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Mortar and Concrete: Precursors to Modern Materials

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ABSTRACT

For thousands of years, mortar-based materials — including bedding mortars, plaster floors, internal wall plasters, external wall renders and stuccos, and concrete — have been key construction materials in many cultures throughout the world. This paper gives an overview and examples of the use and development of mortar-based materials in cultures across the Middle East, Europe, Asia, and Mesoamerica, prior to the development of hydraulic cements in the late 18th and early 19th centuries.

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1. Introduction

For thousands of years, mortar-based materials — including bedding mortars, plaster floors, internal wall plasters, external wall renders and stuccos, and concrete — have been key construction materials in many cultures throughout the world. The composition of these materials varies geographically, and they appear to have emerged independently within cultures with the establishment of permanent architecture. Historically, the type of binder used in mortar-based materials has been dependent on the availability of natural resources and, in the case of lime binders, the technology to burn calcareous materials in sufficient conditions to produce quicklime.

Portland cement has become the most used binder around the world since its invention in the 19th century; it is the primary binder for concrete — by mass, the second most consumed material in the world after water, with annual global production of approximately 30 billion tonnes (Monteiro et al. 2017) — and is widely used in bedding mortars, terrazzo, renders and stuccos.

However, the use of Portland cement today is not without issue. The environmental impact of Portland cement is of concern as its manufacture is responsible for 2–3% of energy use and 8–10% of global anthropogenic CO_2 – approximately one tonne of CO_2 is released for every tonne of Portland cement produced (Monteiro et al. 2017).

Furthermore, modern cement-based materials are widely understood to be incompatible with many traditional building materials found in historical architecture and should not be used in the conservation of these structures.

As such, it is important to investigate the role traditional materials can play, both in terms of conserving existing architectural heritage and as an alternative in new constructions to materials that are more energy and emissions intensive. This paper does not intend to address these questions directly but, instead, provides an overview of the development of mortar-based materials in cultures across the world and examples of their use which have endured for centuries or, in some cases, millennia. These are detailed chronologically within different geographical regions, beginning with the earliest examples that appear in the Middle-East, before moving on to Europe where much of the available literature is focused. Significant developments in Asia and Mesoamerica, which almost certainly occurred independently from those occurring elsewhere, are also discussed. Unfortunately, the extent of this discussion in some cultures is limited due to the relatively small amount of internationally available publications on the subject.

The history of these materials is particularly convoluted due to the fact that they have been written about by authors from several different fields, including archaeology, architectural history, heritage conservation, and material science. Many authors have used terms such as 'concrete' and 'cement' without providing

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a clear definition and used the terms 'cement', 'binder' and 'mortar' interchangeably. As such, it is important to clarify the terminology that will be used in the subsequent discussion.

'Mortar' is a mixture of binder, fine aggregate and water, which hardens (British Standards Institution 2014) — with 'fine aggregate' considered to be that which is ≤4 mm (British Standards Institution 2008).

Clearly defining 'concrete' is more complex. BS ISO 6707–1 (British Standards Institution 2014) defines 'concrete' as a "mixture of aggregate, cement and water, which hardens," and 'cement' as "finely ground inorganic material that, when mixed with water, forms a paste that sets by means of hydration reactions processes, and that, after hardening, retains its strength and stability, even underwater."

However, this definition excludes any material not made with cement. While the term 'cement' has been used by some authors to describe a variety of binders, including nonhydraulic binders, and both naturally and artificially hydraulic binders, it should be reserved for hydraulic binders which do not contain a high enough free lime content to be slaked (Spalding 1903). This presents an issue as the term 'concrete' is widely accepted as being correct when used to describe many materials which are not made with cement, such as those made by the Romans.

One of the earliest uses of lime in construction was for interior floors, often referred to as 'plaster' floors in the literature. However, unlike 'plaster' in the modern sense, these floors often contained 'coarse' aggregate (>4 mm) and were much thicker than the plaster that is used to coat interior walls and ceilings. In these cases, they were not merely a coating but, instead, form a structural element and could be considered to be the first step in the development of the material we know today as 'concrete'. Thus, in the subsequent discussion, the authors propose the term 'concrete' includes all composite materials that serve a structural purpose and are made from a mixture that includes coarse aggregates and binder which hardens.

2. Overview of binder types

2.1. Air lime

Lime, which was first manufactured as early as 12,000 BCE (Kingery, Vandiver, and Prickett 1988), is created by burning limestone ($CaCO_3$) causing it to calcine and form quicklime (CaO). While the decomposition temperature of limestone varies due to factors such as its composition and the ambient pressure during burning, in general, to thoroughly calcine limestone, a constant temperature of 750–850°C is required for several hours (Gourdin and Kingery 1975) and in order to manufacture tonnage quantities of quicklime, temperatures of 800–900°C have to be sustained for three or four days (Kingery, Vandiver, and Prickett 1988). However, the operational temperatures of modern lime kilns are typically in the range of 920–1000° in order to maximise the speed of the process (Artioli, Secco, and Addis 2019).

$$CaCO_3 \rightarrow CaO + CO_2 \tag{1}$$

Quicklime is slaked by the addition of water and forms a paste of calcium hydroxide ($Ca(OH)_2$):

$$CaO + H_2O \rightarrow Ca(OH)_2 \tag{2}$$

This paste sets and hardens slowly, losing its plasticity due to the evaporation of water, and gaining strength over a long period of time by reaction with atmospheric carbon dioxide — producing calcium carbonate. As such, air limes cannot be set under water and therefore are not 'hydraulic'.

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O \tag{3}$$

While the resulting product is identical to the original limestone, both in chemical and crystalline composition, it does have a different microstructure (Kingery, Vandiver, and Prickett 1988).

Today, two sub-families of air lime are recognised in Europe (British Standards Institution 2015); calcium lime (CL), which consists mainly of calcium oxide/hydroxide, and dolomitic lime (DL) which consists mainly of calcium magnesium oxide/hydroxide. These air limes can be supplied in different forms, either as quicklime (lump or powder) or hydrated lime (powder, putty or slurry/milk of lime).

2.2. Gypsum

Gypsum binders require a much lower burning temperature than lime. When pure gypsum (calcium sulfate dehydrate, $CaSO_4 \cdot 2\,H_2O$) is heated in the range 100–190°C three quarters of the chemically combined water is driven off, resulting in hemihydrate, $CaSO_4 \cdot ½H_2O$ — also known as plaster of Paris (Gourdin and Kingery 1975) as it was first imported from the quarries at Montmartre near Paris (Goode 2018).

$$2CaSO_4 \cdot 2H_2O \rightarrow 2CaSO_4 \cdot {}^1/_2H_2O + 3H_2O \tag{4}$$

However, as the temperature is increased, more crystalline water is removed until water-free anhydrite, $CaSO_4$, is formed above 250°C (Goode 2018). While the hydration of plaster of Paris is chemically just the reverse of the above equation, the microstructure of the gypsum formed is very different, and it has a distinctive forest of well-defined, needle-like dihydrate crystals which interlock and cause cohesion. Excess water which is not involved in the chemical reaction then evaporates. While this results in a large amount of open porosity, it also causes the remaining dihydrate in solution between the needle-like crystals to precipitate and bring rigidity to the structure (Lewry and Williamson 1994). While it is easier to form than lime binder, its architectural use is limited due to its fast setting time, water-solubility and the ease with which it is physically damaged (Gourdin and Kingery 1975).

2.3. Lime-pozzolan mixtures

The first great advancement in lime technology came from the addition of pozzolana. Pozzolana, as defined by BS 6100–9 (2007), contains siliceous or siliceous and aluminous materials which combine with calcium hydroxide (portlandite) at ambient temperatures and in the presence of water to produce compounds having the properties of a cement.

$$Ca(OH)_2 + SiO_2 + H_2O \rightarrow Ca(OH)_2 + \text{C-S-H} \quad (5)$$

C-S-H is calcium silicate hydrate — a poorly-crystalline substance with a variable Ca-Si ratio and water content — which makes a significant contribution towards the development of strength. However, not all the portlandite will combine with silica, and the excess will react slowly over time with atmospheric carbon dioxide to produce calcite and, if the pozzolana also contains aluminate, calcium aluminate hydrates will also form in addition to C-S-H (Lechtman and Hobbs 1986).

Natural pozzolanas from a variety of sources and locations around the world have since been used in combination with both lime and modern cements and, more recently, artificial pozzolans such as fly ash, silica fume, and burned clay and shale have also been adopted. In many cultures where natural pozzolanic materials are not available, this reaction has been achieved by adding crushed clay products — such as bricks, tiles, or terracotta — that have been fired at a low temperature (600–900°C) (Baronio and Binda 1997). This material is used throughout the world today and is widely known as 'cocciopesto' in Europe, 'horasan' in Turkey, 'surkhi' in India and 'homra' in Arabic countries (Böke et al. 2006).

2.4. Hydraulic lime

'Natural hydraulic limes' (NHLs) are produced by burning limestone which naturally contains sources of silica, to create a powder which sets and hardens when mixed with water and, later, by reaction with atmospheric carbon dioxide (British Standards Institution 2015).

