STANDARD PROTOCOLS FOR RESTORING HERITAGE CEMENTING MATERIALS

BY

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Declaration

I, Maphole Emelly Loke, declare that the contents of this thesis represent my own unaided work and that the thesis has not previously been submitted for academic examination towards any qualification. Furthermore, it represents my own opinions and not necessarily those of the Cape Peninsula University of Technology.

Signed

03rd February 2020

Date

Dedication

To the first lady of my life, my maker, my guide and role model, the strongest woman I always look up to; my grandmother 'Mats'eliso Loke. For teaching me that all things are possible with God. She taught me all I know about life, and for that, I shall forever be grateful. You are the reason I soldiered on throughout this journey.

You are my hero Motloung. Kea u leboha!
Psalm 116:12

Abstract

The history of the South African construction industry dates as far back as the seventeenth century, with structures such as the Castle of Good Hope, cathedrals, museums and memorials, among many others. Heritage structures represent the history of a country and its development. These structures do not only elaborate on the history, but also embody unique designs and materials used for their construction. This makes historic structures physically distinct from modern constructions. However, heritage structures often show critical signs of deterioration, which threaten their existence thereof. In attempts to rescue these national treasures, the use of Portland cement has been a common practice, but there are problems with using such modern materials. The application of incompatible materials is said to accelerate the deterioration and endanger the authenticity of these monuments. This practice is a result of a misunderstanding of the original material behaviour and has been responsible for the failure of many restoration projects. For developing countries, South Africa in particular, this approach has undoubtedly impacted negatively on the economy, as repeated repairs are necessary.

Characterisation of the original cementing materials is the key to compatible restoration of heritage buildings. The research describes the development of an integrated protocol to establish the properties of the original heritage cementing materials. The key objective for the study was to investigate and standardise an analytical procedure whose primary purpose is the identification of major and minor components of heritage cementing materials and binder-to-aggregate ratio, which would be useful for production of repair materials. The results obtained from the experimental investigation into the physical, chemical and mineralogical properties of the original materials from the Castle of Good Hope and Robben Island are presented in this study. The experimental results were analysed for their ability to provide useful data at minimal costs. After that, a standard protocol was developed, incorporating the procedure for sampling as well as preparation of the sample, material analysis and documentation.

The standard protocol includes the cohesion test, visual investigation and titration test for analysis of the salts and metal oxides present in the materials. These tests provide relevant data for the search of replica repair materials. The standard protocol will be useful for local heritage authorities, as it could be incorporated into the conservation management plans prior to restoration works. This will ensure compatible and sustainable restoration of historic buildings, not only in South Africa, but around the world.

The heritage materials for the two oldest buildings in the Western Cape were made of hydraulic lime-based mortars, seashells, and uniform and well-graded sand. Less than 5% of the materials showed no traces of a binder. The restored areas were restored with cement-based materials. The affordable standard methodology for characterising original heritage cementing materials involved the sample collection that represents the majority of the building, sample preparation, detailed visual investigation and cohesion test, as well as hydraulicity and cementation indices analysis to determine the type of binder. These tests were concluded to be convenient, easy to conduct and cost-effective.

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Nomenclature

Constants

b/a	Binder-to-aggregate ratio
C_c	Coefficient of curvature
CI	Cementation Index
C_u	Coefficient of uniformity
D_{10}	The sieve size when 10% of the particles are still being retained
D_{30}	The sieve size when 30% of the particles are still being retained
D_{60}	The sieve size when 60% of the particles are still being retained
F_s	The quantity of soluble iron
f_{ZnSO_4}	Molarity factor of the $ZnSO_4$ solution 0.025 M
M	Molarity which demonstrates the concentration of a substance (measured by number of moles) in a litre of water
M_1	Constant dry mass
M_2	Saturated mass
M_3	Hydrostatic mass
M_{Mg}	Soluble magnesium
N	Normality which defines chemical activity in terms of concentration (eq/l)
V_{EDTA}	Volume of EDTA solution used for the titration of Fe_2O_3
$V_{EDTA\ Ca + Mg}$	Volume of 0.025 M EDTA used to titrate the soluble Ca and Mg
$V_{EDTA•\ Ca}$	Volume of EDTA 0.025 M solution used for titration of soluble calcium
V_{ZnSO_4}	Volume of $ZnSO_4$ solution used for the titration
W_1	First weight of the original sample
W_2	Second weight after acid dissolution and washing with distilled water
W_3	Third weight after boiling in saturated sodium carbonate, washing with HCl and water

Greek letters

ρ_a	Apparent density
ρ_r	Real density
ε	Porosity

Terms and concepts

AAS	Absorptive Atomic Spectroscopy
ASTM	American Society for Testing and Materials
CMP	Conservation Management Plan
CPUT	Cape Peninsula University of Technology
C-S-H	Calcium-Silicate-Hydrate gel
DSC	Differential Scanning Calorimetry
DTA	Differential Thermogravimetric Analysis
EDS	Energy Dispersive Spectroscopy
EDTA	Ethylenediaminetetraacetic Acid
EDX	Energy Dispersive X-ray
FTIR	Fourier Transform Infra-Red spectrometry
GDP	Gross Domestic Product
HI	Hydraulicity Index
IC	Ion Chromatography
ICCROM	International Centre for Study of the Preservation and Restoration of Cultural Property
ICP- OES	Inductively Coupled Plasma-Optical Emission Spectroscopy
MDT	Minor Destructive Testing
MSDS	Material Safety Data Sheet
NDT	Non-Destructive Testing
NHL	Natural Hydraulic Lime
NHRA	National Heritage Resources Act
OPC	Ordinary Portland Cement
RILEM	International Union of Laboratories and Experts in Construction Materials, Systems and Structures
SABS	South African Bureau of Standards
SAHRA	South African Heritage Resources Agency
SANS	South African National Standards
SEM	Scanning Electron Microscopy
TEA	Triethanolamine
TGA	Thermogravimetric Analysis
UNESCO	United Nations Educational, Scientific and Cultural Organisation
UPV	Ultrasonic Pulse Velocity
XRD	X-Ray Diffraction
XRF	X-Ray Fluorescence

Chapter 1 Introduction

The characterisation of historical materials is a significant aspect that deserves special attention prior to any restoration project, particularly historic buildings. Special attention has been paid to characterising historic mortars in an effort to achieve compatible restoration. The analysis has been conducted using various analytical techniques, which differ among researchers. The current study intended to experimentally investigate a standard protocol for characterising original heritage mortars for sustainable and long-term restoration of historic structures. This chapter discusses the overview of the scope, the aims and objectives, and the organisation of the thesis.

1.1 Background and problem statement

Historical buildings are structures that represent the unique past of a country (Gleize *et al*., 2009; Sandbhor & Botre, 2013; Hormes *et al*., 2016). Historical structures have unique qualities that represent the aesthetic culture, natural heritage, historic religion, military history, political history or special events of the past. These structures are usually distinctive in design, architecture and material used for their construction. In most cases, they were built with materials that no longer exist, or have been abandoned or replaced by the construction industry of the current era. Notable heritage structures around the world include the Colosseum in Rome, the Old Jerusalem City and its Walls in Israel, the Leaning Tower of Pisa in Italy, St Mary's Cathedral and St Michael's Church at Hildesheim in Germany, the Castle of Good Hope in South Africa, the South African Astronomical Observatory in South Africa, nine areas at the Bo-Kaap in South Africa, The Taj Mahal in India, The Forbidden City in China and Kronburg Castle in Denmark, to mention but a few.

Historic buildings are selected and protected by the United Nations Educational, Scientific and Cultural Organisation (UNESCO). The selection of sites is based on their outstanding universal values. As of September 2019, UNESCO selected a total of five cultural sites (Fossil Hominid sites, Mapungubwe Cultural Landscape, Robben Island, Richtersveld Cultural and Botanical Landscape and Khomani Cultural Landscape), four natural sites (Cape Floral Region Protected Areas, iSimangaliso Wetland Park, Vredefort Dome and Barberton Makhonjwa Mountains) and a mixed heritage site (Drakensberg Park) as world heritage sites in South Africa (World Heritage Sites in South Africa, 2018).

In South Africa, heritage sites are declared as either national or provincial, based on their historical significance by an agency of the Department of Arts and Culture known as the South African Heritage Resources Agency (SAHRA). SAHRA was established in terms of Section 11 of the National Heritage Resources Act (NHRA), No. 25 of 1999. The SAHRA website (2018) lists several heritage buildings, namely: The Union Buildings, South African Parliament, SAHRA building, Mandela House, Sharpeville Police Station, Constitutional Hill Precinct, and St. George’s Cathedral as some of the heritage buildings across the country (South African Heritage Resources Agency, 2018). All these buildings are important tourist attractions for the country.

Many of the heritage structures last for centuries and serve as true historical symbols and are valued for their embodiment of historical architecture and culture (Gleize *et al*., 2009; Sandbhor & Botre, 2013; Hormes *et al*., 2016). Meli *et al*. (2007) also suggest that heritage structures tend to last longer than most modern constructions. Historical sites also attract tourists to a country and thus stimulate its economy. In

a survey by the World Travel and Tourism Council (2018), the travel and tourism industry contributed massively towards the gross domestic product (GDP) and employment. The industry has contributed 2.9% (direct) and 8.9% (total) to GDP, 4.5% (direct) and 9.5% (total) employment in 2017, in South Africa (World Travel and Tourism Council, 2018).

Historic buildings deteriorate with apparent signs of material failure which is caused by several factors that include:

Ageing of the structure
Neglect and ignorance on the part of humans
High humidity
High rainfall
Temperature changes due to seasonal changes and drastic fluctuations in temperatures of day and night
Atmospheric moisture
Exposure to soluble salts
Air pollution
Chemical processes
Biological attack by plants, animals and humans (Lawrence *et al*., 2004).

It is critical to guarantee the long-term existence of these structures through protection against the deterioration factors mentioned above. The evidence in literature indicates that the long-term existence of historic structures is achieved through regular compatible restoration. However, most countries still lack understanding of heritage mortar analysis for restoration purposes. According to Lanas and Alvarez (2003), most restorers use Portland cement for the restoration of historic buildings originally made of lime-based mortars. They state that Portland cement possesses different properties from the original materials, thus causing reactions and acceleration of defects. The application of Portland cement-based materials on heritage buildings is common in South Africa as well. This was mentioned in an interview with SAHRA Built Environment Unit Manager Mr Mwasinga, who stated that during most maintenance projects of heritage buildings in South Africa, the common practice was the replacement of the original material with Portland cement material (Mwasinga, 2018).

A study by Hormes *et al*. (2016) emphasises the need to analyse material properties and means of testing (testing techniques) historic mortars prior to restoration work. This would yield the production of a good repair mortar with matching properties to those originally used during construction. In addition to Hormes' recommendation, there is a need for further studies addressing the optimum standard protocol for characterisation of original heritage cementing materials for long-lasting repairs. It is essential to carefully select specific tests to study the components of the original mortar to ensure compatibility with the repair mortar.

1.2 Research problem

Mortar is a vital material that determines the functioning of a masonry wall as it binds blocks together (Salvadori, 1982; Holmes & Wingate, 1997). Several researchers attempted to create a systematic approach and a holistic methodology for analysis of heritage building mortars (Jedrzejewska, 1960;

Charola *et al*., 1986; Middendorf & Knöfel, 1991; Van Balen *et al*., 1999; Middendorf *et al*., 2005; Rampazzi *et al*., 2010; Papayianni *et al*., 2013; Sandbhor & Botre, 2013; Apostolopoulou *et al*., 2017). No standard analytical technique has been documented, except by Middendorf *et al*. (2005), who introduced a summary methodology for both chemical and mineralogical characterisation. It is therefore difficult to find a suitable method for characterising heritage materials in developing countries such as South Africa.

In South Africa, there is no standard procedure or a documented guideline in any of the construction standards (South African Bureau of Standards (SABS) and South African National Standards (SANS) to follow when investigating and analysing heritage cementing material properties *(chemical, physical, mineralogical/petrographic and mechanical).* Sourcing the correct heritage material replacements is a challenge. Generally, there is a lack of technical knowledge and understanding of heritage structure conservation in most developing countries as far as material analysis is concerned. As a result, the heritage building conservation management plans (CMPs) usually lack clarity on material analysis methodology, thus causing sudden failure of repair materials (Mwasinga, 2018).

1.3 Research question

What is the optimum standard protocol (*from a civil and structural engineering perspective*) used for analysing heritage cementing materials?

1.4 The specific need

This research addresses the present concern by the South African Heritage Resources Agency as far as heritage building conservation is concerned. This will benefit the heritage authorities within South Africa and abroad in terms of providing clear methodology to be followed when characterising cementing materials prior to restoration works. The standard protocol will be useful for local heritage authorities through incorporation into the conservation management plans prior to restoration works. This will help the South African Department of Arts and Culture and other heritage authorities in South Africa and abroad to overcome the common challenge facing heritage structures in terms of conservation and preservation. It will ensure that authenticity is maintained on future maintenance projects and avoid the problem of repeated repairs like the case of the Owl House Museum (Eastern Cape, South Africa) and the re-pointing and re-plastering of Robben Island, where incompatible cement materials were used (South African Heritage Resources Agency, 2018; Conditional Assessment of Buildings on Robben Island, 2018).

1.5 Aims and objectives

The major aim of this research is to experimentally investigate the physical, mineralogical and chemical properties of the heritage cementing materials from different centuries and thus provide results related to the optimum procedure to be followed when characterising original heritage mortars for application in restoration practice. This was achieved through experimental characterisation of the original mortar composition of selected heritage structures in the Western Cape Province, South Africa, whose materials belong to the seventeenth, eighteenth and nineteenth centuries. The study provides the minimum tests (time and cost-effectiveness) considered sufficient to identify the materials (binder and aggregate proportions) that were used in the initial construction of heritage buildings. The results can be used as a guide during a search for restoration materials.

The objective of the work has been selected to fill the identified gaps in the characterisation of the heritage cementing materials which are as follows:

- Characterise the heritage cementing materials (chemically, physically and mineralogically) on structures built in the 1600s to 1800s.
- Select and set the standardised optimum number of tests for identifying components of heritage cementing materials.

1.6 Research context and significance

This research study falls mainly within civil and structural engineering regarding construction materials. Special emphasis on maintaining heritage structures' actual appearance and status is made in this research, thus highlighting a historical point of view. Setting the standard protocol for restoring heritage buildings would be a profitable investment for enhancing the country's economy through boosting the Tourism industry, mainly because keeping these structures in the right conditions preserves authenticity throughout the years.

The study investigated time and cost-effective methodologies for characterising heritage cementing materials. The findings from this study will help in the analysis of the components of the original materials, leading to the search of repair mortars using a similar recipe to the original. The standardised methodology could be used in future restoration projects of old buildings in the Western Cape Province and generally in Africa and abroad. Therefore, it is of utmost importance to investigate in detail, the materials originally used in order to match them during maintenance works.

1.7 Scope and delimitation

The scope of this work was restricted to only identifying the composition of the heritage mortars. The production of replica repair mortar was not a part of the present work. This research focused solely on the characterisation of heritage cementing materials for the buildings constructed between 1600 and 1899 in the Western Cape Province in South Africa. Heritage cementing materials were tested chemically, physically and mineralogically, using only non-destructive testing (NDT) and minor-destructive testing (MDT) methods. The study did not investigate the mechanical properties of the heritage mortars, as the tests in this regard require large samples which could not be collected from the heritage buildings. No imported artificial materials were tested in this work. The challenges facing restoration of heritage cementing materials in South Africa are included in this thesis. Such challenges could be alleviated through the practice of material analysis prior to restoration works.

No attempt was made to address any standard protocols for the restoration of defects on any other building sections such as the roof, timber, steel, finishes or any other aspects that do not involve cementing materials. The general material analysis of heritage structures could be a separate project altogether. The scope of the present research did not cover the effects of restoration technologies on the compatible restoration of heritage buildings, nor the impact of weathering on the degradation of heritage cementing materials.

1.8 Assumptions

This study aimed to obtain useful details of original cementing materials used for the construction of the heritage buildings in the Western Cape, South Africa with the use of the minimum number of tests. The following assumptions were made in this study:

The original cementing materials used for the construction of the heritage buildings in this study contain no traces of Portland cement, but use lime as a binder. This is due to evidence in the literature showing that the majority of heritage structures constructed before the mid-1800s were built using lime as a binder, because the use of Portland cement only came afterwards (Sanjurjo-Sánchez *et al*., 2010).

The materials from the same era had not been altered, ensuring that analysis was carried out on the original material, unless otherwise stated in any available literature or evident from the visual investigation of the structures and through interviews with relevant stakeholders.

Weathering factors had only minor effects on the original material composition; hence, their effect on the materials tested was neglected. However, to address the concern on this, material sampling was carried out on unexposed surfaces that were assumed to be unaffected or somewhat less affected by the environmental weathering agents (where possible).

The test methods used in this study was in alignment with the literature studied, with an assumption that such methods align with the material testing standards (RILEM or ASTM) and guidelines and where such does not apply, non-standard testing with reference to literature took place.

1.9 Methodology

The repair of heritage cementing materials requires the use of compatible materials (Gulzar *et al*., 2013). To achieve the objectives for this research, an experimental investigation was used to analyse the physical, mineralogical and chemical properties of heritage buildings' cementing materials in the Western Cape; South Africa. The literature around the characterisation of historic mortars was thoroughly studied and used as a reference for the analytical techniques selected. The materials from the walls and floor of existing buildings (Castle of Good Hope and Robben Island) were collected for analysis after a permit was granted by the South African Heritage Resources Agency (SAHRA). No harm was caused on the structures, as very limited materials were collected by the use of a small chisel and a hammer, and by hand picking on loose surfaces that were already severely damaged. The study employed different analytical techniques such as visual investigation, cohesion test, wet chemical analysis and titration tests. The tests were carried out on non-standard quantities due to the limitations on material extraction from heritage sites. The experiments were conducted in laboratory facilities at the Cape Peninsula University of Technology, Bellville campus. A set of cost-effective, simple yet effective standard protocols for characterisation of heritage materials for future restoration works is presented at the end of this study.

1.10 Organisation of the dissertation

The layout of the thesis is as follows:

Chapter 1: Introduction: This chapter serves as an overall introduction to the research topic. It provides the background and motivation for conducting characterisation of heritage cementing materials, the overall objectives and the items delineated in the study.

Chapter 2: Review of the Literature: The literature related to the characterisation of heritage cementing materials is discussed thoroughly in this chapter. It includes historic mortar characterisation techniques from around the world and case studies in the literature. Gaps in the literature are identified and further emphasis on the need for the present work to address areas that received less attention is made.

Chapter 3: Research methodology: This chapter provides the systematic experimental procedure and equipment used to carry out the experiments, research design, settings, research methodology and justifies the data gathering methods used to physically, mineralogically and chemically characterise the samples. This is done by citing reference literature on the methods and techniques. The criteria for selection of a particular analytical technique over another is outlined.

Chapter 4: Results: This chapter reports on the results of the laboratory experiments conducted. The obtained results are presented and structured in a consistent manner as well as integrated to demonstrate the findings in relation to the research question, aims and objectives of the study. This includes data obtained from laboratory experiments and onsite investigations.

Chapter 5: Discussion: This chapter explores the interpretation and evaluation of the results concerning the theoretical body of knowledge around cementing materials characterisation. An in-depth analysis of the results obtained by the current study in reference to the literature is made. The analysis is in terms of historic material characterisation and the standards for obtaining the properties of cementing materials of historic buildings are presented. The results from Chapter 4 are covered in order to select the optimum set of tests for characterisation of heritage materials.

Chapter 6: Conclusions and Recommendations: This chapter provides a summary of the study and the findings in addressing the research problem. Conclusions are drawn following the findings and literature reviewed. Recommendations are made on future research around heritage structure restoration using material characterisation.

Chapter 2 Literature review and theory

This chapter provides relevant theory on heritage mortars, the characterisation of cementing materials for heritage structures and an overview of previous studies conducted (publications and investigations). The chapter also elaborates on the characterisation methods that were applied in previous research, as well as an assessment of the results obtained in the literature.

2.1 Introduction

Heritage buildings are structures with proven exceptional and significant qualities that represent a country's aesthetic culture, military history, politics and/or special events of the past. All nations around the world have these historical legacies that tell a unique story about the country. Heritage buildings are of national importance and are usually unique in design, architecture and materials used. The selection of world heritage structures is based on their outstanding universal values and is regulated by the United Nations Educational, Scientific and Cultural Organisation (UNESCO).

If these heritage legacies are not properly maintained, the economy suffers because of repeated repairs and loss of authenticity during restoration, which causes a reduction in tourism, hence, their protection with proper maintenance and use of compatible materials need to be an unquestioned priority. This will ensure their continued existence and functionality, which are mainly dependent on proper and regular maintenance. In order to implement the practice of adequate maintenance of heritage buildings, a tremendous amount of research was conducted with the purpose of understanding the characteristics of cementing materials on different heritage buildings around the world as well as compatible restoration of these structures. Examples of such studies are discussed in this chapter.

2.2 Mortar

The term 'mortar' has various definitions. According to Goins (2001), some refer to mortar as a paste mixture made of proportional quantities of binder, aggregate, water and, in some cases, additives. On the other hand, Salvadori (1982); Holmes and Wingate (1997); Arıoglu and Acun (2006) refer to mortar as the compound that binds bricks or blocks together to give strength and stability to masonry. It is regarded as the fundamental and most used building material since ancient times, with different binder types for many different purposes, such as: masonry mortars in between stones and bricks, as internal or external wall finishing materials (plaster) or (render), as foundations, for flooring, as casings of water conduits or jointing compounds for terracotta pipes and decoration mortars, to name a few (Elsen, 2006). Mortar plays a vital role in the functioning and protection of a structure and its decorative elements. For this study, historic mortars refer to those produced before the end of the nineteenth century prior to the introduction of Portland cement into the construction industry. Such mortars are either soil or lime-based.

Literature indicates that historic mortars are made of lime as a binder, sand as aggregates and some additives such as shell pieces, charcoal particles, lime lumps and chips of wood or straw (Stefanidou & Papayianni, 2005; Sanjurjo-Sánchez *et al*., 2010). Mortar plays a vital role in ensuring that structures survive for years after their construction. The components of mortars are discussed in Section 2.2.1 to 2.2.4.

2.2.1 Binders

Binders are defined as materials with bonding properties which make them capable of holding mineral fragments in a coherent mass. Their properties are what actually determine the mortar properties (Palomo *et al*., 2014). There are different binder types present within the construction industry, which are organic and inorganic. The organic binders include bitumen, polymers, animal and plant glues, while the inorganic binders include lime, Portland cement and gypsum. However, in this study, the focus was only on lime and Portland cement. Both these binder types have different components, and their manufacturing processes differ. The differences in chemical components between binders are the basis for careful consideration when selecting repair mortars, not only for historic buildings but modern ones as well. According to Mitchell (2007); Gleize *et al*. (2009), the use of binders that are different from the original has negative repercussions on repaired surfaces, and therefore, more damage than benefit is usually the case.

Lime

According to Mitchell (2007), it is not precisely known when lime mortars were introduced to the construction industry. Nonetheless, present evidence shows that this kind of mortar was used for the construction of historic buildings during Roman times. Elsen (2006) reported that lime binder is estimated to have made its first appearance around the 6th millennium BC, while Aggelakopoulou *et al*. (2011) reported lime mortars to have been used since 7000 BC. Portland cement, however, was introduced to the construction industry during the nineteenth century (Sanjurjo-Sánchez *et al*., 2010). Mortars used before the introduction of Portland cement are therefore termed 'historical' or 'ancient' mortars (Arandigoyen & Alvarez, 2007).

The production process involves heating the raw materials, such as limestone, calcrete and dolomite. Lime appears in two types, namely: hydraulic and non-hydraulic. Palomo *et al*. (2014) list calcium oxide and calcium hydroxide (CaO, $Ca(OH)_2$), magnesium oxide and hydroxide (MgO, $Mg(OH)_2$), silica (SiO_2), alumina (Al_2O_3) and iron oxide (Fe_2O_3) to be the main constituents of construction limes. Lime mortars have the following properties that differ from the Portland cement mortars.

- *Low mechanical strengths* caused by the low attraction between calcite and quartz crystals (1:3 lime mortar of 28 days has only 0.2 - 0.5 MPa) (Zheng, 2011).
- *Slow setting* that makes it easy to work with (workability).
- *Low modulus of elasticity*.
- *High permeability to water and water vapour*.
- *Low resistance to freeze-thaw cycles* (Palomo *et al*., 2014).

Lime production includes the chemical reaction indicated in Equation (2.1).

$$\underset{\text{Calcium Carbonates}}{CaCO_{3\,(s)}} \xrightarrow{\text{Heat}} \underset{\text{Lime}}{CaO_{\,(s)}} + \underset{\text{Carbon Dioxide}}{CO_{2\,(g)}} \qquad (2.1)$$

Portland cement

Portland cement is the binder most often used in modern concrete production. In a study by Gosselin *et al*. (2008), it is stated that natural cement was first produced by end of the nineteenth century. It is manufactured by blending limestone or chalk with organic clays containing silica, lime, iron oxide, alumina, and magnesia. These are heated at high temperatures in a kiln to induce interaction between materials to form calcium silicates (see Figure 2.1) leading to production of clinker that is ground in a mill to form a powder known as Portland cement (Mbasha, 2015). As concluded by Lanas and Alvarez (2003); Mitchell (2007); Marini *et al*. (2018), the introduction of Ordinary Portland Cement (OPC) has led to replacement of lime as a binder for most repairs of historic buildings. The authors indicate that this has obvious repercussions, as the two materials are entirely different in terms of mechanical and physical properties. Therefore, attempting to merge the two is said to yield poor results that, in most cases, have aggravated the damages. To overcome this, Marini *et al*. (2018) propose the use of a newly-introduced technique which incorporates use of improved natural hydraulic lime (NHL).
Portland cement mortars have the following properties:

- *Stability in volume* - With the hydration process, a mortar undergoes shrinkage due to a volume decrease of the hydrated cement phases.
- *Good durability* depending on compaction and binder/water ratio.
- *High (10 MPa and above) mechanical* strengths resulting from a high proportion of Calcium-Silicate-Hydrate (C-S-H) gel.
- *Heat of hydration* - The fast development of heat in the system can lead to quick evaporation of water which results in the formation of cracks in the mortar (Palomo *et al*., 2014)

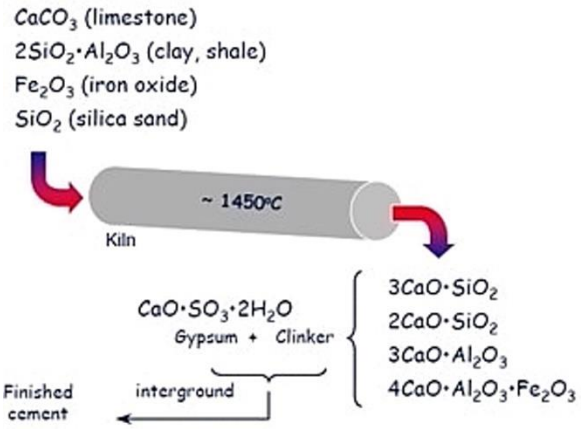


Figure 2.1 Cement compound formation (Mbasha, 2015)

2.2.2 Aggregates

Aggregates contribute significantly to the behaviour of mortar (Isebaert *et al*., 2014) and come in two forms; coarse and fine, depending on the strength requirement for specific mix design. For mortar production, fine sand has been used over the years, with particle size ranging between 75 (or 63 for some) µm and 2 mm. Sand is usually named after its origin or nature (Holmes & Wingate, 1997). Traditionally, the choice of the sand to be used in mortar production was determined by what was available in the neighbourhood of the building site (Ngoma, 2009).

2.2.3 Water

Water is added to solid components of the mortar to provide the plastic properties for mortar and to activate the hydration process of hydraulic components. Water, clean and free from impurities, where possible, is used during mortar production to form a paste that is easy to work with. Careful consideration needs to be taken when selecting the amount of water to be added to the solid components, as too much water causes segregation (separation of components) while too little water reduces mortar workability.

2.2.4 Additives

In addition to the binder and aggregates, there is both organic and inorganic material added to the mortar to modify its properties (plastic and hardened) such as workability, strength, hardening rate and durability among others. The conventionally used materials include marine shells, charcoal particles and straw. On the other hand, modern materials used by the construction industry include accelerators, plasticizers, air-entraining agents and fibrous materials.

2.2.5 Binder identification

The identification of the original binder type through its properties plays a vital role in selecting the most appropriate repair binder. For the purpose of matching the properties of the original, research around the identification of the type of binder used for historic structures has been conducted extensively over the years, mostly in Europe. The process of identifying the type of binder is depicted in Figure 2.2.

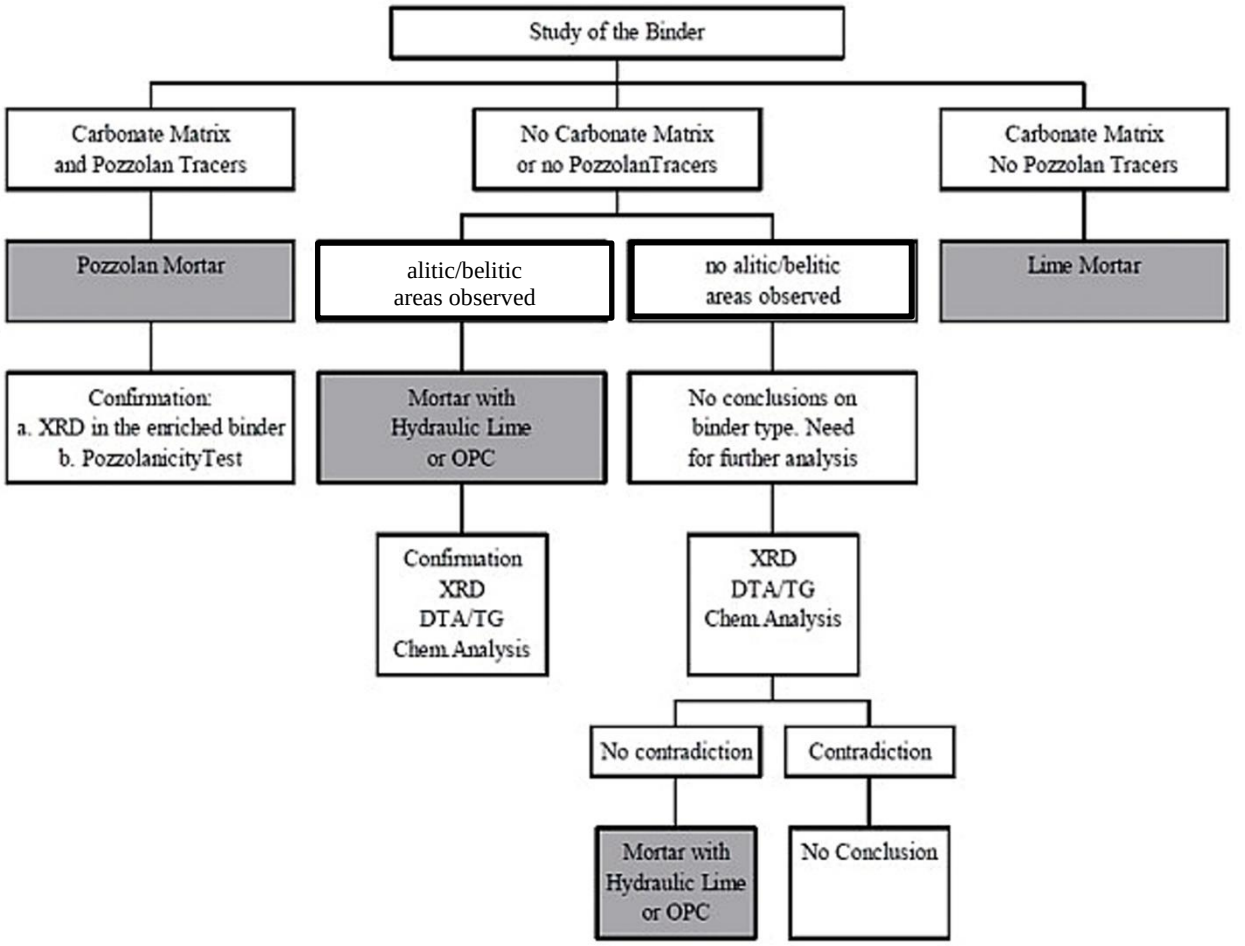


Figure 2.2 Flow chart for binder identification (Van Balen *et al.*, 1999b; Palomo *et al.*, 2014)

2.3 Mortar durability

Durability is defined as the measure of a structure's performance over a specified period (Lawrence *et al*., 2004). There are many factors like human neglect, ignorance, and environmental conditions that affect the durability of building materials on historic buildings (Feilden, 2003). Studies indicate that deterioration of mortar occurs through different chemical, physical, mechanical and biological processes, which often coincide. However, the most significant factors affecting the durability of mortars are considered external, which are environmentally based (Lawrence *et al*., 2004). Due to these factors, the preservation of monuments has become a significant challenge as durability is always compromised. It is therefore advisable that a restoration team takes the external factors into consideration during the design stage of historic repair mortars. The deterioration factors include, but are not limited to those outlined in Section 1.1.

2.3.1 Atmospheric moisture

The presence of water in the atmosphere is the source of most mechanisms that damage mortars (Lawrence *et al*., 2004; Price, 2010). With reference to Camuffo (1995), there are three main ways by which moisture penetrates the mortar during dry seasons, and they are: condensation of water vapour, capillary rise and rainfall. Furthermore, Amoroso and Fassina (1983) elaborate that out of the three forms of moisture; rainfall in particular carries dissolved acid gases to the surface of masonry (forming acid rain), corroding the mortar and possibly the entire masonry. Water harms the mortar as it comes into contact with the hardened surface and spreads through the porous system of the material, dissolving the calcium enriched hydrated phases in the mortar materials.

2.3.2 Soluble salts

The presence of salt constituents in the pores of mortar causes significant damage by weakening the tensile strength leading to the transformation of mortars into a powder. According to a study by Ngoma (2009), these salts are either blown by the wind from the sea (sea-spray), are carried into masonry by rising damp (soil water) or carried through air pollution. Ngoma (2009) further sheds some light on the ways in which salts cause harm to the building materials. The factors he highlighted include crystallisation from solution and hydration.

2.3.3 Atmospheric pollution

As mentioned by Ngoma (2009), there are pollutants that dissolve in water to produce acidic solutions, namely sulphur oxides and nitrogen oxides. These chemicals are said to be occurring naturally; however, due to urbanisation, there is an alarming increase in their emission into the atmosphere. The increase is caused by human activities such as combustion of fossil fuels, for example, petroleum, crude oil and coal, which lead to massive production of the acidic solutions that attack mortars. Additionally, Amoroso and Fassina (1983) report that heritage buildings in Europe are susceptible to air pollutants such as carbon dioxide, nitrogen oxides, ozone, sulphur oxides and particulate matter.

2.3.4 Biological colonisation

According to Ngoma (2009), microorganisms such as algae, lichens and fungi generally cause minor damage to masonry when compared to other factors such as air pollution or soluble salts. In addition to microorganisms, plants tend to grow in between masonry joints as depicted in Figure 2.3. This could lead to mortar chipping off as the plants are de-rooted, thereby affecting the structure's integrity.

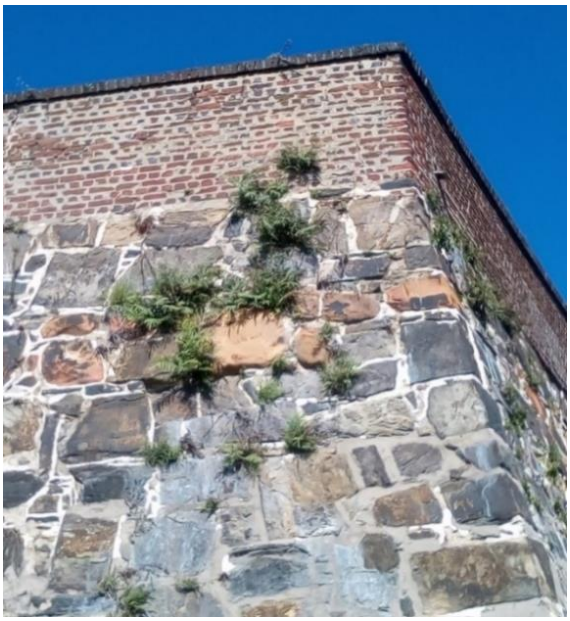


Figure 2.3 Biological colonisation: Castle of Good Hope, South Africa

Despite the factors discussed above, historic buildings' durability is dependent on their regular maintenance. It is of obvious benefit that defects are prevented from occurring beforehand. However, regular maintenance with incompatible materials is equally discouraged.

