Universidade de Évora - Instituto de Investigação e Formação Avançada

Programa de Doutoramento em Ciências da Terra e do Espaço

Área de especialização | Processos Geológicos

Tese de Doutoramento

Diagnosis and characterisation of mortars and concrete materials in buildings awarded with the Valmor Prize for Architecture - State of conservation and contributions for their safeguard

Luís Filipe dos Santos de Almeida

Orientador(es) | José Mirão
Antonio Manuel dos Santos Silva
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A tese de doutoramento foi objeto de apreciação e discussão pública pelo seguinte júri nomeado pelo Diretor do Instituto de Investigação e Formação Avançada:

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Abstract

Instituted over a century ago, the Valmor Prize for Architecture is one of Portugal's most prestigious architectural awards, with its bestowal falling under the responsibility of the Lisbon City Council. Nowadays, there is a pressing need to protect and preserve constructions dating back to the 20th century. The absence of a collective awareness of how modern heritage might be threatened could result in many of these architecturally valuable structures not surviving.

In this doctoral thesis, an analysis of the state of conservation and characterisation of the constituent materials of 17 prize-awarded buildings was conducted. This analysis focused mainly on rendering mortars, plasters, and concrete materials, using a characterisation methodology suitable for the proposed objectives.

The work plan was divided into four parts: (1) Selection of case studies, obtaining permission from the respective owners for their access and study, and gathering information about the construction history, including compiling the constructive transformations and conservation/restoration efforts that occurred throughout the lifespan of the buildings; (2) Identification of anomalies, in-situ testing, and selective sampling of concretes and mortars; (3) Experimental campaign applying a methodology for compositional, mineralogical, chemical, microstructural, physical, and mechanical characterization; and (4) Interpretation and discussion of the results.

The obtained results fill a knowledge gap concerning materials and construction techniques related to the advent of Portland cement usage and the abandonment of traditional lime-based binders during the 20th century. This research deepens the understanding of the chronological application of various binder types and construction techniques associated with modernization during the last century. On the other hand, the results provide a substantial number of parameters and essential characteristics useful for applying compatible materials in repair and conservation actions that may be undertaken, which will contribute to safeguarding this valuable historic architectural heritage.

Keywords: mortars, concretes, renders, plasters, binders, aggregates, diagnosis, characterisation, conservation, compatibility, durability, 20th century, Lisbon, Valmor Prize for Architecture.

Resumo[1]

DIAGNÓSTICO E CARACTERIZAÇÃO DE REVESTIMENTOS DE EDIFÍCIOS GALARDOADOS COM O PRÉMIO VALMOR DE ARQUITECTURA – ESTADO DE CONSERVAÇÃO E CONTRIBUTOS PARA A SUA SALVAGUARDA

Instituído há mais de um século, o Prémio Valmor de Arquitectura é um dos mais prestigiados prémios de arquitectura em Portugal, sendo ai sua atribuição da responsabilidade da Câmara Municipal de Lisboa.

Nesta tese de doutoramento, foi realizada a análise do estado de conservação e a caracterização de materiais constituintes de 17 edifícios galardoados com este prémio, particularmente das argamassas de revestimento interior e exterior, assim como de materiais de betão, através duma metodologia de caracterização adequada aos objectivos propostos.

O plano de trabalhos dividiu-se em quatro partes: (1) Selecção dos casos de estudo, pedido de autorização aos respectivos proprietários para o seu acesso e estudo, recolha de elementos da história construtiva, incluindo a compilação das transformações construtivas e de conservação/reabilitação ocorridas ao longo do tempo de vida dos edifícios; (2) Levantamento de anomalias, ensaios *in-situ* e amostragem selectiva de betões e argamassas; (3) Campanha experimental aplicando uma metodologia para a caracterização composicional, mineralógica, química, microestrutural, física e mecânica e (4) interpretação e discussão dos resultados.

Os resultados obtidos permitem, por um lado, colmatar uma lacuna no conhecimento dos materiais e técnicas construtivas relacionadas com o advento da utilização do cimento Portland e o abandono dos ligantes tradicionais à base de cal, aprofundando o conhecimento sobre a cronologia de aplicação dos vários tipos de ligantes e técnicas associadas à modernização ocorrida durante o século XX. Por outro lado, os resultados aportam uma substancial quantidade de parâmetros e de características fundamentais para a aplicação de materiais compatíveis em acções de reparação e de conservação que possam vir a ser tomadas, contribuindo para a salvaguarda deste valioso património histórico-arquitectónico.

Palavras-chave: argamassas, betões, revestimentos, ligantes, agregados, diagnóstico, caracterização, conservação, compatibilidade, durabilidade, século XX, Lisboa, Prémio Valmor de Arquitectura.

[1] The author does not write according to the 1990 Portuguese spelling agreement.

CHAPTER 1. INTRODUCTION

1.1 OVERVIEW

Interest in conserving buildings constructed in the 20th century has grown, leading to an expanding discussion on strategies for protecting and preserving these buildings. Particularly, buildings that exhibit significant heritage value due to their recognized architectural quality and their representation of specific historical periods are receiving increased attention.

A comprehensive understanding of the construction realities and contexts is essential to valorise the built heritage. It involves acquiring knowledge about aspects related to the construction projects and the materials used, placing them within the contemporary construction trends of each period. Such knowledge serves as a foundation for effective repair and conservation efforts.

The buildings that have been awarded the Valmor Prize for Architecture not only hold historical significance within Portuguese architecture and construction spanning over a century, but they also warrant detailed study. Examining them from a preventive and damage-mitigation perspective is important to ensure their preservation. Considering the study period of one hundred years (1902 - 2002), from the award assignment to the beginning of the 21st century, intentionally established as a sufficiently broad interval regarding potential case studies, and because it is important to encompass various historical-architectural periods up to the contemporary era - accompanied by new trends and technological evolution associated with construction methods - the characterisation of wall renders and plasters will facilitate the understanding of the evolution of the selected materials during each period, as well as the degradation processes associated with a specific period and construction context. In addition to wall renders and plasters, in the same context, it was also important to study the concrete used as both structural support and exposed concrete surfaces, known as architectural concrete, which is not covered by renders. This study holds particular significance as it determines the evolution of the strength and durability of these materials.

By gathering a series of case studies, a characterisation methodology was applied, involving various actions, from collecting historical records and elements throughout the building's lifespan, to on-site analysis and observation of their state of conservation, to diagnose the causes and main degradation agents, as well as the application of chemical, mineralogical, microstructural, physical, and mechanical tests to obtain a wide range of fundamental characteristics for understanding the materials and construction techniques.

Another central aspect of this research was the study of the evolution of construction materials throughout the 20th century and their performance during the building's lifespan. The 20th century warrants further study regarding mortars and concretes, including their production and the chronological evolution of the binders utilized, which are still raising the following question: when and where the use of Portland cement began to prevail over traditional lime? From this perspective, the awarded buildings are considered within the best practices in construction in Portugal. It is expected to find in these materials the paradigm of quality and innovation as hallmarks of the excellent architectural project's execution. It serves as a starting point for acquiring this knowledge.

1.2 Objectives

To create knowledge about the construction materials used and their performance over time, to evaluate the condition of preservation, and to provide guidance for conservation and rehabilitation actions on such significant architectural and cultural heritage, the following objectives were proposed:

1. The inspection of the main anomalies in rendering mortars, plasters and concretes of the award-winning buildings, the degradation processes, and their causes;
2. The mineralogical, chemical, microstructural, physical, and mechanical characterisation of samples of rendering mortars, plasters and concretes;
3. The relationship between degradation processes occurring within a time span of less than 100 years and the materials and construction methods employed;
4. Recommendations for materials to be used in the rehabilitation and preservation of that built heritage.

1.3 Thesis Organisation

The organisation of the thesis follows an unconventional structure for presenting the results and conclusions. It includes the presentation of published and peer-reviewed works in international journals indexed in scientific databases.

This doctoral thesis is structured as follows:

Chapter 1 – This introductory chapter provides an overview of the topic, the motivation behind this research, the established objectives, and the overall organisation of the document.

Chapter 2 – A chapter covering the general concepts of the construction materials studied and their main applications.

Chapter 3 – A chapter that provides a historical review of the Valmor Prize for Architecture in its historical context.

Chapter 4 – A chapter addressing the methodology, the case studies, their contextualization, and the developed research through the four published research papers.

Chapter 5 – A chapter that summarises the main conclusions of the developed work. It also presents suggestions for future developments and relevant aspects requiring further investigation.

Chapter 6 – A final chapter with a summary of the dissemination work and training actions conducted during the research period.

CHAPTER 2. MATERIALS

This chapter covers the concepts and generalities of the materials studied, namely mortars and concretes.

2.1. MORTARS

Rendering and plastering mortars are materials used in construction for coating and finishing exterior and interior walls, respectively. They are typically applied as a layer or a set of layers over a base substrate, such as masonry or concrete, to provide a smooth, even, and decorative surface and the protection of the walls.

In general, mortars can be used to perform several different tasks, being applied as renders to protect the masonries against the external actions or plasters to perform similar tasks in interior conditions, but also to repoint existing joints in which mortars are non-existent or non-functional and as grouts to inject inside masonries with structural problems. Other applications could be referred to, such as the case of bedding mortars - used to create a joint between new elements - or those used for cosmetic stone repair [1].

Mortars are usually mixtures of binder, sand, and additives that are designed to adhere to the substrate and provide a durable and weather-resistant finish.

Among the various types of mortars, the most common types - being those addressed in this research – include the following ones.

2.1.1. Lime-based mortars

Lime mortar is a versatile and enduring construction material with millennia of history. It comprises a mixture of slaked lime, water, and aggregates like sand. Its importance in historical architecture is evidenced by its use in iconic structures. The lime mortar's resilience and compatibility with diverse masonry materials like bricks and stones have rendered it an indispensable building element throughout various historical periods.

Lime is emerging from limestone calcination and slaking processes and encompasses non-hydraulic and hydraulic variants. The former, often called "fat lime" or "air lime," relies on carbonation for setting, gradually absorbing carbon dioxide from the atmosphere and reverting to a limestone-like state. This unique property, i.e. the carbonation, makes non-hydraulic lime mortar breathable and suitable for heritage structures that require moisture regulation to prevent deterioration.

Lime with hydraulic properties, derived from limestone with inherent impurities, can set even in humid conditions. This variant has proven invaluable for constructions subjected to challenging environmental conditions, such as water exposure. Its varying degrees of hydraulicity provide options for achieving specific hardness levels and setting speed.

Various additives or additions are sometimes used in lime mortars to enhance their properties or address specific construction needs. The choice of additives depends on the desired outcome and the project's specific requirements. The most common types of additions in lime mortars include pozzolans. Pozzolans are materials such as volcanic ash, brick dust, or certain types of clay that can be added to lime mortars. They react with lime to form additional chemical bonds, increasing the mortar's strength and durability. Common pozzolans used include metakaolin, fly ash, and natural pozzolans.

While the Industrial Revolution introduced alternative materials like natural and industrial Portland cements, which supplanted lime mortars in many contemporary projects due to their rapid setting and strength, their value endured in restoration and conservation efforts. Conservation architects and builders continue to employ lime mortars to safeguard the authenticity and longevity of historical buildings, as their properties align with preserving original structures. This resurgence showcases the intersection of traditional craftsmanship, sustainable building practices, and the ongoing recognition of lime mortar's role in maintaining architectural heritage.

2.1.2. Cement-based mortars

Until the end of the 18th century, the binders used were limestone-based, either air or hydraulic lime, and could be found in interior and exterior mortars or aesthetic elements [2]. After this period, the development of hydraulic binders led to the appearance of natural cement, particularly with James Parker's 1796 patent for the so-called "Roman cement". This cement results from the burning of marl septarian-type rocks, followed by grinding and producing a brown-coloured powder that sets quickly when mixed with water. Natural cements are characterised by the fact that the composition of the raw materials has not changed. The properties of these binders result from the chemical and mineralogical characteristics of the original materials, which are directly related to the place of extraction and the firing conditions.

The original limestone marls were supposed to have a clay content of 22-35%, resulting in a primarily hydraulic binder due to the high clay content and low free lime content [3-6]. Natural cement is generally produced at lower temperatures, between 850°C and 1100°C, when compared to modern Portland cement, resulting in primarily belitic cement, hence its hydraulicity. At the same time, the formation of calcium aluminates is also typical. The quick setting of natural cement is closely linked to the formation of calcium aluminium oxide carbonate (or carbonate hydroxide) hydrates [7, 8]. Following a dormant period, which can vary based on the origin of the natural cement and how it was processed through calcination, further strength is achieved due to the hydration of calcium silicate hydrates [9].

Natural cement was mainly produced in England from the end of the 18th century to the beginning of the 19th century. From 1802 onwards, industrial production began in France, with various production sites appearing later, from 1940 onwards. At the same time, mass production also began in Germany, after an initial period of imports from England and the first local production in 1820. Austria, one of the largest producers of natural cement, began large-scale production in 1842 [10]. With the development of artificial cements, particularly Portland cement, the production of natural cement decreased, initially affecting English manufacture but eventually limited to a small number of European producers.

Natural cements, which preceded today's ordinary Portland cement, were a significant innovation in the construction industry during the 19th-century Industrial Revolution. Natural cements were known for their quick-hardening and water-resistant properties, unlike hydraulic limes. However, their prominence dwindled with the introduction of Aspdin's ordinary Portland cement in 1824, primarily due to Portland cement's superior rigidity and hardness [11]. John Aspdin obtained a patent for a substance created through the combination of limestone and clay. He named this product "Portland cement" due to its perceived similarity, when solidified, to Portland stone, a type of limestone commonly used in English construction. Aspdin's product might have been under-burned to meet the standards of actual Portland cement. Isaac Charles Johnson may have developed the original prototype in Southeastern England around 1850[2].

In Portugal, the industrial activity related to the manufacturing of hydraulic lime and natural cement had been in operation for several decades. This was the case even though some scholars expressed pessimism about this industry. One notable figure in this context was the engineer José Paixão Castanheira das Neves, who, in 1890, provided an assessment of the state of the national natural cement industry in his report titled "Estudos sobre cimentos naturais” [12, 13]: "(...) *The cement industry in Portugal has so far had very little development* (...) *Our industry is insignificant, which is all the*

[2] In https://www.britannica.com/technology/cement-building-material/History-of-cement.

more surprising given that the consumption of cement and hydraulic lime imported into the country has increased considerably in the last five years, from 1885 to 1889 (...)"[3].

In the historical context of Portugal, a notable factory existed known as the "Empresa de cal hidráulica e cimento natural da Rasca," founded in Quinta da Rasca, Alcântara, (Lisbon) in 1866. This company held the distinction of being the first Portuguese establishment exclusively dedicated to the production of natural hydraulic binding materials. Its primary focus was on extracting and processing marl sourced from the municipality of Setúbal in the Serra da Pedra Branca region. The limitations in the quality of its binder products, attributed mainly to the relatively primitive manufacturing techniques and the absence of robust quality control procedures, the cement sold under the "Rasca" brand faced challenges and was deemed unsuitable for maritime construction projects, leading to its relatively short-lived presence in the market. As a result, the "Empresa de cal hidráulica e cimento natural da Rasca," operated for only 11 years. Initially known as “Fábrica da Rasca” and later as “Fábrica do Outão”, the factory had undergone relocations and transformations. It had previously been situated at the Quinta da Rasca site, with a later unsuccessful attempt at natural cement production in Altinho (Belém/Ajuda) due to inappropriate raw materials sourced from the Alvito quarry in Lisbon. In 1906, this factory made a significant shift by transitioning to the production of Portland cement [14-17].

In 1873, a new company emerged in Cabo Mondego near Figueira da Foz, in the centre of Portugal, where industrial production of natural cement began. The Cabo Mondego Company began by exploiting the bituminous coal and marl-carbonate formations available in the Cabo Mondego region. From 1884 onwards, the Cabo Mondego industries produced white lime, hydraulic lime and Natural cement, probably the best quality natural cement production in Portugal [18].

Despite the advances in cement production in Portugal, there were still many technical difficulties. The high extraction costs and the low quality of coal led to continued imports from France and the United Kingdom [19]. Until the end of the 19th century, Portugal imported most of the cement on the domestic market until the binder production industry began to take hold [15,16].

[3] Cf. Ferreira, Carlos Antero (1989). Betão: a idade da descoberta. Lisboa, Ed. Passado Presente; and Relatorio apresentado à direcção da 3ª circumscripção hydraulica em 21 de Abril de 1890, in “Revista de Obras Publicas e Minas”, Lisboa, XXII, June-Septembre, 1891, p. 181 (translated from portuguese).

From 1881-82 onwards, a scientific approach to cement study began through the Military School's teaching engineers and the materials mechanics department of the Lisbon Harbour Works Directorate (1886). José Paixão Castanheira das Neves (1849-1922) was its first director, presenting various studies on Portuguese building materials to draw up guidelines for Portuguese Portland cement [14].

In 1887, the increase in industrial production of building materials and the need to assess and control their quality led to the development of the first official quality control laboratory. Castanheira das Neves published a relevant study on Portuguese natural cements [14, 18]. This study categorised Portuguese natural cements into two groups: slow-setting natural cements (obtained from the Rasca and Cabo Mondego factories) and fast-setting cements (obtained from Pataias, Maceira and São Pedro de Moel) [14].

Between 1891 and 1894, various studies focused on producing hydraulic lime, natural cement and the characteristics of the materials available on the market. In 1892, the first company to produce artificial Portland cement was founded - Cimento Tejo in Alhandra (now CIMPOR). The licence for production was granted in 1894, marking the beginning of 10 years of exclusivity to produce Portuguese Portland cement [14].

New advances in the research and development of artificial cements and Portland cement led to the latter becoming the recommended cement for most works requiring a hydraulic binder, particularly in reinforced concrete. Throughout the 20th century, a great deal of research was carried out into the most suitable raw materials, the strength of the processed materials, and production methods, culminating in technological advances that have led us to the point where today there are precise standards for the composition, specifications and conformity criteria for ordinary cements [20] to be applied in civil engineering works.

The types of cement defined in EN 197-1:2011 [20] for the European Union include:

- Ordinary Portland Cement (CEM I): This is the most common type of cement and is used for a wide range of construction applications. It can be subdivided into several classes based on initial and final strength.
- Portland Cement with Additions (CEM II): This type of cement includes additions of materials such as limestone filler, slag, fly ash, or pozzolans to improve specific properties of the cement. It is also divided into classes based on the type and quantity of addition.

- Blast Furnace Portland Cement (CEM III): This type of cement is produced with a significant amount of blast furnace slag. It is also subdivided into classes based on the amount of slag.
- Pozzolanic Portland Cement (CEM IV): This cement contains a substantial amount of pozzolan and is used in applications where resistance to chemical attack is essential.
- Composite Portland Cement (CEM V): This type of cement combines slag and pozzolan additions. It is used in specific applications that require resistance to aggressive environments.

In the context of cement classification, specialised or non-standard cements are used for specific and sometimes niche applications. These cements may have unique properties or compositions that suit particular construction needs. Some examples of uncommon cements include:

- Expansive Cements: These cements contain additives that cause the concrete to expand during the early stages of hydration. They are used in applications where controlled expansion is desirable, such as prestressed concrete.
- Sulfate-Resistant Cements: These cements are designed to resist the effects of sulfate attack and are used in environments where the concrete may be exposed to sulfate-rich soil or water.
- White Cements: White cements are used when white or light-coloured concrete is desired for architectural or decorative purposes. They have a lower iron and manganese content, making a whiter appearance.
- Coloured Cements: These cements are blended with pigments to produce concrete in various colours for aesthetic purposes.
- High-Alumina Cements: High-alumina cements have a high alumina content and are used in applications requiring resistance to high temperatures and chemical corrosion, such as refractory linings.
- Calcium Aluminate Cements: These contain calcium aluminate compounds and are used in specialised applications like rapid-setting concrete or as a binder in refractory castables.
- Geopolymer Cements: Geopolymer cements are alternative binders that do not rely on traditional Portland cement chemistry.

Portland cement-based mortars, vital constituents of modern construction, renowned for their strength and versatility, consist of mixtures comprising Portland cement, fine aggregates like sand, water and additions, extensively utilised to bond masonry units such as bricks, stones, and concrete blocks. These mortars are nowadays indispensable, offering reliable adhesion, flexibility, and durability for diverse construction projects, aligning with the demands of residential, commercial, and infrastructural endeavours.

2.1.3. Gypsum-based mortars

Gypsum-based mortars are mixtures of gypsum, water and fine aggregates used in coating walls, ceilings and internal surfaces of buildings. Gypsum is the main binder of these mortars and plays a fundamental role in their ability to harden and adhere. Gypsum mortars are popular as plaster applications, including decorative works, due to their fast-setting properties and ease of application.

The knowledge of plaster production and application was originally introduced to Europe by the Greeks, who had acquired it from the ancient Egyptian and Minoan civilizations. Subsequently, the art of stucco and its intricate techniques were transferred to the Romans, who documented these methods in their construction manuals. Although stucco was utilized during the Roman era and the European Middle Ages, its use gradually declined in medieval Europe, only to experience a revival during the Arab presence from the 8th to the 15th centuries [21, 22].

However, it was not until the 17th century that stucco once again gained prominence in European decorative arts [21]. This resurgence was driven by the aesthetic demands of the Baroque period.

The distinction between gypsum and lime plasters remained unclear until the 18th century. During this period, researchers like Lavoisier began delving into the principles of gypsum technology, and this research continues to the present day. However, the traditional craftsmanship in this field was initially hesitant to embrace these scientific findings. It was not until the latter part of the previous century that this craft evolved into a modern industry, with a similar evolution seen in the mass production of lime [23, 24].

In light of these historical developments, it becomes essential to differentiate between gypsum and lime plasters, especially in conservation and restoration projects. However, it is equally important to acknowledge and leverage their exceptional compatibility. As noted by Gárate-Rojas (1999) [25], the combined use of these two materials leads to the creation of highly versatile products, both in terms of composition and application, offering a wide range of possibilities [24].

In Portugal, gypsum plasters became increasingly prevalent in construction during the latter part of the 18th century. It was commonly employed as the primary binding agent for plasters or in conjunction with calcitic air lime. In Portuguese architecture, the 19th century and the initial quarter of the 20th century witnessed the zenith of gypsum plaster usage. Gypsum plasters found particular favour for their decorative qualities, perfectly harmonizing with the prevalent architectural trends of the era [24].

2.1.4. Blended-binder mortars

Blended mortars, particularly lime-cement mortars, represent a dynamic facet of modern construction, fusing the attributes of multiple materials to achieve specific performance characteristics; these mortars encompass combinations of Portland cement, lime, fine aggregates like sand, and water, often tailored to provide a balance between the strengths of cement-based mortars and the historical preservation qualities of lime-based mortars; their emergence reflects the need to address both structural requirements and the preservation of historical architecture [26].

Historically, lime-cement mortars have evolved as a response to the challenges posed by the interplay of traditional construction and modern demands; they seek to leverage the strengths of both Portland cement and lime, combining cement's rapid setting and compressive strength with lime's flexibility, and compatibility with historic masonry.

The composition of lime-cement mortars can vary widely, depending on the specific application, desired properties, and preservation goals; the ratio of Portland cement to lime, as well as the type of aggregates used, play a pivotal role in determining characteristics such as workability, strength, permeability, and compatibility with the existing structure [26, 27].

2.1.5. General applications of mortars

Rendering mortars are applied to the external walls of buildings to provide protection against weathering, improve aesthetics, and enhance thermal insulation.

Plasters are used to create smooth and even surfaces on interior walls, which can be painted or decorated. Plasters are often used for finishing coats or decorative purposes. Some common types include gypsum-based plasters.

Renders and plasters can be textured, sculpted, or patterned to create decorative effects or mimic other materials such as stone or marble.

Both rendering mortars and plasters play a crucial role in improving the appearance, protection, and longevity of buildings while providing a versatile canvas for various finishing options. The specific choice of mortar or plaster depends on factors such as the substrate, location, desired aesthetics, and functional requirements.

2.2. CONCRETE

Concrete is a composite construction material characterised by its versatile and durable nature. Some of the main applications of concrete include buildings, bridges and infrastructure, roads and pavements, dams and hydraulic structures, marine structures, high-rise buildings, prefabricated elements, retaining walls, foundations, and decorative elements.

It comprises primarily a binder (generally hydraulic, although other binders can be used, eventually air binders), aggregates (such as sand, gravel, or crushed stone), water, and optional additives. It is formed by mixing these components to create a cohesive mixture that can be moulded into various shapes and structures. In the case of Portland cement-based concretes, the hardening is due over time through a process known as hydration, in which the cement particles react with water to form a solid mass.

Reinforced concrete, a material in which concrete hardens around embedded metal, typically steel, is credited with its invention to Joseph Monier, a gardener in Paris who crafted concrete garden pots and tubs reinforced with iron mesh. He received a patent for this innovation in 1867 [28]. The reinforcing steel, whether in rods, bars, or mesh, is critical in providing tensile strength to the material. Ordinary concrete cannot alone effectively withstand various types of stresses, including those caused by wind, earthquakes, vibrations, and bending forces. Consequently, it is often unsuitable for many structural applications. However, in the case of reinforced concrete, the combined tensile strength of steel and the compressive strength of concrete results in a material capable of supporting substantial stresses across significant spans. The fluidity of the concrete mixture allows for precise placement of the steel reinforcement at or near the points where the most significant tensile stresses are anticipated. This integration of concrete and steel reinforcement grants reinforced concrete its exceptional structural capabilities.[4]

Concrete is widely used in construction due to its exceptional strength, durability, and adaptability. It offers significant advantages by comparison with other structural materials

[4] According to the definition of concrete in https://www.britannica.com/technology/concrete-building-material

(wood, masonry, etc.), including the ability to be moulded into intricate shapes, its resistance to fire and weathering, and its capacity to support heavy loads. Additionally, concrete's properties can be adjusted through variations in its composition, allowing for tailored solutions to meet specific project requirements.

Throughout history, concrete's evolution has seen formulation advancements, leading to specialised types such as reinforced concrete, precast concrete, and various high-performance variants. These adaptations have enabled the construction of diverse structures, ranging from buildings, bridges, and dams to roads, pavements, and even artistic installations.

The entrepreneurial skills of François Hennebique, the father of the best-known reinforced concrete patent at an international level, will be fundamental to the widespread use of this new material in construction in Portugal. The first period of use of reinforced concrete in Portugal began in the early 90s of the 19th century. Several references define this period: 1892 (April), which coincides with the granting of the first Cottancin patent in Portugal by José Martins; 1894 (September), when the first Hennebique patent was registered in Portugal [29].

In the history of the advent of reinforced concrete in Portugal, it must be recognised the relationship with the birth of national Portland production was fundamental to its rapid diffusion in the country. 1894 was the year in which it was decided to experiment with the production of artificial Portland. The country's first Portland cement was produced by a cement industry in Alhandra (Fábrica Cimento Tejo) to reduce Portland imports from countries such as France, Germany or England and increase industrial and global development [29].

At the outset of the 20th century, as the number of reinforced concrete structures increased, a necessity arose for the establishment of comprehensive regulations governing this innovative material. The "Regulation for Reinforced Concrete" [30] emerged in response to this need in 1918. This regulation drew its foundation from advanced French technical documents, considered among the most progressive of that era. Further addressing the technical requirements associated with the potential of reinforced concrete, Decree No. 25948, dated 16 October 1935 [31], was introduced. This decree marked the evolution of regulations governing reinforced concrete and led to the creation of the "Reinforced Concrete Regulations" (RBA). The safety evaluation, rooted in the concept of permissible stresses for both steel and concrete, was retained from the initial regulation. Calculations of stresses were based on a linear elastic model.

In accordance with the standard, the permissible stress values had to consider the fatigue limits stipulated for steel and concrete materials.

The advancement of new concepts, calculation methods, and technical guidance for designers saw significant progress, driven both nationally by the establishment of the National Laboratory for Civil Engineering in 1946 and internationally by the formation of the "Comité Européen du Béton" in 1953. This progress justified the publication of a new regulatory framework known as the “Regulation of Reinforced Concrete Structures” (REBA) [32], which superseded the previous RBA [31]. The REBA introduced modifications to several essential aspects in calculating reinforced concrete elements.

On 30 July 1983, Decree-Law No. 349-C/83 [33] came into effect, introducing the “Regulations for Reinforced Concrete and Prestressed Structures” (REBAP). These regulations, coupled with the “Safety and Actions Regulations” (RSA) [34], continued the design philosophy initiated with the REBA [32], which focused on verifying limit states. While the changes from the REBA were not revolutionary, notable differences arose in terms of safety verification criteria and the definition of limit states. Additionally, concepts related to the quantification and combination of actions and safety coefficients were introduced within the REBAP framework.

In the 1990s, the European Union undertook a comprehensive effort to standardize regulations across various sectors. Within the field of civil engineering, and with a specific focus on reinforced concrete, a pivotal moment occurred with the publication of Eurocode 2, commonly referred to as EC2 [35]. This publication marked a significant shift from national regulations to European standards, with adaptations made for each country by defining their respective national annexes.

In Portugal, adopting EC2 [35] brought about distinct changes compared to the existing REBAP [33] regulations. These changes notably maintained the reference classification for various concrete strength requirements, including compressive strength. Revising these strength classifications has been an ongoing process within Portuguese regulations.

CHAPTER 3. THE VALMOR PRIZE FOR ARCHITECTURE

3.1 BACKGROUND

During the last quarter of the 19th century, an attempt was made to expand and reorganise Lisbon's urban area on the initiative of specific sectors of the liberal bourgeoisie interested in the valuation of new land to be urbanised. There was a conflict between modernisation through the new technologies available and the romantic spirit with a revivalist basis. Lisbon left its predisposition to grow along the banks of the Tagus River to give way to a city focused on the occupation of the interior plateaus through the imposition of new concepts of urbanisation that emerged in the second half of the century in Europe as a result of the new social organisation emerging from the Industrial Revolution.

The industrial and financial bourgeoisie appropriated the city and built relatively tall, more profitable buildings along the grand avenues. The change in mentality produced rapid, safer and more profitable construction along the new avenues. The need to build buildings with new functions, housing, and city support facilities caused some disruption. There had to be greater coherence between the materials available for construction and the desire to build in the romantic style that still existed in Portugal. On the one hand, industrial buildings already required materials such as iron, steel, glass, and later reinforced concrete. However, architects and engineers could not apply these materials with the desired plastic quality and speed that industrial equipment showed in its construction. There was an evident lag in the European context, even regarding imported influences. Parallels with the rest of Europe provide a better understanding of the development of 19th-century architecture and town planning in Portugal and the appearance of the Valmor Prize for Architecture [36].

Most construction in Portugal continued to refuse any dialogue between what is considered "Architecture" and what is understood as mere utilitarian construction. It is thus in this complex universe of cultural transformations and the search for new solutions that the Valmor Prize for Architecture was instituted in Lisbon in 1902, based on the legacy of the Viscount of Valmor, Fausto de Queirós Guedes, whose regulations state that:

"*A prize will be given annually in two equal parts to the owner and the architect of the most beautiful building or house built in Lisbon, on condition, however, that this new* *house or restoration of an old building has an architectural style, Classical, Greek or Roman, in short, a style worthy of a civilised city*.[5]"

3.2 EMERGENCE OF THE VALMOR PRIZE FOR ARCHITECTURE

Established by the will of the 2nd Viscount of Valmor, Fausto de Queirós Guedes, the Valmor Prize for Architecture was an instrument for affirming Portuguese architecture throughout the 20th century. Its creation followed Parisian models, showing attention to artistic practice of profound significance and social impact, going through a crucial moment of affirmation, but also of questioning, driven by engineers and the new needs of the industrial and service city that architects, adherents to Art Nouveau and the first modernisms, knew how to assume [37].

These issues, already common in the rest of Europe, were experienced in Portugal tenuously due to its evident backwardness. Although Fausto Queirós Guedes' will dates back to 1898, it was only at Lisbon City Council session of January 21, 1903, that the regulations for what became known as the Valmor Prize were approved. Although it was not a quick process to structure the beginning of the award of the Valmor prizes, due to the Lisbon City Council having to attend to the terms of inventory of assets, a regulation was made by the subsequent Lisbon City Council following the guidelines left by the legacy [36].

Viscount Valmor's testamentary provisions, which were strictly followed, reveal the taste for eclectic forms that were dominant at the time as regards the various artistic styles. The Valmor Prize became an almost obligatory reference and a quality certificate periodically awarded to architecture works. In this context, in 1938, due to the devaluation of that legacy, the Municipality of Lisbon decided to institute a complementary Prize, called the Municipal Prize for Architecture, which was instituted on October 28, 1943, to maintain a material value. The Valmor Prize and the Municipal Prize for Architecture are thus the responsibility of the Lisbon City Council and are awarded jointly [36].

These awards are intended to stimulate the design and construction of architecturally significant buildings in Lisbon, honouring architects and owners. Both prizes reflect the history of the evolution of architecture in Lisbon and in the country, which include and have been considered the best examples. The Municipal Prize for Architecture was

[5] Translated excerpt from the Will of the 2nd Viscount of Valmor, Manuscript (1897), C.M.L. Archives, Historical Fund. Historical Fund.

established in order to prevent the corseting of the clauses of the legacy institution and the monetary erosion caused by the four decades that had elapsed [36].

3.3 THE REGULATIONS OVER TIME

The first amendment to the regulation only took place on April 29, 1942, in which the revision is symptomatic of the care taken to preserve the Viscount legacy, keeping as close as possible to the first regulation, clarifying dubious aspects. It is essential to mention that, even if the vast majority of the awarded works are, in fact, houses or residential buildings (primarily until the end of the 1960s), if the choice were limited to that same category, as some interveners proposed, the opportunity to award works such as the *Igreja do Rosário de Fátima* (1938) or the Headquarters of the newspaper *Diário de Notícias* (1940) – the first and second non-residential buildings awarded, respectively - would have been lost [38].

On November 8, 1982, the Valmor Prize became associated with the Municipal Architecture Prize, but the regulations were also completely revised, and numerous changes were made. The most significant of these was the change in the constitution of the jury and works of public initiative can now be awarded. The remaining changes relate to the pecuniary value that increased significantly, once associated with the Municipal Prize, which was always higher than the Valmor Prize.

Two more regulation amendments followed soon after. Regarding the first, it should be noted that the amount associated with the Municipal Architecture Prize – previously only given to the architects who had authored the work – is now distributed in equal parts to the owners and multiplied six times. It was also clarified that only "new works or remodelling or total and integral restoration of buildings" would be considered. As for the second, the merger between the Valmor Prize and the Municipal Prize for Architecture becomes definitive [36].

The latest regulation, approved on December 16, 2003, with retroactive effect from the 1997 Prize, brought some changes, especially following the construction of the 1998 Lisbon's World Exhibition. The scenario of a set of buildings of unquestionable architectural quality and the result of an urban project of unique dimension and projection in the city led to the introduction of landscape architecture as another of the possible categories to be awarded. The attention given to the restoration or remodelling of buildings is also significantly reinforced at a time when several actions to safeguard heritage in the city centre were taking place, and, for the first time, reference is made to the importance of framing and articulating with the surroundings.

CHAPTER 4. DEVELOPED RESEARCH

4.1. METHODOLOGY

The methodology employed in the diagnosis and characterisation of mortars and concretes is described systematically and in detail in each of the published scientific papers.

The methodology was applied to each type of material in an appropriate manner to the scientific questions posed in the objectives, considering that the study should focus on contemporary building materials applied during the construction of the buildings.

To summarise, the materials characterisation methodology employed comprised four distinct phases in the workflow: (1) compilation of history and constructive records, (2) on-site evaluation and anomalies' survey that intends to be a support to the subsequent characterisation, (3) in situ tests and sampling, (4) experimental campaign in the laboratory.

Preparatory tasks, such as the compilation of elements of the constructive history, descriptive memories and technical specifications, contract documents, drawings and processes for the licensing of works that have taken place over the buildings' lifetime were helpful in making clear the constructive context of each building and helped on obtaining information as valuable as the constructive characteristics and the applied materials, although the first decades of awarded prizes often revealed a lack of these records.

4.2. CASE STUDIES

The list of buildings awarded annually during the 20th century counts 60 awards and 41 honourable mentions. Most of these buildings retain the features and functions they were designed, although a small number have been demolished (note that the award does not directly confer a heritage protection status) or remodelled.

Lack of jury consensus or other socio-economic reasons made recognising works from throughout the 20th century some years impossible. However, the chronological list is reasonably well distributed throughout the 20th century, which has allowed to carry out prior work of choosing the awarded buildings considering the premise of at least one building per decade.

After a previous choice of the buildings, the owners were asked to grant authorisation for the study presented here, which proved ineffective in some cases, leading to the reformulation of the choice of case studies. However, after various endeavours, it was impossible to study buildings awarded in the decades of 1910s and 1960s. Two buildings awarded after 2000 were also studied since their construction began in the 20th century.

Thus, the list of case studies, whose architectural and constructive framework is carried out in this chapter, consists of a total of seventeen award-winning buildings. Table 1 shows the case studies and fieldwork list, particularly the anomalies' survey and sampling. The number of samples tested in the laboratory are also listed by type of characterisation.

Table 1 – List of case studies, fieldwork and samples collected and tested in the laboratory

				Rendering and plastering mortars			**Concrete**	
					Number of tested samples			Number of tested samples
#	Case studies	Acronym (*)	Inspection of anomalies	Number of samples	Chemical, mineralogical, and microstructural characterisation	Physical and mechanical characterisation	Number of samples	Physical and mechanical characterisation
1	*Ventura Terra* Building	CVT (1903)	x	5	5	5	-	-
2	*Malhoa* House	CMAG (1905)	x	-	-	-	-	-
3	*Luiz Rau* Building	AR49 (1923)	x	11	11	8	-	-
4	*Nossa Senhora de Fátima* Church	IRF (1938)	x	8	8	8	4	4
5	*Bernardo da Maia* House	CBP (1939)	x	8	8	6	-	-
6	*Diário de Notícias* Building	DN (1940)	x	12	12	12	4	4
7	*Cristino da Silva* Building	AAC (1944)	x	6	6	6	-	-
8	Laboratories of Pasteur Institute of Lisbon	LIP (1958)	x	2	2	2	7	7
9	*América* Building	EUA53 (1970)	x	6	6	5	2	2
10	*Franjinhas* Building	FRAN (1971)	x	-	-	-	7	7
11	Calouste Gulbenkian Foundation Headquarters and Museum	FCG (1975)	-	1	1	-	7	7
12	*Sagrado Coração de Jesus* Church	ISCJ (1975)	x	-	-	-	1	1
13	Jacob Rodrigues Pereira Institute	JRP (1987)	x	1	1	-	7	7
14	The Knowledge Pavilion	PCV (1998)	x	-	-	-	12	12
15	C8 Building (Faculty of Sciences of the University of Lisbon)	C8 (2000)	x	-	-	-	6	6
16	*Atrium Saldanha* Building	AS (2001)	x	-	-	-	8	8
17	New University of Lisbon Rectory	UNL (2002)	x	1	1	1	6	6
			Total	**61**	**61**	**53**	**71**	**71**

(*) - The acronym consists of an alphanumeric code comprising the abbreviation of the building's name and the year it won the Valmor Prize for Architecture.

4.3. Diagnosis and the state of conservation of renders, plasters and reinforced concrete surfaces of Valmor Prize award-wining buildings

The following research paper presents the main construction types and characteristics of seventeen case studies, along with an inspection of the main anomalies detected in renders, plasters, and concrete surfaces. The applied methodology made it possible to classify plasters, renders, and concrete materials according to their state of conservation. This study aims to contribute to future conservation actions that will guarantee better preservation concerning sustainable materials, i.e., compatible materials to the existing ones that enhance the durability of the old buildings and minimise the use of new materials.

The results point to renders and plasters of the buildings analysed globally in a reasonable state of conservation. However, it was verified that when compared to the Reinforced Concrete Buildings (RCBs), which, in this case, include the buildings whose structure is entirely of reinforced concrete, the Pre-Reinforced Concrete Buildings (PRCBs) presented a greater extent, degree, and severity of degradation.

Since the PRCBs are also the oldest buildings (1903 to 1944), the higher degree and extent of degradation of the assessed materials can be attributed to the more prolonged exposure to the agents of degradation, as well as to the construction typology that makes them particularly vulnerable to the water action and other agents related to water, such as salts crystallisation.

Concerning architectural concrete, which is from the buildings' structure, constructed between 1965 and 2002, the major issues detected are related to reinforcement corrosion. This is primarily caused by low concrete coverings of the reinforcement bars and possibly other factors like concrete porosity that favoured carbonation. However, no direct correlation was found between the average thickness of concrete cover and building age, nor did differences in carbonation depth was evidenced to be linked to concrete quality. Despite the limited corrosion-related anomalies, the overall condition of architectural concrete surfaces remains reasonable.

Regarding non-architectural concrete, from the buildings' structure as well, the carbonation front has typically reached the reinforcement in most cases.

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Article

20th-Century Award-Winning Buildings in Lisbon (Portugal). Study of Plasters, Rendering, and Concrete Materials Aiming Their Sustainable Preservation

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Abstract: Conservation, increasing the useful life period of existing significant buildings with minimum consumption of new materials, as much as possible of low-embodied energy, is an important step towards sustainable rehabilitation, while also contributing to the preservation of the cultural heritage. In the context of 20th-century buildings' conservation, the knowledge of construction techniques and applied materials is essential to pursue sustainable preservation and rehabilitation actions. This paper presents the main construction types and characteristics of a set of architecture award-winning buildings in Lisbon (Portugal) between 1903 and 2002 along with an inspection of the main anomalies detected in renders, plasters, and concrete surfaces. The applied methodology made it possible to classify plasters, renders, and concrete materials according to their state of conservation. The study of 20th-century buildings is justified by the intense renovation activity in the city centres, which leads to the loss of their outer layers and their historical and original values. This study aims to contribute to future conservation actions that will guarantee better preservation concerning sustainable materials, i.e., compatible materials to the existing ones that enhance the durability of the old buildings and minimize the use of new materials.

Keywords: render; plaster; concrete; conservation; sustainable intervention; 20th century; built heritage

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1. Introduction

The conservation of historic buildings is a cultural need, and a sustainable aim, as it allows avoiding demolition and reconstruction, spending high quantities of new raw materials.

Buildings' conservation, especially those with remarkable architectural interest, requires the use of methodologies that should include (1) in-depth knowledge of the social and construction environments, namely the analysis of the project design and the materials used, framing them into the respective constructive periods and (2) support for repair and conservation actions defined by worldwide recognised principles [1–3].

Surveying the building's condition is another step to ensure the right maintenance and comprehensive actions whenever there is a need to intervene in the built heritage for its preservation.

One issue to point out is the replacement of plasters and renders over time. Even if keeping their original constructive characteristics, buildings may undergo changes in their envelope or indoors. Substitutive rendering mortars should be considered only when there is no way to preserve the pre-existing ones or to fulfil lacuna, and the formulation of those new mortars should be compatible with the substrates and with the pre-existent mortars, to enhance the life period of the existing elements simultaneously with keeping their authenticity and cultural value. The development of compatible materials is a complex task that depends on many factors, such as the type of aggregates and binders, the binder content, the aggregate grain size [4], and the masonry characteristics [5,6], amongst others. Additionally, the mortars and concretes compositions must be developed with local raw materials, to avoid distant sources, thus minimising the environmental damage, applying life cycle principles.

Iconic buildings and architectural landmarks from the 20th century, whether as part of national or world heritage, have extreme cultural importance which in some cases led to a high level of protection defined by law to prevent alteration, invasive refurbishment, or demolition. Thus, adequate maintenance actions are mandatory as well as a correct diagnosis of their state of conservation. To confirm that, several studies mention the application of survey methods before the design of any intervention program [7–14].

In Portugal, the importance of the buildings constructed in the 20th century is not much recognised yet, despite the efforts made by some authorities and buildings' owners towards their preservation [15].

The present study concerns masonry and concrete buildings awarded with the Valmor Prize for Architecture in Lisbon, Portugal, aiming their sustainable preservation. The award-winning buildings epitomise the history of Lisbon's unique architecture for more than a century that needs to be studied and maintained. Furthermore, this award establishment coincides with the beginning of the 20th century, when the adaptation of the construction contexts was imposed by the auspices of industrialisation and the advent of new technologies and materials. The introduction of Portland cement and reinforced concrete allowed higher construction speed and major architectural breakthroughs, contributing to the decline of traditional materials, namely the lime-based ones, and leading to a disruption in the building construction paradigm.

Seventeen award-winning buildings from 1903 to 2002 were studied regarding the state of conservation of their renders, plasters, and hardened concrete surfaces. A survey of existing anomalies was carried out using a methodology that comprised visual inspection and, whenever possible, non-destructive in situ tests [16,17].

This work does not intend to be representative of ordinary buildings, but rather, it aims to understand and evaluate the advances achieved in each construction period in Portugal during the 20th century concerning the construction technologies and materials, based on buildings of unquestionable architectural value.

Considering these aspects, a summary of the main characteristics of these buildings will be presented to assess the influence that the materials and technologies used may have in their state of conservation. This work also aims to support future conservation actions according to the best practices towards the preservation of the studied buildings, as they are still in use and have a significant cultural, historical, and architectural importance. In this work, an attempt to correlate the state of conservation with the buildings' age and their typology was also carried out.

This article presents the first results of the evaluation of the current state of conservation of the renders, plasters, and hardened concrete surfaces based on visual inspection. A comprehensive characterisation study of these materials is being carried out to obtain a complete diagnosis that avoids unnecessary demolition of elements and to provide data concerning the choice of compatible and sustainable repair materials. It is expected that the presented results would be complemented and related with data of the ongoing material characterisation to accomplish the following compatibility criteria [18–20]:

(a) Mechanical compatibility. It must be ensured that excessive stress does not develop in covering and jointing cementitious repair materials, failing the support or surrounding pre-existing materials. Excessive stresses should not be transmitted to the pre-existing structural/masonry elements, so the knowledge of the modulus of elasticity, compressive strength, and adhesion characteristics are required.

(b) Physical compatibility. It is related to the capillary rising and drying of water, and the permeability of liquid water and water vapour. In masonry buildings, the water drainage off the support implies that the water vapour permeability must be high, and the capillary absorption of water must be low to moderate with a high drying capacity. Therefore, the porous structure must be evaluated.

(c) Chemical and mineralogical compatibility. It is related to the binder and aggregate types, and their salt content. It is intended that the new mortars and other composite materials used for repair do not give rise to expansive reactions or harmful reaction products, and they do not contain high levels of soluble salts nor favour their crystallisation.

In addition, and to ensure that the substitutive materials have identical characteristics to the original ones, the binders used must be similar, which requires the characterisation of the original binders and the aggregates should have the same colour, nature, shape, and a similar particle size distribution.

These criteria will ensure that the repair materials will not contribute to the degradation of pre-existing elements and will be able to protect the existing walls and structures. They must be reversible, durable, and, finally, they must not harm or deprive the buildings of their architectural character and cultural value [21]. To this extent, the knowledge of the physical, mechanical, chemical, and mineralogical characteristics will make possible the design of a rehabilitation methodology.

2. The Valmor Prize. Historical Background and Case Studies

The 2nd Viscount of Valmor, Fausto de Queiroz Guedes (1837–1898) was an admirable protector of arts. To recognise the artistic values and architectural works, this nobleman left a donation in his will to be managed by Lisbon's city hall to distinguish the best design works and to stimulate the social function of architecture. Through a regulation that has undergone successive changes, the first award was given in 1902 [22], which is an attribution that is still currently the responsibility of the city hall of Lisbon.

The Valmor prizes awarded during the 20th century can be roughly divided into four main periods [22,23]. The first period (1902–1921) valued single-family buildings. In the second period (1923 to 1950), new regulations led to the promotion of nationalist-inspired architecture. It was a period marked by irregularity on the awarding of prizes which comprised a struggle between architectural traditionalism and modernism. The third period (1951–1980) was characterised by a gradual withdrawal of the nationalist regime's architectural practices and also by the decrease in the prestige of the prize that occurred due to many factors, namely its low monetary value and the award irregularity over that period. In 1958, the new regulation established the possibility of non-residential award-winning buildings. In the last period (1982 onwards), the Valmor Prize was merged with the Lisbon City Prize for Architecture, leading to the attribution of a new monetary reward and another regulation update.

Despite a few award-winning buildings' demolitions that occurred in the past century, most still-existing ones keep the main functions for which they were designed. Nevertheless, some changes have been made due to new user requirements and to improve the existing conditions.

2.1. Case Studies Typology and Architectural Features

Case studies were divided into two main groups according to the construction historical context and typology: the pre-reinforced concrete structure buildings (PRCBs) and the reinforced concrete structure buildings (RCBs), which, in this case, include the buildings whose structure is entirely of reinforced concrete (Figures 1 and 2).

Figure 1. PRCBs case studies (acronym and award-winning year): (**a**)—CVT (1903); (**b**)—CMAG (1905); (**c**)—AR49 (1923); (**d**)—CBP (1939); (**e**)—AAC (1944).

Figure 2. RCB case studies (acronym and award-winning year): (**a**)—IRF (1938); (**b**)—DN (1940); (**c**)—LIP (1958); (**d**)—EUA53 (1970); (**e**)—FRAN (1971); (**f**)—FCG (1975); (**g**)—ISCJ (1975); (**h**)—JRP (1987); (**i**)—PCV (1998); (**j**)—C8 (2000); (**k**)—AS (2001); (**l**)—UNL (2002).

The PRCBs set is characterised by generally holding self-supporting masonry structures, although some buildings may already incorporate reinforced concrete elements. The studied buildings began to incorporate reinforced concrete elements in the 1920s.

Most of the buildings studied are located within the limits of the current Lisbon boroughs of Santo António, Avenidas Novas, and Alvalade, as shown in Figure 3. Their location coincides with the expansion axis of the city centre towards the north.

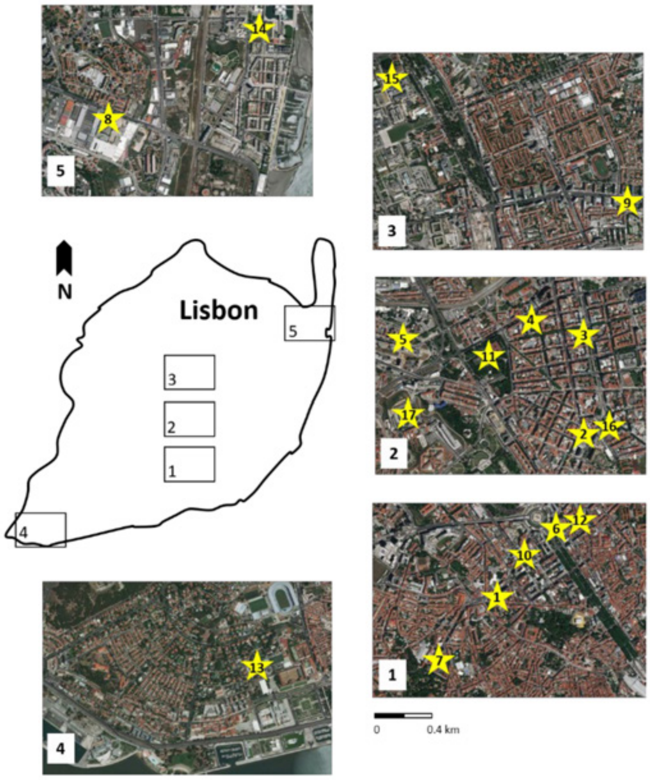


Figure 3. Location of the case studies. Areas 1, 2, and 3 are, respectively, located within the limits of Santo António, Avenidas Novas, and Alvalade boroughs. Case studies: 1—CVT(1903); 2—CMAG(1905); 3—AR49(1923); 4—IRF(1938); 5—CBP(1939); 6—DN (1940); 7—AAC(1944); 8—LIP(1958); 9—EUA53(1970); 10—FRAN(1971); 11—FCG(1975); 12—ISCJ(1975); 13—JRP(1987); 14—PCV(1998); 15—C8(2000); 16—AS(2001); 17—UNL(2002).

"Avenidas Novas" (new avenues) and adjacent neighbourhoods were the main areas of expansion of the city of Lisbon at the beginning of the 20th century. An urban development occurred between the end of the 19th century and the first half of the 20th century at the northern outskirts of the city core. New neighbourhoods and blocks of single-family houses and income buildings were built for the middle and upper-middle classes. The new neighbourhoods were characterised by wide streets, garden areas, and a homogeneous design of the façades. In general, the design of the façades reflected a taste of eclectic architecture and an inspired Art Nouveau outlook.

Throughout the 20th century, the architectural design projects were developed in the modernist style, somewhat embracing the social and political nationalist period installed in the country, gradually abandoned with renewing tendencies, creating new forms of expression, which were formulated as a legacy from the first two post-war generations of architects [24].

The RCBs were built in different locations as the city grew widely. Reinforced concrete developed a very significant route since the beginning of the 20th century in Portugal. Buildings awarded after 1930 confirm a growing trend in the use of reinforced concrete, despite a transition period in which construction mixed tradition with innovation, merging masonry self-supporting walls with reinforced concrete.

Many of the reinforced concrete structure buildings have their surfaces without rendering or coating elements, particularly after the 1970s. In our study, architectural concrete is defined as any visible concrete surface, even from a structural element, which was not deliberately coated. The last award-winning building received its award in the early 21st century; however, as its construction started in the 20th century, we decided to include it in this study.

2.2. Historical Records

Thorough research was done over the Lisbon municipality archives, where the historical collections of buildings' records can be found. However, not all the records have a complete and detailed description of the construction methods and materials applied, especially for the buildings constructed in the first decades of the 20th century. The compilation of the constructive historical elements, namely descriptive memories, specifications, drawings, and work licensing processes (e.g., refurbishment and demolition) that have occurred throughout the buildings' lifetime allow framing the original constructive context, obtaining information as useful as the constructive characteristics, materials used, and the adopted constructive solutions, as well as the original existing renderings.

As a result of this compilation, more detailed knowledge about the use of materials and the main construction characteristics was achieved. Table 1 presents a summary of the main features for each case study, which includes the data collected from the historical records.

Table 1. Summary of case studies' main constructive characteristics.

Case Study	Name	Construction Period	Construction Typology	Award Year	Architect	Structural Characteristics	Main Constructive and Architectonical Features	Main Coatings/ Renderings	Class of Concrete Prescribed in the Design Project
CVT (1903)	Ventura Terra Building	1902–1903	PRCB	1903	Miguel Ventura Terra	Self-supporting masonry walls. Steel beams used as structural elements	Multifamily building. Four floors, basement, and attic. Art Nouveau facade with decorative elements	Limestone coating all over the main facade; polychromatic elements and frieze tiles; rendering mortars	(a)
CMAG (1905)	Malhoa House	1904–1905	PRCB	1905	Manuel Norte Júnior	Self-supporting masonry walls	Single-family house. Two floors and basement. Art Nouveau elements on facades with neo-Romanic decorative elements	Limestone coatings; frieze tiles; rendering mortars	(a)
AR49 (1923)	Luiz Rau Building	1920–1923	PRCB	1923	Porfírio Pardal Monteiro	Self-supporting masonry walls. Concrete slabs and steel-supporting balconies' structure in balconies at rear facade	Dwelling building with five floors, basement, and mansard. East facade adorned with corbels and decorated pilasters. Steel stairs in balconies at rear facade.	Exterior limestone coatings at street level; rendering mortars	According to 1918 regulations (b)
IRF (1938)	Nossa Senhora do Rosário de Fátima Church	1934–1938	RCB	1938	Porfírio Pardal Monteiro	Reinforced concrete structure with brick masonry panes	Modernist architecture religious building. Ogival arches are built in a centered plan.	Indoor mural decoration and white marble coatings. Limestone-coatings in exterior walls	According to 1935 regulations (c)
CBP (1939)	Bernardo da Maia House	1938–1939	PRCB	1939	Carlos and Guilherme Rebelo de Andrade	Self-supporting masonry walls. Reinforced concrete structure in the foundations and basement floor	*Joanino* Baroque-inspired like architectural style Dwelling and office building with two floors, basement, and attic.	Exterior limestone coatings at street level; rendering mortars	According to 1935 regulations (c)
DN (1940)	Diário de Notícias Building	1936–1940	RCB	1940	Porfírio Pardal Monteiro	Reinforced concrete structure with brick masonry panes	Modernist architecture building constructed to house a newsroom and a typography with six upper floors and basement	Most of the main facade area is covered with limestone; ceramic tiles and rendering mortars	According to 1935 regulations (c)
AAC (1944)	Cristino da Silva Building	1942–1944	PRCB	1944	Luís Cristino da Silva	Mixed concrete-masonry structure (concrete slabs and self-supporting masonry walls)	Multifamily building designed with nationalist tendency and composed by three dwelling floors and terrace	Rendering mortars and rock imitating mortars; limestone coatings	According to 1935 regulations (c)
LIP (1958)	Laboratories of Pasteur Institute of Lisbon	1955–1957	RCB	1958	Carlos Manuel Oliveira Ramos	Reinforced concrete structure with brick masonry panes	Modern architecture building for industrial purposes. All the building walls have the functions of dividing the spaces, insulation, and thermal protection	Glazed partitions. Rendering mortars and limestone coatings	According to 1935 regulations (c)
EUA53 (1970)	América Building	1966–1969	RCB	1970	Leonardo Rey Colaço de Castro Freire	Reinforced concrete structure with brick masonry panes	15th floor dwelling and commerce building inserted in residential area	Rendering mortars, limestone coatings, ceramic tiles, rock imitating mortars	B300 [compressive strength = 300 kg/cm^2] (d)
FRAN (1971)	Franjinhas Building	1965–1969	RCB	1971	Nuno Teotónio Pereira; João Braula Reis	Reinforced concrete structure	Building designed for trade and services. Facades in architectural concrete	Concrete precast exterior panels	B300—superstructure [compressive strength = 300 kg/cm^2] (d)

Table 1. *Cont.*

Case Study	Name	Construction Period	Construction Typology	Award Year	Architect	Structural Characteristics	Main Constructive and Architectonical Features	Main Coatings/ Renderings	Class of Concrete Prescribed in the Design Project
FCG (1975)	Calouste Gulbenkian Foundation Headquarters and Museum	1963–1969	RCB	1975	Ruy Athouguia, Alberto Pessoa, Pedro Cid, G. Ribeiro Teles and António Barreto	Reinforced concrete structure	Modernist building's complex, consisting essentially of architectural concrete	Granite coatings	B225—foundations [compressive strength = 225 kg/cm^2]; B300—superstructure [compressive strength = 300 kg/cm^2] (d)
ISCJ (1975)	*Sagrado Coração de Jesus* Church	1966–1970	RCB	1975	Nuno Teotónio Pereira; Nuno Portas: Pedro Almeida; Luís Vassalo	Reinforced concrete structure	Modernist, religious architecture developed on several levels due to the irregularity of the site, featuring architectural concrete	Concrete-based precast exterior panels with visible limestone and marble aggregates	B300 [compressive strength = 300 kg/cm^2] (d)
JRP (1987)	*Jacob Rodrigues Pereira* Institute	1984–1987	RCB	1987	Rui de Sousa Cardim	Reinforced concrete structure	Set of four modernist buildings with several areas in architectural concrete	Rendering mortars and limestone coatings onto concrete buttresses	B225 [compressive strength = 225 kg/cm^2] (e)
PCV (1998)	The Knowledge Pavilion	1996–1998	RCB	1998	João Luís Carrilho da Graça	Reinforced concrete structure	Composed of two bodies of distinct volumetry. Both built in white architectural concrete	Limestone coatings and iron elements	B35 [compressive strength = 35 MPa]—white concrete (f)
C8 (2000)	C8 Building (Faculty of Sciences of the University of Lisbon)	1997–2000	RCB	2000	Gonçalo Byrne	Reinforced concrete structure	Architectural concrete facades	Precast concrete panels applied onto external facades and limestone coatings	B25 [compressive strength = 25 MPa] - Foundations and earth supporting walls; B30 [compressive strength = 30 MPa]-superstructure (f)
AS (2001)	*Atrium Saldanha* Building	1992–1997	RCB	2001	João Paciência and Ricardo Bofill	Reinforced concrete structure	Vertical elements consisting of cylindrical white architectural concrete pillars. Architectural concrete in the underground floors	White and gray architectural concrete, glass and marble coatings	B30 [compressive strength = 30 MPa]; B40 [compressive strength = 40 MPa]—architectural concrete (f)
UNL (2002)	New University of Lisbon Rectory	2000–2002	RCB	2002	Manuel and Francisco Aires Mateus	Reinforced concrete structure	Composed of two bodies of distinct volumetry, featuring architectural concrete in the underground floors	External limestone cladding all over the building; glass	B25 [compressive strength = 25 MPa] Foundations and earth supporting walls; B30 [compressive strength] - superstructure (f)

(a) Not applied; (b) minimum compressive strength at 28 days: 120 kg/cm^2 [25]; (c) minimum compressive strength at 28 days: 180 kg/cm^2 [26]; (d) according to 1967 regulations [27]; (e) according to 1971 regulations [28]; (f) according to 1989 regulations [29].