In addition to NHLs, there are currently two additional sub-families of products in Europe which come under the classification of the wider family of 'lime with hydraulic properties'; 'formulated lime' (FL) and 'hydraulic lime' (HL). FLs consist mainly of air lime and/or NHL with added hydraulic and/or pozzolanic materials and set and harden by both reaction with water and atmospheric CO_2. HLs consist of lime and other materials such as cement, fly ash, blast furnace slag and limestone filler, and set and harden due to the reaction with water — with atmospheric CO_2 only contributing to the hardening (British Standards Institution 2015). As the current classifications of these terms (NHL, FL, HL) are relatively recent, they are not referred to in historic literature, and the term 'hydraulic lime' is typically used to describe what we now refer to as 'natural hydraulic lime' or, in some cases, 'artificial hydraulic lime' which is manufactured when lime is burned with an added source of silica, such as clay. As such, it should be noted that any subsequent discussion of 'hydraulic limes' is referring to the historic terminology and not the current classification of HLs described in current European standards.

While the calcination of limestone can occur at 750–850°C (Gourdin and Kingery 1975), the typical temperature for NHL production is approximately between 950°C and 1250°C (Roberts 1956; Valek et al. 2014). The major hydraulic component of NHL is the dicalcium silicate phase β-belite ($\beta\text{-}C_2S$) which forms at approximately 800°C. While another polymorph of belite, $\gamma\text{-}C_2S$, can form at lower temperatures, it is not hydraulic and therefore does not contribute to setting or hardening (Roberts 1956). If alumina is present, calcium aluminates can also form in the normal temperature range of NHL production and, while not typical in NHLs, if uneven heating in the kiln leads to localised 'hot spots', tricalcium silicate or 'alite' (C_3S) can also form as a minor constituent (Valek et al. 2014). Like modern cements, the hydration of hydraulic limes produces both portlandite and C-S-H, and, if calcium aluminate was produced during the burning process, calcium aluminate hydrates will also be produced. However, unlike modern cements, hydraulic limes contain a high enough proportion of free lime (CaO) to allow them to be slaked (Callebaut et al. 2001; Valek et al. 2014), and it is this ability to slake which is typically considered to be the defining difference between 'hydraulic lime' and 'hydraulic cement' (Spalding 1903).

A comparison of different lime binders is shown in Figure 1.

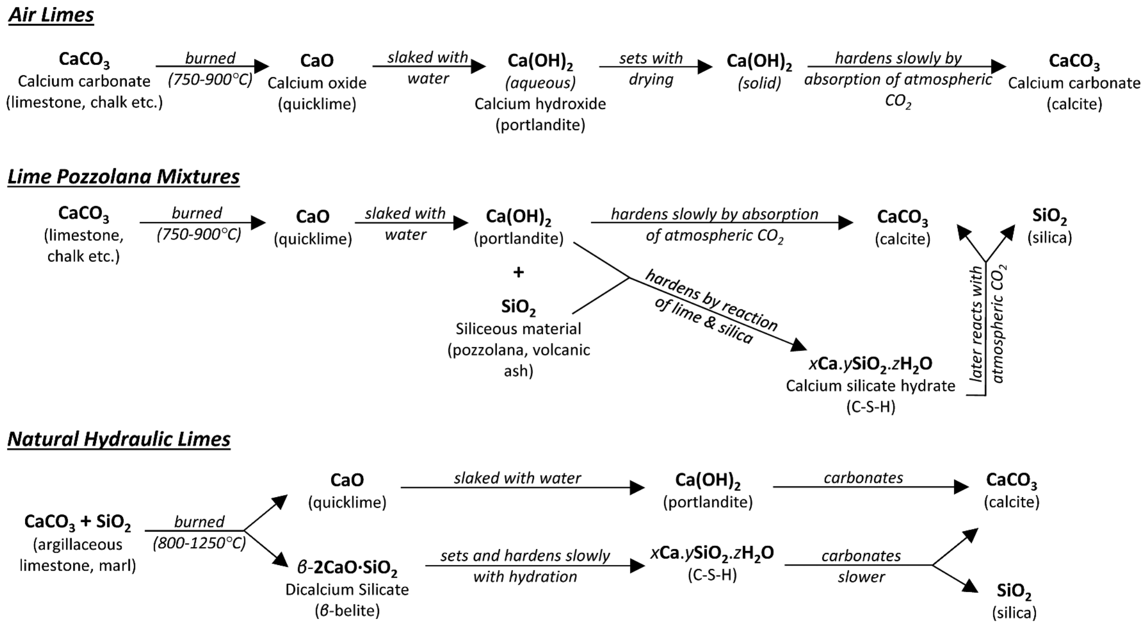

Figure 1. Comparison of different lime binders..

3. Overview of historical developments

3.1. Middle-east

The earliest known uses of lime occurred in the Middle East, with evidence of its manufacture as early as 12,000 BCE (Kingery, Vandiver, and Prickett 1988). The first known architectural use was discovered at the Late Epipalaeolithic site Nahal Ein-Gev II (NEG II), located at the Upper Jordan Valley where an approximately 40 cm thick lime plaster was used to cover bodies in a cemetery dated to c.10,000 BCE (Friesem et al. 2019). This is similar to other early examples of lime in construction, where it was typically used as the binder in flooring materials that archaeologists often refer to as 'plaster' or, if there is evidence of intentional polishing, 'terrazzo'. Discussed here are some of the most significant sites that represent major architectural use or technological change. However, significantly more examples of plaster floors exist at other archaeological sites including ʿAin Ghazal, Aswad, Beidha, Beisamoun, Wadi Ghuwayr, Jericho, Kfar HaHoresh, Ramad, and Shaqarat Mazyrad (Clarke 2012; Garfinkal 1987; Kingery, Vandiver, and Prickett 1988). Descriptions of further sites in the region where lime plaster was used for decorative functions are provided by Gourdin and Kingery (1975) and Kingery, Vandiver, and Prickett (1988).

3.1.1. Göbekli Tepe

Göbekli Tepe (Figure 2) is located in the Şanlıurfa province of south-east Anatolia on top of a large limestone plateau and features monumental round-oval and rectangular megalithic structures that were erected between 9600 and 8200 BCE during the Pre-Pottery Neolithic age (Schmidt, Merbach, and Pant 2017; UNESCO 2022a). The site, which covers about 9 hectares, has only been partially excavated. The round enclosures are typically made of concentric stone walls, connecting a 10–30 m diameter circle of monolithic T-shaped limestone pillars, at the center of which are two freestanding monolithic pillars (Schmidt 2002). The rectangular buildings measure approximately 3 × 4 m, feature stone walls and possess smaller and fewer pillars or no pillars at all. There are many instances of lime plaster floors (Haklay and Gopher 2020; Schmidt, Merbach, and Pant 2017), also referred to as 'terrazzo floors' in the literature, which began to erode following their excavation and have since been covered and backfilled for their conservation. Numerous prehistoric quarries have been identified at the site as the source of limestone for both the monuments and the lime plaster, as have workshop areas which feature possible areas where the lime plaster was mixed (Schmidt, Merbach, and Pant 2017).

3.1.2. Aşıklı Höyük

Aşıklı Höyük (Figure 2) is located in Central Anatolia, 25 km south-east of Aksaray, near the village Kızılkaya, on an alluvial plain on the banks of the Melendiz river, which lies in a basin formed by the eruption of volcanic mountains of Hasandağı and of the Melendiz-range (Esin et al. 1991; Hauptmann and Yalcin 2000). The site, which may have been occupied from approximately 7600–6800 BCE (Todd 1968), includes the remains of rectangular and trapezoidal structures built of mud-brick which have walls approximately 1–1.5 m high, made from bricks approximately 60 cm wide and 5.5 cm thick (Todd 1966). They consist of 1–3 rooms, with each room varying between 2 × 3 m and 4 × 4 m and covering an area of 6–16 m^2. In many cases, the walls and floors are painted with yellow, pink or red clay plaster, and in some cases, the floors were overlain with a layer of highly polished lime plaster approximately 6–8 cm thick (Esin et al. 1991).

Analyses of a sample from a plastered floor at Aşıklı Höyük undertaken by Hauptmann and Yalcin (2000) suggests that the hardening of the plaster was due to a pozzolanic reaction, and the researchers claim this to be the first recorded reaction of its kind. They present two possible explanations for its occurrence. The first, that silicious ignimbrite, which was available in large quantities around the settlement, was mixed with quicklime during slaking, and the second, that calcium-rich clay with a high volcanic ash content was burned with the limestone. However, it is likely that the latter scenario would not result in a pozzolanic reaction but, instead, would result in the manufacture of an artificial hydraulic lime.

3.1.3. Çayönü Tepesi

Çayönü Tepesi, which lies at the foot of the Taurus Mountains and by a tributary of the upper Tigris in south-eastern Turkey, is an archaeological site that was excavated between 1964 and the 1990s, with the majority of the excavated levels dating to around 7250–6750 BCE. Several of the buildings at the site have floors which were coated in a thin layer of lime plaster (Schirmer 1990) and one particular structure at the site, a 9 × 10 m structure known as the ‘terrazzo building’, has a solid rectangular floor that was carefully laid.

The floor varies in thickness from 5 to 20 cm, but typically consists of an approximately 12 cm thick layer of crushed limestone fragments embedded in a lime mortar which had been burned and slaked, set on a bed of coarse limestone fragments, and with a top layer of pink limestone pieces, 1–3 cm in diameter, embedded in a pink lime mortar approximately 1 cm thick. Integrated into this top layer are two sets of parallel white stripes, approximately 5 cm wide and 4 m long, that are made of crushed white limestone that were set into the surface. The top surface was ground and polished after it had hardened, giving it a terrazzo-like appearance (Braidwood et al. 1971; Haklay and Gopher 2019; Schirmer 1990).