2.4 Characterisation of heritage cementing materials

Characterisation of heritage mortars is a process which involves investigating the major components such as aggregates, binder and hydraulic components. It could generally be described as a form of reverse engineering process, with the aim of determining what materials were used during construction instead of deciding what materials to use (mix design). Historic mortars characterisation is a well-researched topic and involves analysis and investigation on properties of hardened mortar collected from existing historic buildings. It incorporates detailed experimental procedures for identification of physical, chemical, mineralogical and mechanical properties of the mortars. It could be carried out for the purpose of either research, documentation or compatibility assessment prior to conservation and restoration of historic structures (Hauková *et al*., 2013).

As pointed by Groot *et al*. (2004); Benedetti and Pelà (2012), the experimental characterisation of the mechanical properties of mortars in existing masonry constructions is complex as decay on mortar surfaces can lead to false results, and the amount of material obtainable is always inadequate. The literature proposes the use of unexposed samples for analysis to overcome the possible errors. According to past studies, mechanical characterisation requires the use of a large quantity of samples to carry out standard testing on standard test specimens. Hence, not much research has been carried out in this context. In most cases, mechanical characterisation is a challenge, as it is too risky for heritage authorities to allow extraction of large quantities of mortar from the joints of existing brickwork. This would cause a disturbance to specimens and the structure itself; and therefore, the mechanical properties are hard to analyse using standard sample sizes.

The reverse engineering process provides an answer to the question: What cementing material (type of binder, aggregates and additives) was originally used for the construction of the heritage building? The process is presented in Figure 2.4. Additionally, a study by Hauková *et al*. (2013) highlights the importance of characterising not only historic building materials but also the modern materials prior to restoration.

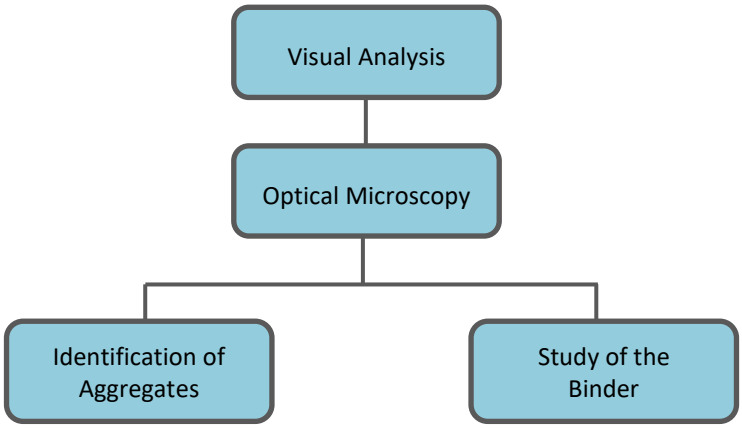


Figure 2.4 Mortar analysis procedure (Van Balen *et al*., 1999; Palomo *et al*., 2014)

There are several properties that are investigated through material characterisation, namely aesthetic, physical, chemical, mineralogical and mechanical properties. These properties play different roles in mortar durability for the sustainability of structures for future years. In order to perform the experimental procedures on materials, the representative samples are collected from existing buildings using safe techniques.

2.4.1 Sampling methodology

Sampling is the acquisition of materials to be investigated from existing masonries of historic buildings. The permission to alter historic buildings is often controlled by the heritage authorities, and no individual is allowed to carry out sampling without consent from heritage authorities. Due to the cultural and historical importance of heritage structures, the sample quantity must be kept to a minimum at all times, as the aim is to always inflict the least possible damage. These limitations make an appropriate sampling of heritage mortars very difficult (Hughes & Callebaut, 1999). Several authors that investigated the characteristics of historic mortars emphasise the need for proper sampling, as this phase can influence the results obtained. The data recording form used for sampling of historic mortars in East Africa is presented in Figure 2.5. Ngoma (2009) further underlines the importance of gathering all relevant information during sampling which would be useful during characterisation.

Sampled structure: **Location:** **Age:** **Photo:** ☐ **Taken** ☐ **Not taken**	**Sample no:** **Sampled by:** **Date:** **Reason for sampling:**
Sampling method: ☐ Hammer & chisel ☐ Drill core ☐ Others (explanation): **Sampling location:** ☐ Wall ☐ Foundation ☐ Roof ☐ Exterior ☐ Interior ☐ At depth:….mm ☐ Others (explanation): **Function of sample:** ☐ Render ☐ Plaster ☐ Joint bedding **Sample condition:** ☐ Hard ☐ Soft ☐ Soft and friable **Carbonation condition:** ☐ Carbonated ☐ Partial ☐ Not yet **Inclusions:** **Sample colour:** **Environmental conditions:** **Water Table:** **Condition of associated building materials:**	

Figure 2.5 Sampling data form (Ngoma, 2009)

In addition to proper sampling which involves the use of a sampling data form to gather all necessary information about the material, the next most crucial aspect linked with sampling is ensuring that the correct mortar quantity is obtained. Like Hughes and Callebaut (1999), the current study used Table 2.1 as a reference for the amount of material to be collected in order to successfully complete the selected analytical techniques.

Table 2.1 Analytical technique versus sample quantity (Hughes & Callebaut, 1999)

Information required	Possible Analytical methods	Minimum QUANTITY and QUALITY of sample
Appearance Qualitative structure/texture. Spatial relations of components. Binder/aggregate ratios	Optical examination Thin sections: optical microscopy Acid dissolution or thin section	In-situ or 10g (intact) ~10g (intact) ~100g (intact, crumbling or powder) depending on aggregate size.
Aggregate grading and colour.	Acid Dissolution and Sieving	~100g
Mineralogy/composition of aggregate and additives.	Thin sections (microscopy) XRD DTA TGA Scanning Electron Microscopy (SEM) IR Spectroscopy	~10g (intact, crumbling or powder.) ~2-5g (powder) ~1g (powder) ~1g (powder) ~2-5g (intact) ~1g (powder)
Mineralogy of carbonate binder.	Optical Microscopy XRD Scanning Electron Microscopy DTA TGA IR Spectroscopy	~10g (intact) ~2-5g (powder) ~2-5g (intact) ~1g (powder) ~1g (powder) ~1g (powder)
Microstructure of binder	Scanning Electron Microscopy (SEM)	~2-5g (intact)
Composition	EDX and Electron Probe analysis	5g (intact)
Organic materials	IR Spectroscopy	~1g (powder)
Major and Trace Elemental concentrations	Atomic Absorption Spectrophotometry ICP X-ray Fluorescence EDX and Electron Probe analysis	5 g (powder) 1g (powder) 10g (powder) 5g (intact)
Anions/cations concentration Salt content.	Ion Chromatography	1g (powder)
Porosity	Mercury intrusion porosimetry Density and water saturation method	10g (intact) 10g (intact)
Free/uncombined moisture content	Moisture content	10g (powder, or intact samples)
Permeability	Surface probe permeability Laboratory permeability	100g intact or in-situ Core sample intact lump
Elasticity	Indirect tensile and bending tests	>100g intact Core samples or intact joint sample.
Bond strength, tensile bending strength of mortar/masonry join.		>100g Including mortar – masonry join. Core possible.
Durability of material	Hardness/ Schmidt hammer test Freeze thaw and salt crystallisation	In-situ or intact 100g pieces. Core or joint sample Large >200g intact samples- chisel, saw or cores.
Compressive strength		100g core samples or intact joint sample.

2.4.2 Aesthetic analysis

The analysis of the colour and texture of the original material is conducted to avoid a mismatch and to preserve the aesthetics represented by a specific colour or texture on historic buildings. In most decorative mortars, such as historical sculptures and decorative plastering/rendering, colour plays a significant role in authenticity; hence, the use of a colour similar to the original material is encouraged. One needs to bear in mind the possibility of original colour change and general material properties over time that is caused by ageing and weathering. To get a sense of the original, the analysis is usually made on the unexposed surfaces (inner part of the mortar). This form of characterisation is performed by the use of instruments such as colorimeters, spectrophotometers and by the human eye with use of a colour scale to define the colour of the mortars. In addition, some researchers such as Schueremans *et al*. (2011) use a stereo microscope to obtain detailed results on a material's aesthetic properties.

Several authors, such as Schueremans *et al*. (2011); Bertolini *et al*. (2013); Drdácky´ *et al*. (2013); Gulzar *et al*. (2013); Lopez-Arce *et al*. (2016); Apostolopoulou *et al*. (2017) present procedures to carry out both visual and microscopic investigations on colours and textures of historic mortars, both in Europe and Asia. An approximate 30% of studied literature conducted visual investigations on colour and cohesion, and went further to analyse the mortars using stereo microscopes. Figure 2.6 demonstrates the typical results of a detailed investigation using a stereo microscope at the laboratory.
Key:
A – 2 mm
B – 1 mm
C – 0.5 mm

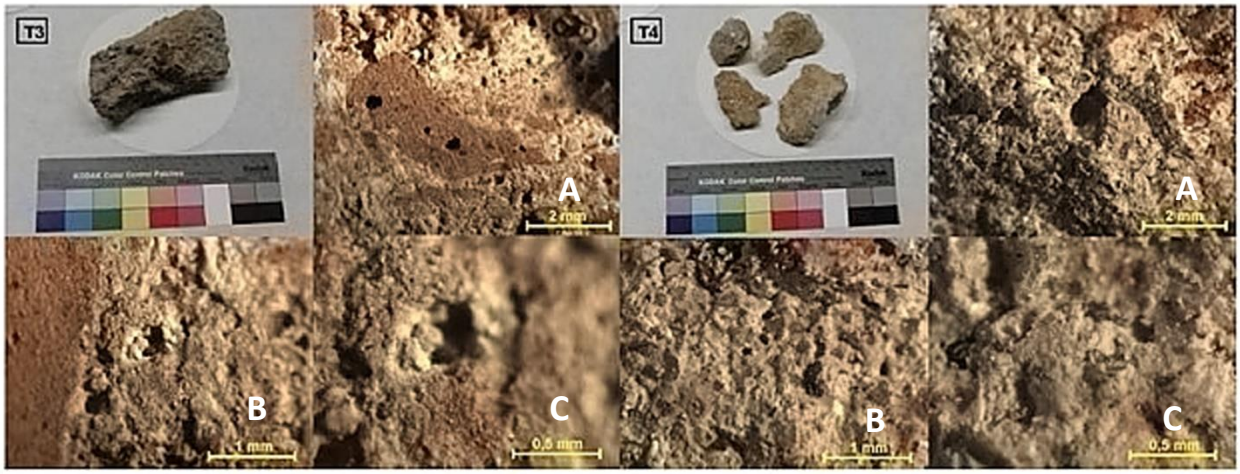


Figure 2.6 Stereo microscopic images: Original (T3) & repair(T4) brick-laying mortars (Schueremans *et al*., 2011)

2.4.3 Physical analysis

The physical analysis stage investigates the properties of the mortars such as open and total porosity, apparent density, frost resistance, water retention and particle size distribution. According to Hauková *et al*. (2013), there are thus far no European standards for carrying out porosimetry analysis of historic mortars. As for determining the grain distribution, the mortar is passed through a nest of sieves before and after being exposed to hydrochloric acid (HCl), and later the cumulative percentage of materials passing the sieves is calculated. A similar procedure could be carried out on dry mortar (without digestion in HCl). The physical properties of cementing materials assist in determining the aggregate type (coarse,

medium, fine, well-graded, or poorly graded sand) that was used during construction, which plays a vital role in restoration. Apostolopoulou *et al*. (2017) highlight the need to consider the effect of weathering and possible alterations on materials throughout the years which could have an impact on the exact gradation curves of the original materials. They, however, conclude that the restoration aggregates gradation curves must be as close to the original as possible, regardless of the possible weathering of original materials.

Authors such as Apostolopoulou *et al*. (2017); Bertolini *et al*. (2013); Corinaldesi (2012); Drdácky´ *et al*. (2013); Gleize *et al*. (2009); Gulzar *et al.* (2013); Labiadh *et al*. (2009); Lopez-Arce *et al*. (2016); Ontiveros-Ortega *et al*. (2016); Özkaya and Böke (2009); Schueremans *et al.* (2011) use different methods to analyse physical properties of heritage mortars. From the case studies reviewed, 60% of the researchers used scanning electron microscopy (SEM), only 10% investigated the thermal conductivity of the samples, 40% checked open and total porosity of the mortars and 40% performed granulometry or particle size distribution.

Thermal conductivity

It refers to the ability of the material to allow heat to pass through (Apostolopoulou *et al*., 2017). The characterisation technique used by Apostolopoulou *et al*. (2017) involved exposing the mortar to different heat ranges and recording the mass lost throughout heat increase for each mortar. The results obtained from this test are shown in Table 2.2.

Table 2.2 Thermal conductivity analysis (Apostolopoulou *et al*., 2017)

Sample	Mass loss for each temperature range (%)				$CO_2/H_2O_{ch.b}$	$CaCO_3$
	<120 $H_2O_{ph.b}$*	120-200 H_2O_{salts}	200-600 $H_2O_{ch.b.}$*	>600 CO_2		
MK1	1,06	1,83	2,14	39,30	18,36	89,21
$MK1_{binder}$	0,85	2,02	2,33	39,22	16,83	89,03
MK2	1,21	2,34	2,03	31,43	15,48	71,35
MK2b	0,42	0,50	3,6	28,39	9,28	64,44
MK4	0,50	0,52	5,12**	39,25	7,67**	89,10
$MK4_{binder}$	0,52	0,45	3,82	40,00	10,47	90,80
MK5a	0,95	2,05	7,36**	27,50	3,74**	62,42
MK5b	0,68	0,90	6,05**	24,55	4,06**	55,73
MK6	0,38	0,54	2,76	20,35	7,37	46,19
MK7	0,50	0,57	5,93**	29,87	5,04**	67,80
$MK7_{binder}$	1,09	0,20	3,14	36,80	11,71	83,54
MK8	0,69	0,96	7,7**	26,48	3,44**	60,11

* $H_2O_{ph.b}$: physically bound water, $H_2O_{ch.b.}$: chemically bound water.
** Samples with the presence of organic matter – the exact amount attributed to chemically bound water cannot be calculated and therefore the reverse hydraulicity index calculated is not indicative.

Open and total porosity

The porosity of historical materials is commonly studied employing mercury porosimetry. The method involves the use of high pressure to force mercury into the pores of the sample and measure the volume of mercury required to fill the pores. The results help determine the pore-radius distribution and the porosity of the material. According to Ngoma (2009), mercury porosimetry is not an ideal method to use on lime-based soft and friable mortars because of the high pressure used, which could damage the texture of the mortar and lead to incorrect estimations of porosity. Therefore, Ngoma (2009) suggests an

alternative method, which saturates a sample with water, and then measuring the weight of a sample before and after full saturation in water. The procedure for this hydrostatic porosity measurement involves oven drying the sample at a maximum temperature of 60 ^{0}C (temperature higher than this could cause thermal damage to a test sample) to obtain a constant mass (M_1), and then immersing the dried sample in tap water for 24 hours. The saturated mass of the sample is then taken (M_2). The third mass (M_3) is finally taken when the previously immersed sample is wiped (to remove surface moisture) and suspended by a wire hook from the plate of the balance into a container of water (hydrostatic weighing). Porosity was calculated by using Equations 2.2 to 2.4 (Ngoma, 2009).

$$\rho_r = \frac{M_1}{M_1 - M_2} \text{ x } 100 \tag{2.2}$$

$$\rho_a = \frac{M_1}{M_3 - M_2} \text{ x } 100 \tag{2.3}$$

$$\varepsilon = \left[1 - \frac{\rho_a}{\rho_r} \right] x\ 100 \tag{2.4}$$

Where:
M_1 - Constant dry mass
M_2 - Saturated mass
M_3 - Hydrostatic mass
ρ_a - Apparent density
ρ_r - Real density
ε - Porosity

In another study, Apostolopoulou *et al*. (2017) determined the mortar porosity using a mercury intrusion porosimetry technique. Ngoma (2009) indicated that this method is inappropriate to use on lime-based, soft and friable mortars as high pressure is used and could damage the texture of the mortar and lead to incorrect estimations of porosity. Therefore, hydrostatic porosity measurement is concluded to be the best option as the mortar texture is not damaged. The results for mercury intrusion porosimetry show the bulk density (g/cm^3), total porosity (%), average pore radius (lm) and specific surface area (m^2/g) as stated in a paper by Apostolopoulou *et al*. (2017).

Scanning electron microscopy (SEM)

Several researchers including Drdácky´ *et al*. (2013); Gulzar *et al*. (2013); Lopez-Arce *et al*. (2016) applied a scanning electron microscopy (SEM) together with energy dispersive spectroscopy (EDS) analysis in order to distinguish between different phases of the binder on image contrast. These techniques also help determine the weathering products caused by decay processes and to identify the type of salt efflorescence and sub-efflorescence on mortars. The SEM-EDS results from most studies showed a general presence of elements such as calcium (Ca), magnesium (Mg), potassium (K), carbon (C), oxygen (O), silicon (Si), sodium (Na), chlorine (Cl) and aluminium (Al) on historic mortars studied. Figure 2.7 demonstrates the typical results of SEM-EDS.

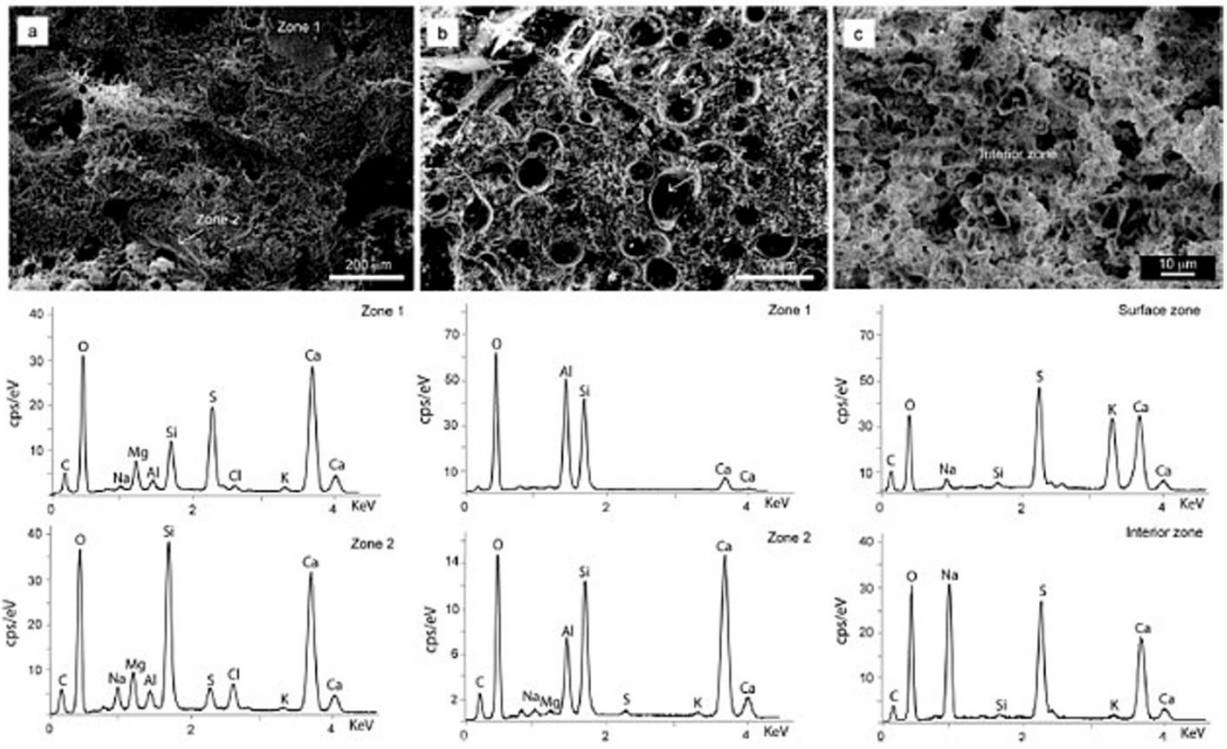


Figure 2.7 SEM-EDS analyses (Lopez-Arce *et al*., 2016)

Physical ageing tests

These incorporate the properties of mortars under extreme conditions and are evaluated using RILEM recommendations outlined in a paper by Papayianni *et al*. (2013). The tests include wetting-drying cycles, cycles of salt crystallisation (10% w/w solutions of Na_2SO_4 and NaCl), as well as freeze-thaw cycles. Papayianni *et al*. (2013) suggest that the results from these tests could be used as compatibility criteria in terms of functional adjustment, stability and resistance to extreme weathering conditions of repair mortars. The different test procedures are as follows:

Wetting-drying cycles: Samples are immersed in water after drying at 100 ^{0}C for 24 hours for 7 days. Their wet weight is measured every day, after that, they are dried at 100 ^{0}C for 24 hours, making a complete wetting-drying cycle.

Salt crystallisation cycles (sulfates and chlorides): Samples are dried at 100 ^{0}C for 20 hours and kept indoors for 2 hours and weighed. They are then immersed in a solution of sodium sulfate (Na_2SO_4) or sodium chloride (NaCl) 10% w/w for 2 hours. The procedure continues by drying the samples at 100 ^{0}C for 20 hours and keeping them indoors at a temperature of ±20 ^{0}C for 2 hours (1 cycle). They are weighed after every cycle.

Freeze-thaw cycles: An article by Křivánková *et al*. (2019) indicates the vulnerability of lime mortars to freeze-thaw cycles. This is due to lime having high porosity and low mechanical resistance. Therefore, the ability of historic mortars to resist frost attack plays a vital role in its durability, particularly in cold environments. The test is conducted by exposing the material to freezing and thawing and assessing its resistance from the formation of cracks or change in weight.

Procedure for the freeze-thaw test: Samples are dried at 100 ^{0}C for 20 hours and kept indoors for 2 hours and weighed. Afterwards, they are put in water at a temperature of -10 ^{0}C for 4 hours, dried at 100 ^{0}C for 19 hours and kept indoors for 1 hour (1 cycle). They are weighed after every cycle (Papayianni *et al*., 2013).

The results obtainable from the physical ageing tests are presented in Figure 2.8, showing the cycles for each test.

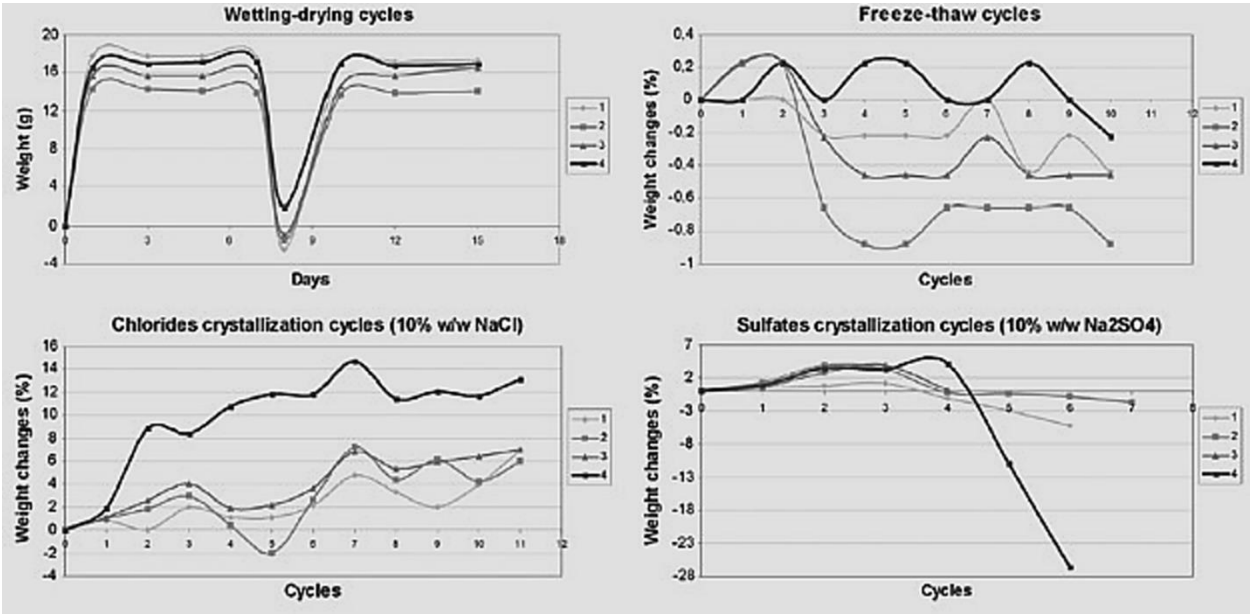


Figure 2.8 Ageing tests of repair mortars (Papayianni *et al*., 2013)

2.4.4 Mineralogical and petrographic analysis

As outlined by Cizer *et al*. (2010); Elsen *et al*. (2011), it is advisable to carry out a mineralogical analysis which is complemented by chemical analysis as the latter alone does not provide all necessary information on original material used (mineral phases, type of aggregate and binder). The minerals such as calcium hydroxide ($Ca(OH)_2$), calcite ($CaCO_3$), quartz (SiO_2), gypsum ($CaSO_4 2H_2O$) and albite ($NaAlSi_3O_8$), as well as the salts namely sulfates, nitrates and nitrites, play an essential role in the way a mortar behaves and reacts with the surrounding environment. The minerals are identified using a technique referred to as x-ray diffraction (XRD) analysis. The method clearly shows the crystalline phases that took place in the binder by determining the mineralogy of clays in the soil.

TGA and FTIR

To assess the degree of hydration and present quantities of calcium hydroxide/slaked lime ($Ca(OH)_2$) and calcium carbonate/limestone ($CaCO_3$), thermogravimetric analysis (TGA) and Fourier transform infrared spectrometry (FTIR) are among other methods used. All these tests provide details of the minerals present and their quantities in a mortar sample, while FTIR gives the ratio between the $CaCO_3$ and SiO_2 content. These two tests require less than 2 mg of samples to conduct. The process of mineralogical and petrographic characterisation of historic mortars is illustrated in Figure 2.9.

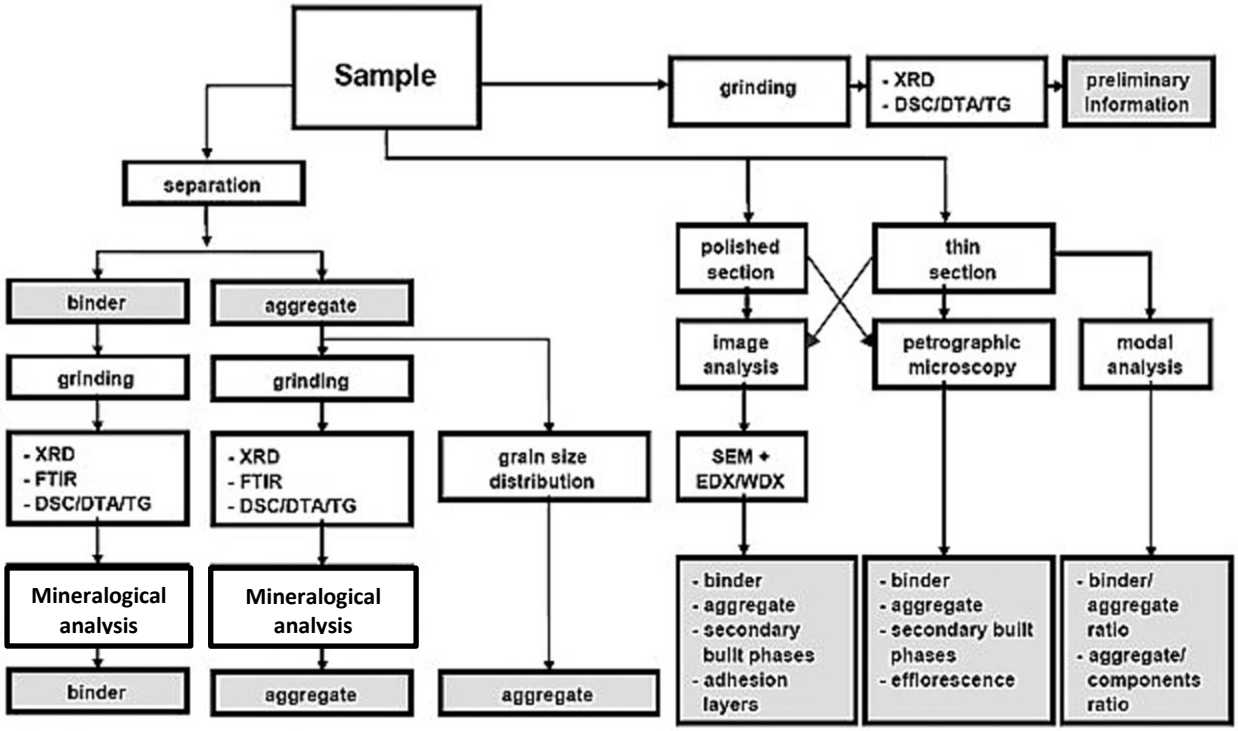


Figure 2.9 Flow chart for mineralogical characterisation (Middendorf *et al*., 2005a)

From the literature reviewed, an approximate 90% of the researchers used XRD to investigate the mineralogical properties of historic mortars. In a study conducted by Schueremans *et al*. (2011), XRD was used and complemented with TGA and DTG. The results of their research indicate calcite and quartz to be the dominant minerals in the mortars with some traces of portlandite.

X-ray diffraction (XRD)

XRD and EDX are the two analytical methods which use an x-ray to determine the crystalline structure of nanomaterials and element contents in materials respectively. The techniques involve placement of the powdered sample in a container, then the use of x-rays of fixed wavelength to reflect the characteristics of the mineralogy of the sample (Ngoma, 2009). The status of elements is marked using the following signs to denote their presence or absence:

+++ dominantly present
++ present
+ traces
? possibly present
- not detected

In addition to the components observed in mortars studied above, other researchers observed significant amounts of ettringite, gypsum, portlandite, calcite and quartz as shown in Figure 2.10.

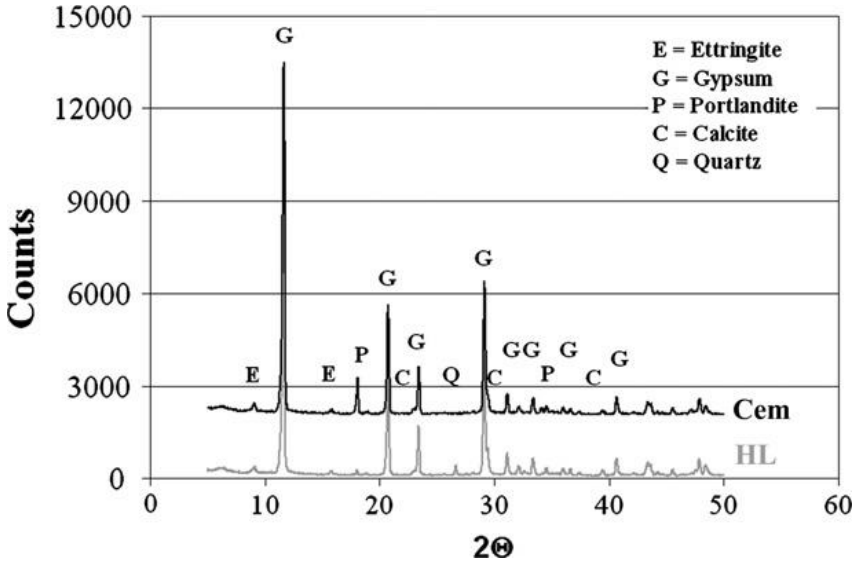


Figure 2.10 XRD analysis (Corinaldesi, 2012)

Thermogravimetric analysis

There are generally three fundamental analyses used to identify binders, aggregates and admixtures as well as the thermal conductivity namely: thermogravimetry (TG), differential thermal analysis (DTA) and differential scanning calorimetry (DSC). TG involves the measuring of weight loss in a sample as it is heated. The gradual increase in temperature that results in weight loss denotes specific physical decompositions in the materials. For instance, the weight loss of approximately 26.5 wt.-% demonstrates the presence of gypsum in a mortar sample. This is different for various compounds.

On the other hand, DTA deals with exposing the sample to heat for the loss of chemically bound components in a mortar sample. These could be water from gypsum or carbon dioxide from calcite and dolomite. DTA is further used to identify endothermic or exothermic transitions of particular minerals. Figure 2.11 shows an example of the results obtained from a TGA/DTG analysis.

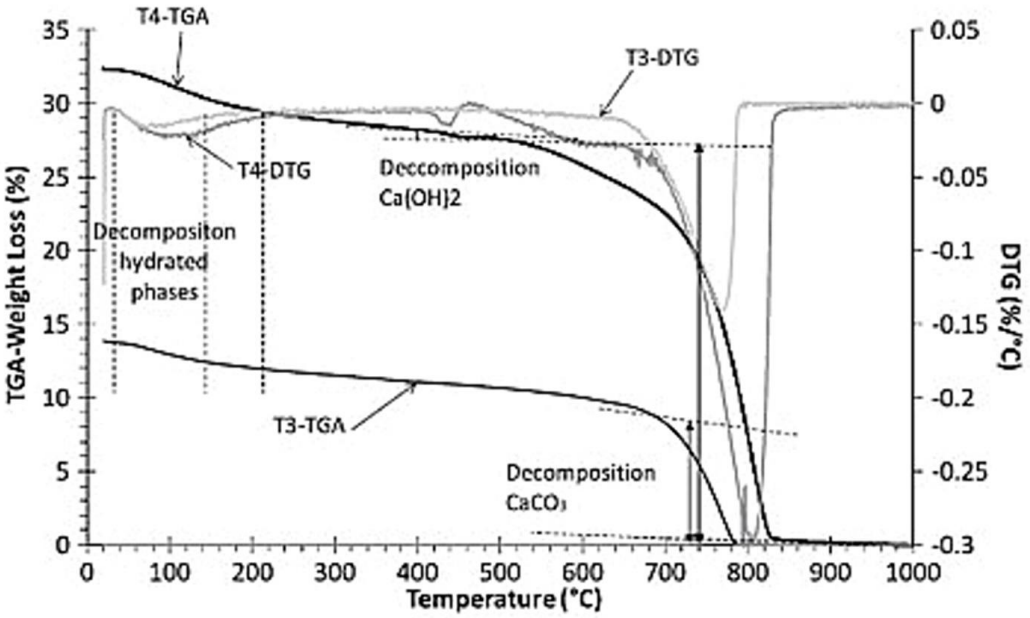


Figure 2.11 TGA/DTG (Schueremans *et al.*, 2011)

Ion chromatography (IC)

Lopez-Arce *et al*. (2016) performed IC to identify and quantify the soluble anions and cations such as chlorides (Cl^-) and sodium (Na^+) in parts per million (ppm) present in the raw anhydrous mortars as indicated in Table 2.3. The procedure to carry out this test is based on the collection of approximately 0.1 g of sample and dissolution in 10 ml of de-ionised or distilled water for 5 minutes. The solution with the solid residue is left to settle down with a minimum rest period of 24 hours at room temperature. The chromatography test strips are used to determine the presence and varying degree of ions through colour change. IC is considered accurate, as well as a reliable method for identifying ions in samples (Ozga, 2009). The other method uses the colorimetric kit. This method works in the same way as the test strips.