3. Methodology

3.1. Visual Inspection, Sampling, and In Situ Testing

A visual inspection was carried out, mainly on the building's envelope and in the indoor spaces whenever access was allowed. Two sets of materials were grouped independently. The first one included renders and plasters and the second one included concrete. This survey was carried out to achieve the following:

- Identify the main macroscopic characteristics of buildings' renderings and plasters, namely thickness, and the number of layers, and measure the cover thickness and the carbonation depth in concrete samples.
- Assess the state of conservation of buildings' renderings, plasters, and visible concrete surfaces.

Visual survey for building anomaly assessment has been widely reported in the literature (e.g., [30–32]), especially those that combines visual survey with inquiries to tenants or owner's representatives and visits to their apartments [33].

The inspection performed in this work comprised visual survey of the external building envelopes and whenever possible visits to apartments or service areas on different floor levels as well as in the common areas.

Sampling was carried out in places that do not compromise the building's safety or aesthetics [34].

Although the results of the materials characterisation are not presented in this document, it is important to mention that in order to assess the original composition of the mortars and concretes and to support conservation and rehabilitation actions, samples were taken to study their physical, mechanical, chemical, and mineralogical properties. The most suitable substitution materials can only be developed after a full characterisation of the original materials according to the following test methodology [21,35–40]:

- Mineralogical analysis by X-ray diffraction (XRD) for phase identification of the binder and the aggregates, complemented with simultaneous thermogravimetry and differential thermal analysis (TG/DTA) to confirm XRD results and to estimate the proportion of some compounds.
- Optical microscopy to identify pozzolanic additives and neoformation products, and the type of the binder since the binder-related particles and raw material remnants can be identified by petrographic observations.
- Microstructural observations with scanning electron microscope equipped with X-ray microanalysis (SEM-EDS) will provide additional information on the morphology and chemical composition of the mortar and concrete constituents.
- Wet chemical analysis to separate the soluble from the insoluble fraction (siliceous sand). The insoluble fraction is also used to obtain the grain size distribution of the sand and to estimate the binder/aggregate ratio. If carbonate aggregate is present, the petrographic point-counting technique will be used to estimate its proportion. Regarding the soluble fraction, atomic absorption spectroscopy (AAS) may be carried out to obtain the content of soluble salts.
- Physical and mechanical tests will be accomplished, and the data extracted for the formulation of a compatible material similar to the pre-existing one must ensure a satisfactory performance. Test methods should at least include capillary water absorption, drying, compressive strength, open porosity, and ultrasonic pulse velocity.

Secondly, non-destructive in situ tests were performed as a complement to the visual inspection. These tests were performed on a non-systematic basis, since several wall zones were not accessible. Then, measurements of moisture content, superficial hardness, and mechanical strength of wall renders and plasters [16] as well as of concrete surfaces were carried out.

In the moisture evaluation [41,42] of renders and plasters, a portable hygrometer was used. This technique is based on the variation of the electrical resistance of the materials according to the respective water content. It is not an absolute evaluation as it assesses a related moisture content and only at the exposed surface. Four moisture content classes have been considered according to the device reference values: dry zones have moisture values below 2% and moderate wet zones vary between 2 and 4%; a range between 4 and 6% is considered for wet zones; and the values between 6 and 6.9% are for very wet zones.

Surface hardness for rendering mortars and plasters was carried out with a shore A durometer, whose procedure was based on ASTM standard D2240-05 [43]. A pendulum sclerometer was used to assess the mechanical strength of rendering mortars and plasters, and the procedure was based and adapted from ASTM C805 standard [44].

Moisture, surface hardness, and mechanical strength were evaluated every 0.3 m on a vertical masonry wall profile, as shown in Figure 4a, with a maximum height of 3 m.

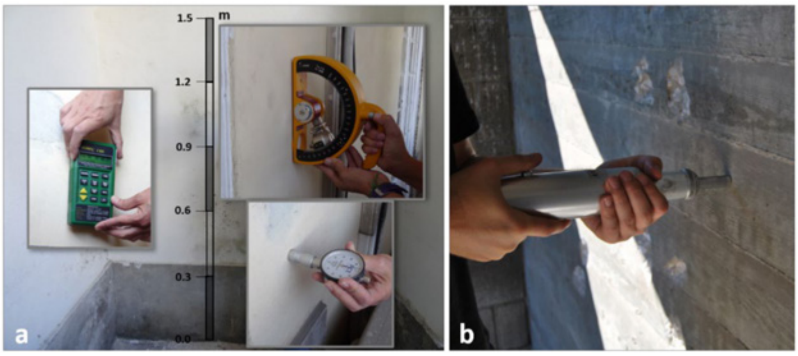


Figure 4. In situ tests. (**a**) Example of a vertical profile to assess moisture, surface hardness, and mechanical strength of renders in a wall. Portable hygrometer, middle left; pendulum sclerometer, top right; shore durometer, bottom right; (**b**) Rebound Schmidt hammer test on a concrete surface.

Table 2 refers to a qualitative classification based on old render studies for hardness and mechanical strength [45] that was adopted in this work.

Table 2. Qualitative classification for hardness and mechanical strength according to Tavares (2009) [45].

Shore A Durometer	Hardness Classification	Rebound (Vickers Degrees)	Mechanical Strength Classification
<30	very weak	<20	very weak
30–50	weak	20–30	weak
51–70	moderate	31–40	moderate
71–87	normal	41–55	normal
88–100	very hard	56–75	hard
		>75	very hard

For concrete surfaces, mechanical tests were carried out using a rebound Schmidt hammer [46] to assess the concrete quality and uniformity.

Concrete cores from architectural and non-architectural concrete (NAC) were extracted in pillars and walls. The measure of reinforcement concrete covering thickness on the outermost rebars using a proper detector was mainly performed at the sampling zones. Carbonation depth was measured directly in cores after sampling by applying a phenolphthalein solution [47].

3.2. Anomalies' Survey

Building materials applied as coatings and/or surfaces are the most susceptible to deterioration [48]. Amongst other degradation causes, salts crystallisation (efflorescences and cryptoflorescences), pollution, water, and biological activity, structural defor-mations, and dimensional variations due to shrinkage or thermal actions can be recognised [49]. External agents, application technology, mixture composition, aggregates and binder type, porous structure, water to binder ratio, and curing period are among the factors that may influence mortars and concrete performance [50].

Cement-based materials, such as concrete and mortar, are subjected to physical, mechanical, and chemical deterioration mechanisms [51]. Physical degradation can be caused by freeze–thaw, thermal effects, salt crystallisation, shrinkage, and erosion. Mechanical deterioration can be caused by impact, overload, movement, explosion, and vibration [52]. Chemical mechanisms can affect both mortars and concrete materials through the same processes, such as sulfate attack, alkali-aggregate reaction, and acid attack. Alkali-aggregate reaction and internal sulfate reaction, both included in the internal expansive chemical reactions, are known to be the cause of expansion, cracking, spalling, loss of strength, and adhesion [51].

The corrosion of steel rebars in reinforced concrete is the major degradation factor of concrete structures and may be induced by carbonation or chloride penetration. Certain exposition environments enhance both processes. Those mechanisms lead to the depassivation of steel rebars that start to be corroded with the production of expansive rust [53]. It generates stresses that cause the cracking and spalling of the concrete, compromising the structures' safety. Corrosion is furtherly dependent on the cover thickness of concrete, binder composition, and moisture content.

The technical restrictions and the lack of authorisation to access all the buildings' areas of all the case studies had conditioned the visual inspection, the anomalies survey, and sampling. For that reason, renders and plasters from some case studies were not inspected. The surveying period was 2 years long, from 2016 to 2018, and was carried out on a non-periodic basis.

Table 3 presents a description of anomalies appended to the main deterioration processes for both renders/plasters and concrete materials. Since many reinforced concrete structure buildings have uncoated surfaces, a subdivision of anomalies' types based on French standard NF P18-503 [54] and CIB report no. 24 [55] is proposed for architectural concrete surfaces, in which three main groups are identified: (1) shape anomalies, includes defects affecting the major geometrical aspects of the concrete element, (2) texture anomalies, including defects that with minor or no concerns to the overall geometry but with impact on surface characteristics and (3) colouration anomalies, taking into account mainly visual anomalies and unexpected heterogeneities.

Table 3. Types, causes, and groups of anomalies related to renders, plasters, and architectural concrete surfaces.

	Renders/Plasters			Architectural Concrete Surfaces		
Causes/Group of Anomalies	**Physical**	**Mechanical**	**Chemical and Biological**	**Shape**	**Texture**	**Colouration**
Types of anomalies	Wear/erosion	Disaggregation	Stains	Wear/erosion	Bug holes	Dirt stains
	Capillary rising water; water retention or infiltration	Detaching	Biological growth	Flatness defects	Mapped cracking	Moisture stains
	Water condensation	Cracking	Cracking	Disaggregation	Dribbling	Corrosion stains
	Efflorescence		Spalling	Spalling	Fastening marks	Biological growth
			Disaggregation	Oriented cracking	Honeycombing	Efflorescence
			Efflorescence	Crusts		
				Formwork incrustation		

3.3. State of Conservation Classification

To assign a state of conservation rating for renders and plasters, a classification based on the concept of severity of the anomalies was adapted from Veiga and Aguiar (2003) [56]. This classification includes the extension of degradation and its severity. Degree and extension of degradation are classified as low (+), medium (++), and high (+++) according to its global persistence in the audited building. Severity is divided into four degrees according to repairability: (1) maintenance and conservation needed; (2) consolidation or localised repair needed; (3) filling gaps needed; and (4) partial substitution needed. Table 4 shows the matrix we propose with a colour code related to the state of conservation evaluation for renders and plasters. The same methodology was applied for architectural concrete surfaces.

Table 4. Matrix used for state of conservation classification.

		Degree and Extension of Degradation		
		+	++	+++
Severity of degradation	1	Good	Reasonable	Reasonable
	2	Reasonable	Reasonable	Poor
	3	Reasonable	Poor	Very Poor
	4	Reasonable	Very Poor	Very Poor

3.4. Age, Materials, and Degradation Relationship over Time

As previously stated, the quality of construction and materials may also influence the deterioration process and the local weather conditions and poor maintenance. The degradation of the exterior surfaces of buildings is a major cause of renovation actions. Studies concerning service life prediction [57–59] have been made to prevent future damage and avoid ruin. A study conducted in several European countries concerning the deterioration of apartment buildings [60], in respect to façade rendering, concluded that approximately 60 years is the average time of service life for the materials, i.e., until deteriorated materials must be replaced, considering a distribution range of 100 years of case studies during the last century. However, it may vary in different countries.

4. Results

In situ observations revealed renders consisting of up to four layers. The maximum thickness per layer is variable reaching up to 50 mm.

Plasters applied until the 1960s normally consist of more than one layer. Thicknesses vary between 5 mm (in outer layers) and 40 mm.

Indoor finishing layers are generally white, probably lime-based, with thicknesses varying between 2 and 5 mm, and stone imitation mortars designated in Portuguese as "marmorite" (meaning similar to marble) with siliceous rolled pebbles or limestone aggregates [61] with thicknesses ranging between 5 and 10 mm. In rendering finishing layers, stone imitation mortars are also present with visible marble aggregates, with thicknesses varying between 5 and 8 mm. Table 5 presents plasters and rendering mortars stratigraphy observed during visual inspection.

Table 5. Stratigraphy of plasters and renders.

Case Study	Construction Period	Plasters	Renders
CVT (1903)	1902–1903	2 to 3 layers (includes finishing white layer)	(a)
CMAG (1905)	1904–1905	(a)	1 to 4 layers: non-original renders (b)
AR49 (1923)	1920–1923	3 layers (includes finishing white layer)	2 layers
IRF (1938)	1934–1938	1 to 2 layers (includes finishing white layer)	(a)
CBP (1939)	1938–1939	2 layers (includes finishing white layer)	(a)
DN (1940)	1936–1940	2 (c) to 4 layers (includes finishing white layer)	(a)
AAC (1944)	1942–1944	2 layers (includes finishing in stone imitation mortar)	2 to 3 layers (includes finishing in stone imitation mortar)
LIP (1958)	1955–1957	(a)	1 layer
EUA53 (1970)	1966–1969	2 layers (includes finishing in stone imitation mortar)	2 layers (includes finishing in stone imitation mortar)
FRAN (1971)	1965–1969	(a)	(a)
FCG (1975)	1963–1969	1 layer	(a)
ISCJ (1975)	1966–1970	(a)	(a)
JRP (1987)	1984–1987	(a)	1 layer
PCV (1998)	1996–1998	(a)	(a)
C8 (2000)	1997–2000	(a)	(a)
AS (2001)	1992–1997	(a)	(a)
UNL (2002)	2000–2002	1 layer	(a)

(a) Not analysed; (b) Non-original renders applied ca. 1980?; (c) Some plasters were applied ca.1998.

Regarding reinforced concrete, the evolution of the performance requirements led to a general increase in compressive strength set due to the complexity of built structures. There has been a progressive increase in the minimum compressive strength requirements over time since the first Portuguese regulation decreed in 1918 [25]. Even though the consulted documents of the buildings' design projects up to the 1960s do not mention values for concrete strength, we considered that for the awarded buildings, they must have been applied under the regulations.

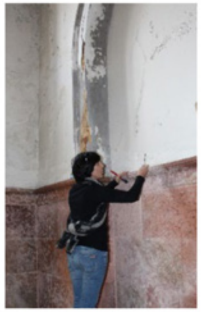
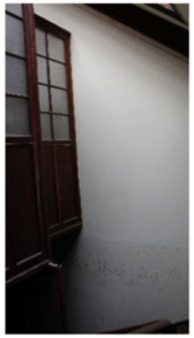
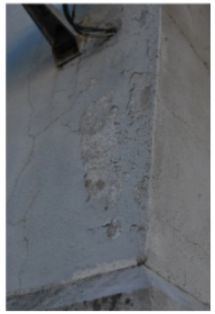
4.1. Diagnosis and State of Conservation Assessment

4.1.1. Renders and Plasters

Wall renders and plasters were mainly surveyed in PRCBs case studies, with few cases in RCBs. Table 6 summarize the main types of anomalies detected by visual inspection, while Table 7 summarizes the results of in situ tests.

Table 6. Examples of main types of anomalies detected in renders and plasters of the cases studies.

a. Examples of main types of anomalies detected in PRCBs' renders and plasters by location.

Case Study	**CVT (1903)**			**CMAG (1905)**		
Zone ID Location	1 Basement	2 Hall entrance	3 Main stairs/roof	1 SE facade	2 NW facade	3 NW facade
Image						
Detected anomalies	Capillary rising water; detaching	Water infiltrations; detaching; efflorescence	Water infiltrations	Cracking	Cracking; biological growth	Detaching; stains; biological growth

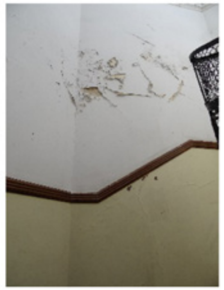
Case Study	**AR49 (1923)**				
Zone ID Location	1 W rear facade. Ground floor level (exterior)	2 E façade. 1st floor	3 Interior stairs. 1st/2nd floor	4 Interior stairs. 4th/5th floor	5 Interior stairs. 5th/6th floor
Image					
Detected anomalies	Capillary rising water	Cracking, stains	Water infiltrations; cracking; detaching and efflorescence		

Table 6. *Cont.*

a. Examples of main types of anomalies detected in PRCBs' renders and plasters by location.				
Case Study	**CBP (1939)**			**AAC (1944)**
Zone ID Location	1 Basement. NE access	2 Basement. Intermediate room	3 Basement. NW access	1 Access to the rear outer space
Image				
Detected anomalies	Capillary rising water	Capillary rising water; cracking;cryptoflorescences; detaching	Capillary rising water; cracking; efflorescence	Stains, biological growth
Case Study	**AAC (1944)**			
Zone ID Location	2 Belvedere	3 Boiler room. Ground floor	4 Access stairs to the terrace	5 W facade. 2nd floor
Image				
Detected anomalies	Erosion; capillary rising water; stains and biological growth	Stains	Cracking; stains	Disaggregation

Table 6. *Cont.*

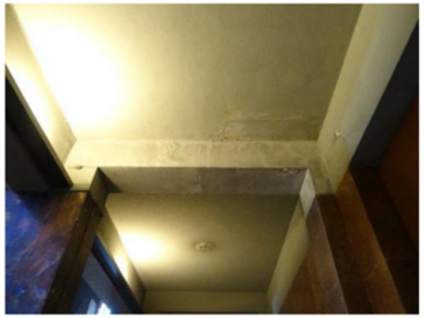
b. Examples of main types of anomalies detected in RCBs' renders and plasters by location				
Case Study	**IRF (1938)**			**DN (1940)**
Zone ID	1	2	3	1
Location	N.S. Piedade Chapel. Upper wall	Right upper gallery	Left-side gallery	SE facade
Image				
Detected anomalies	Water infiltration; cracking and detachment	Cracking; detaching	Water infiltration; cracking	Stains, biological growth; detaching
Case Study	**DN (1940)**	**LIP (1958)**		**EUA53 (1970)**
Zone ID	2	1	2	1
Location	NE facade	Roof's chimney	West side 4th floor bathroom.	Service room corridor. Ground floor
Image				
Detected anomalies	Water retention; stains; biological growth; detaching	Cracking; detaching	Water infiltration; stains; biological growth; detaching	Cracking

Table 7. Results of in situ tests performed in the zones reported in Table 6.

Case Study	**CVT (1903)**		**CMAG (1905)**	**AR49 (1923)**		**CBP (1939)**			**AAC (1944)**			**EUA53 (1970)**
Construction typology	PRCB		PRCB	PRCB		PRCB			PRCB			RCB
Zone ID	1	2	1	1	5	1	2	3	1	2	3	1
						MC						
m	0.0 (>0.9 m)	2.3 (at 1.5 m; 2.1 m)	1.1 (at 2.7 m)	3.9 (at 1.8 m)	4.8 (at 0.3 m)	3.0(at 1.5 m)	1.0 (at 1.8 m)	1.6 (at 1.2 m)	3.4 (at 0.3 m)	3.3 (at 0.9 m)	0.6 (at 1.8 m)	0.0 (>1.5 m)
M	4.1 (at 0.3 m)	4.5 (at 0.6 m)	4.7 (at 0.3 m)	6.9 (at 0.6 m)	5.1 (at 2.1 m)	6.5 (at 0.6 m)	5.3 (at 0.3 m)	4.7 (at 0.6 m)	5.1 (at 1.2 m)	3.7 (at 0.6 m)	2.0 (at 0.3 m)	0.9 (at 0.3 m)
						MS						
m	12 (at 0.3 m)	(a)	36 (at 1.5 m)	(a)	(a)	(a)	(a)	(a)	35 (at 1.2 m)	23 (at 0.6 m)	71 (at 0.9 m)	96 (at 1.5 m)
M	26 (at 0.9 m)	(a)	38 (at 1.2 m)	(a)	(a)	(a)	(a)	(a)	48 (at 0.3 m)	50 (at 0.9 m)	92 (at 0.3 m)	106 (at 1.2 m)
						SH						
m	97 (at 0.3 m)	20 (at 2.1 m)	94 (at 0.9 m; 2.7 m)	(a)	(a)	(a)	(a)	(a)	(a)	(a)	98 (at 0.6 m)	(a)
M	99 (>0.9 m)	100 (at 0.6 m)	97 (at 1.8 m)	(a)	(a)	(a)	(a)	(a)	(a)	(a)	100 (at 0.9 m)	(a)

MC—moisture content (%); MS—mechanical strength (Vickers degrees); SH—surface hardness (Shore A degrees); m—minimum value; M—maximum value; >—above; (a) not available.

The assessment of rendering mortars and plasters shows the prevalence of water as the main degradation agent. As shown in Table 7 and in Figure 5, in situ tests to evaluate the moisture content revealed "wet" to "very wet" zones for all PRCBs assessed. Infiltration, capillary rising water, and moisture stains are the main anomalies related to water and are present in almost all PRCBs evaluated, which is in accordance with the main problems affecting old masonry structures (e.g., [50,62–64]). The water-related anomalies in buildings CVT (1903), AR49 (1923), CBP (1939), and AAC (1944), point out different sources of water. Capillary rising water from the soil and underground through foundations and walls caused loss of adhesion to the substrate that in some cases led to detachment. Apart from building CBP (1939), where none of the following was observed, runoff and infiltration of rainwater caused erosion, loss of cohesion, stains, and biological growth. In both PRCBs and RCBs, localised infiltrations were observed due to the absence or inability of adequate drainage systems or to plumbing defects in interior walls. Cracking was also observed, associated with shrinkage or thermal cycles, which induce internal stresses compromising aesthetic and protective purposes [65,66]. In rare assessed cases, non-oriented cracks (<0.2 mm opening) can be associated with the loss of elasticity of the coating paintings.

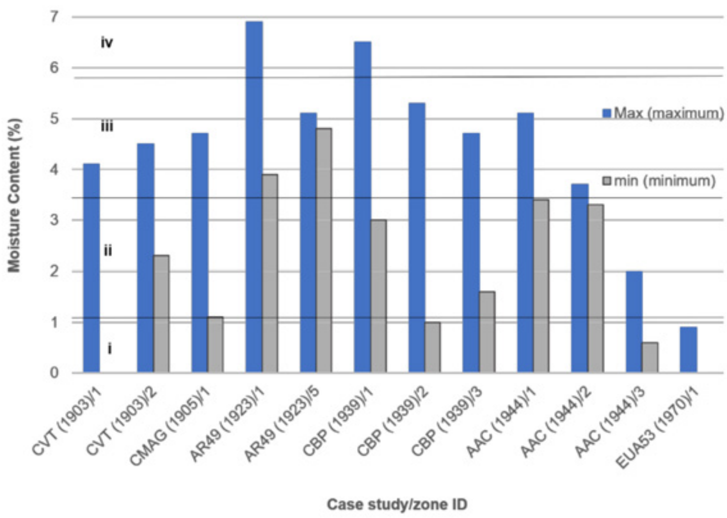


Figure 5. Moisture content (%) by tested zones. Dry zone (**i**); moderate wet zone (**ii**); wet zone (**iii**) and very wet zone (**iv**).

Mechanical strength values obtained by in situ tests in the assessed zones enable us to classify them in the range of very weak to moderate [45] and confirm the existence of degradation associated with the moisture increase, with the lowest values corresponding to the zones where the maximum moisture was registered. However, this relationship was not confirmed for surface hardness, as it may be observed from Table 7.

Table 8 summarises the state of conservation of the renders and plasters according to the proposed classification.

Table 8. State of conservation classification of renders and plasters in Lisbon's awarded buildings (colour code according to Table 4).

Case Study	Construction Typology	Anomaly Type	Mechanism/Cause	Extension and Degree of Degradation	Severity	State of Conservation
CVT (1903)	PRCB	Water infiltrations	Physical	++	1	Reasonable condition
		Capillary rising water		+	1	
		Detaching	Mechanical	+	4	
		Efflorescences	Chemical/Physical	+	1	
CMAG (1905)	PRCB	Cracking	Mechanical	++	2	Reasonable condition
		Detaching		+	4	
		Stains	Chemical	+	1	
		Biological growth		+	1	
AR49 (1923)	PRCB	Capillary rising water	Physical	+	1	Reasonable condition
		Water infiltrations/retention		++	1	
		Cracking	Mechanical	++	3	
		Detaching		+	4	
		Stains	Chemical	+	1	
		Efflorescences	Chemical/Physical	+	1	
CBP (1939)	PRCB	Capillary rising water	Physical	+++	1	Reasonable condition
		Cracking	Mechanical	++	2	
		Detaching		+	4	
		Cryptoflorescences	Chemical/Physical	+	1	
AAC (1944)	PRCB	Capillary rising water	Physical	+	1	Reasonable condition
		Erosion		+	1	
		Disaggregation	Mechanical	+	4	
		Stains	Chemical	++	1	
		Biological growth		++	1	
IRF (1938)	RCB	Water infiltration	Physical	++	1	Reasonable condition
		Cracking	Mechanical	+	2	
		Detaching		+	4	
DN (1940)	RCB	Water retention	Physical	+	1	Reasonable condition
		Detaching	Mechanical	+	1	
		Stains	Chemical	++	1	
		Biological growth		+	4	
LIP (1958)	RCB	Water infiltration	Physical	+	1	Reasonable condition
		Cracking	Mechanical	+	1	
		Detaching		+	4	
		Stains	Chemical	+	1	
		Biological growth		+	1	
EUA53 (1970)	RCB	Cracking	Mechanical	+	1	Good condition

4.1.2. Reinforced Concrete

Spalling was the main anomaly found in the architectural concrete surfaces of RCBs, namely FRAN (1971), ISCJ (1975), JRP (1987), and occasionally in PCV (1998). Building FCG (1975) was not inspected. However, another study [67] refers that it is in a good state of conservation. Anomalies resulting from the patch repairs of spalling and cracking are the only type of defects pointed out. In newer buildings, respectively C8 (2000), AS (2001), and UNL (2002), the detected anomalies are essentially related to the presence of moisture and dirt stains, in which the latter is a sign of ongoing corrosion.

Table 9 shows examples of the main types of anomalies detected, while Tables 10 and 11 show the main characteristics measured during the survey in sampling zones, namely compressive strength, concrete cover thickness, and concrete carbonation depth. The last two were also measured in NAC.

The spalling phenomenon observed is related to a low concrete covering thickness, which provided feeble protection for the reinforcement to corrosion by carbonation.

Based on the experimental data obtained (Tables 10 and 11), there is not a clear relationship between the average reinforced concrete covering thicknesses and the age of buildings, neither with the carbonation depth, reflecting the different concrete's quality. It is also observed that the minimum covering thickness of 20 mm proposed in Portuguese regulations [28,29] was not respected in cases LIP (1958) and FRAN (1971).

Figures 6 and 7 show that the average covering thickness is greater than the average carbonation depth in sampling zones. However, several values of maximum depth of carbonation are higher than the recorded minimum cover, for the same building, indicating a high probability of corrosion.

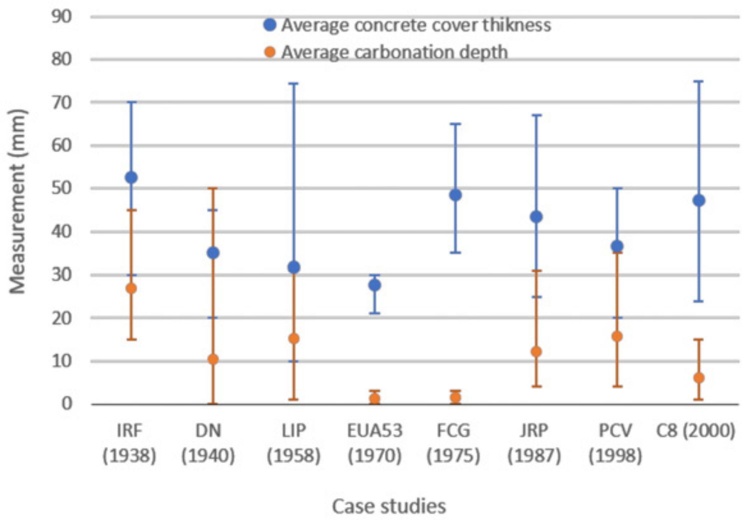


Figure 6. Relationship between concrete cover thickness and the carbonation depth in NAC sampling zones. Range of results: minimum to maximum.

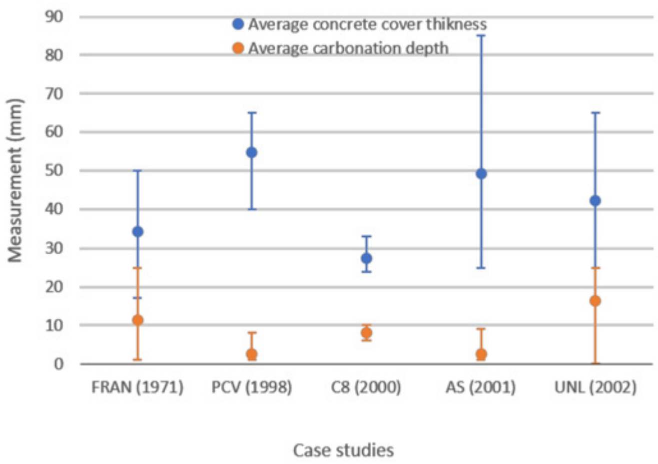


Figure 7. Relationship between concrete cover thickness and the carbonation depth in architectural concrete sampling zones. Range of results: minimum to maximum.

Table 9. Examples of main types of anomalies detected in architectural concrete surfaces of RCBs.

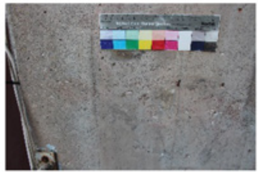
Case Study	**FRAN (1971)**			**ISCJ (1975)**		
Location	E facade. Rear windows' precast panels	E facade	Terrace over the gallery (1st floor)	3rd floor staircase level	6th floor. Entrance door	Nave's ceiling
Image						
Detected anomalies	Biological growth; spalling	Erosion	Spalling	Stains; biological growth; spalling	Spalling	Moisture and corrosion stains
Case Study	**JRP (1987)**					
Location	Indoor garden	Children's day care building	Exterior passage between buildings	Indoor garden	W side building	
Image						
Detected anomalies	Honeycombing; stains	Flatness defects; efflorescences	Oriented cracking (>3mm); spalling; corrosion stains, biological growth	Spalling	Stains; oriented cracking (<0.5 mm)	

Table 9. *Cont.*

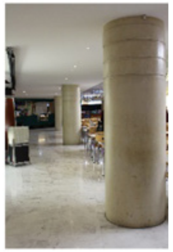
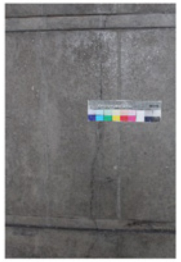
Case Study			**PCV (1998)**				C8 (2000)
Location	E facade	E facade		W facade	N facade	N high block facade	S facade. 7th level
Image							
Detected anomalies	Mapped cracking; bug holes	Oriented cracking (<0.5 mm)		Flatness defects	Crust; corrosion stains	Spalling; flatness defects; dirt stains	Moisture and dirt stains
Case Study		**C8 (2000)**			**AS (2001)**		**UNL (2002)**
Location	N facade	W facade	E facade	Restaurant area, level 0	Car parking, level −3	Car parking, level −4	Technical area, level −1
Image							
Detected anomalies	Moisture and dirt stains		Dribbling; Moisture stains	Wear	Oriented cracking (<0.5 mm)	Dribbling; efflorescences	Fastening marks

Table 10. Summary of the main characteristics of the NAC in sampling zones.

Case Study	IRF (1938)	DN (1940)	LIP (1958)	EUA53 (1970)	FCG (1975)	JRP (1987)	PCV (1998)	C8 (2000)
Age (years)	83	81	64	52	52	34	23	21
Concrete cover thickness (mm)								
min	30.0	20.0	10.0	21.0	35.0	25.0	20.0	24.0
max	70.0	45.0	74.3	30.0	65.0	67.0	50.0	75.0
average	52.5	35.0	31.7	27.7	48.6	43.3	36.5	47.3
Carbonation depth (mm)								
min	15.0	0.0	1.0	0.0	0.0	4.0	4.0	1.0
max	45.0	50.0	31.0	3.0	3.0	31.0	35.0	15.0
average	26.9	10.5	15.3	1.2	1.5	12.2	15.8	6.1

Table 11. Summary of the main characteristics of the architectural concrete in sampling zones.

Case Study	FRAN (1971)	ISCJ (1975)	PCV (1998)	C8 (2000)	AS (2001)	UNL (2002)
Age (years)	52	51	23	21	24	19
Compressive strength (MPa)						
min	32.0	33.0	45.0	40.0	45.0	35.0
max	54.0	42.0	53.0	54.0	54.0	46.0
median	47.0	39.0	49.0	50.0	50.0	42.0
Concrete cover thickness (mm)						
min	17.0	(a)	40.0	24.0	25.0	25.0
max	50.0	(a)	65.0	33.0	85.0	65.0
average	34.1	(a)	54.8	27.3	49.2	42.3
Carbonation depth (mm)						
min	1.0	3.0	1.0	6.0	1.0	0.0
max	25.0	20.0	8.0	10.0	9.0	25.0
average	11.4	10.7	2.5	8.2	2.6	16.3

(a) not measured.

To assess the quality of the concrete and to verify its uniformity throughout the building, surface rigidity tests by Schmidt rebound hammer were carried out. However, it should be noted that the mechanical strength measured by this test may be influenced by the concrete surfaces' conditions, such as carbonation, temperature, degree of saturation, location, and surface texture. The assessment carried out showed FRAN (1971) as the building with the greatest dispersion of results (Figure 8). Case studies PCV (1998), C8 (2000), and AS (2001) have a distribution to approach a central value of 50 MPa.

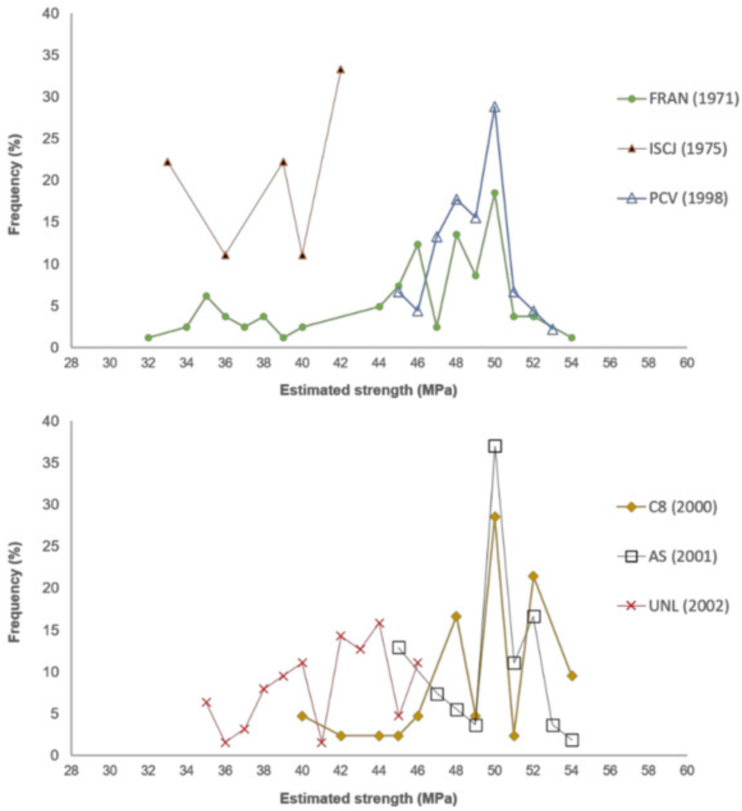


Figure 8. Frequency results of Schmidt rebound tests on architectural concrete surfaces.

As shown in Figures 6 and 7, the carbonation depth range does not vary clearly with the building's age. This can be due to different carbonation rates related to the location of exposed structures, inherited concrete properties (e.g., binder type, porosity, microstructure, moisture), and the existence of coatings or paintings applied on the concrete surfaces, which seems to be frequent in the assessed buildings.

Table 12 summarises the state of conservation of architectural concrete according to the proposed classification.

Table 12. State of conservation classification for architectural concrete of RCBs (colour code according to Table 4).

Case Study	Anomaly Type	Group of Anomalies	Extension and Degree of Degradation	Severity	State of Conservation
FRAN (1971)	Biological growth	Colouration	+	1	Reasonable condition
	Erosion	Shape	+	1	
	Spalling		+	2	
ISCJ (1975)	Moisture stains	Colouration	++	2	Reasonable condition
	Corrosion stains		+	1	
	Biological growth		+	1	
	Spalling	Shape	+	2	
JRP (1987)	Moisture stains	Colouration	++	2	Reasonable condition
	Corrosion stains		+	1	
	Efflorescences		+	2	
	Biological growth		+	1	
	Oriented cracking (<0.5 mm)	Shape	+	2	
	Oriented cracking (>3mm)		+	3	
	Spalling		+	2	
	Flatness defects		+	2	
	Honeycombing	Texture	+	2	
PCV (1998)	Corrosion stains	Colouration	+	1	Reasonable condition
	Dirt stains		+	1	
	Bug holes	Texture	+	1	
	Mapped cracking		+	3	
	Oriented cracking (<0.5 mm)	Shape	+	2	
	Spalling		+	2	
	Flatness defects		+	1	
	Crust		+	2	
C8 (2000)	Moisture stains	Colouration	++	2	Reasonable condition
	Dirt stains		++	2	
	Dribbling	Texture	+	1	
AS (2001)	Efflorescences	Colouration	+	1	Reasonable condition
	Wear	Shape	+	1	
	Oriented cracking (<0.5 mm)		+	2	
	Dribbling	Texture	+	1	
UNL (2002)	Corrosion stains	Colouration	+	1	Reasonable condition
	Fastening marks	Texture	+	2	

5. Discussion

5.1. Renders and Plasters

Renders and plasters thickness and number of layers based on the macroscopic observation were identified during sampling work. Regarding the binder, it can be assumed by the constructive elements consulted, together with visual observation, that mortars are probably aerial lime-based at least until the 1920s in PRCBs.

The state of conservation of existing renders and plasters has been characterised by the types of anomalies and their extension. To assess the state of conservation, a diagnosis based on visual inspection was performed to identify the causes of degradation and their extent. In some cases, complementary in situ tests were carried out. The combined analysis provided the following data:

1. The physical degradation mechanisms due to the water action are the main causes of anomalies found in PRCBs. Though in RCBs, those mechanisms contribute to degradation with a lower prevalence, as seen by the extension of degradation, which is usually lower in comparison to PRCBs;
2. The loss of adhesion and cohesion, which lead respectively to detaching and disaggregation, are the most serious anomalies found in PRCBs. The migration and crystallisation of soluble salts (efflorescences and cryptoflorescences) are sometimes found in the walls due to capillary rising water from the underground on the lower building floors. The inefficient connections between elements (e.g., roofs/eaves; terraces/walls) aggravate the infiltrations, as well as the deficient channeling of water off the buildings;
3. Stains are frequently found in external facades of PRCBs and RCBs due to moisture, dirt, and biological action, as a result of the environmental exposure;
4. Cracking is also present, mainly as a result of shrinkage and water action in the early stages of detaching process. Cracking can also be associated with the presence of salts and corrosion of reinforced concrete elements;
5. A decrease of about 54% in mechanical strength is observed, considering the maximum values as reference (Table 7) in PRCBs surveyed zones, namely in CVT (1903)/1 and AAC (1944)/2. In both cases, this decrease seems to be associated with the observed rising water (Table 6a);
6. The surface hardness results do not corroborate in general the degradation observed, which is probably due to the multilayer system found in the tested wall coverings;
7. No relationship between the age and the state of conservation was found, since the studied buildings have, in general, a reasonable state of conservation. However, in comparison to RCBs, PRCBs' renders and plasters show a higher degree and extension of degradation, including severity as well, which is mainly related to water action as already mentioned;
8. Despite the anomalies surveyed, their degree and extension in both types of buildings are not persistent nor generalised. This condition, despite the age of the buildings, may reflect the good selection of materials and careful construction, as it could be a characteristic of the awarded buildings. In addition, more care with maintenance than in the case of common buildings of the same period would have been beneficial.

5.2. Reinforced Concrete

Considering architectural concrete surfaces, visual inspections performed along with the in situ tests produced the following results:

1. Changes of colour and shape are the most common anomalies detected. Colouration, similar to moisture stains, were mainly caused by water runoff, while the corrosion stains are mainly related to spalling, indicating ongoing corrosion of rebars phenomena, and being more worrying than moisture stains in terms of durability;
2. Shape anomalies, specially spalling, were regularly found in older buildings such as FRAN (1971), ISCJ (1975), and JRP (1987). Spalling, mainly due to the corrosion of the reinforcement, can be attributed to a deficient constructive control associated with a low covering concrete thickness. Nevertheless, covering thicknesses in sampled areas are, on average, higher than the carbonation depth measured in samples (Figure 7);
3. Texture anomalies, which include bug holes, mapped cracking, fastening marks, and honeycombing, are also present in most of the buildings studied but are less frequent than other groups of anomalies. All of these are related to the construction technology, which reveals in some cases lesser care in the application of in situ concrete cast elements than in precast concrete.
4. The main anomalies related to concrete corrosion are spalling and oriented cracking and were found in buildings corresponding to case studies until 1998. These observations are in line with the higher values of carbonation depth for buildings until 1975. After that, it should be mentioned the case study UNL (2002) where the carbonation

depth measured (16.3 mm) was the highest, which was probably due to a different binder type, low binder content, or high water-to-cement ratio;

5. The results of the concrete strength show values between 32 and 54 MPa. It should be mentioned that the case study FRAN (1971) exhibits a higher dispersion of results. As shown in Table 1, only one class of reinforced concrete was prescribed for this building. There would not be expected such a dispersion of results for only one reinforced concrete class, unless (1) a variation in the composition of applied concrete had occurred or (2) due to different carbonation areas.
6. The analysis of carbonation depth and the concrete cover thickness of all the architectural concrete surfaces from the buildings' sampling zones (Table 11 and Figure 7) except for the FRAN (1971) demonstrates that the carbonation did not yet reach the rebars. However, spalling was locally identified in some buildings.
7. All the concrete materials analysed from the studied buildings are in a reasonable conservation state condition (Table 12), according to the proposed classification. Nevertheless, building JRP (1987), showed the largest number of anomaly types, which can be related to lack of quality control during the construction phase and lack of maintenance.

Anomalies detected on architectural concrete are according to the main defects usually found for this material, which includes cracks and spalling (e.g., [68–70]). Despite spalling being reported in the architectural surface of every award-winning building up to 1998, its extension and severity are yet limited.

Regarding non-architectural reinforced concrete, it should be noted that with the exception of buildings EUA53 (1970), FCG (1975), and C8 (2000), the carbonation front has already reached the rebars in some of the analysed areas, which can affect the durability of these buildings. In the case study LIP (1958), it was verified that rebar corrosion originates the cracking and detachment of the renders.

6. Conclusions

In this work, rendering and plaster materials from structural masonry (PRCBs) and reinforced concrete (RCBs) architecture award-winning buildings constructed between 1902 and 2002 in the city of Lisbon (Portugal) were analysed.

The applied methodology for the diagnosis and state of conservation assessment of renders, plasters, and concrete materials was completely adjusted to the intended purposes, using state of conservation classifications to widely reproduce their actual state of conservation based on the severity and the degree and extension of degradation concepts. This objective is of major importance to preserve the building authenticity, avoid demolition, and restrict the need for new materials. In the analysed buildings, which are characterised by above-average design, materials' choice, and careful construction, as testified by the award, the state of conservation seems to be primarily influenced by external rather than intrinsic factors. However, there are types of anomalies that are associated with specific construction technologies.

It was found that the renders and plasters of the buildings analysed are in a reasonable state of conservation, although it was verified that when compared to the RCBs, PRCBs presented a greater extent, degree, and severity of degradation.

Since the PRCBs are also the oldest buildings (1903 to 1944), the higher degree and extent of degradation of the assessed materials can be attributed to the longer exposure to the agents of degradation, as well as to the construction typology that makes them particularly vulnerable to the water action and other agents related to water, such as salts crystallisation.

In addition, regarding the analysed renders and plasters, it was found that until the 1960s, they had a multilayer construction, regardless of whether they were from indoors or outdoors. From the 1970s onwards, there was a change, with plasters and renders becoming monolayers certainly related with being cement-based and thus not as dependent on a multilayer structure for the water protection capacity of the lime-based coverings.

Regarding architectural concrete, i.e., buildings constructed between 1965 and 2002, the main anomalies detected are associated with reinforcement corrosion, mainly due to the low coverings and most likely to other enhancing characteristics, such as porosity, that favoured carbonation. However, a direct relationship between the average thickness of the reinforced concrete cover and the age of the buildings was not proved, nor with the differences in the carbonation depth, which is attributed to the different qualities of the concrete.

Similarly, the conservation state of the architectural concrete surfaces is reasonable, despite the restricted anomalies related to corrosion. Regarding the non-architectural concrete, the carbonation front reached, in most cases, the reinforcement, which may compromise the durability and safety of those buildings.

Since this study addresses 20th-century heritage buildings, it is expected to be reflected into a historical and social (even economic) value through the knowledge of the applied materials that reflect the functionality and aesthetical purposes of the needs of each construction period. To avoid major interventions to this built heritage, the following should be considered:

1. Ongoing investigation on the past interventions should be carried out for in-depth knowledge of the buildings' historical background.
2. Frequent monitoring of the areas that shows anomalies. Increased degradation can lead to the need for complete replacement of the materials, which forces a reduction of the life cycle as well as interrupts the original aesthetic and cultural concept of the buildings.
3. Actions to minimise the damage caused by agents such as water, using water protection capacity systems while preserving the vapour permeability of the walls, namely in the case of "before concrete" buildings (PRCBs).
4. Repair actions on exposed concrete degraded surfaces, due to reinforcement corrosion as a result of carbonation, to prevent the increase in anomalies, using compatible and informed repair materials.
5. Characterising the composition, the physical and the mechanical properties of mortars and concrete, to produce a range of data capable of leading to the informed choice of compatible materials, respecting their typology (e.g., multi or monolayer mortar), with a reduction of the carbon footprint, by performing minimal interventions and using local materials.

Further work includes the completion of the ongoing physical, mechanical, chemical, mineralogical, and microstructural characterisation. The experimental data obtained will allow defining criteria for the formulation of compatible repair materials to be applied in future conservation and rehabilitation actions.

As a final remark, it should be mentioned that for the development of conservation and rehabilitation solutions, data concerning materials' characterisation and compatibility criteria should be disseminated through scientific publications and should be given to the buildings' owners to ensure that they will be guided to applicators and rehabilitation consultants.

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References

1. ICOMOS. The Athens Charter for the Restoration of Historic Monuments. In Proceedings of the First International Congress of Architects and Technicians of Historic Monuments, Athens, Greece, 21–30 October 1931.
2. ICOMOS. The Venice charter for the conservation and restoration of monuments and sites. In Proceedings of the Second International Congress of Architects and Specialists of Historic Buildings, Venice, Italy, 25–31 May 1964.
3. ICOMOS. Krakow charter: Principles for the conservation and restoration of built heritage. In Proceedings of the International Conference on Conservation Kraków 2000, Krakow, Poland, 22–24 November 1998.
4. Santos, A.R.; Veiga, M.R.; Santos Silva, A.; Brito, J. Microstructure as a critical factor of cement mortars' behaviour: The effect of aggregates' properties. *Cem. Concr. Compos.* **2020**, *111*, 103628. [CrossRef]
5. Marques, A.I. Rehabilitation of Old Buildings: Reduction of Seismic Vulnerability by Reinforcing Walls. Ph.D. Thesis, Instituto Superior Técnico, Lisbon, Portugal, 2020. (In Portuguese).
6. Marques, A.I.; Ferreira, J.G.; Candeias, P.X.; Veiga, M.R. Axial compression and bending tests on old masonry walls. In Proceedings of the 3rd International Conference on Protection of Historical Constructions, Lisbon, Portugal, 12–15 July 2017.
7. Haspel, J.; Petzet, M.; Zalivako, A.; Ziesemer, J. *The Soviet Heritage and European Modernism. Heritage at Risk*; Hendrik Bässler Verlag: Berlin, Germany, 2007.
8. Cannata, M. The Repair and Alterations of the De La Warr Pavilion. *J. Arch. Conserv.* **2006**, *12*, 81–94. [CrossRef]
9. Buck, L.S. A Material Evaluation of the Gropius House: Planning to Preserve a Modern Masterpiece. *APT Bull.* **1997**, *28*, 29–35. [CrossRef]
10. Boothby, T.; Parfitt, M.; Roise, C. Case Studies in Diagnosis and Repair of Historic Thin-Shell Concrete Structures. *APT Bull.* **2005**, *36*, 3–11. Available online: https://www.jstor.org/stable/40004699 (accessed on 27 April 2021).
11. Allanbrook, T.; Normandin, K. The Restoration of the Fifth Avenue Facades of the Metropolitan Museum of Art. *Apt Bull.* **2007**, *38*, 45–53. Available online: https://www.jstor.org/stable/i40000278 (accessed on 27 April 2021).
12. Dell'Acqua, A.C.; Franzoni, E.; Sandrolini, F.; Varum, H. Materials and techniques of Art Nouveau architecture in Italy and Portugal: A first insight for a European route to consistent restoration. *Rest. Build. Monum.* **2009**, *15*, 129–144. [CrossRef]
13. Sandrolini, F.; Franzoni, E.; Varum, H.; Nakonieczny, R. Materials and technologies in Art Nouveau architecture: Façade decoration cases in Italy, Portugal and Poland for a consistent restoration. *Inf. Construcción* **2011**, *63*, 5–11. [CrossRef]
14. Marie-Victoire, E.; Cailleux, E.; Texier, A. Carbonation and historical buildings made of concrete. *J. Phys.* **2006**, *136*, 305–318. [CrossRef]
15. Tostões, A.; Vieira Constantino, M.; Casciato, M. *The Buildings*; Lisbon: Calouste Gulbenkian Foundation Central Services: Lisbon, Portugal, 2012.
16. Santos, D.; Veiga, M.R. Test Methods for Existing Coatings. In Situ and Laboratory Tests. In Proceedings of the V Jornadas FICAL, Lisbon, Portugal, 23–25 May 2016. (In Portuguese).
17. Cóias, V. *Inspection and Tests in the Rehabilitation of Buildings*, 2nd ed.; IST Press: Lisbon, Portugal, 2009. (In Portuguese)
18. Veiga, M.R. Conservation of Historic Renders and Plasters: From Laboratory to Site. In *Historic Mortars*; RILEM Bookseries; Válek, J., Hughes, J., Groot, C., Eds.; Springer: Dordrecht, Germany, 2012; Volume 7. [CrossRef]
19. Morgan, D.R. Compatibility of concrete repair materials and systems. *Constr. Build. Mater.* **1996**, *10*, 57–67. [CrossRef]
20. Veiga, M.R.; Fragata, A.; Velosa, A.L.; Magalhães, A.C.; Margalha, G. Lime-Based Mortars: Viability for Use as Substitution Renders in Historical Buildings. *Int. J. Archit. Herit.* **2010**, *4*, 177–195. [CrossRef]
21. Veiga, M.R.; Aguiar, J.; Santos Silva, A.; Carvalho, F. Methodologies for characterisation and repair of mortars of ancient buildings. In Proceedings of the 3rd International Seminar on Historical Constructions, Guimarães, Portugal, 7–9 November 2001.
22. CML. *Valmor Prize*; Silva, A.P., Ed.; Câmara Municipal de Lisboa: Lisbon, Portugal, 2004. (In Portuguese)
23. Bairrada, E.M. *Valmor Prize 1902–1952*; Biblioteca de Lisboa: Lisbon, Portugal, 1988. (In Portuguese)
24. Rodea, J. Apropos of Lisbon's Modern Architecture (1925–1965) in Modern Lisbon. International committee for documentation and conservation of buildings, sites and neighbourhoods of the modern movement. *Docomomo J.* **2016**, *55*, 4–7. [CrossRef]
25. Ministério do Comércio. *Decree nº 4036. Regulation for the Use of Reinforced Concrete*; Diário do Govêrno: Lisbon, Portugal, 1918. (In Portuguese)
26. Ministério das Obras Públicas e Comunicações. *Decree nº25948. Reinforced Concrete Regulation*; Diário do Govêrno: Lisbon, Portugal, 1935. (In Portuguese)
27. Ministério das Obras Públicas. *Decree nº 47723. Regulation of Reinforced Concrete Structures*; Imprensa Nacional de Lisboa: Lisbon, Portugal, 1967. (In Portuguese)
28. Ministério das Obras Públicas. *Decree nº 404/71. Hydraulic Binder Concrete Regulation*; Imprensa Nacional de Lisboa: Lisbon, Portugal, 1971. (In Portuguese)
29. Ministério das Obras Públicas, Transportes e Comunicações. *Decree-Law 445/89. Hydraulic Binder Concrete Regulation*; Imprensa Nacional de Lisboa: Lisbon, Portugal, 1989. (In Portuguese)

30. Rodrigues, M.F.S.; Teixeira, J.M.C.; Cardoso, J.C.P. Buildings envelope anomalies: A visual survey methodology. *Constr. Build. Mater.* **2011**, *25*, 2741–2750. [CrossRef]
31. Marteinsson, B.; Jónsson, B. Overall survey of buildings-performance and maintenance. In Proceedings of the 8th Durability of Building Materials and Components, Vancouver, BC, Canada, 30 May–3 June 1999; Institute for Research in Construction: Ottawa, ON, Canada, 1999.
32. Shohet, I.M.; Puterman, M.; Gilboa, E. Deterioration patterns of building cladding components for maintenance management. *Constr. Manag. Econ.* **2002**, *20*, 305–314. [CrossRef]
33. Jaggs, M.; Palmer, J. Energy performance indoor environmental quality retrofit—A European diagnosis and decision making method for building refurbishment. *Energy Build.* **2000**, *31*, 97–101. [CrossRef]
34. CEN. *EN 16085:2012. Conservation of Cultural Property. Methodology for Sampling from Materials of Cultural Property. General Rules*; European Standard: Brussels, Belgium, 2012.
35. Moropoulou, A.; Bakolas, A.; Bisbikou, K. Characterization of ancient, byzantine and later historic mortars by thermal and X-ray diffraction techniques. *Thermochim. Acta* **1995**, *269–270*, 779–795. [CrossRef]
36. Groot, C.J.W.P.; Bartos, P.J.M.; Hughes, J.J. Characterisation of old mortars with respect to their repair. In Proceedings of the 12th International Brick/Block Masonry Conference, Madrid, Spain, 25–28 June 2000; Volume 2.
37. Elsen, J. Microscopy of historic mortars—A review. *Cem. Concr. Res.* **2006**, *36*, 1416–1424. [CrossRef]
38. Magalhães, A.; Veiga, R. Physical and mechanical characterisation of historic mortars. Application to the evaluation of the state of conservation. *Mater. Construcción* **2009**, *59*, 61–77. [CrossRef]
39. Valek, J.; Veiga, M.R. Characterisation of mechanical properties of historic mortars—testing of irregular samples. In Proceedings of the 9th International Conference on Structural Studies, Repairs and Maintenance of Heritage Architecture (STREMAH 2005), Msida, Malta, 22–24 June 2005.
40. GranDuBé. *Quantities Associated with the Durability of Concrete*; Arliguie, G., Hornain, H., Eds.; Presses de l'Ecole Nationale des Ponts et Chaussées: Paris, France, 2007; Volume 1. (in French)
41. Massari, G.; Massari, I. *Damp Buildings. Old and New*; ICCROM: Rome, Italy, 1993.
42. Henriques, F. *Moisture in Walls*, 3rd ed.; LNEC: Lisbon, Portugal, 2001. (In Portuguese)
43. ASTM. *D2240-05. Standard Test Method for Rubber Property-Durometer Hardness*; ASTM International: West Conshohocken, PA, USA, 2005.
44. ASTM. *C805. Standard Test Method for Rebound Number of Hardened Concrete*; ASTM International: West Conshohocken, PA, USA, 2008.
45. Tavares, M. A The Conservation and Restoration of Exterior Cladding of Old Buildings–A Study and Repair Methodology. Ph.D. Thesis, Faculty of Architecture of the Technical University of Lisbon, Lisbon, Portugal, 2009. (In Portuguese).
46. CEN. *EN 125-04-2:2012b. Testing Concrete in Structures Non-Destructive Testing. Determination of Rebound Number*; European Standard: Brussels, Belgium, 2012.
47. CEN. *EN 14630. Products and Systems for the Protection and Repair of Concrete Structures. Test Methods. Determination of Carbonation Depth in Hardened Concrete by the Phenolphthalein Method*; European Standard: Brussels, Belgium, 2006.
48. Veiga, M.R.; Silva, S. Mortars. In *Long-Term Performance and Durability of Masonry Structures. Degradation Mechanisms, Health, Monitoring and Service Life Design*, 1st ed.; Ghiassi, B., Lourenço, P.B., Eds.; Elsevier. Ltd.: Amsterdam, The Netherlands, 2019; pp. 169–208.
49. Veiga, M.R. Rendered Façades: Anomalies and Resolution Strategies. In *Proceedings of the Façade Rehabilitation Seminar, Portugal, 2016*; Vasconcelos, G., Lourenço, P., Eds.; University of Minho: Guimarães, Portugal, 2016. (In Portuguese)
50. Sousa, V.; Almeida, N.; Meireles, I.; Brito, J. Anomalies in Wall Renders: Overview of the Main Causes of Degradation. *Int. J. Arch. Herit.* **2011**, *5*, 198–218. [CrossRef]
51. CEN. *EN 1504:2005. Products and Systems for the Protection and Repair of Concrete Structures*; European Standard: Brussels, Belgium, 2005.
52. Griffin, I.; Tate, J. Conserving our Wartime Heritage: A Reinforced Concrete Air Raid Shelter in East Lothian, Scotland. *J. Arch. Conserv.* **2012**, *18*, 81–100. [CrossRef]
53. Alonso, C.; Andrade, C. Lifetime of rebars in carbonated concrete. In Proceedings of the Understanding and Prevention of Corrosion, 10th European Corrosion Congress, Barcelona, Spain, 5–8 July 1993; Costa, J.M., Mercer, A.D., Eds.; Institute of Materials: London, UK, 1994.
54. AFNOR. *NF P-18-503. Concrete Surfaces and Facings. Identification Elements*; French Standard: Paris, France, 1989. (In French)
55. CIB. *Report No. 24. Tolerances on Blemishes of Concrete*; Report Prepared by CIB Working Commission W29 "Concrete Surface Finishings"; CIB: Rotterdam, The Netherlands, 1971.
56. Veiga, M.R.; Aguiar, J. Defining Intervention Strategies for Old Building Renders. In Proceedings of the 1st National Pathology and Buildings Rehabilitation Seminar, Porto, Portugal, 18–19 March 2003. (In Portuguese).
57. Serralheiro, M.I.; Brito, J.; Silva, A. Methodology for service life prediction of architectural concrete facades. *Constr. Build. Mater.* **2017**, *133*, 261–274. [CrossRef]
58. Ximenes, S.; Brito, J.; Gaspar, P.; Silva, A. Modeling the degradation and service life of ETICS in external walls. *Mater. Struct.* **2015**, *48*, 2235–2249. [CrossRef]
59. Silva, A.; Gaspar, P.; Brito, J. *Methodologies for Service Life Prediction of Buildings: With a Focus on Façade Claddings*, 1st ed.; Springer International Publishing: Cham, Switzerland, 2016.