3.1.4. Yiftahel

The archaeological site of Yiftahel (Figure 2) is located south of the Bet Netofa valley in Lower Galilee, Israel, and is now thought to have had two major periods of occupation; 8000–7000 BCE (Pre-

Figure 2. Neolithic sites with plaster floors. (a) Göbekli Tepe [Beytullah Eles 2019, CC BY-SA 4.0]. (b) Aşıklı Höyük [Sarah Murray 2010, CC BY-SA 2.0]. (c) Yiftahel [Garfinkel Yosef, CC BY-SA 3.0]. Lepenski Vir [Cvetinovic Dejan 2021, CC BY-SA 4.0].

Pottery Neolithic B) and 3600–3300 BCE (Early Bronze Age IA) (Khalaily et al. 2008). Excavations, which began in 1982, uncovered a rectangular house with stone walls 0.4 m high and a hard plastered floor 4–8 cm thick which covered an area approximately 65 m^2. The plaster consists of two layers: a base layer approximately 45 mm thick and a top finishing layer 5 mm thick. Compressive strength tests carried out on 25 mm cubes gave average results of 33.8 MPa for cubes of the base layer only and 44.8 MPa for cubes of made of both base and finishing layer. Analyses of both layers indicated that they were composed of mainly $CaCO_3$ (Ronen, Bentur, and Soroka 1991).

Subsequent excavations have since revealed more lime plaster floors, and seeds found on them have been dated to 6840 ± 50 BCE (Ronen, Bentur, and Soroka 1991). One particularly large plaster floor was uncovered; produced from a mixture of quicklime, water and stone, it was laid on an even base of sandy clay to form a 180 m^2 floor that varied in thickness between 3 and 8 cm. It is mainly in one homogenous layer which resembles travertine in appearance and had been intentionally polished. It is estimated that more than 9 m^3 of plaster was required to make the floor, with more than 2 metric tons of lime binder (British Cement Association 1999). Laboratory analyses of samples taken from the site revealed that the material had high density, low water absorption and unexpectedly high strength, and most likely contained both aggregate and binder that were calcareous (Malinowski and Garfinkel 1991).

3.1.5. Ancient Egypt

Although it was only moist mud or clay that was used as mortar between sun-dried bricks in most ancient Egyptian construction (Davey 1961, Lucas 1959 [1948]; Moropoulou, Bakolas, and Anagnostopoulou 2005), it has been proposed that lime mortars, gypsum mortars and concrete were incorporated into larger, monumental structures such as the Great Pyramid of Giza (Figure 3). Stanley (1979) claims that a mural in Thebes dating to c.1950 BCE depicts various stages in the process of manufacture and application of mortar and concrete, and provides two pictures which appear to be modern reproductions of the original mural. The first allegedly shows workmen filling earthenware jars with water which is then mixed with lime and used as a mortar for stone masonry, and the second, a section of a concrete wall being constructed and faced with stonework on both sides. However, Stanley's interpretation appears to be speculative and no source confirming it is supplied.

While some authors believe the material used in the Great Pyramid was lime-based, many consider that it was more likely to have been produced from burnt gypsum (Stanley 1979; Davey 1961; Blezard 1998; British Cement Association 1999; Reid 1877, Lucas 1959 [1948]). Lucas (1959 [1948]) states that lime mortar was not used in ancient Egypt before Greco-Roman times, and that no known examples of its use prior to the time of Ptolemy I (323–285 BCE), and reasons that the preference for using gypsum over lime, despite the abundance and accessibility of lime, was due to the scarcity of fuel for lime burning. Some authors (Moropoulou, Bakolas, and Anagnostopoulou 2005) have stated that the purpose of the gypsum mortars between the stone blocks was not to act as a joint but, instead, was mainly used as a lubricating agent to assist in accurately arranging the blocks. Petrographic and X-ray diffraction analysis by Klemm and Klemm (1990) has since proven the use of lime in the Old Kingdom of Egypt. However, this was never seen as a pure lime mortar but, instead, as gypsum/lime mixtures in varying ratios, with the lime content increasing significantly between the 3rd and 5th Dynasties before significant reducing during the 6th Dynasty (c.2345–2181 BCE).

Additional confusion on this issue arises upon the examination of the material by French chemist Louis-Joseph Vicat who, after carrying out chemical analysis of mortar allegedly from the Great Pyramid, claimed that the mortar is lime "*exactly similar to our mortars in Europe*" (Vicat 1837). However, additional notes written by Captain John Thomas Smith (Smith 1837), who translated Vicat's original work into English, claim that Smith conducted his own analysis on mortar from the Great Pyramid and that it was very different from that described by Vicat. Smith concluded that the material he examined himself contained no siliceous matter but was composed of rich lime and coarsely powdered gypsum which was used as a substitute for sand in the mortar, with a ratio of 1:5 of lime to gypsum, and casts doubt on the origins of the mortar submitted to Vicat .

More recent research into the construction of the Great Pyramid has proven controversial. Davidovits (1984) proposed the hypothesis "*that the limestone that constitutes the major pyramids of the Old Kingdom of Egypt is man-made stone*," formed by a complicated geopolymeric system. He supported this by comparing chemical data from X-ray analysis, mineralogical data from X-ray diffraction (XRD) of samples from the pyramid casing stones, quarries local to the pyramids, and his own laboratory reproduction of crushed limestone with a mineral binder composed of synthetic zeolite. Additionally, thin-section analysis of the pyramid casing stones revealed the presence of what are possibly air bubbles and organic matter which he argues are not characteristic of natural stone but, instead, support his hypothesis that the stone blocks are man-made.

While some authors have shown support for Davidovits' theory (Barsoum 2007; Barsoum, Ganguly, and Hug 2006; MacKenzie et al. 2011; Morris 1987, 1991), several rebuttals of Davidovits' work have been published (Folk and Campbell 1991; Harrell and Penrod 1993; Ingram, Daugherty, and Marshall 1993; Jana 2007; Klemm and Klemm 1990), for some of which there have been subsequent counter-rebuttals (Morris 1992, 1993) and there is, at present, no scientific consensus on the issue.

3.1.6. Mesopotamia

Other sources of mortar have also been used extensively throughout the Middle East for thousands of years and can be seen in the excavations of Mesopotamian sites. While evidence of the use of natural asphalt (bitumen) as an adhesive for the construction of tools dates back as far as ≈ 70,000 BCE (Boëda et al. 2008), evidence of its use in construction is much more recent. While asphaltic mortars — which were prepared by mixing bitumen with chopped straw, clay and sand — were occasionally used for roadways and ordinary domestic dwellings, they were predominantly used in the construction of more prestigious structures, such as temples, palaces, terraces and ziggurats. The waterproofing ability of bitumen meant it was also used extensively for lining baskets, jars, water reserves, bathrooms, water pipes, cisterns, boats and sarcophagi (Connan et al. 1999).

Excavations at the archaeological site of Khafaje (Frankfort, Jacobsen, and Preusser 1932), located 15 km east of Baghdad, have shown that a variety of different building materials were in use by the 3rd millennium BCE. These included bricks made from clay, mixed with chopped straw to increase their strength, which were typically sun-dried, but occasionally baked when required — such as when the structure was in contact with water. While sun-dried bricks were typically bonded with a mortar made from the same clay mixture to form a homogenous mass, baked bricks were found laid in bitumen — presumably to make the structures watertight — and both mud plaster and bitumen were used to coat walls and floors. In addition to these, lime plastered floors and walls were also excavated, as well as a jar containing lime. Several kilns were discovered at the site, one of which was determined to be a lime kiln following a chemical analysis of its contents.

The city of Babylon — located 85 km east of Baghdad — experienced significant expansion and rebuilding during the reign of Nebuchadnezzar II (604–562 BCE) and baked bricks largely replaced sun-dried bricks during this period (Pedersén 2021). Davey (1961) claims that a change in mortar also occurred during this period, with asphaltic mixtures being largely replaced by mixtures of hydrated lime, to which clay, bitumen, ashes and other materials were sometimes added. However, Koldeway (1914), who carried out early excavations of Babylon, reports that Nebuchadnezzar only used lime mortar in the latest buildings of his reign, such as the Kasr, the Principal Citadel and Babil. Even in these cases, lime mortar or mixtures of lime and gypsum, typically only replaced asphaltic mortar in the upper parts of structures — as witnessed in the Northern Palace area, and to a limited extent in some upper parts of the Ištar Gate (Pedersén 2021) (Figure 3) — and specifically those not in direct contact with water (State Board of Antiquities and Heritage Iraq 2018). Babylon fell to the Persian Empire in 539 BCE, and Forth (2009) claims that the Persians abandoned the use of bitumen there entirely and, instead, favoured the use of lime mortar.

3.1.7. Persia

A type of mortar known as 'sarooj' has been used since at least 1250 BCE in Iran and has since been used across the Persian Gulf (Masoumi, Banakar, and Boroomand 2015), Oman and Afghanistan (Al-Rawas et al. 1998; Soleymani, Najafgholipour, and Johari 2022). The composition of sarooj appears to vary depending on its intended use, local custom and geographical location.

Masoumi, Banakar, and Boroomand (2015) claim the basic ingredients are lime, clay and sand, though other additions — such as ash from burned animal dung, natural fibres from plants and animals, and organic additives such as milk and egg — have also been reported. Two different types of sarooj are found in Iran: 'sarooj sard' (cold sarooj) and 'sarooj gard' (warm sarooj). Sarooj gard is a hydraulic mortar, obtained from burning lime which contains clay (natural hydraulic lime), and is abundantly found in the south of Iran along the northern coast of the Persian Gulf. Sarooj sard is manufactured by mixing lime, ash and water (lime-pozzolana mixture), with clay and sand also added (Masoumi, Banakar, and Boroomand 2015).