Table 2.3 Ion Chromatography analysis (Lopez-Arce *et al.*, 2016)

Soluble salts		Anions (ppm)				Cations (ppm)					Wt (%)
Samples		Cl^-	SO_4^{2-}	NO_3^-	PO_4^{3-}	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}	
Anhydrous mortar	Lit	5	23	0	0	22	0.00	3	1	129	2
	Art	5	0	0	0	16	0.00	3	1	534	5
	Alt	6	0	0	0	22	0.00	3	3	165	2

Soluble salt analysis

The presence of high amounts of salts (mainly chlorides and sulfates) in soils used for repair mortars can result in premature deterioration of the building material as a result of salt crystallisation (Zinn, 2005). The soluble salts in the mortar samples are tested using a method that involves the use of ion test strips to test for chlorides and sulfates. Another method involves a titration test to determine the presence of chloride (Cl^-) ions. The sample (10 g) is soaked for three hours in 10 ml of deionised water to bring any soluble salts into solution. The test strips for chloride and sulfate ions are then immersed in a solution and

observed for colour changes in the indicators on the strips. The test strip indicators change colour, denoting the presence of specific ions with a varying range depending on the concentration in ppm (Zinn, 2005). The test only provides the range in concentration of significant salts in a sample. It does not give the exact quantities of the salts.

Additional qualitative salt analysis

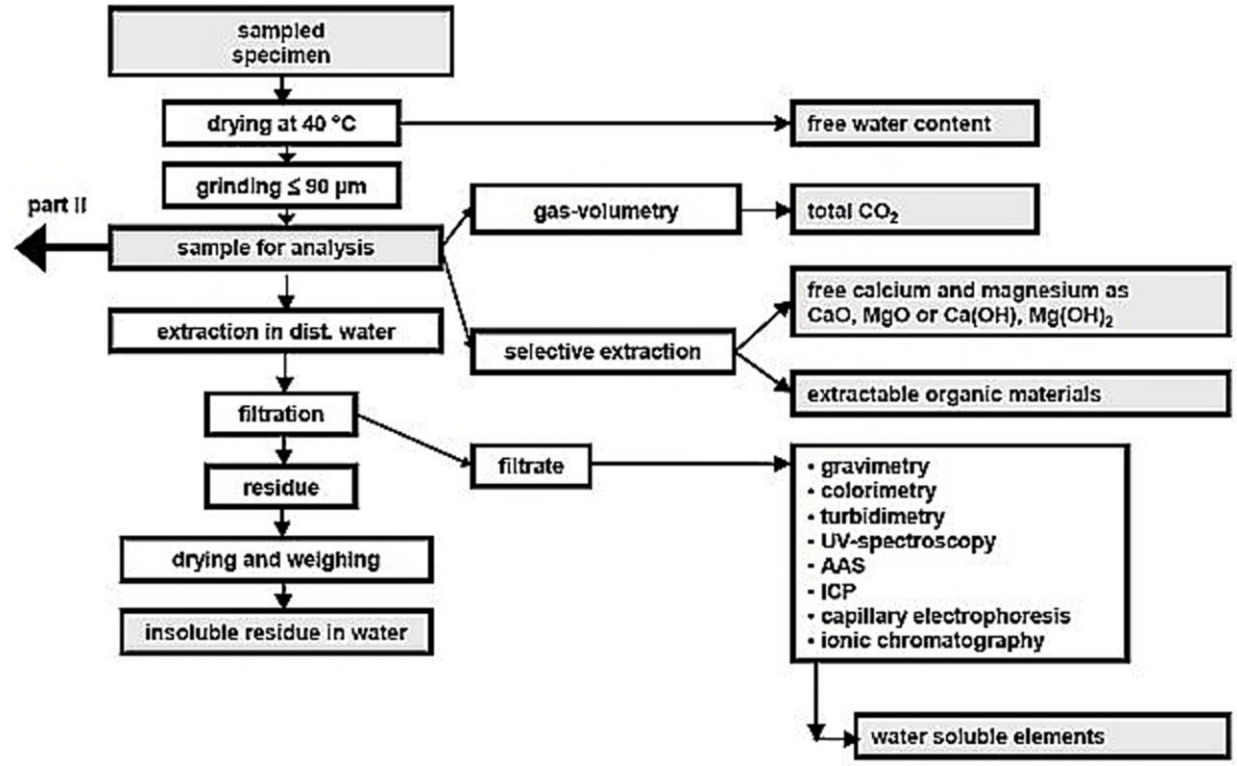
The process only identifies the salts present in masonry mortars and does not provide details of the quantities. According to Teutonico (1988) and Borrelli (1999), the analysis helps understand the deterioration pattern of the masonry which depends mainly on the mortar used. It includes the analysis of sulphates, chlorides, nitrites, nitrates and carbonates by use of chemicals such as diluted hydrochloric acid, nitric acid, acetic acid, sulfamic acid, barium chloride, silver nitrate and zinc powder among others.

Crystallisation cycles

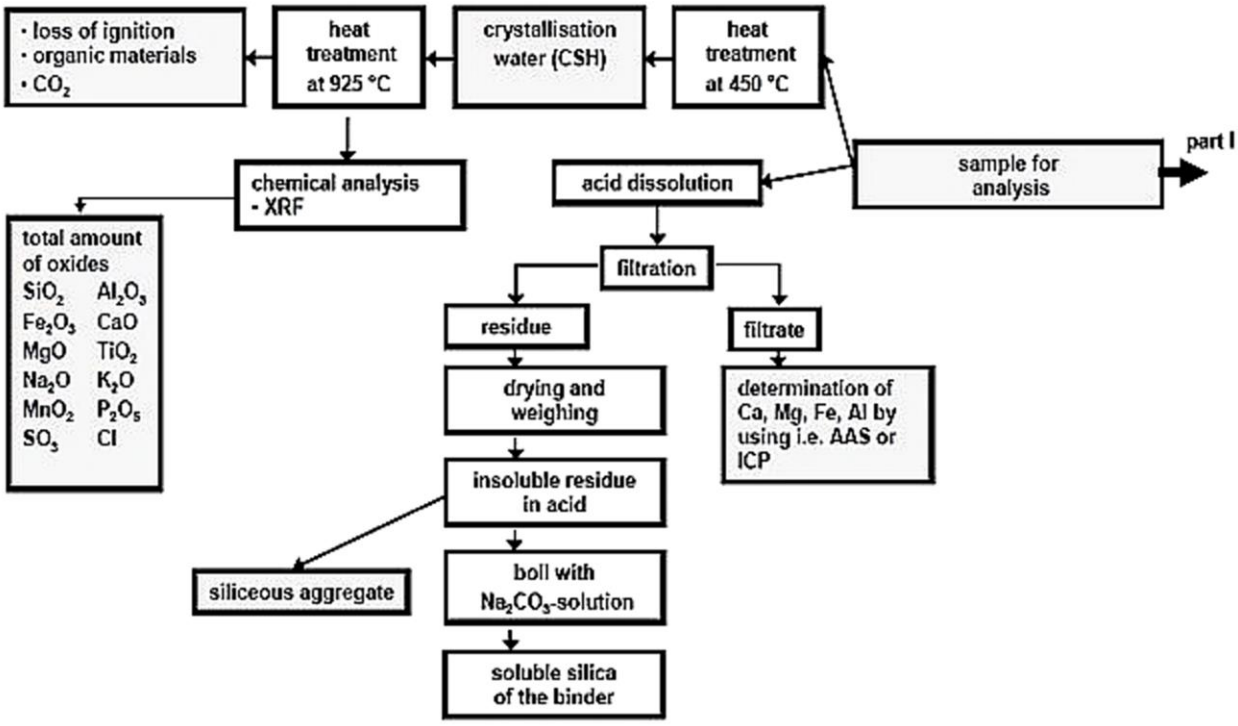
The analysis is conducted to assess the way salts are transported inside the mortar. The presence of salt crystallisation can affect the aesthetic properties of the structure. The test is conducted on 5 cm^3 samples using 10% sodium sulphate solution. The sample is dried at 110 degrees Celsius for 24 hours and left to cool, then immersed in a 10% sodium sulphate solution. The procedure is repeated at least three times, or until there are visible changes in the sample (Teutonico, 1988; Borrelli, 1999).

2.4.5 Chemical analysis

This identifies the chemical composition of historic mortars. It is through the chemical characterisation where the type of binder used and its quantity and the binder-to-aggregate ratio, as well as hydraulicity of materials, are identified. Authors such as Schueremans *et al*. (2011) determine the chemical composition of mortar samples using different methods, such as wet chemical analysis and x-ray fluorescence (XRF). Schueremans *et al*. (2011) demonstrate that using XRF gives the chemical composition of the mortar constituents only. Gulzar *et al*. (2013); Hormes *et al*. (2016) and Ontiveros-Ortega *et al*. (2016) also conduct XRF analysis in determining the chemical properties of historic building mortars in Pakistan, Germany and Spain respectively. The procedures to determine the water (Part II) and acid (Part I) soluble chemical elements in historic mortars are shown in Figures 2.12 a) and b). Part I of the procedure is used to analyse the elements which are soluble in acid using the acid dissolution method together with the atomic absorption spectroscopy (AAS) and inductively coupled plasma (ICP). The second part of the analysis investigates the presence of water-soluble salts using gravimetry, colorimetry, turbidimetry, ICP, AAS and ion chromatography, among other methods.



(a)



(b)

Figure 2.12 a) & b) Chemical characterisation (Middendorf *et al.*, 2005b)

Wet chemical analysis

Some researchers (Özkaya & Böke, 2009; Schueremans *et al*., 2011) propose the use of wet chemical analysis, whereby the binder is dissolved in a solution of hydrochloric acid. This analytical procedure provides details on the chemical composition of the acid-soluble binder and the insoluble aggregate, including their relative proportions. It is, however, to be noted that no international standards exist in relation to the type of acid and concentration to use while carrying out wet chemical analysis (Middendorf *et al*., 2005). As a result, the acid type and concentration differ per country, researcher and laboratory.

Titration test

Cayme and Asor (2017) carried out a simple and inexpensive complexometric titration test using ethylenediaminetetraacetic acid (EDTA) and potassium permanganate ($KMnO_4$) to quantify the amount of calcium in a mortar sample. According to Cayme and Asor (2017), it is essential to quantify the amount of calcium, as it is a primary chemical element found in lime-based mortars and is responsible for binding the stone and sand aggregates in a mortar. Middendorf *et al*. (2005b) recommend the use of the titration method to quantify the oxides of iron, aluminium, calcium, sodium and potassium in a historic mortar sample.

X-ray fluorescence (XRF)

The results of XRF conducted by Schueremans *et al.* (2011) on a church and a castle (constructed during the fourth and twelfth centuries) showed 72% traces of sand, 15% lime, 3% pozzolanic material and a 0.4 binder/aggregate-ratio. The mortar indicated traces of some essential parameters such as insoluble build-up CO_2, cementation (CI) and hydraulicity indices (HI). Other authors also found traces of lime and calcium in the mortars that were used from as far back as the fourth century BC.

The cementation and hydraulicity status of historic mortars are some of the important aspects to look at during the characterisation process. Therefore, in addition to calcium, which is identified as a major element in a lime-based mortar, there are other element oxides such as silica (Si), aluminium (Al), iron (Fe) and magnesium (Mg) that need to be quantified for calculation of the cementation (CI) and hydraulicity (HI) indices (Boynton, 1980; Holmes & Wingate, 1997; Elsen *et al*., 2011; Brosnan, 2014). Both the CI and HI are calculated using Equations (2.5) and (2.6), respectively.

$$\mathrm{CI} = \frac{(2.8 \text{ x } \%\mathrm{SiO_2} + 1.1 \text{ x } \%\mathrm{Al_2O_3} + 0.7 \text{ x } \%\mathrm{Fe_2O_3})}{(\%\mathrm{CaO} + 1.4 \text{ x } \%\mathrm{MgO})} \tag{2.5}$$

$$\mathrm{HI} = \frac{\mathrm{SiO_2} + \mathrm{Al_2O_3}}{\mathrm{CaO}} \tag{2.6}$$

Where:
SiO_2 – Silica
Al_2O_3 – Aluminium Oxide
Fe_2O_3 – Iron Oxide
CaO – Calcium Oxide
MgO – Magnesium Oxide

The CI quantities in Table 2.4 represent different lime and cement contents in mortar samples. According to Groot *et al*. (2004), the information on hydraulicity of the binder is considered a major requirement for compatibility on heritage mortars.

Table 2.4 Cementation index classification (Martínez *et al*., 2013; Brosnan, 2014)

Binder description	CI	Active clay in the limestone
Pure or non-hydraulic (aerial) lime	CI<0.15	Very little clay
Sub-hydraulic lime	0.15 to 0.3	Very little clay
Slightly hydraulic limes	0.3 to 0.5	Around 8%
Moderately hydraulic limes	0.5 to 0.7	Around 15%
Eminently hydraulic limes	0.7 to 1.1	Around 25%
Natural cement	1.7	Up to 45%

AAS and ICP-OES

The oxides of chemical elements are quantified by the use of atomic absorption spectroscopy (AAS) or inductively coupled plasma optical emission spectroscopy (ICP-OES). The analysis takes place on a liquid sample that is fed into the plasma. The molecules are converted to individual atoms and ions using high-temperature ratio frequency induced argon plasma (Elsen *et al*., 2011). The tests require approximately 10 g of the material. The first step for preparation of the sample involves disaggregating the crushed material to separate the binder (<0.063 mm) from the aggregates (0.063-2 mm). This is done when the aggregate is assumed to be soluble in acid; hence, both binder and aggregate are analysed separately. After that, the material is dried at 40 °C and immersed in 200 ml of deionised water or HCl. The filtrate is tested for the presence of element oxides and their quantities. The undissolved residue is considered the aggregate content.

2.4.6 Mechanical analysis

The strength and stiffness properties of mortar in existing buildings are quite challenging to obtain. This is because only a limited quantity of samples can be obtained from the historic buildings, making it difficult to carry out standard tests. Nonetheless, there are non-destructive testing methods used to determine mechanical properties.

Non-destructive testing

Schueremans *et al*. (2011) carried out a surface test on-site using a pendulum Schmidt hammer to provide information on mortar hardness, as depicted in Figure 2.13. The hammer is held at right angle to the tested surface to record the hardness. This test produces surface and pointing hardness which are affected by surface roughness, subsurface conditions such as voids, near-surface cracks, incipient spalls, specimen geometry, vicinity of nearby edges and hammer orientation (Schueremans *et al*., 2011). Hence, careful selection of the testing area needs to be prioritised to avoid misleading results. As depicted in Figure 2.13, lime mortars (T1, T5 and T7) have low hardness and cementation values while, hydraulic mortars have higher values. Schueremans *et al*. (2011) further found alternative tests that involve drilling resistance, tensile strength test, single or double flat jacks, and rebar locator and ultrasonic pulse velocity

(UPV) tests to mention but a few. In addition, Benedetti and Pelà (2012) provided an example of obtaining mechanical properties using an in-situ penetrometer technique. This method is carried out to measure the energy required to penetrate a masonry structure by means of drilling resistance. Due to scattered results, this technique requires caution when combining and analysing results.

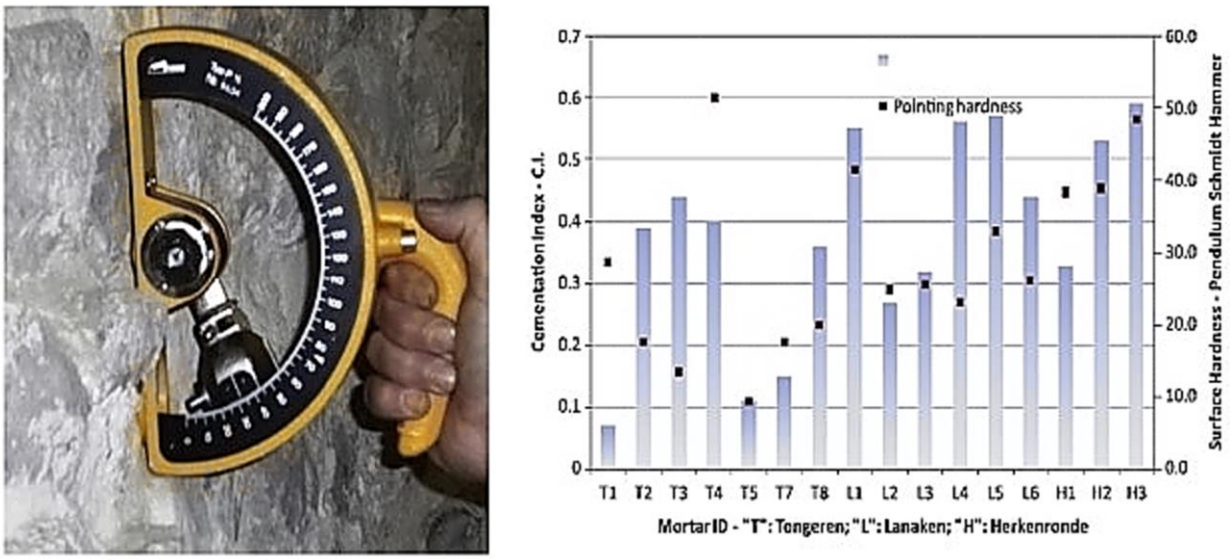


Figure 2.13 Pendulum Schmidt hammer (Schueremans *et al*., 2011)

In addition to the testing techniques outlined by Schueremans *et al*. (2011), a field test (scratch test), shown in Figure 2.14, was suggested. This is an NDT method which formulates a relationship between the scratch index and cement content in historic mortars. The test sensitivity could be altered by changing the form of the probe, the spring force and/or changing the number of turns used to make a measurement. It is to be noted that there is a possible difference in surface and inner mortar hardness due to the susceptibility of surface areas to attack by environmental factors.

Figure 2.14 Mortar Scratch test (Lawrence *et al*., 2004)

Compressive and flexural strength

In addition to the NDT, some authors recommended minor destructive testing to determine the strength and durability of historic mortars. The sample size of the material collected was, however, not enough to

perform standard testing; so, smaller specimens with varying dimensions (non-standard) were collected and tested, and the results are shown in Table 2.5. Corinaldesi (2012); Papayianni *et al*. (2013) manufactured specimens of 40 x 40 x 160 mm and cured them for 28 days for the flexural strength test. They evaluated the strength in accordance with EN 1015-11. Papayianni *et al.* (2013) tested compressive strength in accordance with the American Society for Testing and Materials (ASTM) C191-81, using shaped cubic samples of dimensions 4 x 4 x 4 cm. Their study further tested the modulus of elasticity. A similar procedure was suggested by Ngoma (2009) where a dynamic E-modulus was used to test the strength of the material. The hardness of the material could also be quantified by the use of the Mohs hardness scale or Russack system for brick and mortar description. A different study, which involved curing the specimens for a period of 12 months and determining the strength, thereafter, was conducted by Apostolopoulou *et al*. (2017).

Table 2.5 Flexural strength (Drdácky´ *et al*., 2013)

Specimen number	Specimen width (mm)	Specimen height (mm)	Ultimate load (N)	Bending strength (MPa)
T Narni 1.1-1	29.5	19.7	128.6	1.011
T 1.1-2	24.6	23.2	73.03	0.827
1 Average				0.919
T Narni 1.2-1	16.6	12.8	12.16	0.537
T Narni 1.2-2	25.8	19.3	16.8	0.210
2 Average				0.374
T Narni 1.3-1	30.6	18.6	9.8	(0.083)
T 1.3-2	16.3	15.9	72.8	2.631
T 1.3-3	14.2	10.7	15.51	1.431
T 1.3-4	12.6	12.9	9.5	0.681
3 Average				1.581 (1.207)
T Narni 1.4-1	29.2	19.7	14.4	(0.138)
T Narni 1.4-2	29.5	19.7	25.3	0.238
T 1.4-3	19.8	19.3	33.0	0.674
T 1.4-4	17.6	13.8	4.5	0.199
4 Average				0.370 (0.312)

2.4.7 Analytical techniques summary

There is a great number of analytical methods that have been applied in characterising heritage mortars, as discussed in Sections 2.4.2 to 2.4.6. The experimental work for characterisation of historic mortars as per reviewed literature is shown in Table 2.6. This shows both field and laboratory tests that were conducted by numerous authors around the world. In addition to the tests shown in Table 2.6, there are other tests used to characterise historic mortars that include: electron probe analysis, indirect tensile and bending tests, moisture content, permeability, density, water saturation method, gas chromatography and Hydrogen potential (pH) tests. All these methods provide useful information concerning different aspects of a mortar, and some complement others. It is, therefore, advisable to carefully select the most suitable method based on the type of properties aimed to be achieved at the end of the analysis. The required amount of material for a test also acts as a determining factor for the choice of the analytical technique.

Table 2.6 Characterisation of historic mortar- Analytical methods

Properties	**Aesthetic**		**Physical**				**Chemical**			**Mineralogical**			**Mechanical**		
Author	Visual investigation & cohesion	Microscopy	SEM	Thermal Conductivity	Porosity	Granulometry	Wet chem. analysis	XRF	Soluble salts (AAS/ICP)	FTIR	TGA/DTA	XRD	Compressive test	Flexural test	Surface hardness
Apostolopoulou *et al*. (2017)	✓	✓		✓	✓	✓			✓		✓	✓	✓		
Balksten (2010)			✓					✓				✓			
Bertolini *et al*. (2013)	✓	✓	✓								✓	✓			
Borsoi *et al*. (2010)		✓	✓	✓		✓					✓	✓			
Corinaldesi (2012)					✓	✓						✓	✓	✓	
Drdácky´ *et al*. (2013)	✓	✓	✓		✓						✓		✓	✓	
Fang *et al*. (2015)			✓							✓	✓	✓			
Gleize *et al*. (2009)						✓			✓	✓	✓	✓			
Gosselin *et al*. (2008)			✓								✓	✓			
Gulzar *et al*. (2013)	✓	✓	✓					✓				✓			
Hauková *et al.* (2013)								✓				✓			
Hormes *et al*. (2016)								✓				✓			
Labiadh *et al*. (2009)			✓			✓					✓	✓			
Lopez-Arce *et al*. (2016)	✓	✓	✓		✓							✓	✓	✓	
Marini *et al*. (2018)												✓	✓	✓	
Morricone *et al*. (2013)										✓		✓			
Ontiveros-Ortega *et al*. (2016)			✓		✓			✓				✓	✓		
Özkaya and Böke (2009)			✓			✓	✓				✓	✓			
Schueremans *et al*. (2011)	✓	✓			✓	✓	✓	✓			✓	✓			✓

2.5 Compatibility assessment

Isebaert *et al*. (2014) describe compatibility as "*utilising materials that do not have negative consequences on the original materials*." Additionally, Bertolini *et al*. (2013) describe compatibility as a major factor to assess prior to selection of a mortar to be used along-side suitable procedures for restoration of heritage mortars. Cizer *et al*. (2010) and Singh *et al.* (2014) add that this definition is based on the original mortar's properties, which are compared to the repair mortars. Schueremans *et al*. (2011) recommend that the repair mortars should match the original in terms of physical, mechanical and chemical properties. This is to guarantee the long-term durability and strength of historical structures. It is therefore essential to conduct a detailed assessment of the material properties before carrying out repair works. According to Deshpande (2017), finding a compatible material to duplicate the original is an even-greater challenge globally. To overcome this challenge, Hormes *et al*. (2016) emphasise the need to thoroughly study the material properties and testing technologies. They point out that material analysis assists in the production of a good repair mortar that matches the cementing material used originally.

It is vital to carefully select a number of tests to study the properties of the original mortar to ensure compatibility with the repair mortar. Figure 2.15 summarises the overall conventional characterisation techniques for achieving compatibility on historic mortars. The knowledge of original material properties makes it possible to foresee how the historic structure will react with restoration materials applied to it. Such critical information can only be achieved through original material characterisation.

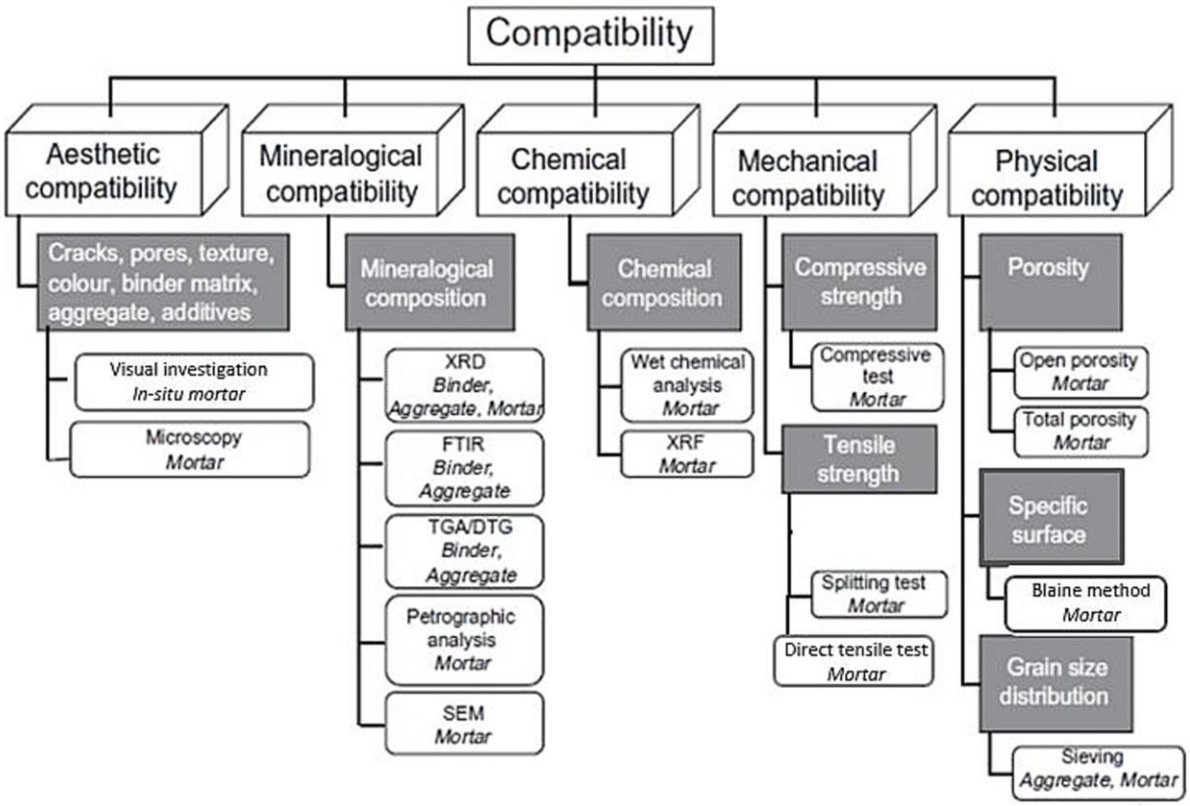


Figure 2.15 Compatibility assessment (Schueremans *et al*., 2011)

2.6 Historic mortar characterisation - Case studies

Characterisation of historic mortars for compatible restoration has been carried out for many years in different countries all around the world. Several studies using various techniques, as shown in Figure 2.15, were conducted in Europe, Asia, South America and (very few) in Africa. The case studies and results based on analytical techniques listed in Table 2.6 are summarised in Table 2.7. All case studies indicated some traces of different lime binders. This section summarises the case studies from the literature studied, indicating the location of study, types of historic structures investigated, their age and the results obtained. All the researchers in this study investigated the properties of heritage mortars by means of different methods, which Palomo *et al*. (2014) propose should be standardised.

2.6.1 Global

According to Bartz and Filar (2010), there is a wide range of publications on historic mortar characterisation which dates as far back as the 1960s. Most researchers have experimentally investigated mortars from different locations (floors, rendering and plastering, laying/bedding) of the monuments in their respective countries, with specific attention to compatible restoration. The literature generally shows case studies on historic buildings, mostly in Europe, Asia and very few in South America and Africa.

Table 2.7 Case studies in literature

Author	Location	Type of structure	Age (Century)	Material tested	Samples tested	Results & Analysis
Apostolopoulou *et al*. (2017)	Greece (Europe)	Church	12th	Internal & external mortar	10	High porous lime-based mortars with minor additions of organic admixtures.
Bertolini *et al.* (2013)	Italy (Europe)	Church	4th & 5th	Mortar	40	Binder was mainly based on magnesian lime. Gypsum was found in most samples.
Borsoi *et al*. (2010)	Portugal (Europe)	Archaeological site	I-IV A.D.	Mortar	11	Traces of aerial calcitic lime with quartz, schistose and granitoid aggregates and artificial pozzolanic materials.
Drdácky´ *et al*. (2013)	Italy (Europe)	Roman arch bridge	27 BC	Roman mortars	4	The strongest mortar was used.
Gleize *et al*. (2009)	Brazil (South America)	9 historic buildings in the State of Santa Catarina	18th – 20th	Rendering mortars	9	Hydrated lime obtained from the burning of seashells was the major binder. It was sometimes mixed with hydraulic materials (clay, ground ceramic tile or brick, and hydraulic lime).
Gosselin *et al*. (2008)	France (Europe)	Cathedral	11th	Pointing mortars	11	Old mortars show traces of siliceous and carbonates aggregates.
Gulzar *et al*. (2013)	Pakistan (Asia)	Mughal Empire	17th	Plaster & mortar	7	Calcitic lime binder produced from calcinations of kankar-$CaCO_3$ from soil horizon. The aggregates included crushed bricks, broken kankar pieces, brick kiln furnace slag and a small fraction of siliceous sand.
Hormes *et al*. (2016)	Germany (Europe)	Cathedral	13th	Mortar	3	High concentration of iron, comparable to that of calcium.
Labiadh *et al*. (2009)	Tunisia (Africa)	Ottomans monuments	15th	waterproof-coating mortars	2	Presence of air-hardening lime, pozzolanic and gypsum, indicating sulfatic but free of ettringite.
Lopez-Arce *et al*. (2016)	France (Europe)	Exhibition hall and museum	18th	Plaster mortars	9	Traces of gypsum, calcium and sodium sulfates, minor amounts of nitrates were found in mortar.
Morricone *et al*. (2013)	Rome (Europe)	Archaeological site of Porta Marina	3rd BC	Pointing mortar	25	Aggregates present in the mortars are essentially pozzolanic.
Ontiveros-Ortega *et al.* (2016)	Spain (Europe)	Theatre	4th BC	Substructure mortars	14	Presence of lime nodules in the mortar, calcareous crust.
Özkaya & Böke (2009)	Turkey (Europe)	Temple	1st BC	Wall mortars	2	Pure lime, coarse aggregates particle sizes > 1 mm and pozzolanic fine aggregates in Roman mortars.
Schueremans *et al*. (2011)	Belgium (Europe)	2 Churches & Castle	4th & 12th	Pointing mortar	9,6,3	Relatively similar properties. Carbonation of lime in the pointing mortar.

2.6.2 Africa

The African continent is home to many heritage sites, as shown in Figure 2.16. In addition to the world heritage sites recognised by UNESCO, respective countries have identified places of historical significance that represent their history. This being the case, there are hundreds of historical sites around Africa, some of which are structurally deteriorating and in need of repairs. However, from the literature explored in this study, a conclusion can be drawn around the topic of historic material characterisation for restoration purposes around Africa. Studies on historic mortars are relatively rare, especially when considering the number of historic buildings present in Africa. This research could only access two studies from all the reviewed literature focusing on material analysis in North and East Africa, and none in Southern Africa.

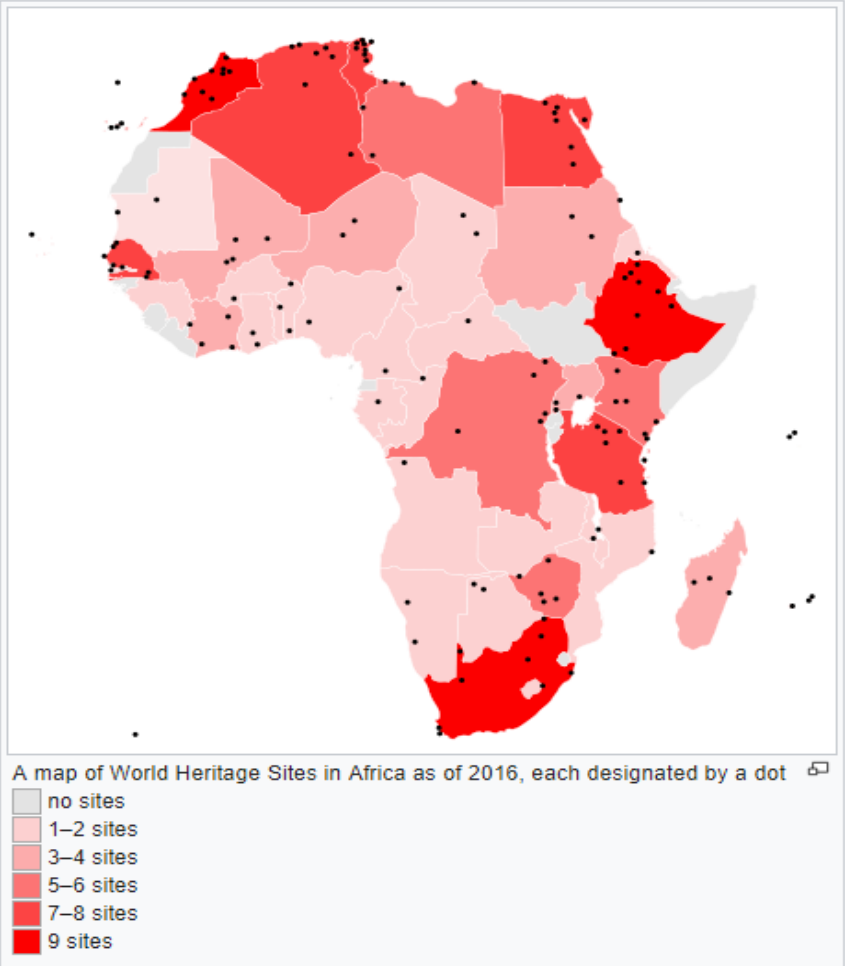


Figure 2.16 World Heritage Sites in Africa (UNESCO, 2018)

2.6.3 South Africa

There are hundreds of heritage sites in South Africa, which were formerly protected by the National Monuments, until the Department of Arts and Culture (under NHRA, No. 25 of 1999) tasked the South African Heritage Resources Agency with identification and management of such sites. These sites include places and objects that have exceptional qualities through their association with historical events,

persons, organisations or have scientific, social or cultural values across the country (SAHRA, 2018). The authenticity and integral protection of the heritage sites are currently governed by the legislation, which offers permanent formal protection under Part 1 of Chapter 2 of the National Heritage Resources Act (NHRA) introduced in April 2000.

For the protection of these heritage sites, SAHRA's mandate is: "*to promote social cohesion in South Africa by identification, conservation and management of heritage resources so that they can contribute to socio-economic development and nation-building*." (SAHRA, 2018). The agency has thus far identified both national and provincial heritage sites which are protected under the terms of the 1999 NHRA. This legislation mandates the requirement of a permit for any changes, destruction, excavations or damage to the sites. However, it was established during several interviews with SAHRA that most heritage restorers still lack understanding where authenticity is concerned on heritage building restoration. This has resulted in failures of restoration works, as material analysis was either not conducted at all or not cried out correctly.

This raises concern, not only for SAHRA, but historians and economists, as the loss of authenticity of heritage sites impacts severely on the history being represented and thus, tourist traffic deteriorates over time. In this study, no records could be found for publications on the characterisation of heritage cementing materials or mortars in the Southern Africa region. According to the Assessment report for Robben Island building precincts (2018), there was a proposal for the collection of existing mortars for analysis. The analysis was proposed for mitigation of the present deterioration on some of the oldest (1700s to early 1900s) buildings on the island. Therefore, to avoid repeated restoration works, incorporation of material analysis procedure into the conservation management plans would undoubtably ensure improved restoration process.

2.7 Optimisation of analytical techniques

The literature has thus far highlighted the analytical procedures used for characterisation of historic mortars, mostly based on an approach proposed by Van Balen *et al*. (1999). The approach combines several analytical methods, as summarised in a paper by Schueremans *et al.* (2011). It simply studies the binder and aggregate of ancient mortars, which are the fundamentals of mortar composition. However, though the approach recommended by Van Balen *et al*. (1999) has been generally accepted and widely used all around the world, the issue of optimisation of the analytical techniques has received almost no attention thus far. The current study seeks to optimise (by means of simple statistical methods) the characterisation techniques from a developing country's perspective, based on the elements discussed in this section. The main objective of this research was to set a standard protocol for characterising heritage cementing materials for compatible restoration.