60. Balaras, C.A.; Droutsa, K.; Dascalaki, E.; Kontoyiannidis, S. Deterioration of European apartment buildings. *Energy Build.* **2005**, *37*, 515–527. [CrossRef]
61. Martinho, C.; Veiga, R.; Faria, P. Marmorite-contribution to a proper preservation of a durable wall coating. *Conserv. Património* **2018**, *28*, 31–38. [CrossRef]
62. Magalhães, A.; Veiga, M.R.; Pina dos Santos, C.; Matias, L.; Vilhena, A. Methodologies for diagnosis of rendering anomalies due to moisture in walls. *Conserv. Património* **2008**, *7*, 45–54. [CrossRef]
63. Veiga, M.; Fragata, A.; Tavares, M.; Magalhães, A.C.; Ferreira, N. Inglesinhos convent: Compatible renders and other measures to mitigate water capillary rising problems. *J. Build. Apprais.* **2009**, *5*, 171–185. [CrossRef]
64. Franzoni, E. Rising damp removal from historical masonries: A still open challenge. *Constr. Build. Mater.* **2014**, *54*, 123–136. [CrossRef]
65. Veiga, M.R. Influence of application conditions on the cracking susceptibility of renderings. *Concr. Sci. Eng.* **2000**, *2–7*, 134–140.
66. Veiga, M.R.; Stefanidou, M.; Balksten, K.; Alvarez, I.; Nunes, C.; Theoridoridid, M.; Faria, P.; Papayianni, I.; Van Hees, R. Durability of lime-based renders: A review of some degradation mechanisms and assessment test methods. In Proceedings of the RILEM SMSS19 Rovinj Proceedings, Rovinj, Croatia, 18–22 March 2019; RILEM Publications SARL: Cachan, France, 2019.
67. Valença, J.; Almeida, C.; Botas, J.; Júlio, E. Patch Restoration Method: A new concept for concrete heritage. *Constr. Build. Mater.* **2015**, *101*, 643–651. [CrossRef]
68. Emmons, P.H. *Concrete Repair and Maintenance Illustrated*; RS Means: Kingston, NY, USA, 1993.
69. Courard, L.; Van der Wielen, A.; Darimont, A. From defects to causes: Pathology of concrete and investigation methods. In Proceedings of the 17th Slovenski Kolokvij o Betonih, Ljubljana, Slovenia, 19 May 2010.
70. Courard, L.; Gillard, A.; Darimont, A.; Bleus, J.M.; Paquet, P. Pathologies of concrete in Saint-Vincent Neo-Byzantine Church and Pauchot reinforced artificial stone. *Constr. Build. Mater.* **2012**, *34*, 201–210. [CrossRef]

4.4. CHARACTERISATION OF RENDERING AND PLASTERING MORTARS

4.4.1. Chemical, mineralogical, and microstructural analysis

The following research paper reports on the study of renders and plasters from eleven 20th century award-winning buildings with the Valmor Prize for Architecture between 1903 and 2002. The mortars have been investigated through XRD, optical and electronic microscopy (SEM-EDS), thermal analysis (TGA/DTA), wet chemical analyses and Atomic Absorption Spectroscopy (AAS).

Identifying the binders in mortars, especially when designing a plan for conserving and restoring the built heritage, is crucial for a correct intervention. Therefore, the characterisation of building materials has been part of the solution in the context of proper rehabilitation. Among other techniques used in the characterisation of binders, the importance of those related to microscopy (both optical and electronic) are particularly relevant. Microscopy and its combined techniques can be, indeed, the keys to such identification, as the classical approaches to mineralogical identification are not sufficiently conclusive in the investigation of the types of hydraulic binders in mortars. Since more than one binder can be applied simultaneously, it is unproductive to investigate them without microscopic techniques.

This work used microscopic techniques (petrography combined with SEM-EDS) as a fundamental part of the characterisation. These techniques are essential for distinguishing between the various types of binders and are irreplaceable as they promote disambiguation and clarify doubts about the application of the binders.

The main results reveal that the use of air lime lasted until the 1940s. It also highlights the beginning of the use of Portland cement in mortars in the 1930s, mixed with air lime, and the abandonment of mortar formulations solely based on air lime. This study highlights the use of interior finishing gypsum-lime-based mortars until the 1940s and different types of Portland cement from the 1940s onwards. Portland cement was the main binder for the analysed stone imitating mortars from 1940s to 1970s.

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Composition of renders and plasters of award-winning buildings in Lisbon (Portugal): A contribution to the knowledge of binders used in the 20th Century

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ABSTRACT

This paper reports on the study of renders and plasters from 20th-century award-winning buildings in Lisbon (Portugal) with the Valmor Prize for Architecture. The mortars have been investigated through XRD, optical and electronic microscopy (SEM-EDS), thermal analysis (TGA/DTA), wet chemical analyses and AAS. The results reveal that the use of air lime lasted until the 1940s. It also highlights the beginning of using of Portland cement in mortars in the 1930s, mixed with air-lime, and the abandonment of mortar formulations solely based on air lime. This study highlights the use of finishing lime-gypsum-based mortars until the 1940s and different types of Portland cement from the 1940s onwards. Portland cement was the main binder for the analysed stone-imitating mortars from 1940s to 1970s. Finally, salt contamination was occasionally found in mortars, which generally reveal a good state of conservation.

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20th century; aerial mortars; characterisation; hydraulic mortars; microscopy; mineralogy; renders; plasters

1. Introduction

Lisbon (Portugal's capital city) experienced considerable development in the last century. The increased industrial facilities and population growth caused a need to open the city boundaries widely. Given such events, Lisbon's architectural and constructive challenges grew consistently with the trend of ongoing population growth, as more dwellings should be built.

Design trends were recorded in works of high architectural value, recognised by the Lisbon city council through an architecture award, established in 1902 and designated as the Valmor Prize for Architecture. The use of new construction materials, such as Portland cement, occurred in Portugal with some delay compared to other European countries (Oliveira 1995). The use of Portland cement-based composite materials began with the importation of Portland cement from abroad in the 1860s (Mateus 2018) due to the adaptation to technological advances of the late 19th century. A progressive abandonment of aerial binders, namely air lime, rose as a significant architectural breakthrough and construction fastness even though most traditional techniques used in the 19th century have survived and lasted during the 20th century (Mateus 2002).

Historically, air lime began to be used from immemorial times and hydraulic lime was already used from the Roman and Greek times. A method to create hydraulic binders involved mixing air lime with natural or artificial pozzolans to produce mortars or concrete capable of hardening when exposed to water (Callebaut et al. 2001).

The hydraulic properties of binders have been gradually discovered over time. In 1756, John Smeaton discovered the hydraulic properties of products obtained by burning impure limestones (Callebaut et al. 2001). In 1796, James Parker patented Roman cement, also known as Natural cement (Parker 1796). It was obtained by burning a septarian marlstone that was grounded before use. This binder was produced without additions and required grinding rather than slaking, as in the case of hydraulic lime. In 1824, Joseph Aspdin patented "Portland cement", a material produced by firing a mixture of finely-ground clay and limestone (Winter 2012). Still, this newfound product was produced at a lower temperature than the modern Portland cement. Only William Aspdin and Isaac Johnson's late efforts, between 1844 and 1845, had led to an improvement by reaching the sintering temperatures (>1300°C) to produce clinker, a very reactive cementitious material (Blezard 1998; Winter 2012). Technological advances since the 19th century have produced significant changes in cement properties.

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One of the properties with different results achieved in the 20th century was compressive strength, with Portland cement of the second half of the 19th century being less resistant than those of the 20th century (Blezard 1998). Despite the increasing efforts made between the 19th century's last quarter and the beginning of the 20th century to manufacture quality Portland cement in vertical kilns, Portland cement manufacturing in the country only gained its status from 1923 on with the inauguration of the most extensive factory line at the time (Coimbra, Moreira dos Santos, and Braz de Oliveira 1995). In addition to the calcination technology, cement grinding is one of the main determining factors regarding cementitious properties. Using ball mill technology increased fineness in 20th-century cement (Vidovszky and Pintér 2018). The properties of the hydraulic binders, namely Hydraulic lime — either a natural product (NHL) or an artificial product obtained by calcination of a mixture of clay and limestone (HL) -, Roman or Natural Cement and Portland Cement, are dependent on the composition of the raw materials. The manufacturing technology, i. e., type, time, and calcination temperature, will define characteristic chemistry. Considering that the maximum temperature was kept below 1250°C, thus below the sintering point, critical to the clinker's production, the reactive phases of hydraulic lime, Natural cement and early PC were mainly silicates and aluminates (hydraulic phase) and unbound CaO (air-hardening phase). The hydraulic properties of hydraulic lime can be primarily attributed to the di-calcium silicate (C_2S — belite) and, to some extent, to the tri-calcium aluminate (C_3A) (Boynton 1980; Brown and Clifton 1988; Czernin 1980; Lea 1988). In comparison, the predominant reactive component of Portland cement is the tri-calcium silicate (C_3S — alite), responsible for its quicker hardening and higher ultimate strength (Bye 1999; Lea 1988; Taylor 1990; Zacharopoulou 2009). Composite cement has been used over time as mineral additions can replace clinker, offering considerable opportunities to optimise workability, strength development and durability (Deolalkar 2015). Among the mineral additions, divided into hydraulic, pozzolanic and almost inert filler (Kurdowski 2014), the latent hydraulic ground granulated blast furnace slag (GGBFS) has simultaneously pozzolanic properties. Portland cement with slag was first produced in Germany in 1883 (Papadakis and Venuat 1964), and its use began to spread after the First World War. In contrast, the pozzolanic nature of fly ash was first identified in the US in 1914 (Anon 1914). In Portuguese cement factories, granulated slag began to be incorporated in 1961 (Coutinho 2012). The most used additions in Portugal are limestone filler and fly ash. Regarding the latter, its application was first regulated by a national decree published in 1980 (MHOP - Ministério da Habitação e Obras Públicas 1980). A memorandum was promulgated between Portuguese coal power plants and the cement industry for the supply of fly ash and its full use (MPAT - Ministério do Plano e da Administração do Território, da indústria e Comércio e das Obras Públicas, Transportes e Comunicações 1987).

Hydraulic binders have spread out in construction for structural elements and coatings. It is unknown when the air lime ceased to be used in rendering mortars and plasters or if it ceased entirely. However, lime as one of the main constituents was reported in gypsum-based plasters of the interior Portuguese 20th-century finishing coatings (Freire et al. 2019), denoting a trend to preserve the old constructive techniques.

At the beginning of the 20th century, a finishing mortar was developed in Central Europe in which Portland cement allowed façades to have an imitation stone appearance. Since imitation render was a qualitative and less expensive alternative to natural stone, this finishing mortar was applied in many constructions (Govaerts et al. 2014). Stone-imitating mortars emerged in Portugal between 1940–1970. Those that are characterised by the visible aggregates on the surface due to a washing technique that removes the superficial slurry, exposing the aggregates, are called *Marmorite*. Some of these coatings have a flat surface due to polishing for functional or aesthetic reasons. The first *Marmorite* mortars were made of several aggregates, mainly crushed stone and stone powder, with air lime as the main binder (Martinho, Veiga, and Faria 2018; Veiga et al. 2019). White cement is also an excellent option since it provides a neutral base for stone-imitating mortars, whether white or pigmented (Dekeyser, Verdonck, and De Clercq 2017). However, it should be mentioned that the Portuguese production of white Portland cement was only established in 1944, and the first ignition of the rotary kiln took place at the end of 1949 (Oliveira 1995).

2. Aim of the study

The characterisation of renders and plasters of Valmor prize for architecture award-winning buildings was carried out. This work does not aim to deal with ordinary buildings but to assess the progress achieved in each period of construction in Portugal during the 20th century, based on buildings of undisputed architectural value, which, in general, were built using state-of-the-art technology of their time. The results of the characterisation through X-ray diffraction (XRD), simultaneous thermogravimetry and differential thermal analysis (TGA/DTA), petrography, scanning electron microscopy with energy dispersive X-ray spectrometry (SEM-EDS), wet chemical analyses and atomic absorption spectroscopy (AAS) are presented and discussed in this paper. The results are meant to shed light upon the evolution of the applied renders and plasters during the 20th century, their main properties and state of conservation, and provide data concerning compatibility issues. It is known that a complete characterisation of the original composition is required to ensure a good restoration whenever it is needed. An appropriate restoration strategy must include using building materials compatible with the original ones (Aggelakopoulou, Bakolas, and Moropoulou 2011; Middendorf et al. 2005a; Veiga 2012; Veiga and Santos Silva 2019; Veiga et al. 2001).

3. Materials and methods

3.1. Case studies and sampling

Samples were collected in locations that did not compromise the buildings' structural safety or cause visual impact. Non-disturbed or unchanged locations were identified according to the oral information from the building owners and their records, as well as the construction work's records, previously consulted in the historical municipality archives. However, mortars applied in refurbishment works during the 20th century were also sampled. That is the case of some samples from the *Diário de Notícias*, the 1940 award-winning building, which application period is known.

Sixty-one renders and plaster samples were collected in eleven buildings constructed in the 20th century. A brief description of the studied buildings is presented in Table 1.

For technical and constructive reasons (e.g. inaccessible/unauthorised zones and external façades without rendering mortars), most of the samples were collected by hammer and chisel at indoor positions. As this work is part of a broader study also involving the characterisation of the employed concretes, some samples were collected while sampling concrete cores. Samples were also extracted in areas where detachments occurred to avoid physical damage or adverse visual impact, as all the buildings are still in use.

The multi-layered samples are often finished with white thin-layer plasters (Figure 1) and stone imitation mortars (Figure 2). A multi-layer system often consists of a layer applied on the substrate, one or several intermediate layers and, finally, the finishing layer. A painting coating covers the finishing layer of twenty-two samples, which are identified in Table 2. All samples are identified by an alphanumeric code (Table 2), in which letters placed at the end of the ID are related to the stratigraphic position. Thus, the letter (A) corresponds to the outermost layer. Table 2 presents the locations of the samples and their macroscopic description.

3.2. Characterisation methodology

The characterisation methodology comprises a range of complementary techniques (Middendorf et al. 2005a, 2005b; Veiga et al. 2001), which aim to identify mortar characteristics (e.g. binder and aggregate nature) and the decay processes.

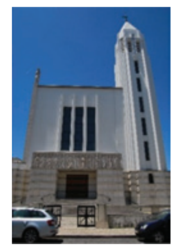
After drying at 40°C in a drying oven for 48 hours, each sample layer from the multi-layer mortar system was carefully separated from the whole set with the help of mechanical tools. Paint coatings were removed on most mortars except one *Marmorite* sample (EUA53-3A), whose paint could not be removed by scraping. Each layer was carefully crushed with a rubber hammer and sieved to separate the binder-rich fraction from the remaining. In the case of white thin-layer smooth finishing plasters, their homogeneity and the absence of aggregates did not allow the separation into fractions. The finer fraction, i.e., the binder-rich fraction with a higher binder concentration, was obtained by extracting the fines passing a 106 μm sieve directly from the bulk mortar and was characterised by X-ray diffractometry (XRD).

The overall fraction corresponding to the samples as collected was obtained by crushing and grinding to pass through a 106 μm sieve and was then analysed by XRD. All the ground samples were then divided for the remaining tests, namely thermal analysis, wet chemical analyses, and Atomic Absorption Spectrophotometry (AAS). Additionally, undisturbed fragments were chosen to manufacture thin sections for petrographic analyses and polished surfaces for SEM-EDS analyses.

3.2.1. Mineralogical characterisation

XRD was carried out in a Bruker D8 Discover diffractometer with CuKα radiation operating at 40 kV and 40 mA. The XRD patterns were measured between 3° to 75° 2θ, with a step size of 0.05° and a recording time per step of 1s. The crystalline phases were identified with the PDF-ICDD Powder Diffraction Database (International Centre for Diffraction Data), using the PANalytical X'Pert HighScore Plus software.

Table 1. Buildings' summary description.

Case study ID	Prize-winning year	Case study description	Image of buildings' façades
CVT (1903)	1903	*Ventura Terra* building • Architectural Style: Art Nouveau. • Use: residential building. • Construction period: 1902–1903. • Construction system: Self-supporting masonry walls. • Studied samples: five samples from indoors.	
AR49 (1923)	1923	*Luiz Rau* building • Architectural Style: Eclectic. • Use: residential building. • Construction period: 1920–1923. • Construction system: Mainly self-supporting masonry walls. • Studied samples: seven samples from indoors and four from the outdoors.	
IRF (1938)	1938	*Nossa Senhora de Fátima* Church • Architectural Style: Modern. • Use: Church. • Construction period: 1934–1938. • Construction system: Reinforced concrete structure. • Studied samples: eight samples from indoors.	
CBP (1939)	1939	*Bernardo da Maia* House • Architectural Style: Baroque-inspired. • Use: housing and office space. Recently adapted to school. • Construction period: 1938–1939. • Construction system: Hybrid concrete-masonry structure. • Studied samples: eight samples from indoors.	
DN (1940)	1940	*Diário de Notícias* Building • Architectural Style: Modern. • Use: newspaper writing and typesetting. Recently adapted to residential building. • Construction period: 1936–1940. • Construction system: Reinforced concrete structure. • Studied samples: twelve samples from indoors.	
AAC (1944)	1944	*Cristino da Silva* Building • Architectural Style: *Português Suave.* • Use: residential building. • Construction period: 1942–1944. • Construction system: Hybrid concrete-masonry structure. • Studied samples: two samples from indoors and four from the outdoors.	
LIP (1958)	1958	*Laboratories of Pasteur Institute of Lisbon* • Architectural Style: Modern Industrial. • Use: Pharmaceutical laboratories. It was later adapted for use as an educational establishment. • Construction period: 1955–1957. • Construction system: Reinforced concrete structure. • Studied samples: two samples from outdoors.	

(*Continued*)

Table 1. (Continued).

Case study ID	Prize-winning year	Case study description	Image of buildings' façades
EUA (1970)	1970	*America* building ● Architectural Style: Modern. ● Use: residential building. ● Construction period: 1966–1969. ● Construction system: Reinforced concrete structure. ● Studied samples: four samples from indoors and two from outdoors.	
FCG (1975)	1975	*Calouste Gulbenkian Foundation Headquarters and Museum* ● Architectural Style: Modern. ● Use: Offices and museum. ● Construction period: 1963–1969. ● Construction system: Reinforced concrete structure. ● Studied samples: one sample from an outdoor wall.	
JRP (1987)	1987	*Jacob Rodrigues Pereira Institute* ● Architectural Style: Modern. ● Use: Educational institution. ● Construction period: 1984–1987. ● Construction system: Reinforced concrete structure. ● Studied samples: one sample from a render of an external element (terrace guardrail).	
UNL (2002)	2002	*New University of Lisbon Rectory* ● Architectural Style: Modern. ● Use: Educational institution. ● Construction period: 2000–2002. ● Construction system: Reinforced concrete structure. ● Studied sample: one sample from indoors.	

3.2.2. Thermal analysis

The overall fraction of each sample was analysed in a simultaneous thermogravimetric and differential thermal analysis (TGA/DTA) SETARAM unit, under an inert atmosphere (argon, 3 L/h) with a uniform heating rate of 10°C/min, from room temperature to 1000°C. The CO_2 content was obtained from the mass loss in the range of 520 to 900°C.

TGA was also used to determine the gypsum content of finishing white smooth mortars. The mass loss obtained between 85–250°C, corresponding to gypsum dehydration, was converted into $CaSO_4.2\,H_2O$, multiplying by 4.78 (ratio between the molecular masses of $CaSO_4.2\,H_2O$ and $2\,H_2O$).

3.2.3. Microstructural analyses

3.2.3.1. Petrographic observations. The petrographic observations were performed on thin polished sections under a petrographic microscope Olympus BX60, equipped with four magnification lenses: 5×, 10×, 20×, and 40 × . These observations were carried out to complement the XRD data regarding the identification and the nature of the aggregates, but also as an indispensable tool for the identification of the binder type. Although XRD is an excellent method for determining mineral phases, residues in the mortar matrix are only visible through microscopic tools that can indicate the type of binder used (Walsh 2007).

3.2.3.2. SEM-EDS. Scanning Electron Microscopy with Energy Dispersive Spectrometry (SEM-EDS) was performed using a Hitachi S3700N coupled to a microanalysis Quantax EDS system equipped with a Bruker XFlash® Silicon Drift Energy Dispersive Detector. The EDS data were processed using the Esprit software from Bruker. The microscope was operated with a 20 kV accelerating voltage and a variable 20–40 Pa chamber atmosphere, and the analyses were performed on uncoated specimens. One of the main purposes for applying SEM-EDS was to complement petrographic analyses, namely, to confirm the type and nature of the binders and to detect the occurrence of neoformation and alteration products in the binder pastes.

3.2.4. Wet chemical analyses and AAS

A cold nitric acid dilution (1:50) separated siliceous sand from the binder. The residue of the acid attack — the insoluble residue (IR) — was calcined at 1000°C,

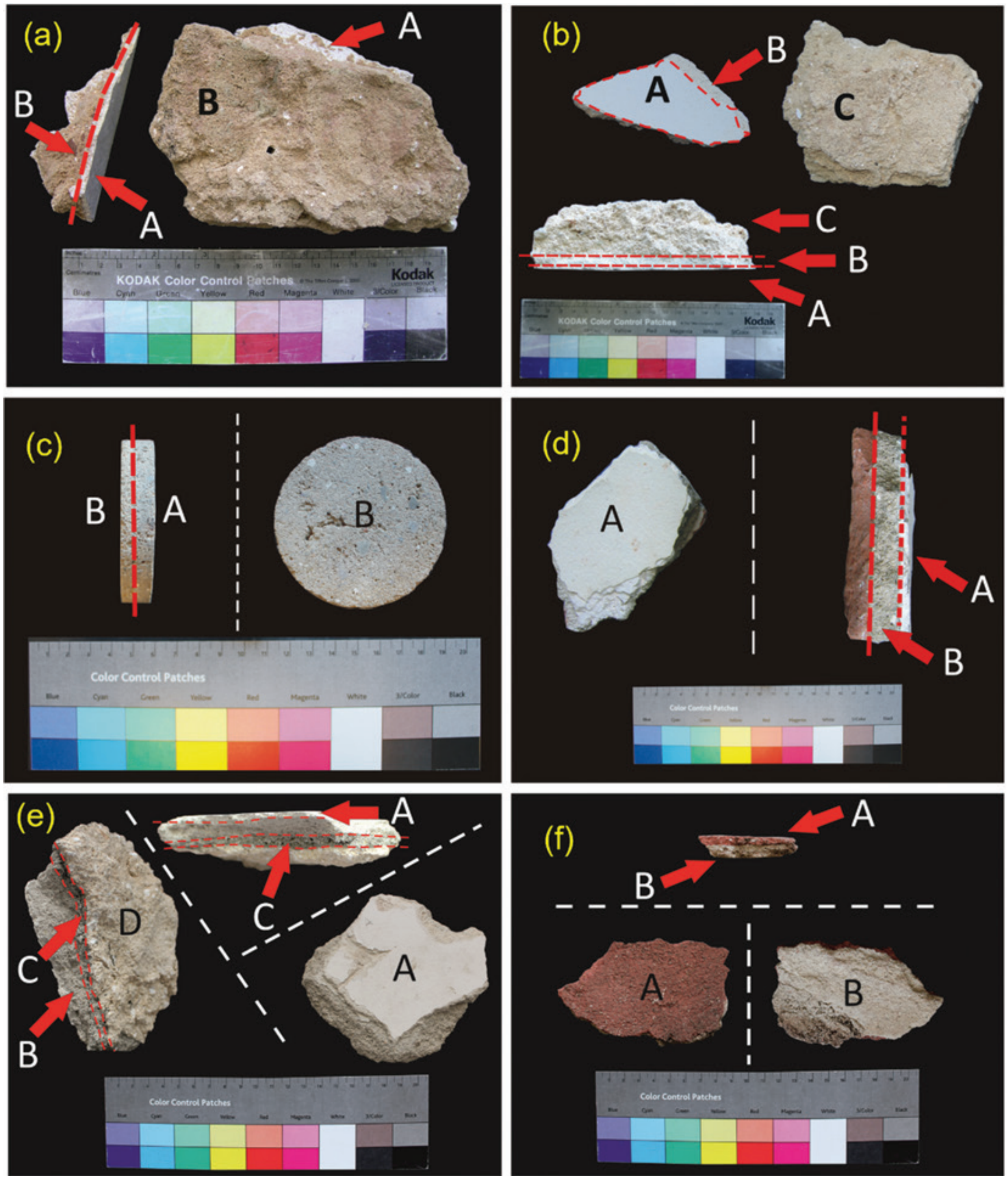


Figure 1. Multi-layered samples from six case studies. (a) A - CVT3A, B - CVT3B; (b) A - AR49-15A, B - AR49-15B, C - AR49-15C; (c) A - IRF7A, B - IRF7B (d) A - CBP4A, B - CBP4B; (e) A - DN19A, B - DN19B, C - DN19C, D - DN19D; (f) A - AAC1A, B - AAC1B.

corresponding to the siliceous aggregate content in the analysed sample (CEN 2014). This procedure was carried out for all samples except the white thin-layer finishing plasters since no sand aggregate was reported after the visual observations.

In the filtered solutions resulting from the nitric acid attack, the alkalis (sodium and potassium) content was determined by AAS following EN 196–2 (CEN 2014). The analysis was performed in a Shimadzu AA-6300 flame spectrophotometer. Hollow cathode lamps of each element and an air-acetylene flame were used.

Soluble silica was determined by the polyethylene oxide method, and sulphate content was determined by the gravimetric method with barium chloride. All these methods comply with EN 196–2 (CEN 2014). Chlorides were determined by direct potentiometry (CEN 2007).

3.2.5. Binder to aggregate ratio

Binder to aggregate ratio (b:a) was calculated using the values of IR obtained by wet chemical analysis and the CO_2 content obtained by TGA. IR corresponds to siliceous aggregate, and its mortar content was computed to determine the b:a ratio. The lime content in the binder was calculated, considering the lime was utterly hydrated and not carbonated. The amount of CO_2 will correspond to the

Table 2. Samples identification, description and sampling zones.

Case study	Location of the samples	Sampled element	Application technique	Sample ID	Th.	Summary macroscopic description
CVT (1903)	Basement-Entrance Hall	Interior wall	Multi-layer plaster	CVT1A	2	White thin-layer smooth finishing plaster
				CVT1B	5	White mortar with siliceous sand
				CVT1C	20	Brownish mortar with lime lumps and siliceous sand
	Ground floor - Entrance Hall. Adornment arch	Interior wall	Multi-layer plaster	CVT3A	5	White thin-layer smooth finishing plaster (a)
				CVT3B	20	Orange-brownish, friable mortar with lime lumps and siliceous sand
AR49 (1923)	Courtyard Access. Ground floor. Ceiling	Interior wall	Multi-layer plaster	AR49-2A	4	White thin-layer smooth finishing plaster (a)
				AR49-2B	4	Whitish mortar with fine siliceous sand
		Exterior Wall	Multi-layer render	AR49-6C	10*	Orange-brownish, friable mortar with lime lumps and siliceous sand
		Exterior wall	Multi-layer render	AR49-7B	6	Whitish mortar with fine siliceous sand
	Balcony on the 5th floor. East Façade	Exterior Wall	Multi-layer render	AR49-8A	4	Whitish mortar with fine siliceous sand (a)
				AR49-8B	12	Orange-brownish, friable mortar with lime lumps and siliceous sand
	West-facing wall between 5th and 6th stair floor landing	Interior wall	Multi-layer plaster	AR49-11A	3	White thin-layer smooth finishing plaster (a)
				AR49-11B	5	White mortar with siliceous sand
	Stairs' window between the 4th and 5th floor	Interior window -lintel	Multi-layer plaster	AR49-15A	3	White thin-layer smooth finishing plaster (a)
				AR49-15B	5	White mortar with siliceous sand
				AR49-15C	25	Orange-brownish, friable mortar with lime lumps and siliceous sand
IRF (1938)	Sacristy	Interior wall	Multi-layer plaster	IRF1B	10	Brownish mortar with lime lumps and siliceous sand
	Nossa Senhora da Piedade Chapel	Interior wall	Multi-layer plaster	IRF2A	5	White mortar with fine siliceous sand (a)
				IRF2B	3*	Orange-brownish mortar with siliceous sand
	Main chapel gallery/roof access	Interior wall	Multi-layer plaster	IRF3A	4	White mortar with fine siliceous sand (a)
				IRF3B	5*	Orange-brownish mortar with lime lumps and siliceous sand
	Interior access to the bell tower	Interior wall	Monolayer plaster	IRF4A	10	Single-layer, grey-brown mortar with lime nodules and siliceous sand (a)
		Interior wall	Multi-layer plaster	IRF7A	10	Brownish-grey mortars with lime lumps and siliceous sand (a)
				IRF7B	8	Brownish-grey mortars with lime lumps and siliceous sand
CBP (1939)	Basement. Staff room	Interior wall	Monolayer plaster	CBP1A	15	Single-layer, whitish mortar with lime lumps and siliceous sand (a)
	1st floor activity room	Interior wall	Multi-layer plaster	CBP4A	3	White thin-layer smooth finishing plaster (a)
				CBP4B	15	Greyish mortar with siliceous sand
	1st floor. Corridor to the activity's room	Interior wall	Multi-layer plaster	CBP6A	4	White thin-layer smooth finishing plaster (a)
				CBP6B	15	Whitish mortar with lime lumps and siliceous sand
		Interior wall–ceiling	Multi-layer plaster	CBP7A$_1$	5	External layer of moulded on-site plaster element (crown moulding)
				CBP7A$_2$	5	Internal layer of moulded on-site plaster element (crown moulding)
				CBP7B	20*	Whitish mortar with lime lumps and siliceous sand

(*Continued*)

Table 2. (Continued).

Case study	Location of the samples	Sampled element	Application technique	Sample ID	Th.	Summary macroscopic description
DN (1940)	Level −2. Technical rooms' corridor	Interior wall/column	Monolayer plaster	DN9A**	31	Greyish mortar with siliceous sand (a)
		Interior wall/column	Monolayer plaster	DN10A**	31	Greyish mortar with siliceous sand
		Interior wall/column	Multi-layer plaster	DN11A**	20	Greyish mortar with siliceous sand (a)
				DN11B**	6	Greyish mortar with siliceous sand.
	Level −2. Warehouse room	Interior wall/column	Multi-layer plaster	DN12A	20	Greyish mortar with siliceous sand (a)
				DN12B	30	Brownish mortar with lime lumps and siliceous sand
				DN12C	15	Compact grey mortar with fine siliceous sand
				DN12D	15	Whitish mortar with lime lumps and siliceous sand
	5th floor. Office room. North wall	Interior wall	Multi-layer plaster	DN19A	4	White thin-layer smooth finishing plaster (a)
				DN19B	12	Greyish mortar with siliceous sand.
				DN19C	5	Greyish mortar with grains of siliceous sand
				DN19D	20	Light brownish mortar with lime lumps and siliceous sand
AAC (1944)	Side access to the ground floor	Exterior wall	Multi-layer render	AAC1A	7	Rough red-coloured mortar with fine siliceous sand
				AAC1B	7	Greyish mortar with siliceous fine sand
	Ground floor outdoor render	Exterior wall	Multi-layer render	AAC2A	5	Stone-imitating mortar with projected limestone aggregates
				AAC2B	15	Greyish mortar with siliceous sand
	Boiler room	Interior wall	Multi-layer plaster	AAC3A	5	Stone-imitating mortar *Marmorite* type with white and bluish limestone aggregates
	Step coating of inside stairs	Interior wall	Multi-layer plaster	AAC4A	5	Stone-imitating mortar *Marmorite* type with white limestone aggregates
LIP (1958)	Chimney render	Exterior wall	Monolayer render	LIP1A	20	Grey mortar with siliceous sand (a)
	Ground floor. South building. west façade	Exterior wall	Monolayer render	LIP9A	7	Grey mortar with siliceous sand (a)
EUA53 (1970)	Common interior staircase wall. 3rd floor	Interior wall	Multi-layer plaster	EUA53-2A	10	Stone-imitating mortar *Marmorite* type with quartzite aggregates
				EUA53-2B	15	Grey mortar with siliceous sand
	Chimney render	Exterior wall	Multi-layer render	EUA53-3A	8	Yellow stone-imitating mortar *Marmorite* type with white limestone aggregates (a)
				EUA53-3B	50	Compact grey mortar with siliceous sand
	Corridor of the technical area	Interior wall	Multi-layer plaster	EUA53-4A	5	White stone imitating mortar *Marmorite* type with white limestone aggregates
				EUA53-4B	20	Grey mortar with siliceous sand
FCG (1975)	Ventilation shaft	Exterior wall	Monolayer render	FCG4A	10	Single-layer greyish mortar with siliceous sand (a)
JRP (1987)	Terrace guardrail render	Exterior concrete pre-cast element	Monolayer render	JRP2A	10	Single-layer greyish mortar with siliceous sand (a)
UNL (2002)	Air treatment unit room	Interior wall	Monolayer plaster	UNL3A	15	Single-layer greyish mortar with siliceous sand (a)

Notation: Th — Average thickness of the sample (mm); * Measurement performed directly on the sample (lower than the total thickness of the corresponding layer); ** Samples of the plasters applied during a refurbishment action in 1998; (a) covered by a painting coating.

Table 3. Qualitative mineralogical composition by XRD.

Samples	Case study	Identified crystalline compounds: Overall fraction: Quartz	Calcite	Feldspars	Muscovite	Kaolinite	Others[1]	Binder-rich fraction: Quartz	Calcite	Feldspars	Muscovite	Kaolinite	Others[1]
CVT1B	CVT (1903)	++/+++	++	+++	++	?	G (++)	+/++	+++	+/++	+/++	trc	G (++/+++)
CVT1C		+++	++	+++	+/++	trc	G (trc)	+/++	+++	trc/+	+	trc	G (+); Ha (trc)
CVT3B		+++	+++	+	trc	?	Ha (trc/+)	+	+++	?	?	trc	Ha (trc/+); G (trc)
AR49-2B	AR49 (1923)	+++	++/+++	++/+++	+/++	trc	Ha, G (trc)					(a)	
AR49-6C		+++	++	++/+++	+/++	?	G (+/++); Ha(?)	+/++	+++	+	+	-	G (++)
AR49-7B		++/+++	++	++/+++	+/++	trc	Ha (trc); G (+)	+	+++	+	+	?	Ha (trc/+); G (+/++)
AR49-8A		+++	++	++	+/++	trc	P (+/++)	+/++	+++	+	trc/+	-	P (++); V (trc)
AR49-8B		+++	++	++	trc/+	-	-	+/++	+++	+	+	trc	-
AR49-11B		+++	++	++/+++	+/++	trc	Ha (?)	+/++	+++	+	++	trc	Ha (trc)
AR49-15B		+++	++/+++	+/++	+	-	G (trc)					(a)	
AR49-15C		+++	++	++	+	?	G (trc)	+/++	+++	trc	+	-	G (trc)
IRF1B	IRF (1938)	+++	++	++/+++	++	?	G (trc)	++	+++	+/++	+/++	trc	G (trc/+)
IRF2A		+++	++/+++	++/+++	++	trc	G (?)	+/++	+++	+/++	+	trc	G, CP (?)
IRF2B		+++	++	+/++	trc	trc	G, CP (?)	++	+++	++	+/++	trc/+	G (trc/+)
IRF3A		++/+++	++/+++	++	+	?	CP (?)	+/++	+++	+/++	trc	?	G, CP (?)
IRF3B		+++	++	+/++	trc	trc	-	+/++	+++	+/++	trc/+	trc	Mc (trc)
IRF4A		+++	++	++/+++	++	?	-	++	+++	+/++	++	trc	G (trc)
IRF7A		+++	++	++	+	-	CP (trc)	+/++	+++	trc/+	trc	trc	Mc (?); CP (trc/+); V, A (trc)
IRF7B		+++	++	++	+/++	-	CP (trc)	++	+++	trc/+	trc	trc	A, Mc (trc); CP (+)
CBP1A	CBP (1939)	+++	++	+/++	+	?	-	+/++	+++	trc	trc/+	trc	-
CBP4B		+++	+/++	++/+++	+/++	trc	Cl (trc/+); V (trc); CP (?)	++	++/+++	+/++	+	trc/+	V (+); G (trc); CP (trc/+)
CBP6B		++/+++	++	++	+	trc	-	+/++	+++	+/++	+	trc/+	-
CBP7B		++/+++	+/++	++/+++	+	trc	P (trc)	+/++	+++	++/+++	+/++	trc	P (+); G (+/++)
DN9A	DN (1940)	++/+++	++	+/++	+	-	CP (trc/+)	+/++	++/+++	+	trc	trc	P, V (trc); CP (trc/+)
DN10A		+++	++	++	trc/+	trc	V (?); CP (trc)	+/++	++/+++	+	trc	trc/+	P (trc); V, CP (trc/+)
DN11A		+++	++	++	+	trc	V, CP (trc)	+/++	++/+++	+	trc	trc	V (+); CP (trc/+)
DN11B		+++	++	++	trc/+	trc	Cl, E (trc); P, CP (trc/+)					(a)	
DN12A		+++	++	+/++	+/++	+/++	V (+); G (trc/+); CP (trc)	+/++	++/+++	+/++	++	++	P (trc); V (++); G, E (trc/+); CP (+)
DN12B		+++	+/++	+/++	+	trc	P (+/++); V (trc/+); Hc (trc); Mc (+); G (?); E, CP (trc/+)	+/++	+/++	+	?	trc	P (++); V (trc/+); Hc (trc); G (?); Mc, E (+/++); CP (+)
DN12C		+++	+/++	++	+/++	trc	Cl, Hc, Mc, E, CP (trc/+); V (trc); G (?)	++	++	+/++	+	-	V, Hc (trc/+); Mc, CP, E (+); G (?)
DN12D		++/+++	++	++/+++	+	-	V (+); CP (?)	+/++	+++	+/++	+	trc	V (+/++); A (trc); CP (?)
DN19B		+++	++	+/++	+	+	V (trc/+); Mc (trc); CP (?)	+/++	++/+++	+	+	+/++	D (?); V (+/++); Mc (+); CP (vtg/+)
DN19C		+++	++	++	trc	trc	P, Mc (trc); D (?); CP (trc/+)					(a)	
DN19D		+++	++	+/++	trc/+	trc	V (trc/+); Mc (?); CP (?)	+/++	+++	+	trc	trc/+	V (+); Mc, CP (?)

(*Continued*)

Table 3. (Continued).

		Identified crystalline compounds											
		Overall fraction						Binder-rich fraction					
Samples	Case study	Quartz	Calcite	Feldspars	Muscovite	Kaolinite	Others[1]	Quartz	Calcite	Feldspars	Muscovite	Kaolinite	Others[1]
AAC1A	AAC (1944)	+++	++	++	+	trc/+	H (trc/+); CP (trc)	+/++	+++	+/++	+/++	+	H (+); CP (trc/+)
AAC1B		+++	++	++	+	trc/+	PC (trc)	+/++	+++	+/++	+	+/++	Hc (trc); CP (trc/+)
AAC2A[2]		+	+++	trc	trc	-	D (+); V, G, CP (trc); A (+/++)	++/+++	++	++	trc/+	trc	D, A, CP (trc/+); V (+); G (trc)
AAC2B		+++	++	++	+	trc	D, V, A, G (trc); CP (trc/+)	++	++/+++	+	trc	trc	D (+); V, CP (trc/+); A, G (trc)
AAC3A[2]		trc/+	+++	trc	trc	trc	P (+); Hc, E (trc); CP (trc/+)					(a)	
AAC4A[2]		trc/+	+++	trc	-	trc	P (trc/+); Hc, E (trc); CP (+)					(a)	
LIP1A	LIP (1958)	+++	++	+	trc	-	Cl (trc/+)	++	+++	+/++	+	trc	V, CP (trc)
LIP9A		++/+++	++	++/+++	+	trc	Cl (trc); V (?); CP (vtg/+)	+/++	++/+++	+	trc	trc	; V (?); A (trc/+); CP (+/++)
EUA53-2A[2]	EUA53 (1970)	+++	++/+++	-	-	-	V (vtg); CP (+)	+	+++	-	-	-	V (+); CP (+/++)
EUA53-2B		+++	++	++	+/++	trc	Cl, CP (trc/+); Mc (trc)	++	++/+++	+/++	+	trc/+	; CP (+); Mc (trc)
EUA53-3A[2]		trc	+++/++++	-	-	-	Go (?); D, CP (trc); A (trc/+); Mc (?)	+	+++/++++	-	-	-	Go, CP (trc); D (trc/+); A (+); Mc (?)
EUA53-3B		+++	++	+++	+/++	trc	Cl (trc); CP (trc/+); Mc (?)	++	++	++	+	trc	; CP (trc/+); Mc (?)
EUA53-4A[2]		trc	+++/++++	-	-	-	V, CP (trc)					(a)	
EUA53-4B		+++	++	++/+++	+/++	trc	Cl (trc); CP (?)	+/++	+++	trc/+	+	trc/+	A, V, Mc (?); CP (trc/+)
FCG4A	FCG (1975)	+++	++/+++	++/+++	+/++	trc	Cl, CP (trc)	+/++	+++	+	+	trc/+	CP, Alh (trc); A, Mc (?)
JRP2A	JRP (1987)	++/+++	++/+++	+/++	+	-	Cl, Mc, CP (trc); P (+); E (?)	+/++	++	+/++	+	trc	Hc, Mc (trc); E (trc/+); CP (+); P (++)
UNL3A	UNL (2002)	++/+++	++	++/+++	+	-	Cl (?); CP (trc)	++	++/+++	trc	trc/+	-	CP (trc/+)

Notation used in XRD peak analysis: ++++ Very high proportion (predominant compound), +++ High proportion, ++ Medium proportion, + Weak proportion, trc Traces, - Not detected; [1] A — Aragonite; Alh — Calcium aluminate hydrate; Cl — Clinochlore; CP — Clinker Portland anhydrous compounds (C_3S, C_2S, C_4AF); D — Dolomite; E — Ettringite; G — Gypsum; Go — Goethite; H — Hematite; Ha — Halite; Hc — Hydrocalumite; Mc — Monocarboaluminate; P — Portlandite; V — Vaterite; a) not performed due to the small amount of sample available; [2] – stone-imitation mortar; (a) — not available amount of calcium in the binder, which can then be converted to $Ca(OH)_2$ (Middendorf et al. 2005b), that all of the CO_2 came from the decomposition of $CaCO_3$ in the lime mortar. For this purpose, Equation (1) was applied to calculate the percentage by mass of the dry lime content originally used, which corresponds to the binder part, assuming all of the $CaCO_3$ present in the sample was converted to $Ca(OH)_2$ during the reaction.

$$Ca(OH)_2 = CO_2 \times \frac{MM\left[Ca(OH)_2\right]}{MM(CO_2)} \quad (1)$$

Where:

1. $Ca(OH)_2$ = Hydrated lime (HL) content in percentage by mass.
2. CO_2 is the obtained by TGA.
3. MM = Molar Mass.

For cementitious mortars, regardless of whether they contain other binders, the Portland cement content (PC) was obtained according to equation (2) (Arliguie 2007). equation (2) does not consider the binder additions, so the calculations of Portland cement as a binder were performed by default.

$$PC = 100 - [IR + LOI + ((CO_2) - 1) * 1.27] \quad (2)$$

LOI corresponds to loss on ignition determined by TGA and obtained from the mass loss between 20–1000°C.

Regarding stone-imitating mortars containing carbonates, the binder to aggregate ratio was obtained by point counting in thin sections, according to RILEM Technical Committee TC167-COM Lindqvist and Sandström (2000). Point counting was carried out using an automatic point counter from PELCON, coupled to a petrographic microscope LEICA DM 2500P. The counted points of each constituent were converted into percentages. The percentage of each constituent corresponds to the volume in percentage occupied in the thin section. The volumes were then converted into mass according to Lindqvist and Sandström (2000) and considering the bulk densities of 1500 kg/m^3 and 1800 kg/m^3 for blended lime-cement and the sole cement mortars, respectively.

Regarding mortars containing mineral additions, the b:a ratio reflects only the cement content, as it is not possible to determine the content of supplementary cementitious materials.

4. Results and discussion

The characterisation of renders and plasters, i.e., coating mortars of the building's exterior and indoors, also includes decorative stone-imitating mortars. The study of finishing white smooth plaster mortars will be presented in a separate section.

4.1. Renders and plasters

4.1.1. Study of the overall fraction and aggregates

The qualitative mineralogical composition determined by XRD in the overall fraction, which is highly influenced by the aggregates' content, showed that quartz and feldspar are the main minerals, followed by muscovite and scarce or traces of kaolinite (Table 3), despite the calcite content which is primarily related to the binder.

Clay minerals are associated with siliceous sand, as seen by optical and scanning electron microscopy (Figure 3a,b). For example, in case study AR49 (1923), clays are present in aggregate interfaces, which indicate the use of unwashed sands.

Gypsum is present in the mortars from the case studies CVT (1903) and AR49 (1923), possibly due to impurities in the raw materials used to produce the binder in those cases. However, the intentional use of gypsum cannot be ruled out in the CVT1B, AR49-6A and AR49-7B samples, where it was found in more significant proportion. After the 1944 case study, the presence of gypsum was not detected in the studied samples.

Halite, a soluble salt, is present only in the samples from the first two awarded buildings CVT (1903) and AR49 (1923), with sample CTV3B containing the highest proportion.

Anhydrous calcium silicates of Portland cement are generally present in all samples from the IRF (1938) case study onwards. Besides, hydrated Portland cement compounds such as portlandite, ettringite, hydrocalumite, calcium aluminate hydrate and monocarboaluminate have been identified. The only exception is the sample AR49-8A, where no evidence of Portland cement was found but contained a high proportion of portlandite. The occurrence of portlandite in this specific case must be attributed to the presence of a whitewashed external layer, which contaminated the sample.

Vaterite was identified in almost all samples in which Portland cement was detected. The occurrence of vaterite may be related to the carbonation of cement pastes since $CaCO_3$ can precipitate with different morphologies (vaterite, calcite, aragonite), depending on the environmental conditions (e.g. temperature, humidity) and the chemical composition of the pore solution (Shtepenko et al. 2006). Vaterite has also been identified in an air lime mortar (DN12D), possibly due to the dissolution-precipitation processes (Grilo et al. 2014).

Aragonite occurs in some samples from the IRF (1938), AAC (1944) and EUA53 (1970) case studies. In *Marmorite* samples AAC2A and EUA53-3A, its presence can be related to the type of limestone used as

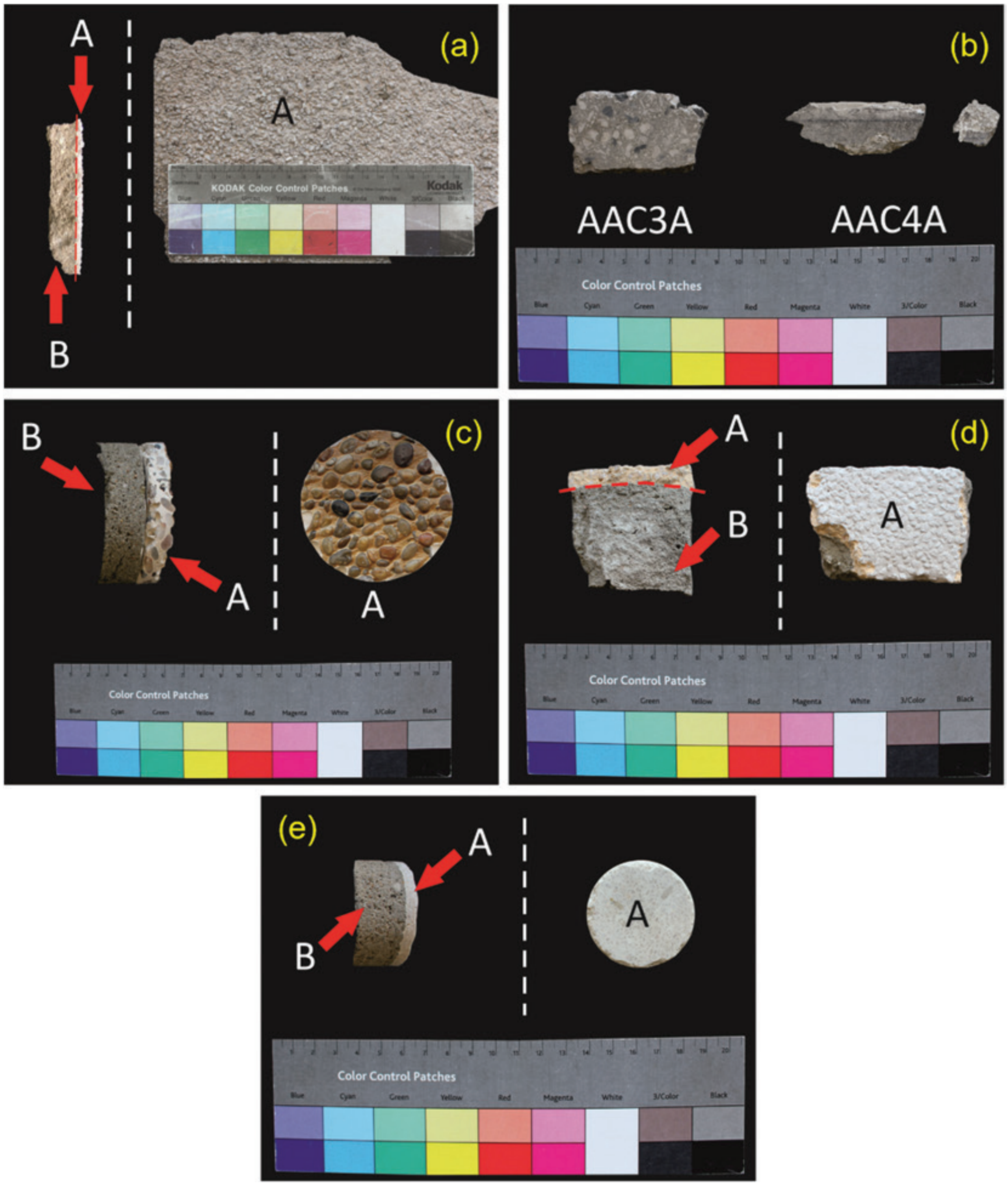


Figure 2. Multi-layered samples with stone-imitating mortars as finishing layers ("A" layers). (a) AAC2A, AAC2B; (b) AAC3A, AAC4A; (c) EUA53-2A, EUA53-2B; (d) EUA53-3A, EUA53-3B; (e) EUA53-4A, EUA53-4B.

aggregate, since dolomite, a carbonate containing magnesium, also occurs in these samples.

Sample AAC1A is the only one that shows red colouration, which is attributed to the presence of hematite that must have been incorporated into the binder as a pigment.

Figure 3 shows thin-section micrographs of mortars from several case studies to illustrate the nature of the siliceous aggregates present. The aggregates are, therefore, mainly composed of subangular to subrounded monocrystalline quartz grains, although polycrystalline grains could be present. Feldspar and muscovite grains are also visible.

Figure 4 shows selected micrographs of the so-called *Marmorite* mortars. Although this coating is quite durable (Martinho, Veiga, and Faria 2018; Veiga et al. 2019), sometime in the 20th century, its proper maintenance stopped, and paintings started to be applied massively over the existing stone-imitating mortars, which led to loss of character. Sample EUA53-3A is an example of this practice, where a painting was applied over the mortar (Figure 4g).

Marmorite mortars have a high proportion of their aggregate content of a carbonate nature. These include crushed marble and crystalline limestone aggregates. In the case study EUA53 (1970), the presence of a

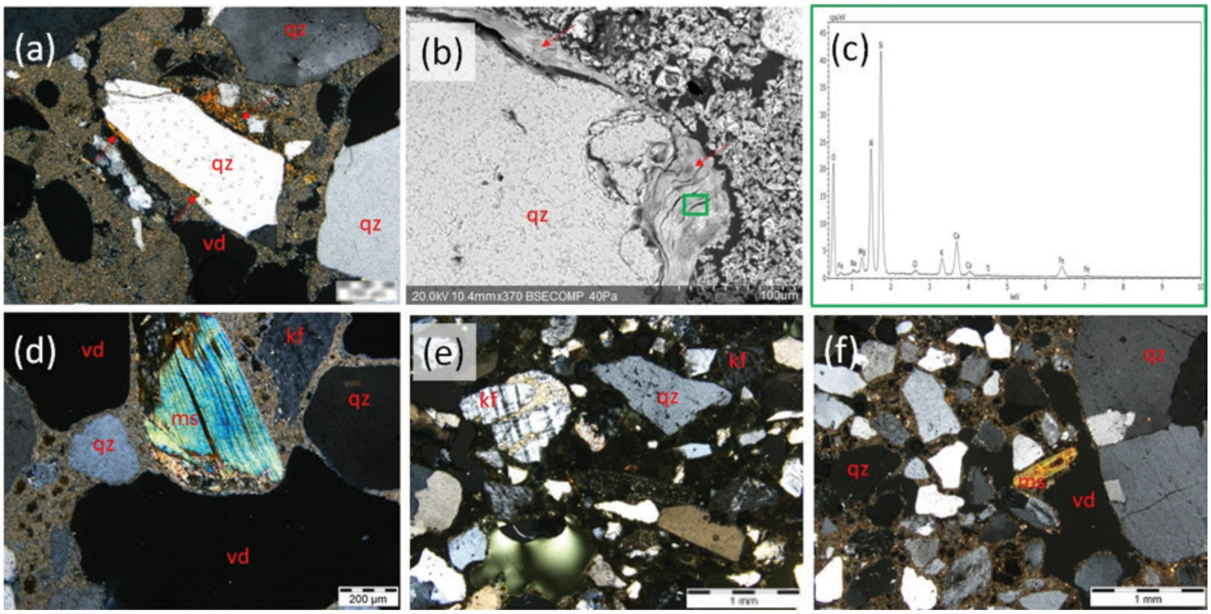


Figure 3. Thin sections micrographs in crossed polarised light (a, d, e, f) and a detail of the sample showed in (a) in backscattered mode at SEM (b). Sample AR49-15C (a, b, and c) and EDS spectrum (c) of clays around the edge of a quartz grain, area marked in (b). Samples CBP4 (d); EUA53-2C (e); UNL3A (f). Notation: qz – Quartz; kf – K-feldspar; ms – Muscovite; vd – voids; red arrows – clay minerals.