However, other authors (Makarchian and Khodaverdian 2011) have claimed that the word sarooj is derived from the middle Persian word 'charook', which means something compounded of four different materials — in this case lime, ash, water and cattail flower. Makarchian and Khodaverdian (2011) also describe two different types of sarooj: 'air-setting sarooj' and 'hydraulic sarooj'. Air-setting sarooj is typically used for ordinary buildings and is made from a mixture of quicklime, ash and water, and forms a stiff paste which is mixed and compacted vigorously for 12 h before being applied to the work. Hydraulic sarooj, which is used for structures in contact with water, has burnt clay powder added.

Several authors (Al-Rawas et al. 1998; Hago, Al-Rawas, and Al-Riyami 2002; Hago, Al-Rawas, and Al-Sidairi 2002; Meddah et al. 2020) report that, in Oman, 'sarooj' is a local term referring specifically for an artificial pozzolana produced by calcining clay. This material is mixed with lime and water to make a hydraulic mortar, which has been used in Oman for thousands of years in buildings, forts, and water channels.

While sarooj mortar was typically used to plaster hydraulic structures (Soleymani, Najafgholipour, and Johari 2022), it was also used as a masonry mortar in structural elements. In Iran, it was often used in masonry bridges prior to the introduction of Portland cement (Makarchian and Khodaverdian 2011), such as at Si-o-se-pol (also known as the Allahverdi Khan Bridge) built 1599–1602 (Figure 3). Other examples of historic monuments to incorporate sarooj are the Anahita temple and the ziggurat Chogha Zanbil (Soleymani, Najafgholipour, and Johari 2022) (Figure 3) — probably the oldest structure known to incorporate sarooj mortar, dating to approximately 1250 BCE (Masoumi, Banakar, and Boroomand 2015).

3.1.8. Cyprus

Lime and gypsum plasters have been used on the island of Cyprus since the Neolithic period and appear to be connected to the emergence of permanent architecture. As in other cultures in the region at this time, lime plasters were used extensively to form floors and to coat walls. While the earliest lime plaster floors were typically thin layers approximately 5 mm thick laid over a base layer of mud, floors of multiple layers have also been observed and are the result of long periods of reuse leading to many successive coatings and production of thicker floors over time. During the Chalcolithic period (c.5th – 3rd millennium BCE), the hardness and thickness of lime plaster floors increased significantly and likely coincides with a thorough dissemination of lime burning technology during this time (Philokyprou 2012).

While natural pozzolans were not available in Cyprus, artificial pozzolans in the form of baked clay or ceramics have been used since the Late Bronze Age (1200 BCE), coinciding with a period of prosperity and the emergence of urban centres with important buildings which had sanitary facilities and drainage systems (Philokyprou 2012; Theodoridou, Ioannou, and Philokyprou 2013). Due to the seemingly purposeful use of these mortars to line hydraulic structures, such as wells and water canals, it can be concluded that there must have been at least an empirical understanding of their waterproofing ability (Theodoridou, Ioannou, and Philokyprou 2013).

Figure 3. (A) the Great Pyramid [Nina Aldin Thune 2005, CC BY-SA 3.0]. (b) Excavation of the Ištar Gate [David Stanley 2016, CC BY-SA 2.0]. (c) Chogha Zanbil [Carole Raddato 2019, CC BY-SA 2.0]. (d) Si-o-se-pol [Ninara 2012, CC BY-SA 2.0].

3.2. Europe

The use of lime appears to have occurred in Europe several thousand years later than it did in the Middle East. Even then, the use of lime does not appear to be widespread throughout Europe during the Neolithic era, and only a few examples are known from this period. However, the developments in lime and concrete technology that would eventually take place in Rome would far surpass anything that had come before, with many great concrete structures constructed across the empire — some of which are still in use today.

3.2.1. Lepenski Vir

Excavations at Lepenski Vir (Figure 2) have discovered red lime plaster that had been used to make hut floors. Lepenski Vir lies on the banks of the Danube, and the red lime was brought from almost 200 miles upstream, suggesting its users had some knowledge of its properties, and mixed it with sand, gravel and water to produce what could be considered to be the earliest known concrete in Europe (British Cement Association 1999; Stanley 1979). This material, which dates to around 5600 BCE, was laid and compacted to form a floor ranging from 1 to 25 cm thick and incorporated a stone hearth at one end.

In the English translation of their book (Srejović and Edwards 1972), describe the floors at the archaeological site as being constructed of a mass of sandy, marly red limestone from the Koršo hills. The floors were prepared by baking the limestone and then adding water, sand and gravel to form a mixture that was poured over the foundations and embedded all the construction details (the stone blocks which form the hearth, the thresholds, a rounded boulder in the centre and the stone slabs which line the post holes that support the upper construction). The floor was then smoothed before it had hardened and decorated with a thin red or white composition.

3.2.2. Ancient Greece

The use of lime plaster for floors in Greece dates back to at least the Late Neolithic period (6th millennium BCE) and has been found at archaeological sites at Makri, Thrace, and Drakaina Cave, the Ionian Islands (Karkanas 2007). The floors at Makri are described as being a sequence of whitish hard floors and brownish-grey compact layers, the thickness of each layer varying from 1 to 5 cm. Petrographic analyses concluded that these consisted of pure lime plaster with large amounts of lime lumps and half-burnt tufa remnants, and impure lime floors consisting of a mixture of burnt lime, anthropogenic debris, and siliciclastic sediment. The floors at Drakaina Cave are described as being whitish hard plaster floors, found to be made of almost pure lime plaster, though containing large amounts of lime lumps, half-burnt fossiliferous marl remnants and occasional clay admixtures.

The use of lime became more prevalent in ancient times, with notable examples of its extensive use in multiple structures at the UNESCO World Heritage Sites of Mycenae and Tiryns (1600–500 BCE) (Greek Ministry of Culture 1998a, 1998b; ICOMOS 1999; Schliemann 1885). During the excavations of Tiryns, Schliemann (1885) notes many instances of lime concrete floors, some of which were patterned and coloured. Chiotis et al. (2001) describes the lime plaster floor of the Tiryns Palace courtyard as being composed of a reddish undercoat 10–15 mm thick, rich in fragments of terracotta, and a white superficial lining up to 0.5 mm thick.

In addition to the numerous examples of floors, lime plaster was used on the walls in many places at Tiryns (Schliemann 1885) and an underground reservoir constructed at Mycenae in the 12th century BCE has walls which are described as being "*clad in hydraulic cement*" (ICOMOS 1999), and "covered with a thick coat of hard, watertight stucco, which has a fine smooth upper layer laid over a rather coarse backing" (Wace 1949). Analyses of the cistern plaster by Chiotis et al. (2001) revealed that it is composed of a thin (0.2 mm) two layer surface lining, on top of a 20–30 mm thick undercoat consisting of lime mixed with rounded aggregates 1–10 mm in diameter. There is no indication that hydraulic materials were used in the undercoat to make it watertight. Instead, the porosity of the undercoat was reduced by abrading it smooth once it had set, thus causing mechanical interlocking of the grains, and then a first waterproofing layer of almost pure lime was spread, followed by a second thinner layer of siliceous composition — possibly a mixture of lime, calcined gypsum and clay which may have also been calcined to produce a pozzolanic reaction. However, samples from another ancient Greek cistern analyzed by Chiotis et al. (2001) — an open-air cistern at the Argive Heraion, located between Mycenae, Tiryns and Argos — were rich in fragments of terracotta and pulverised terracotta which, in addition to the same smoothing technique seen at the cistern in Mycenae, improved its waterproofing. The use of mixtures of lime and crushed terracotta has also been reported by Shaw (2009) who gives examples of its use in wall and floor plasters in Minoan Crete as early as the late bronze age (c.1200 BCE).

By 500 BCE lime concrete was being used in ancient Greece with a relatively high degree of skill and with knowledge of the effects of highly siliceous, volcanic Santorin earth which started to be used sometime between 500 and 300 BCE (Idorn 1997). Evidence of the ancient Greek skill and knowledge of concrete was discovered during the archaeological excavation of the ancient city of Kamiros on the island of Rhodes, where a great waterstorage tank with a capacity of 600 m^2 was unearthed close to the temple of Athena of Kamiros (Koui and Ftikos 1998). The concrete used in the water tank construction combined a mixture of siliceous gravel, granular intermediate calcareous aggregates and fine-grained aggregates with a binder consisting of volcanic earth and lime; forming a concrete of such high quality that it was found to have excellent physical and mechanical properties, despite three millennia of weathering.

The ancient Greeks reportedly also made use of lime-based compositions as a render for porous limestone used in temples, as a binding material between bricks and stone, and to cover walls of sun-dried bricks (British Cement Association 1999) — with bricks being their preferred method for constructing walls and used to construct private houses, public buildings, and royal homes (Pliny 1898 [c.77 CE]; Vitruvius 1914 [c.25 BCE]). Vitruvius describes the house of King Mausolus at Halicarnassus as being decorated throughout with Proconnesian marble, but with walls built of brick of extraordinary strength and covered with stucco *"so highly polished that they seem to be as glistening as glass"*. Vicat (1837) claims that the houses of ordinary citizens in Athens were decorated with calcareous stucco which, in terms of whiteness, hardness and polish, was comparable to Parian marble.

3.2.3. Ancient Rome

The invention of concrete is often incorrectly attributed to the Romans. However, the word 'concrete' does come from the Latin 'concretus' meaning 'grown together' or 'compounded' (Stanley 1979), and perhaps the most significant period in the history of concrete began around 300 BCE when the Romans began to develop and use a form of structural concrete, known as '*opus caementicium*', for ambitious construction projects.