2.8 The standard protocol

The literature emphasises the importance of achieving some degree of universality for the approach to characterising heritage mortars. This research takes a similar tack to that in a PhD thesis submitted by Ngoma (2009), where he reports that there is still a gap in the literature when it comes to a standard method used for analysing historical masonry and mortars internationally. As a result, developing a standard protocol from the existing techniques of material analysis would solve the majority of restoration

problems facing the construction industry in terms of appropriate repairs. The present analytical techniques somewhat complement each other, making it challenging to select an optimum methodology. The selection of the appropriate methodology requires careful consideration of the aspects mentioned in this section.

2.8.1 Time and cost

Research by Hauková *et al.* (2013) shows that a majority of the analytical methods identified by most studies are too costly to conduct regularly, especially for conservation and restoration of historic buildings. Since the main objective for carrying out heritage mortar characterisation is sustainable compatible restoration, it is considered extremely important to assess the cost-efficiency before carrying out scientific characterisation. The assessment could involve the equipment and laboratory arrangement costs, the expertise required as well as cost of time. We seek to achieve the standardised cost and time effective characterisation techniques and procedures from the current outlined analytical methods. This will not only be a credible source for restorers when attempting restoration of heritage buildings, but will also play an important role in economic sustainability, particularly for developing countries.

2.8.2 Amount of material required

The places of historic significance are protected by heritage authorities, which means alterations on these buildings are always monitored, with policies in place indicating restrictions to proposed alterations on these structures. It is therefore advisable to carry out experiments using as little materials as possible. Therefore, the choice of analytical methods needs to consider the amount of mortar required to successfully complete the tests. The less the material required, the better.

2.8.3 Technical data quality

Hauková *et al*. (2013) mention that some of the methods listed and used by a majority of the researchers rarely provide useful technical information, yet they are generally very costly. Thus, one needs to take a closer look at the results/information obtainable from a certain technique and their importance or application in producing a replica mortar (Hughes & Callebaut, 1999). It is imperative to determine the necessity of a test in relation to its significance in terms of providing necessary information, as opposed to non-applicable details in restoration practices.

2.8.4 Ease of use

The main purpose of material characterisation is to collect the essential information for restoration using a limited sample size, time and budget (Hauková *et al.*, 2013). It is suggested that the use of a simplified analytical procedure would yield better results. To select the most complex procedures which require a high level of expertise is discouraged, because this will cause shortcomings in terms of cost in acquiring such expertise. This simplicity is applicable not only to the characterisation of the materials, but also to the results obtained. It serves no purpose to gather complicated results that would require a high level of expertise to analyse and interpret, especially since the results are intended for application in restoration.

2.8.5 Acceptance by other researchers

Another factor that needs to be considered when determining the type of methodology to use for the characterisation of heritage cementing materials is the preference for the technique by other researchers. This requires careful consideration, since there are some factors such as the location of the study and the economic standing of the country (developed or developing) that need to be considered before making a decision on analytical methods to use. It is to be noted that the technique might be popular in terms of application among researchers, but considered costly or inaccessible in terms of availability of laboratory facilities or equipment in developing countries.

2.9 Characterisation for compatible restoration

According to Sandbhor and Botre (2013), *restoration* is: "the act or process of accurately depicting the form, features and character of a property as it appeared at a particular period of time." The need to augment the strength of historic masonries dates back hundreds of years. Fang (2015) points out that the conservation and preservation of heritage buildings should be an important focus, since heritage buildings represent the beauty of history. The need for restoration of heritage structures is further emphasised by the fact that a majority of the heritage buildings are in major cities and take up a lot of space where developers would rather create new structures. Some heritage structures have become safety hazards, as they have deteriorated severely, showing risks of collapse. It is therefore important to ensure that these structures continue to exist and are protected. A study by Gulzar *et al*. (2013) concludes that restoration and protection of historic structures require the application of materials which match the original ones.

According to Penelis (1996), it is more challenging to repair historic buildings than ordinary buildings. This is because the attention when restoring historic buildings is focused mainly on maintaining the historic originality, which becomes a significant challenge for most African countries that lack understanding and research around historic material analysis for restoration interventions. Dolar-Mantuani (1984) emphasises the need to consider not only the appearance of the material but also the properties (strength, adhesion, flexibility and porosity) and the structure's future maintenance. He further specifies the negative consequences associated with inappropriate or poor execution of repairs of historic mortars which include premature damage and accelerated deterioration of the original building fabric. Often, more damage is done during the restoration of heritage buildings, increasing the cost of future maintenance and repair works.

A study by Papayianni and Pachta (2017) indicates that the use of compatible and locally available materials has been introduced for the restoration of old earth block masonry buildings. This includes the development of useful repair materials and techniques to utilise when repairing these structures. In a study by Lourenço (2006), he raises a concern as far as durability, strength and architecture in restoration are concerned. He highlights the importance of differentiating between the science of construction and the art of conservation and restoration, as these two are considered independent aspects that require separate attention. The repair and restoration of heritage structures is a major challenge, as most historic buildings were built using materials that have been superseded by modern construction materials. There is currently an influx of new materials because of enormous growth in the construction industry over the years. This is the result of new and innovative construction materials and techniques being developed and applied all around the world.

The typical procedure for restoration of heritage cementing materials is through the application of Portland cement, but the approach often fails (Lanas & Alvarez, 2003; Martínez *et al*., 2013; Marini *et al*., 2018). The failure of Portland cement-based mortar when repairing structures where historic lime-based mortars were used is due to the shortcomings of Portland cement itself, which involve quicker cracking and corrosion of the original fibres, as the cement contains more soluble salts, which result in salt crystallisation (Marini *et al*., 2018). The substitution of original material with Portland cement has been found to intensify damage to the original fibres in the mortar. The damage is a result of the following properties portrayed by Portland cement: high compressive strength and modulus, large thermal expansion coefficient, low porosity, and mainly consists of microscopic pores which hinder water movement and air circulation in the masonry, which in turn harms the original fabric (Rodriguez-Navarro *et al*., 1998; Martínez *et al*., 2013; Gulbe *et al*., 2017).

The use of Portland cement for the repair of heritage buildings was reported in the Conditional Assessment Report for Robben Island (2018). The report indicates that some of the oldest buildings on the island appeared to have lost their historic features due to previous maintenance, which did not consider air circulation within the wall structures. The conclusion made in this report was that most buildings' plaster and raised pointing needed to be removed, as they were suspected to be silicone and cement, which are detrimental to the overall fabric. It was therefore recommended to replace the plaster and the pointing material with lime-based material which matches the original material. This damage would have been avoided if the original materials were analysed prior to application of repair materials.
Singh *et al*. (2014) and Apostolopoulou *et al*. (2017) state that a study on restoration of historic mortars may include information on aesthetic and material property compatibility (*chemical, physical and mechanical*). According to these studies, several researchers fail to explain the context of compatibility when it comes to restoration of historic mortars. The compatibility of heritage materials is entirely dependent on original material characterisation prior to application in restoration.

2.10 Summary

Heritage structures have a positive impact on the history, culture, and economy of their respective countries and therefore, their care and maintenance are considered important. The concept of ancient mortar characterisation started around the 1960s and has been explored extensively on a global scale; but there is no literature related to this topic in South Africa. An investigation of the properties of heritage cementing materials is required, because appropriate restoration of heritage buildings contributes to the country's economy through travel and tourism. A holistic approach is necessary to provide comfortable and tailor-made cementing materials for restoration of heritage buildings so as to meet expectations at a lower cost.

Originally used materials can only be identified through experimental characterisation. Characterisation of heritage cementing materials involves use of methods such as visual investigation, wet chemical analysis, x-ray diffraction (XRD), x-ray fluorescence (XRF), atomic absorption spectroscopy (AAS) and scanning electron microscopy (SEM), among others. The restoration of heritage structures is carried out with careful consideration of the original properties of the materials used in the structure. According to the literature, mortars and grouts used for repair of historic buildings must have properties that are compatible to the original to merge in the right way: equal in strength, adhesion, flexibility and porosity. The materials distinguish structures from one another in terms of the unique story a building tells about

the history it represents; and therefore, characterisation of original materials helps maintain the authenticity of these structures.

In all the presented case studies, the main focus was on the physical, mineralogical and chemical characterisation of the cementing materials, with less focus on mechanical properties. It can also be seen from the presented case studies that most heritage buildings were constructed using lime mortars, not Portland cement. Hence, the use of modern ordinary Portland cement (OPC) for production of heritage repair mortars would only cause harm to the original surface with time, as the binder properties do not match. Therefore, for the effectiveness of the repairs, there is a need to identify and develop a standardised test methodology which will be used for characterising the cementing materials prior to the restoration process.

2.11 Conclusion

Although extensive and detailed work pertaining to historic mortar characterisation has been undertaken, there are still some critical areas that have received less attention and need to be addressed. A majority of the researchers have only shed light on the process of characterising heritage mortars but have not provided a standard procedure for carrying out such characterisation. No literature could be found concerning studies providing guidelines for a standard procedure to characterise heritage materials. There are a few cases where researchers have attempted to create a systematic approach for analysis of heritage mortars. Nonetheless, the subject of guidelines for characterisation of heritage cementing materials has received almost no attention thus far.

The literature highlights the proposed test methods, some of which are lengthy, costly (especially for developing countries) and require dedicated instruments and skilled operators (research experts). This raises the question as to what is then considered the most economically effective and easy-to-conduct method that provides sufficient information on properties (physical, mineralogical and chemical) of heritage cementing materials for repair purposes. Since there is a lack of academic publications on the issue of heritage mortar characterisation in South Africa, obtaining accurate solutions to the problem of decay on these buildings becomes an everlasting challenge. It results into use of incompatible materials during the restoration process. Therefore, there is an indisputably need to develop and standardise a methodology to approach restoration interventions on historical mortars.

It is clear, from the literature reviewed that we know very little about the composition of original heritage cementing materials. The overall challenge is incorporating the old mortars (with unknown properties) with new materials to solve the problem of decay on heritage cementing materials. While there are more publications relating to heritage mortar characterisation in most European countries, it is evident that providing a standard protocol for characterising these materials has been given minimal attention, even in Europe. The choice of analytical techniques is totally at the researcher's discretion and personal preference (subjective). The objective of this work was to come up with an optimised protocol to characterise the original materials, especially for South Africa, as there is no data available in this regard. The implementation of specific methodologies could be incorporated into conservation management plans; under material analysis of original materials. This will be a worthwhile investment, as it will help augment the life span of historic buildings through compatible restoration.

Chapter 3 Research methodology

3.1 Introduction

The process of characterising historic mortars has been carried out over the years throughout the world, using various techniques and methods, mostly non-destructive. There are, however, no standardised methodologies for characterising cementing materials for their restoration purposes. This raises a concern in terms of selecting repair materials. Therefore, this study intends to characterise the heritage cementing materials on the structures constructed between the 1600s and 1800s. It further investigates the standard analytical methodology for obtaining the properties of existing cementing materials.

This chapter presents a detailed methodology used to answer the research question of this study. It presents the procedure followed for collection and analysis of data. The chapter provides an approach that was used to complete the experimental study procedure. It encompassed data collection through personal interviews and literature reviews, field and laboratory testing on original materials for aesthetic, physical, mineralogical and chemical properties. Aspects including equipment, data collection and analysis are also discussed.

3.2 Research design

A field assessment and an experimental procedure were carried out for this study. The cementing material samples from the existing buildings were taken (within the limitations inherent to historic structures) and their composition studied to identify mortar constituents: nature of binder, aggregate, additives, and their relative proportions. The physical, mineralogical and chemical properties were evaluated using the RILEM Standards for material testing, where the sample amount was adequate, though non-standard tests were conducted where there were limited samples. The study was in alignment with the methodological literature as reference.

3.3 Methodology

The goal of this project was to develop a standard protocol for the analysis of heritage mortar components. The protocol was developed from the existing techniques for characterisation of historic cementing materials. The work involved an experimental investigation into the properties of the heritage cementing materials of the selected heritage sites in the Western Cape Province, South Africa. The representative structures contained sections that were constructed between the 17th and 19th century, which qualified them to be the subject of this research. As indicated in Table 3.1, the methodology matrix included analyses of materials from different areas of the structure (floor, plaster, render, and bedding). This was conducted with the purpose of fully understanding the material properties of the materials used on the structure prior to the emergence of Portland cement as the mortar of choice.

Table 3.1 Methodology summary

Material era	Sample location	Test	Material required (g)	Tested paramenters	Materials & Equipment	
1600s 1700s 1800s	S1 - Floor S2 - Joints & pointing S3 - Plaster (All these per century material)	Visual investigation & microscopic imaging	10	Appearance colour i.e. Reddish brown, grey	Naked eye, Color scale, optical microscope	*Compare and analyse cost, time, complexity, accuracy & results, then select appropriate optimisation technique to select a set of standard protocols/guideline for characterising heritage cementing materials.*
		Cohesion test	10	Texture & feel, behavour towards application of finger pressure	Human fingers	
		Sieve analysis	300	Grain size distribution & particle shapes i.e. 10% fines, 40% aggregates, grain distribution curve	Sieve stack, analytical scale,	
		Titration for qualitative analysis of salts	0.1	Carbonates, sulfates, chlorides, nitrates, nitrites	analytical grade chemicals, reagents, indicators, test tubes, beakers	
		Titration for quantitative analysis of element oxides	10	Element oxides, binder to aggregate ratio	pipette, burette, beakers, oven, magnetic stirrer, analytical grade chemicals, reagents, indicators	

3.4 Study area

From the literature reviewed, it is evident that the majority of research was conducted on churches, museums and castles, built from as far back as the 1st century BC. This is most certainly because heritage materials act as a reminder of the past religious, architectural and political history. The selected heritage sites studied are located in the Western Cape Province, South Africa (see Figure 3.1) built and date from the 17th century onwards. The sites included the oldest colonial building in South Africa (Castle of Good Hope) and the world-renowned Robben Island. Both these structures are in Cape Town.

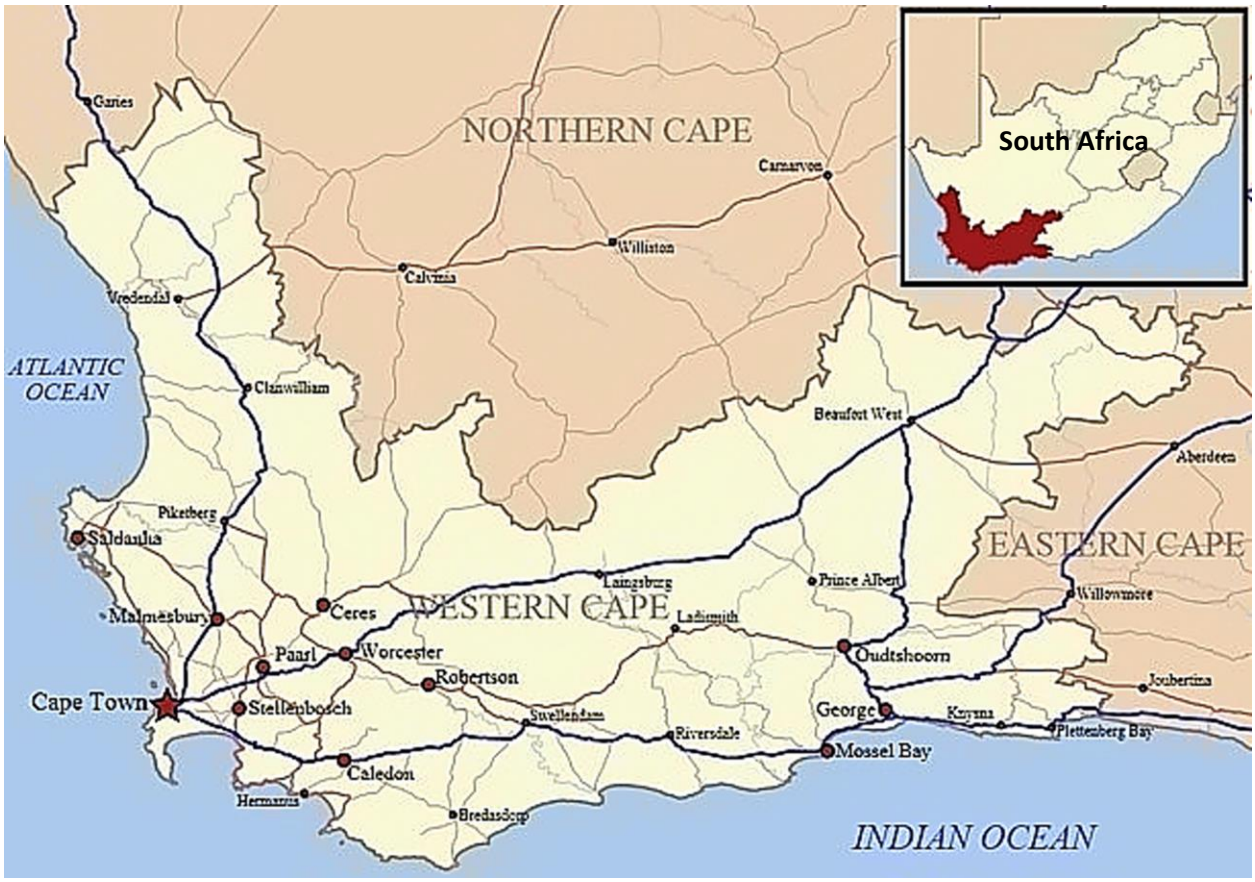


Figure 3.1 Study area: Western Cape, South Africa (Patterns and characteristics of migration to the Western Cape, South Africa, 2019)

3.5 Building Selection

There are hundreds of heritage structures around South Africa. However, very few met the selection criteria, which is depended on various factors discussed below:

The study focussed only on declared (by SAHRA) heritage buildings in the Western Cape province in South Africa.

There was no criterion set for the level of the declaration; both provincial and national heritage structures were eligible for selection in this study.

Only stone and brick masonry structures were identified for the next selection phase. This was a result of the need to study the cementing materials that bind the stone/brickwork.

For the building to be on the selection list, the minimum possible information with relation to its construction period had to be somehow accessible from the archives. This was a condition set to ensure the use of only the structures from the 1600s to 1800s. Additional information regarding the previous restoration, alterations, conservation and repairs had to be available.

Even if the structure met the above criteria, further investigation was carried out on the original historical status of the structure. The buildings which have not entirely lost their original appearance because of repairs and modifications were identified through interviews with SAHRA and site visits to inspect the buildings.

After such buildings were identified, prior to sampling, it was imperative to ensure that the site owners would allow sampling of materials; hence, requests to sample were proposed. In addition to sampling, unlimited access to the site had to be guaranteed for the duration of the study to avoid interruptions.

The potential structures had to be those in which the day-to-day activities of the facility would not be interfered with by the study.

The availability of deteriorated cementing materials to allow sampling that would not harm the areas that are currently in good conditions was another condition for selection.

The Castle of Good Hope and Robben Island met the above criteria and were selected for the study.

3.5.1 Castle of Good Hope

The Castle is the second fort at the Cape which was built from 1666 to 1679 by the Dutch East India Company, making it the oldest existing colonial building in South Africa (A brief history of the Castle of Good Hope, 2018). It was originally located on the coastline of Table Bay, but, after land reclamation activities, is now some distance from the coast (Castle of Good Hope, 2018). The Castle is characterised as a National Heritage Site by SAHRA. It has a pentagonal shape and is made of stone and block masonry, as shown in Figure 3.2. This structure was constructed in various stages from 1666 to 1679; but, no specific properties of the Castle's original mortar have been characterised and documented in scholarly publications. There is a great possibility that different mortar types or mixes were used at the Castle since it was built in stages. At present, the Castle houses the administration and organisational elements of the military (Headquarters of Western Cape Command), while at the same time hosting tourists.

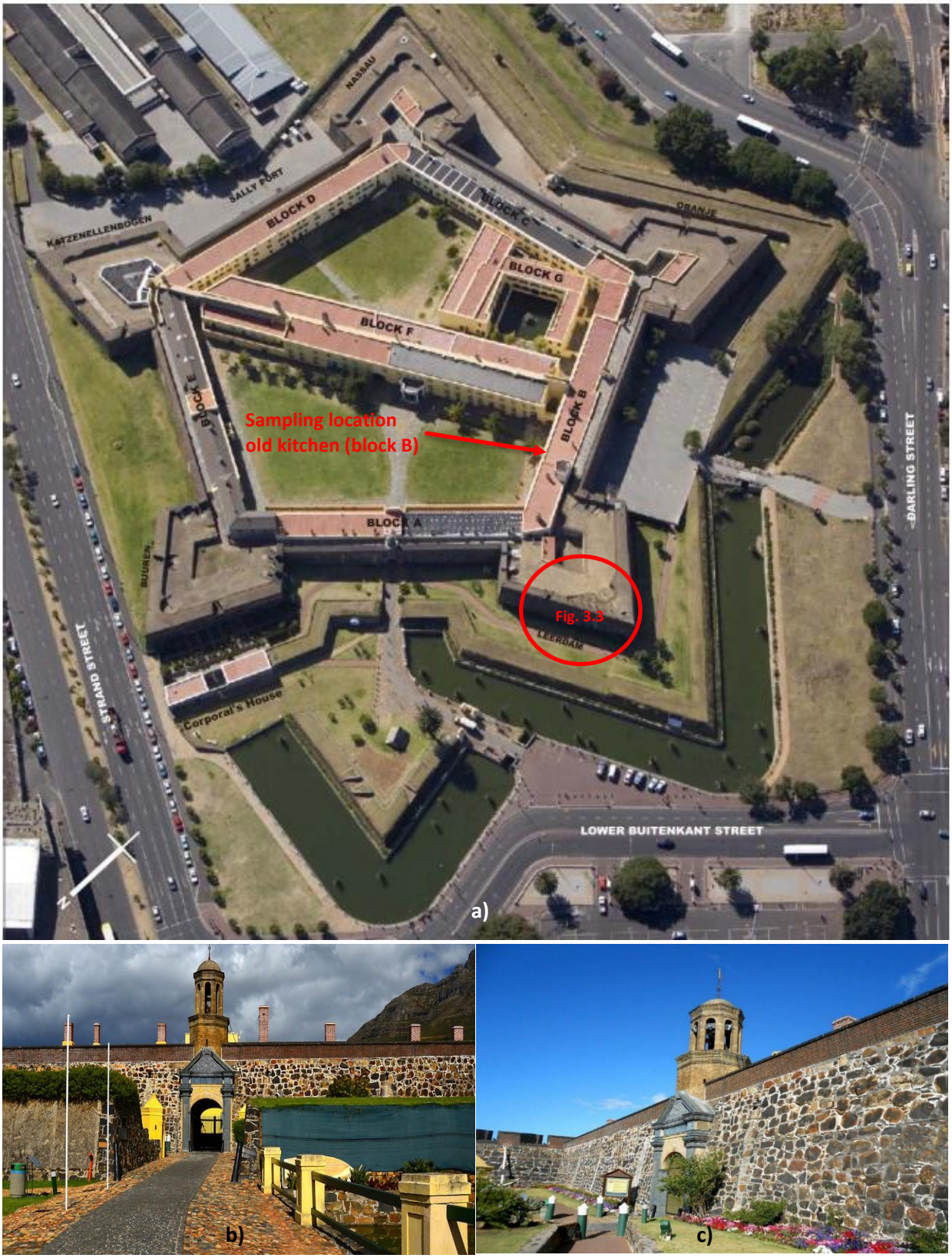


Figure 3.2 a) Contextual aerial photograph b) Entrance c) Elevation (Castle of Good Hope, 2018)

Castle restoration history

The castle, like any other old building, experienced deterioration, threatening its continued use and even existence. These conditions saw the commissioning of Gawie Fagan in the mid-to-late 1960s to make it habitable (Gilbert, 1994). Gilbert (1994) indicates that according to the assessment report by Fagan, the castle, in order to survive another century, had to be restored. The alternative was demolition. The decision by the then Department of Public Works was to carry out a complete restoration, not demolition. Some of the most recent cementing material repairs on the castle are clearly visible even from a distance, as depicted in Figure 3.3.

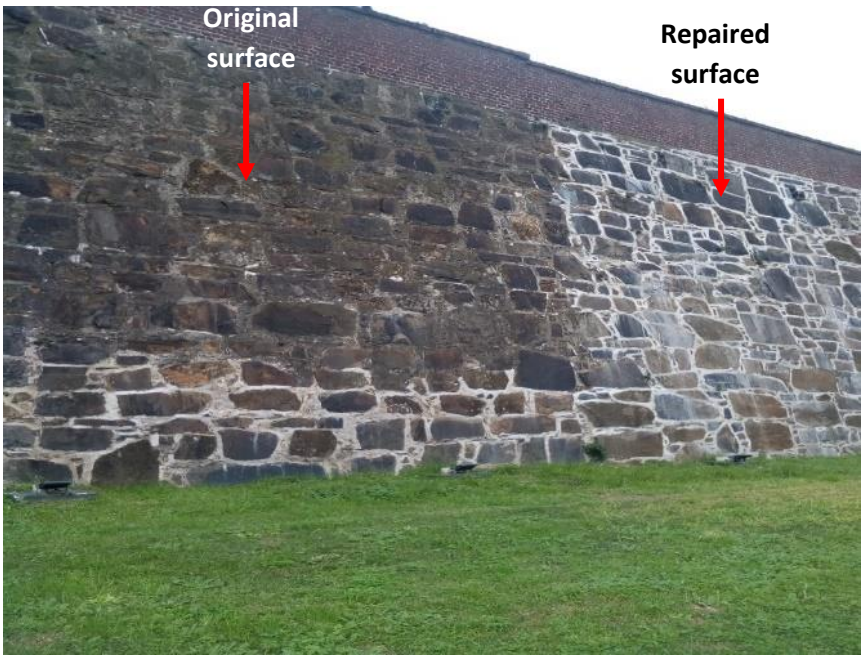


Figure 3.3 Wall section: Castle of Good Hope, South Africa

3.5.2 Robben Island

Robben Island is also categorised by SAHRA as a National heritage site and is one of the ten UNESCO World Heritage Sites in South Africa (Robben Island, 2018). This justifies the need for regular and proper maintenance of this site, as it represents the country and its history, both locally and abroad. The site has a political history dating back to the 17th century when it was used for isolation of political prisoners. The island became known internationally when it served as an isolation prison for liberation leaders and activists, including the first democratically elected black president in South African history; Nelson Mandela. Robben Island tells a sad but victorious story of the Apartheid governance and racism which came to an end as democracy and freedom came to life in South Africa in 1990s. The island attracts thousands of tourists from within Africa and around the world, thereby promoting economic growth for the country through travel and tourism, as well as employment.

This world heritage site is located in Table Bay. It was used as a maximum-security prison, with some additions made during the 19th century, and is now a museum. Like the castle, Robben Island was constructed with stone masonry, as depicted in Figure 3.4. The different material eras were collected for analysis and comparison from the structure.

Figure 3.4 a) Robben Island elevation b) Plan c) Entrance side view (Robben Island, 2018)

Robben Island restoration history

It was evident from visual inspections and the contents of the Conditional Assessment of Buildings on Robben Island (2018) by Charles Consult Consortium that the island has undergone maintenance and repair works in recent years (1999 onwards). In this assessment report, it was stated that the repairs on the wall pointing did not consider how air circulates through wall sections and how condensation and dampness were to be treated. It has been concluded that the repair work caused further damages to wall sections of the island. The need for material analysis prior to restoration of heritage structures is emphasised further in the assessment report. SAHRA has proposed maintenance work to rectify the previous restoration mistakes on the island. The deterioration seen on some of the oldest buildings on the island is shown in Figure 3.5. Regrettably, the material used for repair proved to be incompatible with both the original mortar and the masonry surface itself, and thus are seen to be detaching from the original surface on both the maximum-security prison (1700s) and the pre-primary school building (1800's). The original material underneath the repair material was lime-based while the repair materials are Portland cement which are impermeable. This has caused the moisture movement problems on the wall structure and repair materials peeling off.

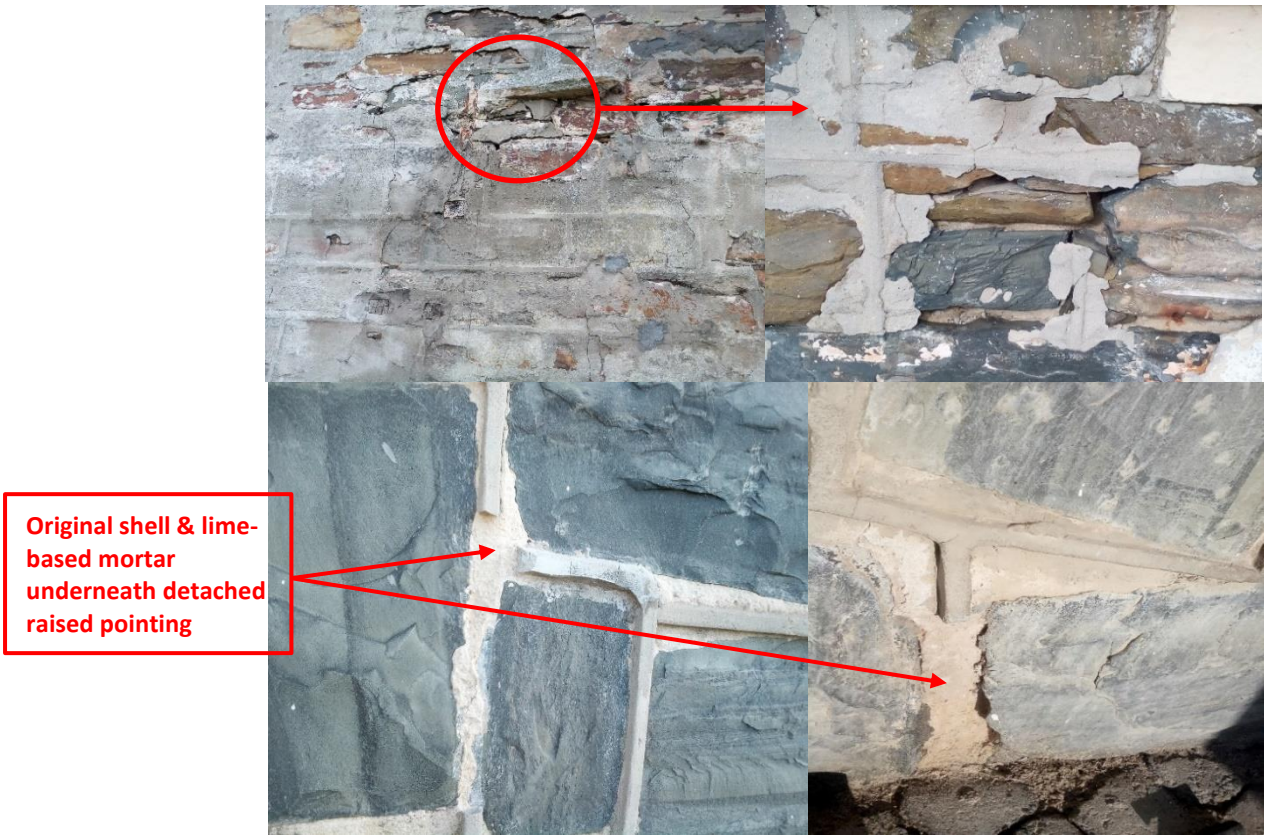


Figure 3.5 Recent failing repairs – Robben Island

3.6 Selection of analytical techniques

The selection of the preliminary set of tests for the investigation of historic mortars was carried out based on the details discussed in this section. The representative test procedures were selected from a list of analytical techniques providing similar results. Each test was critically assessed in terms of its advantages and disadvantages as discussed by Hughes and Callebaut (1999); Schueremans *et al*. (2011) and Hauková *et al*. (2013). A summary of the discussion can be found in Table 3.2.

The preliminary selection criteria used for analysing the cementing materials from the Castle of Good Hope and Robben Island included the following:

A. Time taken to conduct a test.
B. The cost of the test (equipment or expertise).
C. Availability of testing equipment within the current research area.
D. The quantity of material required for a test.
E. Technical data quality required in answering the set goals for analysis.
F. The ability of the method to give information on the mortar composition.

After completion of the preliminary minimum tests (highlighted in Table 3.2), the final tests were standardised based on their merits in relation to the results they provided. Some techniques complement each other, while others provide quantitative rather than qualitative results.

Table 3.2 Analytical methodology optimisation

Analysis	Method	A	B	C	D (g)	E	F	Advantages	Disadvantages
Aesthetic	Cohesion	<1 hr	None	Yes	10	Adequate	Yes	Specimen may be used in further analysis, technically easy to perform	Results may differ according to personal views (subjective)
	Colour test (colorimeter)	<1 hr	Fair. Colorimeter	No	<1	Adequate	No	Produces accurate colour measurements	Cannot be used for colourless compounds, similar colours can produce errors in results
	Optical microscopy	<1 hr	Fair. Microscope	Yes	10	Adequate	Yes (limited)	Allows determination of grain size distribution	1 day for preparation of thin section hence, time-consuming
	Visual investigation (Colour chart)	<1 hr	Low	Yes	10	Adequate	Yes (limited)	Specimen may be used in further analysis, technically easy to perform	Results may differ according to personal views (subjective)
Physical	Frost resistance/freeze-thaw	2 days	Fair. Freezer	No	>200	Adequate	No	Specimen may be used in further analysis, technically easy to perform	Time-consuming
	Moisture content (Gravimetric drying)	2 days	Fair	Yes	3 cm^3	Adequate	No	Specimen may be used in further analysis	Time-consuming
	Permeability (surface probe)	1 hr	Fair	No	3 cm^3	Adequate	Yes (limited)	Specimen may be used in further analysis	Time-consuming
	Porosity (mercury intrusion porosimetry)	1 hr	High. Equipment	No	10	Adequate	Yes (limited)	Rapid & accurate testing procedure, determines pore size distribution	Expensive lab equipment is needed; specimen is destroyed
	Porosity (Water absorption by immersion)	2 days	None	Yes	3-5 cm^3	Adequate	Yes (limited)	Technically easy to perform	Time-consuming
	Scanning electron microscopy (SEM)	<1 hr	High. Equipment	No	2-5	Adequate	Yes (elements)	Less preparation time of the sample	Expensive lab equipment & operations are needed, time-consuming in preparation of sample
	Sieve analysis (using HCl)	1-2 hrs	Low. Sieves, HCl	Yes	100	Adequate	Yes (Aggreg.)	Technically easy to perform	Large material quantity needed; sample is destroyed
	Sieve analysis (dry)	1 hr	Low. Sieves	Yes	50-100	Adequate	Yes (Aggreg.)	Technically easy to perform, sample can be used in future	Separation of the aggregate, large material quantity needed

Analysis	Method	A	B	C	D (g)	E	F	Advantages	Disadvantages
	Thermal conductivity	7 days	Fair. Oven	Yes	>200	Adequate	No	Technically easy to perform	Very time-consuming
	Water desorption test	2 days	None	Yes	3-5 cm^3	Adequate	Yes (Approx.)	Technically easy to perform	Time-consuming
Chemical	Atomic absorption spectroscopy (AAS)	1-2 hrs	High. Equipment	No	5-10	Adequate	Yes (elements)	High sensitivity, technically easy to perform	Expensive element, time-consuming when preparing the sample
	Hydrogen potential test	<1 hr	Low. pH meter	Yes	5-10	Inadequate	No	Technically easy to perform	Incomplete analysis details
	Induced coupled plasma (ICP)	1-2 hrs	High. Equipment	No	1-10	Adequate	Yes (elements)	High accuracy	Expensive lab equipment needed; time-consuming when preparing the sample
	Titration of element oxides	1 hr	Low. Chemicals	Yes	5-10	Adequate	Yes (Binder)	No need for high end equipment, easy to perform	Some level of operator skill required, can be time-consuming
	X-ray fluorescence (XRF)	1 hr	High. Equipment	No	1-10	Adequate	Yes	High accuracy & easy sample preparation	Expensive lab equipment needed
Mineralogical	Differential thermal analysis (DTA)	<1 hr	High. Equipment	No	1	Adequate	No	Highly sensitive	Expensive lab equipment needed
	Fourier transform infra-red spectrometry (FTIR)	<1 hr	High. Equipment	No	<2mg	Adequate	No	Highly sensitive	Expensive lab equipment needed
	Gas chromatography	<1 min	High. Equipment	No	<1	Adequate	No	High accuracy in few minutes	Expensive lab equipment needed
	Ion chromatography (IC)	24 hr	Fair. Ion strip	No	0.1	Adequate	Yes (ions)	Accurate as well as reliable	Time-consuming
	Salt crystallisation	<1 hr	Low. Burner	Yes	>200	Adequate	Yes (limited)	Technically easy to perform	Large quantity of sample required; results are based on assumptions
	Soluble salt analysis (titration)	<1 hr	Low. Chemicals	Yes	0.1	Adequate	Yes (major salts)	Technically easy to perform	Qualitative not quantitative analysis
	Thermogravimetric analysis (TGA)	3 hrs	High. Equipment	No	<2mg	Adequate	No	Small consumption of material	Expensive lab equipment needed
	X-ray diffraction (XRD)	<1 hr	Fair. Equipment	No	2-5	Adequate	Yes	Considered least expensive	Relatively low sensitivity, qualitative

3.7 Experimental procedure

An experimental approach to analysing heritage cementing materials was determined. The characterisation procedure was based primarily on the adaptation of testing procedures cited in the literature, as well as the RILEM standards and ASTM standards, where such applied. The proposed tests were performed based on non-destructive and minor destructive testing. They were carried out with consideration to health and safety precautions of the Republic of South Africa in a well-monitored environment. A series of aesthetic, physical, mineralogical and chemical tests, as summarised in Figure 3.6 were carried out in the Soils and Hydraulics laboratories of the Cape Peninsula University of Technology (CPUT), Bellville campus. Due to a limited quantity of samples collected, non-standard dimensions were used. The sample quantities were decided with reference to the literature studied. As mentioned in the scope of the study, no mechanical properties of cementing materials were investigated in this work.