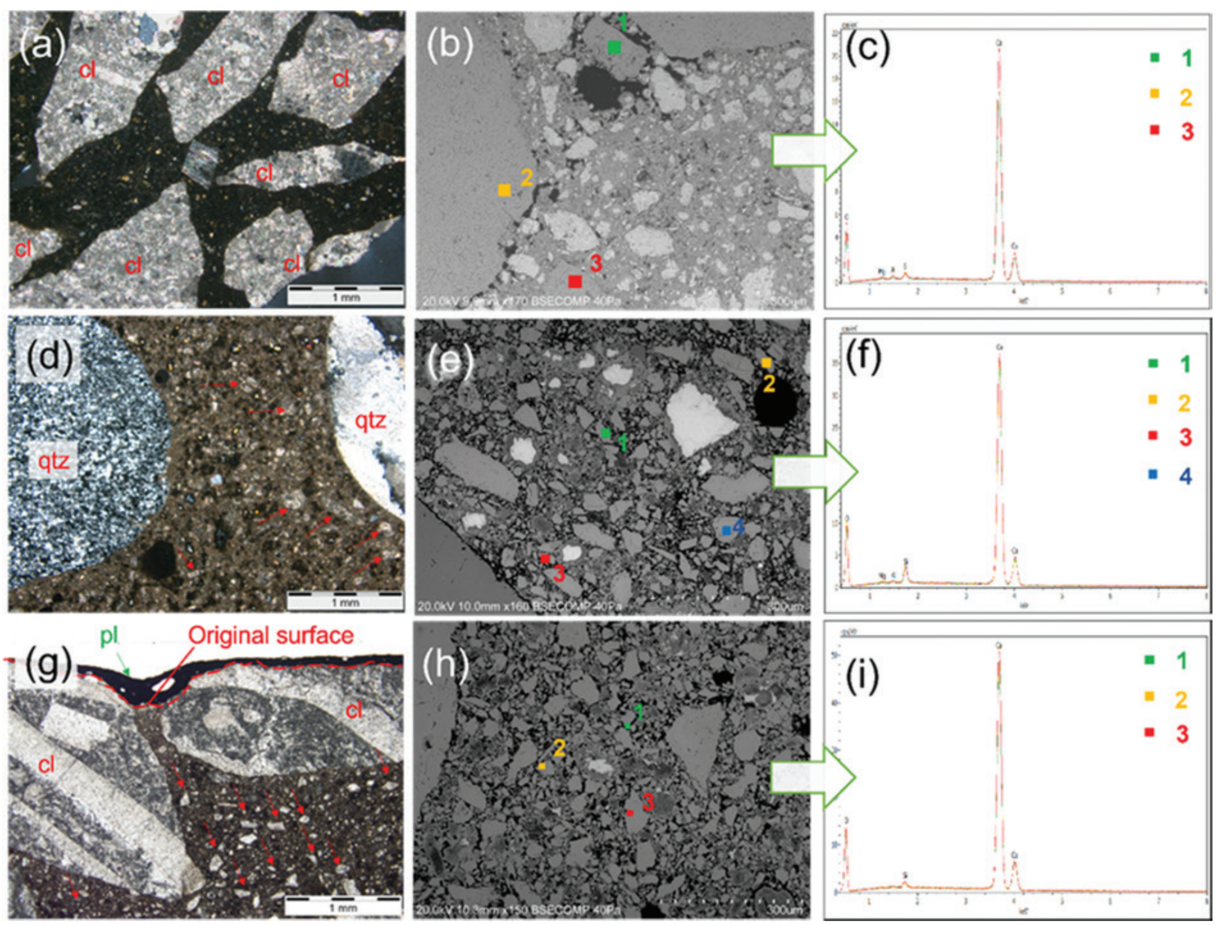


Figure 4. Micrographs of stone-imitating mortars. Samples AAC4A (a, b), EUA53-2A (d, e), and EUA53-3A (g, h). Thin sections in crossed polarised light (a, d) and in plane polarised light (g). SEM backscattered images on the middle column (b, e and h) in selected zones of the same samples displayed in the left-side column; the light grey grains correspond to cement residues. EDS spectra on the right-side column of the random limestone fine aggregates whose elemental composition highlights the Ca content. Notation: cl - crystalline limestone (often sparitized); qtz – quartzite aggregates; red arrows – indication of fine limestone aggregates (limestone/marble dust); pl - painting layer.

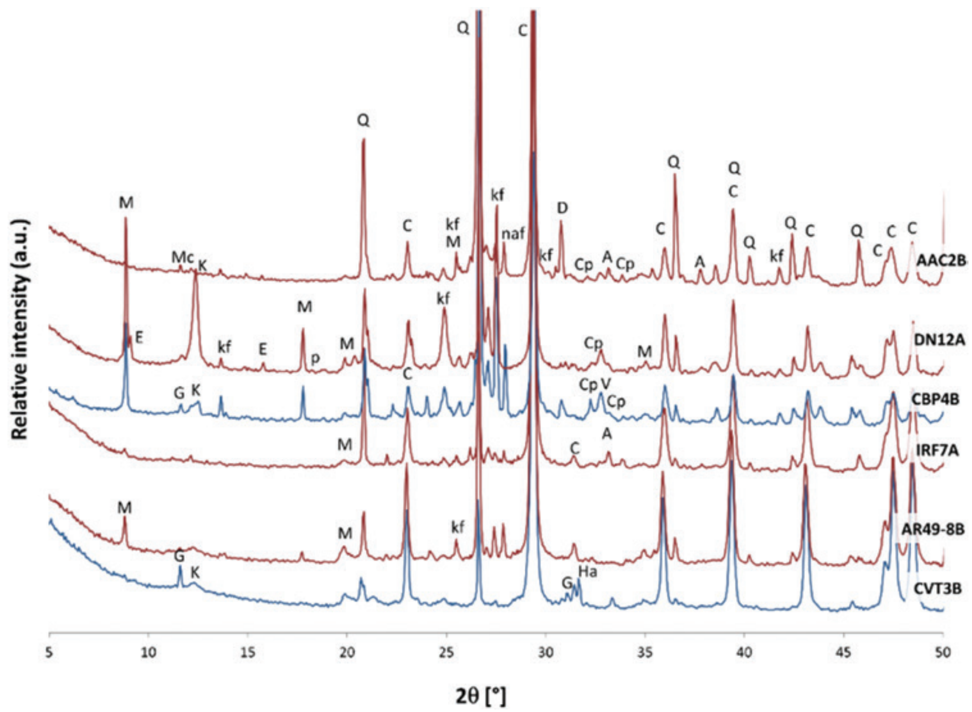


Figure 5. XRD patterns of the binder-rich fraction of the samples from the 1st half of the 20th century buildings. Notation: M – Muscovite; E – Ettringite; Mc – Monocarboaluminate; G – Gypsum; K – Kaolinite; Kf – Microcline; Q – Quartz; naf – Albite; C – Calcite; Ha – Halite; a – Aragonite; Cp – Clinker Portland anhydrous compounds (C_3S, C_2S, C_4AF); V – Vaterite.

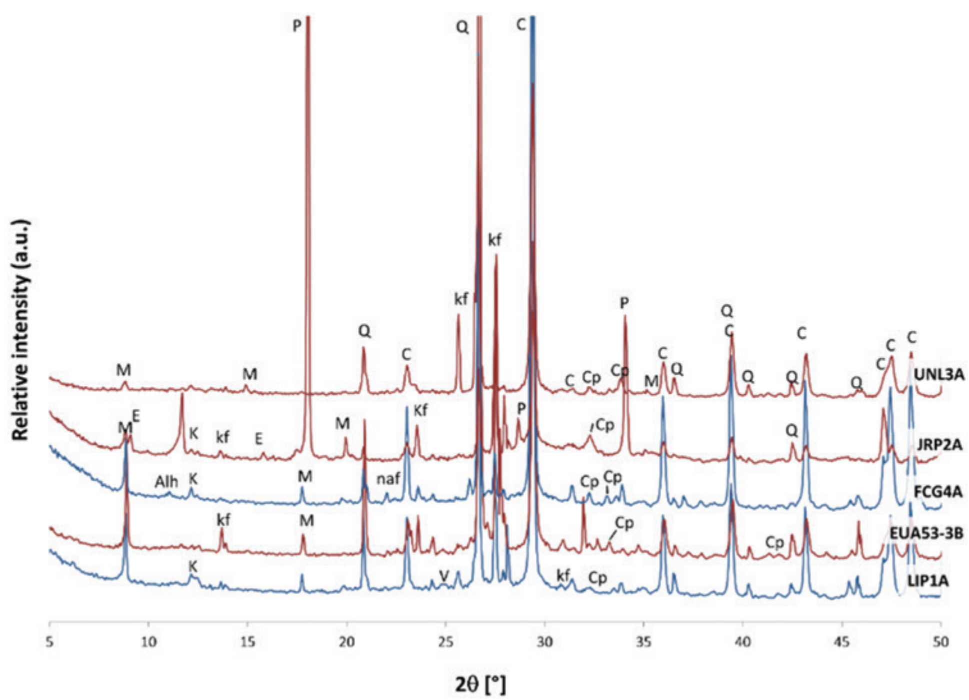


Figure 6. XRD patterns of binder-rich fraction of the samples from the 2nd half of the 20th century buildings. Notation: M – Muscovite; Mc – Monocarboaluminate; K – Kaolinite; Alh - Calcium aluminate hydrate; Kt – Katoite; P – Portlandite; E – Ettringite; Kf – Microcline; Q – Quartz; naf – Albite; C – Calcite; Cp – Clinker Portland anhydrous compounds (C_3S, C_2S, C_4AF); V – Vaterite.

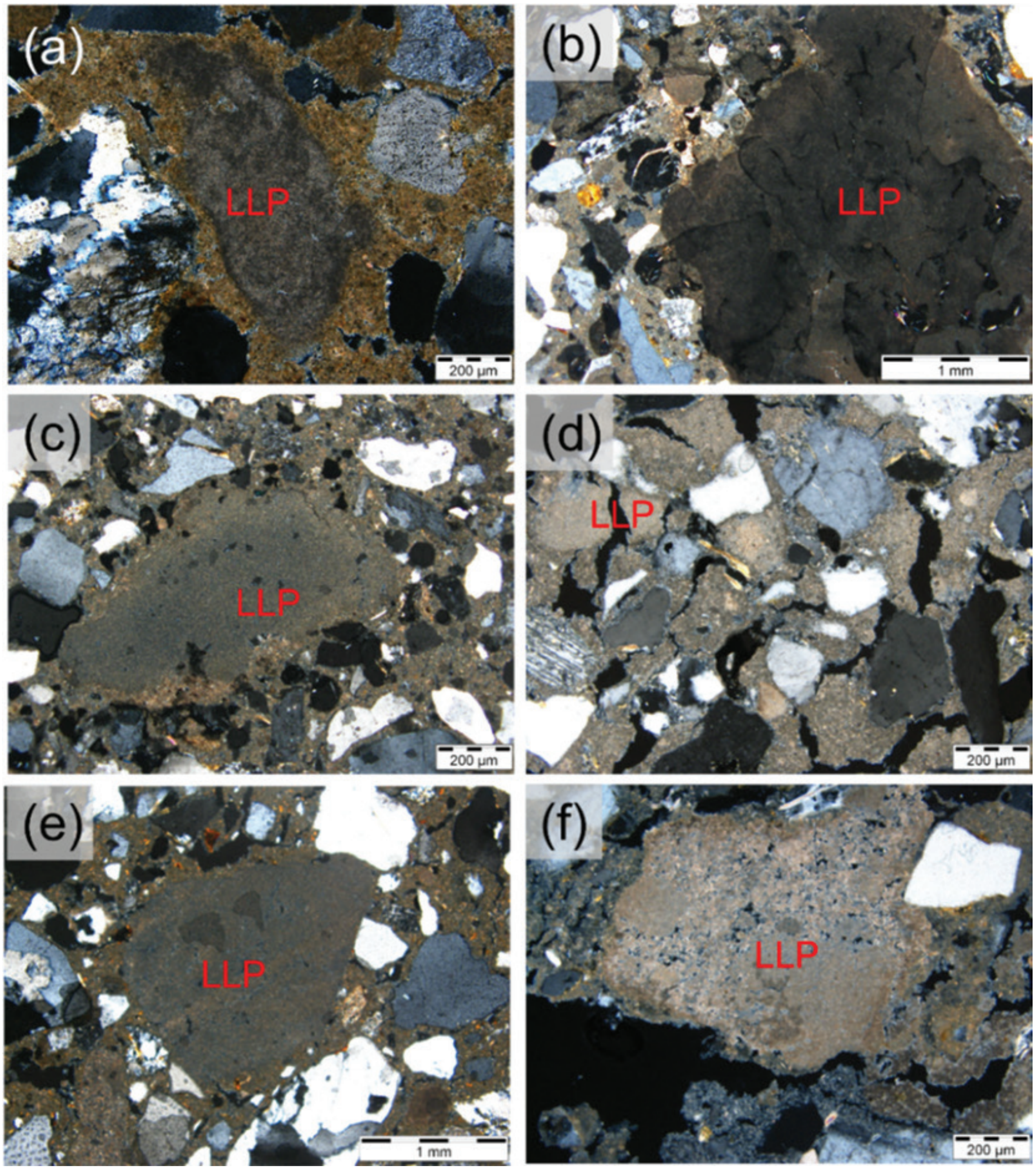


Figure 7. Micrographs of air lime mortars displaying lime lumps (LLP). Thin sections in cross-polarised light from the case studies CVT (1903) to DN (1940). Sample CVT3A (a); sample CVT1C (b); sample AR49-6C (c); sample CBP6B (d); sample IRF3B (e); sample DN12D (f).

limestone filler is clear, as seen in Figure 4d–i. In contrast, no clear evidence of limestone presence was observed in the *Marmorite* mortar from the AAC (1944) case study (Figure 4a). The binder paste is homogeneous and almost isotropic, not revealing the existence of fine aggregate in high proportion as in its counterparts from the 1970 awarded building. Sample EUA53-2A, however, does not contain calcareous sand but rounded quartzite aggregates, being the only sample that contains aggregates of this lithology. Backscattered SEM images (Figure 4b,e,h) confirm the increment in limestone aggregate from 1944 to the 1970 case study samples. In contrast, the cement paste decreased, as can be seen by the decrease of the light grey grains which correspond to the cement residues. Typically, mineral fillers are added as a substitute for cement or aggregate in mortar production, as incorporating such fine fillers can improve mortar performance (Cepuritis et al. 2014; Korjakins et al. 2008; Li et al. 2019; Mňahončáková et al. 2008; Peng and Jacobsen 2013). In the case of the *Marmorite* mortars from the EUA53 (1970) case study, it can be verified that the fine limestone grains have similar chemical composition regardless of the nature of the sand (Figure 4f,i).

4.1.2. Study of binders

The qualitative mineralogical composition determined by XRD (Table 3) in the binder-rich fraction showed that

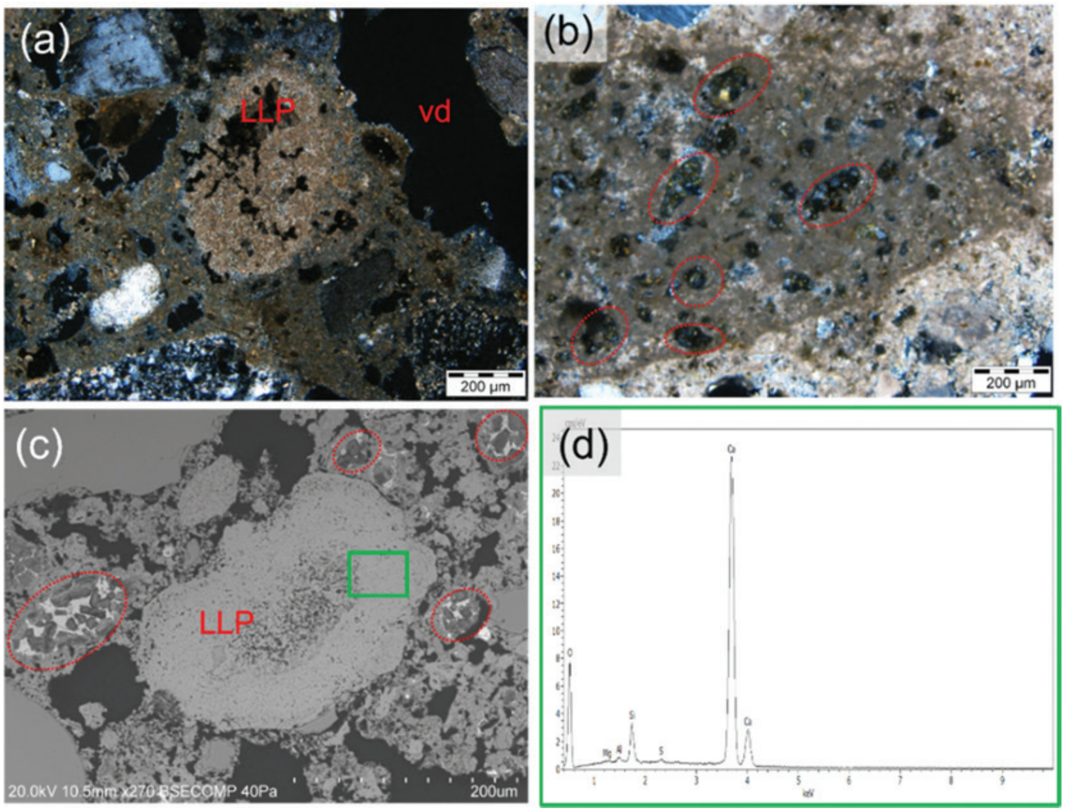


Figure 8. Lime-Portland cement mortar from IRF (1938) case study. Thin section micrographs in cross-polarised light of IRF7B sample (a) and (b); backscattered SEM micrograph (c) and EDS spectrum (d) of a lime lump (LLP) area marked in (c) by a green square. Notation: vd – air void; red dotted circle-shape figures – Portland cement clinker grains.

calcite is the main mineral in almost every analysed sample. Calcite is present mainly due to the carbonation of lime since scarce or none carbonate aggregates were found, except for the *Marmorite* mortars. The occurrence of unslaked lime in the form of nodules (lime lumps) marks a trail of air lime used as a binder, often detectable macroscopically. Non-hydrated hydraulic phases (e.g. alite-C_3S, belite-C_2S or brownmillerite-C_4AF), typically from Portland cement clinker (Lea 1988), were detected in several samples, notably from the IRF (1938) case study onward (Table 3, Figures 5 and 6), as well as ettringite, hydrocalumite, monocarboaluminate and portlandite.

However, some of these anhydrous and hydrated minerals can be associated with other hydraulic binders, namely natural cement and hydraulic lime (Lea 1988; Weber et al. 2007; Zacharopoulou 2009). The combination of mineralogical and microstructural characterisation is essential to clear up any doubts. It is known that the occurrence of certain crystalline phases detected in XRD allows differentiating the types of hydraulic binders. Gehlenite (C_2AS) can be considered as an indicator for lower burning temperatures with a stability range between 900°C and 1150°C (Callebaut et al. 2001), as well as wollastonite (CS), another anhydrous phase typical of a moderate firing of natural cement (Bouichou et al. 2019). C_2AS is possibly formed as an intermediate compound during the production of Portland clinker but does not occur in the final product (Taylor 1990). As the natural hydraulic lime (NHL) is calcinated at moderate temperatures (<1250°C), gehlenite is still present (Callebaut et al. 2001). Besides, it is common both in hydraulic lime and in natural cement, low or non-reactive compounds assembled in binder-related particles (lumps or nodules) as a result of a low calcination temperature. Most of these compounds are of non-crystalline nature, comprising solid solution silicate phases as well as crystalline CS, coarse C_2S and C_2AS (Gadermayr, Pintér, and Weber 2012). Neither gehlenite nor wollastonite was detected by XRD in the studied samples, which suggests that, among the various hydraulic binders, only Portland cement was used.

Despite the absence of these compounds, further microscopic investigation was carried out. Figures 7-11 show examples of different samples in which the microscopic analysis allowed clarifying the type of binder used.

The binder-rich fraction contains kaolinite, mainly in trace amounts. It also contains muscovite, although in more significant quantities than kaolinite. The sample

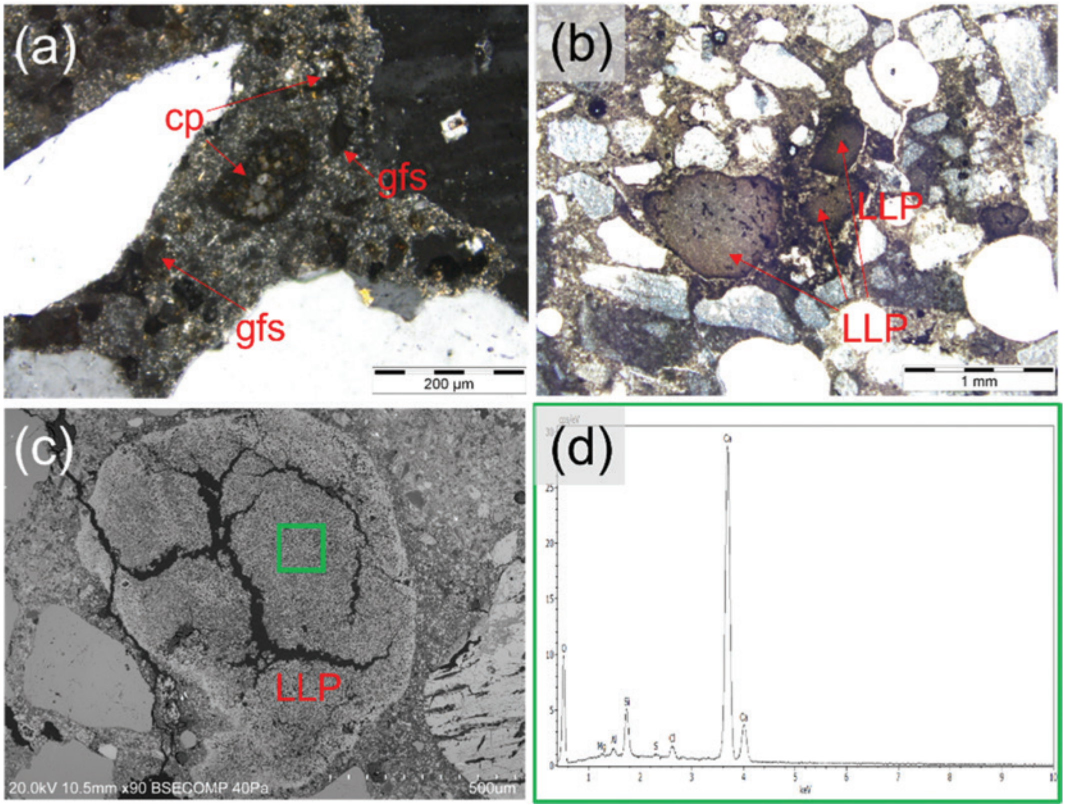


Figure 9. Lime-Portland cement mortar from DN (1940) case study. Thin section micrographs of DN19D sample: cross polarised light (a), and plane polarised light (b); backscattered SEM micrograph (c) and EDS spectrum (d) of a lime lump (LLP) marked in (c) by a green square. Notation: cp – unhydrated cement grains; gfs – granulated blast furnace slags.

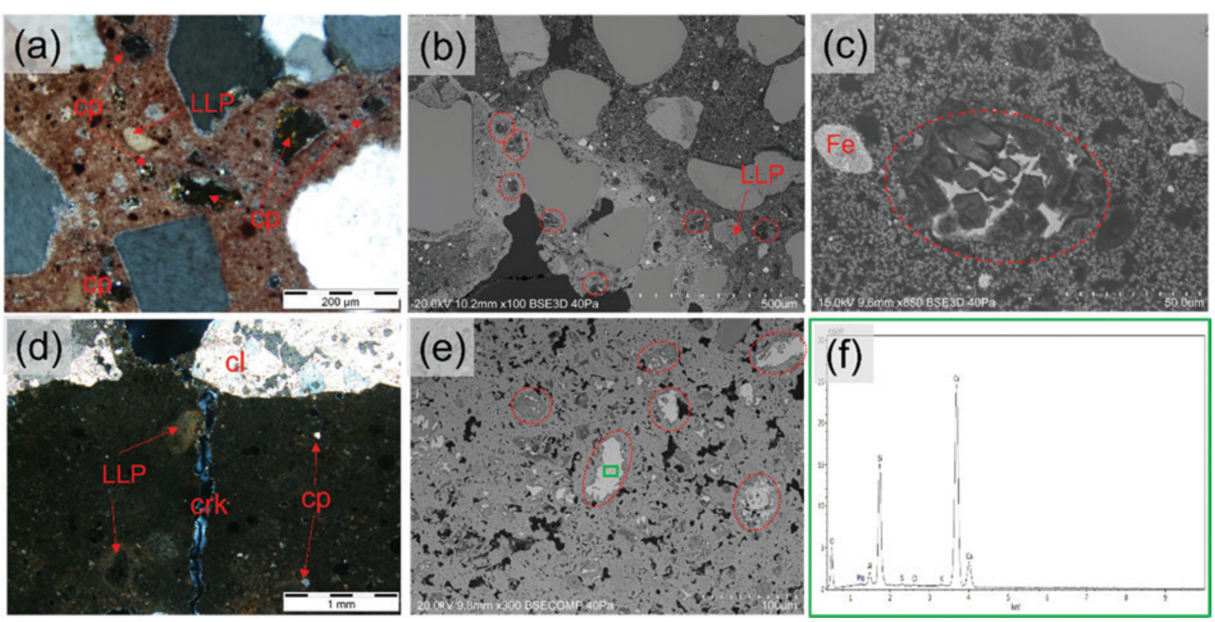


Figure 10. Lime-Portland cement blended mortar from AAC (1944) case study. Thin section micrograph of the red-pigmented sample AAC1A in cross polarised light (a), and at SEM in backscattered mode (b, c). Thin section micrograph of sample AAC2A in cross polarised light (d), and in backscattered mode (e). SEM micrographs (b) and (c) displaying calcium-depleted cement grains inside the red dotted circle-shape figures and the largest unhydrated cement grains in (e). EDS spectrum (f) of a C_3S phase marked in (e) by a green square. Notation: cp – Portland cement grains; LLP – lime lumps; Fe – Iron-rich grain (hematite); cl – crystalline limestone; crk – crack.

preparation methodology is relevant since the samples are manually disaggregated with a rubber hammer to avoid breaking the aggregates to obtain small lumps, which are then sieved in the 106 μm sieve. Since micas are soft minerals with evident cleavage, they are easily breakable and transposable through the sieve mesh. The mesh size allows the passage of particles such as very fine sand, silt, and clay particles according to the sedimentological classification of Wentworth (1922) concerning particle diameter. Minerals from the mica and clay groups can therefore also be expected to occur in the binder-rich fraction.

The microscopic analyses proved that rendering mortars and plasters applied in CVT (1903) and AR49 (1923) case studies are of aerial nature, i.e., no pozzolanic additions or hydraulic compounds were observed. To corroborate this assumption regarding the nature of the binder, Figure 7 shows several examples of binder residues as unslaked lime lumps. Figure 7d also shows a remarkable shrinkage cracking microstructure pattern, often visible in air lime mortars.

Case study IRF (1938) also contains air lime-based plasters; however, it was the first case study where Portland cement was found blended with air lime. Lime-Portland cement mortars have many Portland cement clinker residues composed of belite and alite. Belite presents rounded crystals and interference amber to grey colours, whilst alite often presents grey to black interference colours with hexagonal crystals (Figures 8b,9a,11b). Calcium depletion of Portland cement clinker unhydrated phases (Shtepenko et al. 2006; Weber et al. 2015) has occurred due to carbonation. It evolved the replacement of residual clinker particles by calcium silicate hydrate (C-S-H) of low Ca/Si ratio, which approaches hydrous silica, yet still preserves the crystals' morphology (e.g. Figure 10c). In some cases, the

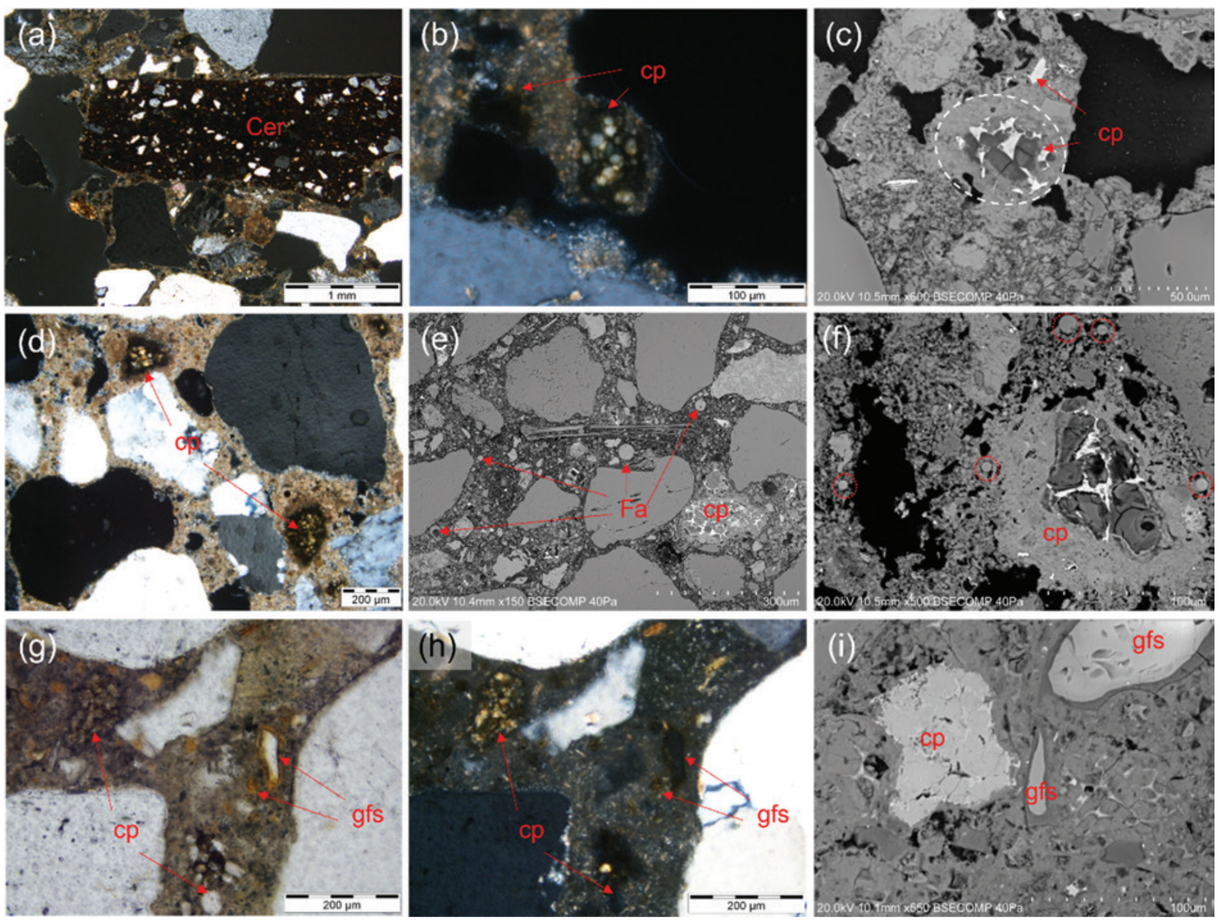


Figure 11. Optical microscopy and SEM images of samples mortars from CBP (1938), DN (1940) and JRP (1987) case studies. Thin section micrographs in cross polarised light of sample CBP4B showing the presence of ceramic fragments and Portland cement clinker grains (a, b); Backscattered SEM image (c) showing Portland cement clinker grain affected by carbonation (white dotted circle); Thin section micrograph in cross polarised light of sample DN19B (d) showing Portland cement clinker grains; Backscattered SEM micrographs (e, f) highlighting the presence of fly ash (red arrows and red dotted circles); Thin sections micrographs of sample JRP2A in plane polarised light (g) and in cross polarised light (h) displaying Portland cement clinker grains and granulated blast furnace slags; SEM image (i) with high magnification of the same sample. Notation: Cer – Ceramic aggregate; cp – Portland cement clinker grain; Fa – Fly ash; gfs – Granulated blast furnace slag.

calcium depletion phenomenon can be seen through the black interference colours inside the Portland clinker residues, which leads to quasi-isotropic behaviour in optical microscopy under crossed polarised light (Figure 10a).

Air lime-Portland cement mortars can also be found in renders and plasters from the DN (1940) and AAC (1944) case studies (Figures 9 and 10) and with the same distinctive features. The lime lumps present have calcium-rich compositions with low impurities, as exemplified by the EDS spectra in Figures 8d and 9d.

CBP (1939) case study is the first one in the chronological scale of the buildings studied to show solely Portland cement mortars. In this context, AAC (1944) is the last to contain air lime-Portland cement blended mortars.

All the *Marmorite* mortars — from case studies AAC (1944) and EUA53 (1970) — were formulated with Portland cement. The sample AAC2A, which has a superficial layer of projected crushed crystalline limestone onto the binder paste after its application (Figure 10d), is the only stone-imitating mortar to incorporate lime and Portland cement.

The *Marmorite* samples from the case study EUA53 (1970) showed unhydrated cement Portland residues containing no tetra-calcium aluminoferrite (C_4AF) phase (Figure 10e), a feature consistent with the use of white Portland cement.

Composite Portland cement mortars were identified in two case studies, namely DN (1940) and JRP (1987) (Figure 11). The presence of fly ash in sample DN19B points to a mortar not contemporary with the building's construction (1936–1940, see Table 1). It should be noted that fundamental studies of fly ash mixed with Portland cement were carried out in 1937 (Davis, Carlson, and Kelly 1937) and that these additions only began to be applied in Portugal in the 1980s (Coutinho 2012). On the other hand, sample DN19D incorporates GGBFS, an addition that began to be produced in Portugal in the 1960s; it is expected that this sample is also not contemporary with the construction unless the cement applied was imported, which was not possible to clarify. Thus, the whole set constituted by the samples DN19A, DN19B, DN19C, and DN19D, may be considered non-contemporary with the construction and possibly the result of a refurbishment of which record could not be found.

It should also be noted that the first use of white cement was found in a stone-imitating mortar, AAC2A. Since the construction period of the respective building was before the beginning of the production of white cement in Portugal, it is evident that the cement applied was imported. The CBP4B sample from the CBP case study (1939) is also noteworthy as it incorporates ceramic aggregates in its composition (Figure 11a,b,c).

4.1.3. Binder to aggregate ratio and chemical analyses

Tables 4 and 5 present the chemical constituents analysed and the binder and aggregate content of the renders, plasters, and stone-imitating mortars.

In order to verify the existence of a trend of increase or reduction of the aggregates' content in plasters and renders over time, Figures 12, 13 present the evolution over the period under analysis of the aggregate portion in the mix, which corresponds to the denominator of the b:a ratio (Table 4). The plasters' aggregate portion in the mix does not show a clear trend. However, a very slight tendency of increasing the aggregate portion over the studied period for the outermost or monolayered mortars is shown (Figure 12), which is considered to be dependent on the higher content of the aggregates in the samples CBP4B (aggregate portion = 20.3) and DN19B (aggregate portion = 25.2). In CBP4B, the increase in the mass of aggregates is due to the incorporation of

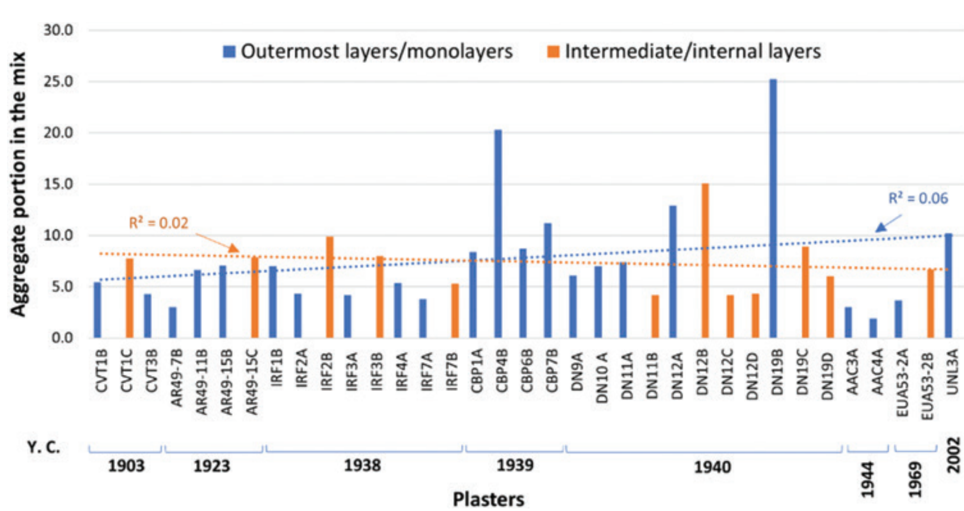


Figure 12. Aggregate portion in the mix (the denominator for the b:a ratio) of plasters over the studied period. Notation: Y. C. – building's year of completion; blue dotted line — linear regression for outermost layers/monolayers; orange dotted line — linear regression for intermediate/internal layers.

Table 4. Constituents (wt%) and binder to aggregate ratio of renders and plasters.

Case study	Sample Id.	BL	Binder type[i]	IR	CO_2 [ii]	HL	LOI	PC[iii]	SiO_2 Soluble	Na_2O equivalent[iv]	Chlorides (Cl^-)	Sulphates (SO_3)	b:a[v]
CVT (1903)	CVT1B	I	AL	72.788	7.99	13.45	8.43	(b)	0.19	0.07	0.03	3.60	1 : 5.4
	CVT1C	I	AL	80.16	6.13	10.32	11.08	(b)	0.35	0.87	0.54	0.08	1 : 7.8
	CVT3B	I	AL	75.26	10.48	17.64	6.36	(b)	0.19	0.48	0.07	0.10	1 : 4.3
AR49 (1923)	AR49-6C	E	AL	73.54	7.53	12.68	12.17	(b)	0.41	0.45	0.09	2.93	1 : 5.8
	AR49-7B	I	AL	62.62	12.42	20.91	7.64	(b)	0.37	0.94	0.37	0.89	1 : 3
	AR49-8A	E	AL	62.40	12.86	21.65	10.72	(b)	0.40	0.08	0.02	0.13	1 : 2.9
	AR49-8B	E	AL	83.86	4.46	7.51	13.69	(b)	0.44	0.06	0.03	0.08	1 : 11.2
	AR49-11B	I	AL	80.53	7.19	12.1	10.74	(b)	0.24	0.84	0.10	0.11	1 : 6.7
	AR49-15B	I	AL	69.81	5.85	9.85	6.73	(b)	0.09	0.31	0.06	0.14	1 : 7.1
	AR49-15C	I	AL	82.38	6.21	10.45	4.92	(b)	0.15	0.33	0.06	0.15	1 : 7.9
IRF (1938)	IRF1B	I	AL+OPC	82.57	6.97	11.73	6.56	1.42	0.47	0.07	0.02	0.08	1 : 0.1 : 7
	IRF2A	I	AL	74.34	10.17	17.12	5.47	(b)	0.27	0.61	0.02	0.47	1 : 4.3
	IRF2B	I	AL	85.43	5.11	8.6	11.09	(b)	0.35	0.36	0.05	0.10	1 : 9.9
	IRF3A	I	AL	74.07	10.53	17.73	10.90	(b)	0.26	0.06	0.06	0.16	1 : 4.2
	IRF3B	I	AL	83.70	6.19	10.42	9.97	(b)	0.47	0.32	0.04	0.20	1 : 8
	IRF4A	I	AL+OPC	74.73	8.28	13.94	10.04	5.3	0.80	0.12	0.02	0.12	1 : 0.4 : 5.4
	IRF7A	I	AL+OPC	67.30	10.65	17.93	6.28	6.75	1.16	0.15	0.01	0.19	1 : 0.4 : 3.8
	IRF7B	I	AL+OPC	74.85	8.38	14.11	10.21	5.04	0.97	0.12	(c)	0.09	1 : 0.4 : 5.3
CBP (1939)	CBP1A	I	AL	83.66	5.92	9.97	10.04	(b)	0.10	0.07	0.06	0.24	1 : 8.4
	CBP4B	I	OPC	87.76	3.35	(b)	11.47	4.33	0.67	0.10	0.06	0.20	1 : 20.3
	CBP6B	I	AL	85.89	5.88	9.9	6.19	(b)	0.25	0.06	0.04	0.21	1 : 8.7
	CBP7B	I	AL	87.87	4.66	7.85	9.39	(b)	0.21	0.06	0.11	0.10	1 : 11.2
DN (1940)	DN9A	I	OPC	68.71	8.02	(b)	9.20	11.29	1.97	0.17	0.03	0.62	1 : 6.1
	DN10 A	I	OPC	69.83	8.29	(b)	9.55	10.02	1.42	0.17	0.04	0.38	1 : 7
	DN11A	I	OPC	72.23	7.36	(b)	8.46	9.72	1.71	0.16	0.01	0.38	1 : 7.4
	DN11B	I	OPC	66.92	6.65	(b)	36.45	15.86	3.24	0.10	0.03	0.23	1 : 4.2
	DN12A	I	OPC	83.73	3.75	(b)	12.01	6.50	1.12	0.07	0.01	0.05	1 : 12.9
	DN12B	I	AL+OPC	76.74	3.02	5.08	37.98	10.49	2.63	0.04	0.01	0.03	1 : 2.1 : 15.1
	DN12C	I	OPC	69.60	3.95	(b)	35.03	16.61	2.11	0.17	0.04	0.49	1 : 4.2
	DN12D	I	AL	72.57	9.94	16.73	8.36	(b)	0.20	0.76	0.01	0.09	1 : 4.3
	DN19B	I	PCC	83.13	6.81	(b)	9.41	3.30 (a)	1.02	0.12	(c)	0.15	1 : 25.2 (a)
	DN19C	I	OPC	74.55	7.05	(b)	12.81	8.37	1.30	0.17	0.02	0.28	1 : 8.9
	DN19D	I	AL+PCC	77.83	7.75	13.05	6.95	4.39 (a)	1.04	0.06	0.02	0.19	1 : 1 : 6
AAC (1944)	AAC1A	E	AL+OPC	79.56	7.75	13.05	42.08	2.32	0.25	0.06	(c)	0.05	1 : 0.2 : 6.1
	AAC1B	E	AL+OPC	81.37	6.94	11.68	12.36	2.62	0.19	0.08	(c)	0.07	1 : 0.2 : 7
	AAC2B	E	OPC	74.67	9.09	(b)	8.18	3.05	1.45	0.34	0.01	0.10	1 : 24.5
LIP (1954)	LIP1A	E	OPC	75.5	5.89	(b)	10.32	9.93	0.59	0.20	0.03	0.08	1 : 7.6
	LIP9A	E	OPC	72.13	6.97	(b)	8.43	10.88	1.79	0.13	0.04	0.19	1 : 6.6

(*Continued*)

Table 4. (Continued).

Case study	Sample Id.	BL	Binder type[i]	IR	CO_2 [ii]	HL	LOI	PC[iii]	SiO_2 Soluble	Na_2O equivalent[iv]	Chlorides (Cl^-)	Sulphates (SO_3)	b:a[v]
EUA53 (1970)	EUA53-2B	I	OPC	76.65	4.85	(b)	6.36	11.51	1.53	0.17	0.01	0.31	1 : 6.7
	EUA53-3B	E	OPC	64.27	9.11	(b)	7.64	13.07	0.81	0.20	0.01	0.38	1 : 4.9
	EUA53-4B	I	OPC	78.39	6.21	(b)	13.69	6.82	1.15	0.13	0.01	0.11	1 : 11.5
FCG (1975)	FCG4A	E	OPC	77.5	6.47	(b)	10.74	7.59	0.51	0.44	(c)	0.15	1 : 10.2
JRP (1987)	JRP2A	E	PCC	74.74	2.64	(b)	6.73	15.30 (a)	3.17	0.06	0.01	0.51	1 : 4.9 (a)
UNL (2002)	UNL3A	I	OPC	73.15	8.39	(b)	4.92	7.14	3.04	0.12	(c)	0.36	1 : 10.2

Legend: BL – Building's location (exterior – E; interior/indoors – I); AL – air lime; OPC – Ordinary Portland cement; PCC – Portland composite cement; AL+OPC – blended air lime and ordinary Portland cement; IR – insoluble residue; i – the binder type was previously identified by microstructural analyses combined with XRD; ii – obtained by TGA; HL – hydrated lime ($Ca(OH)_2$) obtained by the Equation (1); iii – Portland cement content calculated according to the Equation (2); iv – alkalis content through Na_2O equivalent; LOI: loss on ignition obtained by TGA; v – binder to aggregate ratio (HL:OPC:aggregate - for air lime with by mixed ordinary Portland cement; HL:aggregate - for air lime mortars; OPC or PCC:aggregate - for cement mortars); (a) – overestimated aggregate content, since it is a mortar with mineral additions; (b) – not applicable; (c) – not detected.

Table 5. Chemical contents in percentage by weight and binder to aggregate ratios of stone-imitating mortars.

Case study	Sample Id.	BL	Binder type[i]	IR	CO_2 [ii]	$CaCO_3$ [iii]	$CaCO_3$ (from paste) [iv]	$CaCO_3$ (from aggregates) [iv]	TAA	HL [v]	LOI	PC [vi]	SiO_2 Soluble	Na_2O equivalent [vii]	Chlorides (Cl^-)	Sulphates (SO_3)	b:a[viii]
AAC (1944)	AAC2A	E	AL +WPC	15.71	30.41	69.16	48.25	20.91	36.62	41.26	41.44	10.49	0.62	0.21	0.01	0.33	1 :0.3 :1.0
	AAC3A	I	OPC	4.04	33.29	75.71	28.99	46.72	50.76	(a)	7.96	16.97	3.62	0.12	0.03	0.34	1 : 3.0
	AAC4A	I	OPC	2.80	30.70	69.82	25.10	44.72	47.52	(a)	7.88	24.45	4.99	0.13	0.01	0.47	1: 1.9
EUA53 (1970)	EUA53-2A	I	WPC	57.33	11.48	26.11	21.49	4.62	61.95	(a)	11.08	16.55	1.12	0.04	(b)	0.34	1 : 3.7
	EUA53-3A	I	WPC	4.13	40.54	92.20	27.99	64.21	68.34	(a)	12.17	3.57	0.30	0.08	0.01	0.26	1 : 19.1
	EUA53-4A	I	WPC	3.74	39.17	89.08	22.27	66.81	70.55	(a)	10.72	6.35	1.31	0.05	(b)	0.22	1 : 11.1

Legend: BL — Building's location (exterior — E; interior/indoors — I); AL — air lime; OPC — Ordinary Portland cement; WPC — White Portland cement; AL+WPC — blended air lime and white Portland cement; IR — insoluble residue; I — the binder type was previously identified by microstructural analyses combined with XRD; ii — obtained by TGA; iii — obtained by multiplying $\%CO_2$ by the ratio between the molecular masses of $CaCO_3$ and CO_2; iv — estimated by point counting and normalised for the $CaCO_3$ content obtained by TGA; TAA — total amount of aggregates = [IR + $CaCO_3$ (from aggregates)]; HL — Hydrated lime ($Ca(OH)_2$) obtained by the Equation (1); v — considering $CaCO_3$ from paste; LOI: loss on ignition obtained by TGA; vi — Portland cement content calculated by Equation (1); vii — alkalis content through Na_2O equivalent; viii — binder to aggregate ratio (HL: WPC:aggregate — for air lime with by mixed white Portland cement; OPC:aggregate — for ordinary Portland cement mortars); (a) — not applicable; (b) — not detected.

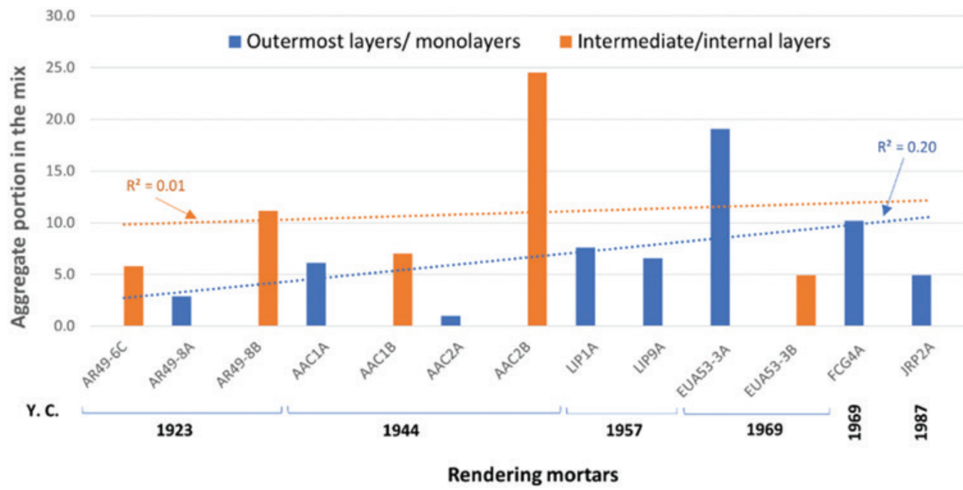


Figure 13. Aggregate portion in the mix (the denominator for the b:a ratio) of renders over the studied period. Notation: Y. C. – building's year of completion; blue dotted line - linear regression for outermost layers/monolayers; orange dotted line - linear regression for intermediate/internal layers.

ceramic elements, as shown in Figure 11a. Regarding sample DN19B, the binder may be undervalued since it contains GGBFS. In this case, the application of Equation (2) overestimates the aggregate content.

Figure 13 shows the same for rendering mortars, in this case regarding the outermost and monolayer rendering mortars. This very slight linear trend is greatly influenced by the aggregate's proportion of the EUA53-3A, which contains a limestone filler. For internal layers, there is a need to have more data to conclude about the evolution of the aggregate content over the period under analysis.

Despite the attempted interpretation presented above regarding plasters and rendering mortars, there is no evidence or statistical support to validate any trend in the evolution of the aggregates' content over the period under analysis.

Figure 14 shows the plot of soluble silica vs. alkalis content of the mortars under analysis, which corroborates the evidence of hydraulicity of the respective mortars. The evidence suggests this could be due to C-S-H in the pastes that contain Portland cement. Soluble silica has a widespread distribution between 0.3 and 5% in Portland cement mortars, while its maximum value is 0.5% in lime mortars. The alkalis could be related to the presence of feldspars, as indicated by the results of mineralogical characterisation by XRD, rather than to alkali-rich binders.

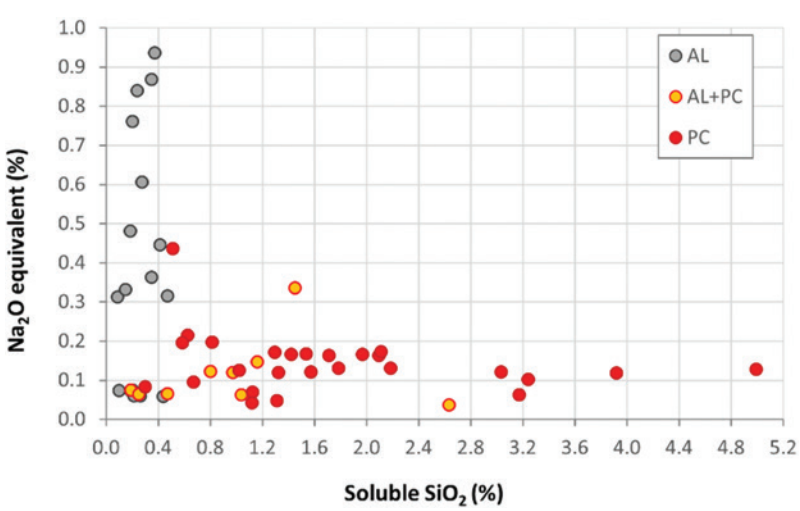


Figure 14. Content of soluble SiO_2 against Na_2O equivalent content. Notation: Al — air lime mortars; AL+PC — air lime with mixed ordinary Portland cement, including mortars with supplementary cementitious materials; PC — white and ordinary Portland cement mortars and mortars with supplementary cementitious materials.

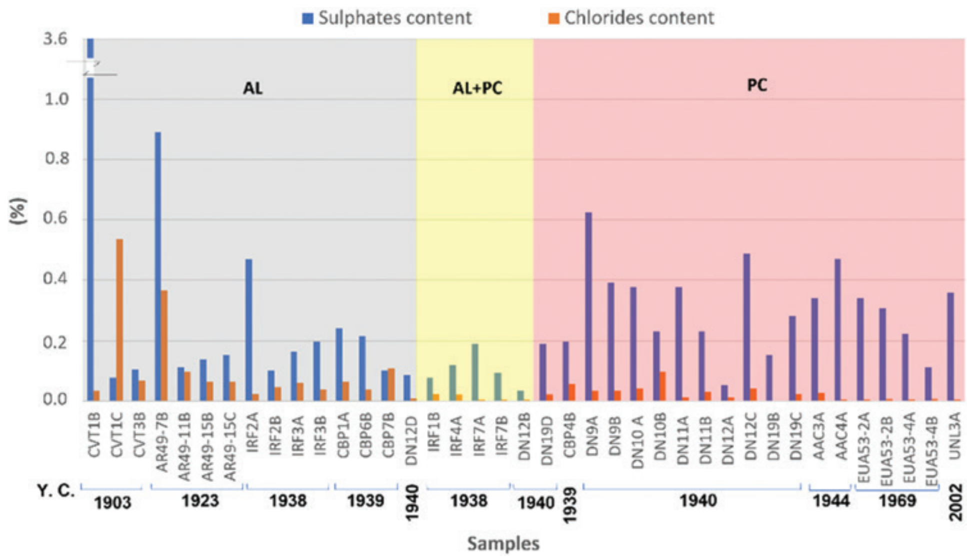


Figure 15. Sulphates and chloride content in plasters. Notation: Y. C. – building's year of completion; Al – air lime mortars; AL+PC – air lime with by mixed Portland cement, including mortars with supplementary cementitious materials; PC – white and ordinary Portland cement mortars and mortars with supplementary cementitious materials.

The potential degradation phenomena were evaluated through the content of sulphates and chlorides. The samples with the higher values in these two constituents were collected in areas where degradation had already occurred, namely in the case studies awarded in 1903 and 1923. Air lime mortars CVT1B; AR49-6C and AR49-7B contain 3.6%; 2.93% and 0.89% of sulphates, respectively, whereas CVT1C and AR49-7B contain the highest chloride content (0.54% and 0.37%, respectively). However, these chloride contents are low. The sulphate content may be related to the settlement of gypsum by water transport which affected the smooth white outermost finishing layers from the surface of the detaching zones, while the origin of chlorides is more uncertain.

Mortars containing Portland cement show values lower than 0.6% for sulphate, which is expected since calcium sulphate hydrate is used as a setting retarder.

Ageing did not produce differences during the analysed period, and the exposure to pollutants does not seem to have produced effects either (Figures 15 and 16). Considering the assessment of chloride and sulphate content, it can be concluded that all mortars are in a good state of conservation since their values are generally low.

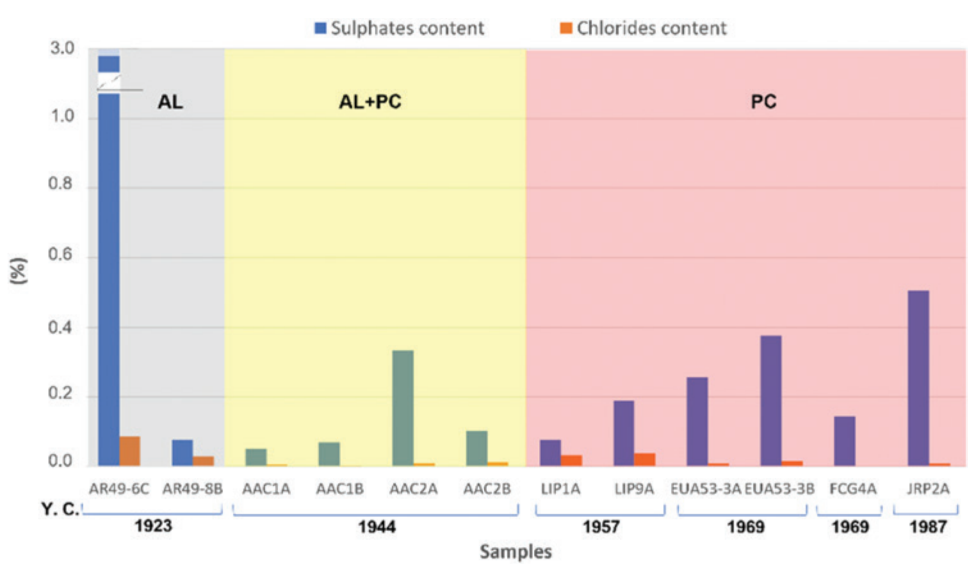


Figure 16. Sulphates and chloride content in renders. Notation: Y. C. – building's year of completion; Al – air lime mortars; AL+PC – air lime with by mixed Portland cement, including mortars with supplementary cementitious materials; PC – white and ordinary Portland cement mortars and mortars with supplementary cementitious materials.

