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Microstructural characteristics of mortars prepared by hot lime mix

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Original scientific paper

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Microstructural characteristics of mortars prepared by hot lime mix

The effect of lime characteristics and hot lime mix method on hydraulic, microstructural and mechanical properties of mortars is determined by producing mortars from quicklimes of two different marbles and two limestones. Results of SEM-EDS, XRD and TGA analyses reveal that the porous microstructure of mortars and spongy texture of calcite crystals are the indicators of the hot lime mix method. This study shows that characteristics of limestones used for the production of limes, as well as the preparation method, directly affect hydraulic, mechanical and microstructural properties of mortars.

Key words:
marble, limestone, lime, mortar, hot lime, microstructure, SEM-EDS

Izvorni znanstveni rad

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Mikrostrukturna svojstva mortova koji se pripremaju od mješavine živog vapna

U radu su, na temelju mikrostrukturnih svojstava morta koji se priprema od vapna dvaju različitih mramora, te dva vapnenca utvrđen učinak vapna na hidraulična, mikrostrukturna i mehanička svojstva morta. Rezultati analize materijala pretražnim elektronskim mikroskopom i elementne analize (SEM-EDS), analize rendgenske difrakcije (XRD) i termogravimetrijske analize (TGA) pokazali su poroznu mikrostrukturu morta i spužvastu teksturu kristala kalcita kao indikatore metode mješavine živog vapna. Istraživanje je pokazalo da svojstva vapnenca koji se koristi u proizvodnji vapna, kao i metoda pripreme, izravno utječu na hidraulična, mehanička i mikrostrukturna svojstva morta.

Ključne riječi:
mramor, vapnenac, vapno, mort, živo vapno, mikrostruktura, SEM-EDS

Wissenschaftlicher Originalbeitrag

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Mikrostrukturelle Eigenschaften von Mörteln, die aus einer Branntkalkmischung hergestellt wurden

Basierend auf den mikrostrukturellen Eigenschaften des Mörtels, der aus dem Kalk zweier verschiedener Marmorarten und zwei Kalksteinen hergestellt wurde, wurde die Wirkung von Kalk auf die hydraulischen, mikrostrukturellen und mechanischen Eigenschaften des Mörtels bestimmt. Die Ergebnisse der Materialelektronenmikroskopie und Elementaranalyse (SEM-EDS), Röntgenbeugung (XRD) und thermogravimetrischen Analyse (TGA) zeigten eine poröse Mikrostruktur des Mörtels und eine schwammige Calcit-Kristallstruktur als Indikatoren für die Branntkalkmischungsmethode. Untersuchungen haben gezeigt, dass die Eigenschaften von Kalkstein, der bei der Kalkherstellung verwendet wird, sowie die Herstellungsmethode die hydraulischen, mechanischen und mikrostrukturellen Eigenschaften von Mörtel direkt beeinflussen.

Schlüsselwörter:
Marmor, Kalkstein, Kalk, Mörtel, Branntkalk, Mikrostruktur, SEM-EDS

1. Introduction

Lime has been the most frequently used traditional material in construction works for more than three thousand years [1]. It is produced by first heating calcareous stones to convert calcium carbonate into calcium oxide (quicklime) and then by slaking the quicklime with water [1, 2]. Different types of limestone sources, marble elements of structures, and marble sculptures, were used as raw materials in lime production in ancient times [3]. According to their chemical compositions and hardening process, limes can be classified into two fundamental types: non-hydraulic limes and hydraulic limes [4-6]. Non-hydraulic limes are mainly composed of calcium oxide and magnesium oxide added in different ratios. They harden through a carbonation reaction by absorption of CO_2 from the air. During the reaction, CO_2 reacts with lime and produces calcium carbonate. The reaction begins on the surface of the lime sample and progresses through to the inside. The rate of carbonation reaction mainly depends on the CO_2 concentration, relative humidity and temperature of air, and moisture content of lime [7, 8]. High concentration of CO_2 and ambient relative humidity accelerate the carbonation rate of lime.

Natural hydraulic limes contain CaO, MgO and active compounds of calcium silicates and aluminates, which are formed during calcination of limestones containing impurities such as alumina, iron, and silica [1, 5, 9]. These compounds ensure that lime sets and hardens in the presence of water, which represents hydraulic character. Therefore, hydraulic limes have been used in mortars in water constructions such as bridges, drainage systems, cisterns, foundations, etc. [10].

Lime mortars were traditionally produced by using either slaked lime or quicklime as binding material and several types of aggregates of different properties. The production method involving the use of quicklime is generally known as "hot lime mix" [11, 12]. In hot lime mix, quicklime is mixed with aggregates and, afterwards, water is gradually added to the mixture, until an appropriate workability is obtained [13]. During this process, better bonding between lime and aggregates is obtained as a result of the heat given off and lime expansion. Improved mechanical properties are obtained by improved bonding of lime and aggregates, and lower amount of water required in hot lime mix [11, 13].

Studies concerning traditional lime mortar production generally focus on the effects the lime-aggregate ratio, type of aggregate, and curing time, have on carbonation, mechanical strength, physical properties, microstructural properties, mineralogical composition, and chemical composition of lime mortars produced in laboratory conditions [13-19]. However, the effects of production methods and long-term effects of lime types on mortar characteristics are generally disregarded although they are of critical importance for their application in the conservation of historic buildings.

The aim of this study is to identify the effects of lime characteristics on the hydraulic, microstructural and mechanical properties of mortars prepared by quicklime and aggregates (hot lime mix). It also aims to determine microstructural changes that occur due to hot lime mixing. Chemical, mineralogical and microstructural characteristics of limes produced from marbles and limestones, and their effects on mortars, are investigated in this study. The results of this study could also guide the selection of limestones to be used for the production of intervention mortars in the restoration of historic buildings.