While there is no clear date for the first use of lime or concrete in Rome, the writings of Cato (1935 [c.160 BCE]), detail the construction and operation of a large lime kiln — ten feet across, twenty feet high, and with sides sloping to a width of three feet at the top — and the use of lime mortar for the construction of walls and foundations. Some of the earliest examples of the use of concrete in floors and foundations can be found at the temples of Castor (rebuilt 117 BCE) (Figure 4) and Concord (rebuilt 121 BCE) (Davey 1961; Frank and Stevens 1925). Even at this early stage, the amount of concrete used in the construction could be significant, with some masses of concrete at the Temple of Castor measuring as much as 16 m wide and some almost 4 m deep (Frank and Stevens 1925).

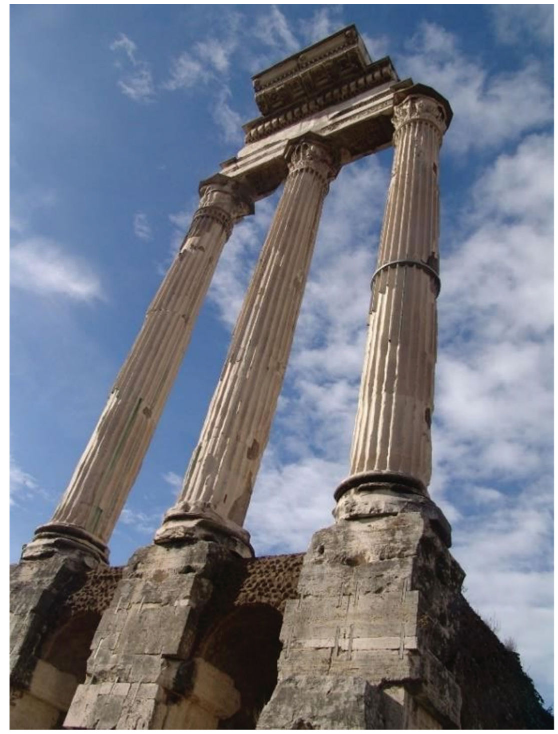

Figure 4. Temple of Castor and Pollux [Rüdiger Marmulla 2017, CC BY-SA 4.0.

By the middle of the first-century CE, masonry-faced concrete was used extensively for the construction of public buildings (Brune and Perucchio 2012; Lechtman and Hobbs 1986). While removable wooden formwork was used by the Romans for foundations, as well as for centring vaults and domes, it was rarely used for making concrete walls or piers (Lechtman and Hobbs 1986) as it is today. Instead, Roman builders created a technique which involved creating permanent masonry walls and filling the gap between the walls with lime mortar and rubble aggregate, which would harden to form a solid concrete core. Originally, this involved rough courses of irregular stone and was known as '*opus incertum*' but, by the time of Vitruvius, it was far more common to lay courses of dressed masonry, known as '*opus reticulatum*' (Figure 5). However, while *opus reticulatum* was far more aesthetically pleasing than the *opus incertum*, it was also recognized that the *opus incertum* technique created a wall which was far stronger (Vitruvius 1914 [c.25 BCE]).

The practice of concrete making in Roman times also differs from modern concrete construction as the mixture of binder and aggregate was not mixed prior to casting and was, instead, mixed in-situ, within the wall (Adam 2005). The aggregates themselves were also significantly different from those used today and included large pieces volcanic rock and coarse brick, known as '*caementa*', which have been found to be as large as 10–20 cm (Brune and Perucchio 2012; Jackson et al. 2009; Lancaster et al. 2011).

The works of Vitruvius (1914 [c.25 BCE]) give no indication that the Roman's had knowledge of hydraulic limes and, instead, relied on the use of pozzolana to make their binders hydraulic. At some point in the second-century BCE, Roman builders began to use a pink volcanic ash containing silica and alumina, quarried from several different places around the Bay of Naples; mixing it with lime as they believed it to be sand. They discovered that this mixture resulted in a much stronger concrete than anything they had been able to previously produce. The best source of this volcanic material was found to come from Pozzuoli and, as a result of this, the material became known as pozzolana — a material which would significantly alter the future of concrete construction. Vitruvius described it as "*a kind of powder which from natural causes produces astonishing results*," and wrote that "*This substance, when mixed with lime and rubble, not only lends strength to buildings of other kinds, but even when piers of it are constructed in the sea, they set hard under water*". In translations of his work (1898 [c.77 CE]), it is described by Pliny as a substance is "*forming a barrier against the waves of the sea, becoming changed into stone the moment of its immersion, and increasing in hardness from day to day*".

It is possible that this pozzolana was first used at Puteoli and Cosa, north of Rome, to make hydraulic mortar for marine concrete (Idorn 1997; Lechtman and Hobbs 1986) — a technique which later spread north and was used in many Roman harbour sites on the west coast of Italy (Jackson et al. 2013). In some cases, the use of concrete in these projects was massive, with blocks formed from 30 to 125 m^2 of concrete, poured into wooden formwork which was assembled on shore and then floated out to sea where it was filled until it eventually sank in place (Lechtman and Hobbs 1986).

Pozzolana was later used in large-scale projects such as the theatre in Pompeii (Idorn 1997), constructed in 75 BCE, where concrete was, again, used as an infill material in walls with a stone or brick facade in the *opus incertum* technique. When Rome was reconstructed in the first-century CE, pozzolana concrete was widely used (Newby 2001), and many of those structures are still in existence today. In areas where pozzolana was not available, Roman builders would instead crush tiles, bricks or pottery into a powder and add this to lime to produce a similar effect (Blezard 1998, Vicat 1837; Pliny 1898 [c.77 CE]). In these circumstances, the presence of fine powder from the crushed material would discolour the lime, tinging it red or yellow depending on the colour of the material used (Vicat 1837).

The creation of this material is described by Pliny (1898 [c.77 CE]):

> Even broken pottery has been utilized; it being found that, beaten to powder, and tempered with lime, it becomes more solid and durable than other substances of a similar nature; forming the cement known as the 'Signine' composition, so extensively employed for even making the pavements of houses.

Some confusion has since arisen regarding the naming of this material, and the terms '*opus signinum*' and '*cocciopesto*' are often used interchangeably to describe mortar which contains lime and crushed tile or pottery. However, this is at odds with the descriptions of '*signinum* work' by Vitruvius (1914 [c.25 BCE]), which is specified for cisterns:

> Signinum work is made as follows. In the first place, procure the cleanest and sharpest sand, break up lava into bits of not more than a pound in weight, and mix the sand in a mortar trough with the strongest lime in the proportion of five parts of sand to two of lime.

Further confusion occurs when comparing these definitions to those used within the field of mosaic conservation, where '*opus signinum*' has been defined as a "*pavement made of lime mortar mixed with ceramic fragments into which quadrangular tesserae or small stone fragments are inserted, either randomly or to form geometric designs*," and 'cocciopesto' as a "*pavement made of lime mortar mixed with ceramic or stone fragments without the insertion of other elements*" (Alberti et al. 2013). While the manufacture of pressing-room floors from broken pottery laid over a bed of lime was described by Cato (1935 [c.160 BCE]) prior to Vitruvius or Pliny, he gives no indication that this was done to produce any kind of chemical reaction or hydraulic improvement to the mortar and, instead, it seems this was done to produce a durable ceramic top layer which could be rubbed down to form a smooth layer.

While the development of pozzolanic concrete was a great achievement, the Romans also experimented with other concrete construction techniques which, while less prevalent in their construction than pozzolanic concrete, certainly show no less ingenuity and forward thinking. For instance, petrographic examination of Roman concrete has provided evidence of what has been concluded to be intentional air-entrainment similar to that found in modern concrete (Idorn 1959), which is likely a result of the addition of organic materials that were known to be used by the Romans. Pliny the

Elder claims that the Temple of Minerva, for example, was plastered with a mortar that was blended with milk and saffron. Translations of his work (1898 [c.77 CE]) describe a material called 'maltha' which is *"a cement prepared from fresh lime; lumps of which are quenched with wine, and then pounded with hogs' lard and figs, both of them, mollifying substances. It is the most tenacious of all cements and surpasses stone in hardness. Before applying the maltha, the substance on which it is used must be well rubbed with oil."*

Roman builders also attempted to reinforce some of their structures with metal reinforcement. Evidence of its use has been found at several locations, in particular in the remains of concrete vaulting of important thermae built under imperial patronage such as the Baths of Caracalla (Figure 5), the Baths of Trajan, the Baths of Diocletian, the Large Baths, and the Heliocaminus Baths at Hadrian's Villa (Yegül 1992). The Baths of Caracalla are described in the biography of Roman emperor Caracalla in '*Historia Augusta*' (Magie 1993 [c.4th century CE]):

> For it is said that the whole vaulting rested on gratings of bronze or copper, placed underneath it, but such is its size, that those who are versed in mechanics declare that it could not have been built in this way.

During the excavations of the baths, many tons of fragments of iron girders were found. These compound girders were riveted together perpendicularly, then cased in bronze to form a lattice-work ceiling which may have formed panels that were filled with lightweight concrete, coated with fine stucco that was painted and gilt (Middleton 1892).

The lack of tensile reinforcement in Roman concrete structures meant that they had to be designed in such a way that load was carried in compression, resulting in walls of massive thickness — sometimes in excess of 8 m. Consequently, lightweight concrete was developed to reduce the need for such massive buttresses and walls (British Cement Association 1999), with early attempts made by casting hollow clay jars into walls and arches, and later by introducing crushed porous volcanic rock (pumice) as a lightweight aggregate (Chandra and Berntsson 2002; Stanley 1979). Lightweight concrete was subsequently used in two of the most prestigious and notable Roman structures: the Colosseum, completed in 82 CE, and the Pantheon, completed in 127 CE — both of which have endured to the present as a testament to Roman engineering.