Table 6. Qualitative XRD composition, calculated gypsum and calcite contents by TGA-DTA and gypsum to calcite ratio of the analysed samples.

		Identified crystalline compounds					Calculated contents (wt. %)			
Sample	Case study	Calcite	Gypsum	Anhydrite	Quartz	Others[1]	Gypsum	Calcite	Portlandite	Gypsum : Calcite
CVT1A	CVT (1903)	+++	trc/+	-	trc/+	F, M(trc)	1.5	74.6	-	1 : 50.4
CVT3A		++	+++	trc	trc	-	63.6	35.0	-	1 : 0.6
AR49-2A	AR49 (1923)	+++	+++	-	trc	Ha (+)	34.6	59.5	-	1 : 1.7
AR49-11A		+++	++/+++	-	trc	Sy (trc); Eu (+)	29.4	55.7	-	1 : 1.9
AR49-15A		+++	+++	-	-	-	32.4	62.5	-	1 : 1.9
CBP4A	CBP (1939)	++/+++	+++	+	trc	P (++)	45.2	39.4	5.1	1 : 0.9
CBP6A		+++	+/++	-	+	M (trc); P (+)	17.0	65.3	1.8	1 : 3.8
$CBP7A_1$		++/+++	++	trc	-	M (trc); P (+/++)	38.8	51.0	3.5	1 : 1.3
$CBP7A_2$		++/+++	++	trc	-	M (?); P (+/++)	33.3	57.8	3.1	1 : 1.7
DN19A	DN (1940)	+/++	+++	++	trc/+	M, Bs (trc); F (?)	55.4	12.0	-	1 : 0.2

Notation used in XRD peak analysis: +++ High proportion, ++ Medium proportion, + Weak proportion, trc Traces, - Not detected; [1] F – Feldspars; M – Muscovite; *P* – Portlandite; Ha – Halite; Sy – Syngenite; Eu – Eugsterite; Bs – Bassanite.

4.2. Finishing white smooth plaster layers

4.2.1. Mineralogical characterisation and microscopic observations

The qualitative mineralogical composition determined by XRD showed that gypsum and calcite are the main mineral compounds (Table 6 and Figure 17). Portlandite is present in weak to medium proportions in all CBP (1939) case study samples. The trace occurrence of quartz and other silicates, namely feldspars and muscovite minerals, aligns with Freire et al. (2019), which stated that they are relatively common in raw materials.

The presence of anhydrite ($CaSO_4$), found in some samples, can be either due to the calcination process, where a given amount of over-burnt gypsum is always present (Cardoso and Pye 2017; Sanz 2009), or/and to the raw material, as a common impurity of the gypsum deposits (Freire et al. 2019). The amounts of gypsum and calcite were calculated from the mass losses obtained by TGA-DTA (Table 5). DTA curves (Figure 17) show a peak (G1) related to the transformation of gypsum (dihydrate) into bassanite (hemihydrate), which comprises the removal of 3/4 of the water of crystallisation. In some cases, DTA curves show another peak (G2) related to losing the remaining water from hemihydrate to form soluble anhydrite (Adams, Kneller, and Dollimore 1992; Borrachero et al. 2008; SNIP 1982).

Concerning the decarbonation of calcite, the temperature range varies according to its content (referred to as C in Figure 17). Higher amounts of calcite correspond to broader intervals of 600–900°C (e.g. CVT1A, CBP6A). Besides these two significant compounds (gypsum and calcite), a peak (P) around 450°C is also perceptible (Figure 17), which corresponds to the dehydration of portlandite ($Ca(OH)_2$), corroborating the XRD data. The calcite proportion was found to be higher than gypsum, apart from samples CVT3A, CBP4A and DN19A. The

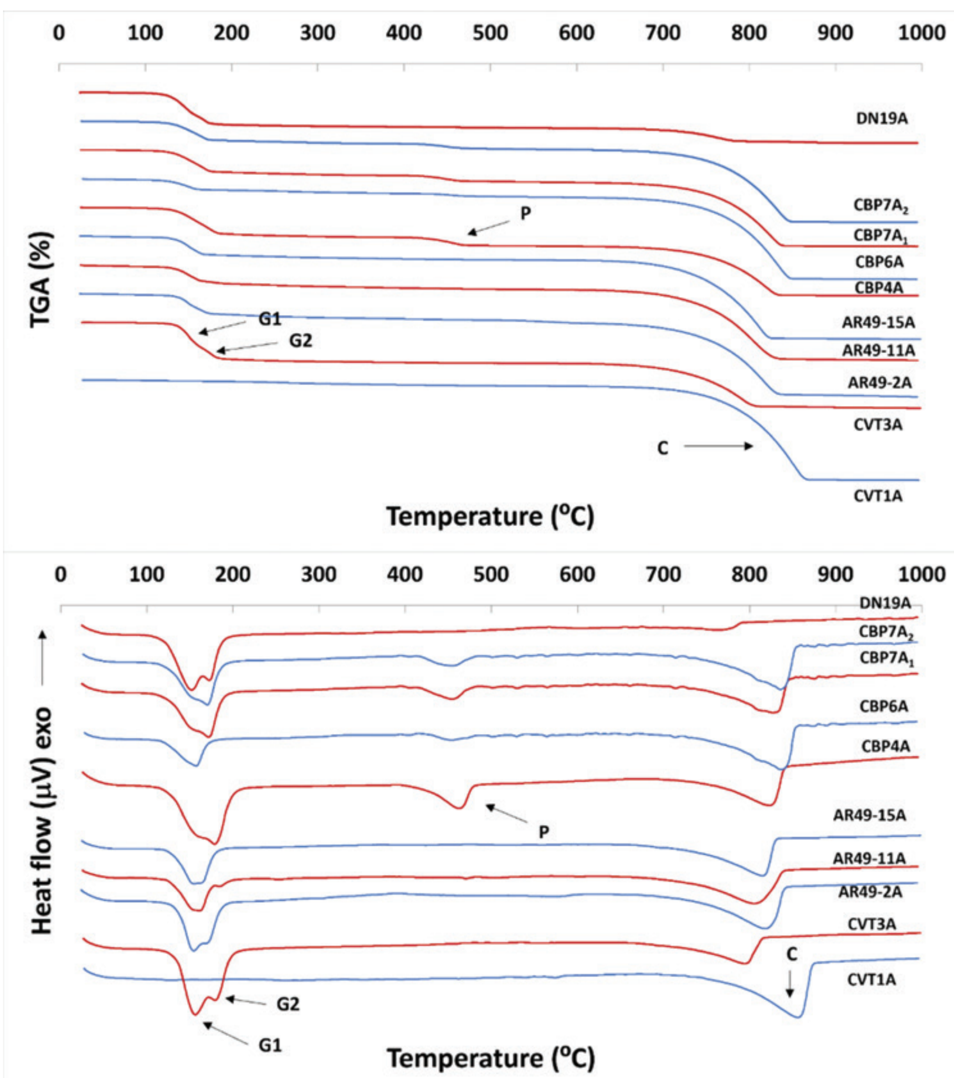


Figure 17. TGA and DTA curves of samples' compositions.

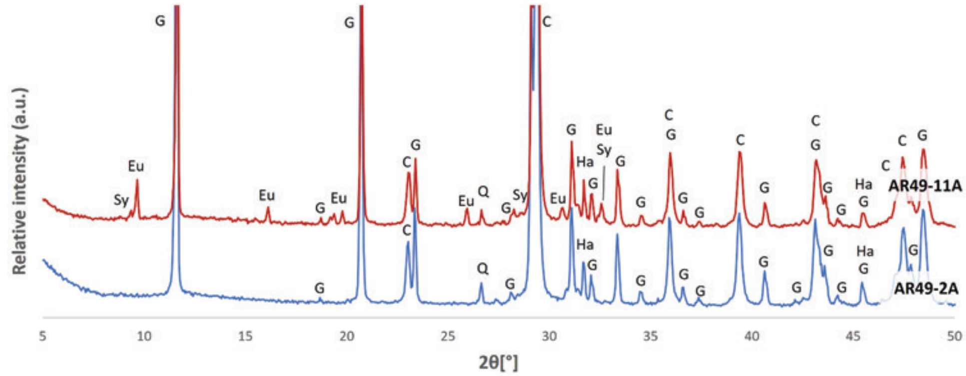


Figure 18. XRD patterns of the samples AR49-2A and AR49-11A. Notation: G – Gypsum; Q – Quartz; naf – Albite; C – Calcite; Ha – Halite; Sy – Syngenite; Eu – Eugsterite.

results of the mineralogical and chemical characterisation demonstrated that the finishing layers of the wall plasters and moulded elements are gypsum and lime based.

Finally, the presence of salt efflorescences (halite: NaCl; syngenite: $K_2Ca(SO_4)_2{\bullet}H_2O$; eugsterite: $Na_4Ca(SO_4)_3{\bullet}2(H_2O)$ detected by XRD in the samples AR49-2A and AR49-11A (Figure 18) should be addressed, indicating contamination, which is consistent to what was reported relating to decay evidence in walls of AR49 (1923) case study by a previous study (Almeida et al. 2021).

The microscopic observations allowed confirming the mineralogical results regarding the composition and binders.

The gypsum crystals are hardly detected and challenging to distinguish in the matrix of the CVT1A sample (Figure 19a,b), which is consistent with XRD and TGA-

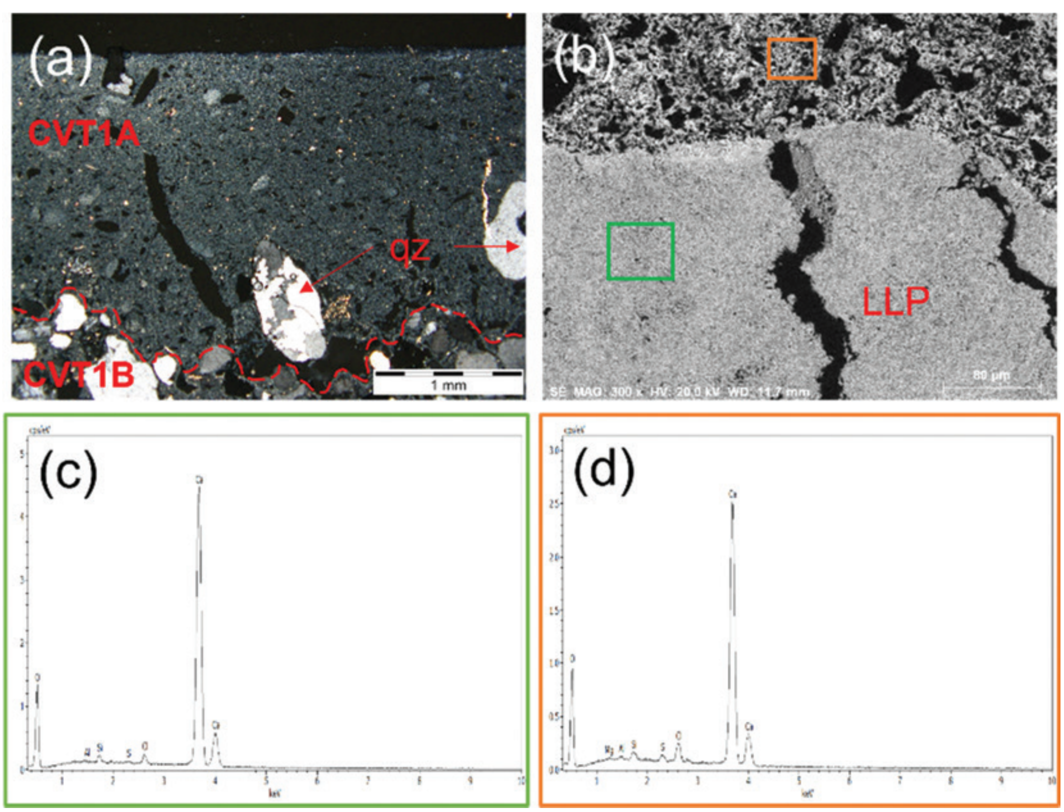


Figure 19. Micrographs (a, b) and EDS spectra (c, d) from the areas marked with green and orange frames in (b). Crossed polarised light image of the interface (red dashed line) between CVT1A and CVT1B (multi-layer samples) with the indication of quartz grains (a); Backscattered SEM micrograph and respective identification by EDS (b); Calcium-rich lime lump (LLP) (c); calcium-rich matrix(d).

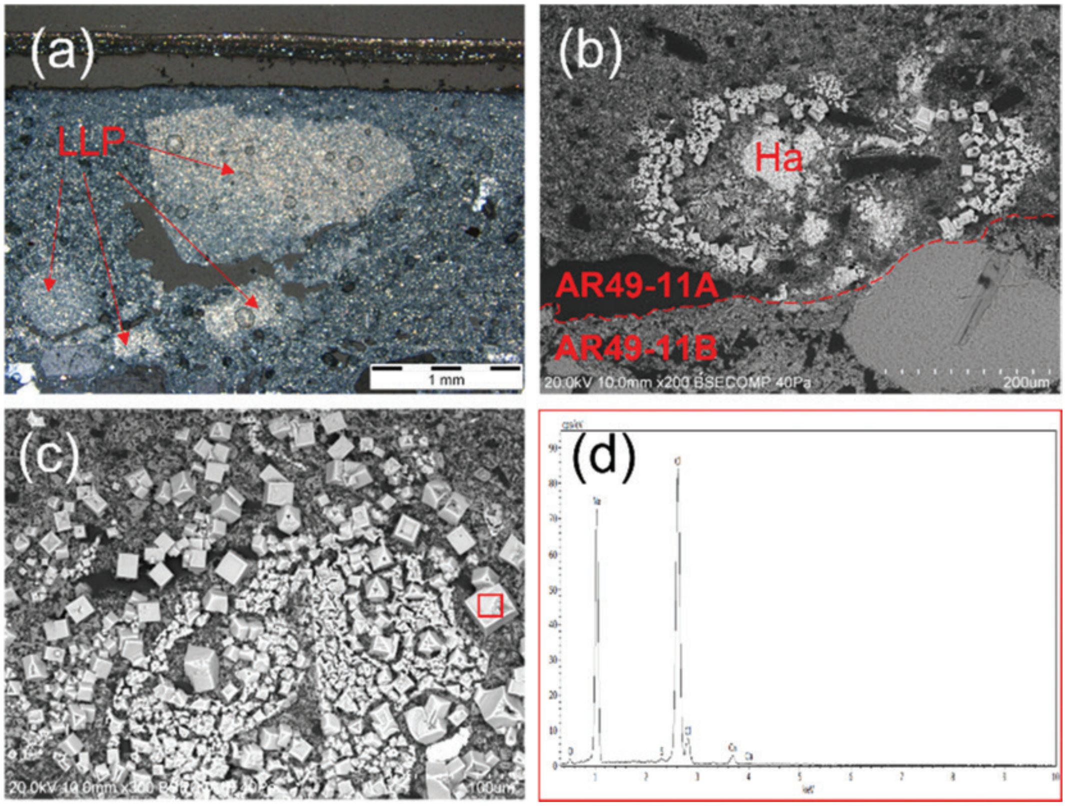


Figure 20. Micrographs (a, b, c) and EDS spectrum (d) from the area marked with a red frame in (c). Crossed polarised light image with the indication of lime lumps (LLP) in AR49-11A (a); backscattered SEM micrograph of salt contamination (Halite: Ha) near the interface of the samples AR49-11A and AR49-11B (b); detail of the salt (NaCl) efflorescence and respective identification by EDS (c); EDS spectrum of the analysed area (d).

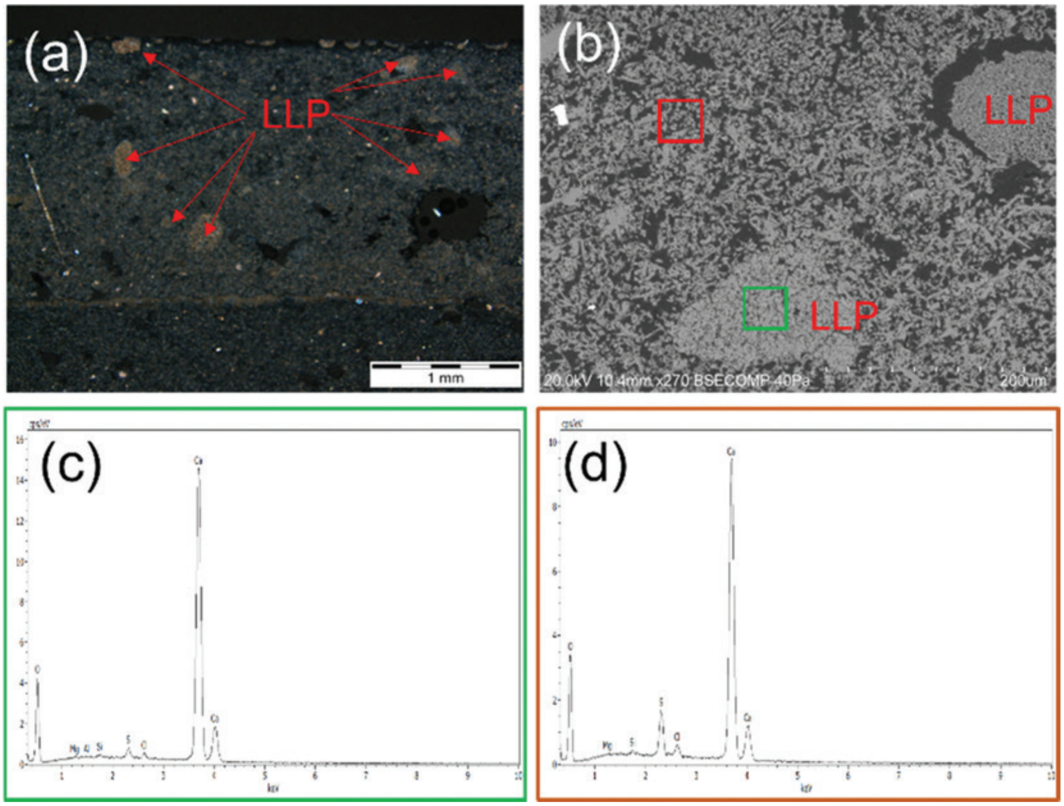


Figure 21. Micrographs of the sample CBP7A$_1$ (a, b) and EDS spectra from the areas marked with a green and a red frame in (b). Crossed polarised light image with the indication of lime lumps (LLP) (a); backscattered SEM micrograph with the evidence of needle-like crystals and the identification of EDS areas (b); calcium-rich lime lump (LLP) (c) and calcium-rich matrix with sulphur (from gypsum) as one of the compositional elements (d).

DTA data, where less than 1.5% gypsum was incorporated. On the contrary, needle-like gypsum crystals are visible in samples where gypsum has a higher presence, such as in CBP7A1 (Figure 21b). In general, a lime-gypsum blended matrix are difficult to distinguish by light microscopy, as both minerals have low birefringence, so SEM must be used to individualise the two constituents.

Many halite crystals are detected in sample AR49-11A (Figure 20), which XRD also detected. It denotes a previous contact with saline water, probably in wet/dry cycles.

Microscopy also shows non-hydrated calcium-rich lime lumps in all samples (Figures 20 and 21), confirming the use of air lime in addition to gypsum.

5. Conclusions

This paper presents and discusses the results of the analytical characterisation of 61 mortar samples from Lisbon's 20th-century architectural awarded buildings. The main conclusions are:

(a) The aggregates of the renders and plasters are mainly of siliceous origin.
(b) The aggregates of the stone-imitating mortars include crushed limestone or rounded quartzite particles. Some also include limestone filler.
(c) The binders used followed a chronological line of application in the studied buildings: Air lime was the only binder used until 1938, with Portland cement employed afterwards.
(d) The stone-imitating mortars studied were formulated with Portland cement, except in one case, when a mixture of air lime and white Portland cement was used.
(e) Portland cement mortars with supplementary cementitious materials were also used, namely GGBFS and fly ash, but only after the 1960s.
(f) Salt ingress by chlorides and sulphates was detected, but in low content. The highest values were found in the oldest buildings studied: CVT (1903) and AR49 (1923). It mainly affects buildings whose structure is of masonry (pre-reinforced concrete structures), which are more susceptible to degradation by the action of water.
(g) Finishing smooth white plasters were produced using a mix of gypsum and air lime.
(h) Lime is the main constituent in the thin-layer plasters used to finish smooth surfaces, which is consistent with the literature for Portuguese gypsum-lime-based plasters. The presence of aggregates was occasionally detected.
(i) Occasional salt contaminations were detected in finishing plasters, which are in line with the above stated in f).

The results obtained allow the first approach for recommending repair and restoration materials. The repair materials must be based on the same binders and aggregates for each mortar type and in proportions similar to pre-existent ones. In addition, the new materials must also have similar physical and mechanical characteristics, which should be detailed in further work.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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References

Adams, J., W. Kneller, and D. Dollimore. 1992. Thermal analysis (TA) of lime and gypsum based medieval mortars. *Thermochimica Acta* 211:93–106. doi:10.1016/0040-6031(92)87010-8.

Aggelakopoulou, E., A. Bakolas, and A. Moropoulou. 2011. Properties of lime–metakolin mortars for the restoration of historic masonries. *Applied Clay Sciences* 53 (1):15–19. doi:10.1016/j.clay.2011.04.005.

Almeida, L., A. S. Silva, M. R. Veiga, J. Mirão, and M. Vieira. 2021. 20th-Century award-winning buildings in Lisbon (Portugal). Study of plasters, rendering, and concrete materials aiming their sustainable preservation. *Buildings* 11 (8):359. doi:10.3390/buildings11080359.

Anon, A. 1914. Investigation of the Pozzolanic nature of coal ashes. *Engineering News* 71:1334–35.

Arliguie, G. 2007. *"Grandubé: Grandeurs associées à la durabilité des bétons"*. 1st ed. Paris: Presses Ecole Nationale Ponts Chaussees.

Blezard, R. G. 1998. The history of calcareous cements. In *Lea's chemistry of cement and concrete*, ed. P. C. Hewlett, 1–23. London: Elsevier. doi:10.1016/B978-075066256-7/50013-8.

Borrachero, M. V., J. Paya, M. Bonilla, and J. Monzo. 2008. The use of thermogravimetric analysis technique for the characterisation of construction materials. *Journal of Thermal Analysis and Calorimetry* 91 (2):503–09. doi:10.1007/s10973-006-7739-3.

Bouichou, M., E. Marie-Victoire, A. Texier, and T. Blondiaux. 2019. How to identify a natural cement: case study of the vassy Church, France. In *Historic Mortars*, ed. J. Hughes, J. Válek, and C. Groot, 3–19. Springer International Publishing. doi:10.1007/978-3-319-91606-4_1

Boynton, R. S. 1980. *The chemistry and technology of lime and limestone*. 2nd ed. New York: J. Wiley & Sons.

Brown, P. W., and J. R. Clifton. 1988. Mechanisms of deterioration in cement-based materials and lime mortar. *Durability of Building Materials* 5 (3–4):409–20.

Bye, G. C. 1999. *"Portland cement: Composition, production and properties"*. 2nd ed. London: Thomas Telford Ltd.

Callebaut, K., J. Elsen, K. Van Balen, and W. Viaene. 2001. Nineteenth century hydraulic restoration mortars in the Saint Michael's Church (Leuven, Belgium): Natural hydraulic lime or cement? *Cement and Concrete Research* 31 (3):397–403. doi:10.1016/S0008-8846(00)00499-3.

Cardoso, I. P., and E. Pye. 2017. Gessoes in Portuguese Baroque gilding grounds, part 1: A study of historical documentary sources. *Studies in Conservation* 62:185–209. doi:10.1080/00393630.2015.1130774.

CEN. 2007. *EN 14629. Products and systems for the protection and repair of Concrete structures. Test methods. Determination of chloride content in hardened concrete*. Brussels: European Committee for Standardization.

CEN. 2014. *EN 196-2. Method of testing cement – Part 2: Chemical analysis of cement*. Brussels: European Committee for Standardization.

Cepuritis, R., B. J. Wigum, E. J. Garboczi, E. Mørtsell, and S. Jacobsen. 2014. Filler from crushed aggregate for concrete: Pore structure, specific surface, particle shape and size distribution. *Cement and Concrete Composites* 54:2–16. doi:10.1016/j.cemconcomp.2014.03.010.

Coimbra, V. L. M., M. J. Moreira dos Santos, and G. Braz de Oliveira. 1995. *A indústria portuguesa do cimento: um século de história 1890-1990*, Vol. 1, 1st ed. Lisboa: Cimpor.

Coutinho, A. S. 2012. *Fabrico e propriedades do betão*, Vol. 1, 5th ed. Lisboa: LNEC.

Czernin, W. 1980. *Cement chemistry and physics for civil engineers*. 2nd ed. London: Georg Godwin.

Davis, R. E., R. W. Carlson, and J. W. Kelly 1937. "Properties of cement and concretes containing fly ash". *Journal Proceedings of the American Concrete Institute*, 33 (5): 577–612. doi:10.14359/8435.

Dekeyser, L., A. Verdonck, and H. De Clercq. 2017. Belgian craftsmanship in the interwar period: Formulas and application techniques of decorative cement-based wall finishes. *International Journal of Architectural Heritage* 11 (5):621–35. doi:10.1080/15583058.2015.1017780.

Deolalkar, S. P. 2015. *Designing green cement plants*. 1st ed. Oxford: Butterworth-Heinemann. doi:10.1016/B978-0-12-803420-0.00001-9.

Freire, M. T., A. Santos Silva, M. R. Veiga, and J. Brito. 2019. Studies in ancient gypsum-based plasters towards their repair: Mineralogy and microstructure. *Construction and Building Materials* 196:512–29. doi:10.1016/j.conbuildmat.2018.11.037.

Gadermayr, N., F. Pintér, and J. Weber. 2012. Identification of 19th century Roman cements by the phase composition of clinker residues in historic mortars . In *Proceedings of the 12th International Congress on the Deterioration and Conservation of Stone*. New York: Columbia University.

Govaerts, Y., A. Verdonck, W. Meulebroeck, and M. de Bouw 2014. "Damage assessment of early 20th century stone imitating mortars" in *Proceedings of the 1st International Conference on Ageing of Materials & Structures*, K. Van Breugel, and E. A. B. Koenders ed., Delft.

Grilo, J., A. Santos Silva, P. Faria, A. Gameiro, R. Veiga, and A. Velosa. 2014. Mechanical and mineralogical properties of natural hydraulic lime-metakaolin mortars in different curing conditions. *Construction and Building Materials* 51:287–94. doi:10.1016/j.conbuildmat.2013.10.045.

Korjakins, A., S. Gaidukovs, G. Šahmenko, D. Bajāre, and D. Pizele. 2008. Investigation of alternative dolomite filler properties and their application in concrete production. *Construction Science* 24 (9):64–71.

Kurdowski, W. 2014. Mineral additions for cement production. In *Cement and Concrete Chemistry*, 1st ed, 533–83. Dordrecht: Springer. doi:10.1007/978-94-007-7945-7_7.

Lea, F. M. 1988. *The chemistry of cement and concrete*. 3rd ed. London: Arnold Ltd.

Lindqvist, J. E., and M. Sandström. 2000. Quantitative analysis of historical mortars using optical microscopy. Characterisation of old mortars. *Materials and Structures/Matériaux et Constructions, RILEM TC*. 167 (33):612–17. Paris: RILEM. doi:10.1007/BF02480600.

Li, L. G., Y. M. Wanga, Y. P. Tana, and A. K. H. Kwanb. 2019. Filler technology of adding granite dust to reduce cement content and increase strength of mortar. *Powder Technology* 342:388–96. doi:10.1016/j.powtec.2018.09.084.

Martinho, C., R. Veiga, and P. Faria. 2018. *Marmorite* – contributo para a correta conservação deste durável revestimento de paredes. *Conservar Património* 28:31–38. doi:10.14568/cp2017026.

Mateus, J. M. 2002. *Técnicas Tradicionais de Construção de Alvenarias. A literatura técnica de 1750 a 1900 e o seu contributo para a conservação de edifícios históricos*. Lisboa: Livros Horizonte.

Mateus, J. M. 2018. A História da Construção em Portugal. In *A História da Construção em Portugal – Consolidação de uma disciplina*, ed. J. M. Mateus, 13–28. Lisboa: By the Book.

MHOP - Ministério da Habitação e Obras Públicas. 1980. *Decreto Regulamentar 4/80. Diário da República n.º 61/1980, I.*

Middendorf, B., J. J. Hughes, K. Callebaut, G. Baronio, and I. Papayianni. 2005a. Investigative methods for the characterisation of historic mortars—Part 1: Mineralogical characterisation. *Materials and Structures* 38 (8):761–69. doi:10.1007/BF02479289.

Middendorf, B., J. J. Hughes, K. Callebaut, G. Baronio, and I. Papayianni. 2005b. Investigative methods for the characterisation of historic mortars—Part 2: Chemical characterisation. *Materials and Structures* 38 (8):771–80. doi:10.1007/BF02479290.

Mňahončáková, E., M. Pavlíková, S. Grzeszczyk, P. Rovnanı, and R. Černý. 2008. Hydric, thermal and mechanical properties of self-compacting concrete containing different fillers. *Construction and Building Materials* 22 (7):1594–600. doi:10.1016/j.conbuildmat.2007.03.016.

MPAT - Ministério do Plano e da Administração do Território, da indústria e Comércio e das Obras Públicas, Transportes e Comunicações. 1987. Despacho conjunto. *Diário da República* II (100): 5581–5582.

Oliveira, G. B. 1995. *A indústria portuguesa do cimento: Um século de história 1890-1990*, 1st ed., Vol. 2. Lisbon: Cimpor.

Papadakis, M., and M. Venuat. 1964. *Fabrication et utilisation des liants hydraulics*. 1st ed. Paris: Imprimerie Jouve.

Parker, J. 1796. A certain cement or terras to be used in aquatic and other buildings and stucco work. *British Patent* n.2120. 27, July. https://www.cementkilns.co.uk/cemkilndoc006.html

Peng, Y., and S. Jacobsen. 2013. Influence of water/cement ratio, admixtures and filler on sedimentation and bleeding of cement paste. *Cement and Concrete Research* 54:133–42. doi:10.1016/j.cemconres.2013.09.003.

Sanz, D. 2009. "Análisis del yeso empleado en revestimientos exteriores mediante técnicas geológicas". PhD Thesis, Polytechnic University of Madrid, Spain.

Shtepenko, O., C. Hills, A. Brough, and M. Thomas. 2006. The effect of carbon dioxide on β-dicalcium silicate and Portland cement. *Chemistry Engineering* 118 (1–2):107–18. doi:10.1016/j.cej.2006.02.005.

SNIP. 1982. *Le plâtre : physico-chimie, fabrication et emplois*. Paris: Eyrolles.

Taylor, H. F. W. 1990. *Cement Chemistry*. New York: Academic Press.

Veiga, M. R. 2012. Conservation of historic renders and plasters: From laboratory to site. historic mortars: characterisation, assessment, conservation and repair. In *RILEM Bookseries*, ed., Valek, Hughes, and Groot, Vol. 7. doi:10.1007/978-94-007-4635-0_16.

Veiga, R., J. Aguiar, A. Santos Silva, and F. Carvalho 2001. "Methodologies for characterisation and repair of mortars of ancient buildings", in *Proceedings of the 3rd International Seminar on Structural Analysis of Historical Constructions*, Guimarães, Portugal, ed. by P. B. Lourenço and P. Roca, 353–62. University of Minho.

Veiga, M. R., and A. Santos Silva. 2019. *"Mortars", in* long-term performance and durability of masonry structures degradation mechanisms, health monitoring and service life design. Ghiassi, B., Lourenço, P. B, eds. 169–208. Woodhead Publishing, Elsevier. 10.1016/B978-0-08-102110-1.00006-6.

Veiga, M. R., A. Santos Silva, C. Martinho, and P. Faria 2019. "Decorative renders simulating stone of middle 20th century in the region of Lisbon", in *Proceedings of the 5th Historic Mortars Conference, Pamplona, Spain*, Álvarez, J. I., Fernández, J. M., Navarro, I., Durán, A., and Sirera R. ed., Paris: RILEM.

Vidovszky, I., and F. Pintér. 2018. An Investigation of the application and material characteristics of early 20th-Century Portland cement-based structures from the historical campus of the Budapest University of Technology and Economics. *International Journal of Architectural Heritage* 14 (3):358–75. doi:10.1080/15583058.2018.1545059.

Walsh, J. J. 2007. Petrography: Distinguishing natural cement from other binders in historical masonry construction using forensic microscopy techniques. *Journal ASTM International* 4 (1):100674. doi:10.1520/JAI100674.

Weber, J., A. Baragon, F. Pintér, and C. Gosselin 2015. "Hydraulicity in ancient mortars: Its origin and alteration phenomena under the microscope" in *Proceedings of the 15th Euroseminar on Microscopy Applied to Building Materials*, Delft.

Weber, J., N. Gadermayr, R. Kozłowski, D. Mucha, D. Hughes, D. Jaglin, and W. Schwarz. 2007. Microstructure and mineral composition of Roman cements produced at defined calcination conditions. *Materials Characterisation* 58 (11–12):1217–28. doi:10.1016/j.matchar.2007.04.025.

Wentworth, C. K. 1922. A scale of grade and class terms for clastic sediments. *The Journal of Geology* 30 (5):377–92. doi:10.1086/622910.

Winter, N. B. 2012. *Understanding cement. An Introduction to Cement Production, Cement Hydration and Deleterious Processes in Concrete*. Woodbridge, UK: WHD Microanalysis Consultants Ltd.

Zacharopoulou, G. 2009. Interpreting chemistry and technology of lime binders and implementing it in the conservation field. *Conservar Património* 10:41–53. doi:10.14568/cp10-4.

4.4.2. Physical and mechanical characterisation

The following paper addresses the study of renders and plasters' physical and mechanical characteristics from nine award-winning buildings. Due to constraints such as limited authorization for sample collection in all seventeen case studies and the presence of coverings in inaccessible areas, a more extensive sampling effort proved unfeasible. The characterisation was done to understand mortars' physical and mechanical properties and their evolution during the 20th century. These characteristics will also help determine compatibility requirements for future conservation and restoration interventions. Fifty-three samples were studied through capillary water absorption, drying rates, open porosity, dynamic modulus of elasticity and compressive strength. The results showed different ranges of quantitative values for these tests, whether the mortars are lime, gypsum, cement-based or do have lime-cement blended formulations.

The results show that the air lime mortars existing in the oldest buildings, between 1903 and 1944, have the highest values of capillary absorption and simultaneously the highest drying rates and present the lowest values of compressive strength and dynamic modulus of elasticity, which is expected for this type of mortars.

The blended lime–cement mortars in the buildings constructed between 1938 and 1944 have intermediate capillary absorption and drying rates in comparison to lime and Portland cement mortars. Compressive strength values of blended lime–cement mortars and multi-layer mortars with different binders in each layer are variable. In general, an increase in the Dynamic Modulus of Elasticity values is related to the introduction of Portland cement. The Portland cement mortars applied in buildings erected after 1939 show the lowest values of capillary absorption and the highest values of mechanical strength.

Despite the limitations in obtaining samples and preparing them in the laboratory, particularly in cases where it was not feasible to separate and test each layer of the multi-layer system in an independent way, for compatibility purposes it is prudent to consider as a good approximation the properties obtained for the whole set of layers.

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20th Century Mortars: Physical and Mechanical Properties from Awarded Buildings in Lisbon (Portugal)—Studies towards Their Conservation and Repair

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Abstract: This paper addresses the study of renders and plasters' physical and mechanical characteristics from selected buildings awarded during the 20th century with a renowned architectural prize in Lisbon, Portugal. The characterisation was done to understand mortars' physical and mechanical properties and their evolution during the 20th century. These characteristics will also help determine compatibility requirements for future conservation and restoration interventions. Since these buildings have a heritage great interest status, the need to preserve them is a paramount issue. Fifty-three samples from nine case studies were studied via capillary water absorption, drying rates, open porosity, dynamic modulus of elasticity, and compressive strength. There were limitations in sample collection due to the buildings being in service and technical constraints regarding sample quantity for testing and separating layers of the multi-layer mortar system. Nevertheless, the results showed different ranges of quantitative values for these tests, whether the mortars were lime, gypsum, cement-based or had lime–cement blended formulations.

Keywords: mortars; renders; plasters; water absorption; drying rates; mechanical properties; open porosity; air lime; lime–cement; Portland cement; 20th century; compatibility

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1. Introduction

The 20th century Valmor Prize for Architecture award-winning buildings are testimonies of Lisbon's cultural, architectural, and constructive heritage [1,2] that should be studied and understood to be better preserved and valued. The knowledge of the characteristics of the building materials in their historical context enables a more effective response to the conservation and restoration issues that arise from ageing or lack of maintenance. Designing mortars for restoration is critical in any conservation project [3,4]. Composite materials, in particular mortars, are complex materials that depend on (i) the raw materials used and (ii) the design parameters. In the case of monument protection and historical buildings, it is essential to design mortars with the characteristics required to ensure their compatibility with existing materials and their effectiveness in physical and mechanical performance [3,5]. Although the rendering mortars and plasters of most of the studied buildings have generally shown a reasonable to good state of conservation, as reported previously by Almeida et al. [6], it is helpful for an in-depth study of their characteristics to choose which materials should be used in case the original materials have to be replaced so that they can be compatible with the substrate and with the background pre-existent materials. Any intervention that requires a partial replacement of the renders should consider several requirements to be taken into account. These requirements mainly concern water resistance and chemical and mechanical behaviour, besides aesthetic compatibility [7].

The mechanical compatibility of the mortar means essentially that the flexural and compressive strengths, as well as the elastic parameters, should be similar, or lower, in order not to transmit tensions to the old ones over a level that can contribute thoughtfully to their cracking, delamination, or rupture [7,8]. Thus, the modulus of elasticity of the compatible materials must not be higher than that of the existing materials [9].

The hardening of the mortar, whether by hydration, carbonation, or other reactions, its shrinkage and relaxation capacity, which is not characterised by the modulus of elasticity or by instantaneous measurements of other characteristics, will influence the transmission of stresses that occur over a certain period. Insofar as concerns physical characteristics, namely water capillary absorption, adsorption, and diffusion, all these interactions with liquid and vapour of water phases should also be identical (or higher), i.e., the compatible mortar materials for restoration should not impair the water transport in vapour or liquid state thus forcing it to circulate via the historic materials preferentially. This compatibility should be verified to prevent the exposure of wall components to excess and/or long-term humidification periods [10].

Besides physical and mechanical compatibility of new materials to pre-existing ones, chemical compatibility is acceptable to meet the requirements set out above and prevent the formation and/or contamination with non-desirable substances (e.g., soluble salts) [11].

The selection of raw materials used according to functional requirements has led, in the past, to the application of techniques that allowed the differentiated use of aggregates and binders mixed in different proportions. The use of successive layers (multi-layer system), with different thicknesses and with the reduction of the average size of aggregates and layer thickness towards the surface, particularly in lime mortars, was beneficial to avoid the ingress of moisture into the structure [12] to minimise the shrinkage tendency and to optimise the carbonation of lime [13]. Coating systems replaced these traditional systems with artificial hydraulic lime of higher performance and, later, in the 20th century, with Portland cement [13–15], optimised by single-layer systems, with pre-dosed mixtures ready to be applied.

This paper presents the results of the physical and mechanical characterisation of mortars and plasters from a set of nine Valmor Prize for Architecture award-winning buildings in Lisbon constructed between 1903 and 2002. The Valmor Prize still has today, since its first attribution in 1902, an annual base for its attribution to promote and encourage architectural quality, which has invariably been reflected in the quality and constructive solutions adopted. Its regulations were remodelled several times, and the Valmor Prize was merged with the Municipal Prize for Architecture in 1982 after its establishment in the 1940s [1,2]. The construction of all the studied buildings began in the 20th century, and they represent a sample of the best construction practices and features, highly relevant for the study of the state of the art of 20th century construction in the city and the same time in the country. For this reason, this work does not intend to study ordinary buildings. Legal frameworks protect some of these buildings, which are generally recognised for their architectural and aesthetic excellence. Studying them offers insights into what is considered a high standard in design and urban planning. These buildings often feature a careful selection of materials, according to the higher patterns of their lifetime, aligning with the architectural design and technical requirements.

Many award-winning projects incorporate innovative and sustainable materials, contributing to advances in the construction industry. Analysing the materials used in these buildings provides valuable information on the performance of the materials and their long-term durability in different environmental conditions. In order to gain a deeper understanding of the evolution of the materials used over a century, it was decided to choose at least one award-winning building from each decade of the 20th century. However, not all decades have award-winning buildings, and this study was not permitted in some cases.

The mineralogical, chemical, and microstructural characterisation, fundamental for the knowledge of the binders and aggregates' nature and other crucial aspects to determine their state of conservation was already performed [16].

The results will generate a significant set of critical elements for understanding the evolution of mortar typologies and applications throughout the 20th century in Portugal. The consistent data set should also be considered in the design of repair mortars compatible with the original and still preserved ones. The information generated will also allow future comparisons with similar materials from other countries/regions.

2. Materials and Methods

2.1. Materials

The physical and mechanical characterisation results of fifty-three samples of renders and plasters from nine case studies will now be presented. Samples were mainly collected by hammer and chisel, although some were collected by core drilling during a concrete sampling campaign, whose study is not addressed in this paper. Table 1 refers to the collected samples' location, constructive element, and application technique. A sample's short description is presented in the same table.

The samples, mainly multi-layered (Figure 1), are often finished with smooth, white, thin plasters (Figure 2) when it comes to indoor walls and stone-imitating mortars (Figure 3). However, in several cases, the finishing layer is a painting coating. An alphanumeric sample code identifies the layer position towards the surface (Table 1), which means the letter A stands for the outermost layer.

Figure 1. Multi-layer sample set DN12ABCD. A paint finish layer (red arrow) can be seen covering the most superficial layer (A). The sample layers are labelled A–D.

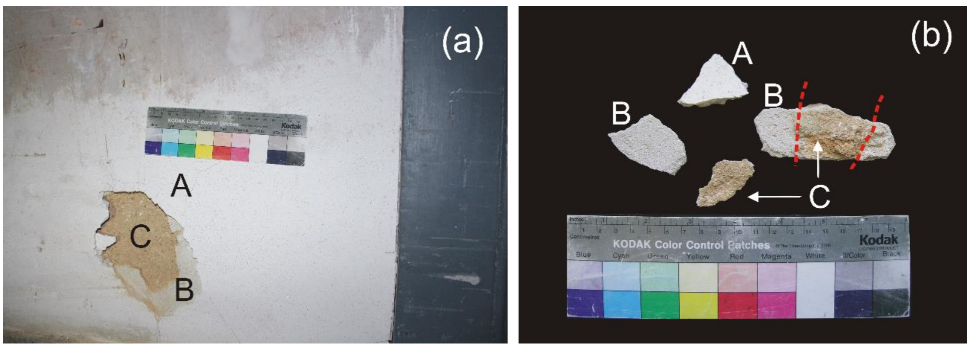

Figure 2. Multi-layered sample CVT1ABC: (**a**) sampling on a wall with pre-existing detachment; (**b**) layers' specimens A, B and C. The white finishing smooth thin plaster corresponds to layer A. Red dots divide layers B and C.

Table 1. Building location and general characteristics of the samples according to Almeida et al. [16].

Case Study	Building Name and Location (WGS84 Coordinates)	Awa. Yr.	Comp. Yr.	Location of the Samples/Sampled Element/Application Technique	Samples ID	Samples Description	Th. (mm)
CVT (1903)	*Ventura Terra* Building (38.72082, −9.15319)	1903	1903	Basement–entrance hall/Internal wall/Multi-layer plaster	CVT1A	Gypsum-air lime-based plaster. Finishing layer	2
					CVT1B	White mortar with siliceous sand	5
					CVT1C	Brownish mortar with lime lumps and siliceous sand	20
				Ground floor–Adornment arch of the Entrance hall/Internal wall/Multi-layer plaster	CVT3A	Gypsum-air lime-based plaster	5
					CVT3B	Orange-brownish, friable mortar with lime lumps and siliceous sand	20
AR49 (1923)	*Luiz Rau* Building (38.73872, −9.14668)	1923	1923	Courtyard access. Ground floor. Ceiling/External wall/Multi-layer render	AR49-6C	Orange-brownish, friable mortar with lime lumps and siliceous sand	10 *
				Balcony on the 5th floor. East façade/External wall/Multi-layer plaster	AR49-8A	Whitish mortar with fine siliceous sand	4
					AR49-8B	Orange-brownish, friable mortar with lime lumps and siliceous sand	12
				West-facing wall between 5th and 6th stair floor landing/Internal wall/Multi-layer plaster	AR49-11A	Gypsum-air lime-based plaster	3
					AR49-11B	White mortar with siliceous sand	5
				Window located on the stairs between the 4th and 5th floor/Internal window-lintel/Multi-layer plaster	AR49-15A	Gypsum-air lime-based plaster	3
					AR49-15B	White mortar with siliceous sand	5
					AR49-15C	Orange-brownish, friable mortar with lime lumps and siliceous sand	25
IRF (1938)	*Nossa Senhora do Rosário de Fátima* Church (38.74005, −9.15051)	1938	1938	Sacristy/Internal wall/Multi-layer plaster	IRF1B	Brownish mortar with lime lumps and siliceous sand	10
				Nossa Senhora da Piedade Chapel/Internal wall/Multilayer plaster	IRF2A	White mortar with fine siliceous sand	5
					IRF2B	Orange-brownish mortar with siliceous sand	3 *
				Main chapel gallery (roof access)/Internal wall/Multi-layer plaster	IRF3A	White mortar with fine siliceous sand	4
					IRF3B	Orange-brownish mortar with lime lumps and siliceous sand	5 *
				Interior access to the bell tower/Internal wall/Monolayer plaster	IRF4A	Single-layer, grey-brown mortar with lime nodules and siliceous sand	10
				Interior access to the bell tower/Internal wall/Multi-layer plaster	IRF7A	Brownish-grey mortars with lime lumps and siliceous sand	10
					IRF7B		8

Table 1. *Cont.*

Case Study	Building Name and Location (WGS84 Coordinates)	Awa. Yr.	Comp. Yr.	Location of the Samples/Sampled Element/Application Technique	Samples ID	Samples Description	Th. (mm)
CBP (1939)	*Bernardo da Maia* House (38.73867, −9.16181)	1939	1939	Basement. Staff room/Internal wall/Monolayer plaster	CBP1A	Single-layer, whitish mortar with lime lumps and siliceous sand	15
				1st floor activity's room/Internal wall/Multi-layer plaster	CBP4A	Gypsum-air lime-based plaster	3
					CBP4B	Greyish mortar with siliceous sand	15
				1st floor. Corridor to the activity's room/Internal wall/Multi-layer plaster	CBP6A	Gypsum-air lime-based plaster	4
					CBP6B	Whitish mortar with lime lumps and siliceous sand.	15
					CBP7B	Whitish mortar with lime lumps and siliceous sand	20 *
DN (1940)	*Diário de Notícias* Building (38.72376, −9.14810)	1940	1940	Level 2. Technical rooms' corridor/Internal wall–column/Monolayer plaster	DN9A **	Greyish mortar with siliceous sand	31
					DN10A **	Greyish mortar with siliceous sand	31
				Level 2. Technical rooms' corridor/Internal wall–column/Multi-layer plaster	DN11A **	Greyish mortar with siliceous sand	20
					DN11B **	Greyish mortar with siliceous sand	6
				Level 2. Warehouse room/Internal wall–column/Multi-layer plaster	DN12A	Greyish mortar with siliceous sand	20
					DN12B	Brownish mortar with lime lumps and siliceous sand	30
					DN12C	Compact grey mortar with fine siliceous sand	15
					DN12D	Whitish mortar with lime lumps and siliceous sand	15
				5th floor. Office room–North wall/Internal wall/Multi-layer plaster	DN19A	Gypsum-air lime-based plaster	4
					DN19B	Greyish mortar with siliceous sand	12
					DN19C	Greyish mortar with siliceous sand	5
					DN19D	Light brownish mortar with lime lumps and siliceous sand	20

Table 1. *Cont.*

Case Study	Building Name and Location (WGS84 Coordinates)	Awa. Yr.	Comp. Yr.	Location of the Samples/Sampled Element/Application Technique	Samples ID	Samples Description	Th. (mm)
AAC (1944)	*Cristino da Silva* Building (38.71676, −9.15777)	1944	1944	Side access to ground floor/External wall/Multi-layer render	AAC1A	Rough, red-coloured mortar with fine siliceous sand	7
					AAC1B	Greyish mortar with siliceous fine sand	7
				Ground floor outdoor render/External wall/Multi-layer render	AAC2A	Stone-imitating mortar with projected limestone aggregates	5
					AAC2B	Greyish mortar with siliceous sand	15
				Boiler room/Internal wall/Multi-layer render	AAC3A	Stone-imitating mortar "Marmorite" with white and blueish limestone aggregates	5
				Step coating of inside stairs/Internal wall/Multi-layer render	AAC4A	Stone-imitating mortar "Marmorite" with white limestone aggregates	5
LIP (1958)	Laboratories of Pasteur Institute of Lisbon (38.75730, −9.10695)	1958	1957	Chimney render/External wall/Monolayer render	LIP1A	Grey mortar with siliceous sand	20
				Ground floor. South building. west façade/External wall/Monolayer render	LIP9A	Grey mortar with siliceous sand	7
EUA53 (1970)	*América* Building (38.74877, −9.13695)	1970	1969	Common interior staircase wall. Third floor/Internal wall/Multi-layer render	EUA53-2A	Stone-imitating mortar "Marmorite" with quartzite aggregates	10
					EUA53-2B	Grey mortar with siliceous sand	15
				Chimney render/External wall/Multi-layer render	EUA53-3A	Yellow stone-imitating mortar "Marmorite" with white limestone aggregates	8
					EUA53-3B	Compact grey mortar with siliceous sand	50
				Corridor of the technical area/Internal wall/Multi-layer plaster	EUA53-4B	Grey mortar with siliceous sand	20
UNL (2002)	New University of Lisbon Rectory (38.73440, −9.16026)	2002	2002	Air treatment unit room/Internal wall/Monolayer render	UNL3A	Single-layer greyish mortar with siliceous sand	15

Notation: Awa. Yr.—Award year; Comp. Yr.—Year of completion of building construction; Th—Average thickness of the sample (mm); * Measurement performed directly on the sample (lower than the total thickness of the corresponding layer); ** Samples of the plasters applied during a refurbishment action in 1998.

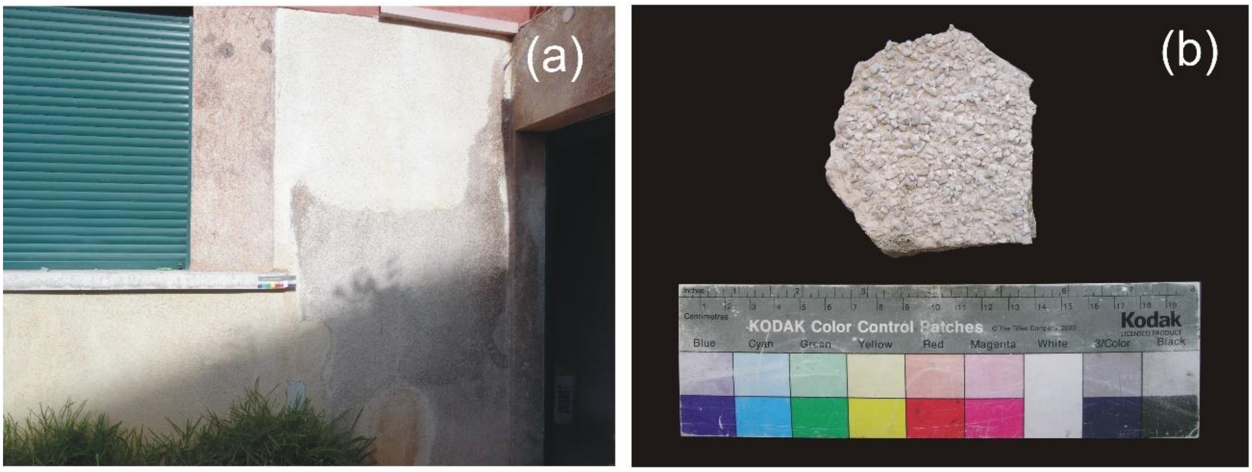

Figure 3. Stone-imitating mortar applied on the rear façade of the case study AAC 1944 (**a**), and a sample render (AAC2A, see Table 1) was collected in the same building (**b**).

Mineralogical and microstructural characterisation [16] showed that the studied mortars have different types of binders, from air lime and gypsum to Portland cement, as well as blended binders, that is, air lime mixed with Portland cement (Table 2).

2.2. Limitations of the Study

The sample program was designed considering the preservation of the building's aesthetic meaning and value. Mortars were collected aiming to have enough material for testing, which was only sometimes possible as most buildings were in service. Most of the samples were collected indoors for several reasons, such as the building's inaccessibility, forbidden areas, or external façades having no rendering mortars.

In the multi-layered samples, attempts were made to carefully separate each layer from the other mechanically, sometimes without success. Nevertheless, due to the inability to preserve the integrity of the detached thin layers (e.g., thin, smooth layers) from the whole set, it was decided to test the entire set when it was not possible to separate each layer or when it was foreseeable that the layers would not have enough dimension to be tested alone. When a limited number of samples was available, a methodology was adopted that included phased testing of the same sample, starting with non-destructive tests, such as ultrasound pulse velocity to evaluate the dynamic modulus of elasticity, and ending with compressive strength. Some samples were fragmented or cut into several specimens, one for each test; when an abundant sample was available, several specimens were tested. As the samples have non-standard and irregular shapes, the laboratory characterisation required adapted test methods that were developed and validated in previous works [17–19].

2.3. Experimental Work

2.3.1. Capillary Water Absorption and Drying Capacity Test

It is essential to ensure that after an intervention, the wall will have a similar hygric behaviour as the wall with its original materials to achieve compatibility between the old and the replacement mortar. The previous hygrometric characteristics should be maintained or slightly modified, i.e., similar capillary and water vapour coefficients or higher [5].

Table 2. Summary of results obtained for the analysed samples (by layer or by sets of layers).