According to the literature (Table 2.6), there are several techniques available for the analysis of historic mortars that many researchers have used in the past. However, it was concluded that not all could be considered economically viable. Thus, in order to achieve the objectives of this study, the experiments conducted were identified as the affordable (cost, time and expertise-wise) using the selection criteria in Table 3.2. The first phase of the research involved an experimental characterisation using the preliminary techniques, followed by the final optimisation of the optimum methods.

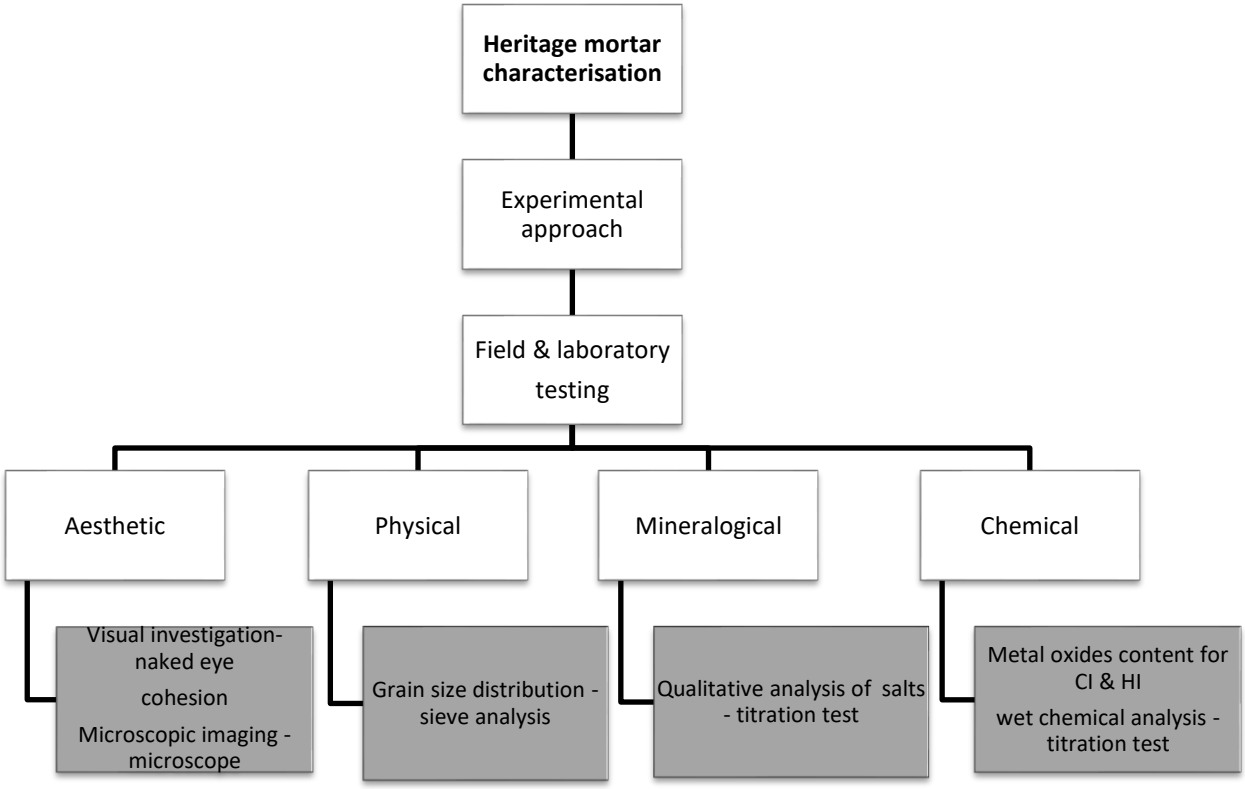


Figure 3.6 Experimental flow chart

3.7.1 Sample collection and preparation

The collection and preparation of samples is the most important stage of material analysis. Again, careful consideration needs to be taken when sampling historical materials. Additionally, minimal harm should be caused on historic structures while at the same time gathering enough material to carry out analysis successfully. In terms of the National Heritage Resources Act (Act No. 25 of 1999), Section 34, a permit must be obtained from the relevant heritage authority before altering any heritage site. A permit was granted by SAHRA to collect hardened cementitious material samples. A maximum of 1 kilogram of representative samples from existing heritage buildings constructed between the 1600s and 1800s in South Africa; Western Cape (Robben Island, The Castle of Good Hope) were collected, where possible. The materials incorporated three different mortar types sampled from different locations of a building (floor, plaster/renders and pointing, joints/bedding).

The choice to use a range of materials from different centuries was in order to evaluate any trends regarding heritage cementing materials used over the years. A sampling of these materials was carried out using a chisel and a small hammer, with the main target being the inner (original) material. This was conducted to avoid sampling of materials which had been altered by either weathering or pollutants in the atmosphere. The sample information consisted of details similar to those in a data sheet used by Ngoma (2009), as stated in Section 2.4. The information obtained during sampling is summarised in Table 3.3, showing details of the samples and their age.

The sampling locations in both the Castle of Good Hope and Robben Island are noted in Figures 3.2 and 3.7, respectively. A total of 9 samples from the Castle (old kitchen building) and 15 from Robben Island were investigated. On Robben Island, materials were collected from the locations that were scheduled for maintenance by SAHRA, as indicated in Figure 3.7. The materials were collected from three of the five building precincts namely: The old maximum security prison and old school (pre-primary school), which were scheduled for restoration by the appointed consulting firm.

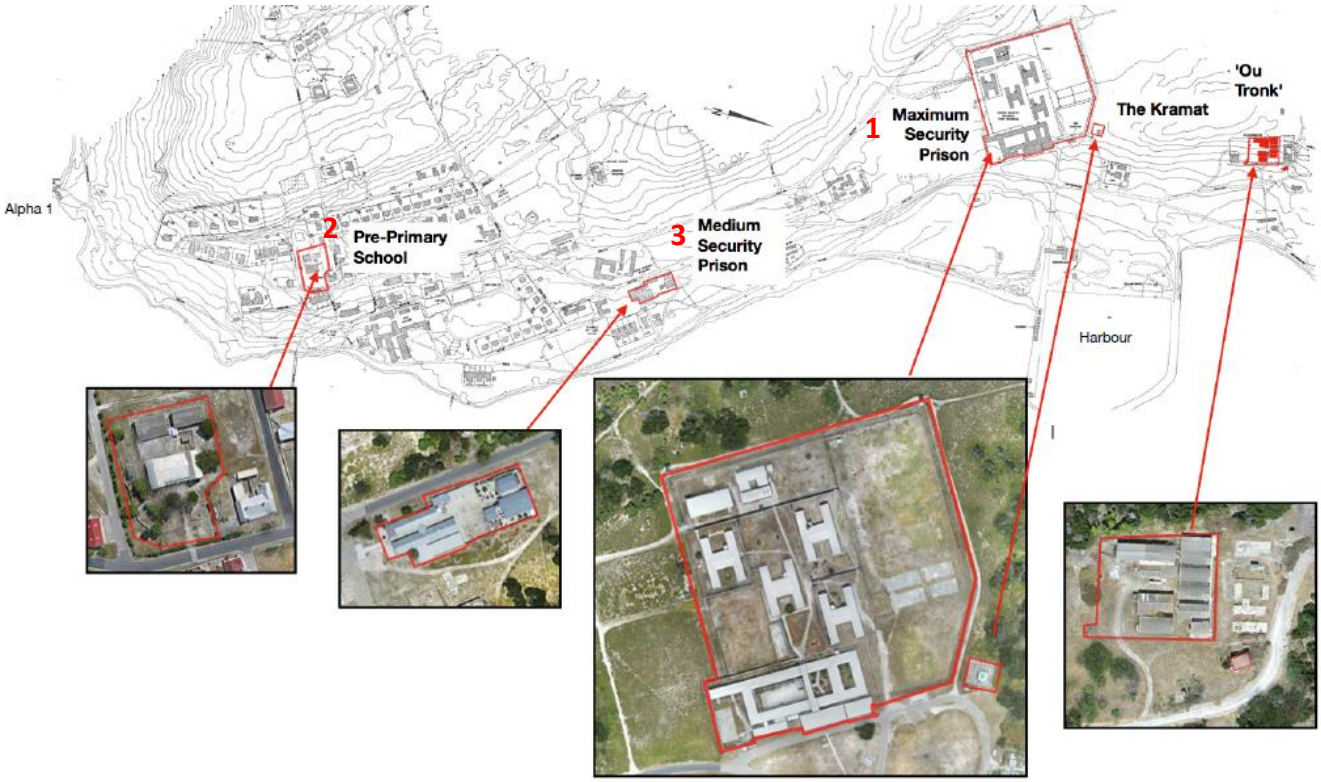


Figure 3.7 Sampling locations (Conditional Assessment of Buildings on Robben Island, 2018)

As the materials came in a chip (intact hard) form, careful consideration while breaking them down into powder was taken. Hard breaking would break up larger particles by mistake and affect particle distribution and results of the further analyses, which depend on particle sizes. After sampling of materials was completed, the tests to determine aesthetic, physical, mineralogical and chemical properties were performed, as outlined in sections 3.4.2 to 3.4.5. The following codes were used for ease of material reference: SK - Sample from Kitchen, MX - Maximum Security Prison, MD - Medium Security Prison and PS - Primary School.

3.7.2 Aesthetic characterisation

The characterisation of heritage mortars followed a similar procedure recommended by Van Balen *et al*. (1999); Palomo *et al*. (2014) which identifies the unknown sample components. The procedure consists of the study of the proportions of both the aggregate and the binder. According to Groot *et al*. (2004), the characterisation of historic mortars for restoration purposes provides the quantities of aggregates and binder that help identify all necessary data to produce a compatible or at least matching repair material. This is undertaken to avoid harm to the original material and to preserve the aesthetic effect created by a specific colour or texture. In addition to aesthetic characterisation, it is always necessary to conduct further investigation on other properties to complement what the colour and texture of the mortar truly represent.

Visual investigation - Colour

The colour and style of pointing have significant influence on the aesthetic properties of a masonry structure (RILEM, 2016). Before carrying out the analysis of the sampled material, a general visual investigation on-site was carried out. This assesses the materials' visual properties as they appear on the structure itself. From this survey, it is possible to distinguish between the original and the repair material without even carrying out any laboratory experiments. The colour difference helps track down the possible application of the modern material on the structure. It can be clearly seen on the Castle's wall section in Figure 3.3 and the island's pointing and render mortar in Figure 3.5. The difference seen is a clue for the investigator in terms of the possible difference in materials, though this hypothesis, needs to be tested with further analysis of the materials in the laboratory facility. A close look at the colour of the material gives beneficial information on the expected material components. As shown in Figure 3.8, the colour provides details on the prediction of the used binder (lime or cement), the aggregate type (sand or limestone), as well as possible additives. The visual investigation is thus a very useful stage of material characterisation.

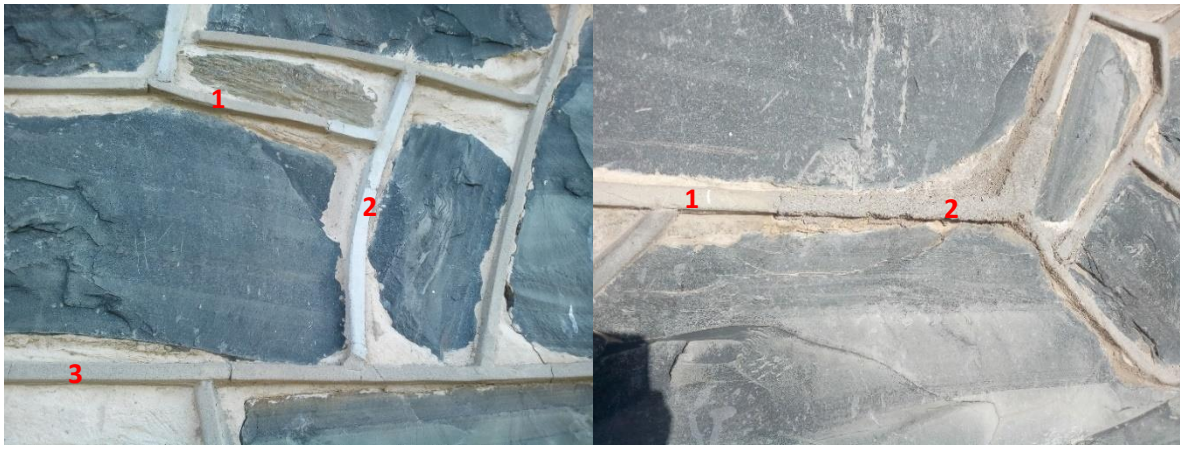


Figure 3.8 Different material colour and texture (Robben Island)

After a careful survey of the structures, and studying the pattern used on the masonry materials, samples were collected for the characterisation procedure similar to the one by Schueremans *et al*. (2011) and Bertolini *et al*. (2013). The process involves use of approximately 10 grams of mortar to carry out visual assessment by use of the naked eye and fingers to define the colour and cohesion of the mortar samples. The mortar chip is carefully assessed from the surface to the core in order to get the exact colour and texture. The colour as seen by naked eye was evaluated in accordance with the method outlined by Schueremans *et al*. (2011). The method involves using a colour scale to define the colour of the samples. It is from this test where the possible salt and chemical elements present in a mortar sample are hypothesised, though they need to be confirmed through mineralogical and chemical analysis.

In addition, the choice of further analytical techniques could be based on the colour of the material. Some tests cannot be performed on certain materials. For example, calcium-containing mortars cannot be analysed using hydrochloric acid, as the results would be inaccurate. The visual investigation formulates a relationship between the colour and the chemical composition of the material. For confirmation on the visual conclusions, confirmatory titration tests were performed on the samples.

Visual investigation - Cohesion

This characterisation procedure ensures the prediction of the material composition without even collecting the material to analyse at the laboratory. Just visually inspecting the material provides much information on the composition of such mortar. The roughness would denote a high content of aggregates in the mortar, while a smooth mortar surface indicates high binder content. The cohesion test procedure adopted from the work of Bertolini *et al*. (2013) was used for testing the samples' cohesion. The test was conducted by attempting to break the sample with human fingers while assessing its behaviour as gentle pressure is applied to it. Bertolini *et al*. (2013) mention that mortars exhibit different behaviours when pressure is applied to them, namely: very tough - does not break; tough - breaks without crumbling; friable - crumbles and incoherent - inconsistent to the touch.

Microscopy analysis

The procedure by Schueremans *et al*. (2011) and Bertolini *et al*. (2013) was referenced for a detailed investigation of the samples at the laboratory using a stereo microscope. The test analyses the colour, binder, and aggregate particle size of polished samples of approximately 5 cm^3 or smaller under a stereo microscope. Polished mortars are placed under a microscope before grinding to detect the special features at relatively low magnifications (10x to 70x). Traces of seashells of varying particle sizes and organic content were also visible under a microscope. High-resolution images of magnified mortars showing these features were captured (shown in Section 4.2).

Table 3.3 Sample data sheet & aesthetic properties

Sample ID	Age	Sample type	Sample description	Cohesion	Description
Castle of Good Hope					
SK1	1666	Plaster	Whitish mortar with black and reddish aggregates	Tough	Predicted shell and lime-based plaster material
SK2		Plaster	Whitish grey mortar with finer black grey aggregates	Tough	Predicted shell and lime-based plaster material
SK3		Plaster	Whitish yellow mortar with black grey aggregates	Tough	Predicted shell and lime-based plaster material
SK4		Floor	Whitish grey mortar with dense black grey aggregates	Very tough	Predicted shell and lime-based floor material
SK5		Bedding /joints	Whitish mortar with black grey aggregates	Tough	Predicted shell and lime-based bedding material
SK6		Bedding /joints	Cream white mortar with black grey aggregates	Tough	Predicted shell and lime-based bedding material
SK7		Bedding /joints	Light yellowish soil mortar	Friable	Material did not show the presence of binder
SK8		Bedding /joints	Reddish brown soil mortar	Friable	Material did not show the presence of binder
SK9		Bedding /joints	Yellowish orange soil mortar	Friable	Material did not show the presence of binder
Robben Island					
MX1	1700	Raised pointing	Grey material	Very tough	Material detached from white surface bedding. Suspected to be repair material having high cement content. It is smooth/fine
MX2		Joints/bedding	Whitish material	Tough	Predicted original lime-based material
MX3		Joints/bedding	Whitish to grey material	Tough	Predicted original lime-based material
PS1	Before/around 1846	Rendering	Dark grey with 2nd layer of whitish grey material attached to it	Tough	Displays two layers of different material. Inner glittery layer
PS2		Rendering	Dark grey with whitish grey layer attached to it	Very tough	Detached from the surface with a layer of different material attached to it
PS3		Rendering	Dark grey mortar	Very tough	Cement-like based mortar detached from the original surface
PS4		Plaster	Whitish grey with dense white & grey shell fragments. (shells medium crushed and slate traces)	Very tough	Shell and lime-like based plaster material

Sample ID	Age	Sample type	Sample description	Cohesion	Description
PS5		Rendering	Grey mortar with slate particles	Very tough	Seems to be cement based material with rough texture
PS6		Plaster on steps	Greyish with brown aggregates and traces of white finely crushed shells	Very tough	Traces of coarse aggregates other than sand visible
PS7		Plaster	Grey mortar	Very tough	Seems to be cement based material with medium texture. The chip was detached with part of the bedding material
PS8		Floor	Grey mortar with large stone(slate) aggregates	Very tough	Has rough texture, it is unclear whether it is cement or lime based
PS9		Plaster on steps	Whitish grey with traces of fine crushed shells	Very tough	Seems to be lime-based material
PS10		Rendering	Whitish grey with white medium sized shells & slate aggregate (not dense)	Very tough	Seems to be lime-based material. Has smooth texture
PS11		Rendering	Whitish grey with white medium sized shells (very dense)	Very tough	Has smooth texture
PS12		Floor	Grey mortar with medium crushed slate aggregates and minor traces of white shells	Very tough	The material has medium to rough texture and shows traces of shell fragments.

3.7.3 Physical characterisation

Physical characterisation of historic mortars acts as a guide for the analysis of aggregates (sand). Apostolopoulou *et al*. (2017) mention the need to consider the effects of weathering and possible alterations on materials throughout the years, which could impact on the exact gradation curves of the original materials. It was concluded that the restoration aggregates gradation curve must be as close to the original as possible, regardless of the possible weathering on original materials.

The same samples used for identifying aesthetic properties were tested for the physical properties. Since the sample sizes were limited, only non-standard material quantity (300 grams per test) was used for the sieve analysis test. The test was only carried out on some samples, as it was not possible to get more than 100 grams of certain materials.

Granulometry/sieve analysis

The granulometry of aggregates is considered a key factor in determining the quality of the mortar mix (Borges *et al*., 2010). The pattern in which soil particles are distributed in a mortar and their relative sizes are the indicators of mortar engineering properties which include: load capacity and mechanical resistance, mortar cohesion and hardness, hydraulic conductivity, shrinkage, compressibility and shear strength (Arizzi & Cultrone, 2013; Cayme & Asor, 2017). The authors further emphasise the role played by a well-graded aggregate on the porosity and retraction of mortars which gives homogenous and consistent mortars. It is to be noted that the engineering behaviour of mortars cannot be based entirely on the gradation of aggregates, but also on other factors such as effective stress, stress history, mineral type, plasticity and geologic origins of aggregates among others. The materials from the island could not be tested using this method, due to restrictions in sampling quantity. The granulometric analysis was performed with reference to Schueremans *et al*. (2011), using the ASTM D422-63 Standard as a guide. The procedure is as follows:

- Oven drying the gently ground mortar samples to obtain the dry sample weights without any microscopic moisture.
- Passing the dry powder mortar samples through a nest of sieves and agitating the sieves.
- Taking out the material retained on each sieve to a dish and,
- Using an analytical scale (close to 0.01 g) to weigh the amount of material retained on each of the sieves placed in ascending order of size.
- The cumulative percentage of materials passing the sieves was calculated and,
- Plotted on a graph of percentage passing versus sieve size, as shown in Section 4.2.

3.7.4 Mineralogical characterisation

After conducting physical tests on samples, complementary qualitative analysis of salts was further carried out to identify the sulfates, chlorides, nitrites and nitrates present in mortar samples. The microchemical tests used in this study required little expertise and were considered inexpensive, despite providing results in a few minutes. The test uses simple chemical reactions to identify the salts by use of reagents and indicators.

Qualitative analysis – Water soluble salts

Qualitative analysis of water-soluble salts is said to help identify the type and cause of deterioration on the masonry (Teutonico, 1988; Borrelli, 1999). The analysis includes identification of the ions such as the sulfates (gypsum, magnesium, ammonium and calcium sulfates), chlorides, nitrites, nitrates and carbonates on additives, aggregates and a binder (material passing 0.063 sieve). The test only provides the range in concentration of significant salts in a sample. It does not give details on salts contents. Teutonico (1988) stated that cement contains other soluble salts besides the ones mentioned here. These salts get washed onto the original mortars and cause crystallisation or efflorescence that is said to be destructive to the masonry. This is one of the shortcomings that make cement unsuitable for application on historic lime-based structures.

The laboratory procedure for identifying the salts was adopted from Teutonico (1988) and Borrelli (1999) and required use of only 0.1 g of the sample. For this study, the binder was analysed separately from the aggregates. The preparation of the sample prior to testing is as follows:

- The sample (0.1 g) is finely ground and divided into two parts in test tubes (for testing and for control).
- Add 5 ml distilled or deionised water to the first test tube which is being used for analysis and shake gently to dissolve the sample.
- Allow the insoluble part of the sample to settle at the bottom of the test tube. A few minutes is required for the solution to become clear or use a fine filter paper and a small funnel to obtain the clear filtrate.
- The insoluble residue is conserved for analysis of carbonates.
- The clear filtrate is divided into 4 equal parts in different test tubes for the analysis of salts.

The qualitative analysis for the salts is outlined below.

Qualitative analysis of Sulfates (SO_4)

Take the filtrate in test tube 1.
Add 1 or 2 drops of dilute hydrochloric acid (HCl 2N) and 1 or 2 drops of a 10% solution of barium chloride ($BaCl_2$).
If the reaction gives a white precipitate of barium sulphate ($BaSO_4$) that is insoluble in dilute nitric acid, the sample contains sulfates. It is advised to gently stroke the test tube walls with a glass rod to help the nucleation of crystals leading to the formation of the precipitate. The chemical reaction is shown in Equation 3.1.

$$SO_4^- + BaCl_2 \longrightarrow BaSO_4 + 2Cl^- \quad (3.1)$$

Sulfate in solution — white precipitate

Qualitative analysis of Chlorides (Cl^-)

Take the filtrate in test tube 2.

Add 1 or 2 drops of dilute nitric acid (HNO_3 2N) and 1 or 2 drops of a solution (0.1 N) of silver nitrate ($AgNO_3$).
If the reaction gives a whitish-blue precipitate of silver chloride (AgCl), the sample contains chlorides. The chemical reaction is shown in Equation 3.2.

$$\underset{\text{Chloride in solution}}{Cl^-} + AgNO_3 \longrightarrow \underset{\text{white-blue precipitate}}{AgCl} + NO_3^- \qquad (3.2)$$

Qualitative analysis of Nitrites (NO_2^-)

Take the filtrate in test tube 3.
Add 1 or 2 drops of Griess-Ilosvay's reagent and 1 or 2 drops of dilute acetic acid (CH_3COOH 2N).
The reaction takes some time to take place (10 minutes or more), so allow sufficient time to characterise the colour change as either visible or negative.
If the reaction gives a more or less intense pink colour, this indicates the presence of nitrites.

Qualitative analysis of Nitrates (NO_3^-)

A. If the test for nitrites was negative (did not show the pink colour), add a small quantity of zinc powder to the same solution (in test tube 3).
The reaction takes some time to take place (10 min or more), so allow allow sufficient time to characterise the colour change as either visible or negative.
If the test yields a pink colour after the addition of zinc powder, the sample contains nitrates.

B. If the test for nitrites was positive (showed pink colour in test tube 3).
Take the filtrate in test tube 3.
Add 1 or 2 crystals of sulfamic acid (HSO_3NH_2 or H_3NSO_3) to eliminate the nitrites. Keep adding as necessary until the pink colour disappears, though excess sulfamic acid should be avoided.
Add 1 or 2 drops of acetic acid (CH_3COOH 2N) and 1 or 2 drops of Griess-Ilosvay's reagent and a small quantity of zinc powder.
If the reaction yields a more or less intense pink colour, the sample contains nitrates.

Qualitative analysis of Carbonates (CO_3^-)

Take the insoluble residue obtained from filtration of the solution of the crushed sample.
Add 1 or 2 drops of concentrated HCl to the test tube.
If the reaction yields the bubbles of gas (CO_2), the sample contains carbonates. The chemical reaction is shown in Equation 3.3.

$$\underset{\text{Insoluble}}{CaCO_3} + 2HCl \longrightarrow \underset{\text{soluble}}{CaCl_2} + H_2O + CO_{2(gas)} \qquad (3.3)$$

The procedure for the qualitative analysis of salts is depicted in Figure 3.9.

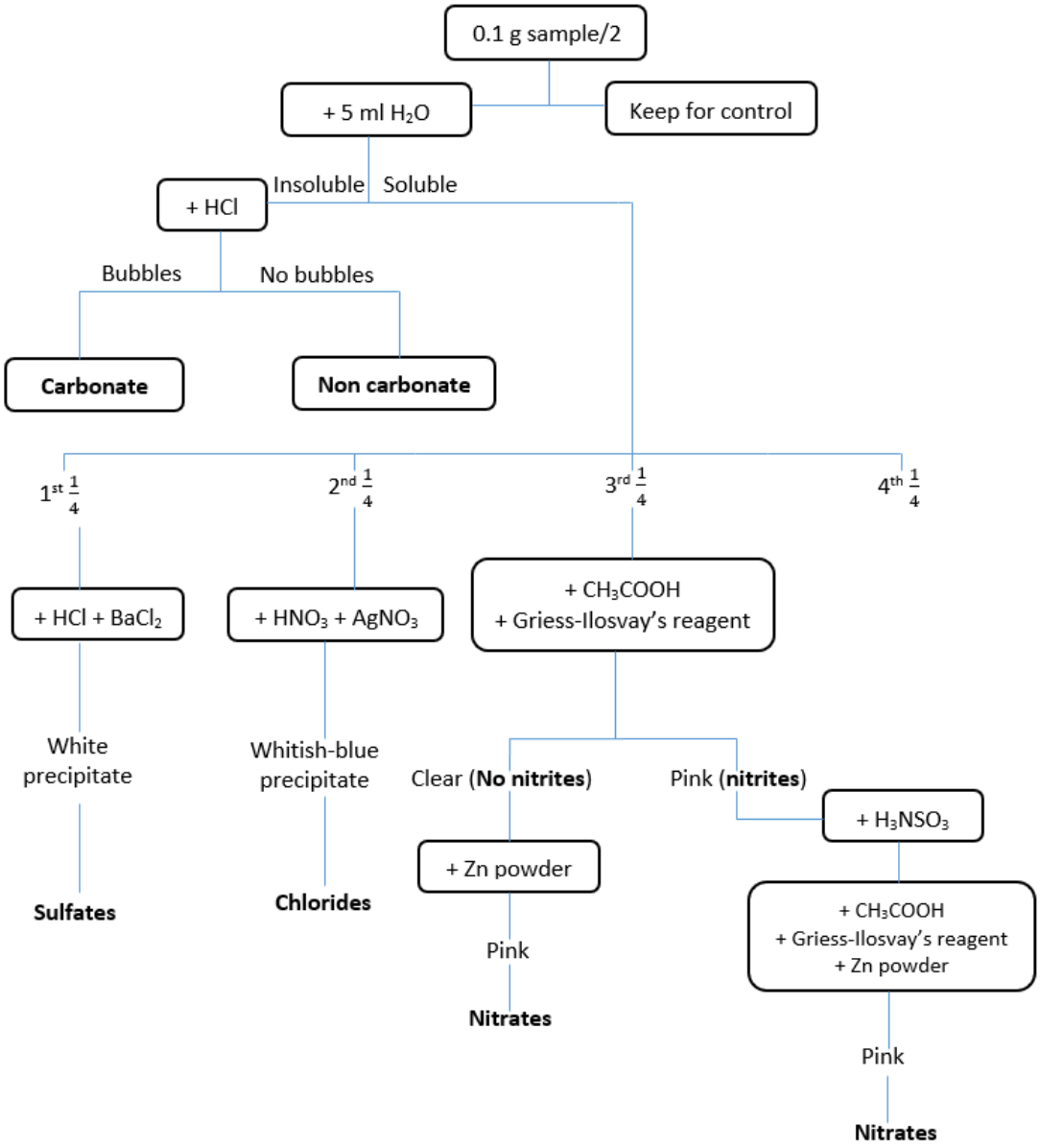


Figure 3.9 Soluble salts analysis (Teutonico, 1988; Borrelli, 1999)

3.7.5 Chemical characterisation

After carrying out mineralogical characterisation, Middendorf *et al*. (2005) suggest confirming the results with a chemical analysis. Starting with mineralogical analysis provides information on aggregate solubility in acid and the binder hydraulicity; hence, determining the separation technique to use in chemical analysis. Chemical characterisation helps identify the element oxides present in the original mortar. The chemical composition of the mortars was determined through an experimental procedure involving acid dissolution/separation of the binder from the aggregate and analysing the filtrate containing oxides of elements present in the sample. The aim was to identify the presence of calcium, magnesium, iron, silica and aluminium oxides for calculation of the cementation index.

The elements play a major role in determining the cementation and hydraulicity indices. The first set of experiments was used to determine the binder-to-aggregate ratio of collected samples using acid dissolution method. The method involves dissolution of samples in deionised water and into volume ratio of diluted hydrochloric acid. The procedure of obtaining a filtrate was similar to the one standardised by Middendorf *et al*. (2005b) in Figure 2.12. It is worth noting the differences in opinions among researchers as to what amount of HCl acid should be used. There is no standard as far as the ratio and temperature of HCl are concerned.

Wet chemical separation – binder/aggregate ratio

The method disaggregates the binder from the aggregate by dissolving the binder in a sample using a diluted acid. The behaviour of the sample, when attacked with an acid, gives an idea of the possible binder type used in the original production of the mortar. As stated by Middendorf *et al*. (2005b), some assumptions are usually made during the binder dissolution. The most significant assumption is that the binder dissolves in acid while the siliceous aggregate remains unharmed. However, in cases where the sample contains the acid-soluble silica, the mineralogical analysis intervenes in terms of identifying the possibility of salt traces in a sample. The presence of silica does not provide conclusions on the use of a hydraulic binder for production of the original material: the silica could be from brick dust, pozzolan/volcanic ash cement, flint or clay minerals. Therefore, the mineralogical analysis helps trace the origin of the silica. As stated by Middendorf *et al.* (2005b), assigning a point of origin for the silica based on mineralogical analysis requires some background knowledge of the origin of the heritage building's construction materials. The use of the acid dissolution technique requires knowledge of aggregate solubility in hydrochloric acid prior to analysis of heritage cementing materials of unknown components (Middendorf *et al*. 2005b).

There are two methods applicable for the determination of the binder-to-aggregate ratio. They differ based on the aggregate solubility in acid. Alvarez *et al*. (1999) and Middendorf *et al.* (2005b) report on the two methods. For the analysis of materials containing aggregates with insoluble aggregates in acid, the powdery sample is attacked with acid directly after crushing and drying. The latter method requires mechanical separation of the binder from the partially or completely acid-soluble aggregate using the sieving process. According to Alvarez *et al*. (1999); Middendorf *et al*. (2005b) and Cayme and Asor (2017), the mechanical separation gives material passing 75 or 63 µm sieves, which are assumed to be enriched with the binder, while the retained material is the aggregate.

Acid digestion for the binder-to-aggregate ratio and element oxides

The determination of the binder-to-aggregate ratio was carried out using the procedure in Figure 3.10.

Determination of b/a through binder digestion

Dry the crushed sample at 40°C and take a constant 1st weight.
Put the sample in a 200 ml beaker with addition of 40 ml of 2N HCl and stir for 5 minutes.
Allow gas evolution to decay, then,
Heat the sample quickly until weak boiling for 5 minutes.
Filter the hot solution through a medium textured paper.
The filtrate is saved to be used for analyses of calcium and magnesium oxides.

Wash the residue with distilled hot water (at least three times).
Dry the residue and weigh it again (2nd weight).
For the calculation of the binder-to-aggregate ratio of the mortar, Equation 3.4 was used.
The soluble content is considered the acid-soluble binder, while the insoluble part is considered acid-insoluble aggregate.

$$b/a = \frac{(W_1 - W_2)}{W_2} \quad (3.4)$$

Where:
b/a - binder-to-aggregate ratio
W_1 - First weight of the original sample
W_2 - Second weight after acid dissolution and washing with distilled water
W_3 - Third weight after boiling in saturated sodium carbonate, washing with HCl and water

Determination of the soluble SiO_2 soluble silica (C-H-S) in the binder

Boil the insoluble part (aggregate) with 30 ml of saturated sodium carbonate (Na_2CO_3) solution for about 4 minutes.
Filter and wash the residue with HCl 4N.
Rewash it at least three times with distilled water.
Dry the insoluble content at 40°C until mass constancy is reached (3rd weight).
Because of the fact that by using saturated Na_2CO_3 solution, the CSH-phases will be cracked, the insoluble lost weight, which reflects the content of hydraulic components, calculated using Equation 3.5.

$$\%\ \text{Vol.}\ SiO_2 = \frac{(W_2 - W_3)\ x\ 100}{W_1} \quad (3.5)$$

Determination of the soluble Fe_2O_3

Transfer 10 ml of filtrate from the determination of soluble silica into a 200 ml beaker.
Bring the solution to a pH = 2 ± 0.1 (use diluted HCl).
Heat to 50-55°C and add 1 ml of the indicator (dissolve 5g of sulphosalicylic acid in 100 ml of distilled water).
Titrate with 0.025 M EDTA until colour changes from pink-violet to yellow.
Preserve this solution for the subsequent determination of Al_2O_3 using Equation 3.6.

$$F_s = 0.998\ x\ V_{EDTA} \quad (3.6)$$

Where:
F_s - the quantity of soluble iron
V_{EDTA} = volume of EDTA solution used for the titration, expressed in % (mass/mass) of Fe_2O_3.