The Colosseum (Figure 5), an oval 190 by 130 m, was the largest of Rome's amphitheatres with seating capacity for 50,000 spectators. It has foundations made of dense concrete, but arches and vaults constructed of lightweight concrete, which have survived despite lightning strikes, earthquakes and vandalism (British Cement Association 1999).

The Pantheon (Figure 5) was one of the few buildings in Rome to have survived intact after the fall of the Roman Empire. Its unique domed roof is 43.4 m in diameter with a 9 m diameter oculus to allow light to

Figure 5. Examples of Roman concrete. (a) Opus reticulatum at Villa Adriana, Tivoli, built 117–138 CE [Camelia Boban 2011, CC BY-SA 3.0]. (b) Dome of the Pantheon [Bruno Hautzenberger 2012, CC BY-SA 3.0]. (c) the Colosseum [David Iliff 2007, CC BY-SA 3.0]. (d) the Baths of Caracalla [Veronika Janssen 2009, CC BY-SA 3.0].

enter (Adam 2005; Newby 2001) and is constructed from lightweight concrete in which crushed pumice was used as an aggregate (Stanley 1979). At the time, the dome was three times larger than any other built (Newby 2001) and remained the largest in the world until the 20th Century (British Cement Association 1999).

As the Roman Empire expanded, Roman engineers carried their knowledge of cement and concrete with them. Due to the difficulty of transporting pozzolana from Rome, most of the Roman concrete used in Britain was a lime concrete, making use of the local materials which were available (Stanley 1979); although ground tiles were sometimes added as an artificial pozzolan to produce a higher quality material (Blezard 1998). In Britain, the Roman's used concrete in walls, foundations and floors — some of which were overlaid with elaborate, decorative mosaics. Roman constructions in Britain which incorporated concrete include Pharos lighthouse, Dover (c.50–138 CE) (Figure 6) — a four storey, 13 m tall lighthouse with sandstonefaced walls which had a lime and rubble core — and Portchester castle, Hampshire (c.250–300 CE) (Figure 6) — a large fort with concrete foundations and walls over 2 m thick with a concrete core (British Cement Association 1999).

However, perhaps the most significant Roman construction in Britain is Hadrian's Wall (122–130 CE) (Figure 6), a stone and concrete wall which reached up to 4 m tall and stretched 120 km from the Solway Firth to Tyne and included 16 forts — each housing 500 to 800 men — 80 smaller forts, or 'milecastles', and 158 towers (Mallinson and Davies 1987; Stanley 1979). In places, the wall was built with the '*opus incertum*' technique, with stone facing and a core of lime concrete or clay. Cores of the infill concrete were taken by the Building Research Establishment (BRE) and were found to contain large, irregular stone aggregate (local sandstone and igneous rock from the Whin Sill) up to 30 cm across. Several fragments of fired clay products were identified in this particular sample of mortar. However, the quantity was not thought to be sufficient to produce a significant pozzolanic effect. Despite this, the sample contained abundant C-S-H but no significant carbonation, suggesting the formation of C-S-H was the main setting mechanism. However, this was most likely a product of a reaction between the lime and chert present in the coarse aggregate (Mallinson and Davies 1987).

3.2.4. Medieval - Renaissance era Europe

It appears that most of the Roman knowledge and skill regarding concrete construction and pozzolanic materials disappeared almost completely following the fall of the Roman Empire (Stanley 1979). Despite being recorded by authors such as Vitruvius, the fact that it was written in Latin and most people had limited access

Figure 6. Early concrete in Britain. (a) the Roman 'Pharos' Lighthouse [Alexander Kachkaev 2012, CC BY-SA 3.0]. (b) Hadrian's Wall [Carole Raddato 2017, CC BY-SA 2.0]. (c) the exterior walls of Portchester Castle [Jamie Heath 2021 CC BY-SA 2]. (d) Remains of the concrete core of Reading Abbey [Hugh Llewelyn 2018, CC BY-SA 2.0].

to these records meant that this knowledge was largely confined to the Catholic Church (Idorn 1997).

This was certainly the case in Britain, where concrete work during the Saxon era was limited (British Cement Association 1999) and relatively primitive when compared to that which had occurred during Roman times. Examples of lime concrete floors have been identified at several Saxon sites dating from the 6th – 9th century CE, including St. Peter's Street, Northampton (Williams 1979); Bamburgh Castle (Kirton and Young 2012); Lyminge, Kent (Thomas 2018); and Wearmouth and Jarrow, Northumberland (Cramp 2005). The remains of large mortar mixers have also been found at some of these sites (Cramp 2005; Kirton and Young 2012; Williams 1979), all with similar designs consisting of a roughly circular cavity, typically 2–3 m in diameter, with a central pivot on which a bar and paddles were mounted, allowing large quantities of mortar to be mixed at once.

More significant concrete works began to appear in Britain following the Norman conquest, and it became common to use lime concrete infill in walls, piers and buttresses from the 12th century onwards (Cowan 1977). Founded in 1121 by Henry I, the third Norman king of England and fourth son of William the Conqueror, Reading Abbey (Figure 6) is perhaps the most recognised structure in Britain that utilised concrete during this period. It was built in a similar manner to that of the Roman Empire, with permanent masonry walls that were filled with a core of lime mortar and large pieces of aggregate — in this case flint and limestone. The facing stones have since been removed and much of the abbey destroyed, with only the solid concrete cores from some of the walls remaining. Analyses of samples from Reading Abbey confirmed them to be carbonated lime concrete with no evidence of C-S-H in the binder (Mallinson and Davies 1987).

During the medieval period, a wide variety of organic admixtures were used in lime mortars in Britain. A detailed literature review conducted by Sickels (1981a) concluded that these were used largely by communities with limited access to high-quality limes and so masons experimented with locally available organic materials to improve weak lime or clay mortars. Some examples of organic additives that were used are beer, beeswax, blood, eggs, egg white, fruit juice, malt, rice, sugar, urine, and wort. Specific examples where organic admixtures were allegedly used include; Queen Eleanor's cross at Charing Cross, London, which incorporated the egg whites and wort of malt with lime and Calais sand; Rochester Cathedral, the mortar and stucco of which contained bullocks' blood; Rockingham Castle where melted wax was used; and in King Edward II's work at Westminster, where pitch is said to have been mixed in the stucco and mortar (Bankhart 1908; Sickels 1981a, 1981b).

While air limes were the predominant binders in Britain during this period, there is some evidence of the use of pozzolanic materials. Originally commissioned by Henry III in the late 14th Century, the Cosmati Pavement in the sanctuary of Westminster Abbey was built with materials and possibly artisans imported from Rome, using mortars of lime and pozzolanic crushed terracotta that were intended to cope with the damp conditions caused by its location on the Thames riverbank. Analyses of samples from the floor has revealed that the mortars were hot mixed, with shards of terracotta and locally sourced aggregates added to the lime at the time of slaking (Siddall 2013).

There is more substantial evidence that the use of the pozzolanic materials continued in other parts of Europe following the fall of the Roman Empire in the 5th century CE, and they appear to have been in common use throughout the Byzantine and Ottoman Empires.

Analyses and characterization of hydraulic mortars from ancient cisterns and baths in Greece (Stefanidou et al. 2014) has shown the use of pozzolanic materials in both Byzantine and Ottoman structures. Samples analysed from the Byzantine bath in Thessaloniki (11th – 12th century CE), the Byzantine Castle of Servia (10th century CE) and the Ottoman Pazar Hamam in Thessaloniki (15th century CE), all contained lime and pozzolana, and samples obtained from the Byzantine Castle of Servia, which were from an internal cistern, also contained brick fragments as aggregates.

Analyses of brick-lime plasters and mortar from three Ottoman bath buildings located in Turkey and constructed in the 14th and 15th century have shown the use of pozzolanic bricks as aggregates. While the bricks used as aggregate in the mortar and plaster were found to have good pozzolanicity, those used to construct the domes of the bath buildings did not and this suggests the intentional selection of bricks that would create a hydraulic binder (Böke et al. 2006).

While surviving literature on the topic from this period is sparse, during the Renaissance period, Italian architect Leon Battista Alberti wrote about the use of building materials in his books, the compilation of which can be found in the English translation, '*The Architecture of Leon Batista Alberti in Ten Books*' (Alberti 1755 [1485]). In these writings, Alberti dedicates a whole chapter to lime and plaster of Paris, in which he describes the nature of lime, its uses and types. He also quotes the work of both Vitruvius and Pliny, and it is clear that there is an understanding of the use of both pozzolana and cocciopesto at this time:

> There are other ancient pavements made all of one piece, which I suppose, was a mixture of lime, sand, and pounded brick, of each a third part: which may be made more strong and lasting yet, by the addition of one fourth part of 'Tyber' – stone, beat to powder. Others in this sort of plaster mightily commend the sand of Pozzuolo, which they call 'Rapillo'. Plaster that is designed for pavements must be tried by continual beating, whereby it will daily acquire greater stiffness and hardness, till it comes to be in a manner firmer than stone itself and it is certain, that if this plaster is sprinkled with lime-water, and linseed-oil, it will grow almost as hard as glass, and defy all manner of weather.