Case Studies	Samples	Layers' Set	D1	D2	Ccc	UPV	E_{dus}	P_0	ρ	CS	Binder Type (Per Layer) [(2)]	b/a [(2)]
			$kg.m^{-2}.min^{-1}$	$kg.m^{-2}.min^{-0.5}$		$m.s^{-1}$	MPa	%	$kg.m^{-3}$	MPa		
CVT (1903)	CVT1A	(b)	0.0021	0.0539	0.39						GP	(b)
	CVT1AB	Set A + B				1670.05	3155.11 [(1)]				GP (A); AL (B)	(b)
	CVT1C	(b)	(a)		1.22			33.80	1664.94		AL	1:7.8
	CVT3A	(b)	0.0024	0.0066	0.16	2011.49	4600.96	40.48	1263.48		GP	(b)
	CVT3B	(b)	0.0036	0.0144	1.47	1164.88	2073.30	30.77	1697.67	2.57	AL	1:4.3
	CVT3AB	Whole set								6.17	GP (A); AL (B)	(b)
AR49 (1923)	AR49-6C	(b)	0.0013	0.0225	0.69			32.05	1687.79		AL	1:5.8
	AR49-8B	(b)						26.89	1759.82		AL	1:11.2
	AR49-8AB	Whole set	0.0116	0.0798	0.83	1725.33	4606.04	26.83	1719.26		AL (A); AL (B)	(b)
	AR49-11A	(b)						47.44	1293.50		GP	(b)
	AR49-11AB	Whole set	0.0009	0.0263	0.78	2166.51	4319.57 [(1)]				GP (A); AL (B)	(b)
	AR49-15C	(b)	0.0129	0.0294	1.68						AL	1:7.9
	AR49-15ABC	Whole set	0.0030	0.1195	1.35	1476.89	3385.24	30.76	1724.46	1.77	GP (A); AL (B); AL (C)	(b)
IRF (1938)	IRF1B	(b)	0.0057	0.1291	0.59	1427.56	3092.04	31.12	1685.82	2.09	AL + OPC	1:1:7
	IRF2AB	Whole set	0.0050	0.1330	0.76	1868.21	5129.75 [(1)]				AL	(b)
	IRF3A	(b)	0.0019	0.0459	0.28	830.50	1054.57	33.04	1698.83		AL	1:4.2
	IRF3B	(b)	0.0068	0.1252	1.62			31.02	1706.65		AL	1:8
	IRF4A	(b)	0.0016	0.0749	0.54	1022.73	1814.01	22.68	1926.98		AL + OPC	1:5.4
	IRF7AB	Whole set	0.0023	0.0813	0.63	1477.76	3722.03	24.07	1893.78	18.37	AL + OPC (A); AL + OPC (B)	(b)

Table 2. *Cont.*

Case Studies	Samples	Layers' Set	D1	D2	Ccc	UPV	E_{dus}	P_0	ρ	CS	Binder Type (Per Layer) [(2)]	b/a [(2)]
			$kg.m^{-2}.min^{-1}$	$kg.m^{-2}.min^{-0.5}$		$m.s^{-1}$	MPa	%	$kg.m^{-3}$	MPa		
CBP (1939)	CBP1A	(b)	0.0033	0.1176	1.25	1418.81	3077.71	28.31	1698.79	2.27	AL	1:8.4
	CBP4B	(b)	0.0041	0.4590	2.05			25.97	1832.76		OPC	1:20.3
	CBP4AB	Whole set	0.0043	0.1153	1.91	1726.22	4579.32	33.17	1602.72	5.48	GP (A); OPC (B)	(b)
	CBP6AB	Whole set	0.0016	0.0892	1.28	1437.19	2942.01	29.47	1707.52	2.69	GP (A); AL (B)	(b)
	CBP7B	(b)	0.0031	0.1099	2.22	1207.73	2359.58	25.92	1797.43	1.26	AL	1:11.2
DN9 (1940)	DN9A	(b)	0.0016	0.0706	0.31	1994.65	7184.41	18.34	2006.40	10.59	OPC	1:6.1
	DN10A	(b)	0.0023	0.0706	0.23	2109.27	8049.49	18.25	2010.30	22.39	OPC	1:7
	DN11AB	Whole set	0.0023	0.0065	0.23	1984.13	7150.79	18.43	2018.24	33.81	OPC	(b)
	DN12ABCD	Whole set	0.0012	0.0903	0.39	2173.91	7502.27			12.34	OPC (A); AL + OPC (B); OPC (C); AL (D)	(b)
	DN12A	(b)				2057.60	7281.23	23.66	1910.91		OPC	1:12.9
	DN12B	(b)				1848.02	5640.63	23.06	1835.15		AL + OPC	1:2.1:15.1
	DN12C	(b)				2149.77	8120.51	17.58	1952.35		OPC	1:4.2
	DN12AB	Set A + B	(c)		0.55						OPC (A); AL + OPC (B)	(b)
	DN12CD	Set C + D	(c)		0.42						OPC (C); AL (D)	(b)
	DN19A	(b)	(c)		0.70	1995.44	4495.09 [(1)]				GP	(b)
	DN19B	(b)	(c)		0.50	2317.20	9346.00	22.24	1933.82		PCC	1:25.2
	DN19C	(b)	(c)		0.22						OPC	1:8.9
	DN19D	(b)	(c)		0.58	2170.77	7744.08	26.76	1826.43		AL + PCC	1:1:6
	DN19ABCD	Whole set	0.0028	0.0861	0.21					9.71	GP (A); PCC (B); OPC (C); AL + PCC (D)	(b)
AAC (1944)	AAC1A	(b)	(c)		0.53						AL + OPC	1:0.2:6.1
	AAC1B	(b)	(c)		0.35						AL + OPC	1:0.2:7
	AAC1AB	Whole set	0.0027	0.0083	0.37	1197.60	2233.14	31.20	1730.00		AL + OPC (A); AL + OPC (B)	(b)
	AAC2AB	Whole set	0.0017	0.0255	0.19	2654.63	13369.52	14.11	2107.97	33.78	AL + WPC (A); OPC (B)	(b)
	AAC3A	(b)	0.0008	0.0151	0.10	1863.35	6383.21 [(1)]				OPC	1:3.0
	AAC4A	(b)	0.0015	0.0211	0.14						OPC	1:1.9

Table 2. *Cont.*

Case Studies	Samples	Layers' Set	D1	D2	Ccc	UPV	E_{dus}	P_0	ρ	CS	Binder Type (Per Layer) (2)	b/a (2)
			$kg.m^{-2}.min^{-1}$	$kg.m^{-2}.min^{-0.5}$		$m.s^{-1}$	MPa	%	$kg.m^{-3}$	MPa		
LIP (1958)	LIP1A	(b)	0.0020	0.0377	0.63	2281.79	14841.50	22.64	1919.64	16.58	OPC	1:7.6
	LIP9A	(b)	0.0013	0.0252	0.11	1533.74	6040.63 (1)				OPC	1:6.6
EUA53 (1970)	EUA53-2A	(b)	0.0013	0.0197	0.08	1580.61	5050.13	13.20	2246.00	65.15	WPC	1:3.7
	EUA53-2B	(b)	0.0069	0.0096	0.64	1460.56	3764.49	20.13	1960.75	11.90	OPC	1:6.7
	EUA53-3B	(b)	0.0039	0.0415	0.11			15.20	2066.00	28.72	OPC	1:4.9
	EUA53-3AB	Whole set	0.0013	0.0310	0.07	1916.81	7024.69 (1)				WPC (A); OPC (B)	(b)
	EUA53-4B	(b)				1675.98	4843.92	21.92	1916.10	12.50	OPC	1:11.5
UNL (2002)	UNL3A	(b)	0.0025	0.0414	0.44	2336.09	9701.89	17.40	1975.30	18.62	OPC	1:10.2

Notation and remarks: (a) test stopped due to sample breakage; (b) not applicable; (c) layers could not be tested per se, nor partial sets of more than one layer could be tested. Result for the whole set; (1) The bulk density used in the calculation was obtained by dividing the mass of the specimen by the product of the average dimension of width, length and depth; (2) According to [16]; AL—Air lime; GP—Gypsum-air lime-based plaster; OPC—Ordinary Portland cement; PCC—Portland composite cement; WPC—White Portland Cement; b/a—binder to aggregate ratio by mass [hydrated lime (HL)/OPC/aggregate—for air lime with by mixed ordinary Portland cement; HL/aggregate—for air lime mortars; OPC/aggregate and WPC/aggregate—for cement mortars]. Blank fills to non-performed tests.

Capillary water absorption was determined using a test procedure developed for historic mortars [17]. All the capillary water absorption tests were performed in a controlled environment (T = 20 ± 2 °C and RH = 65 ± 5%) and using the capillary absorption by contact technique (Figure 4a,b). For friable samples, this technique consists of placing the samples in baskets with a bottom lined with a geotextile sheet to avoid material loss during the test. In contrast, non-friable samples were placed directly in contact with water (Figure 4a). Non-friable samples and the sets consisting of sample, basket, and geotextile sheet were held on two narrow acrylic strips in a tub with enough water to keep the contact samples' surface or the geotextile sheets wet. The samples were weighed at 0, 2, 5, 7, 10, 15, 20, 25, 30, 35, 40, 60, 90, 180, 300, 480, and 1440 min (24 h) and then every 24 h until saturation. The capillary absorption coefficient by contact (Ccc in kg.m^{-2}.min$^{-0.5}$), which refers to the initial rate of water absorption is measured by the slope of the initial phase of the curve based on linear regression as determined by EN 15801 [20]. The baskets with the samples were taken out of the tub after the samples reached saturation and were placed in the same environment to dry on acrylic strips to prevent contact with any other surface (Figure 4c). The drying rates (corresponding to the first drying phase D1 and the second drying phase D2) were evaluated via the weighing procedure at 30, 60, 90, 270, 480, and 1440 min and every 24 h until the test specimens achieved constant weight, as determined by EN 16322 [21].

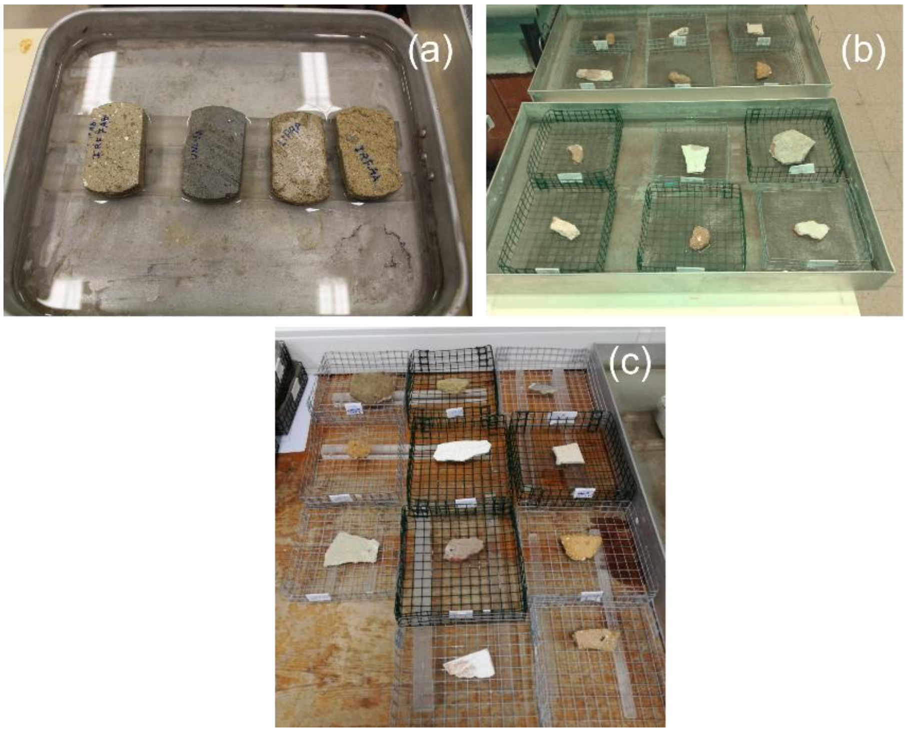

Figure 4. Determination of the capillary water absorption by contact of non-friable (**a**) and friable samples (**b**); and drying procedure (**c**).

2.3.2. Open Porosity and Bulk Density

The study of open porosity deepens the understanding of the pore structure, namely the continuous network of pores that allows liquid and gas circulation inside the material [22]. The determination of the open porosity and the bulk density by hydrostatic weighing followed the EN 1936 standard [23]. This method consists of eliminating the air in the pores, followed by filling them with water using a desiccator coupled to a vacuum pump (Figure 5a), ending with the hydrostatic weighing (Figure 5b) and the determination of the immersed and still saturated samples' mass. The ratio between the volume of open pores and the apparent volume of the sample obtains open porosity. It is calculated by

Equation (1), while bulk density is obtained by the ratio between the mass of the dry sample and its apparent volume (Equation (2)).

$$P_0 = \frac{m_s - m_d}{m_s - m_h} \times 100 \quad (1)$$

$$\rho_b = \frac{m_d}{m_s - m_h} \times \rho_{rh} \quad (2)$$

where

P_0—open porosity [%];
m_s—mass of the saturated sample [g];
m_d—mass of the dry sample [g];
m_h—mass of the immersed sample [g];
ρ_b—bulk density [kg.m^{-3}];
ρ_{rh}—real density of water [kg.m^{-3}].

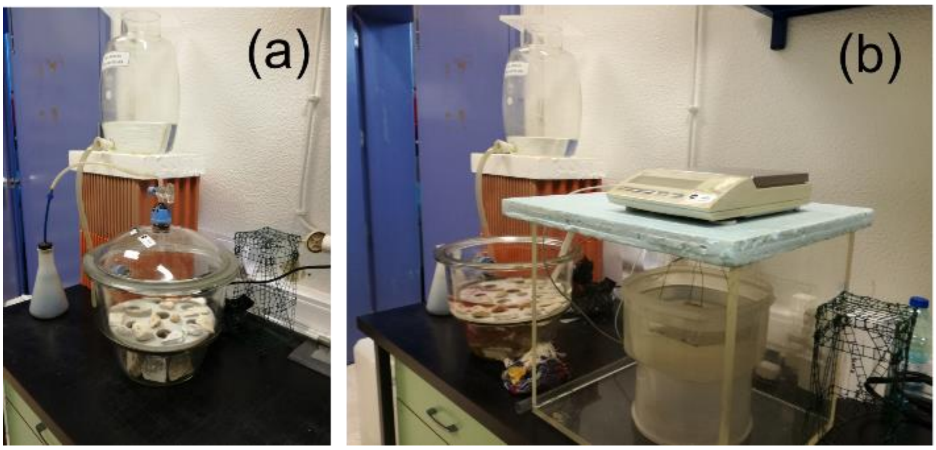

Figure 5. Determination of the open porosity and the bulk density: (**a**) air-to-water replacement procedure and (**b**) hydrostatic weighing.

Due to the small number of specimens available to perform all the programmed experimental campaigns, the open porosity test was performed in most cases on unaltered and unaffected fragments provided after the compression test. Separating the multi-layer samples into single layers made it unfeasible to test each layer, as it could crush or significantly reduce the size during the procedure; in such cases, it was decided to perform the test on the whole set.

2.3.3. Dynamic Modulus of Elasticity

When old mortars, especially the lime-based ones, are subjected to conservation interventions, the dynamic modulus of elasticity (E_d) should be considered as the experimental values obtained in characterisation must be respected to assure stiffness compatibility between the old and the new substitution mortars.

The E_d was determined according to Equation (3), which is based on the measurement of the ultrasonic pulse velocity (UPV—velocity of high-frequency sound waves) via the material [24] and is expressed in Pa. Two measurement methods, namely direct and indirect transmission methods were applied. In the direct transmission method, the transducers with pointed ends are placed on opposite sides of the sample (Figure 6b). In contrast, the transducers are placed on the same specimen surface in the indirect method. In this case, the acquisition is made by fixing the transmitter transducer at a specific point. At the same time, the receptor moves over a marked row at the surface of the specimen (Figure 6a,c) and at different distances (with a 1 cm increment for each acquisition). To perform this test, based on measuring the speed of propagation of longitudinal ultrasonic waves in microseconds, an Ultrasonic Tester Steinkamp BP-7 model was used. Equation (3) was applied.

$$E_d = v^2 \rho K \tag{3}$$

where

v—velocity of the ultrasound waves via the material or ultrasound pulse velocity (UPV);
ρ—bulk density;
K—constant depending on the coefficient of Poisson (φ).

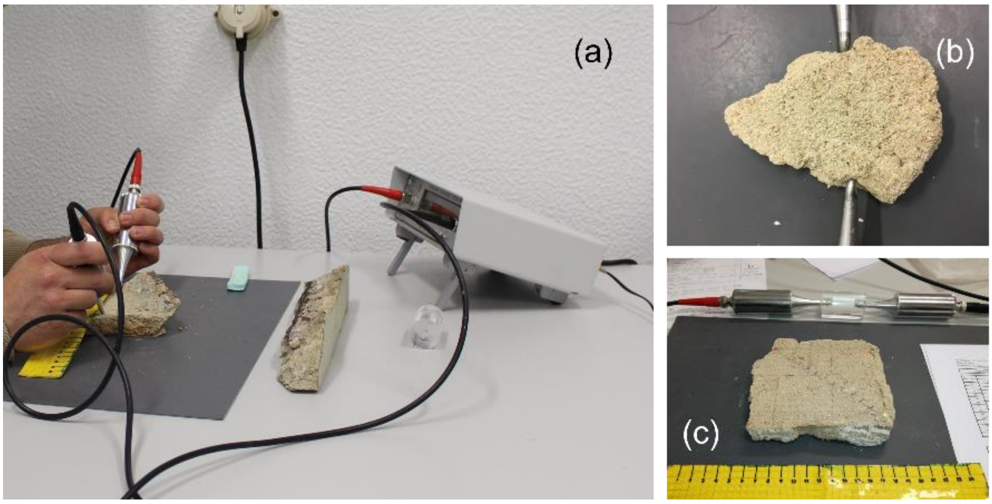

Figure 6. Determination of the dynamic modulus of elasticity: (**a**) apparatus for the determination by the indirect method; (**b**) direct method—the transducers are placed in sample's opposite surfaces; and (**c**) indirect method—sample marked with a line segment for the indirect method.

Bulk density (ρ) was obtained by Equation (2) for samples subjected to open porosity and bulk density tests. For those where this property could not be obtained, a simple mathematical approach was used to find the bulk density by dividing the mass of the specimen by the product of the average dimension of the three directions: width, length, and depth, measured with a calliper.

The value of 0.2 was assumed for the coefficient of Poisson (φ) of mortars since it has yet to be precisely known. The constant K was calculated by Equation (4) as follows:

$$K = \frac{(1+\varphi)(1-2\varphi)}{(1-\varphi)} \tag{4}$$

The indirect method was used on the multi-layer samples, which are supposed to express the UPV of the whole set and applied at the largest possible dimension over the specimen's surface. The direct method used the largest distance between the transducers to characterise the single layers.

For calculating the dynamic modulus of elasticity considering the indirect method, removing the influence of the remaining layers from the outermost one is impossible as it is directly related to the bulk density. Despite being determined by adapted methods, which proved to be adequate and reliable, the remaining layers contribute to the rise in the overall bulk density of these types of samples and, consequently, the calculated values of the modulus [8]. Thus, UPV could be a more reliable result in multi-layer samples since it does not involve calculations using the bulk density.

2.3.4. Compressive Strength

The compressive strength (CS) test was carried out to establish the limits of strength that must be respected to ensure compatibility between the old and the replacement mortars.

After the complete drying, the samples' surfaces were regularised with a high-performance rotary tool, so they were entirely in contact with the load cell during the test. A direct compression test was carried out (Figure 7), giving compressive strength values by dividing the compressive force that produces rupture of the sample by a 40 mm × 40 mm area of force application [18,19]. No samples that were less than 20 mm thick were tested. An electromechanical testing device compliant with EN 1015-11: 1999 [25], ETI, model HM-S with a load cell of 200 kN, was used. The load rate was adjusted so that failure occurred within no longer than 90 s, varying between 50 $N.s^{-1}$ and 100 $N.s^{-1}$ and, in a few cases, with a value of 200 $N.s^{-1}$.

Figure 7. Compressive strength test.

3. Results and Discussion

This section presents and discusses the results of the physical and mechanical characterisation. Table 2 summarises the results of the characterisation performed.

The results obtained for the whole sets should be analysed as indicative since the layers have different binder/aggregate ratios and, in some cases, different binders [16], which influences the results [26]. It should be noted that most of the mortars analysed were collected in interior walls, where it was common to find in buildings constructed until the 1940s fragile finishing layers based on lime–gypsum whose separation was challenging to perform per se.

3.1. Capillary Absorption and Drying of Absorbed Water

The pore system of old lime mortars is composed of a high proportion of wide pores [27], which, associated with the reduced thickness of the samples (see Table 1), leads to maximum absorption in the first few minutes. In general, and in most cases, saturation is quick and takes place within the first 24 h (1440 min or 37.95 $min^{0.5}$), but a substantial slope reduction can be observed between 6 min and 2 h, depending on the sample (see Figure 8). To be meaningful as a rate of water absorption, the range of points considered in the calculation of the Ccc must be in the straight part of the plot, as settled in EN 16322 [21]; thus, it was adjusted on a case-by-case basis, considering that this is the most significant stage of absorption. In the case of samples with hydraulic binders, the Ccc are necessarily different from those of air lime mortars, theoretically lower due to the slower absorption. The slower absorption is related to the volume and pore size of the capillary porosity and its connectivity. The capillary pores that most affect the capillary water absorption coefficient range between 0.1 µm and 5 µm [28], while in lime mortars, there is an essential range of pores which are coarser than the capillary range, with a larger diameter, up to 10 µm or more [29].

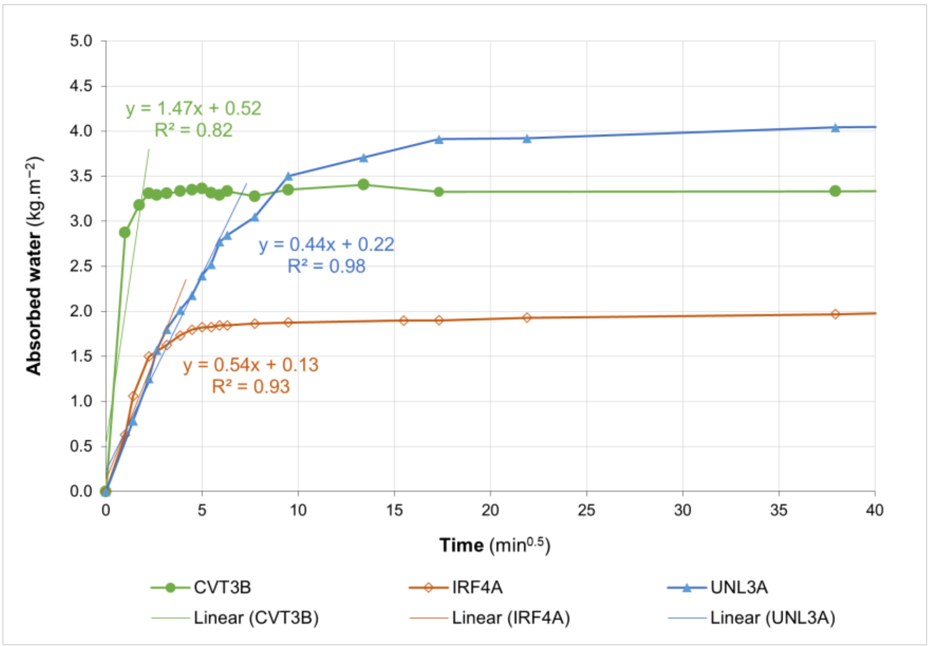

Figure 8. Capillary absorption curves of monolayer selected samples and respective slopes (Ccc).

Figure 8 shows the capillary absorption plots of three selected monolayer samples, each with a different binder (CVT3B: air lime; IRF4A: air lime and Portland cement; UNL3A: Portland cement) demonstrating typical water absorptions for that kind of binder. In the case of the air lime sample, it is evident the fast water absorption until saturation materialised by the highest Ccc (1.47 kg.m^{-2}.min$^{-0.5}$), which corresponds to maximum absorption and consequent saturation after the first 5 min, unlike what happens with the air lime–Portland cement and cement mortar samples, which, in turn, show lower absorption values than expected. In the case of the two air lime–Portland cement monolayer mortars, Ccc values vary between 0.35 kg.m^{-2}.min$^{-0.5}$ (AAC1B) and 0.59 kg.m^{-2}.min$^{-0.5}$ (IRF4A). However, the higher proportion of cement in IRF4A did not contribute to reducing Ccc, as other factors related to the aggregate and the w/b ratio may have a decisive influence. In the case of Portland cement mortars, despite the poor correlation, the trend of Ccc reduction over the analysed period is noticeable (Figure 9), probably due to the adjustment of the water/cement ratio in the mix design and the increase of cement fineness that led to the development of a less sorbing pore structure and eventually to the optimisation of the particle size distribution of aggregates that influences compactness [28]. Although these characteristics were not thoroughly investigated, it should be noted that in the case of the stone-imitating mortar EUA53-2A, which presents the lowest Ccc (0.08 kg.m^{-2}.min$^{-0.5}$), the contribution of a limestone filler, as reported by Almeida et al. [16] may have contributed to the low absorption, which is also corroborated by the lowest value of the open porosity (13.2%). These properties may also be related to another factor: the construction technique. The technique of application of these mortars, also known by the Portuguese term Marmorite, foresaw the tightening with metallic rollers still in the fresh state [30], which might have produced a porosity reduction.

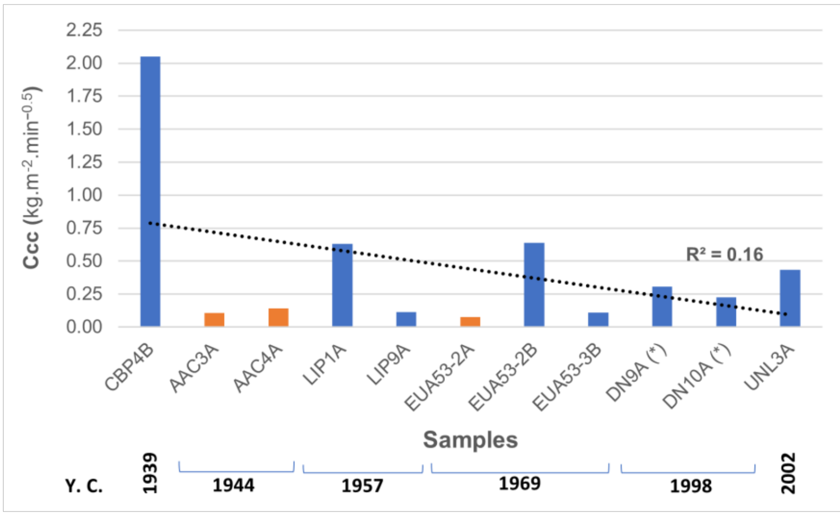

Figure 9. Evolution of Ccc of Portland cement mortars regardless of the Portland cement type (orange columns—*marmorite* samples). Y. C.—Building's year of construction completion; (*) mortars from the 1940 award-winning prize.

Sample CBP4B shows the highest Ccc amongst the Portland cement mortars (2.05 kg.m^{-2}.min$^{-0.5}$) (Figure 9), which is a value more typical of lime mortars. It also presents a high open porosity of approximately 26%, which suggests a high water-to-cement ratio or can be justified by the meagre binder-to-aggregate ratio as shown in Table 2, which also has influenced the mechanical performance, as demonstrated by the low compressive strength. Although the result refers to the CBP4AB set, we can consider the influence of the white, smooth, thin outermost layer as negligible for this parameter due to its reduced thickness.

Figure 10 shows the plot of two selected multi-layer samples in which the phased development of absorption is observed. It allowed separating the graphical events corresponding to water absorption in distinct layers of the same set. In the case of the DN12ABCD set, it was possible to separate the two absorption events by the inflexion zone. This zone corresponds to the physical separation of the half assemblies (DN12AB plus DN12CD) that were not fully bonded. The visual observation of the water rises during the test also verified the resumption of the absorption event.

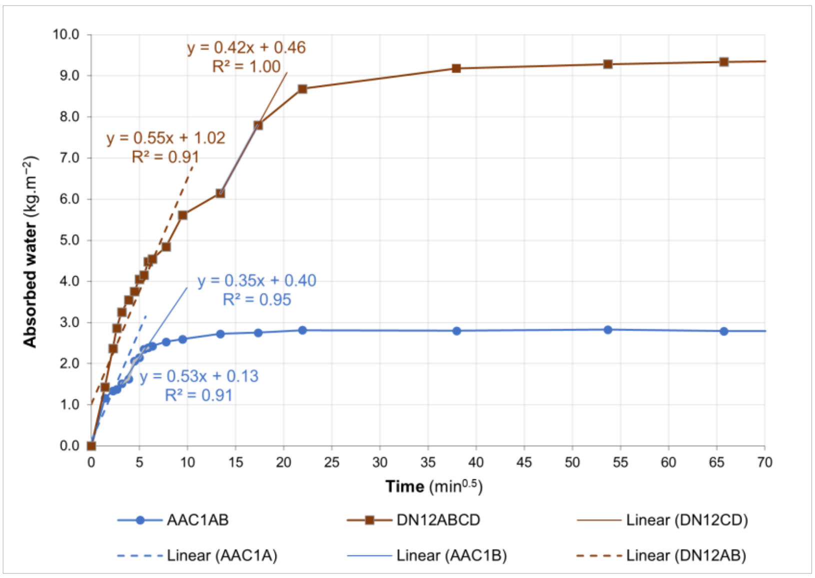

Figure 10. Capillary absorption curves of multi-layer selected samples and respective slopes (Ccc).

The multi-layer samples were tested with the exterior face of the outermost layer in contact with water; still, only in some capillary water absorption tests was it possible to verify the differentiated effect of the absorption. However, it was found that the layers subjected individually to the test present higher Ccc values than the sample set to which the respective layer belongs. Using as an example the sample CBP4AB, it was found that layer A, a smooth lime–gypsum-based plaster, lowered the Ccc due to being in contact with water. Its physical characteristics, namely the pore structure contributed to a delay in water absorption, as expected for these thin layers [8]. The effect of the interface zones that introduces some discontinuity may also contribute to reducing the absorption rate.

Among the air lime mortars, sample IRF3A has the lowest Ccc value. Still, it has a high open porosity value (33.04%). These results do not match because this sample, which has a low thickness, has a paint layer that was impossible to remove before the test. The paint layer may have delayed the water percolation.

Excluding the previously mentioned sample because it is an isolated case, and once the binder of the mortars is known, it is possible to establish ranges of Ccc values for each type of mortars. Hence, based on the results for individual samples and monolayers, we can group them as follows:

- White smooth thin layers (gypsum–lime-based): 0.16 < Ccc < 0.70 ($kg.m^{-2}.min^{-0.5}$);
- Air lime mortars: 0.69 < Ccc < 2.22 ($kg.m^{-2}.min^{-0.5}$);
- Air lime–Portland cement mortars: 0.35 < Ccc < 0.59 ($kg.m^{-2}.min^{-0.5}$);
- Portland cement mortars:
    (a) Cementitious stone-imitating mortars—referred to as *Marmorite*: 0.08 < Ccc < 0.14 ($kg.m^{-2}.min^{-0.5}$);
    (b) Remaining mortars (excluding CBP4B): 0.11 < Ccc < 0.64 ($kg.m^{-2}.min^{-0.5}$).

Figure 11 shows the typical drying curve from which it was possible to compute the D1 rate, in this case, for the DN12ABCD set. For the same set, Figure 12 shows the curve as a function of the square root of time from which the value for D2 was estimated.

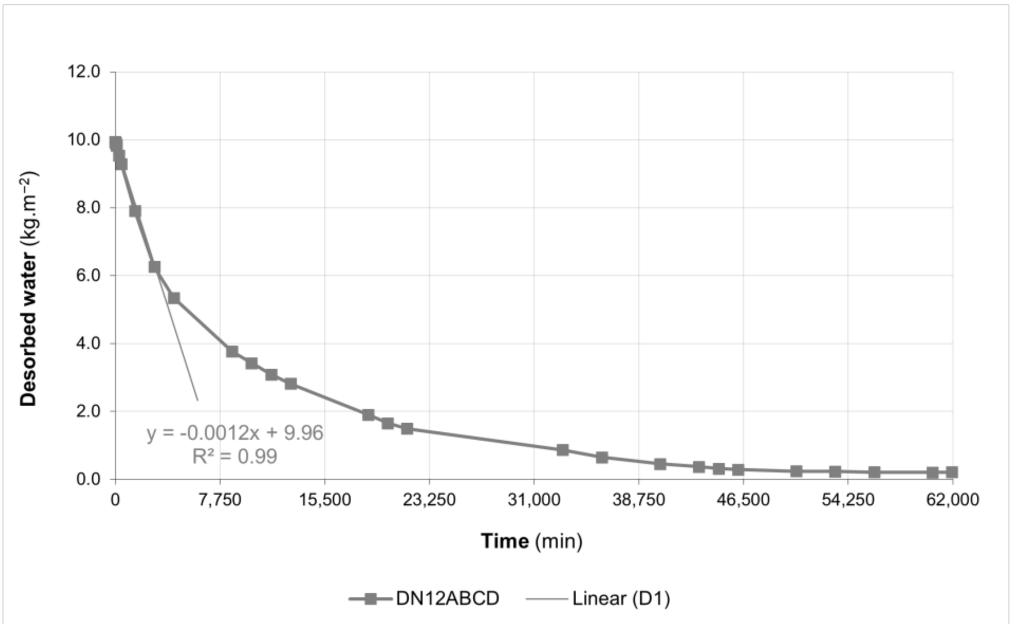

Figure 11. Drying curve and first drying phase D1 representation.

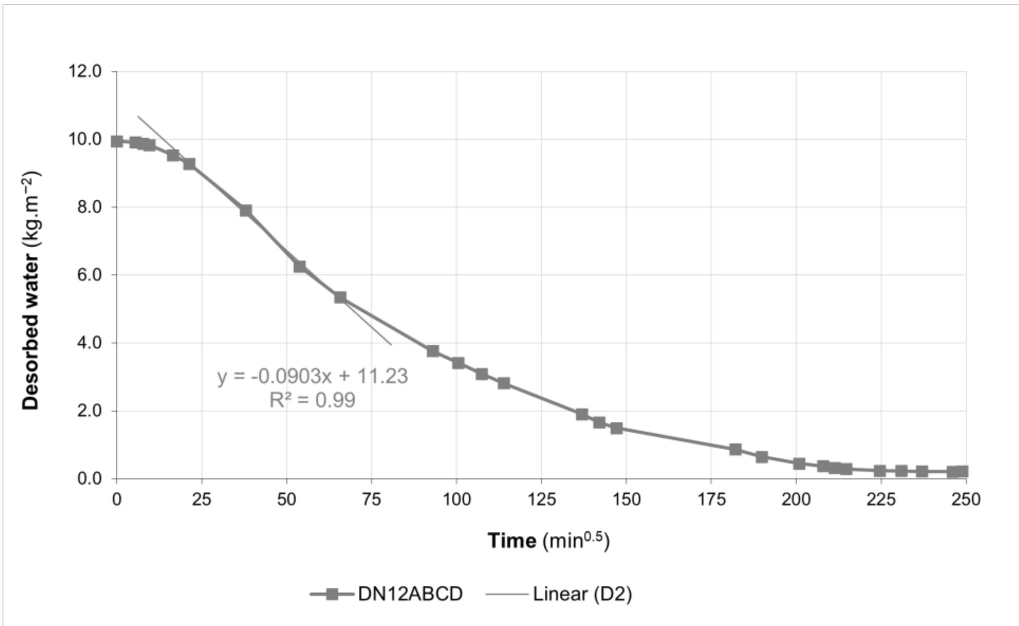

Figure 12. Drying curve and second drying phase D2 representation.

Figure 13 shows the distribution of the drying rate by liquid water transport D1 and the drying rate of the second phase by mixed liquid water and water vapour transport, D2, in monolayer samples and samples' individualised layers.

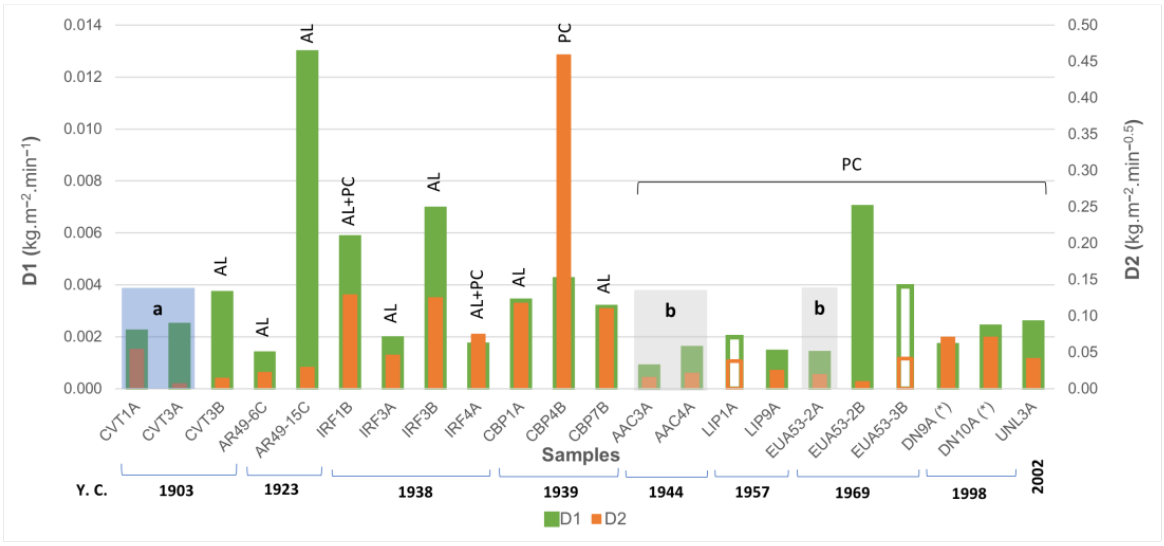

Figure 13. Comparison of the two main drying phases (D1 and D2) of individualised and monolayer renders (columns without colour fill) and plasters (coloured columns). Notation: a—white smooth thin plasters (gypsum–air-lime-based plasters); b—stone-imitating mortars; AL—air lime; PC—encompasses all types of Portland cement (ordinary; composite and white Portland cement); Y. C.—Building's year of construction completion or mortars' execution year; (*) mortars from the 1940 award-winning prize.

Lower D1 values were generally observed in Portland cement mortars, except for sample EUA53-2B. The air lime mortars show higher D1, denouncing a higher capacity of liquid water transport during the drying process to the surface of the sample followed by evaporation with some variability, as shown by the D2 values, being, in general, higher for air lime mortars which are consistent with their high Ccc and open porosity, possibly related to the predominance of macropores, typical for air lime mortars. Sample AR49-15C presents a low D2 value, which points to a low evaporation rate, producing some retention of vapour inside the pores. As there is no evidence of salts or other contaminants [16], it cannot be said that they have influenced the vapour permeability. Mosquera et al. [31] demonstrated that the porosity does not significantly influence the diffusivity. Instead, the pore radius controls the diffusivity, which, in our case, is unknown and may explain the reduced value of D2.

The same happens in the other air lime mortars built in 1903 and 1923 regarding water vapour diffusion. However, the low thickness of the samples may imply that most water is removed in the liquid phase, and the remaining humidity left for the phase related to water vapour diffusivity is low. The air lime–Portland cement or simply lime–cement (also labelled as AL + PC in some plots) samples analysed in the case study IRF (1938) show a difference in the D1 value. The sample IRF1B presents less cement in the lime-to-cement ratio than the sample IRF4A. Lime–cement mortars reduce both their pore volume and their pore size as cement content in the mix increases [31], as expected in these mortars, which present open porosity values of 31.12% and 22.68% (IRF1B and IRF4A, respectively).

As for the white smooth thin layer samples, they denote a very compact microstructure. In the case of sample CVT3A, the capillary absorption coefficient is low, but the drying rate D1 is high. This fact indicates the presence of some pores larger than the capillary range, which facilitate liquid water drying but do not contribute to absorption.

Finally, Portland cement mortars, applied from the 1930s onwards, show lower D1 rate values for stone-imitating mortars, consistent with the lower Ccc. Between renders and plasters, there are no considerable differences in terms of drying rates, which is also the case for Ccc. Among all the samples with Portland cement binder, CBP4B has the highest D2 ratio, and the sample EUA53-2B has the highest D1 ratio and the lowest D2 ratio. In the first case, the sample shows simultaneously relatively high D1 and very high D2, indicating high transport of liquid water to the sample's surface, followed by very high evaporation, consistent with their high Ccc and medium open porosity results. In the second case, considering the open porosity result (20.13%) and the low Ccc value (0.64 $kg.m^{-2}.min^{-0.5}$), the high D1 rate should be related to the pore size and not to its volume.

The drying of the multi-layered samples, as observed in Figure 14, only demonstrates the result for the whole set drying; not possible to individualise the drying effect for each layer in the plot. Still, it is possible to observe that the AR49-8AB set has the highest D1, which is consistent with the type of binder since both layers (A and B) have air lime as a binder. Samples with at least one air lime layer show relatively high D1 values, unlike the AR49-11AB set, as layer A, a lime–gypsum-based, has almost the same thickness as layer B. Layers with gypsum–air lime-based binders seem to produce the effect of lowering the drying rates, particularly the D2 rate, as can be seen in the CBP4AB set by comparison to sample CBP4B (Figure 13).

3.2. Mechanical Characterisation

The individualised (single layer) and monolayer dynamic modulus of elasticity results are the lowest for the lime mortars (until the end of the 1930s) since these samples have higher porosity, being more permeable to the fluid circulation, including air and, therefore, with reduced UPV. In opposition, Portland cement mortars exhibit the highest values (Figure 15) since they present higher compactness than air lime mortars. The white smooth thin plasters present E_d values relatively close (ca. 4601 MPa—CVT3A; 4495 MPa—DN19A), which indicates identical porosity values in both samples, although the porosity of sample DN19A is not known. It is also evident that the influence of Portland cement content on the compactness of the air lime-Portland cement mortars, in which the increase of E_d is notorious for a lime-to-cement ratio above 1:1 (samples DN12B and DN19D). Although sample DN12B presents a lower ratio than sample DN19D, i.e., a higher proportion of Portland cement, the velocity of ultrasound wave propagation is lower. Regarding the sample DN19D, which incorporates ground granulated blast furnace slag (GGBFS), as demonstrated in [16], once this kind of addition is finer than the Ordinary Portland cement (OPC) clinker, it led to a less porous structure with fewer capillary pores. Consequently, there would also be a finer distribution of pores [32].

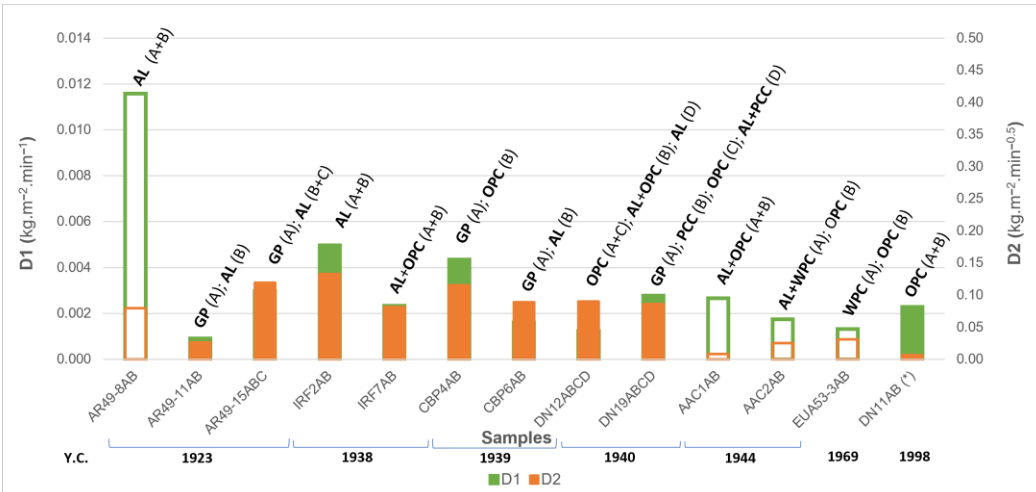

Figure 14. Comparison of the two main drying phases (D1 and D2) of multi-layer renders (columns without colour fill) and plasters (coloured columns). Notation: Binder types per layer (layers A–D in brackets)—GP: Gypsum—air lime-based plasters (white smooth thin plasters); AL: Air lime; OPC: Ordinary Portland cement; WPC: White Portland cement; PCC: Portland composite cement; Y. C.—Building's year of construction completion or mortars' execution year; (*) mortars from the 1940 award-winning prize.

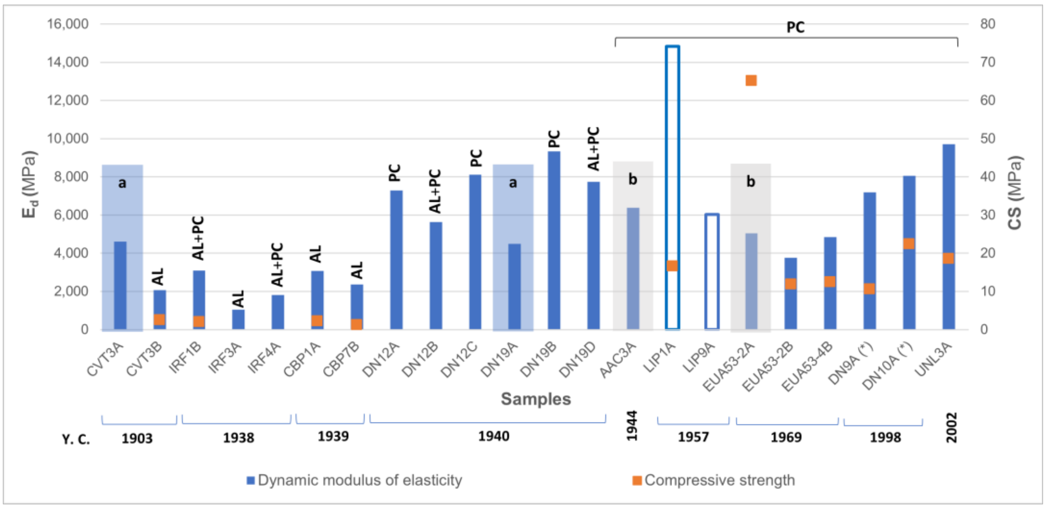

Figure 15. Dynamic modulus of elasticity (E_d) and compressive strength (CS) plot of the individualised and monolayer renders (columns without colour fill) and plasters (blue-coloured columns). Notation: a—white smooth thin plasters (gypsum–air-lime-based plasters); b—stone-imitating mortars; Binder types—AL: Air lime; PC: encompasses all types of Portland cements (ordinary; composite and white Portland cements); Y. C.—Building's year of construction completion or mortars' execution year; (*) mortars from the 1940 award-winning prize.

Regarding the E_d, the most significant contrast is verified in sample LIP1A, relative to its Portland cement counterparts by having the highest value. Compared with samples DN19B and UNL3A, whose UPV values are of the same magnitude, LIP1A presents the highest E_d value, which can be influenced by the aggregate content producing higher compactness, as it is the smallest of the three samples. This one and LIP9A are both rendering mortars of the same building. However, this sample shows higher compactness, which is inconsistent with its UPV result.

The bulk density calculation used may explain this discrepancy of values. The calculation of this property should be, whenever possible, performed via tests, avoiding the lack of accuracy that is characteristic of calculations involving the size and mass of the specimen directly, i.e., dividing the mass of the specimen by the product of the average dimension of width, length, and depth. In these cases, the bulk density should only be considered an approximate value to the real one and, consequently, an approximate value of the E_d.

The remaining samples, whose binder is only composed of Portland cement, do not show significant disparities in the E_d values, presenting higher ranges of values than the lime mortars but sometimes lower than the mixed air lime and Portland cement samples.

Regarding the compressive strength results, the difference between lime and Portland cement mortars is clear. As expected, the open porosity (Figure 16) is consistent with the type of mortars, i.e., lime mortars have higher open porosity and consequently lower compressive strength. If the sample EUA53-2A is excepted, the average value of the compressive strength of the Portland cement samples tested is approximately eight times higher than that of the air lime mortars. Sample EUA53-2A is an outlier since its compressive strength value is the highest (65.15 MPa). Besides having an average thickness of 10 mm, which was an a priori condition to exclude it from being tested, it was found that the rupture stress was conditioned by the size and nature of the quartzite aggregates, some with the major axis dimension close to the thickness of the tested sample.

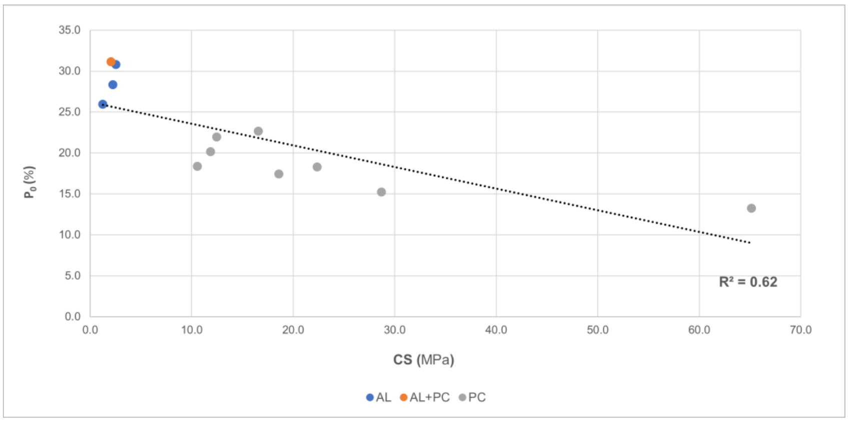

Figure 16. Correlation between open porosity (P_0) and compressive strength (CS). AL—air lime; PC—encompasses all types of Portland cement (ordinary; composite and white Portland cement).

Figure 17 shows the dynamic modulus of elasticity and compressive strength plots for the multi-layer samples. Most sets containing Portland cement have the highest values of the modulus of elasticity. A general tendency is observed for the dynamic modulus of elasticity and compressive strength to increase over the period under analysis, influenced by the introduction of Portland cement as in the case of individualised and monolayer renders and plasters (Figure 15). The sets containing lime mortars maintain the trend observed for the single layers tested, which is that of lower modulus of elasticity values for renders until the 1940s. It must be mentioned that the application of the indirect method to calculate the dynamic modulus of elasticity of each layer is, however, conditioned by the presence of the successive layers, and it is not possible to quantify the influence of each layer on this mechanical property results.

The compressive strength results show that the sets essentially composed of lime as a binder present the lowest values (AR49-15ABC and CBP6AB). The presence of the superficial white smooth thin layer, i.e., the lime–gypsum-based layer (A), should not influence the results that much, as low values of compressive strength are expected for these materials, as demonstrated by Freire et al. [8]. The authors reported average values for compressive strength of 2.26 MPa, which is an intermediate value to those tested on samples

AR49-15ABC (1.77 MPa) and CBP6AB (2.69 MPa). The Portland cement mortars show compressive strength values above 30 MPa, except for the CBP4AB set, whose low result directly correlates with the high porosity and the water absorption. The pore structure will necessarily influence that result. A possible explanation for this performance may lie in the water-to-cement ratio employed and either in the hydration or curing conditions of the sample, which are unknown at this point.

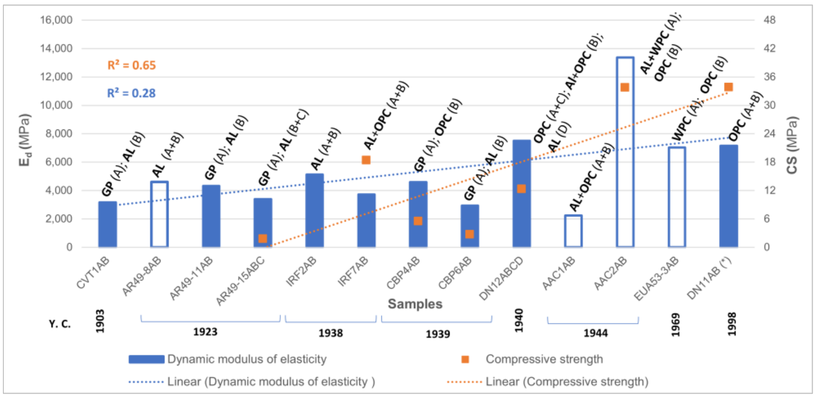

Figure 17. Dynamic modulus of elasticity (E_d) and compressive strength (CS) plot of multi-layer renders (columns without colour fill) and plasters (blue-coloured columns). Notation: Binder types per layer (layers A – D in brackets))—GP: Gypsum–air lime-based plasters (white smooth thin plasters); AL: Air lime; OPC: Ordinary Portland cement; WPC: White Portland cement; Y. C.—Building's year of construction completion or mortars' execution year; (*) mortars from the 1940 award-winning prize.

The lime–cement sample sets, IRF7AB and DN12ABCD, show compressive strength values between 12 and 19 MPa. These values are closer to Portland cement mortars. However, IRF1B, another lime–cement mortar, shows a value closer to lime mortars (2.09 MPa) (Figure 15 and Table 2). Some authors have concluded that the presence of lime implies variations in compressive strength; that is, the presence of lime in lime–cement mortars reduces compressive strength [26,33], but also the binder-to-aggregate ratio and the type of cement [34] can influence the result of compressive strength. In this case, not only does the binder-to-aggregate ratio vary, but the type of cement used and its physical and mechanical properties still need to be known. It is, however, known that the fineness of older cement is higher than the current ones [35]. However, the combination of all these different parameters makes it difficult to have a more consistent interpretation based on the results obtained for these three samples.

4. Requirements for a Compatible Restoration

The requirements should be considered case-by-case since the plasters and renders analysed are from buildings constructed in different periods throughout the 20th century. The different types of binders, compositions and formulations require such an approach. However, the work already carried out by other authors that established or analysed compatibility parameters [8,11,36], whose application is more relevant in heritage buildings, should be considered. Nevertheless, the parameters investigated in the laboratory should be respected, advising the use of mortars with identical binder characteristics and proportions, similar grain size distribution and aggregate mineralogy. In the case of Portland cement mortars, since the specifications of cement used are not known, the use of OPC (and WPC in the due cases) is proposed in all cases, despite evidence of the use of composite cement [16] in two samples, provided that they do not exceed the quantified values for the physical and mechanical characterisation.

Table 3 shows the ranges of values obtained for the assessed characteristics to be considered in the compatibility requirements. The results are organised by binder and coating type.

Table 3. Ranges of values obtained for the assessed physical and mechanical characteristics to be considered in the compatibility requirements.

Type of Binder	**Type of Mortar**	**Case Study**	**Quantitative Ranges**							**Mortar Mix Design**
			Physical Characteristics and Water Behaviour				**Mechanical Characteristics**			**Binder to Aggregate Ratio**
			Ccc	**D1**	**D2**	**P_0**	**UPV**	**Ed**	**CS**	**(b/a) (3)**
			$kg.m^{-2}.min^{-0.5}$	**$kg.m^{-2}.min^{-1}$**	**$kg.m^{-2}.min^{-0.5}$**	**%**	**$m.s^{-1}$**	**MPa**		
gypsum-air lime	Plasters (w.s.t.l.)	CVT (1903)	0.16–0.39	0.0021–0.0024	0.0066–0.539	40.5 (1)	2011 (1)	4601 (1)		n.a.
		DN (1940)	0.70 (1)				1995 (1)	4495 (1)		n.a.
Air lime	Plasters	CVT (1903)	1.22–1.47	0.0036 (1)	0.0144 (1)	30.8 (1)	1165 (1)	2073 (1)	2.6 (1)	1:0:4.3–1:0:7.8
		AR49 (1923)	1.68 (1)	0.0129 (1)	0.0294 (1)	30.8 (1,2)	1477 (1,2)	3385 (1,2)	1.8 (1)	1:0:7.9 (1)
		DN (1940)	0.58 (1)			23.1–26.8 (1)	1848–2171 (1)	5641–7744 (1)		1:1:6–1:2:15.1
		IRF (1938)	0.76–1.62	0.0050–0.0068	0.1252–0.1330	31.0–33.0	831–1868 (2)	1055–5130 (2)		1:0:4.2–1:0:8
		CBP (1939)	1.25–2.22	0.0016–0.0033	0.0892–0.1176	28.3–29.5	1208–1419	2360–3078	1.3–2.7	1:0:8.4–1:0:11.2
	Renders	AR49 (1923)	0.69–0.83	0.0013–0.0116	0.0225–0.0798	26.9–32.1	1946 (1,2)	4463 (1,2)		1:0:5.8–1:0:11.2
Lime–cement	Plasters	IRF (1938)	0.54–0.63	0.0016–0.0057	0.0813–0.1291	22.7–31.1	1023–1478 (1,2)	1814–3722 (1,2)	2.1–18.4	1:0.1:7–1:0.4:5.4
	Renders	AAC (1944)	0.35–0.53	0.0027 (1,2)	0.0255 (1,2)	31.2 (1,2)	1198 (1,2)	2233 (1,2)		1:0.2:2.7–1:02:6.1
Portland cement (*)	Plasters	CBP (1939)	2.05 (1)	0.0041 (1)	0.4590 (1)	33.17 (1,2)	1726 (1,2)	4579 (1,2)	5.5 (1,2)	0:1:20.3 (1,2)
		DN (1940)	0.22–0.50			17.6–22.2	1984–2317	7151–9346	10.6–33.8	0:1:4.2–0:1:25.2
		EUA (1970)	0.64 (1)	0.0069 (1)	0.0096 (1)	20.1–21.9	1461–1676	3764–4844	11.9–12.5	0:1:6.7–0:1:11.5
		UNL (2002)	0.44 (1)	0.0025 (1)	0.0414 (1)	17.4 (1)	2336 (1)	9702 (1)	18.6 (1)	0:1:10.2 (1)
	Plasters (s.i.m.)	AAC (1944)	0.10–0.14	0.0008–0.0015	0.0151–0.0211		1863 (1)	6383 (1)		0:1:1.9–0:1:3
	Renders (s.i.m.)	EUA53 (1970)	0.08 (1)	0.0013 (1)	0.0197 (1)	13.2 (1)	1581 (1)	5050 (1)		0:1:3.7 (1)
	Renders	AAC (1944)	0.19 (1,2)	0.0017 (1,2)	0.0255 (1,2)	14.1 (1,2)	2655 (1,2)	13370 (1,2)	33.8 (1,2)	n.a.
		LIP (1958)	0.11–0.63	0.0013–0.0020	0.0252–0.0377	22.6 (1)	1534–2282	6041–14842	16.6 (1)	0:1:6.6–0:1:7.6
		EUA (1970)	0.07 (2)–0.11	0.0013 (2)–0.0039	0.0310 (2)–0.0415	15.2 (1)	1917 (1,2)	7025 (1,2)	28.7 (1)	0:1:4.9 (1)

Notation: (1) Range of values not defined because only one sample or specimen was tested; (2) Value obtained in a multi-layer sample set or average value of all sets tested in the same case study. Applicable only when the characteristics of a single layer are not known or when only the sets of layers with the same type of binder were characterised, disregarding the white smooth thin plaster and stone-imitating mortar layers (in this case the layer A from the set AAC2AB); (3) according to [16]; (*) encompasses all types of Portland cement; b/a—binder to aggregate ratio by mass [hydrated lime: Portland cement (regardless the type): aggregate]; n.a.—not applicable.; w.s.t.l.—white smooth thin layers; s.i.m.—stone-imitating mortar. Blank fills to non-determined characteristics.

Chemical and mineralogical characterisation should be included in the context of compatibility; however, these aspects do not fall within the scope of this manuscript.

5. Conclusions

This paper deals with the physical and mechanical characterisation of fifty-three mortar samples from buildings built in Lisbon in the 20th century and awarded with one of the most significant architectural prizes in Portugal. The results allow compatibility criteria to be established if restoration or conservation actions are required.

The originality and value of this built heritage require developing preservation strategies that involve proactive and routine maintenance, followed by occasional interventions that do not de-characterise the surroundings. To this end, adopting new materials compatible with the originals is essential. Constant monitoring of the state of conservation is also essential to establish the basis for early detection of defects, thus minimising the need for physical interventions.

Via physical and mechanical characterisation, the study made it possible to have a better knowledge of the properties of mortars applied throughout the 20th century, in a period that needs further research. With this study, it was possible to clarify how the techniques evolved, knowing from the start the age of the case studies and, consequently, the age of the samples studied. Despite the difficulty in obtaining samples of sufficient size to perform all the programmed tests, it was still possible to characterise mortars with a high degree of reliability.