Determination of the soluble Al_2O_3

Add to the solution previously used for the soluble iron oxide determination, 3 ml of 0.025 M EDTA.
Bring to pH = 5 ± 0.1 by using a solution of hydrated potassium hydroxide (4N KOH).
Heat the solution near boiling point for about 3 minutes.
The beaker should be covered. Cool the solution to a temperature of 25-30°C and add 1 to 3 drops of an indicator (xylenol orange).
Titrate over a magnetic stirrer with a 0.025 M zinc sulphate solution until colour changes from yellow to pale pink.
The amount of Al_2O_3 (%) can be determined by using Equation 3.7.

$$Al_2O_3 = \left(3 - V_{ZnSO_4} \bullet f_{ZnSO_4}\right) \bullet 0.632 \quad (3.7)$$

Where:
V_{ZnSO_4} - volume of $ZnSO_4$ solution used for the titration of EDTA excess in the soluble Al_2O_3 determination
f_{ZnSO_4} - molarity factor of the $ZnSO_4$ solution which is 0.025 M

Determination of soluble CaO

Take 5 ml of solution from the flask containing the filtrate of the determination of unreactive silica (filtrate from b/a ratio determination) and put it into a beaker (200 ml).
Dilute with 15 ml of distilled water.
Add 5 ml of TEA (triethanolamine) and bring to pH 12.5 ± 0.2 by using a KOH solution (4 N).
Add the reagent methylene blue and titrate by using a magnetic stirrer with 0.025 M EDTA until the colour changes from blue to yellow.
The amount of CaO is determined using Equation 3.8.

$$CaO = 1.402 \text{ x } V_{EDTA.Ca} \quad (3.8)$$

Where:
$V_{EDTA\bullet Ca}$ - volume of EDTA 0.025 M solution used for titration of soluble calcium

Determination of soluble CaO and MgO

Take 5 ml of solution from the flask containing the filtrate of the determination of unreactive silica (filtrate from b/a ratio determination) and put it into a beaker (200 ml).
Dilute with 15 ml of distilled water.
Add 0.5 ml of TEA (triethanolamine) and bring to pH 10.1 ± 0.1 by using ammonia solution (NH_3).
Add the indicator methylene blue and titrate by using a magnetic stirrer, then add a 0.025 M EDTA solution until colour changes from blue to yellow.
The percentage of soluble magnesium (M_{Mg}) is shown in Equation 3.9.

$$M_{Mg} = 1.008 \left(V_{EDTA\ Ca+Mg} - V_{EDTA\ Ca}\right) \quad (3.9)$$

Where:

$V_{EDTA\ Ca+Mg}$ - volume of 0.025 M EDTA used to titrate the soluble Ca and Mg

$V_{EDTA\ Ca}$ - volume of 0.025 M EDTA used to titrate only Ca

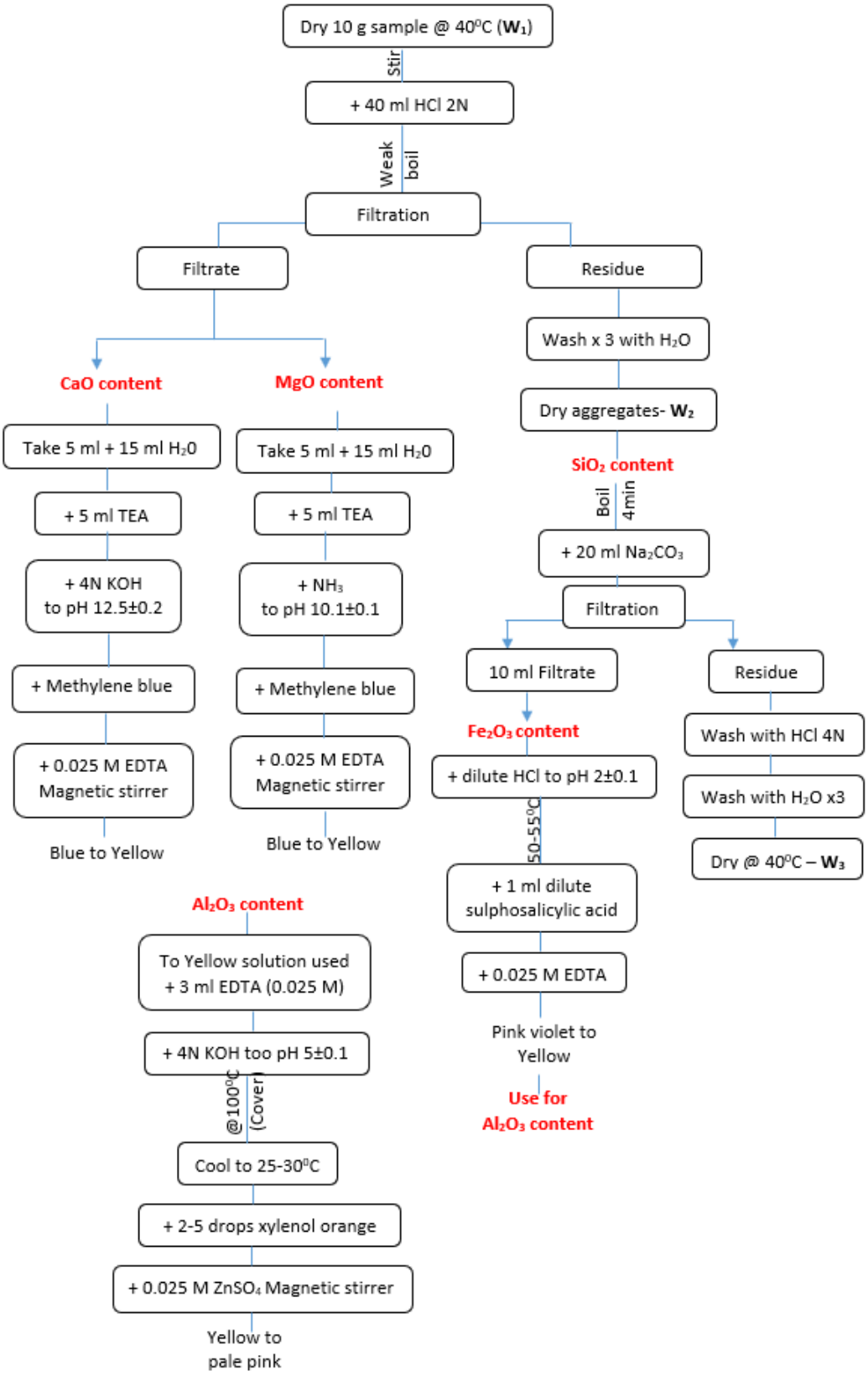


Figure 3.10 Determination of the element oxides (Middendorf *et al.*, 2005b)

Test limitations

The titration method is considered the most cost-effective analytical technique used for both mineralogical and chemical characterisation of historic cementing materials. However, this method, like any other experimental technique has limitations, which impact on its use under certain conditions. One of the major limitations is the application of the wet chemical analysis for analysis of calcareous mortars that use limestone, shell, or coral sands, as these aggregates will dissolve in the acid (Alvarez *et al*., 1999). For the current study, mechanical separation of calcareous material prior to testing was conducted in order to analyse the binder separately from the aggregate.

The analysis gets tricky if different binders (lime and cement) or aggregates were used for the mortar production. This was not the case in this study, as only one binder type was used per mortar mix, as evident from the visual analysis.

As reported by Middendorf *et al*. (2005b) and Ngoma (2009), the wet chemical analysis procedure has not yet been standardised on an international scale, leading to the use of different methods depending on researcher discretion and different locations. This has brought up some debate as to what amount of acid to use, the concentration required, and the temperature for the acid dissolution. A number of different authors such as Alvarez *et al*. (1999); Van Balen *et al*. (1999); Middendorf *et al*. (2005b) voice their opinions on the concentrations of the acid used in dissolution. Besides the argument in the acid use, there are differences in use of chemical indicators for the titration method. For example, Alvarez *et al*. (1999) used barium diphenylaminosulphonate and ditizone as indicators for the titrations of iron and aluminium oxides, while Middendorf *et al*. (2005b) proposed use of sulphosalicylic acid and xylenol orange as indicators. A similar difference was spotted between Cayme and Asor (2017); Middendorf *et al*. (2005b) for the quantification of calcium. The former titrated the mortar using potassium permanganate, while the latter titrated using methylene blue as an indicator.

3.7.6 Health and safety

The use and handling of chemicals require careful attention, as working with acids and reagents is associated with a degree of risk, and could cause accidents to users and people on site. These accidents could be fatal, especially where acids and chemicals are involved. For the purpose of the mineralogical and chemical characterisation of heritage cementing materials using the procedures above, safety training was provided prior to experimental testing. The training was based on the CPUT safety policy, rules and regulations. It covered the major aspects as follows:

- Before the start of a laboratory work, the locations of the emergency tools such as first aid kit and fire extinguisher must be well established. This includes the knowledge of emergency procedures and contacts in case of accidents at the laboratory.
- The laboratory work must be completed by a minimum of two people who are both familiar with the CPUT safety regulations.
- Studying the material safety data sheet (MSDS) for each chemical is a requirement, as all the chemicals need to be regarded as dangerous.
- The availability of a source for running water in case of spillage of chemicals on the skin I essential. Preventative measures must be prioritised, whereby direct contact with the skin needs to be absolutely avoided.

- The following activities are strictly prohibited in the lab: running, sleeping, eating, drinking from laboratory glassware and smoking.
- The use of safety attire which includes the laboratory coat, safety glasses, protective shoes and latex gloves among others, is mandatory.
- Beware of hot surfaces and glassware, as no visual difference between hot and cold is evident.
- Handling of chemicals should be undertaken with all due caution, including the transportation and handling of bottles and containers to avoid possible spillage.
- Mixing and diluting of acids should only occur using *the acid to water* rule.
- Labelling of glassware to avoid mix-ups is strictly advised.
- To avoid contamination, excess acid is disposed of down the drain and not returned to the original container.
- Washing and rinsing of glassware must be conducted on individual apparatus, not all in one bath.

3.7.7 Data collection and analysis

This section presents relevant data pertaining to physical, mineralogical and chemical properties of the binder and aggregates in historic samples. Their interpretations and analysis are also discussed within the scope of this research.

Data collection

The relevant data pertaining to the behaviour of samples collected from historic buildings from the 17th to the 19th century is presented in this section. The tests performed produce data as follows:

- Binder and aggregate colour, texture and particle sizes.
- The presence of dominant salts in a mortar.
- The chemical composition of the mortar constituents in the form of oxides and their approximate quantities.

Data analysis

The results obtained from the experimental procedure give the matrix of the materials used and their development over the centuries (from 17th to 19th). From these results, one can delineate a trend in materials used for both the Castle and the Island, as well as possible repairs that could have been made on these structures. The data obtained is a guide to deciding on the methodology for analysis of other heritage structures based on the following key aspects:

- *Cost* – the cost for conducting the experiment in terms of expertise, equipment and laboratory expenses.
- *Time* – the duration the test takes with consideration to urgency for characterising with the aim of restoring.
- *Complexity* – knowledge of the analytical techniques alone is not sufficient, a misunderstanding of the results may cause a confusion in terms of material composition (Feilden, 2003). Hence, the choice of analytical procedures must take into consideration the simplicity of the test for future use by restorers.

- *Convenience/access* – The availability of the test equipment and material.
- *Material quantity*- How much material is required to conduct the test.
- *Usefulness/parameters obtained* – How results will help restorers conduct repairs.

Experimental errors

The possible errors include slight differences on estimated aggregate, content due to possible dissolution of some parts of the aggregate in acid during chemical analysis. The difference is, however, ignored for the purpose of this study. The other possible error is the effects that weathering, and aging have on the samples. This could lead to material properties which differ slightly from the original. However, the aim is to get to as close as possible, to the properties of the original material, accepting the possibility of alterations due to weathering factors. Human error during testing, especially during the separation of aggregates from binder, are also considered a factor that could lead to inaccuracy.

3.7.8 Instrumentation and material used

The samples were tested using the equipment from CPUT laboratories with the instrumentation and apparatus mentioned in this section. All the equipment and instruments were calibrated prior to use.

Instrumentation and apparatus

The apparatus listed below were employed to achieve the aim and objectives of this research during the experimental work. These instruments were calibrated according to specified standards before testing and serviced in accordance to manufacturer recommendations.

Sampling equipment: Chisel and hammer, sample bags and tags/labels, pen and marker.

Physical and aesthetic testing equipment included: colour scale, optical hand-held microscope, glass plates.

Mounting and polishing equipment included: jars, universal sieves, mortar and pestle.

Weighing and sample preparation instruments: analytical balance (0.01 g precision), weighing scale, petri dishes, sample bottles, drying oven/desiccators, sample labelling stickers.

Chemical and mineralogical analysis apparatus: glass beakers, glass funnels, filter paper, burette (including stand, rings, clamps and pump), volumetric pipette, microlitre pipette and tips, spatula, measuring cylinders, thermometer, burner, pH meter, boiling flasks (Florence flasks, Erlenmeyer or conical flasks), volumetric flasks, test tubes, tongs and racks, stop watch, test tubes, porcelain crucibles, wash bottles, sample bottles, magnetic stirrer.

Personal safety equipment: safety googles, latex lab gloves, nose masks, safety boots.

Testing matrix

The laboratory instruments used in this research are depicted in Figure 3.12

Figure 3.11 Testing equipment

Chemicals

In addition to the instruments shown, the following chemicals, reagents and indicators were used: distilled water, dilute acetic acid (2N CH_3COOH), ammonia solution (NH_3), 10% barium chloride solution ($BaCl_2$), ethylenediaminetetraacetic acid (0.025 M EDTA), Griess-Ilosvay's reagent, concentrated hydrochloric acid (HCl), dilute hydrochloric acid (2N HCl), methylene blue ($C_{16}H_{18}ClN_3S$), dilute nitric acid (2N HNO_3), hydrated potassium hydroxide (4N KOH), silver nitrate (0.1N $AgNO_3$), saturated sodium carbonate (Na_2CO_3), sulfamic acid (H_3NSO_3), sulphosalicylic acid ($C_7H_6O_6S$), triethanolamine (TEA) $C_6H_{15}NO_3$, xylenol orange ($C_{31}H_{32}N_2O_{13}S$), zinc powder, zinc sulphate solution (0.025 M $ZnSO_4$). All chemical reagents were analytical grade. The preparation calculations of the desired concentrations for the acids, indicators and reagents are attached in Annexure B.

3.8 Summary

The use of incompatible materials for repair of heritage buildings has been a concern for a long time. This is usually caused by lack of knowledge and understanding of the original material properties. In order to avoid disparity in materials during repair of historic structures, there is a need to characterise the original materials using a standard methodology, as there are several methods available. The chapter has elaborated on the procedure for the selected methods in this study. The methods carried out in this study were in alignment to those indicated in Table 2.6 by various authors in locations shown in Table 2.7.

Chapter 4 Results

The results obtained from experiments conducted in this investigation are presented in this chapter. The optimum analytical techniques, such as visual investigation to test colour, the cohesion test to identify the texture of the materials, microscopic analysis for the materials composition at magnification, sieve analysis for particle size distribution and wet chemical analysis for chemical and mineralogical composition, were carried out on materials which originated from the 1600s to 1800s. The results represent the characteristics of the representative selected sections of heritage cementing materials from the Castle of Good Hope and Robben Island in the Western Cape, South Africa.

4.1 Sampling

The materials were sampled and recorded on a data sheet similar to the one used by Ngoma (2009). The details for one of the samples are shown in Figure 4.1. The recording of all major details of the samples and the conditions around the sampling location provides an understanding of some of the properties that may be observed during the characterisation of the collected materials. The results from the material analysis could be influenced by several factors, such as the method used for sampling, the sampling location, for example, the height from the floor or in some cases, the exposure to deterioration factors described in Section 1.1, and finally, the environmental conditions around the structure, which could have an impact on the actual material properties.

Sampled structure: CASTLE OF GOOD HOPE
Location: BLOCK B - KITCHEN
Age: 353 YEARS
Photo: ☑ Taken ☐ Not taken

Sample no: SK1
Sampled by: M. LOKE
Date: 21 AUGUST 2018
Reason for sampling: MATERIAL COMPOSITION ANALYSIS

Sampling method: ☑ Hammer & chisel ☐ Drill core ☐ Others (explanation):

Sampling location: ☐ Wall ☐ Foundation ☐ Roof ☐ Exterior ☑ Interior ☐ At depth: 1200 mm ☐ Others (explanation):

Function of sample: ☐ Render ☑ Plaster ☐ Joint bedding

Sample condition: ☑ Hard ☐ Soft ☐ Soft and friable

Inclusions: SEASHELLS

Sample colour: WHITISH WITH BLACK AND REDDISH AGGREGATES

Environmental conditions: INLAND WITH HUMID TEMPERATURE, CLOUDY.

Condition of associated building materials: MAROON CLAY BRICKS IN GOOD CONDITIONS

Notes: THE PLASTER MATERIAL WAS THICK (20 - 40 mm) AND TOUGH TO BREAK WHEN PRESSURE WAS APPLIED TO IT. IT DISPLAYED LIME PRESENCE.

Figure 4.1 Sample data recording (SK1)

4.2 Visual and microscopy analysis

The results from the visual investigation showed the colour of the binder matrix to be predominantly whitish, whereas the cohesion was classified as tough and very tough, except for a few samples, which were evaluated as friable. The visual investigation showed no signs of the binder presence in samples SK7-SK9, implying that these samples consisted of simply mud. The colour for the three samples did not show any traces of either lime or cement, and this could be the reason why they were crumbling. The absence of a binder makes the aggregate particles loose, for obvious reasons. All the other samples showed signs of the presence of a binder, aggregate (sand) and for some samples, the additives (seashells and slate).

The cohesion test results showed that the sand particles ranged from fine to coarse. This was concluded from the broken sample pieces and the texture of the samples. Some of the mortars from the castle and the island contained fragments of black seashells, while others had white seashells with varying sizes ranging between 0.2 and 25 mm. Figures 4.2 and 4.3 show stereo-microscopy images of polished surfaces of the mortars from the castle and island, respectively. Samples SK1, SK2 and SK3 showed similar shell distribution as well as size (between 15 and 25 mm) while, sample SK4 showed finely crushed seashells (less than 5 mm) as compared to samples SK5 and SK6 (5 to 15 mm). However, the other samples (SK7, SK8, SK9, MD1, MD2, MD3, MX1, MX2 and MX3) did not show any presence of the seashells. Materials from Robben Island indicated the presence of shells ranging between 0.2 and 10 mm in diameter.

The binder used was mostly whitish, which denotes the use of lime as a binder. The grey binder material that was visible on some surfaces areas of the structures was suspected to be a repair material. This is probably because, in these materials, the intense grey colour differed from the major areas of the structure, especially the vast area of surface pointing. It was therefore concluded to be repair materials, which were mainly cement-based rather than lime-based. The suspicion was supported by the shared restoration history of the structures.

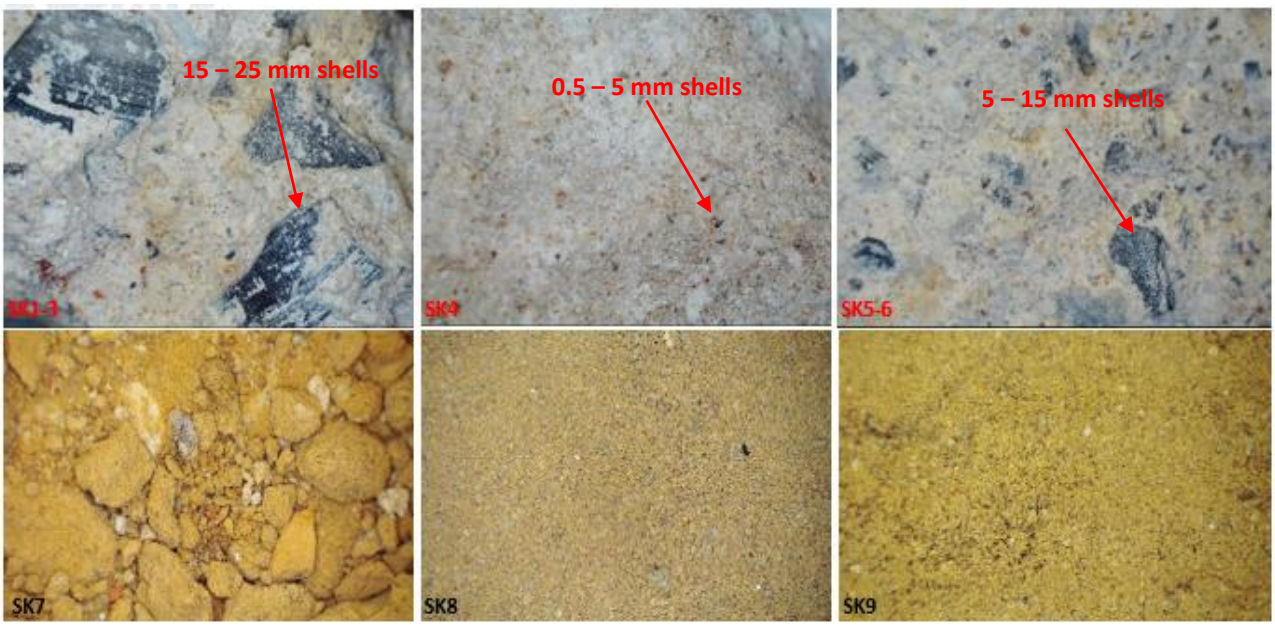


Figure 4.2 Stereo microscopy images (Castle of Good Hope)

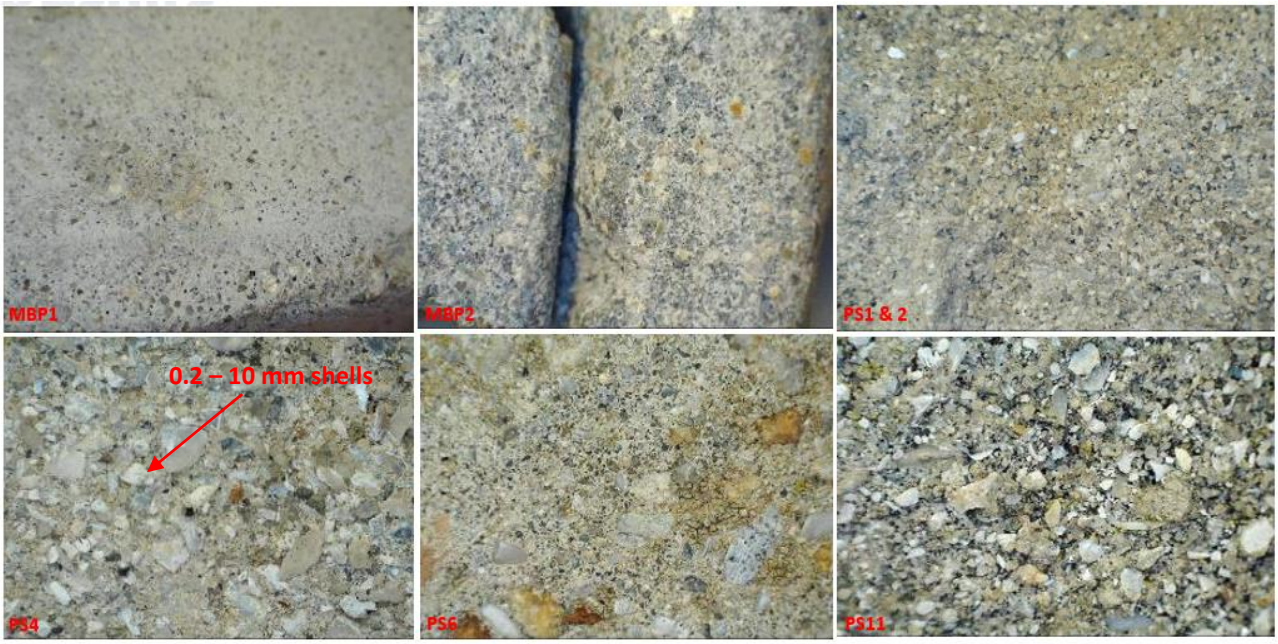


Figure 4.3 Stereo microscopy images (Robben Island)

4.3 Sieve analysis

After conducting the sieve analysis test on the castle samples with quantities higher than 1 kg, the average mass retained on the respective sieves was recorded, as indicated in Table 4.1. Due to limitations on sample quantities, samples SK4, SK6, SK7 and all samples from Robben Island could not satisfy the minimum required quantity (300 g) to perform non-standard sieve analysis, hence, those are excluded in this study. The detailed particle size distribution tables for the samples are attached as Annexure C.

Table 4.1 Castle of Good Hope particle distribution

Sieve size (mm)	Average mass retained (g)					
	SK1	SK2	SK3	SK5	SK8	SK9
9.500	0	0	0	0	0	0
6.700	3.44	0	0	0.90	0	29.12
4.750	9.11	1.48	4.19	7.17	0	8.15
2.360	54.81	13.92	39.96	62.19	0.57	17.82
2.000	23.60	13.68	20.89	21.57	1.54	7.08
1.180	23.76	23.65	26.63	24.34	5.09	5.54
0.600	62.77	98.02	73.28	59.62	21.70	35.39
0.425	21.92	37.83	21.27	13.90	41.09	21.96
0.300	20.75	36.35	24.34	22.29	52.28	22.18
0.150	34.64	48.75	35.35	35.19	102.81	79.20
0.075	27.41	17.85	28.36	30.89	56.91	44.11
Pan	17.28	8.48	25.50	21.89	16.11	27.60
Total	299.49	300.00	299.77	299.95	298.10	298.13

In order to determine if the old mortar samples were produced with good representations of each particle size, a gradation curve was plotted, as shown in Figure 4.4. The plot is derived from the sieve size percentage retained (Table 4.1).

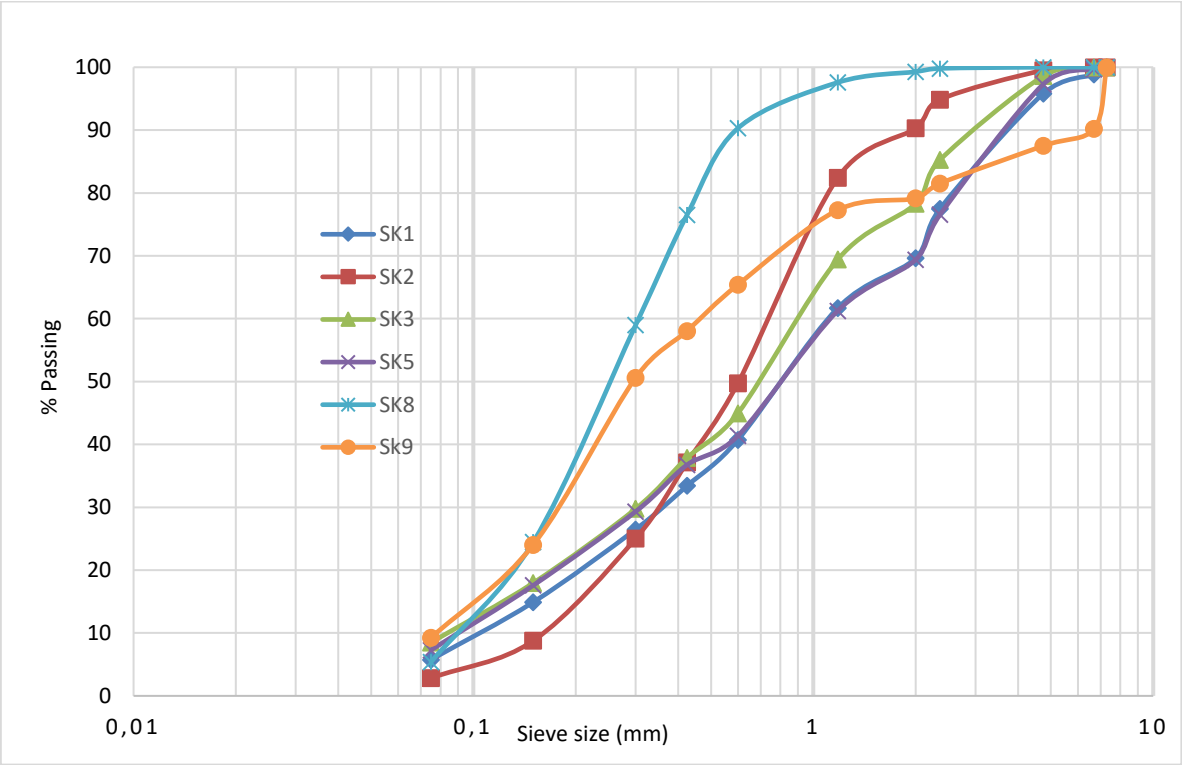


Figure 4.4 Particle distribution curves

The parameters used for the classification of soils were computed from the logarithmic plot in Figure 4.4. These parameters include the uniformity coefficient (C_u) and coefficient of curvature (C_c) which are computed from the extrapolation of the 10%, 30% and 60% materials passing the corresponding sieve sizes. The results for these parameters are presented in Table 4.2 for each mortar sample.

Coefficient of uniformity

The coefficient of uniformity (C_u) is a parameter that evaluates the consistency in the particle size in accordance with Equation 4.1 (Ontiveros-Ortega *et al.*, 2016; Cayme & Asor, 2017).

$$C_u = \frac{D_{60}}{D_{10}} \tag{4.1}$$

Coefficient of curvature

The coefficient of curvature (C_c) evaluates the variation in size of soil particles, and therefore the gradation of different particle size ranges expressed using Equation 4.2. The C_u of 1 indicates grain sizes, which are all the same (poorly graded material), $C_u > 1$ indicates grain sizes that span within a large range (uniform material). For the soil to be classified as well-graded, it must meet the criteria:
$C_u > 1 < C_c < 3$ (Ontiveros-Ortega *et al.*, 2016; Cayme & Asor, 2017).

$$C_c = \frac{D_{30}{}^2}{D_{10} \text{ x } D_{60}} \quad (4.2)$$

Where:
D_{10} – the sieve size when 10% of the particles are still being retained
D_{30} – the sieve size when 30% of the particles are still being retained
D_{60} – the sieve size when 60% of the particles are still being retained

Table 4.2 shows the values for the coefficients of uniformity and curvature and it can be concluded that the mortars from the kitchen wall section (see Figure 4.5) at the Castle were made of uniform soils except for sample 8, which showed a well-graded soil as per the C_u and C_C criteria.

Figure 4.5 The investigated section, Castle of Good- Hope-South Africa

Table 4.2 Soil classification parameters mortar sample

Sample ID	D_{10}	D_{30}	D_{60}	C_u	C_c	Material gradation
SK1	0.075	0.170	1.100	14.7	0.4	Uniform
SK2	0.075	0.170	0.730	9.7	0.5	Uniform
SK3	0.075	0.170	0.900	12.0	0.4	Uniform
SK5	0.075	0.170	1.100	14.7	0.4	Uniform
SK8	0.075	0.170	0.300	4.0	1.3	Well-graded
SK9	0.075	0.170	0.460	6.1	0.8	Uniform

4.4 Quantities of mortar components

Based on the sieve analysis conducted on the Castle materials, the respective quantities of mortar constituents are as presented in Table 4.3. The samples are generally composed of less than 10% binder. This is not the case for samples SK7 to SK9, where the visual investigation did not indicate the presence of a binder. The materials <63 µm could be the clay materials instead of the binder for samples SK7 to SK9. However, the confirmatory wet chemical analysis will provide details on these samples' chemical composition, which will confirm whether or not this is the case.

Table 4.3 Mortar estimated binder, sand and additive %

Sample ID	Estimated quantities (% mass)		
	<63 µm (Binder)	63 µm-2mm (Sand)	>2mm (Additives)
SK1	5.8	71.7	22.5
SK2	2.8	92.0	5.1
SK3	8.5	76.8	14.7
SK5	7.3	69.3	23.4
SK8	5.4	94.4	0.2
SK9	9.3	72.3	18.5

4.5 Qualitative analysis of soluble salts

The titration procedure was conducted on all the mortar samples as proposed by Teutonico (1988) and Borrelli (1999). The salts were analysed separately on materials considered to be aggregates, as well as the binders, in accordance with their particle sizes. The analysis was on materials below the 0.063 mm particle sizes which are considered to be rich in binder content (Cayme & Asor, 2017) and the aggregates retained on a 0.063 mm sieve. The qualitative results for the major soluble salts (sulfates, chlorides, nitrates, nitrites and carbonates) are shown in Tables 4.4 and 4.5 respectively with the picture gallery in Annexure D. The presence of salts was indicated using the symbols as shown below:

- \- Indicates the absence of an ion
- ± Indicates the presence of an ion at the limit of perceptibility
- \+ Indicates the presence of an ion
- ++ Indicates the presence of an ion in notable quantity
- +++ Indicates the presence of an ion as a principal component

Table 4.4 Salt contents in mortars – Dust or binder (< 0.063 mm)

Sample ID	Sulfates	Chlorides	Nitrites	Nitrates	Carbonates	Notes on carbonates
SK1	+++	++	±	-	+	-
SK2	+++	+++	+	-	+++	-
SK3	++	+++	+++	+	+++	-
SK4	++	++	+	-	+++	-
SK5	±	+++	+	±	+++	-
SK6	+	+++	-	-	+++	-
SK7	+++	++	-	-	++	-
SK8	±	++	-	+	±	-
SK9	±	++	±	-	+	-
MX1	+	++	+	±	+++	-
MX2	+	+++	±	-	++	-
MX3	+	+++	±	±	+++	-
MD1	+++	+	±	±	+++	Smell produced
MD2	+++	++	++	-	+++	-
MD3	+++	++	+	-	+++	Smell produced
PS1	-	±	±	-	++	-
PS2	±	++	±	-	+++	-
PS3	-	++	+	-	+++	-
PS4	+	++	+	-	+++	Smell, colour change from whitish to brown blackish
PS5	++	+++	+++	-	+++	-
PS6	++	+++	+++	-	+++	-
PS7	+	-	±	-	++	-
PS8	±	+	++	-	+++	-
PS9	±	±	±	-	+++	-
PS10	-	-	++	-	++	-
PS11	±	±	+	-	+++	-
PS12	+	+	±	-	+++	-

Table 4.5 Salt contents in mortars – Aggregates (> 0.063 mm)

Sample ID	Sulfates	Chlorides	Nitrites	Nitrates	Carbonates	Notes on carbonates
SK1	+	++	±	-	++	-
SK2	-	++	+	-	++	-
SK3	+	++	±	-	+++	Smell produced
SK4	-	+	±	-	+++	Smell, colour change from whitish to blackish residue
SK5	-	+	-	-	+++	-
SK6	-	+	±	-	+++	Smell produced
SK7	±	+	-	-	+++	Smell produced
SK8	±	++	±	-	-	-
SK9	±	±	+	±	+	-
MX1A	±	+++	+	-	+++	Smell produced
MX2	+	++	±	-	+++	-
MX3	±	+++	±	±	+++	Smell produced
MD1	++	±	±	-	++	Smell produced
MD2	++	+	±	-	++	Smell produced
MD3	++	+	+	-	+++	Smell produced
PS1	-	±	-	-	-	-
PS2	-	+	±	-	++	-
PS3	-	+	±	-	+++	-
PS4	-	+	++	-	+++	-
PS5	-	+	-	±	+++	-
PS6	-	++	±	±	+++	-
PS7	-	±	±	-	+++	-
PS8	+	-	±	-	+++	Colour change from grey to white
PS9	-	-	-	-	++	-
PS10	-	±	±	-	+++	-
PS11	±	±	±	-	+++	-
PS12	-	-	±	-	++	Smell, colour change from grey to greenish

4.6 Wet chemical analysis

The metal oxides used for determination of the binder type through the use of the cementation index are shown in Table 4.6. The values for the cementation and hydraulicity indices were computed using Equations 2.5 and 2.6 respectively, with the classification criteria in Table 2.4 as a basis for the binder type description. The detailed calculations for the binder-to-aggregate ratio and the metal oxides are attached in Annexures E and F respectively.