Almost a century later, Andrea Palladio would also discuss the writings of Vitruvius (Palladio 1738 [1570]), which suggests that, by this point, knowledge of the material was widespread within Italy:

> The also dig out of the earth in Terra di Lavoro, in the territories of Baia and Cuma, a sort of sand, called 'pozzolana' by Vitruvius, which immediately cement in the water, and makes buildings very strong.

The ancient Roman use of concrete was also influential on Italian architects at this time, and structures such as the Pantheon and Colosseum are said to have inspired its use by Donato Bramante to construct the four piers that would support the dome of St. Peter's Basilica at the Vatican, as well as friezes and cornices on the exterior of the building. However, it has been alleged that Michelangelo was unhappy with the work as it did not meet the accepted proportions of binder to sand — Bramante's workers had used proportions of 1/10 or 1/12 instead of 1/3 or 1/4 – and remedial work eventually had to be undertaken to rectify the poor workmanship. Despite this, the four masonry-adorned concrete piers were complete up to the cornice, including the arched connecting structure by the time Bramante died in 1514 and Michelangelo was appointed to continue the work (Steiger 1996).

3.3. Asia

Despite being home to some of the oldest civilisations in the world, there appears to be relatively little literature available in English that details the historic use of mortar-based materials in Asia. It is unclear to what extent this is due to a lack of available English translations of existing literature, and what is due to a lack of their use in regions outside those discussed. However, it is clear that, in the countries for which research is available, the use of lime was very often in conjunction of with the use of organic additives to alter the properties of the binder.

3.3.1. China

A form of ancient concrete has also been discovered in the Gansu Province of northwest China, at the Dadiwan site in Qin'an County, which dates to the Yangshao Period (5000–3000 BCE). As with other examples of ancient concrete of this era, it was used as a floor material in residential structures. Analyses of two different floors (Li, Zhao, and Li 2012), designated F-405 and F-901, has shown that they were made of a lightweight concrete formed by the mixture of calcined ginger nut, red clay and hollow calcined calcareous nodules known as 'kunkur'. Carbon-14 dating of F-405 and F-901 place the age of the floors as 6769 ± 312 and 6137 ± 159 years old, respectively. The calcined ginger nut was the main bonding agent in the floors, while the calcined kunkur acted as a lightweight aggregate. It is likely that these materials were calcined in one of the 38 pottery kilns that were excavated from the Dadiwan site, and it is estimated that the temperature for firing pottery at that time was about 840-1040°C. The kunkur was fairly pure $CaCO_3$, while the ginger nut was 70–80% $CaCO_3$ and 20–30% clay and other minerals. This means the mixture set by two mechanisms. Firstly, the β-belite that will have been formed by calcining the ginger nut would have hydrated to form C-S-H and calcium aluminate silicate hydrate (C-A-S-H.). Secondly, additional strength would have developed from the hydration and subsequent carbonation of the CaO from both materials to form $CaCO_3$ (Li, Zhao, and Li 2012).

In literature, the application of lime in China can be traced back to the Xia Dynasty (2070–1600 BCE) (Zhang et al. 2014), and archaeological records show its regular use by Chinese builders as early as the Qin Dynasty (221–206 BCE) (Zhao et al. 2015). However, while the use of pozzolana spread in Europe during this time, there is a lack of evidence to suggest that hydraulic mortar technology was developed in ancient China, and this is partly due to the absence of natural hydraulic materials like volcanic ash (Xiao et al. 2014; Zhang et al. 2014). Instead, there arose the use of organic-inorganic hybrid lime mortars with admixtures of natural organic compounds such as rice starch, egg white, brown sugar, tung oil, plant extracts, and animal blood (Xiao et al. 2014; Dai et al. 2019, S. Q.; Fang et al. 2014, S.; Fang et al. 2014, 2015; Zhao et al. 2015). These admixtures, when added in the correct quantities, can have several beneficial effects on lime mortars, including accelerated setting and hardening (rice starch; tung oil; pig blood), increased compressive strength (rice starch; pig blood), improved water resistance (tung oil; pig blood) and air-entrainment (pig blood) (Zhao et al. 2015).

From at least as early as the Western Zhou Dynasty (1046–771 BCE), Chinese builders began to use a composite material of lime, sand and clay, known as 'sanhetu' or 'tabia' to fulfil the construction requirements that lime alone could not meet (Zhang et al. 2014). This material was widely used in structures in ancient China from the Han Dynasty (206 BCE – 220 CE) onwards, before becoming increasingly replaced by Portland cement concrete in the last two centuries (Dai et al. 2019). While sanhetu was unable to gain high levels of strength or durability, the addition of natural organic materials, such as rice starch, egg white, brown sugar, tung oil, plant extracts and animal blood, resulted in a material with excellent performance, and this was used in many important structures, including city walls, bridges, dams and tombs — many of which survive to this day (Dai et al. 2019; Zhang et al. 2014). Analysis undertaken on samples from an ancient tomb located in Anhui province reveal a mineralogical composition of quartz, kaolinite, calcite and calcium silicate hydrate, and compressive strength tests on samples cut to 100 × 100× 100 mm gave an average strength of 21 MPa, which is much higher than that of the organic-inorganic hybrid lime mortars previously mentioned, probably due to the addition of clay to the system (Dai et al. 2019).

3.3.2. India

Lime plaster has been used in India for centuries, often with additives to improve its properties. While additions such as jute fibres and rice husks are typical in many Indian mortars and plasters (Singh, Waghmare, and Vinodh Kumar 2014), research into the decorative lime wall plasters at the Ellora Caves (6th – 12th century CE) World Heritage Site, Maharashtra (Figure 7), revealed the use of a feebly dolomitic lime binder with fine to medium grained siliceous aggregate and an organic filler of hemp (*Cannabis sativa*). This is possibly the earliest authenticated report of the use of hemp fibres as a filler in lime plaster and suggests that hempcrete may have been manufactured as early as the 6th century CE (Singh, Vinodh Kumar, and Waghmare 2015).

There is also evidence of the use of hydraulic binders in India, often in conjunction with natural additives, and this has been found at several important monuments. One such example is the Vadakumnathan temple, Kerala, which is a Hindu temple dedicated to Shiva, and was built approximately 1300 years ago. Analyses of mortar samples (Thirumalini et al. 2015) from the temple suggests that finely ground shell lime, rich in clay minerals, may have been burnt with limestone to make hydraulic mortar. The formation of the hydration phases suggests the use of hot lime technology and organic materials in the form of carbohydrates, proteins and fats were identified in the mortar. These organic

Figure 7. Sites in India which incorporate lime binders with natural additives. (a) Ellora Caves [Shivaji Desai 2020, CC BY-SA 4.0]. (b) Charminar [Didier Tais 2008, CC BY-SA 3.0]. (c) the Taj Mahal [Yann 2010, CC BY-SA 4.0]..

additives were likely added intentionally to improve its properties — with carbohydrates enhancing carbonation upon fermentation by providing a continuous supply of CO_2 to the inner part of the mortar (Ravi, Thirumalini, and Taher 2018; Thirumalini et al. 2015), protein causing air entrainment and increasing workability, and fat acting as a water-proofer and to control water movement (Thirumalini et al. 2015).

Other examples include the Daulatabad Fort in Maharashtra, where research into the lime plaster of the 13th – 16th century structure points to the deliberate use of hematite iron ore as pozzolanic filler to make the finishing plaster harder, more compact, and less permeable, as well as the inclusion of hemp fibres (Singh and Vinodh Kumar 2018). Another is Charminar, Hyderabad (Figure 7), which was constructed in 1591 CE by the emperor Qutb Shah to mark the beginning of the second Islamic millennium year. Analyses of mortar (Ravi, Thirumalini, and Taher 2018) from Charminar indicate the use of a hydraulic mortar with additions of jaggery (refined sugar), *Terminalia chebula*, and egg white. Analyses of plaster from the Taj Mahal (Aslam 1990) (Figure 7), built between 1631 and 1648, indicate the use of a partially hydraulic binder which also contained fibres of sisal (*Agave sisalana*), jute (*Corchorus*) and hemp (*Cannabis sativa*). Characterization of lime plaster from the 17th century Mughal monument of Bibi Ka Maqbara revealed the addition of zeolites, which appear to have been added intentionally to impart hydraulicity to the lime mortar and enhance its strength, setting and durability (Singh, Waghmare, and Vinodh Kumar 2014).

3.4. Mesoamerica

Lime has been used by different cultures in Mesoamerica for thousands of years, with evidence of the use of lime plaster floors dating back to at least 1500–1150 BCE (Flannery and Marcus 2005). However, there is also evidence of lime-pozzolan mixtures and lightweight concrete (Cabrera, Rivera-Villarreal, and Sri Ravindrarajah 1997; River-Villarreal and Krayer 1996; Rivera-Villarreal and Cabrera 1998) the development of which took place independently from those that occurred in Europe. It has been speculated that the pozzolanic reaction's discovery was a result of the preparation of dough for tortillas (River-Villarreal and Krayer 1996; Rivera-Villarreal and Cabrera 1998). Mesoamericans had to boil corn kernels in a mixture of water and quicklime to remove the cuticles. The wastewater from this process was then disposed of directly onto local soil, large tracts of which contain pozzolanic volcanic ash, causing the soil to harden. It is also possible that natural materials, such as honey and extracts from the bark of trees, were added to mortars and plaster in ancient times, as this is a traditional practice that still exists in parts of Mesoamerica today (Littmann 1957).