The main conclusions are as follows:

- The air lime mortars existing in the oldest buildings, between 1903 and 1944, have the highest values of capillary absorption and simultaneously the highest drying rates and present the lowest values of compressive strength and dynamic modulus of elasticity, which is expected for this type of mortars.
- The blended lime–cement mortars in the buildings constructed between 1938 and 1944 have intermediate capillary absorption and drying rates. Compressive strength values of blended lime–cement mortars and multi-layer mortars with different binders in each layer are variable. In general, an increase in Ed values is due to the introduction of Portland cement.
- The Portland cement mortars applied in buildings erected after 1939 show the lowest values of capillary absorption and the highest values of mechanical strength.
- The results of lime–gypsum-based plasters align with those found in the literature for white smooth thin layers applied in Portugal.
- The stone-imitating mortars (Marmorite type) showed the lowest capillary absorption and, consequently, the lowest open porosities, which points to the governance of the construction technique on reducing these parameters, either by incorporating fillers or by tightening the mortar during their application.
- No significant differences were found in the physical and mechanical characteristics between the samples of renders and plasters; thus, being intended for internal or external application was not a crucial parameter for the choice of the material.
- The physical and mechanical values obtained in this study constitute a basis for the definition of compatibility requirements for restoration mortars in the group of buildings studied.
- For compatibility purposes, the range of values obtained on multi-layer samples, though indicative, should be considered a good approximation of the whole coating properties if there is no possibility of individualising each layer and testing them independently.

6. Future Research

Future research should encompass not only a broader range of mortars within the buildings already under study, where feasible sample collection is possible but also extend to other award-winning structures that remain unexplored, contingent upon the willingness of property owners to grant access and authorisation.

Moreover, the critical importance of including Portuguese standard buildings from other regions in these research endeavours is worth noting. Comparing the mortars and construction techniques employed in award-winning buildings to those found in standard structures from the 20th century can yield valuable insights. This comparative approach will contribute significantly to our understanding of the evolution of construction practices and the materials used throughout this pivotal century in architectural history.

This expanded passage underscores the necessity of ongoing research and the potential benefits of comparing award-winning buildings to standard constructions for a comprehensive understanding of 20th-century mortar properties.

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References

1. CML. *Prémio Valmor [Valmor Prize]*; Silva, A.P., Ed.; Câmara Municipal de Lisboa: Lisbon, Portugal, 2004.
2. Pedreirinho, J.M. A Critical History of the Valmor Prize. *Argumentum* **2018**, 1–175.
3. Apostolopoulou, M.; Moropoulou, A. Mortars for Restoration: Set-up Parameters and Developing Mortar Design Areas. In *Conserving Stone Heritage. Cultural Heritage Science*; Gherardi, F., Maravelaki, P.N., Eds.; Springer: Cham, Switzerland, 2022. [CrossRef]
4. Moropoulou, A.; Bakolas, A.; Moundoulas, P.; Aggelakopoulou, E.; Anagnostopoulou, S. Design and evaluation of restoration mortars for historic masonry using traditional materials and production techniques. *MRS Online Proc. Libr.* **2002**, *712*, 27. [CrossRef]
5. Veiga, M.R.; Silva, S. Mortars. In *Long-Term Performance and Durability of Masonry Structures, Chapter 6. Degradation Mechanisms, Health, Monitoring and Service Life Design*, 1st ed.; Ghiassi, B., Lourenço, P.B., Eds.; Elsevier Ltd.: Amsterdam, The Netherlands, 2019; pp. 169–208.
6. Almeida, L.; Silva, A.S.; Veiga, M.d.R.; Mirão, J.; Vieira, M. 20th-Century Award-Winning Buildings in Lisbon (Portugal). Study of Plasters, Rendering, and Concrete Materials Aiming Their Sustainable Preservation. *Buildings* **2021**, *11*, 359. [CrossRef]
7. Veiga, M.R.; Velosa, A.; Magalhães, A. Evaluation of mechanical compatibility of renders to apply on old walls based on a restrained shrinkage test. *Mater. Struct.* **2007**, *40*, 1115–1126. [CrossRef]
8. Freire, M.T.; Santos Silva, A.; Veiga, M.R.; Brito, J. Studies in ancient gypsum based plasters towards their repair: Physical and mechanical properties. *Constr. Build. Mater.* **2019**, *202*, 319–531. [CrossRef]
9. Papayanni, I. Design and manufacture of repair mortars for interventions on monuments and historical buildings. In *Workshop "Repair Mortars for Historic Masonry"*; RILEM, Ed.; TC RMH: Delft, The Netherlands, 25–28 January 2005.
10. Veiga, M.R. Conservation of historic renders and plasters: From laboratory to site. Historic Mortars: Characterisation, Assessment, Conservation and Repair. In *RILEM Bookseries*; Válek, J., Groot, C., Hughes, J.J., Eds.; Springer: Berlin/Heidelberg, Germany, 2013; Volume 7, 464p. [CrossRef]
11. Veiga, M.R.; Fragata, A.; Velosa, A.L.; Magalhães, A.C.; Margalha, G. Lime-based mortars: Viability for use as substitution renders in historical buildings. *Int. J. Archit. Herit.* **2010**, *4*, 177–195. [CrossRef]
12. Groot, C.; Veiga, R.; Papayianni, I.; Hees, R.V.; Secco, M.; Alvarez, J.I.; Faria, P.; Stefanidou, M. RILEM TC 277-LHS report: Lime-based mortars for restoration–a review on long-term durability aspects and experience from practice. *Mater. Struct.* **2022**, *55*, 245. [CrossRef]
13. Nogueira, R.; Pinto, A.P.F.; Gomes, A. Design and behavior of traditional lime-based plasters and renders. Review and critical appraisal of strengths and weaknesses. *Cem. Concr. Compos.* **2018**, *89*, 192–204. [CrossRef]

14. Zacharopoulou, G. The Renascence of Lime Based Mortar Technology. An Appraisal of a Bibliographic Study. In *Compatible Materials Recommendations for the Preservation of European Cultural Heritage*; Technical Chamber of Greece: Athens, Greece, 1998; pp. 89–114.
15. Callebaut, K.; Elsen, J.; Van Balen, K.; Viaene, W. Nineteenth century hydraulic restoration mortars in the Saint Michael's Church (Leuven, Belgium) Natural hydraulic lime or cement? *Cem. Concr. Res.* **2001**, *31*, 397–403. [CrossRef]
16. Almeida, L.; Silva, A.S.; Veiga, R.; Mirão, J. Composition of renders and plasters of award-winning buildings in Lisbon (Portugal): A contribution to the knowledge of binders used in the 20th Century. *Int. J. Archit. Herit.* **2023**. [CrossRef]
17. Veiga, R.; Magalhães, A.; Bosilikov, V. Capillarity tests on historic mortar samples extracted from site. Methodology and compared results. In Proceedings of the 13th International Masonry Conference, Amsterdam, The Netherlands, 4–7 July 2004; Martens, D., Vermeltfoort, A., Eds.; Eindhoven University of Technology: Eindhoven, The Netherlands, 2004.
18. Magalhães, A.; Veiga, R. Physical and mechanical characterisation of ancient mortars. Application to the evaluation of the state of conservation. *Mater. Construcción* **2008**, *59*, 61–77. [CrossRef]
19. Válek, J.; Veiga, R. sCharacterisation of mechanical properties of historic mortars—Testing of irregular samples. *Adv. Archit. Ser.* **2005**, *20*, 365–374.
20. *CEN-EN 15801:2009*; Conservation of Cultural Property. Test Methods. Determination of Water Absorption by Capillarity. European Standard: Brussels, Belgium, 2009.
21. *CEN-EN 16322:2013*; Conservation of Cultural Heritage—Test, Methods—Determination of Drying Properties. European Standard: Brussels, Belgium, 2013.
22. Damas, A.L.; Veiga, M.R.; Faria, P.; Santos Silva, A. Characterisation of old azulejos setting mortars: A contribution to the conservation of this type of coatings. *Constr. Build. Mater.* **2018**, *171*, 128–139. [CrossRef]
23. *CEN-EN 1936:2006*; Natural Stone Test Methods—Determination of Real Density and Apparent Density, and of Total and Open Porosity. European Standard: Brussels, Belgium, 2006.
24. *CEN-EN 12504-4:2007*; Testing Concrete in Structures—Part 4: Determination of Ultrasonic Pulse Velocity. European Standard: Brussels, Belgium, 2007.
25. *CEN-EN 1015-11:1999*; Methods of Test for Mortar for Masonry. Part 11: Determination of Flexural and Compressive Strength. European Standard: Brussels, Belgium, 1999.
26. Arandigoyen, M.; Alvarez, J.I. Pore structure and mechanical properties of cement-lime mortars. *Cem. Concr. Res.* **2007**, *37*, 767–775. [CrossRef]
27. Lanas, J.; Alvarez, J.I. Masonry repair lime-based mortars: Factors affecting the mechanical behavior. *Cem. Concr. Res.* **2003**, *33*, 1867–1876. [CrossRef]
28. Santos, A.R.; Veiga, M.R.; Santos Silva, A.; Brito, J. Microstructure as a critical factor of cement mortars' behaviour: The effect of aggregates' properties. *Cem. Concr. Compos.* **2020**, *111*, 103628. [CrossRef]
29. Santos, A.R.; Veiga, M.R.; Santos Silva, A.; Brito, J.; Álvarez, J.I. Evolution of the microstructure of lime based mortars and influence on the mechanical behaviour: The role of the aggregates. *Constr. Build. Mater.* **2018**, *187*, 907–922. [CrossRef]
30. Laboratório Nacional de Engenharia Civil. *Execução de Marmorites [Marmorite Execution]*; Especificações E 5-1952; LNEC: Lisboa, Portugal, 1952.
31. Mosquera, M.J.; Silva, B.; Prieto, B.; Ruiz-Herrera, E. Addition of cement to lime-based mortars: Effect on pore structure and vapor transport. *Cem. Concr. Res.* **2006**, *36*, 1635–1642. [CrossRef]
32. Loke, C.K.; Lehane, B.; Aslani, F.; Majhi, S.; Mukherjee, A. Non-Destructive evaluation of mortar with ground granulated blast furnace slag blended cement using ultrasonic pulse velocity. *Materials* **2022**, *15*, 6957. [CrossRef]
33. Haach, V.G.; Vasconcelos, G.; Lourenço, P.B. Assessment of compressive behavior of concrete masonry prisms partially filled by general mortar. *J. Mater. Civ. Eng.* **2014**, *26*, 04014068. [CrossRef]
34. Ramesh, M.; Azenha, M.; Lourenço, P.B. Quantification of impact of lime on mechanical behaviour of lime cement blended mortars for bedding joints in masonry systems. *Constr. Build. Mater.* **2019**, *229*, 116884. [CrossRef]
35. Vidovszky, I.; Pintér, F. An Investigation of the Application and Material Characteristics of Early 20th-Century Portland Cement-Based Structures from the Historical Campus of the Budapest University of Technology and Economics. *Int. J. Archit. Herit.* **2018**, *14*, 358–375. [CrossRef]
36. Pacheco-Torgal, F.; Faria, J.; Jalali, S. Some considerations about the use of lime–cement mortars for building conservation purposes in Portugal: A reprehensible option or a lesser evil? *Constr. Build. Mater.* **2012**, *30*, 488–494. [CrossRef]

4.5. PHYSICAL AND MECHANICAL CHARACTERISATION OF CONCRETE MATERIALS

The following paper deals with the physical and mechanical characteristics of reinforced concrete materials from twelve award-winning buildings constructed between the 1930s and the end of the 20th century in Lisbon, Portugal.

The use of concrete materials in Portugal, namely reinforced concrete, began in the 19th century. However, only during the 20th century, the increase in the application of concrete, alongside the use of hydraulic binders, led to a disruption of traditional construction techniques and enhanced generalised application in concrete structures, combining aesthetics with functionality.

The results are vital to evaluate their durability and will contribute to the knowledge of the current state of conservation of these materials and will allow an understanding of the evolution in the application of national regulations during this period.

The results point to an evolution in the characteristics over the period under analysis, which embodies the application of the national regulations. The physical and mechanical properties of the analyzed concrete materials reproduce an evolution towards the safety and durability requirements imposed by the national regulations on account of the advancement in the knowledge of structural performance and the scientific knowledge acquired throughout the 20th century. Moreover, the results showed, in general, a good durability condition, as far as the physical and mechanical characteristics point out to a good performance, not indicating degradation, considering the age of the buildings and that they are still in use. However, the 1987 award-winning building demonstrated that its overall performance could compromise durability, requiring monitoring actions to prevent degradation.

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Physical and Mechanical Properties of Reinforced Concrete from 20th-Century Architecture Award-Winning Buildings in Lisbon (Portugal): A Contribution to the Knowledge of Their Evolution and Durability

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Abstract: The use of concrete materials in Portugal, namely reinforced concrete, began in the 19th century. However, during the 20th century, the increase in the application of this composite material, alongside the use of hydraulic binders, led to a disruption of traditional construction techniques and enhanced generalized application in concrete structures, combining aesthetics with functionality. In this paper, the authors will present and discuss several physical and mechanical characteristics of reinforced concrete materials from 12 award-winning architectural buildings constructed between the 1930s and the end of the 20th century in Lisbon, Portugal. These results are vital to evaluate their durability, as those buildings have an undiscussable heritage value in the context of 20th-century buildings' valorization. Furthermore, the results will contribute to the knowledge of the current state of conservation of these materials and will allow an understanding of the evolution in the application of national regulations during this period.

Keywords: concrete; award-winning buildings; 20th century; heritage; Lisbon; durability; national regulations

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1. Introduction

Reinforced concrete elements are an essential part of the building structures of the 20th century. In the context of enhancing and preserving built heritage, it is increasingly necessary to know the characteristics of this composite material since little is known about the criteria of the constructive design of a significant proportion of the buildings built in the early 20th century.

However, there has been concern about studying reinforced concrete structures in the international context. These studies often relate construction materials to construction methods, manufacturing processes, performance associated with applying standards, or by approaching their context from the perspective of historical appreciation. Some of them may be exemplified by several works [1–5].

Maintaining concrete structures to extend their service life is a mandatory condition. For the structural integrity of the buildings, durability is a critical factor.

The durability of reinforced concrete structures depends on several factors, such as weathering action, chemical attack, and abrasion, while maintaining its desired design properties. It usually refers to the duration of the life span of trouble-free performance. According to Mather [6], concrete is "durable" if, in its environment, it has provided the desired service life without the high cost of maintenance and repair due to degradation or deterioration.

The evolution of construction processes during the 20th century, associated with the massification of the use of Portland cement, forced the processes' standardization and the creation of national regulations. In 1918 the first Portuguese regulation on reinforced concrete was published [7], which allowed technological and broad harmonization of the use of this composite construction material. Until 1918, public construction was carried out according to the French regulations published in 1906 [7]. To understand the importance of the use of reinforced concrete at the beginning of the 20th century, more specifically between 1903 and 1911, we must mention the publication of the first regulations in various countries, such as Switzerland, Prussia, France, Italy, England, Austria, Russia, Denmark, and the United States [8–21].

In 1935, the so-called reinforced concrete regulation (*Regulamento do Betão Armado—RBA 1935*) [22] revoked the first published regulation. Between the publication of these two documents, which lasted about 17 years, the research and technology applied to increase the knowledge of this composite material have worldwide evolved enormously.

In Portugal, one of the aspects to highlight as an upgrade of regulation is the transition from the use of smooth to ribbed rebars, which was defined by the regulation of reinforced concrete structures published in 1967 (*Regulamento de Estruturas de Betão Armado—REBA 1967*) [23]. The use of plain rebars has implications for the efficiency of crack control and the fixing length. Compared to plain rebars, the ribbed steel ones have greater efficiency in controlling crack openings. After 1967, the Reinforced and Prestressed Concrete Structures regulations were published in 1983 (*Regulamento de Estruturas de Betão Armado e pré-esforçado—REBAP 1983*) [24].

In addition to the reinforced concrete structures regulations, regulations for hydraulic binder's concretes were published in 1971 [25] and 1989 [26], the latter being an updated version of the former. Hydraulic binder concretes are widely used in construction, assuming a relevant role in structures. For that reason, their characteristics and application conditions have a significant impact on the economy and safety of the works.

Table 1 display the concrete characteristics considered in the different regulations published and applied during the 20th century.

Table 1. Evolution of concrete characteristics through regulations applied in Portugal during the 20th century.

Regulations	Main Characteristics
Regulation of 1918	Prescribed dosage in the regulation: 300 kg of cement, 400 L of sand, and 800 L of gravel. There is no concept of resistance class. Minimum compressive strength: 120 kg/cm^2, at 28 days (through cubes).
RBA 1935	The dosage prescribed in the regulation (300 kg of cement, 400 L of sand, and 800 L of gravel). There is no concept of resistance class. Minimum compressive strength value: 180 kg/cm^2, at 28 days (through cubes).
REBA 1967	Resistance classes B180, B225, B300, B350 and B400 (compressive strength in kg/cm^2 = numeric part). Characteristic resistance in kg/cm^2 at 28 days (through cubes).
RBLH 1971 (updated by RBLH 1989)	Two types of concrete: B for resistance requirement and BD1, 2, and 3 for special durability requirement.
REBAP 1983	Resistance classes from B15 to B55, with the resistance increasing by 5 MPa to each class (compressive strength in Mpa = numeric part). Classes defined in international units (MPa). Characteristic strength in MPa (cubic test pieces).

The architectural quality of Lisbon buildings awarded with the Valmor Prize for Architecture [27–29], which is the object of this study, is of great patrimonial interest.

Thus, studying their construction materials is essential to support future conservation and restoration actions. This work does not intend to represent ordinary buildings but to understand and evaluate the advances achieved in each period of construction in Portugal during the 20th century, based on buildings of unquestionable architectural value, which, in general, were built using edge technology of their time. It is crucial to characterize the properties of the employed concretes using a methodology that allows us to provide a set of data regarding their physical and mechanical characteristics. These characteristics should be related to the existing regulations at the construction time and will allow us to infer the quality of the concretes applied.

Different authors have published several studies [30–32] demonstrating the importance of preserving reinforced concrete heritage since the beginning of the 20th century and applying appropriate methodologies to its investigation. A proper assessment of the properties of old concrete is needed to ensure the extended working life and the safe use of old facilities [30]. The study of physical and mechanical characteristics is critical to evaluating the performance of old structures, as demonstrated by Ambroziak et al. [30] in a study on the durability of a 95-year-old concrete built-in bridge. Sena-Cruz et al. [2] studied the physical and chemical characteristics of a reinforced concrete bridge built in 1907. Ambroziak et al. [31] studied the durability and strength of the reinforced concrete properties of a 70-year-old concrete structure in an office building. Sohail et al. [32] investigated the outcomes of concrete degradation in structural concrete elements in the harsh climates of the Arabian Gulf between the 1960s and the 1980s.

This work is part of a more extensive study comprising chemical, mineralogical, and microstructural characterization, whose data will complement the results presented here. The results will allow establishing criteria for maintenance and conservation of this heritage, contributing to its safeguard. The data obtained will also contribute to the knowledge of the evolution of materials in the built heritage of the 20th century, which is attracting more and more interest.

2. Materials and Methods

2.1. Case Studies and Sampling

Twelve buildings were studied (Table 2). The first award-winning building was prized in 1938, and the last one was prized in 2002. These buildings' main architectural and constructive characteristics can be found elsewhere [33–46]. The studied buildings do not present degradation signs that may affect their structural integrity, nor are they continuously monitored.

Concrete sampling was carried out in places that did not compromise the building's safety or aesthetics [47]. Samples were mainly taken from architectural and non-architectural reinforced concrete columns and walls using a diamond core driller equipped with a 75 mm diameter core bit (Figure 1). Due to technical constraints, sometimes core samples were collected at half the diameter, in which case, no mechanical tests were performed.

Table 2. Case studies, sampling zones and samples collected.

Case Study (Award Year)	Name	Image of the Case Study	Construction Year (Completion)	Sampling Zones (Interior/Exterior)	Structural Element	Number of Samples		Type of Coatings/Samples' Distance to the Surface
						Architectural Concrete	Non-Architectural Concrete	
IRF (1938)	*Nossa Senhora do Rosário de Fátima* Church		1938	Belltower (interior)	Columns	n.a.	4	Plasters and painting layers/up to 10 mm
DN (1940)	*Diário de Notícias* Building		1940	Basement. −2 floor (interior)	Columns	n.a.	4	Plasters/26 to 80 mm
LIP (1958)	Laboratories of Pasteur Institute of Lisbon		1957	1st floor. Chemical laboratory and technical area (interior)	Columns	n.a.	6	Painting layers/up to 1 mm
				2nd floor. West façade (exterior)	Beam	n.a.	1	Rendering mortar/7 mm
EUA53 (1970)	*América* Building		1969	Stairs. Between the 3rd and 4th floor (interior)	Wall	n.a.	1	Plasters/25 mm
				Corridor. 2nd-floor technical room (interior)	Column	n.a.	1	Plasters/25 mm

Table 2. *Cont.*

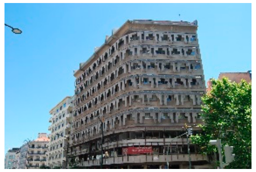
Case Study (Award Year)	Name	Image of the Case Study	Construction Year (Completion)	Sampling Zones (Interior/Exterior)	Structural Element	Number of Samples		Type of Coatings/Samples' Distance to the Surface
						Architectural Concrete	Non-Architectural Concrete	
FRAN (1971)	*Franjinhas* Building		1969	External gallery. East façade (exterior)	Column	1	n.a.	No coatings/0 mm
					Wall	1	n.a.	No coatings/0 mm
				Garage. −2 floor (interior)	Columns	4	n.a.	No coatings/0 mm
					Wall	1	n.a.	No coatings/0 mm
FCG (1975)	Calouste Gulbenkian Foundation Headquarters and Museum		1969	Auditorium ventilation shafts. −2 floor (interior)	Walls	n.a.	4	Plasters and painting layers/up to 10 mm
				Headquarters garage. Technical room. −2 floor (interior)		n.a.	3	Plasters/up to 35 mm
ISCJ (1975)	*Sagrado Coração de Jesus* Church		1970	7th-floor terrace (exterior)	Wall	1	n.a.	No coatings/0 mm
JRP (1987)	*Jacob Rodrigues Pereira* Institute		1987	Swimming pool surrounding area (interior)	Columns	n.a.	7	Painting layers/up to 1 mm

Table 2. *Cont.*

Case Study (Award Year)	Name	Image of the Case Study	Construction Year (Completion)	Sampling Zones (Interior/Exterior)	Structural Element	Number of Samples		Type of Coatings/Samples' Distance to the Surface
						Architectural Concrete	Non-Architectural Concrete	
PCV (1998)	The Knowledge Pavilion		1998	2nd-floor terrace (exterior)	White concrete walls	4	n.a.	No coatings/0 mm
				Ground floor. South façade (exterior)		2	n.a.	No coatings/0 mm
				Garage. −1 floor (interior)	Columns	n.a.	4	Plasters/up to 5 mm
				Technical room. −1 floor (interior)		n.a.	2	Plasters/up to 5 mm
C8 (2000)	C8 Building (Faculty of Sciences of the University of Lisbon)		2000	1st floor. Structure A (interior)	Walls	n.a.	1	Painting layers/up to 1 mm
				1st floor. Structure B (interior)		n.a.	2	Painting layers/up to 1 mm
				1st floor. Structure C (interior)		n.a.	1	Painting layers/up to 1 mm
				1st floor. Structure D (interior)		n.a.	2	Painting layers/up to 1 mm
AS (2001)	*Atrium Saldanha* Building		1997	5th floor. Hub 2 (interior)	White concrete column	2	n.a.	No coatings/0 mm
				Garage. −4 floor (interior)	Column	1	n.a.	No coatings/0 mm
					Walls	5	n.a.	No coatings/0 mm
UNL (2002)	Rectory of the New University of Lisbon		2002	Air treatment unit. 1st-floor (interior)	Walls	n.a.	2	Plasters/up to 30 mm
				Ground floor storage (interior)		1	n.a.	No coatings/0 mm
				Garage. −1 floor (interior)	Earth supporting walls	1	n.a.	No coatings/0 mm
				Garage. −2 floor (interior)		2	n.a.	No coatings/0 mm

Notation: n.a.—not applicable.

Figure 1. Images of concrete sampling campaign: (**a**) IRF (1938); (**b**) FRAN (1971); (**c**) JRP (1987); (**d**) PCV (1998).

2.2. Characterization Methodology

The characterization methodology included observing samples to record the evolution of the dimension of the largest crushed aggregates over time. This evolution is essential to relate it to physical characteristics, such as the compacity, which is also assessed through ultrasonic pulse velocity tests and water absorption by capillary rising, open porosity, and bulk density tests. Carbonation depth was directly measured in core samples so that it can be correlated to the mechanical and physical properties. The mechanical behavior was evaluated through compressive strength and dynamic modulus of elasticity in compression tests to determine their evolution over the analyzed period. Finally, to evaluate the quality of the concrete, the compressive strength results were used to estimate by modelling, through the application of Eurocode 2 [48], the corresponding compressive strengths at 28 days.

Considering the proposed characterization methodology, most of the samples collected are over 150 mm long. As the availability of samples was limited, the core samples were cut in half, and the ends rectified to reach a flat surface and regular dimension. In these cases, capillary water absorption, open porosity and bulk density tests were performed on one of the specimens. Ultrasonic pulse velocity and compressive strength tests were performed in the other specimen, with a length/height equal to the diameter. The dynamic modulus of elasticity in compression was performed on other samples with 150 mm in length, also with the rectified ends.

Figure 2 refer to the main apparatus and testing machines used during the testing campaign. Figure 2a show a tray filled with samples during the water absorption by the capillary rise test. Figure 2b display a weighing apparatus used to estimate the hydrostatic mass during the evaluation of open porosity and bulk density. Figure 2c show a portable ultrasonic pulse velocity tester, and Figure 2d,e exhibit, respectively, the compressive strength and dynamic elastic modulus test machines.

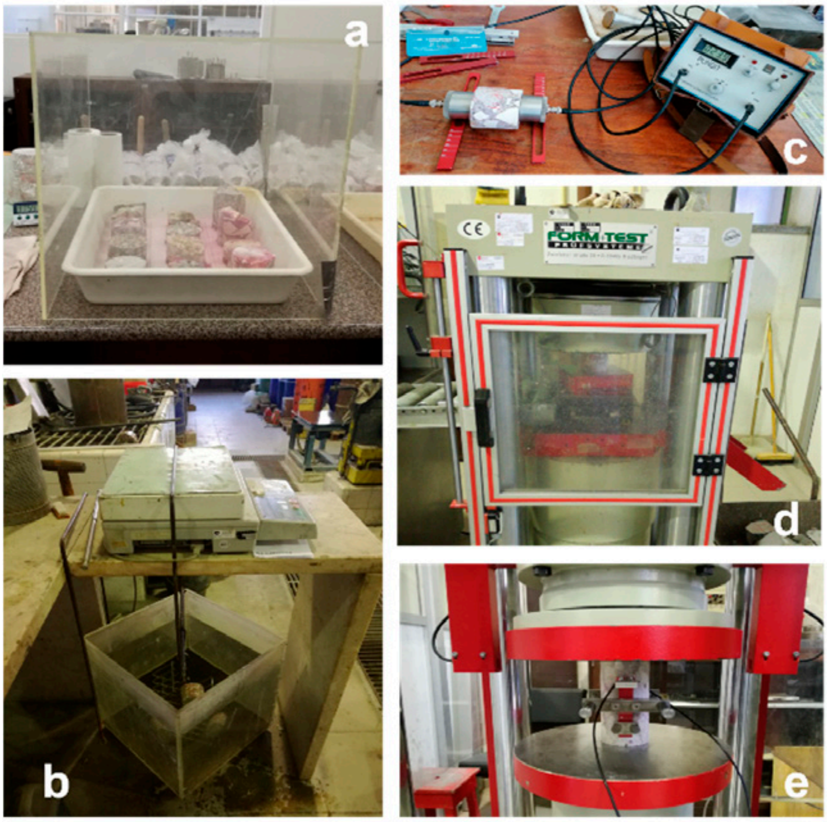


Figure 2. Apparatus for concrete testing: (**a**) capillary water absorption test; (**b**) open porosity and bulk density; (**c**) ultrasonic pulse test; (**d**) compressive strength test; (**e**) dynamic elastic modulus test.

2.2.1. Macroscopic Observation of Cores and Carbonation Depth Assessment

After sampling, the cores were photographed and macroscopically observed to register some characteristics, such as the type of coarse aggregates, presence of cracks, gels, and deposits. The size of the largest coarse aggregates was measured with a digital caliper, and the concrete carbonation depth was measured by applying a phenolphthalein alcoholic solution directly to the core samples [49], whose results have already been published elsewhere [46].

2.2.2. Capillary Water Absorption Test

The water absorption by capillary rise was determined according to LNEC Specification E393 [50]. The test protocol consists of drying a concrete sample, placing it in an oven at a temperature of 40 ± 5 °C for 14 days, and weighing the initial mass (*M0*). Then, the sample is placed inside a tray, filling it carefully with water until the level reaches 5 ± 1 mm above the lower face of the sample, avoiding wetting the other faces.

The tray and the samples were covered with a hood to keep the water level constant during the entire test. The measurements (*Mi*) are made at regular time intervals. To calculate the capillary absorption at a given time, divide the mass increase (*Mi-M0*) by the sample area in contact with the water.

2.2.3. Open Porosity and Bulk Density Test

The open porosity corresponds to the water absorption by immersion under a vacuum. The water absorption test [51] was performed after drying the samples at a temperature of 105 °C until a constant mass was obtained (*Md*). The samples were placed in a receptacle in a vacuum chamber in which the air pressure was brought down to an absolute value of not more than 1 kN/m^2 and held in a vacuum for 24 h. Water was then slowly introduced into the chamber so that the samples were completely immersed, maintaining the 0 for

24 h. The samples were kept immersed for another 24 h at atmospheric pressure and then weighed in water to obtain the hydrostatic mass (*Mh*). Finally, the samples were removed from the water, and their surface was dried rapidly with an absorbent cloth or a natural sponge to remove all surface water to be weighed (*Ms*) to obtain the mass of the saturated samples in a vacuum.

The open porosity (P_0) was then calculated according to the following Equation (1)

$$P_0 = \frac{Ms - Md}{Ms - Mh} \times 100 \tag{1}$$

The bulk density (*Pb*) was calculated according to the following Equation (2).

$$Pb = \frac{Md}{Ms - Mh} \times \rho \tag{2}$$

ρ is the water volumetric mass density at room temperature.

2.2.4. Ultrasonic Pulse Velocity Test

The ultrasonic pulse velocity test was carried out according to EN 12504-4 [52]. Ultrasonic pulse velocity (V) was determined directly using a PUNDIT 6 portable ultrasonic non-destructive tests of CNS electronics, with a measurement range from 0.1 µs to 9999 µs, which has two transducers working in a 54 kHz frequency, placed at the ends of the sample. The velocity of propagation is calculated by the following Equation (3).

$$V = \frac{L}{T} \tag{3}$$

where L is the path length, and T is the time it takes for the ultrasonic pulse to traverse the path length.

The samples were previously rectified by grinding to obtain flat end surfaces. As the grinding was carried out with a water aid, the samples were dried in an oven at 40 °C for 72 h before the test.

2.2.5. Compressive Strength and Dynamic Modulus of Elasticity in Compression

The compressive strength test was performed according to the EN 12390-3 [53] procedure in a FORM+TEST STM 3000 S testing machine featuring a maximum test load of 3000 kN.

The modulus of elasticity in compression was carried out according to E397-1993 [54] in a FORM+TEST Alpha 20–600 testing machine. The test equipment applies and maintains the required load with an accuracy of not less than 1%. The instruments for measuring changes in length (the strain transducers) were placed at equal distances from the ends of the test piece and at least 1/4 of the height from the ends. The measuring instruments enabled the length to be determined with an accuracy of not less than 5×10^{-6}.

A constant load speed within the range of 0.6 ± 0.2 MPa/s was applied. The load was increased continuously, starting from 0.5 MPa until it reached 1/3 of the rupture strain, which was known after the compressive strength test was carried out in other samples of the same building. Six loading cycles were carried out for each test.

2.2.6. Quality Evaluation of the Hardened Concrete

The standard CEN EN 1992: Eurocode 2 [48] was applied to calculate the compressive strength that concrete would have at 28 days of age, considering the concrete class prescribed in the construction design project for each case study [46] and thus evaluating the quality of construction at the time of concrete application.

For this calculation, Equation (4) was used. It was deduced from Equations (5) and (6) of the CEN EN 1992: Eurocode 2, where $f_{ck}(28d)$ is the characteristic value of the compressive strength applied in structures and $f_{cm}(28d)$ is the value obtained by applying

Equation (5) takes into account a standard deviation of 4 MPa, which was considered current in older concrete productions.

$$f_{ck}\ (28d) = f_{cm}(28d) - (1.64 \times 4) \tag{4}$$

$$f_{cm} = f_{cm}(t)\ \beta_{cc}(t)^{-1} \tag{5}$$

with

$$\beta_{cc}(t) = \exp\left\{ s\left[1 - \left(\frac{28}{t} \right)^{1/2} \right] \right\} \tag{6}$$

where f_{cm} is the mean compressive strength at 28 days and $f_{cm}(t)$ is the compressive strength obtained by the test, with t being the buildings' age expressed in days. $\beta_{cc}(t)$ is the coefficient that depends on the age of the concrete t, and s is the coefficient that depends on the type of cement. Since the type of cement used in the production of the concrete is not known, a coefficient s = 0.20 was adopted, according to the CEN EN 1992 standard [41], as older cement presented slower strength increases compared to nowadays.

For both IRF (1938), DN (1940), and LIP (1958) case studies, the presented results of compressive strength at 28 days equals f_{cm} (the mean value). Since the concrete class was prescribed in the construction, the design was defined according to the 1935 regulation [22]. For the definition of the strength limits to be applied, this regulation refers only to minimum values of compressive strength, while in later regulations [23,24], which were applied in the remaining case studies, strength classes are defined using the criterion of the characteristic strength value f_{ck}.

3. Results and Discussion

3.1. Macroscopic Observation of Cores and Carbonation Depth

The macroscopic observation of the concrete cores showed large coarse aggregates composed of white limestone, sometimes fossiliferous, and rarely clayey (Figure 3). The first case study, IRF (1938), exhibited coarse volcanic aggregates, and the second one, DN (1940), also had chert aggregates. Most of these aggregates are compatible with the lithotypes explored to the north of the Lisbon region. No gels, deposits, or cracks were detected in the samples.

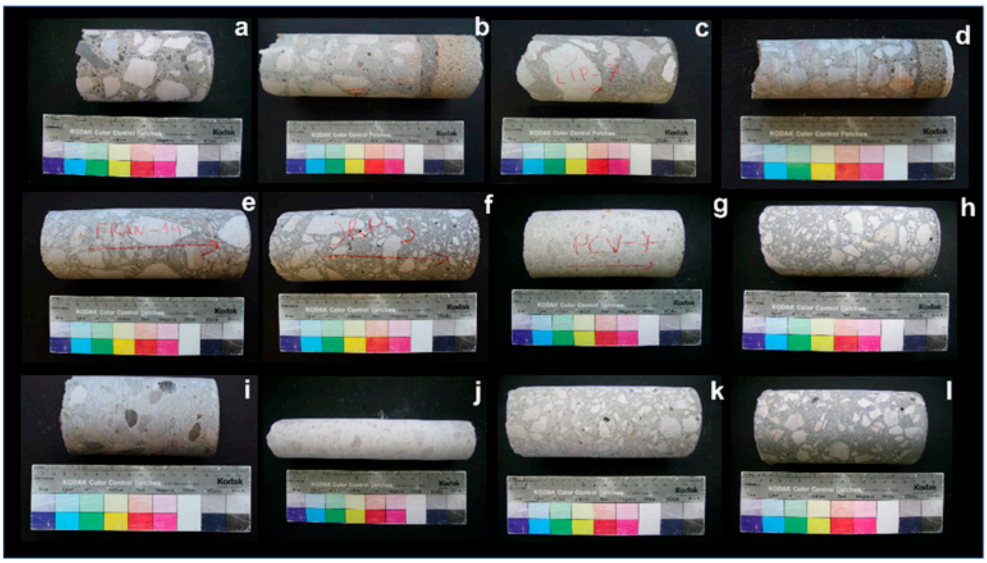


Figure 3. Sample cores from the following case studies: (**a**) IRF (1938); (**b**) DN (1940)—coated with plasters; (**c**) LIP (1958)8; (**d**) EUA53 (1970)—coated with plasters; (**e**) FRAN (1971); (**f**) JRP (1987); (**g**) PCV (1998)—white concrete; (**h**) PCV (1998); (**i**) C8 (2000); (**j**) AS (2001)—white concrete; (**k**) AS (2001); (**l**) UNL (2002).

The average dimension of the largest aggregate (Table 3) showed a reduction during the analyzed period, as displayed in Figure 4. The maximum values were recorded in LIP (1958) concretes. Their reduction started in the late 1960s, as exemplified by the FCG (1975), following the regulations [23,25]. The 1935 regulation [22] limited the maximum size to 40 mm, except for significant elements and massive structures where the coarse aggregates could be larger. The subsequent national regulation to recommend aggregates' dimension criteria was published in 1971 [25]. It mentioned using a maximum dimension of 38.1 mm, should the dimension be lower when the reinforcement would be dense. After these two decrees, further regulation [24] established dimension criteria depending on the reinforcement design.

Table 3. Dimension of the largest aggregate.

Parameters	Gray Concrete												White Concrete	
Case study	IRF (1938)	DN (1940)	LIP (1958)	EUA53 (1970)	FRAN (1971)	ISCJ (1975)	FCG (1975)	JRP (1987)	PCV (1998)	C8 (2000)	AS (2001)	UNL (2002)	PCV (1998)	AS (2001)
Average dimension (mm)	50.0	42.5	60.3	50.0	46.0	45.0	30.6	32.9	30.0	24.7	22.7	22.5	11.7	22.5
S.D. (σ)	8.2	2.9	17.4	14.1	17.2	2.5	3.5	11.0	6.3	4.5	2.6	5.2	2.6	3.5

Notation: S.D.—standard deviation.

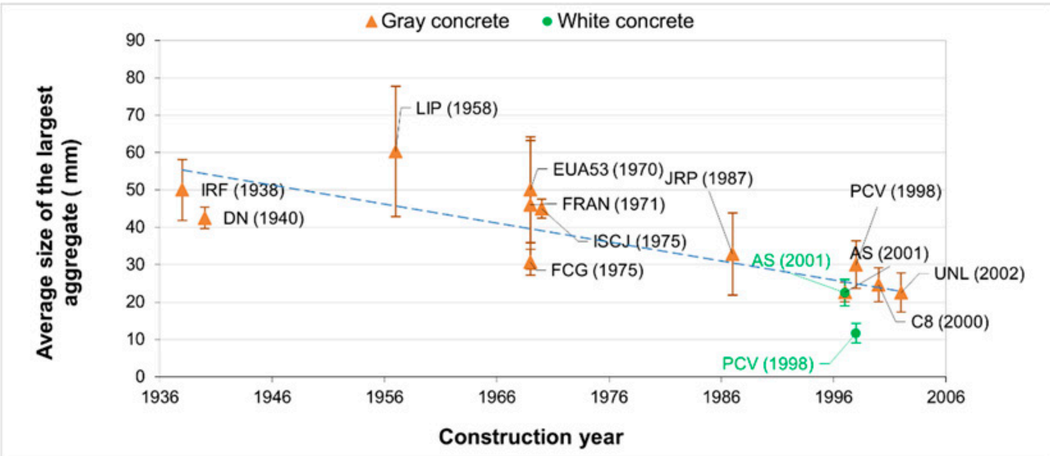


Figure 4. Evolution of the largest aggregate dimension over the period analyzed.

The carbonation depth (Table 4 and Figure 5) of architectural and non-architectural concrete shows a decreasing trend over time, which is expected with concrete ageing.

Table 4. Carbonation depth in the architectural and non-architectural concrete samples.

Parameters	Non-Architectural Concrete									Architectural Concrete				
Case study	IRF (1938)	DN (1940)	LIP (1958)	EUA53 (1970)	FCG (1975)	JRP (1987)	PCV (1998)	C8 (2000)	UNL (2002)	FRAN (1971)	ISCJ (1975)	PCV (1998)	AS (2001)	UNL (2002)
Carbonation depth (mm)	26.9	10.5	15.3	1.2	1.5	12.2	15.8	6.1	16.8	11.4	10.7	2.5	2.6	15.2
S.D. (σ)	10.4	10.2	9.4	1.2	0.5	8.3	9.9	4.2	8.7	6.6	5.1	2.1	1.8	5.7

Notation: S.D.—standard deviation.

The size reduction of crushed coarse aggregate over time is a consequence of the standardization and the optimization of the mixing control. The coarse aggregate plays a vital role in determining the mechanical behavior of concrete as it occupies about 70% of the concrete volume [55,56]. The mechanical properties of concrete from older case studies may be conditioned by the volume occupied by these aggregates and, consequently, by the interfacial zone (ITZ) area, which might evolve to the formation and propagation of microcracks. Similarly, the carbonation depth, which also tends to decrease towards the end of the analyzed period, may be favored by the development of microcracking in the dependence on the ITZ. Concretes from the oldest case study, IRF (1938), have a higher carbonation depth than any other, which is understandable, presumably due to the more prolonged exposure to CO_2. The carbonation depths of other concretes are quite variable due to the protection provided by the coatings. The coatings, whose typology, thickness (Table 1), and the related physical and chemical properties provided different types of protection, conditioned the penetration of CO_2 and the moisture transport capability.

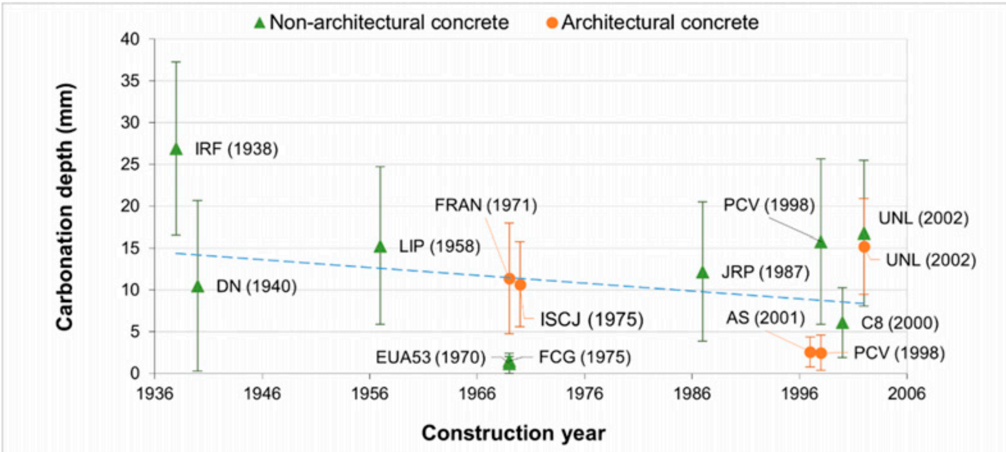


Figure 5. Evolution of the carbonation depth over the analyzed period.

3.2. Physical Characterization

Table 5 show the results of the physical characterization obtained for the open porosity, the ultrasonic pulse velocity, and water absorption by capillary tests.

Table 5. Average results of physical properties of concrete samples.

Parameters	Gray Concrete											White Concrete	
Case study	IRF (1938)	DN (1940)	LIP (1958)	EUA53 (1970)	FRAN (1971)	FCG (1975)	ISCJ (1975)	JRP (1987)	PCV (1998)	C8 (2000)	AS (2001)	UNL (2002)	PCV (1998)	AS (2001)
P_0 (%)	13.78	13.38	10.82	11.60	13.02	13.64	n.a.	20.02	14.86	14.21	15.54	15.75	13.77	13.30
S.D. (σ)	1.51	2.14	1.61	n.a.	2.04	0.67	n.a.	2.60	1.30	0.53	0.41	0.78	n.a.	n.a.
Pb (kg/m^3)	2302.27	2286.01	2379.81	2363.18	2306.50	2279.22	n.a.	2110.31	2258.08	2267.04	2220.48	2229.24	2262.81	2300.23
S.D. (σ)	49.72	66.74	42.37	n.a.	61.87	22.99	n.a.	74.70	35.55	13.31	16.50	23.65	n.a.	n.a.
V (m/s)	4103.20	4093.41	4652.52	4512.94	4816.02	4853.85	n.a.	3792.04	4555.49	4415.99	4512.20	4862.32	4684.49	4406.98
S.D. (σ)	805.37	270.77	169.43	n.a	297.55	180.46	n.a.	456.72	124.04	207.92	102.79	191.88	n.a.	n.a.
W.A. at 15 min (Kg/m^2)	1.18	1.27	1.16	0.41	0.61	0.46	n.a.	1.45	0.51	0.68	0.58	0.40	0.55	0.45
S.D. (σ)	0.01	0.93	0.47	n.a.	0.18	0.14	n.a.	0.49	0.10	0.16	0.15	0.08	n.a.	n.a.
W.A. at 60 min (Kg/m^2)	1.88	1.97	1.80	0.74	0.93	0.76	n.a.	2.52	0.84	1.15	0.97	0.61	0.90	0.91
S.D. (σ)	0.05	1.36	0.55	n.a.	0.24	0.21	n.a.	0.84	0.18	0.29	0.18	0.12	n.a.	n.a.
W.A. at 1440 min = 24h (Kg/m^2)	5.41	4.45	3.63	3.15	2.64	2.09	n.a.	9.13	2.48	4.19	3.47	1.61	2.71	3.33
S.D. (σ)	1.29	1.40	0.52	n.a.	0.67	0.49	n.a.	2.35	0.60	1.11	0.58	0.42	n.a.	n.a.

Notation: P_0—open porosity; S.D.—standard deviation; *Pb*—bulk density; *V*—ultrasonic pulse velocity; W.A.—water absorption by capillary rising; n.a.—not available.

The combined results show that the average of the open porosity values varies between 10.82% and 20.02%. The slight increasing trend over the period under analysis is shown in Figure 6.

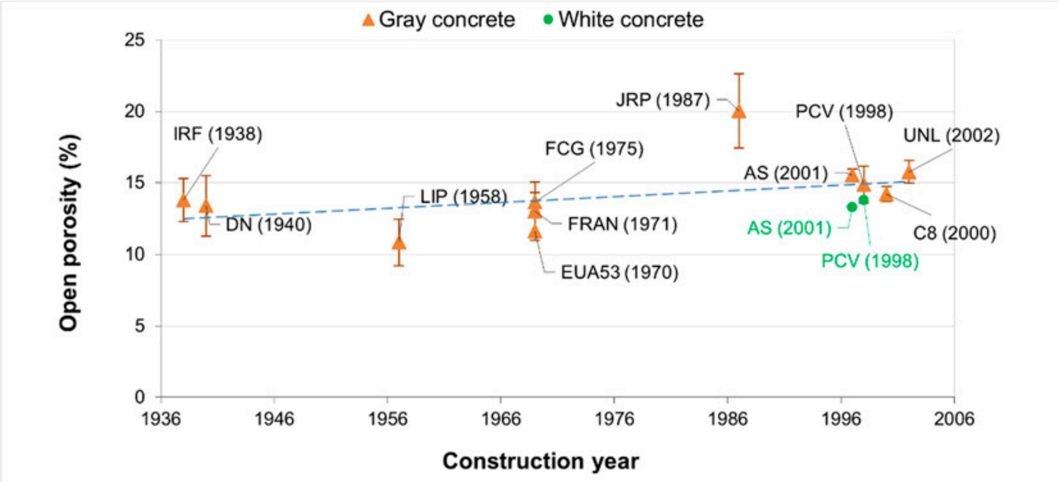


Figure 6. Evolution of the open porosity over the analyzed period.

Regarding the average ultrasonic pulse velocity, the results point to the quality of concrete material in a range between good and excellent, considering the classification of Whitehurst, 1951 [57]. The most significant variations in the results were observed in the case studies IRF (1938) and JRP (1987), as shown in Figure 7. In the first case, coarser aggregates may explain such variations. In contrast, in the second case, the higher open porosity influences the obtained result, resulting in the lower value of ultrasonic pulse velocity and, therefore, the compacity.

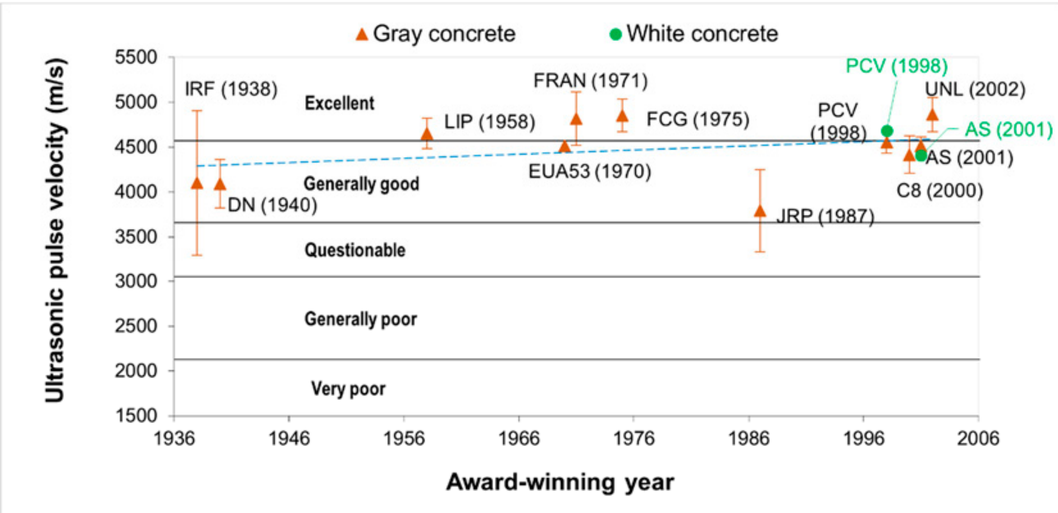


Figure 7. Evolution of the ultrasonic pulse velocity over the analyzed period. Quality classification according to [57].

The water absorption results by capillarity also show a reduction trend along the period under analysis (Figures 8 and 9). The concrete of the case study JRP (1987) shows the highest values of capillary absorption, corroborating the results of the high open porosity and the lowest values of bulk density and ultrasonic pulse velocity. Regarding white concretes, the values of capillary absorption are similar between samples of the two case studies analyzed (Figure 9): PCV (1998) and AS (2001).

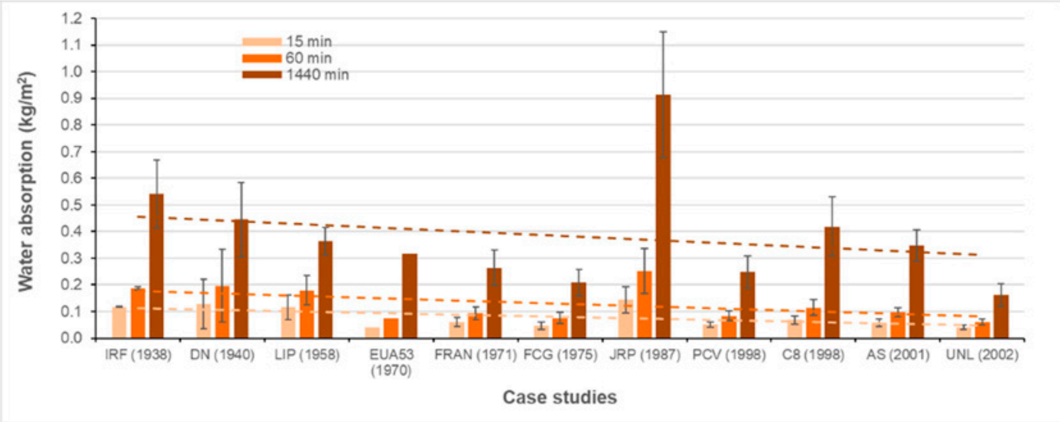


Figure 8. Capillary water absorption values at 15, 60, and 1440 min for gray concrete samples.

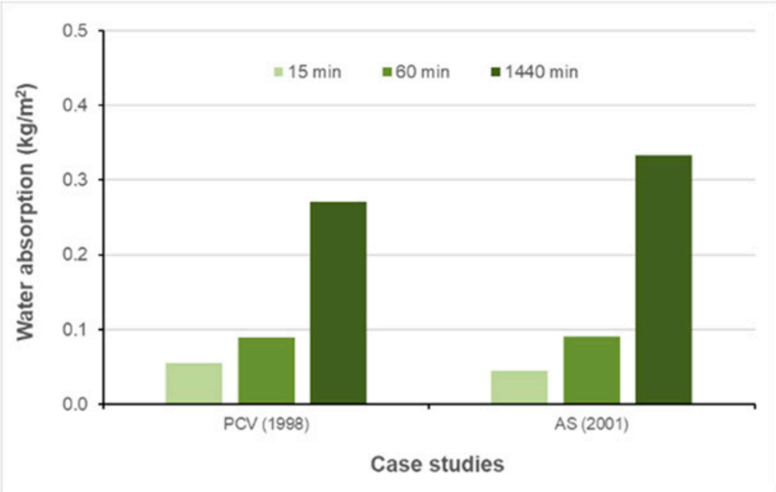


Figure 9. Capillary water absorption values at 15, 60, and 1440 min for white concrete samples.

The results of physical characterization show a tendency to reduce water absorption over time and, consequently, an increase in the compacity of the concrete, as proven by the results of the ultrasonic pulse velocity. The increase in ultrasonic pulse velocity is indicative of the reduction of the total porosity of the tested medium. However, the results obtained for the open porosity show an opposite trend, i.e., an increase over time, albeit slight. The reduction of the maximum size of the crushed aggregates and the greater homogeneity of the concrete favored the increase of compacity. On the other hand, there are exceptions, and the lowest values of open porosity were recorded in older concretes, which have coarse aggregates of larger dimensions. Variations in porosity and water absorption may be linked to cement type and dosage, as well to the volume occupied by the aggregates, namely the coarse aggregates, as it occurred in buildings constructed until the 1960s. The size-effect and the volume occupied by large aggregates in the tested specimens may be at the origin of this trend since the inherent porosity of these aggregates may significantly influence the results. It should be noted that limestones from the north region of Lisbon, one of the most extensive exploration centres in Portugal, present values of open porosity not exceeding 1.2% [58]. Nevertheless, a reduction in cement fineness is assumed over time [59,60]. It is observed that concretes with larger aggregates, more precisely those from buildings awarded up to 1998, usually show lower open porosities than concretes with smaller coarse aggregates. Hence, it can be assumed that the open porosity is influenced by the volume occupied by the coarse aggregates and their inherent low porosity.

From the point of view of the material's durability, the reduction in water absorption is a favorable outcome since it increases the resistance to sulphate attack [61] and may avoid the water ingress into concrete.

White cement should have identical behavior to its gray counterparts of the same type and strength class. As for physical characteristics, there are two differences directly related to each other: fineness and the beginning of the setting. White cement is generally thinner and has a greater specific surface. With greater cement fineness comes greater mechanical resistance, particularly at younger ages. On the other hand, as the cement is made of smaller particles, the amount of water required to achieve certain workability is higher, leading to an increase in porosity [42]. However, there was no open porosity increase compared to gray cement concrete for the same buildings.

3.3. Mechanical Characterization

The mechanical characterization results (Table 6) show a trend toward an increase in compressive strength and dynamic modulus of elasticity in compression throughout the period under analysis, as displayed in Figures 10 and 11. This trend is shown in any concrete, regardless of the structural element considered.

Table 6. Average results of mechanical tests of reinforced concrete samples.

Parameters	**Gray Concrete**													**White Concrete**	
	Superstructure												**S.W.**	**Superstructure**	
Case study	IRF (1938)	DN (1940)	LIP (1958)	EUA53 (1970)	FRAN (1971)	FCG (1975)	ISCJ (1975)	JRP (1987)	PCV (1998)	C8 (2000)	AS (2001)	UNL (2002)	UNL (2002)	PCV (1998)	AS (2001)
f_c (MPa)	28.30	32.10	35.80	n.a	60.43	69.58	n.a	27.17	57.13	60.20	61.90	67.47	76.10	65.00	n.a
S.D. (σ)	4.81	n.a	7.39	n.a	9.46	14.90	n.a	13.32	7.03	14.45	2.26	4.42	n.a	n.a	n.a
E_c (GPa)	18.50	17.30	28.60	n.a	33.80	37.20	n.a	17.55	34.37	28.10	31.63	37.50	n.a	35.50	n.a
S.D. (σ)	n.a	n.a	0.71	n.a	1.70	1.57	n.a	9.40	1.96	n.a.	0.81	0.00	n.a	n.a	n.a

Notation: S.W.—supporting walls; f_c—compressive strength; S.D.—standard deviation; E_c—dynamic modulus of elasticity; n.a.—not available.

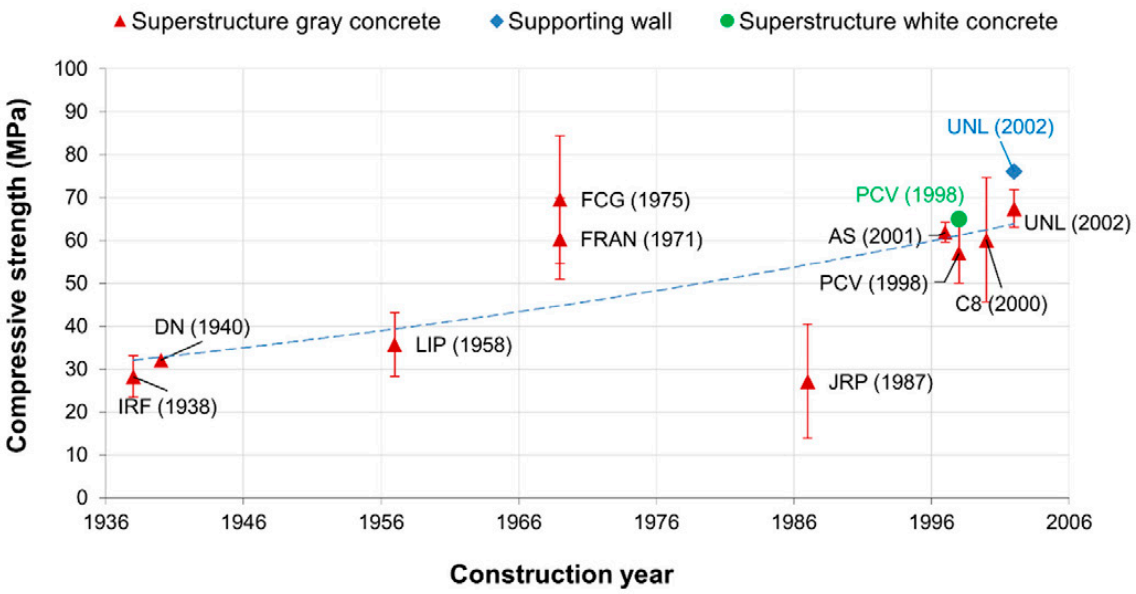


Figure 10. Evolution of the compressive strength over the analyzed period.