Table 4.6 Chemical analysis

Sample ID	b/a	HI	CI	Clay content	Binder type
SK1	0,53	0,15	0,30	Very little clay	Sub-hydraulic lime
SK2	0,25	0,19	0,38	Around 8%	Slightly hydraulic lime
SK3	0,57	0,18	0,20	Very little clay	Sub-hydraulic lime
SK4	0,53	0,23	0,63	Around 15%	Moderately hydraulic lime
SK5	0,56	0,18	0,46	Around 8%	Slightly hydraulic lime
SK6	0,59	0,07	0,24	Very little clay	Sub-hydraulic lime
SK7	0,23	0,42	1,18	Up to 45%	Natural or Portland cement
SK8	0,04	0,42	1,08	Around 25%	Eminently hydraulic lime
SK9	0,14	0,99	2,41	More than 45%	Natural or Portland cement
MX1A	1,06	0,37	1,01	Around 25%	Eminently hydraulic lime
MX2	0,79	0,35	0,89	Around 25%	Eminently hydraulic lime
MX3	0,79	0,36	0,89	Around 25%	Eminently hydraulic lime
MD1	1,18	0,34	0,75	Around 25%	Eminently hydraulic lime
MD2	0,74	0,44	0,86	Around 25%	Eminently hydraulic lime
MD3	0,81	0,30	0,64	Around 15%	Moderately hydraulic lime
PS1	0,61	0,15	0,55	Around 15%	Moderately hydraulic lime
PS2	0,57	0,17	0,60	Around 15%	Moderately hydraulic lime
PS3	1,34	0,40	1,17	Up to 45%	Natural or Portland cement
PS4	0,36	0,21	1,05	Around 25%	Eminently hydraulic lime
PS5	0,44	0,18	0,74	Around 25%	Eminently hydraulic lime
PS6	0,42	0,18	0,66	Around 15%	Moderately hydraulic lime
PS7	0,48	0,18	0,77	Around 25%	Eminently hydraulic lime
PS8	0,10	1,06	2,61	More than 45%	Natural or Portland cement
PS9	0,37	0,14	0,32	Around 8%	Slightly hydraulic lime
PS10	0,43	0,18	0,78	Around 25%	Eminently hydraulic lime
PS11	0,41	0,20	0,54	Around 15%	Moderately hydraulic lime
PS12	0,44	0,17	0,23	Very little clay	Sub-hydraulic lime

4.6.1 Acid insoluble aggregate

This section provides the results related to the aggregates of the materials after acid digestion. The materials which have been digested during identification of the soluble silica are considered binder, while the acid-insoluble materials are considered aggregates. The analysis assists in determining the properties of the samples whose quantity could not allow for the sieve analysis. Hence, sieve analysis during heritage mortar analysis could be replaced with acid digestion. However, since the smallest amount of sample was used, particle distribution using the gradation method would be impossible. Only visual estimations could be carried out, as shown in Table 4.7.

Table 4.7 Sample characterisation data sheet

Sample ID	Aggregate description
SK1	Fine sand with medium-to-large-sized shells with equal quantities.
SK2	White coarse sea sand with glittering particles and medium-to-large-sized shells.
SK3	Fine sand with medium-to-large-sized shells.
SK4	Fine sand dominated by finely crushed shells.
SK5	Fine sand dominated by medium crushed shells.
SK6	Fine sand with medium shells.
SK7	Very fine clayey sand.
SK8	Clayey Soil.
SK9	Clayey Soil.
MX1A	Fine to coarse sand.
MX2	Fine to coarse sand.
MX3	Fine to coarse sand.
MD1	Fine to coarse sand.
MD2	Fine to coarse sand.
MD3	Fine to coarse sand.
PS1	Fine sand with equal portion of medium coarse crushed stone.
PS2	Fine sand with an equal portion of medium coarse crushed stone.
PS3	Fine sand with lesser quantities of medium coarse crushed stone.
PS4	Fine sand with medium to large crushed stone in large quantities. Traces of crushed slate.
PS5	Fine sand with significant quantities of medium crushed stone.
PS6	Fine sand with equal portion of medium crushed stone.
PS7	Fine sand with medium crushed mortar not stone.
PS8	Fine sand with medium to large crushed stone in large quantities. Traces of crushed slate.
PS9	Coarse sand with traces of coarsely crushed stone
PS10	Fine sand with medium to large crushed stone in fewer quantities. Traces of crushed slate.
PS11	Fine sand with medium to large crushed stone in fair quantities.
PS12	Fine white sand which predominantly high quantities of medium to coarse crushed slate.

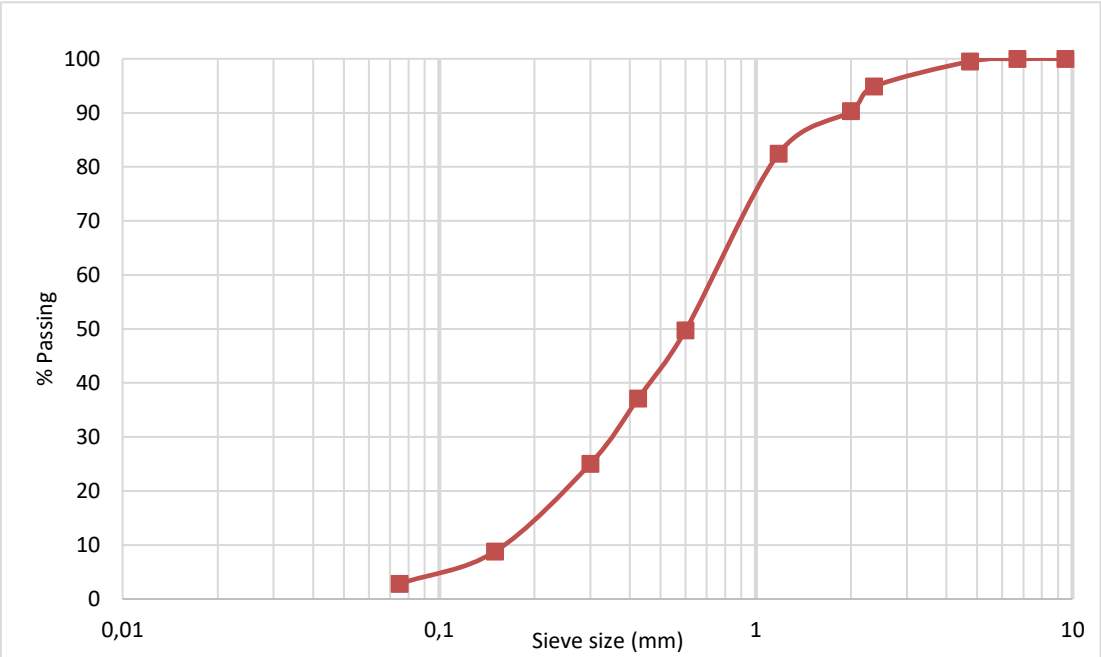
4.7 Material characterisation summary sheet

The properties of Sample 2 from the Castle of Good Hope are summarised in Table 4.8. Table 4.8 could be used as a guiding tool for a search of repair materials for the heritage building in question. The characterisation data sheet was designed to provide as much required data on the original as possible.

Table 4.8 Sample characterisation data-sheet

Sample ID: SK2

Particle distribution



Binder	Slightly hydraulic Lime
Aggregates	White coarse sand
Additives	Seashells
b/a	0.25
Soluble salts	
Carbonated material high in sulfates and chlorides with traces of nitrites.	

Colour	Whitish grey mortar with fine black grey seashells.
Notes	Uniform graded material with very low clay content and no organic content.

4.8 Conclusion

It is vital to investigate and understand the properties of the original mortars before carrying out any restoration on heritage buildings. It provides valuable information on material composition, which would work as a guide for searching for equal or better repair materials. However, the concept of historic material characterisation is yet to be explored in detail on the African continent. Only a few studies have analysed the heritage material properties for restoration interventions in Africa. Looking at the analysis results for both the Castle of Good Hope and Robben Island, the original materials, which go as far back as 1666, were made of lime-based mortars while the recent (late 1900s) repairs were conducted with the application of Portland cement materials.

Chapter 5 Discussion

The characterisation of historic mortars follows a sequence for mixing of concrete which involves the analysis of coarse and fine aggregates, the binder and binder-to-aggregate ratio. This chapter provides an in-depth analysis and comparison of the methods used for the analysis of heritage cementing materials and the results obtained during the characterisation of mortars collected from the Castle of Good Hope and Robben Island. The discussion is divided into two parts: the first part critically assesses the historic mortar characterisation theory studied in comparison to the data obtained from the experiments. The second part compares the results of the analysis for the materials from the different eras in the Western Cape Province, South Africa.

5.1 Introduction

The effects of the various deterioration factors on historic building materials, specifically the mortars, have resulted in the use of different repair materials for these structures. However, research has proven that most of the applied modern materials fail to remedy the problem. This is due to the significant differences between the properties of the original materials and the chosen repair materials. The proposed solution for this common challenge facing historic buildings is the use of the cost-effective methodology to be proposed in this study to analyse the original materials before restoration on any parts of the historic masonry is undertaken. Besides the outlined methods for performing this assignment, there generally exist a vast number of analytical techniques which various researchers selected for their analyses in different countries for different purposes. The methods shown in Section 2.4.2 to 2.4.7 are compared with the ones applied in this study. Their capabilities, effectiveness in providing the original mortar constituents and the quality of data are the aspects evaluated in detail in this chapter. Since there is not much work found on the standard procedure for characterising historic mortars for restoration interventions, discussion is necessary for this area. Thus, it is included in this section.

5.2 Sampling procedure

The method of data capturing during sampling used the form designed by Ngoma (2009), depicted in Figure 4.1. The details on the form are useful and adequate, as they provide insights into the factors that could have an impact on the samples. As seen on the form, all the relevant information, from the sample description to the methodology and the visual investigation, is entered in the form. The information is useful for documentation purposes and could be used as a reference in the future. The following aspects were observed to be essential during the sampling process:

- Historic data collection: This includes collection of all data related to the historic structure as gathered from the oral interviews with relevant authorities, stakeholders, structure owners, as well as the review and evaluation of historic documents (maps, reports, drawings, articles, newspaper reports, archives).
- Formation and statement of the main aim of sampling of materials: This helps with the decision on which analytical techniques to perform for the analysis. For example, if the purpose of sampling is said to be the determination of the deterioration factors and the extent of deterioration on the structure, carrying out salt analysis would be more suitable than determining the hydraulicity index through titration, as this test leads to the determination of the binder type

used, which in this case is not the main focal point. Therefore, the clear objective of the analysis is an important aspect to clarify even before sampling could be undertaken.

- Visual analysis and documentation of the surrounding materials near the sample location: This involves noting the type of bricks/blocks or stones that are joined by the material being sampled for analysis. It was observed that the surrounding materials might, one way or the other, have an impact on the properties of the mortar. For example, the mortar may indicate the presence of silica, which could be associated with hydraulicity, when in fact the silica traces are from the brick powder. Thus, noting the surrounding materials either in a photographic or report format would help answer and clarify some confusion that may arise during the analysis of the results.
- The environmental conditions where the structure is located. This also, like the other factors, has impacts on the results of the analysis and must be noted for future reference.

5.3 Part A - Analytical methods comparison

The characterisation of historic mortars involves the use of analytical methods to provide information about the original historic materials. The obtained information helps one to understand the mortar mix design originally used during the construction. The accuracy of the details is, however, not very good, due to the ageing of the structure. There are various historic mortar analysis techniques, as discussed in Section 2.4 that researchers use to characterise historic mortars for different purposes. However, these methods have not been standardised at any level. Therefore, researchers use different methods for various reasons (Ngoma, 2009). These methods can be divided into two major categories, namely:

- Wet chemical analysis methods: these are rapid, inexpensive and easy-to-perform tests. They include, but are not limited to, different acid digestion tests and gas collection methods. The interpretation of the results from this method requires some level of expertise in terms of material components (Ngoma, 2009).
- Instrumental analysis methods: these methods require the use of expensive equipment and high levels of expertise to operate the equipment and interpret the results. Despite the cost factor associated with the instrumental analysis, the methods provide detailed results about the mortar composition. They include methods such as polarised microscopy analysis, x-ray diffraction analysis, x-ray fluorescence and induced couple plasma, among many (Elsen, 2006).

The current study selected wet chemical analysis over the instrumental analysis based on cost and availability. The titration method is accurate for mineralogical and chemical analysis. However, there are limitations when characterising calcareous mortars with shells, therefore, mechanical separation (sieving) prior to testing was carried out to address this limitation (Alvarez *et al.*, 1999). This section compares and discusses the results obtainable from both methods as observed in the literature and during the experimental work, the quality of results and the shortcomings of each method over the other.

5.3.1 Aesthetic characterisation optimisation

From the results of the cohesion, visual investigation and microscopy analysis, the cohesion and visual investigation alone could have been enough to give details of the mortar composition in terms of particle size. Analysis using these two tests provides enough data in terms of the colour of the mortar, the texture, and the prediction of the binder type. The results from the microscopy analysis do not add any technical

value to what the other tests provide. The shell fragment sizes could have been visually investigated without the use of the microscope, and thus the cost associated with obtaining a microscope could be eliminated.

As was stated in Section 2.4.2, only 30% of the studied literature analysed the mortars using stereo microscopes after they conducted the simple, non-instrument physical characterisation. This could be because microscopy analysis was found to be non-essential by some researchers, as it provides results that could be obtained without use of instruments. Microscopy analysis only complements the visual investigation. The aesthetic characterisation is sufficient without the use of the stereo microscope even though aesthetic characterisation is dependent on the observer. None of the literature conducted colorimetry analysis using instruments, therefore, the visual colour analysis with a colour chart is sufficient to provide details of the mortar colour. However, the results of this method could vary from person to person.

5.3.2 Physical characterisation optimisation

In the literature, physical properties of historic mortars were investigated using scanning electron microscopy (SEM), thermal conductivity, porosimetry, wetting and drying cycles, salt crystallisation and freeze cycles, as discussed in Section 2.4.3. Some of these analytical techniques are either costly or time-consuming, as shown in Table 3.2. According to the results obtained from these tests, the methods could be considered unsuitable for the analysis of mortars for the purpose of determining the original constituents. Instead, the aforementioned tests rather provide some properties and elements which are not essential details for the identification of the binder type. For example, the SEM-EDS analysis (see Figure 2.7) gives elements such as carbon, potassium, chlorine and sodium which, for the purpose of determining the mortar components, are not necessary. This also applies to the physical ageing tests as discussed in Section 2.4.3. The tests could be used as compatibility criteria in terms of functional adjustment, stability and resistance to extreme weathering conditions of repair mortars, but, not for the analysis of mortar constituents. Therefore, they serve a different purpose from the aim of this research and were excluded.

The results of these techniques are more useful for providing data concerning deterioration of the original materials than they are for the restoration interventions using original material composition. As for the freeze-thaw action, it could be applied in areas where temperature ranges are extreme. South Africa rarely or never experiences such weather conditions, thus the analysis for this property is not applicable. The other physical tests, such as the porosity test (water absorption by immersion) could be performed instead, but, the analysis does not give details on particle distribution. It only provides the porosity of the material, from which the relationship between the material porosity and the particle size could be deduced. The search for replica aggregate material using this test would therefore lack precision.

Sieve analysis, on the other hand, is technically easy to perform and the sample is not destroyed during analysis. The technique not only yields particle distribution details, but also provides rough estimates of the mortar constituents (binder, aggregate, additives), based on the particle size of each material retained on respective sieves. The technique further predicts the expected mortar behaviour, which is estimated based on the coefficients of uniformity and curvature. The two coefficients provide details of the material gradations which are associated with the mortar properties.

The results from the sieve analysis test could be used to estimate the expected behaviour of the mortar being analysed. According to Ontiveros-Ortega *et al.* (2016), a well-graded aggregate has some effect on the porosity and retraction of mortars. The mortar produced from a well-graded aggregate is usually homogenous and consistent with pervious permeability, excellent shear strength and negligible compressibility when compacted, while high content of fine aggregates in a mortar provides the opposite properties to well-graded sand-produced mortars. The fine aggregates tend to decrease the mortar porosity, permeability and internal friction. Sieve analysis, being of proven effectiveness, as shown above, has a shortcoming in terms of the quantity of materials required, which is usually impossible to obtain from historic buildings.

5.3.3 Mineralogical characterisation optimisation

Generally, there are eight methods used for the identification of minerals, salts and ions in historic mortars. The analysis in this regard is performed to assess the mortar behaviour with regard to the environmental conditions surrounding the structure. The most commonly used method is XRD, which is normally complemented by use of TGA and DTA. The XRD analysis indicates the presence of the elements in a mortar sample through use of x-rays. The symbols indicated in Section 2.4.4. are used to denote the presence of the minerals. In addition to XRD, the thermal analyses are used for evaluation of the thermal conductivity of historic mortars. However, XRD and thermal analyses are considered expensive, as they require costly laboratory equipment and are also complex to perform. Therefore, they require detailed training and experience (Middendorf *et al*., 2005; Ngoma, 2009).

The salts crystallisation method requires quite large quantities (5 cm^3) of samples and it is a time-consuming exercise and therefore not selected for this study. Ion chromatography, on the other hand, provides reliable quantitative results and could have been selected, but, due to the lengthy procedure required for this test, it failed to pass the set criteria. Therefore, soluble salt analysis using a simple qualitative analysis by Teutonico (1988); Borrelli (1999) was the best alternative option for this phase of characterisation.

5.3.4 Chemical characterisation optimisation

The chemical analysis of heritage mortars is carried out to reveal in detail, the type of binder used for construction of a heritage building. The chemical analytical techniques involve wet chemical analysis which uses chemicals, reagents and indicators to determine the type of binder originally used. The other option is the use of the instruments such as XRF, ICP and AAS, as discussed in Section 2.4.5. The titration method is a relatively cost-effective method when compared to the instrumental methods which require expensive equipment to execute. However, the former gives less accurate results as compared to the latter. The current study chose wet chemical analysis for reasons of both cost and the availability of the required instruments within the vicinity of the study area.

Section summary

A conclusion can be drawn in relation to the choice of analytical techniques: some methods produce similar results, while others provide additional data that could not be obtained from other methods. Table 5.1 provides a summary of these methods of mortar composition analysis.

Table 5.1 Analytical methods used

Component	Method used	Alternative method
Binder	Visual analysis	Colorimetry
Binder	Optical microscopy	X-ray diffraction
Binder	Wet chemical analysis	Atomic absorption spectrometry, induced coupled plasma, scanning electron microscopy
Aggregate	Optical microscopy	X-ray diffraction
Aggregate	Sieve analysis	X-ray diffraction, scanning electron microscopy
Mix proportion	Sieve analysis	-
Mix proportion	Wet chemical analysis	-
Inclusions	Optical microscopy	X-ray diffraction

5.4 Part B – Discussion of results

This section compares the data obtained from the current study and that found in the literature, using various methods. It goes on to discuss and interpret the different era's materials to identify the similarities or differences, if any, in terms of heritage cementing materials development over the centuries. A conclusion is made in terms of the history of cementing materials in the Western Cape Province, taking the two structures (Castle of Good Hope and Robben Island) as representatives of heritage structures over hundred years old.

5.4.1 Aggregates

The results obtained from the wet chemical analysis for the acid-insoluble aggregates show materials that are fine to medium-graded. Samples like SK6, which represented the coarse aggregates, were the exception. As described in Table 4.7, all the samples had different grades of sand as aggregates, while some included shell particles, as was observed during visual investigation. The salt analysis of the aggregates (material retained on sieve size 0.063) shown in Table 4.5 indicated the majority of materials to be either free of sulfates and chlorides or containing only minor traces of these salts. The few samples from the maximum-security prison (1700) on the Island contained very high amounts of chlorides which, according to Zinn (2005), can cause premature deterioration of building materials due to salt crystallisation. Sample MX1, which, according to the visual investigation, was suspected to be repair material that showed signs of spalling from the original surface contained high amounts of chlorides. This makes the aggregate used for this mortar unsuitable for the surface it is applied to. Around 90% of the aggregates were rich in carbonate salts while roughly 15% indicated the presence of nitrates in limited quantities.

5.4.2 Binder

None of the materials from the Castle and Island were produced from pure lime. This can be seen in Table 4.6, where all the CI limits were higher than 0.30, as classified by Boynton (1980). According to Martínez

et al. (2013), the materials with a CI value closer to 1 are equivalent to Portland cement. These are the materials which according to Boynton (1980), have high (up to 45%) clay content. The results have confirmed that all the heritage cementing materials prior to the introduction of Portland cement in the mid-1800s were made of lime. For this study, the high-clay and cement materials were indeed the restored areas, as reported earlier on the visual analysis, as they have shown signs of damage caused by the use of cement in the restoration of heritage buildings.

As was predicted during the physical analysis, the samples SK7 to SK9 were mud without any additions of a binder. The results in Table 4.6 dispute this assertion. These samples indicated a high cementation index, which, according to Middendorf *et al*. (2005b), could be due to silica from brick dust or clay minerals, which have a very high clay content. This justification of the high CI content in these samples is just an assumption, and could be verified based on some background knowledge of the origin of the heritage building's construction materials.

5.4.3 Different Era materials

As indicated in Table 2.7, the majority of historic buildings were constructed using different lime binders. This was found to be the case for the materials from the seventeenth to the nineteenth century in Western Cape of South Africa. The summaries of the material composition in the literature for the three era materials are presented in Table 5.2.

Table 5.2 materials per century

Author	Location	Type of structure	Age (Century)	Material composition
Gulzar *et al*. (2013)	Pakistan (Asia)	Mughal Empire	17th	Calcitic lime binder produced from calcinations of kankar-$CaCO_3$ from soil horizon. The aggregates included crushed bricks, broken kankar pieces, brick kiln furnace slag and a small fraction of siliceous sand.
Current study	South Africa (Africa)	Castle	17th	Sub-hydraulic lime mortars which are low in clay content. The aggregates generally included fine to medium sea sand with inclusions of fine to medium crushed seashells. The other three samples, however, were mud.
Lopez-Arce *et al*. (2016)	France (Europe)	Exhibition hall and museum	18th	Traces of gypsum, calcium and sodium sulfates, minor amounts of nitrates found in mortar.
Current study	South Africa (Africa)	Island	18th	The original materials were made of moderate hydraulic lime, except the repair materials made of cement and high in clay content. The aggregates were generally fine sand. No traces of nitrates found in mortar; however, minor traces of nitrites were observed.
Gleize *et al*. (2009)	Brazil (South America)	9 historic buildings in the State of Santa Catarina	18th-20th	Hydrated lime obtained from the burning of seashells was the major binder. It was sometimes mixed with hydraulic materials (clay, ground ceramic tile or brick, and hydraulic lime).
Current study	South Africa (Africa)	Island	19th	Similar to the 18th century materials, the original materials were made of moderate hydraulic lime, except the repair materials made of cement and high in clay content.

5.5 Conclusion

The main purpose of this work was to develop a standard protocol for determining the components of historic mortars. The results pertaining to the simplest methodology were critically evaluated for their competence in providing the required information on the original mortar recipe. The findings related to the various material properties using various methods were reported and compared. It was observed that the wet chemical analyses techniques are preferable to the instrumental analyses due to superior cost-effectiveness. It can also be concluded that the heritage cementing materials used from as far back as the 1600s were indeed lime-based, as visual investigation predicted. The lime mortars were replaced with cement-based materials on the two heritage buildings in Cape Town, South Africa. From the separated aggregates from the binder through acid dissolution, the particle distribution pattern can be estimated, and hence the aggregate type. Therefore, it is not entirely necessary to carry out sieve analysis, which requires large material quantities.

Chapter 6 Conclusions and recommendations

Many heritage buildings are in dilapidated conditions which warrant research into their sustainable repair. The area which is most susceptible to deterioration is the masonry, which often shows the weakening of cementing materials in between the blocks, bricks or stones. With this being the case, the masonry becomes weak, thus, threatening the continued existence of the historic buildings due to increased likelihood of collapse. Therefore, the protection of heritage structures against deterioration factors needs special attention, as these structures contribute significantly towards economic growth for their home countries. This chapter presents a summary of work carried out to accomplish a standard protocol for characterising heritage mortars for their compatible restoration. It reflects on the objectives and evaluates them against the outcomes. It also entails recommendations for further research in the area of heritage material characterisation.

6.1 Conclusions

The majority of heritage buildings, not only in South Africa but abroad, undergo deteriorations which according to the literature, are often repaired using modern materials which are proven to possess different behaviours to the ones they are replacing. This common application of materials which differ from the original has cost South Africa not only time but also the resources and money involved in repeated repairs of heritage buildings. Even though the legislation states the need to compile a conservation management plan prior to conservation and restoration of heritage properties, there is still a gap in terms of detailed material characterisation procedures to be followed when analysing original materials before embarking on the restoration journey. The literature does emphasise the need to characterise historical materials for achieving long-lasting repairs that match the original surfaces. However, the procedures for carrying out the analysis are often either expensive or time-consuming. This raised the question of what could then be the alternative route in ensuring that the same results are obtained using a methodology that is affordable and technically easy to perform.

The conservation and restoration of historic buildings, as described by Fagan in an interview, is not as simple and straight forward as most appointed conservation and restoration teams assume it to be. Mrs Fagan described the process as follows: "*Dealing with conservation and restoration work on heritage buildings is similar to treating a patient, you do not follow what is on your mind, you get the history (when was it built, by whom, using what materials, what is the problem and the cause) of the building. Then use the material that was used originally. The decision is then made as to what age you are restoring the building to, and the reasons for such choice are provided.*" (Fagan, 2018). The description encourages material characterisation in restoration practices, so does this study. For practice of compatible restoration, the study characterised materials from the Castle of Good Hope and Robben Island, with a focus on aesthetic, physical, mineralogical and chemical properties that hold engineering significance during conservation and restoration.

The procedure to obtain this information commenced with the detailed optimisation of the analytical techniques for selection of the suitable method for application in this study. The merits and shortcomings of each method were outlined and assessed. Thereafter, the suitable techniques were applied to test their competence in providing technical and reliable results in relation to material constituents. The study went on to test the methods, which involved the onsite visual investigation of the materials representing the

different eras (1600s to 1800s), cohesion test, colour test, microscopy analysis, salt analysis and titration tests on similar materials sampled from the heritage buildings.

For aesthetic and physical characterisation, the selected 3 tests were all found to be necessary and worth conducting. However, examining the results from all the three tests discussed in Section 4.2 to 4.4, it is concluded that the optical characterisation alone could provide sufficient information on the mortar properties without the use of a microscope. It is possible to observe the colour, the particle distribution estimate values, the cohesion and the particle sizes from these tests without the use of expensive laboratory equipment. Hence, microscopic analysis can be excluded. As for the investigation of the mineralogical and chemical properties, wet chemical analysis alone provided informative results to be used from the engineering point of view during the restoration of heritage cementing materials. Therefore, this method is preferred over the rest.

A conclusion was made regarding the choice of analytical methods which depend on the deterioration factors or damage present on the structure. For example, we cannot address the problem of moisture movement by simply carrying out colour tests or titration. We need to carry out moisture tests such as permeability or porosity tests for such masonry problems. With this being the case, a careful decision has to be made, since there could be a chance that the original materials do not provide adequate engineering properties in the mortar. In such cases, modifications are necessary to make the optimum repairs.

In general, heritage restoration is a teamwork initiative which requires a common understanding between all the relevant stakeholders. The roles played by different stakeholders, namely: engineers, contractors, conservators, historians, architects, archaeologists, and heritage authorities ensure increased lifespan of these national treasures. The appointment of competent restoration team together with incorporation of the original materials characterisation in the conservation management plans (CMPs) would ensure compatible restoration. It is therefore every stakeholder's responsibility to implement the application of compatible materials as stated in respective site CMP.

6.2 Standard protocol

For determination of the original material properties, a detailed yet inexpensive protocol is proposed. The protocol involves manual sampling of 5 to 15 g of the original material from the heritage structures. The materials are taken for analysis at the laboratory for the binder-to-aggregate ratio, binder and aggregate types and the possible salt contents in the aggregates and the binder itself. The analysis could be performed by any individual with limited knowledge of chemistry using the procedure outlined below.

A. Collection of history

Collect as much history on the building from the stakeholders and archives as possible.

B. Sampling

Identify deteriorated sections to sample from.
Capture images of the section on the building to be sampled.
Capture images of mortars in-situ before sampling.

Use a small hammer to gently remove a maximum of 15 g of the in-situ material from the masonry which represents the section targeted, sample from numerous sections depending on the visibility of material differences.
Store samples in separate sealable bags or bottles with the description (mortar colour, texture, additives, location and the surrounding materials, the environmental conditions, the sampling date, and details of the person carrying out the assignment name, sample number, building name, building location, sample location, mortar type, date of sampling) on the containers.
Keep the samples uncovered to allow them to dry for almost 24 hours following analysis.

C. Laboratory preparation

After procurement of chemicals and necessary glassware, prepare the standard solutions as indicated in Annexure B.
Label all the glassware to be used in accordance to sample identifications and weigh the beakers for mass calculations of binder-to-aggregate ratios.
Printout the methodology to follow during analysis and prepare a template to record the results.

D. Sample preparation

Apply gentle pressure to the mortar chip to determine its resistance to pressure and note the behaviour.
Gently crush 5 to 10 g of the samples with mortar and pestle to form powder and leave other samples for reference. Avoid breaking the aggregate and additive particles, if any.
Oven-dry the sample to ensure the release of microscopic moisture until mass consistency is reached.

E. Testing

Record the colour of the intact mortar against the colour chart.
Separate part (2 g or less) of the crushed material into two parts (binder and aggregate) using standard sieve (0.063 mm) for analysis of salts on two portions of 0.1 g each.
Carry out the procedure illustrated in Figure 3.9.
Record the results for sulfates, chlorides, nitrates, nitrites and carbonates as visible on the test tubes.
Take 5 to 10 g of the powder (before separation) and express the binder-to-aggregate ratio using the procedure summarised in Figure 3.10. This procedure is also followed for determination of the binder type and aggregates type.
Take the residue from acid dissolution and provide descriptive information on the aggregate.

6.3 Recommendations for future research

This study demonstrates clearly that the study of historical buildings is very important for the formulation of intervention techniques and materials. Research should therefore not only be limited to the analysis of physical, chemical and mineralogical properties, but also mechanical properties, material strength, elasticity and shrinkage properties. There is also a need to integrate the general construction material analysis with construction techniques used for the construction of historic buildings. The properties of other materials matter in the analysis of heritage/historical buildings, as they affect the way the cementing materials behave. This will be used to further indicate the technique suitable for restoration of historic structures.

The original material characterisation for restoration interventions in the South African heritage requires enforcement through mandatory inclusion in conservation management plans (CMPs). South African legislation (NHRA, 1999) states that each heritage site must have a CMP prior to restoration, but it does not outline what a CMP must contain (contents of the CMP). The following are the areas that warrant research as far as heritage material characterisation is concerned:

Design and production of suitable heritage restoration mortars for materials from different era, based on their original properties.
Investigation of improved heritage restoration mortar for all eras.
Investigating the environmental impacts on heritage building materials.
Non-destructive testing of heritage buildings for mechanical properties.

References

A Brief History of The Castle of Good Hope, Cape Town. 2018. Available at: https://theculturetrip.com (accessed in September 2018).

Aggelakopoulou, E., Bakolas, A. & Moropoulou, A. 2011. Hygric Properties of Lime Based Mortars used for Restoration Interventions on Historic Structures. *International Conference on Durability of Building Materials and Components*, Porto- Portugal, April 12 -15.

Alvarez, J.I., Martín, A., García Casado, P.J., Navarro, I. & Zornoza, A. 1999. Methodology and validation of a hot hydrochloric acid attack for the characterization of ancient mortars. *Cement and Concrete Research,* 29: 1061–1065.

Amoroso, G. G., Fassina, V. 1983. Stone Decay and Conservation. Elsevier Science, Amsterdam. *Journal of Aerosol Science,* 16.1 (1985): 87–87.

Apostolopoulou, M., Aggelakopoulou, E., Siouta, L., Bakolas, A., Douvika, M., Asteris, P. G. & Moropoulou, A. 2017. A methodological approach for the selection of compatible and performable restoration mortars in seismic hazard areas. *Construction and Building Materials,* 155: 1–14.

Arandigoyen, M. & Alvarez, J. I. 2007. Pore structure and mechanical properties of cement-lime mortars. *Cement and Concrete Research,* 37:767–75.

Arıoglu, N. & Acun, S. 2006. A research about a method for restoration of traditional lime mortars and plasters: A staging system approach. *Building and Environment,* 41: 1223–1230.

Arizzi, A. & Cultrone, G. 2013. The influence of aggregate texture, morphology and grading on the carbonation of non-hydraulic (aerial) lime-based mortars. *Q. J. Engineering Geology Hydrogeology*. 46: 507-520.

Balksten, K. 2010. Understanding Historic Mortars and their Variations – a Condition for Performing Restorations with Traditional Materials. *2nd Historic Mortars Conference HMC2010 and RILEM TC 203-RHM Final Workshop* 22-24 September 2010, Prague, Czech Republic.

Bartz, W. & Filar, T. 2010. Mineralogical characterization of rendering mortars from decorative details of a baroque building in Kożuchów (SW Poland). *Material characterization*, 61: 105-115.

Benedetti, A. & Pelà, L. 2012. Experimental characterization of mortar by testing on small specimens. *15th International Brick and Block Masonry Conference*, Florianópolis – Brazil.

Bertolini, L., Carsana, M., Gastaldi, M., Lollini, F. & Redaelli, E. 2013. Binder characterisation of mortars used at different ages in the San Lorenzo church in Milan. *Material Characterization*, 80: 9-20.

Borges, C., Santos Silva, A. & Veiga, R. 2010. Ancient mortars under action of marine environment: a physico-chemical characterization. 2nd Conference of Historic Mortars - HCM 2010 & RILEM TC 203-RHM Final Workshop, Bagneux, pp. 29-41.

Borrelli, E. 1999. ICCROM *Arc Laboratory handbook. Conservation of architectural heritage, historic structures and materials*. Rome, Italy. Volume 1/99.

Borsoi, G., Silva, A. S., Menezes, P., Candeias, A. & Mirão, J. 2010. Chemical, Mineralogical and Microstructural Characterization of Historical Mortars from the Roman villa of Pisoes, Beja, Portugal. *2nd Historic Mortars Conference HMC2010 and RILEM TC 203-RHM Final Workshop 22-24 September 2010,* Prague, Czech Republic. Pp. 43 – 54.

Boynton, R.S. 1980. *Chemistry and Technology of Lime and Limestone*. (2nd edition) Wiley, New York, USA.

Brosnan, D. A. 2014. Characterization and Degradation of Masonry Mortar in Historic Brick Structures. Hindawi Publishing Corporation, *Journal of Structures*, volume 2014, Article ID: 859879, pp. 1-7.

Camuffo, D. 1995. Physical weathering of stones. *The Science of the Total Environment*, 167: 1-14.

Castle of Good Hope. 2018. Available at https://castleofgoodhope.co.za (accessed in September 2018).

Cayme, J.M.C. & Asor, A.N. 2017. Calcium content of lime mortars from 9th century church ruins in the Philippines using volumetric analysis. *Malaysian Journal of Analytical Sciences*, 21(5): 1080-1090.

Charola, A., Dupas, M., Sheryll, R. & Freund, G. 1986. Characterization of Ancient Mortars: Chemical and Instrumental Methods. Scientific Methodologies Applied to Works of Art. *Proceedings of the Symposium*, Milan, pp. 28-33.