3.4.1. Zapotec

Although less intensively studied than the Aztec or Maya, the Zapotec of Oaxaca produced one of the first civilisations of ancient Mexico through thousands of years of social evolution. They were among the first Native Americans to use adobe, stone masonry, and lime plaster in their constructions, and to orientate their public buildings astronomically (Marcus and Flannery 1996). Evidence of lime-plastered wattle-and daub construction and lime plaster floors for public buildings, thought to be ceremonial 'men's houses', date back to the Tierras Largas phase (1500–1150 BCE) and the first known use of adobe in Oaxaca to the end of the San José phase (1150–850 BCE) when it was used in the construction of large pyramidal temple platforms built of planoconvex adobe bricks and earthen fill, with sloping walls of dry-laid stone masonry (Flannery and Marcus 2005). By the period of 100 BCE – 200 CE, many Zapotec nobles lived in palaces made of adobe brick and lime plaster over stone masonry foundations (Marcus and Flannery 1996).

3.4.2. Maya

The use of lime has been documented at several Maya sites across Mesoamerica. Some of their earliest uses of lime are evident at the site of Cuello, 5 km west of Orange Walk Town in northern Belize (c.1200 BCE – 1250 CE). The first part of the Early Period of architectural development at Cuello took place between 1100 and 600 BCE. During this time, platforms were constructed of a core of small stone and earth within a low retaining wall of limestone cobbles, which were coated with compacted earth and/or a plaster of lime or lime mixed with clay, sand, or other inert filler, and applied over a rough course of stone. During the second half of the Early Period (600–400 BCE), straight-walled masonry superstructures were built — laid in rows of limestone cobbles, packed with earth, and faced with lime plaster. The Late Period (400 BCE – 250 CE) saw the introduction of terraced pyramids, faced with shaped blocks of limestone and finished with lime plaster (Hammond and Gerhardt 1990).

A variety of flooring materials were used at the ancient Maya center of Holmul, northern Guatemala, during the Late Pre-classic period (400 BCE – 250 CE), including lime plaster, clay plaster and compacted sascab (Ahern 2021) — a soft limestone conglomerate or unconsolidated limestone found in the limestone of Yucatan, that typically appears as a white to reddish, compacted powder, frequently containing rounded pebbles or stones, and is often used within the region as a substitute for sand in the preparation of mortars and plasters (Littmann 1958a).

Plaster was commonly used by the Maya to protect and adorn masonry structures during the Late Classic period (600–900 CE), and at the Classic Maya Kingdom of Piedras Negras, Guatemala, high quality masonry structures, plastered with lime, were built at the site's center, the 'Acropolis', between 650–800 CE. It is likely that the limestone was burned on wooden pyres, or 'caleras', rather than in kilns or the pits that were more typically used in the Early Classic period, and any single masonry structure was built with lime from a single source of limestone and aggregate. The aggregate was naturally decomposed local sascab, and it has been estimated that some of the structures may have required 30–40 m^3 of lime plaster (Abrams et al. 2012).

Analyses of samples from the Maya site of Comalcalco (c.550–1000 CE), Tabasco, Mexico (Figure 8), revealed several different architectural uses of lime, including mixtures of lime and aggregates that formed monolithic masses, mortars between brickwork, protective plaster coatings on both walls and floors, cast or modelled decorative stuccos, and thin (<1 mm) lime wash coats which were applied to both horizontal and vertical surfaces. In many cases, these were applied in a multi-layer system (Littmann 1957, 1958b). Due to the absence of local limestone, the abundance of shell in the samples, and the high ratio of calcium to magnesium, it was concluded that shell from nearby waters was used as the source of lime (Littmann 1957). The same elements and systems found at Comalcalco were also found in different structures at Palenque (c.250–800 CE), Chiapas, Mexico (Littmann 1959). However, unlike at Comalcalco, the source of lime was locally available dolomitic limestone, blocks of which were also used for the walls, roofs, and floors at Palenque (Riquelme et al. 2012).

There is also evidence to suggest the use of lime-pozzolana mixtures in architectural Maya plasters. Examination of plasters from the Calakmul, Campeche, Mexico, and Lamanai, Orange Walk District, Belize (Figure 8), shows the inclusion of volcanic ash and glass that were likely added with the intention of making hydraulic mortar (Villasenor and Graham 2010).

3.4.3. Teotihuacan

Teotihuacan (Figure 8) was a large and influential city and state located in the northeastern Basin of Mexico. While its chronology is a point of contention, with little known about its early phase, it experienced a period of explosive urban growth during the subsequent Patlachique phase (100–1 BCE) (Nichols 2016). The city greatly expanded over the next six centuries, with the construction of large pyramids (the Sun and Moon pyramids) and more than 2000 domestic, administrative and ritual compounds, over an area of 20 km^2 (Barca et al. 2019; Miriello et al. 2021). Lime plaster was used extensively throughout Teotihuacan to cover most building surfaces, including floors, walls and roofs (Miriello et al. 2021; Pecci et al. 2016), and it has been estimated that over 12 million square meters of architectural surfaces across the city were covered with lime plaster (Barba et al. 2009).

The valley in which Teotihuacan lies is surrounded by volcanoes (Miriello et al. 2021), and the use volcanic cinders rich in rhyolitic glass shards as aggregate in plasters was a widespread practice in Teotihuacan — with material often transported up to 180 km away along trade routes to the Gulf Coast (Barca et al. 2019; Barca, Crisci, and Miriello 2019). The plasters are typically composed of two layers: a lower layer, called 'firme', of crushed volcanic scoria (tezontle) mixed with a mud-based binder, and a superficial surface layer, called 'enlucido', of lime mixed with volcanic glass shards. While these plasters have been found to be hydraulic, analyses have demonstrated that it is not the presence of glass shards in the external surface layer which produce hydraulicity and this is, instead, due to the reactivity of the tezontle present in the firme layer (Miriello et al. 2021). The floors of Teotihuacan typically had a third layer — a base of layer of compacted volcanic tuff (15–20 cm) — onto which the firme layer (6–10 cm) was applied, before finishing with the thin lime-based surface layer which would be polished (Barba et al. 2009).

3.4.4. El Tajín

One of the most archaeologically significant sites in Mesoamerican is the Prehispanic city of El Tajín (Figure 8), located in northern Veracruz, Mexico. While it was previously thought that El Tajín was occupied during three phases between 100 BCE and 1200 CE, it is now believed there was only one phase of occupation that lasted from 800 to 1200 CE, after which time it was abandoned and partly destroyed (UNESCO 2022b).

Both normal and lightweight concrete were used in the construction of El Tajín. While normal weight concrete, made of rounded limestone coarse aggregate and a lime-pozzolan binder, was used for flooring, it could not be used for ceiling slabs. Instead, builders used lightweight aggregates, rounded pumice up to 100 mm in size, and produced concrete that ranged from 1050 to 1100 kg/m^3, which allowed the construction of upper floors and flat roofs (River-Villarreal and Krayer 1996). These lightweight aggregates and the natural pozzolanic

Figure 8. Mesoamerican archaeological sites. (a) Comalcalco [Alfonso Bouchot 2012, CC BY-SA 3.0]. (b) Lamanai High Temple [Bernt Rostad 2010, CC BY-SA 2.0]. (c) Teotihuacan [Jack Hynes 2006, public domain]. (d) El Tajín [Arian Cigarroa am 2012, CC BY-SA 4.0]..

material used were locally available due to frequent volcanic activity (Rivera-Villarreal and Cabrera 1998).

The builders also developed formwork and placing techniques that allowed them to construct large roofs that were 250–350 m^3. Analyses of samples taken from the upper and middle layer of a slab, showed porosities of 25–39.3% and 45.5%, respectively, and that the upper surface was made of thin layers or fine-grained mortars which were polished to reduce permeability (River-Villarreal and Krayer 1996). In addition to providing structural support, the flat lightweight concrete roofs, which varied in thickness from 0.6 to 1.0 m (Cabrera, Rivera-Villarreal, and Sri Ravindrarajah 1997), also provided good insulation (Rivera-Villarreal and Cabrera 1998).

4. Conclusion

Mortar-based materials have been a key construction material in many cultures throughout the world for thousands of years. They were developed independently in many cultures as they began to build permanent architecture and were used to create hard and durable floors, and as bedding mortars, internal wall plasters and external wall renders and stucco.

While most famously associated with Roman construction, the development of hydraulic binders from lime and pozzolanic materials occurred independently in different regions around the world where natural pozzolanic materials were available. Evidence suggests the intentional manufacture of hydraulic binders may have taken place even earlier and more widely by adding crushed clay products, such as bricks, tiles or terracotta. These lime-pozzolana binders were utilised in the creation of both watertight renders and underwater concrete structures. In many cultures, modifications to the properties of lime binders were made through the addition of various locally available organic additives.

Concrete and lightweight-aggregate concrete were used to create massive structures in ancient cities such as Rome and El Tajin, and Roman engineers even attempted to reinforce some of their concrete with metal. While some of their structures still stand today, the collapse of these civilizations resulted in a loss of knowledge regarding construction techniques, and structural concrete was not used again for several centuries. Even then, the level of sophistication did not match that which had been previously seen, and it would not be until during the industrial revolution, when hydraulic cements were invented, that concrete and, later, reinforced concrete, would undergo significant development.

While this paper gives a brief overview of the historic use of mortars-based materials, ongoing research is required. Firstly, with regards to the conservation of historic sites, it is essential to fully understand the sites and the materials used in order to determine how they will weather in the future, how susceptible they are to the effects of climate change, and which materials are appropriate and compatible for use in their repair and preservation. Secondly, as there is growing concern about the sustainability of Portland cement-based materials, revisiting and understanding the use of historical, locally sourced materials and traditional construction techniques may provide long-term alternative solutions.

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