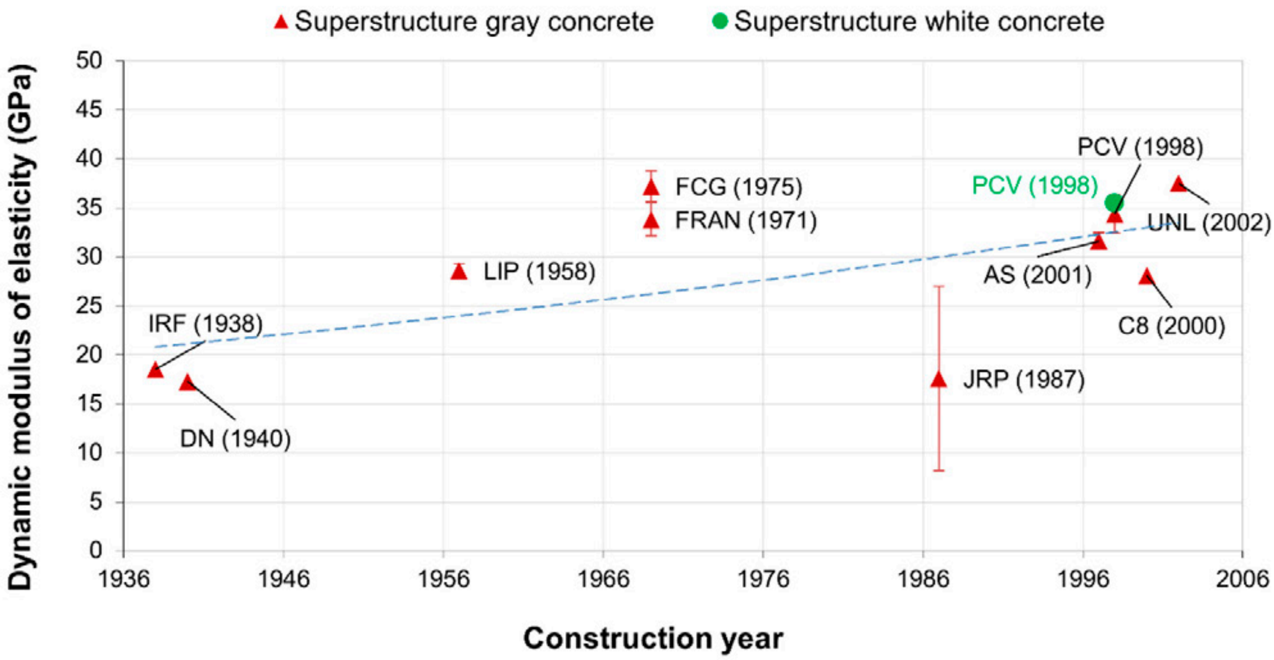


Figure 11. Evolution of the dynamic modulus of elasticity in compression over the analyzed period.

The maximum values of the mechanical characteristics were obtained in the UNL (2002) case study, followed by FCG (1975), respectively, 76.10 MPa and 69.58 MPa—compressive strength values. Dynamic modulus of elasticity values for both case studies are, respectively, 37.50 GPa and 37.2. The case study JRP (1987), on the contrary, presents the lowest values of these characteristics, registering 27.17 MPa and 17.55 GPa, respectively, for compressive strength and the dynamic modulus of elasticity.

It is reported that the compressive strength of concrete increases with the increase of the coarse aggregate size [56]. This relationship was not verified in this study since one or more types of concrete with different characteristics and strength classes were employed in each case study. However, the increasing trend of the compressive strength throughout the period under study accompanies the increase in compacity and the decrease in water absorption. All these properties are pore size structure-dependent, whereas an increase in the fineness of the cement or a decrease in the water to cement ratio (w/c) are expected to occur throughout the 20th century [1]. An evolutive correlation between compressive strength and open porosity shows no clear relationship (Figure 12).

The most relevant source of porosity refers to w/c. When this ratio becomes higher, the porosity of the cement paste in the concrete also upsurges, and the compressive strength reduces as the porosity increases. It is not possible to state a cause–effect relationship between porosity and compressive strength of the concretes up to the 1960s case studies. The concretes of the award-buildings until the 1960s have the larger crushed aggregates. The porosity should be influenced by the coarse aggregate's porosity, resulting in a decrease in the open porosity of the concrete. On the contrary, this relationship is observed in the concrete of the building JRP (1987).

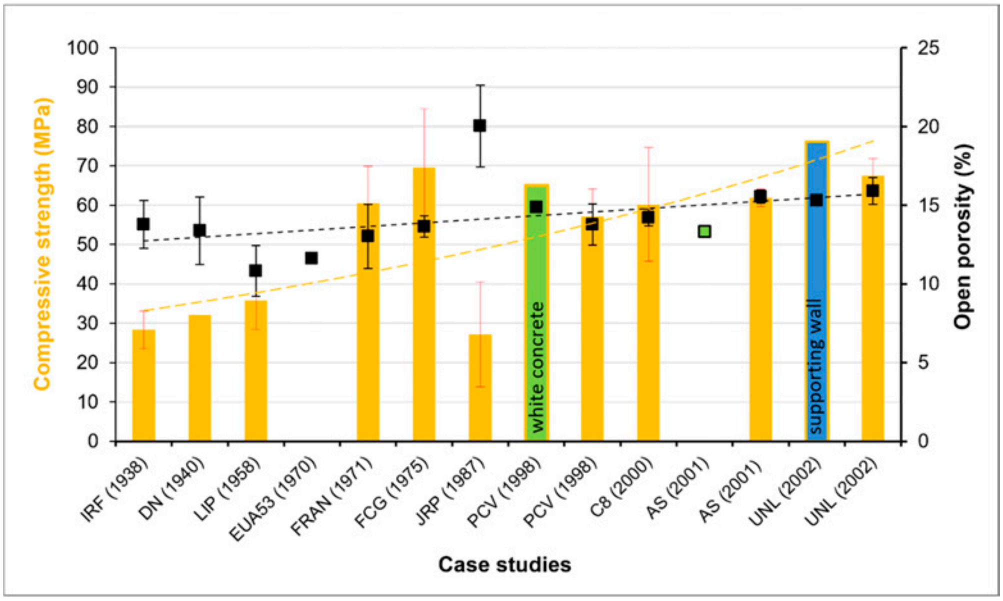


Figure 12. Correlation between compressive strength (bars) and open porosity (black squares) of superstructure white and gray concrete, and concrete from UNL (2002) supporting wall. Standard deviation of compressive strength is represented at the top of the bars by the red lines.

3.4. Quality Evaluation of the Hardened Concrete

The compressive strength was estimated for 28 days of curing, as described in Section 2.2.6, to assess the applied concrete materials' quality at the time of construction and their initial performance.

The results obtained indicate that most of the case studies would have a compressive strength higher than the prescribed at the construction time, as shown in Table 7 and Figure 13. Only two cases exhibited an estimated compressive strength lower than the prescribed: AS (2001) and JRP (1987). A difference of about 3 MPa was registered in the first case study, and the second revealed a difference of 9.5 MPa.

Table 7. Average results of mechanical properties of reinforced concrete samples.

Parameters	**Gray Concrete**												**White Concrete**
	Superstructure											**S.W.**	**Superstructure**
Case study	IRF (1938)	DN (1940)	LIP (1958)	FRAN (1971)	FCG (1975)	ISCJ (1975)	JRP (1987)	PCV (1998)	C8 (2000)	AS (2001)	UNL (2002)	UNL (2002)	PCV (1998)
t (days) *	30,295	29,565	23,360	18,980	18,980	18,615	12,410	8395	7665	8760	6935	6935	8395
Prescribed concrete class	(a)	(a)	(a)	B300	B300	B300	B225	n.a.	B30	B40	B30	B25	B35
β_{cc}	1.214	1.214	1.213	1.212	1.212	1.212	1.210	1.207	1.207	1.208	1.206	1.206	1.207
Prescribed compressive strength (MPa)	17.65	17.65	17.65	29.42	29.42	29.42	22.06	n.a.	30.00	40.00	30.00	25.00	35.00
f_{cm} (t) ** (MPa)	28.30	32.10	35.80	60.43	69.58	n.a.	27.17	57.13	60.20	61.90	67.47	76.10	65.00
f_{cm} (28d) (MPa)	19.81	22.48	25.09	42.36	48.81	n.a.	9.11	(b)	42.40	43.57	53.64	47.58	45.76
f_{ck} (28d) (MPa)	(c)	(c)	(c)	35.80	42.25	n.a.	12.55	(b)	35.84	37.01	47.08	41.02	39.20

Notation: S.W.—supporting walls; * building's age by the end of the year 2021 (considering the completion year of construction); n.a.—not available; ** tested compressive strength = f_c values in Table 6; (a) according to 1935 regulation [22]; (b) no result; (c) The regulation does not mention the characteristic value of the compressive strength, only the minimum value, which implies considering f_{cm} *(28d)* instead of f_{ck} *(28d)*.

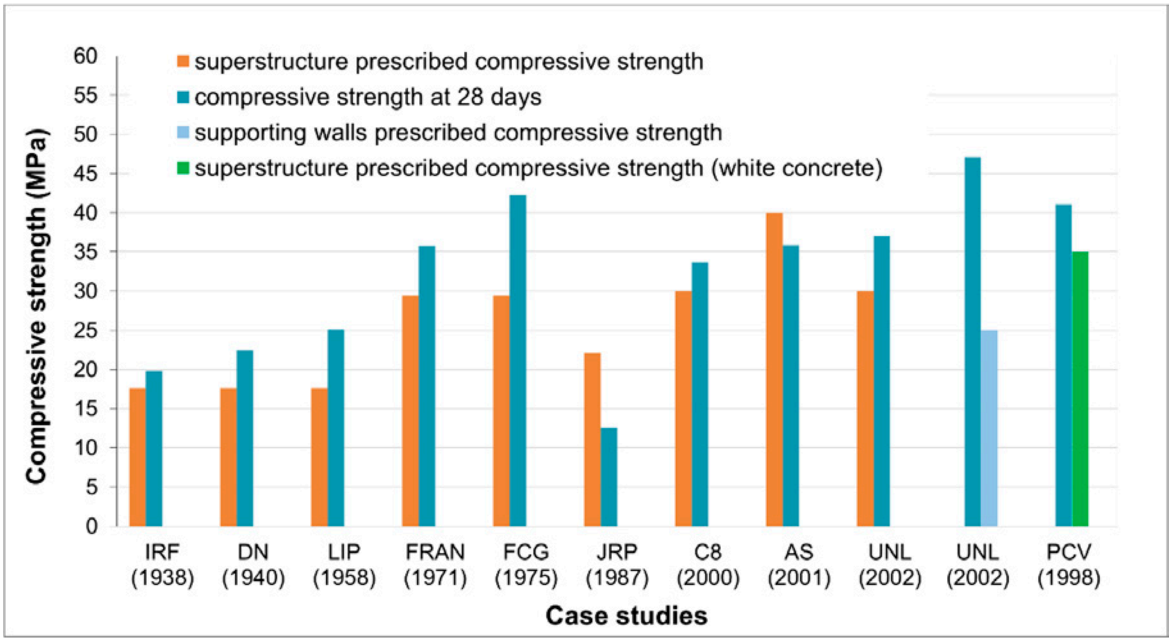


Figure 13. Prescribed compressive strength vs. calculated compressive strength at 28 days.

The concrete of the supporting walls of UNL (2002) presented the best performance, with a difference of 16.2 MPa between the prescribed and the calculated strength, followed by FCG (1975) with a difference of 12.8 MPa.

The evaluation of the concrete quality by estimating the compressive strength at 28 days of curing showed that the project design was followed up successfully. It demonstrates the great care taken during the construction process. It also highlights the actual condition of the structures, which enhances their durability. Although the AS (2001) case study shows a slight difference between the prescribed concrete compressive strength and the estimated one for 28 days of curing (<5 MPa), this difference is not as striking as in the JRP (1987) case study. All the physical and mechanical results obtained for JRP (1987) reveal a worse condition, as its performance is doubly different, which implies a questionable quality of the materials applied, corroborating the ultrasonic pulse velocity results whose dispersion of results places it in the range between the generally good to questionable quality class (Figure 7).

4. Conclusions

The present study made it possible to assess concrete's main physical and mechanical characteristics from a set of 20th-century award-winning architecture buildings in Lisbon. This study is a pioneer one on buildings that have an awarded architectural quality. The systematic studies on this kind of construction materials in Portugal are still scarce.

The results obtained point to an evolution in the characteristics over the period under analysis, which embodies the application of the national regulations. The physical and mechanical properties of the analyzed concrete materials reproduce an evolution towards the safety and durability requirements imposed by the national regulations on account of the advancement in the knowledge of structural performance and the scientific knowledge acquired throughout the 20th century.

The evolution of the physical and mechanical characteristics studied can be listed as follows:

1. The crushed coarse aggregate, mainly composed of limestone, had its maximum size reduced, having decreased from the late 1960s onwards, as exemplified by the case study FCG (1975), as set out in current Portuguese regulation by the time of construction.

2. The carbonation depth shows a decreasing trend, which is expected with concrete ageing. Although it is quite variable as the presence of coatings may play an important role.
3. The open porosity and bulk density values did not show very significant variations. A slight tendency towards a reduction in bulk density and increase in porosity may be related to the variation in the maximum size of the largest aggregate, which varies in the same direction as compacity.
4. Water absorption by capillary rising for all types of concrete studied (white and gray) does not show a consistent trend in the same direction as the open porosity.
5. Open porosity slightly increases towards the end of the analyzed period, implying that this is not exclusively due to the characteristics of the binder but to the whole composite material itself.
6. The mechanical characteristics, except for the building awarded in 1987, show a clear trend towards an increase in the values of the compressive strength and the dynamic modulus of elasticity.
7. Except for the building awarded in 1987, the estimation of the compressive strength at 28 days of curing showed that the project design had been accomplished.

The results allow us to conclude that, in general, the materials show a good durability condition, as far as the physical and mechanical characteristics point out to a good performance, not indicating degradation, considering the age of the buildings and that they are still in use. However, the 1987 award-winning building demonstrated that its overall performance could compromise durability, requiring monitoring actions to prevent degradation.

This study, being part of a more significant characterization underway, contributes to the necessary in-depth knowledge of the physical and mechanical characteristics to apply in conservation and restoration actions over the built heritage in the 20th century.

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References

1. Wilkie, S.; Dyer, T. Design and durability of early 20th century concrete bridges in Scotland: A review of historic test data. *Int. J. Archit. Herit.* **2021**, 1–21. [CrossRef]
2. Sena-Cruz, J.; Ferreira, R.M.; Ramos, L.F.; Fernandes, F.; Miranda, T.; Castro, F. Luiz Bandeira Bridge: Assessment of a historical reinforced concrete bridge. *Int. J. Archit. Herit.* **2013**, *7*, 628–652. [CrossRef]
3. Vidovszky, I.; Pintér, F. An investigation of the application and material characteristics of early 20th-century Portland cement-based structures from the historical Campus of the Budapest University of Technology and Economics. *Int. J. Archit. Herit.* **2020**, *14*, 358–375. [CrossRef]
4. Marcos, I.; San-José, J.T.; Garmendia, L.; Santamaría, A.; Manso, J.M. Central lessons from the historical analysis of 24 reinforced-concrete structures in northern Spain. *J. Cult. Herit.* **2016**, *20*, 649–659. [CrossRef]

5. Carmona-Quiroga, P.; Pachón-Montaño, A.; Queipo-de-Llano, J.; Martín-Caro, J.; López, D.; Paniagua, I.; Martínez, I.; Rubiano, F.; García-Lodeiro, I.; Fernández-Ordóñez, L.; et al. Characterisation and diagnosis of heritage concrete: Case studies at the Eduardo Torroja Institute, Madrid, Spain. *Mater. Constr.* **2021**, *71*, e262. [CrossRef]
6. Mather, B. Concrete durability. *Cem. Concr. Compos.* **2004**, *26*, 3–4. [CrossRef]
7. Ministério do Comércio. *Decree n° 4036. Regulation for the Use of Reinforced Concrete*; Diário do Govêrno: Lisbon, Portugal, 1918. (In Portuguese)
8. Kurrer, K.-E. *The History of the Theory of Structures: From Arch Analysis to Computational Mechanics*; Ernst & Sohn: Berlin, Germany, 2008.
9. Mesnager, A. *Reinforced Concrete Course*; Dunod: Paris, France, 1921. (In French)
10. Ministerium der Öffentlichen Arbeiten. *Provisions for the Execution of Reinforced Concrete Structures in Tall Buildings*; Zentralblatt der Bauverwaltung: Berlin, Germany, 1904. (In German)
11. Ministerium der Öffentlichen Arbeiten. Prussia. In *Provisions for the Execution of Reinforced Concrete Structures in Tall Buildings*; Zentralblatt der Bauverwaltung: Berlin, Germany, 1907. (In German)
12. Martin, N. *The Properties and Design of Reinforced Concrete*; France Commission du Ciment Armé, Ed.; D. Van Nostrand Company: New York, NY, USA, 1912.
13. Gazzetta Ufficiale del Regno de Italia. *Standard Test Methods for Hydraulic Binders, Standard Requirements for the Execution of Reinforced Concrete Works and Technical Conditions with which Supplies of Hydraulic Binders must Comply*; Gazzetta Ufficiale del Regno de Italia: Rome, Italy, 1907. (In Italian)
14. Chrimes, M.M. The development of concrete bridges in the British Isles prior to 1940. *Proc. Inst. Civ. Eng. Struct. Build.* **1996**, *116*, 404–431. [CrossRef]
15. Witten, A. The Concrete Institute 1908–1923, precursor of the Institution of Structural Engineers. *Proc. Inst. Civ. Eng. Struct. Build.* **1996**, *116*, 470–480. [CrossRef]
16. Nielsen, M.P.; Hoang, L.C. *Limit Analysis and Concrete Plasticity*, 3rd ed.; CRC Press: Boca Raton, FL, USA, 2011. [CrossRef]
17. Souponitski, S.; Sniatkov, S.; Grigoriev, S. Early reinforced concrete constructions in Russia: Specific faults and causes of failure. *Eng. Fail. Anal.* **2001**, *8*, 201–212. [CrossRef]
18. Mändel, M.; Orro, O. The marvellous reinforced concrete shells of Tallinn sea-plane hangars in the context of early concrete architecture in Estonia. *Constr. Hist. Soc.* **2012**, *27*, 65–85.
19. Emperger, F. *Manual for Reinforced Concrete Construction*; Wilhelm Ernst & Sohn: Berlin, Germany, 1909. (In German)
20. NACU. *Requirements for Reinforced Concrete or Concrete-Steel Constructed Buildings*; National Association of Cement Users: Philadelphia, PA, USA, 1908.
21. NACU. *Standard Building Regulations for the Use of Reinforced Concrete*; National Association of Cement Users: Philadelphia, PA, USA, 1910.
22. Ministério das Obras Públicas e Comunicações. *Decree n° 25948. Reinforced Concrete Regulation*; Diário do Govêrno: Lisbon, Portugal, 1935. (In Portuguese)
23. Ministério das Obras Públicas. *Decree n° 47723. Regulation of Reinforced Concrete Structures*; Imprensa Nacional de Lisboa: Lisbon, Portugal, 1967. (In Portuguese)
24. Ministério da Habitação, Obras Públicas e Transportes. *Decree n° 39-C/83. Regulation of Reinforced and Prestressed Concrete Structures*; Imprensa Nacional de Lisboa: Lisbon, Portugal, 1983. (In Portuguese)
25. Ministério das Obras Públicas. *Decree n° 404/71. Hydraulic Binder Concrete Regulation*; Imprensa Nacional de Lisboa: Lisbon, Portugal, 1971. (In Portuguese)
26. Ministério das Obras Públicas, Transportes e Telecomunicações. *Decree n° 445/89. Hydraulic Binder Concrete Regulation*; Imprensa Nacional de Lisboa: Lisbon, Portugal, 1989. (In Portuguese)
27. CML. *Valmor Prize*; Silva, A.P., Ed.; Câmara Municipal de Lisboa: Lisbon, Portugal, 2004. (In Portuguese)
28. Bairrada, E.M. *Valmor Prize 1902–1952*; Biblioteca de Lisboa: Lisbon, Portugal, 1988. (In Portuguese)
29. Pedreirinho, J.M. *A Critical History of the Valmor Prize*, 1st ed.; Argumentum: Lisbon, Portugal, 2018.
30. Ambroziak, A.; Haustein, E.; Niedostatkiewicz, M. Chemical, Physical, and mechanical properties of a 95-year-old concrete built-in arch bridge. *Materials* **2021**, *14*, 20. [CrossRef] [PubMed]
31. Ambroziak, A.; Haustein, E.; Kondrat, J. Chemical and Mechanical Properties of 70-Year-Old Concrete. *J. Mater. Civ. Eng.* **2019**, *31*, 04019159. [CrossRef]
32. Sohail, M.G.; Kahraman, R.; Ozerkan, N.G.; Alnuaimi, N.A.; Gencturk, B.; Dawood, M.; Belarbi, A. Reinforced Concrete Degradation in the Harsh Climates of the Arabian Gulf: Field Study on 30-to-50-Year-Old Structures. *J. Perform. Constr. Facil.* **2018**, *32*, 04018059. [CrossRef]
33. AML. *Construction Work 47105*; Process 650/SEC/PG/1934; Municipal Archive: Lisbon, Portugal, 1934. (In Portuguese)
34. AML. *Construction Work 50576*; Process 10460/DAG/PG/1936; Municipal Archive: Lisbon, Portugal, 1936. (In Portuguese)
35. AML. *Construction Work 51405*; Process 15050/DAG/PG/1937; Municipal Archive: Lisbon, Portugal, 1937. (In Portuguese)
36. AML. *Construction Work 27545*; Process 13668/DAG/PG/1955; Municipal Archive: Lisbon, Portugal, 1955. (In Portuguese)
37. AML. *Construction Work 52761*; Process 57854/DAG/PG/1963; Municipal Archive: Lisbon, Portugal, 1963. (In Portuguese)
38. AML. *Construction Work 54898*; Process 39190/1965; Municipal Archive: Lisbon, Portugal, 1965. (In Portuguese)
39. AML. *Construction Work 52469*; Process 1868/1966; Municipal Archive: Lisbon, Portugal, 1966. (In Portuguese)

40. AML. *Construction Work 52743*; Process 1268/DAG/PG/1966; Municipal Archive: Lisbon, Portugal, 1966. (In Portuguese)
41. Proença, J.; Neves, O. *Casa Pia de Lisboa. Structure Project*; Jacob Rodrigues Pereira Institute: Lisbon, Portugal, 1982. (In Portuguese)
42. Fonseca, A.A.; Nunes, A. White concrete. Manufacture, characteristics and structural use. In Proceedings of the 2nd Conference on Concrete Structures, FEUP, Porto, Portugal, 24 April 1998. (In Portuguese).
43. Mendonça, J.; Villar, L. *C8 Building—Chemistry and Physics Department of the Faculty of Sciences of the University of Lisbon. Foundations and Structures Design*; Betar—Estudos e Projectos de Estabilidade, Lda.: Lisbon, Portugal, 1995. (In Portuguese)
44. AML. *Construction Work 62971*; Process 3256/OB/1992; Municipal Archive: Lisbon, Portugal, 1992. (In Portuguese)
45. Mendonça, J.; Villar, M.; Santos, S. *Rectory Building of the New University of Lisbon. Detailed Design of Foundations and Structures*; Betar—Estudos e Projectos de Estabilidade, Lda.: Lisboa, Portugal, 1998. (In Portuguese)
46. Almeida, L.; Silva, A.S.; Veiga, M.d.R.; Mirão, J.; Vieira, M. 20th-Century Award-Winning Buildings in Lisbon (Portugal). Study of Plasters, Rendering, and Concrete Materials Aiming Their Sustainable Preservation. *Buildings* **2021**, *11*, 359. [CrossRef]
47. *CEN-EN 16085:2012*; Conservation of Cultural Property. Methodology for Sampling from Materials of Cultural Property. General Rules. European Standard: Brussels, Belgium, 2012.
48. *CEN-EN 1992-1-2*; Eurocode 2: Design of Concrete Structures. European Standard: Brussels, Belgium, 2012.
49. *CEN-EN 14630*; Products and Systems for the Protection and Repair of Concrete Structures. Test Methods. Determination of Carbonation Depth in Hardened Concrete by the Phenolphthalein Method. European Standard: Brussels, Belgium, 2006.
50. *E393*; Concrete—Determination of Capillary Water Absorption. LNEC: Lisboa, Portugal, 1993. (In Portuguese)
51. *E395*; Concrete—Determination of Water Absorption by Immersion. Vacuum Test. LNEC: Lisboa, Portugal, 1993. (In Portuguese)
52. *CEN-EN 12504-4*; Testing Concrete in Structures—Part 4: Determination of Ultrasonic Pulse Velocity. European Standard: Brussels, Belgium, 2007.
53. *CEN-EN 12390-3*; Tests for Hardened Concrete; Part 3: Compression Strength of Test Specimens. European Standard: Brussels, Belgium, 2011.
54. *E397*; Concrete—Determination of Modulus of Elasticity in Compression. LNEC: Lisbon, Portugal, 1993. (In Portuguese)
55. Chen, B.; Liu, J. Effect of aggregate on the fracture behavior of high strength concrete. *Constr. Build. Mater.* **2004**, *18*, 585–590. [CrossRef]
56. Fazli, H.; Yan, D.; Zhang, Y.; Zeng, Q. Effect of size of coarse aggregate on mechanical properties of metakaolin-based geopolymer concrete and ordinary concrete. *Materials* **2021**, *14*, 3316. [CrossRef] [PubMed]
57. Whitehurst, E.A. Soniscope tests concrete structures. *J. Amer. Conc. Inst.* **1951**, *47*, 433–444.
58. Carvalho, J.; Manuppella, G.; Casal Moura, A. Portuguese decorative limestones. *Bol. Minas.* **2001**, *37*, 223–232. (In Portuguese)
59. Corish, A.T.; Jackson, P.J. Portland cement properties—Past and present. *Concrete* **1982**, *16*, 16–18.
60. Nixon, P.J. *Changes in Portland Cement Properties and Their Effects on Concrete*; Building Research Establishment: Watford, UK, 1988.
61. Zhang, S.P.; Zong, L. Evaluation of relationship between water absorption and durability of concrete materials. *Adv. Mat. Sci. Engin.* **2014**, *2014*, 650373. [CrossRef]

CHAPTER 5. CONCLUSIONS AND FUTURE WORK

5.1. CONCLUSIONS

This chapter presents the research conclusion for a set of buildings constructed in the 20th century and awarded with the Valmor Prize for Architecture, one of the most reputed awards in Portugal and one of the oldest awards in this area worldwide. The results increased the knowledge about the characteristics of the constructive materials used, namely renders, plasters and concretes, and their evolution throughout the studied period. They also provided clues about their state of conservation and durability, contributing to heritage valorisation, and enhancing aspects such as preserving historical-architectural heritage.

The work developed is described in four papers published in international journals indexed in ISI databases. In this chapter, the main conclusions drawn are summarised below, for each specific objective identified in the introduction chapter.

(1) Inspection of the main anomalies in renders, plasters and concretes. The degradation processes and their causes.

1. In the analysed buildings, which are characterised by above-average design, materials choice, and careful construction, as testified by the award, the state of conservation is primarily influenced by external rather than intrinsic factors. However, some types of anomalies are associated with specific construction technologies.
2. In the examined buildings a prevalent pattern was observed until the late 1960s, wherein the mortar coatings exhibited a multilayer configuration, occasionally comprising three or four distinct layers. Subsequently, the analysis was limited to coating mortars from only three case studies that postdated the 1960s: FCG (1975), JRP (1987), and UNL (2002). Although the sample space was limited, the findings indicated a shift towards simplified coatings characterized by a monolayer system, which in this case does not include painting layers. However, it is essential to exercise caution when attributing this reduction in the number of layers in construction works, leading to the adoption of monolayers, to the period after the late 1960s, which was not possible to ascertain with this study. This transformation coincided with the widespread adoption of Portland cement in

construction, since Portland cement mortars are not as dependent on a multilayer structure for the water protection capacity as the lime-based mortars.

3. Regarding architectural concrete, i.e., in buildings constructed between 1965 and 2002, the prominent anomalies detected are associated with reinforcement corrosion, mainly due to the low concrete coverings of rebars, favouring carbonation. However, a direct relationship between the average thickness of the reinforced concrete cover and the age of the buildings was not proved, nor with the differences in the carbonation depth, which is attributed to the different qualities of the concrete. Similarly, the conservation state of the architectural concrete surfaces is reasonable despite the restricted corrosion-related anomalies.

4. Despite the above-stated conclusions regarding the concrete materials, based primarily on observations and in-situ tests, the experimental campaign, in general, revealed that the materials show a good durability condition, as far as the physical and mechanical characteristics point out to a good performance, not indicating degradation, considering the age of the buildings and that they are still in use. However, the 1987 award-winning building demonstrated that its overall performance could compromise durability, requiring monitoring actions to prevent degradation.

(2) Characterisation of rendering mortars, plasters and concrete.

(2.1) Mineralogical, chemical and microstructural characteristics of renders and plasters

1. The aggregates of the renders and plasters are mainly of siliceous origin, including quartz, feldspars, and phyllosilicates, some of them from the clay group associated with the interface of sand grains in the older case studies (CVT (1903) and AR49 (1923)), indicating unwashed aggregates.

2. The aggregates of the stone-imitating mortars include crushed carbonate rocks (limestone and calcite crystals) or rounded quartzite particles. In some samples, limestone filler is also present.

3. The binders used followed a chronological application line in the studied buildings: air lime was the only binder used until 1938, with Portland cement employed afterwards.

4. Blended air lime-cement mortars were found in buildings constructed between 1938 and 1944. In these buildings, there are mortars formulated solely with air lime and others formulated solely with Portland cement.

5. The stone-imitating mortars studied correspond to the period during which their use prevailed in Portugal, often associated with the modernist style. Their common application spanned at least from the 1940s to the 1970s. *Marmorite* - one of the most common types of stone-imitating mortars in Portugal - were originally formulated using air lime; however in the case study AAC (1944) Portland cement was the binder used, and in the other stone-imitating mortars formulated with projected aggregates the binder was white cement mixed with air lime. In the case study EUA (1970) the *marmorites* were also formulated with Portland cement, as expected for this decade, given that the widespread use of Portland cement during this period.

6. Supplementary cementitious materials were also used, namely granulated blast furnace slags and fly ash, but only after the 1960s.

7. Salt ingress by chlorides and sulphates was detected but in low content. The highest values were found in the oldest buildings studied: CVT (1903) and AR49 (1923). It mainly affects buildings whose structure is of masonry (PRCB), which are more susceptible to capillary rising and to degradation by the action of water.

8. Finishing smooth white plasters were produced using a mix of gypsum and air lime (most likely as lime putty, according to literature). The latter is the main constituent, which is consistent with the literature for Portuguese gypsum-lime-based plasters in the 19th and 20th centuries. In this type of plasters, aggregates were occasionally detected. Trace constituents like anhydrite, quartz and muscovite were also identified by XRD, but they were typically found to be impurities present in the raw materials or byproducts of the manufacturing process.

9. Occasional salt contaminations were detected in finishing plasters.

(2.2) Physical and mechanical characteristics of renders and plasters

1. The mortars of the oldest buildings, between 1903 and 1944, which are made of air lime have the highest values of capillary absorption and simultaneously the highest drying rates and present the lowest values of compressive strength and dynamic modulus of elasticity, as expected for this type of mortars.

2. Concerning capillary water absorption, the blended lime-cement mortars in the buildings constructed between 1938 and 1944 have moderate values, i.e. these mortars fall between air lime and cement mortars. The lowest absorption value observed (Ccc = 0.35 $kg.m^{-2}.min^{-0.5}$) is higher than that of cement mortars (Ccc = 0.07 $kg.m^{-2}.min^{-0.5}$), but lower than the highest detected in air lime mortars (Ccc = 0.58 $kg.m^{-2}.min^{-0.5}$). Conversely, the highest absorption observed (Ccc = 0.63 $kg.m^{-2}.min^{-0.5}$) is lower than the highest value in air lime mortars (Ccc = 2.22 $kg.m^{-2}.min^{-0.5}$). In comparison to the maximum absorption values of Portland cement mortars, lime-cement mortars generally exhibit lower values of Ccc. There is only one exception, which was found in a sample from the 1939 award-winning building, where the recorded value for Ccc was 2.05 $kg.m^{-2}.min^{-0.5}$.
3. The introduction of Portland cement has increased the dynamic modulus of elasticity of the mortars.
4. The Portland cement mortars, applied in buildings erected after 1939, show the lowest values of capillary absorption and the highest values of mechanical strength.
5. The results of lime-gypsum-based plasters align with those found in the literature for white smooth thin layers applied in Portugal concerning capillary absorption with low values for Ccc < 1.00 $kg.m^{-2}.min^{-0.5}$.
6. The stone-imitating mortars showed the lowest capillary absorption and, consequently, the lowest open porosities. The reduction in these properties points to a direct relationship with the construction technique, which summarises the incorporation of fillers, as well as the tightening of the mortar during its application.
7. No significant differences were found in the physical and mechanical characteristics between the samples of renders and plasters; thus, being intended for internal or external application was not a crucial parameter for the choice of the material.

(2.3) Physical and mechanical characterisation of concrete materials

1. The coarse aggregates, mainly composed of crushed limestone particles, had their maximum size reduced, having decreased from the late 1960s onwards, as

exemplified by the case study FCG (1975), as set out in current Portuguese regulation by the time of construction.

2. The open porosity and bulk density values did not vary significantly. A slight tendency towards a reduction in bulk density and increase in porosity may be related to the variation in the maximum size of the coarse aggregate, which varies in the same direction as compacity.
3. Water absorption by capillary rising for all types of concrete studied does not show a consistent trend in the same direction as the open porosity.
4. Open porosity slightly increases towards the end of the analysed period, implying that this is not exclusively due to the characteristics of the binder but to the whole composite material itself.
5. The mechanical characteristics, except for the building awarded in 1987, show a trend towards an increase in the compressive strength values and the dynamic modulus of elasticity.
6. Except for the building awarded in 1987, the estimation of the compressive strength at 28 days of curing showed that the project design had been accomplished.

(3) The relationship between degradation processes occurring within a time span of less than 100 years and the materials and construction methods employed.

1. It was found that the renders and plasters of the buildings analysed are in a reasonable state of conservation. However, it was verified that when compared to the reinforced concrete structure buildings (RCB), which, in this case, include the buildings whose structure is entirely of reinforced concrete, the pre-reinforced concrete structure buildings (PRCB), presented a greater extent degree, and severity of degradation.
2. Since the PRCB, i.e. the buildings whose structures are made of non-reinforced concrete materials, are also the oldest buildings (1903 to 1944), the higher degree and extent of degradation of the assessed materials can be attributed to the more prolonged exposure to the agents of degradation, as well as to the

construction typology that makes them particularly vulnerable to the water action and salts crystallisation.

3. Regarding the RCB, the degradation processes are mainly related to concrete carbonation and consequent corrosion of the reinforcement. Nevertheless, the physical and mechanical results point to an evolution in the characteristics over the period under analysis, which embodies the application of the national regulations. The physical and mechanical properties of the analysed concrete materials reproduce an evolution towards the safety and durability requirements imposed by the national regulations on account of the advancement in the knowledge of structural performance and the scientific knowledge acquired throughout the 20th century.

(4) Recommendations for materials to be used in the rehabilitation and preservation of that built heritage.

1. For mortars, the requirements for compatibility purposes (i.e. conservation, repair and preservation works) should be considered case-by-case since the plasters and renders analysed are from buildings constructed in different periods throughout the 20th century.
2. The parameters investigated in the laboratory should be respected, advising the use of mortars with identical binder characteristics and proportions and aggregate mineralogy as defined in the developed research. In the case of Portland cement mortars, since the specifications of cement used are not known, the use of ordinary Portland cement and white Portland cement in the due cases is proposed in all cases, despite evidence of the use of composite cement in two samples, provided that they do not exceed the quantified values for the physical and mechanical properties.
3. The ranges of values obtained for the physical and mechanical characteristics to be considered in the compatibility requirements are parametrised in the paper referred to in section 4.4.2, where the results are organised by the binder and coating type.
4. The ranges of values obtained on multi-layer samples should be considered a good approximation of the whole coating properties when there was no possibility of individualising each layer or testing it independently.

5. In order to mitigate the escalation of defects resulting from corrosion-related degradation in concrete surfaces undergoing repair or restoration, it is advisable to utilise repair materials that are both compatible and well-informed. Consideration should be given to mineralogically adapted Portland cement and aggregate (fine and coarse) compositions that mirror the original materials, guaranteeing colour uniformity and consistent physical and mechanical properties.

5.2. FUTURE WORK

Based on the experimental work carried out, it is recommended to consider publishing a paper on concrete characterization. This article should encompass the outcomes of chemical, mineralogical, and microstructural characterization in addition to the physical and mechanical properties hereby presented. Some of these results have been preliminarily published in the proceedings of conferences and scientific meetings attended and presented by the author. These are mentioned in Chapter 6.

Future work also entails the dissemination of the research findings from this doctoral thesis to the stakeholders of the case studies. This aims to raise awareness of the results and potentially initiate conservation and rehabilitation efforts for their historic heritage.

In the context of understanding building materials from the early 20th century, expanding the research presented in this study remains necessary to encompass additional ordinary structures and potentially award-winning buildings. This expansion will widen the scope of research in this interdisciplinary field.

The knowledge of materials from the early 20th century and the evolution of using raw materials and manufacturing processes require specific and ongoing research. For older reinforced concrete structures, for example, it remains essential to have a more profound grasp of the materials employed, their alignment with the regulations during construction, and how they behave and adapt to present-day demands.

This study addressed the characterisation of materials applied in architecturally and historically significant buildings by providing straightforward answers to the questions raised. However, there is still a need to delve deeper into the use of other types of binders, such as natural hydraulic binders, especially natural cement, in Portugal.

In this regard, and as a contribution to conservation and restoration studies, it is crucial to identify works in which these materials have been used to examine them further. This work will fill a gap in the construction chronology, which is gradually being reduced, and to which this thesis has contributed. A methodology capable of clearly identifying the types of binders is essential, involving optical and petrographic microscopy and microstructural analysis supported by chemical microanalysis. One of the tasks performed involved the development of microscopic and microanalytical analysis of reference samples to define elemental chemical clusters, supplemented with petrographic information. In this scope, increasing the study on reference samples and continuing to develop and optimise this methodology is mandatory.

As for concrete, this thesis paves the way for characterising other historical or ordinary reinforced concrete structures. It goes beyond understanding their properties, evolution, and durability; it also involves seeking sustainable and compatible materials with reduced carbon footprint in their production. It is well-known that the current environmental concerns necessitate finding solutions that minimise environmental impact while meeting the established aesthetic and structural criteria for a building's intended function. The prohibition of Portland cement additives obtained from fossil fuel combustion (e.g. fly ash) requires both an adaptation of the industry and an increase in research and development activities to find suitable alternatives.

CHAPTER 6. DISSEMINATION WORK AND TRAINING ACTIONS

During the period in which the research grant was awarded, between 2016 and 2020, and during the period of the subsequent research work, the beneficiary and author of this doctoral thesis attended several national and international congresses and scientific meetings, presenting papers related to the topic of his thesis and which are included in the respective proceedings' books.

The publications in the respective congresses and others made as part of the developed research are listed below.

- Almeida, L., Santos Silva, A., Mirão, J., Veiga, M. R. (2016). Caracterização de revestimentos de edifícios galardoados com o Prémio Valmor de Arquitetura (1902 a 2002). In R, Póvoas, J. Mascarenhas-Mateus, (Eds.) *Proccedings of the 2º Congresso Internacional de História da Construção Luso-brasileira – Culturas partilhadas, setembro 2016*. V. 1, (pp. 153 – 161). Faculdade de Arquitetura da Universidade do Porto. Porto.

- Almeida, L., Santos Silva, A., Mirão, J., Veiga, M. R. (2017). Characterization of renders from buildings awarded with Lisbon's Valmor prize of architecture. In Mazzolani, F., Lamas, A., Calado, L., Proença, J. M., Faggiano, B. (Eds.) *Proceedings of the 3rd International Conference on Protection of Historical Constructions, 12-15 July 2017* (pp. 277 – 278) IST, Lisbon.

- Almeida, L., Santos Silva, A., Veiga, M. R., Mirão, J. (2017). As argamassas de cal no contexto e na construção de edifícios do século XX distinguidos com prémio Valmor de Arquitectura em Lisboa. *Comunicação ao 1º Simpósio Ibérico da Cal in livro de resumos,* 19 -20 outubro. Évora.

- Almeida, L., Santos Silva, A., Veiga, M. R., Mirão, J., Vieira, M. (2018). Betões de edifícios galardoados com o Prémio Valmor de Arquitectura. Caracterização e contributos para a sua salvaguarda. *Proceedings of Encontro Nacional Betão Estrutural, 7-9 de novembro 2018* (pp. 1195-1204). GPbE, LNEC. Lisboa.

- Almeida, L., Santos Silva, A., Mirão, J., Veiga, M. R. (2019). Evolution of mortars composition and characteristics during the 20th century – Study of Portuguese buildings awarded with Architecture Valmor Prize. In Álvarez, J., Fernández, J., Navarro, I., Durán, A., Sirera, R. (Eds.) *Proceedings of 5th Historic Mortars Conference. 19th – 21st June 2019*. *Pamplona, Spain* (pp. 959 – 972). RILEM, France.

- Almeida, L., Santos Silva, A., Veiga, M. R., Mirão, J. (2020). Evolução das técnicas e materiais de construção durante o século XX em Portugal. Aplicação a edifícios Prémio Valmor de Arquitetura. In M. Menezes, M. R. Veiga, A. Santos Silva, L. Nunes, J. S. Machado (Eds.) *Proceedings of 4º encontro de conservação e reabilitação de edifícios – ENCORE 2020. 3rd – 6th novembre 2020* (pp. 155-158) LNEC. Lisboa. DOI: 10.34638/yzys-hn57

- Almeida, L., Santos Silva, A., Veiga, M. R., Mirão, J., Vieira, M., (2020). Caracterização de betões de edifícios premiados com o Prémio Valmor de Arquitectura durante o séc. XX. Contribuição para a sua salvaguarda. *Proceedings of DEGRADA 2020 - 4.º Encontro Luso-Brasileiro de Degradação em Estruturas de Betão. 18th-19th de march 2021* (pp. 1 – 12) Universidade de Aveiro. Aveiro

- Almeida, L., Santos Silva, A., Veiga, M. R., Mirão, J., Vieira, M., (2021). Caracterização de argamassas e betões de edifícios do século XX galardoados com o Prémio Valmor de Arquitectura. Estado de conservação e contributos para a sua salvaguarda. In Oliveira Santos, L., Júlio, E., Sena Cruz, J., (Eds.) *Proceedings of R&BE 2020 - Reabilitar & Betão Estrutural 2020. Congresso Nacional, 3rd-5th novembre 2021* (pp. 703-713). LNEC. Lisbon.

- Almeida, L., Vieira, M. (2021). A afirmação do betão armado como valor patrimonial durante o século XX através de edifícios Prémio Valmor e Municipal de Arquitetura. *Cadernos do Arquivo Municipal*. 2ª Série Nº 16, pp. 205-235. CML. Lisboa.

The author also attended an international course entitled: Microscopic Techniques to study mineral materials in cultural heritage, held at the University of Applied Arts, Vienna, on 24th – 28th September 2018.

The author was part of the working group involved in the research project funded by FCT called DB-HERITAGE (Database of Construction Materials of Historical and Heritage Interest - PTDC/EPH-PAT/4684/2014), actively participating in the construction of a repository for historical materials, including samples and data provided by this thesis.

After finishing his PhD grant, the author was awarded a grant under the FCT CEMRESTORE project (ref. POCI-01-0145-FEDER-0316–2) - Mortars for the conservation of early 20th century buildings - Compatibility and Sustainability, between 2020 and 2021, where he collaborated on experimental work on topics related to the characterisation of mortars and concretes from Portuguese buildings at the advent of the introduction of Portland cement in Portugal. The work contributed to developing scientific knowledge related to the central aspects of his doctoral thesis.

As a result of his participation in this research project, he authored and co-authored the following publications:

- Figueiredo, C., Moutinho S., Pimenta do Vale, C., Andrejkovičová, S., Velosa, A., Tavares, A., Almeida, L., Santos, A. R., Santos Silva, A., Vieira, M., Veiga, R. (2020). A produção e utilização dos cimentos entre o final do século XIX e o primeiro quartel do século XX. In M. Menezes, M. R. Veiga, A. Santos Silva, L. Nunes, J. S. Machado (Eds.) *Proceedings of 4º encontro de conservação e reabilitação de edifícios – ENCORE 2020. 3rd – 6th novembre 2020* (pp. 155-158) LNEC. Lisboa. DOI: 10.34638/yzys-hn57

- Almeida, L., Santos Silva, A., Figueiredo, C., Moutinho, S., Andrejkovičová, S., Vale, P. C., Veiga, M., R., Costa, A. T., Velosa, A. (2021). Argamassas e ligantes do Mercado do Bolhão e Teatro Nacional de São João no Porto – variabilidade e implicações para a sua reabilitação. In Costa, A., Tavares, A, Rodrigues, H, Lapa, J. (Eds.) *Proceedings of CONREA'21 – Congresso de Reabilitação. Universidade de Aveiro, 29th, 30th june and 1st july 2021* (pp. 435 - 437). UA editora, DOI: 10.48528/gy68-v843; http://hdl.handle.net/10773/31632

- Almeida, L., Silva, A. S., Figueiredo, C., Moutinho, S., Andrejkovičová, S.; Vale, C. P., Veiga, M. R., Costa, A. T., Velosa, A. (2021). Argamassas e Ligantes do Mercado do Bolhão e Teatro Nacional de São João, no Porto: variabilidade e implicações para a sua reabilitação. *Al-Madan*. 2.ª Série, 24 (pp. 87 - 92). Centro de Arqueologia de Almada. Almada

- Almeida, L., Santos, A.R., Santos Silva, A., Veiga, M.d.R., Velosa, A. (2023). Characterization of Mortars and Concretes from the Mirante da Quinta da Azeda, Setúbal (Portugal). A Case Study from the Beginning of the 20th Century. In Bokan Bosiljkov, V., Padovnik, A., Turk, T. (Eds.) *Conservation and Restoration of Historic Mortars and Masonry Structures. HMC 2022. RILEM Bookseries*, V. 42 (pp. 243 - 257). Springer, Cham. https://doi.org/10.1007/978-3-031-31472-8_19

The author collaborated in the organisation of an international Webinar entitled CEMRESTORE - 'Mortars for the conservation of early 20th century buildings - Compatibility and Sustainability'. This webinar occurred on 14 April 2021, jointly organised by LNEC with the Faculty of Architecture of the University of Porto and the University of Aveiro. This webinar aimed to publicise the project's objectives and was attended by several national and international guests who gave an overview of the use of cement at the beginning of the 20th century, focusing on essential aspects such as their development and application in various international contexts.

REFERENCES

[1] Henriques, F. M. A. (2005). Challenges and Perspectives of replacement mortars in architectural conservation. In C. Groot (Ed.) *Proceedings of* the *International RILEM Workshop on Repair Mortars for Historic Masonry. Delft, The Netherlands, 26th - 28th January 2005* (pp. 143 – 161). RILEM.

[2] Damas, A. L.; Veiga, R.; Faria, P. (2016). Caraterização de argamassas antigas de Portugal – contributo para a sua correta conservação. In M. Menezes, J. Delgado Rodrigues, D.Costa (Eds.) *Proceedings of Congresso Ibero-Americano "Património, suas Matérias e Imatérias", Lisbon, LNEC, 2nd -3rd November 2016* (pp. 1-19). LNEC.

[3] Hughes, D. C.; Jaglin, D.; Kozłowski, R.; Mucha, D. (2009). Roman cements — Belite cements calcined at low temperature, *Cement and Concrete Research*, 39(2), 77-89. https://doi.org/10.1016/j.cemconres.2008.11.010.

[4] Kozłowski, R.; Hughes, D.; Weber, J. (2010). Roman cements: key materials of the built heritage of the 19th century. In M. B. Dan, R. Přikryl, Á. Török, (Eds) *Materials, Technologies and Practice in Historic Heritage Structures* (pp. 259 - 277). Springer, Dordrecht. https://doi.org/10.1007/978-90-481-2684-2_14.

[5] Cantisani, E.; Falabella, A.; Fratini, F.; Pecchioni, E.; Vettori, S.; Antonelli, F.; Giamello, M.; Lezzerini, M. (2018). Production of the Roman Cement in Italy: characterization of a raw material used in Tuscany between 19th and 20th century and its comparison with a commercialized French stone material, *International Journal of Architectural Heritage*, 12(6), 1038-1050, DOI: 10.1080/15583058.2018.1431730

[6] Weber, J.; Gadermayr, N.; Kozłowski, R.; Mucha, D.; Hughes, D.; Jaglin, D.; Schwarz, W. (2007). Microstructure and mineral composition of Roman cements produced at defined calcination conditions, *Materials* Characterization, 58(11–12),1217-1228. https://doi.org/10.1016/j.matchar.2007.04.025.

[7] Hughes, D. C.; Swann, S.; Gardner, A. (2007). Roman cement: Part one: Its origins and properties. Journal of Architectural Conservation. 13(1), 21 - 36, https://doi.org/10.1080/13556207.2007.10784986

[8] Vyskocilova, R.; Schwarz, W.; Mucha, D.; Hughes, D.; Kozłowski, R.; Weber J. (2007). Hydration processes in pastes of Roman and American natural cements. *Journal of ASTM International*, 4 (2), https://doi.org/10.1520/JAI100669.

[9]. Hughes, D. C.; Weber, J.; Kozlowski, R. (2010). Roman Cement for the Production of Conservation Mortars. In J. Válek, C. Groot & J.J. Hughes (Eds.) *Proceedings of 2nd Historic Mortars Conference & Rilem TC 203-RHM Repair Mortars for Historic Masonry Final Workshop, Prague, 22nd-24th August.2010* (pp. 1043 – 1050). RILEM.

[10] Gurtner, C.; Hilbert, G.; Hughes, D.; Weber, J.; Kozłowski, R. (2012). Manual on best practice in the application of roman cements. Roman cement, past and present. ROCARE.

[11] Varas, M. J., Alvarez de Buergo, M., Fort, R. (2005). Natural cement as the precursor of Portland cement: Methodology for its identification. *Cement and Concrete Research*, 35, 2055 – 2065, doi:10.1016/j.cemconres.2004.10.045.

[12] Ferreira, C. A. (1989). Betão: a idade da descoberta. Lisboa, Passado Presente.

[13] Neves, J. C. (1891). Relatório apresentado à direcção da 3ª circumscripção hydraulica em 21 de Abril de 1890. In “Revista de Obras Publicas e Minas”, Lisboa, XXII, June-Septembre, 1891

[14] Mascarenhas-Mateus, J. & Castro, C. (2018). The Portland cement industry and reinforced concrete in Portugal. In B. Wouters, S. Voorde, I. Bertels, B. Espion, K. Jonge & D. Zastavni (Eds.) *Building Knowledge, Constructing Histories, Proceedings of the 6th International Congress on Construction History*, V. 2, (pp. 903-911). London, CRC Press /Taylor & Francis. https://doi.org/10.1201/9780429446719

[15] Rocha, D. V. (2015). Argamassas de cimento romano utilizadas em edifícios do início do século XX. Master thesis. Universidade de Aveiro. Aveiro.

[16] Diogo, M. (2000). Indústria e engenheiros no Portugal de fins do século XIX: o caso de uma relação difícil. *Scripta Nova, Revista Eletrónica de Geografía y Ciencias Sociales*. 69(6). https://www.ub.edu/geocrit/sn-69-6.htm.

[17] Neves, P. (2007). Grandes empresas industriais de um país Pequeno: Portugal. Da década de 1880 à 1ª Guerra Mundial. PhD thesis. Universidade de Lisboa. Lisboa.

[18] Neves, J. C. (1894). Estudos sobre cimentos estrangeiros importados em Portugal. *Revista de Obras Públicas e Minas*, 25, 265-371.

[19] Callapez, P. M.; Pinto, J. M. S.; Santos, V. F.; Brandão, J. M. (2014). A mina de carvão do Cabo Mondego e a paleontologia portuguesa. In J. M. Brandão (Ed.) *Memórias do carvão: jornadas internacionais: resumos*. Batalha, (pp. 1 -9).

[20] CEN (2011). EN 197-1:2011 Cement Composition, specifications and conformity criteria for common cements. European Committee for Standardization.

[21] Freire, M. T., Santos Silva, A., Veiga, M., Brito, J., Schlütter, F. (2016). Natural or Artificial? Multi-Analytical Study of a Scagliola from Estoi Palace Simulating Imperial Red Porphyry. *Microscopy and Microanalysis*, 22(6), 1281–1303. https://doi.org/10.1017/S1431927616011909

[22] Lorena, A. (2007). António Domingos Meira decorador e estucador de eleição. C. M. de Cascais (Ed.). *A presença do estuque em Portugal. Do Neolítico à época contemporânea*, (pp. 208–217). Câmara Municipal de Cascais: Cascais.

[23] Wirsching, F. (2005). Calcium Sulfate. In Ullmann’s Encyclopedia of Industrial Chemistry. Wiley-VCH Verlag GmbH & Co. KGaA.

[24] Freire, M. T. (2016). Restoration of ancient Portuguese interior plaster coatings: Characterization and development of compatible gypsum-based products. PhD thesis, Instituto Superior Técnico. Lisboa.

[25] Gárate-Rojas, I. (1999). Artes de los Yesos. Yeserias e Estucos Munilla-Lería, Madrid: Instituto Español de Arquitectura, MRRP, Universidad de Alcalá.

[26] Cizer, O., Van Balen K., Van Gemert, D., Elsen J. (2008). Blended lime-cement mortars for conservation purposes: Microstructure and strength development. Structural Analysis of Historic Construction – D’Ayala & Fodde (Eds.). Taylor & Francis Group, London.

[27] Silva, B. A., Ferreira Pinto, A. P., Gomes, A. (2015). Natural hydraulic lime versus cement for blended lime mortars for restoration works. Construction and Building Materials, 94, 346-360. https://doi.org/10.1016/j.conbuildmat.2015.06.058.

[28] Bosc, J.-L., Chauveau J.-M., Clément, J., Degenne, J., Marrey, B., Paulin, M. (2001). Joseph Monier et la Naissance du Ciment Armé, 180p. Éditions du Linteau, Paris.

[29] Licordari, M. (2016). A Propagação do Betão Armado em Portugal e as Primeiras Indústrias de Cimento do País. In R, Póvoas, J. Mascarenhas-Mateus, (Eds.)

Proccedings of the 2nd Congresso Internacional de História da Construção Luso-brasileira – Culturas partilhadas, setembro 2016. V. 1, (pp. 1189 – 1201). Faculdade de Arquitetura da Universidade do Porto. Porto

[30] Decreto nº 4036 (1918). Regulamento para o Emprego do Betão Armado, Ministério do Comércio, abril de 1918. Lisbon

[31] Ministério das Obras Públicas (1935). Decreto nº 25948, Outubro de 1935, corrigido pelo Decreto nº 33021, Regulamento do Betão Armado, Ministério das Obras Públicas, setembro de 1943. Lisbon

[32] Ministério das Obras Públicas. (1967). Decreto nº 47723. Regulamento de Estruturas de Betão Armado; Imprensa Nacional de Lisboa. Lisbon.

[33] Ministério das Obras Públicas (1983). Decreto-Lei nº 349-C/83, Regulamento de Estruturas de Betão Armado e Pré-Esforçado, Julho de 1983. Lisbon

[34] Ministério da Habitação, Obras Públicas e Transportes (1983). Decreto nº 235/83, Regulamento de Segurança e Acções para Estruturas de Edifícios e Pontes, Maio de 1983. Lisbon

[35] CEN (2010). EN 1992-1-1 Eurocode 2: Design of concrete structures – Part 1-1: General rules and rules for buildings. European Committee for Standardization.

[36] David, R. M. F. S. (2016). A ação cultural e mecenática de Fausto de Queirós Guedes, 2º Visconde de Valmor (1902-1943) e o prémio Valmor de Arquitetura. PhD Thesis. Faculdade de Letras da Universidade de Lisboa.

[37] Montez, P. (1984). O prémio Valmor e a Evolução da Arquitetura. Academia Nacional de Belas Artes. Lisboa

[38] Miranda, R. M. (2010). Os últimos vinte anos do Prémio Valmor e Municipal de Arquitectura. Master Thesis. Faculdade de Arquitectura da Universidade do Porto. Porto.