Cizer, Ö., Schueremans, L., Serre, G., Janssens, E. & Van Balen, K. 2010. Assessment of the Compatibility of Repair Mortars in Restoration Projects. *Advanced Materials Research*, 133-134: 1071-1076.

Conditional Assessment of Buildings on Robben Island: Outline Description of Material Testing Required. 2018. Cape Town, South Africa. Available at: https://sahris.sahra.org.za (accessed in March 2019).

Corinaldesi, V. 2012. Environmentally friendly bedding mortars for repair of historical buildings. *Construction and Building Materials,* 35: 778–784.

Deshpande, U. L. 2017. Repair, restoration and strengthening of buildings. *International Journal of Innovations in Engineering Research and Technology,* 4(3): 121 - 124.

Dolar-Mantuani, L. 1984. Handbook of Concrete Aggregates, A Petrographic and Technological Evaluation, Noyes Data Corporation, United States, pp. 79-125.

Drdácky´, M., Fratini, F., Frankeova, D. & Slizˇková, Z. 2013. The Roman mortars used in the construction of the Ponte di Augusto (Narni, Italy)– A comprehensive assessment. *Construction and Building Materials*, 38: 1117-1128.

Elsen, J. 2006. Microscopy of historic mortars - a review. *Cement and Concrete Research*, 36: 1416-1424.

Elsen, J., Mertens, G. & van Balen, K. 2011. Raw materials used in ancient mortars from the Cathedral of Notre-Dame in Tournai (Belgium). *Mineralogical Sciences and Archaeology*, 23: 871–882.

Fagan, G. 2018. Interview with Castle of Good Hope restoration architect on 11 September 2018, Cape Town; South Africa.

Fang, S., Zhang, K., Zhang, H. & Zhang, B. 2015. A study of traditional blood lime mortar for restoration of ancient buildings. *Cement and Concrete Research*, 76: 232 - 241.

Feilden, B. M. 2003. Conservation of Historic Buildings. 3rd edition, Architectural press, Elsevier.

Gilbert, C. 1994. The Castle of Good Hope: An Examination of Controversies and Conflicting Perceptions – A Case Study in Public History. An Honours degree thesis, University of Cape Town, South Africa.

Gleize, P. J. P., Motta, E. V., Silva, D. A. & Roman, H. R. 2009. Characterization of historical mortars from Santa Catarina (Brazil). *Cement and Concrete Composites*, 31: 342–346.

Goins, E. 2001. A standard method for the characterization of historic cementitious materials. US/ICOMOS Scientific Journal: Historic Mortars & Acidic Deposition on Stone, 3 (1):6-7.

Gosselin, C., Verges-Belmin, V., Royer, A. & Martinet, G. 2008. Natural cement and monumental restoration. *Materials and Structures*, 42: 749-763.

Green, D.W. & Perry, R.H. 2008. Perry's Chemical Engineers' Handbook, McGraw-Hill, (2) 99-118.

Groot, C.J.W.P, Ashall, G.J., Hughes, J.J. & Bartos, P.J.M. 2004. Characterisation of old mortars with respect to their repair. *RILEM TC 167-COM: State of the art Report*.

Gulbe, L., Vitina, I. & Setina, J. 2017. The influence of cement on properties of lime mortars. *Procedia Engineering,* 172:325 – 332.

Gulzar, S., Chaudhry, M. N., Burg J. P. & Saeed S. A. 2013. Characteristics of Ancient Mortars and Plasters from the Archaeological Site of Akbari-Serai (Pakistan). *Journal of Chemistry*, 25(15): 8484-8488.

Hauková, P., Frankeová, D. & Slizˇková, Z. 2013. Characterisation of Historic Mortars for Conservation Diagnosis. *Proceedings of the 3rd Historic Mortars Conference,* Glasgow Scotland, 11-14 September. Pp. 109-117.

Holmes, S. & Wingate, M. 1997. Building with Lime. *Intermediate Technology Publications*, London. Pp. 306.

Hormes, J., Diekamp, A., Klysubund, W., Bovenkamp, G. L. & Börste, N. 2016. The characterization of historic mortars: A comparison between powder diffraction and synchrotron radiation-based X-ray absorption and X-ray fluorescence spectroscopy. *Microchemical Journal*, 125: 190–195.

Isebaert, A., Van Parys, L. & Cnudde, V. 2014. Composition and compatibility requirements of mineral repair mortars for stone – A review. *Construction and Building Materials*, 59: 39–50.

Jedrzejewska, H. 1960. Old Mortars in Poland: A New Method of Investigation. *Studies in Conservation,* 5(4): 132-138.

Křivánková, D., Nunes, C.L., Slížková, Z., Frankeová, D., Niedoba, K. 2019. *High-Performance Repair Mortars for Application in Severe Weathering Environments: Frost Resistance Assessment*. In: Hughes J., Válek J., Groot C. (eds) Historic Mortars. Springer, Cham.

Labiadh, M. R., Ouezdou, M. B., Hajjem, B. T. & Mensi, R. 2009. Characterization of waterproof-covering mortars on Ottoman monuments of ‘‘Ghar El Melh” (Tunisia) *Construction and Building Materials*, 23: 423–433.

Lanas, J. & Alvarez, J. I. 2003. Masonry repair lime-based mortars: Factors affecting the mechanical behaviour. *Cement and Concrete Research,* 33: 867–1876.

Lawrence, S. J., Samarasinghe, W. & Guirguis, S. 2004. Mortar durability- Development and standardization of test methods. *13th International Brick and Block Masonry Conference,* Amsterdam, July -7.

Lopez-Arce, P., Tagnit-Hammou, M., Menendez, B., Mertz, D., Guiavarc'h, M., Kaci, A., Aggoun, S. & Cousture, A. 2016. Physico-chemical stone-mortar compatibility of commercial stone-repair mortars of historic buildings from Paris*. Construction and Building Materials*, 124: 424–441.

Lourenço, P. B. 2006. Recommendations and Advances in Research and Practice. *Construction and Building Materials*, 20(4): 239 - 251.

Marini, A., Cominelli, S., Zanotti, C. & Giuriani, E. 2018. Improved natural hydraulic lime mortar slab for compatible retrofit of wooden floors in historical buildings. *Construction and Building Materials*, 158: 801 - 813.

Martínez, I., Castillo, A., Martínez, E. & Castellote, M. 2013. Physico-chemical material characterization of historic unreinforced masonry buildings: The first step for a suitable intervention. *Construction and Building Materials*, 40: 352–360.

Mbasha, W. M. 2015. The effect of Portland cements and superplasticisers on the rheology of cement paste. Master's thesis, Cape Peninsula University of Technology, Cape Town, South Africa.

Meli, R., Roberto, M. T. & Sánchez-Ramírez, N. 2007. Criteria and experiences on structural rehabilitation of stone masonry buildings in Mexico City. *International Journal of Architectural Heritage*, 1(1): 3 -28.

Middendorf, B. & Knöfel, D. 1991. Investigation of Mortars from Medieval Brick Buildings in Germany. *Proceeding of the 13th International Conference on Cement Microscopy*, Tampa, pp 118-123.

Middendorf, B., Hughes, H., Callebaut, K., Baronio, G. & Papayianni, I. 2005a. Investigation Methods for the Characterisation of Historic Mortars, Part 1: Mineralogical Characterisation. *Materials and Structures*, 38: 761-769.

Middendorf, B., Hughes, H., Callebaut, K., Baronio, G. & Papayianni, I. 2005b. Investigation Methods for the Characterisation of Historic Mortars, Part 2: Chemical Characterisation. *Materials and Structures*, 38: 771-780.

Mitchell, D. S. 2007. The use of lime & cement in traditional buildings*,* Edinburgh: Technical Conservation, *Research and Education Group*.

Morricone, A., Macchia, A., Campanella, L., Massimiliano, D., de Togni, S., Turci, M., Maras, A., Meuccid, C. & Roncad, S. 2013. Archeometrical analysis for the characterization of mortars from Ostia Antica Andrea. *Procedia Chemistry,* 8: 231 – 238.

Mwasinga, B. 2018. Interview with South African Heritage Resources Agency Built Environment Manager on 26 July 2018, Cape Town; South Africa.

Ngoma, A. M. K. 2009. Characterisation and Consolidation of Historical Lime Mortars in Cultural Heritage Buildings and Associated Structures in East Africa. PhD thesis, University of Dar-es-Salaam, Tanzania.

Ontiveros-Ortega, E., Rodríguez-Gutiérrez, O. & Navarro, A. D. 2016. Mineralogical and physical–chemical characterisation of Roman mortars used for monumental substructures on the Hill of San Antonio, in the Roman city of Italica (prov. Baetica, Santiponce, Seville, Spain). *Journal of Archaeological Science*, 7: 205–223.

Ozga, I. J. 2009. Multi-pollutants impact on modern cement built heritage. PhD thesis, University of Bologna.

Özkaya, Ö. A. & Böke, A. 2009. Properties of Roman bricks and mortars used in Serapis temple in the city of Pergamon. *Material characterization*, 60: 995-1000.

Palomo, A., Blanco-Varela, M. T., Martinez-Ramirez, S., Puertas F. & Fortes, C. 2014. Historic Mortars: Characterization and Durability. New Tendencies for Research. Eduardo Torroja Institute, Spain. Pp. 1-20.

Papayianni, I., Pachta, V. & Stefanidou, M. 2013. Analysis of ancient mortars and design of compatible repair mortars: The case study of Odeion of the archaeological site of Dion. *Construction and Building Materials*, 40: 84–92.

Papayianni, I. & Pachta, V. 2017. Earth block houses of historic centres. A sustainable upgrading with compatible repair materials. *Procedia Environmental Sciences*, 38: 274 - 282.

Patterns and characteristics of migration to the Western Cape, South Africa. 2019. Available at: https://www.urbanafrica.net (accessed in March 2019).

Penelis, G. G. 1996. Techniques and Materials for Structural Restoration. Paper No. 2089. *Eleventh World Conference on Earthquake Engineering*, ISBN: 0 08 042822 3.

Price, C. A. 2010. Stone Conservation. *An Overview of Current Research. The Getty Conservation Institute*, 2nd Edition, Los Angeles, United States of America.

Rampazzi, L., Corti, C., Colombo, C., Conti, C. & Realini, M. 2010. Development of an Analytical Protocol for Characterisation of Historical Mortars. An abstract: *2nd Historic Mortars Conference HMC2010 and RILEM TC 203-RHM Final Workshop* 22-24 September 2010, Prague, Czech Republic.

Robben Island. 2018. Available at: http://whc.unesco.org/en/list/ (accessed in September 2018).

Rodriguez-Navarro, C., Hansen, E. & Ginell, W.S. 1998. Calcium hydroxide crystal evolution upon aging of lime putty, *Journal of American Ceramics Society*, 81(11): 3032-3034.

Salvadori, M. 1982. Why Buildings Stand Up, The Strength of Architecture, McGraw Hill, pp. 311.

Sandbhor, S. & Botre, R. 2013. A systematic approach towards restoration of heritage buildings – A case study. *International Journal of Research in Engineering and Technology*, ISSN: 2319-1163, volume 2: issue 3, pp. 229-238.

Sanjurjo-Sánchez, J., Trindade, M. J., Blanco-Rotea, R., Benavides Garcia, R., Fernández Mosquera, D., Burbidge, C., Prudêncio, M. I. & Dias, M. I. 2010. Chemical and mineralogical characterization of historic mortars from the Santa Eulalia de Bóveda temple, NW Spain. *Journal of Archaeological Science,* 37: 2346-2351.

Schueremans, L., Cizer, Ö., Janssens, E., Serré, G. & Van Balen, K. 2011. Characterization of repair mortars for the assessment of their compatibility in restoration projects: Research and practice. *Construction and Building Materials,* 25: 4338–4350.

Singh, M., Waghmare, S. & Kumar, V. 2014. Characterization of lime plasters used in 16th century Mughal monument. *Journal of Archaeological Science*, 42: 430-434.

South Africa, 1999. National Heritage Resources Act of 1999, Volume 25, Section. 35 (4). *Government Gazette,* Pretoria.

South African Heritage Resources Agency. Declared Heritage Sites in South Africa. 2018. Available at: https://www.sahra.org.za/sahris/declaredsites (accessed in Aug 2018).

South African Heritage Resources Agency. Owl house museum site report. June 2018.

Stefanidou, M. & Papayianni, I. 2005. Inclusions in Selected Mortars of Ancient Technology. *Proceeding of the 10th Euroseminar on Microscopy Applied to Building Materials*, Paisley, Scotland, June 21- 25.

Teutonico, J. M. 1988. ICCROM Arc laboratory manual for architectural conservators. Rome, Italy.

Van Balen, K., Ercan, S. &. Patricio, C. 1999. Compatibility and Retreatability Versus Reversibility: A Case Study at the Late Hellenistic Nymphaeum of Sagalassos (Turkey). The Use of and Need of Preservation Standards in Architectural Conservation. ASTM STP 1335, Sickels-Taves (ED). West Conshohocken, PA, pp. 105-118.

Van Balen, K., Toumbakari, E. E., Blanco, M. T., Aguilera, J., Puertas, F., Sabbioni, C., Zappia, G., Riontino, C. & Gobbi, G. 1999. Procedure for mortar type identification: *A proposal, International RILEM workshop on historic mortars: Characteristics and Tests; Paisley,* Scotland, 12th - 14th May, edited by P. Barton, C. Groot and J. J. Hughes. Cachan, France: RILEM Publications.

World Heritage Sites in South Africa. 2018. Available at: http://whc.unesco.org/en/list/ (accessed in September 2018).

World Travel and Tourism Council. 2018. Travel & Tourism, Economic Impact, South Africa. Available at: https://www.wttc.org (accessed in August 2018).

Zheng, H. 2011. Building Materials in Civil Engineering. *Woodhead Publishing Series in Civil and Structural Engineering*. Pp. 29-45,423.

Zinn, W.A. 2005. Cement Modified Earthen Mortar – An Investigation of Soil-Cement Performance Characteristics at Three Southwestern National Monuments. PhD thesis, University of Pennsylvania.

Appendices

Appendix A. Samples gallery

This section shows the samples collected from the Castle f Good Hope and the Robben Island using the manual hammer-and-chisel method.

Figure A.1 Castle of Good Hope samples

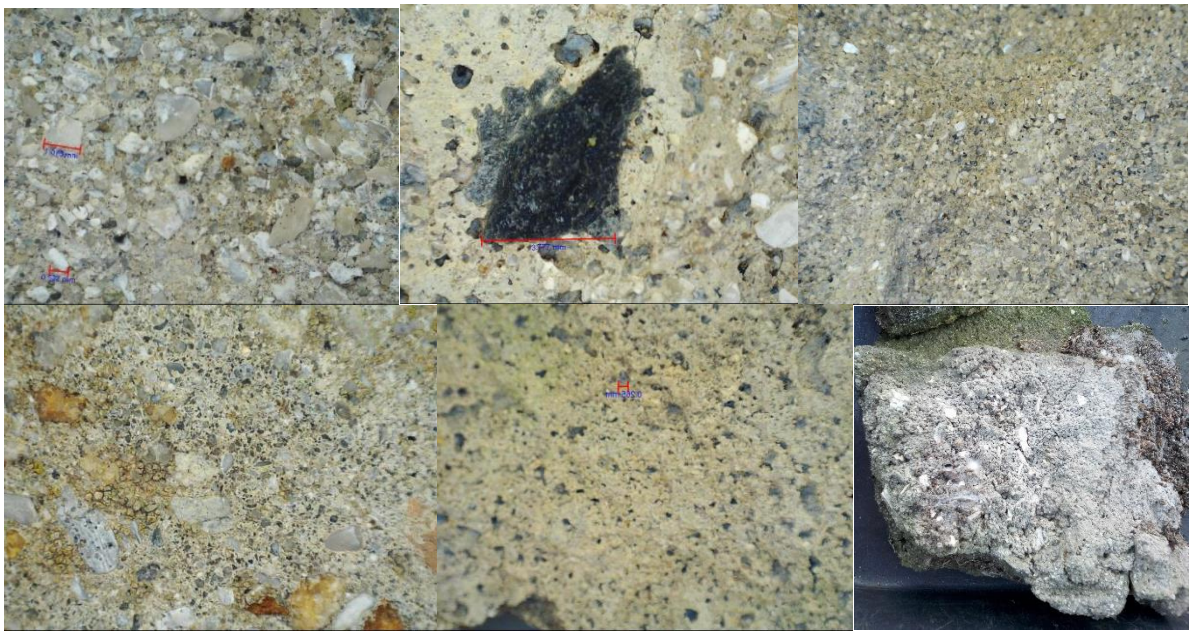


Figure A.2 Robben Island samples

Appendix B. Preparation of solutions

The analytical reagents used in this project were prepared as indicated in this section. Manual mixing of solutions was used, whereby the volumetric flask is closed with a stopper and shaken, holding the flask with both hands. The shaking of the volumetric flask, while holding the neck of the flask or closing the flask with a hand or finger is strongly prohibited. The prepared solutions were then transferred into a clean dry bottle and labelled accordingly.

2N HCl in 250 mL volumetric flask

To prepare 250 mL of a 2 N solution of hydrochloric acid, dilute 47.45 mL of 33 % HCl to a final volume of the volumetric flask with deionised (distilled) water.

10% of $BaCl_2$ solution in 250 mL volumetric flask

To prepare 250 mL of a 10 % solution of barium chloride, dissolve 32.0909 g of $BaCl_2 \times 2H2O$ (99.8 % purity) in deionised or distilled water. After the solid is completely dissolved, dilute the solution to the mark with deionised (distilled) water.

2N of HNO_3 in 250 mL volumetric flask

To prepare 250 mL of a 2 N solution of nitric acid, dilute 34.84 mL of 65 % HNO_3 to a final volume with deionised (distilled) water.

0.1 N of $AgNo_3$ in 250 mL

To prepare 250 mL of a 0.1 N solution of silver nitrate, dissolve 4.2554 g of $AgNO_3$ (99.8 % purity) in deionised or distilled water. After the solid is completely dissolved, dilute the solution to a final volume with deionised (distilled) water.

2 N of CH_3COOH in 250 mL volumetric flask

To prepare 250 mL of a 2 N solution of acetic acid, dilute 28.61 mL of 99.8 % CH_3COOH to a final volume with deionised (distilled) water.

4 N of KOH in 50 mL volumetric flask

To prepare 50 mL of a 4 N solution of potassium hydroxide, dissolve 11.221 g of KOH (100 % purity) in deionised or distilled water. After the solid is completely dissolved, dilute the solution to a final volume with deionised (distilled) water.

Saturated Na_2CO_3 solution in 250 mL volumetric flask

To prepare 250 mL of a 1 M (saturated) solution of sodium carbonate, dissolve 26.497 g of Na_2CO_3 (100 % purity) in deionised or distilled water. After the solid is completely dissolved, dilute the solution to a final volume with deionised (distilled) water.

Sulphosalicylic acid in 100 mL

To prepare 100 mL of sulphosalicylic acid, dissolve 5 g of sulphosalicylic acid in deionised or distilled water. After the solid is completely dissolved, dilute the solution to a final volume with deionised (distilled) water.

0.025 M EDTA in 250 mL

To prepare 250 mL of a 0.025 M solution of EDTA disodium salt, dilute 25 mL of 0.25 M $Na_2C_{10}H_{14}N_2O_8$ to a final volume with deionised (distilled) water.

0.025 M $ZnSO_4$ in 250 mL

To prepare 250 mL of a 0.025 M solution of Zinc sulfate, dissolve 1.7971 g of $ZnSO_4 \times 7H_2O$ (100 % purity) in deionised or distilled water. After the solid is completely dissolved, dilute the solution to a final volume with deionised (distilled) water.

Appendix C. Particle size distribution

The tables below represent the detailed calculations for the particle distribution of the materials from the castle.

Table C.1 SK1 sieve analysis

Sieve size (mm)	Mass 1 retained (g)	Mass 2 retained (g)	Mass 3 retained (g)	Ave. Mass retained (g)	% mass retained	Cumulative mass retained (%)	% passing
9.500	0	0	0	0	0	0	100.00
6.700	2.071	1.928	6.319	3.439	1.15	1.15	98.85
4.750	14.547	4.443	8.331	9.107	3.04	4.19	95.81
2.360	55.604	47.735	61.093	54.811	18.30	22.49	77.51
2.000	23.027	22.597	25.180	23.601	7.88	30.37	69.63
1.180	20.761	28.514	22.013	23.763	7.93	38.30	61.70
0.600	61.267	66.208	60.830	62.768	20.96	59.26	40.74
0.425	21.376	25.305	19.078	21.920	7.32	66.58	33.42
0.300	18.558	22.816	20.863	20.746	6.93	73.51	26.49
0.150	35.111	34.870	33.952	34.644	11.57	85.08	14.92
0.075	28.282	28.240	25.712	27.411	9.15	94.23	5.77
Pan	18.50	17.342	16.003	17.282	5.77	100.00	0
Total				299.492			

Table C.2 SK2 sieve analysis

Sieve size (mm)	Mass 1 retained (g)	Mass 2 retained (g)	Ave. Mass retained (g)	% mass retained	Cumulative mass retained (%)	% passing
6.700	0	0	0	0	0	100.00
4.750	0.749	1.453	1.476	0.49	0.49	99.51
2.360	13.088	14.746	13.917	4.64	5.13	94.87
2.000	13.000	14.364	13.682	4.56	9.69	90.31
1.180	21.641	25.666	23.654	7.88	17.57	82.43
0.600	102.434	93.605	98.020	32.67	50.24	49.76
0.425	37.250	38.408	37.829	12.61	62.85	37.15
0.300	36.185	36.504	36.345	12.12	74.97	25.03
0.150	48.640	48.853	48.747	16.25	91.22	8.78
0.075	17.930	17.777	17.854	5.95	97.17	2.83
Pan		8.407	8.476	2.83	100.00	0
Total			300.000			

Table C.3 SK3 sieve analysis

Sieve size (mm)	Mass 1 retained (g)	Mass 2 retained (g)	Mass 3 retained (g)	Ave. Mass retained (g)	% mass retained	Cumulative mass retained (%)	% passing
6.700	0	0	0	0	0	0	100.00
4.750	3.965	3.352	5.261	4.193	1.40	1.40	98.60
2.360	41.654	40.227	38.001	39.961	13.33	14.73	85.27
2.000	23.935	16.523	22.216	20.891	6.97	21.70	78.30
1.180	25.742	28.491	25.652	26.628	8.88	30.58	69.42
0.600	73.623	71.031	75.196	73.283	24.45	55.03	44.97
0.425	20.391	23.172	20.238	21.267	7.09	62.12	37.88
0.300	23.580	24.735	24.689	24.335	8.12	70.24	29.76
0.150	35.785	35.043	35.210	35.346	11.79	82.03	17.97
0.075	25.797	31.560	27.727	28.361	9.46	91.49	8.51
Pan	24.836	25.866	25.810	25.504	8.51	100.00	0
Total				299.769			

Table C.4 SK5 sieve analysis

Sieve size (mm)	Mass retained (g)	% mass retained	Cumulative mass retained (%)	% passing
9.500	0	0	0	100.00
6.700	0.903	0.30	0.30	99.70
4.750	7.166	2.39	2.69	97.31
2.360	62.194	20.74	23.43	76.57
2.000	21.569	7.19	30.62	69.38
1.180	24.341	8.12	38.74	61.26
0.600	59.622	19.88	58.62	41.38
0.425	13.896	4.63	63.25	36.75
0.300	22.287	7.43	70.68	29.32
0.150	35.194	11.73	82.41	17.59
0.075	30.885	10.30	92.71	7.29
Pan	21.886	7.30	100	0
Total	299.943			

Table C.5 SK8 sieve analysis

Sieve size (mm)	Mass 1 retained (g)	Mass 2 retained (g)	Mass 3 retained (g)	Ave. Mass retained (g)	% mass retained	Cumulative mass retained (%)	% passing
4.750	0	0	0	0	0	0	100.00
2.360	0.443	0.725	0.531	0.566	0.19	0.19	99.81
2.000	1.296	1.525	1.807	1.541	0.52	0.71	99.29
1.180	6.241	5.172	3.862	5.092	1.71	2.42	97.58
0.600	23.056	18.759	23.297	21.704	7.28	9.7	90.3
0.425	41.863	42.112	39.291	41.089	13.78	23.48	76.52
0.300	50.658	51.495	54.678	52.277	17.54	41.02	58.98
0.150	104.033	100.811	103.581	102.808	34.49	75.51	24.49
0.075	53.585	60.447	56.703	56.912	19.09	94.60	5.4
Pan	16.706	15.488	16.140	16.111	5.40	100.00	0
Total				298.100			

Table C.6 SK9 sieve analysis

Sieve size (mm)	Mass 1 retained (g)	Mass 2 retained (g)	Mass 3 retained (g)	Ave. Mass retained (g)	% mass retained	Cumulative mass retained (%)	% passing
9.500	0	0	0	0	0	0	100.00
6.700	22.798	17.180	47.385	29.121	9.77	9.77	90.23
4.750	7.318	9.530	7.594	8.147	2.73	12.50	87.5
2.360	18.473	14.811	20.176	17.820	5.98	18.48	81.52
2.000	7.249	6.629	7.348	7.075	2.37	20.85	79.15
1.180	5.728	5.865	5.013	5.535	1.86	22.71	77.29
0.600	40.460	31.811	33.884	35.385	11.87	34.58	65.42
0.425	24.857	23.806	17.222	21.962	7.37	41.95	58.05
0.300	22.749	28.353	15.440	22.181	7.44	49.39	50.61
0.150	64.005	91.444	82.146	79.198	26.56	75.95	24.05
0.075	51.198	45.993	35.145	44.112	14.80	90.75	9.25
Pan	31.636	24.211	26.935	27.596	9.26	100.00	0
Total				298.132			

Appendix D. Qualitative analysis of salts

Figure D.1 indicates the different colours observed during the analysis of the different salts in mortar samples: whitish precipitate for sulfates, whitish-blue for chlorides and pink for nitrites and nitrates. The figures also demonstrate the formation of bubbles for the carbonate-enriched samples.

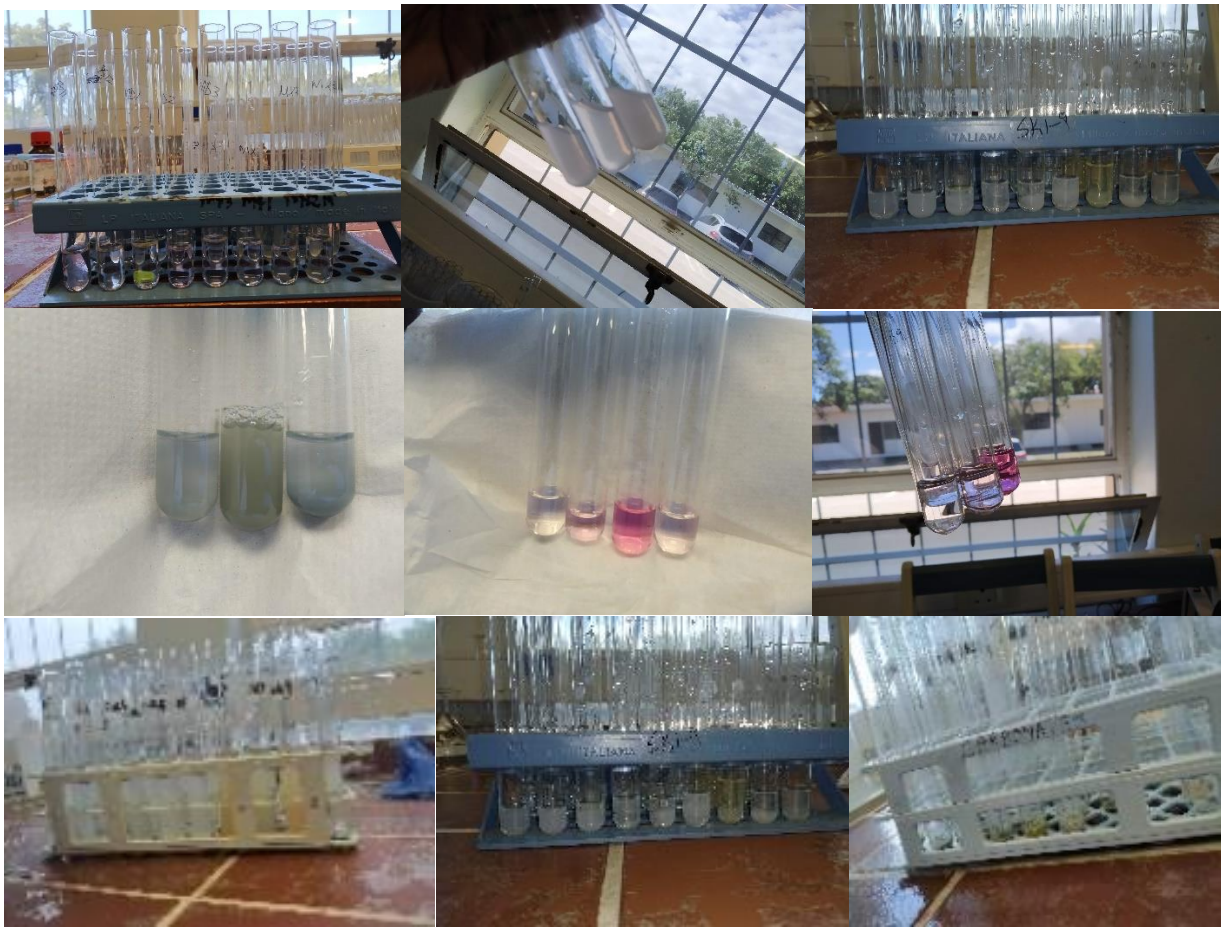


Figure D.1 Colour observations during titration

Appendix E. Binder-to-aggregate ratio

The calculations for binder-to-aggregate ratio using an acid dissolution method are detailed in this section. The advisable way to measure the weight of the materials as accurately as possible is to measure the beaker with the sample and the filter paper containing the residue. This reduces the chances of losing some materials during transfer into weighing instruments. Table E.1 indicates the material weights before and after acid dissolution, which are used in the calculation of the binder-to-aggregate ratio.

Table E.1 Binder-to-aggregate ratio

Sample ID	W_1 = binder + aggregate	W_2 = residue after boiling in 2N HCl & wash with H_2O = acid insoluble aggregate	W_3 = insoluble boil in Na_2CO_3 & wash in HCl then with H_2O = hydraulic components	$b/a = \frac{(W_1 - W_2)}{W_2}$
SK1	10,0844	6,5870	6,3626	0,53
SK2	10,0074	7,9851	7,6754	0,25
SK3	10,0092	6,3646	6,1778	0,57
SK4	10,0094	6,5534	6,0938	0,53
SK5	10,0325	6,4108	6,070	0,56
SK6	10,0331	6,3072	6,3040	0,59
SK7	10,0311	8,1500	7,1758	0,23
SK8	10,0722	9,6727	8,8979	0,04
SK9	10,0625	8,8314	6,4996	0,14
MX1A	5,0595	2,4598	2,1658	1,06
MX2	5,0110	2,7922	2,5913	0,79
MX3	5,0343	2,8158	2,6122	0,79
MD1	5,0908	2,3403	2,1491	1,18
MD2	5,0066	2,8749	2,6390	0,74
MD3	5,0220	2,7740	2,6119	0,81
PS1	10,0340	6,2265	5,9157	0,61
PS2	10,0322	6,3716	5,9761	0,57
PS3	5,0994	2,1815	1,8386	0,57
PS4	10,0051	7,3546	6,9896	1,34
PS5	10,0365	6,9828	6,7801	0,36
PS6	10,0305	7,0717	6,8351	0,44
PS7	10,0211	6,7486	6,4569	0,42
PS8	10,0615	9,1412	6,8923	0,48
PS9	10,0507	7,3556	7,2380	0,10
PS10	10,0316	7,0058	6,7847	0,37
PS11	10,0736	7,1425	6,9078	0,43
PS12	10,0653	6,9878	6,8276	0,41

Appendix F. Quantitative analysis of oxides

The calculations for the oxides and the picture gallery of the procedure are attached in this section. Tables F.1 and F.2 show the volumes of the indicators as obtained during the titration process, as well as the calculated oxides using the Equations 3.5 to 3.9. The samples whose end points during titration did not indicate any expected colour change were regarded to have no traces of the metal oxide, in accordance with the method used. Therefore, such samples are concluded to have zero content of the metals in question.

Table F.1 Titration using EDTA and $ZnSO_4$

Sample ID	$V_{EDTA.Ca}$	$V_{EDTA.Ca+Mg}$	V_{EDTA}	V_{ZnSO_4}
SK1	27,50	20,04	No pink colour	No pink colour
SK2	26,20	20,20	No pink colour	No pink colour
SK3	20,00	25,40	No pink colour	9,40
SK4	28,75	22,30	7,00	No pink colour
SK5	29,00	26,20	7,80	No pink colour
SK6	20,30	20,10	7,80	19,10
SK7	27,70	20,70	7,30	No pink colour
SK8	22,30	20,40	10,90	15,70
SK9	25,20	20,90	5,80	15,50
MX1A	20,10	17,55	9,80	15,00
MX2	16,10	15,20	8,70	21,10
MX3	15,40	15,60	9,20	23,30
MD1	16,00	18,10	9,50	13,90
MD2	15,00	18,80	10,50	0,00
MD3	16,50	17,90	7,60	16,00
PS1	32,30	21,40	8,40	5,80
PS2	32,00	20,70	6,80	17,70
PS3	21,00	20,30	11,00	18,10
PS4	25,00	20,10	7,30	25,50
PS5	20,00	20,00	5,70	19,20
PS6	22,00	20,00	9,30	14,50
PS7	25,00	20,00	7,30	16,50
PS8	22,7	20,10	13,00	12,90
PS9	21,00	20,00	7,80	5,12
PS10	21,00	20,50	12,50	20,90
PS11	20,00	20,00	10,60	9,70
PS12	20,10	20,00	17,70	0,00

Table F.2 Quantification of metal oxides

Sample ID	SiO_2 $\% \mathbf{Vol.SiO_2} = \frac{(W_2 - W_3)\ x\ 100}{W_1}$	CaO = 1.402 x $V_{EDTA.Ca}$	MgO = 1.008 ($V_{EDTA.Ca+Mg}$ -$V_{EDTA.Ca}$)	Fe_2O_3 = 0.998 x V_{EDTA}	Al_2O_3 = (3 - V_{ZnSO_4} x f_{ZnSO_4}) x 0.632
SK1	2,23	38,56	7,46	0,00	1,90
SK2	3,09	36,73	6,00	0,00	1,90
SK3	1,87	28,04	5,40	0,00	1,75
SK4	4,59	40,31	6,45	6,99	1,90
SK5	3,40	40,66	2,80	7,78	1,90
SK6	0,48	28,46	0,20	7,78	1,59
SK7	9,71	38,84	7,00	7,29	1,90
SK8	7,69	31,26	1,90	10,88	1,65
SK9	23,17	35,33	4,30	5,79	1,65
MX1A	5,81	28,18	2,55	9,78	1,66
MX2	4,01	22,57	0,90	8,68	1,56
MX3	4,04	21,59	0,20	9,18	1,53
MD1	3,76	22,43	2,10	9,48	1,68
MD2	4,71	21,03	3,80	10,48	1,90
MD3	3,23	23,13	1,40	7,58	1,64
PS1	3,10	45,28	10,90	8,38	1,80
PS2	3,94	44,86	11,30	6,79	1,62
PS3	6,72	29,44	0,70	18,06	1,61
PS4	3,65	35,05	4,90	25,45	1,49
PS5	2,02	28,04	0,00	19,16	1,59
PS6	2,36	30,84	2,00	14,47	1,67
PS7	2,91	35,05	5,00	16,47	1,64
PS8	22,35	31,83	2,60	12,87	1,69
PS9	1,17	29,44	1,00	5,11	1,82
PS10	2,20	29,44	0,50	20,86	1,57
PS11	2,33	28,04	0,00	9,68	1,74
PS12	1,59	28,18	0,10	0,00	1,90

Figure F.1 Colour observations during titration