2. Experimental methods

Experimental studies can be grouped in two parts. The initial part involves selection of marble and limestone samples and manufacturing of lime from these samples. Microstructural properties, as well as chemical and mineralogical composition of uncalcined, calcined, slaked and carbonated stones, are determined in this part. The second part involves preparation of mortar samples by using limes manufactured in the first part as binder with limestone aggregates. Uniaxial compressive strengths and mineralogical composition of lime mortars tested at 3, 6, 12 and 140 months were identified.

2.1. Selection of marble and limestone samples and their calcination, slaking and carbonation

Two types of marbles from Marmara Island and Muğla province (Turkey) and two types of limestones containing low and high silica from Urla/Izmir (Turkey) were selected as raw materials for the production of lime. Calcination temperatures of selected marbles and limestones were determined by thermogravimetric analysis (TGA) carried out using Perkin Elmer-Diamond TG/DTA in static nitrogen atmosphere between temperatures 25-1000 °C at 10°C/min. Based on TGA results, crushed marble and limestone samples with particle sizes between 0.250-0.125 mm were calcined in a furnace at 850°C for 12 hours, and kept in the desiccator until they reached room temperature. Lime was produced by slaking calcined products (quicklime) with distilled water in a glass beaker. The water to quicklime ratio was nearly four to produce lime [20]. After slaking, lime putties were spread on the glass slides and kept at room conditions for one month for carbonation. Then, the carbonated samples were dried in a laboratory oven at 40°C for 24 hours.

Mineralogical composition of the uncalcined, calcined, slaked, and carbonated samples was identified by X-Ray Diffraction (XRD - Philips X-Pert Pro X-ray diffractometer) using CuKα between 5-60 2°Theta with 1.6°/min scan, operating at 40 kV and 40 mA.

Their microstructural properties and chemical composition were determined by the Scanning Electron Microscope (SEM) coupled with X-Ray Energy Dispersive System (EDS) (Philips XL 30S FEG) operated at different magnifications. Chemical compositions of carbonated samples were also used to determine their hydraulic properties by calculating the hydraulic (H.I.) and cementation (C.I.) indices according to Boynton formula , Eq. (1) and Eq. (2). [21]:

$$H.I. = (\%Al_2O_3 + \%Fe_2O_3 + \%SiO_2) / (\%CaO + \%MgO) \quad (1)$$

$$C.I. = (2.8\ \%SiO_2 + 1.1.\ \%Al_2O_3 + 0.7\ \%Fe_2O_3) / (\%CaO + 1.4\ \%MgO) \quad (2)$$

2.2. Preparation of lime mortars by hot lime mix method and determination of their microstructural and mechanical characteristics

Mortars are prepared by the "hot lime mix" method using calcined limestones (quicklime) and marble aggregates with a binder/aggregate ratio of 1/3 by weight.

The main reason for using quicklime was to obtain a homogenous mixture and strong bond between the binder and aggregates as a result of the "hot lime mix" [11, 13]. The reason for selecting marble aggregates was that they do not react with lime and have the same chemical composition as carbonated lime. Thus it was ensured that the effect of aggregate characteristics on the prepared mortars was negligible, while the effects of different types of limes on mortar properties were determined. Aggregate particle sizes ranged between 1000-2000, 500-1000, 250-500 μm, and equal quantities were used for each size. Mortars were prepared by mixing with a Kitchenaid® mini mixer. The water to quicklime ratio of 4/1 was selected, which was higher compared to the water needed for slaking quicklime, in order to obtain workable mortars [20]. Mortar mixtures were cast in cylindrical PVC moulds 4.5 cm in diameter and 5 cm in height. Samples were removed from moulds after 48 hours and kept for about 28 days at a nearly 100 % relative humidity in a desiccator. After this period, samples were kept in laboratory for 140 months to enable carbonation reaction of lime. ASTM C109 [22] and ASTM C593 [23] were applied for the moulding and storage of mortar mixtures.

The uniaxial compressive strengths values of mortars aged for 3, 6, 12 and 140 months were determined by mechanical tests (Shimadizu AG-I Mechanical Test Instrument) with the maximum force of 15 kN force, at the speed of 1mm/min. The tests were conducted on three parallel samples at specified intervals for each mortar type. The only exception was the test at 140 months of a mortar sample prepared of white limestone, which could not be conducted due to its loss in the long experimental process. The test results were displayed and recorded via the software of the mechanical test instrument.

Hydraulic properties of mortars were determined by thermal analysis conducted on 1 gram of powdered samples in a high-temperature furnace. During the analysis, weight losses between temperatures 200-600°C and 600-900°C were measured by means of a precision balance with a sensitivity of 0.0001 gr. TGA was not preferred for determination of hydraulic properties since very small amounts of samples (~1 mg) required by TGA are not representative for mortars.

XRD patterns of lime mortars broken during mechanical tests were used to establish a relationship between their carbonation degrees. Height ratios of intense diffraction peaks of $CaCO_3$ (29.4 2θ) and $Ca(OH)_2$ (34.0 2θ) were compared for a semi-quantitative evaluation of mortar carbonation for certain time intervals.

3. Results and discussion

3.1. Characteristics of uncalcined, calcined, slaked and carbonated limes made of selected marbles and limestones

Lime is produced out of four different marbles and limestones and used in mortar preparation by hot lime mix method. Microstructural, mineralogical, and chemical characteristics of marbles and limestones and calcined, slaked, and carbonated limes, were determined by SEM-EDS, XRD, and TGA.

Microstructural characteristics of marbles and limestones were identified by SEM analysis. Analysis results showed that Marmara and Mugla marbles (Figure 1a, 1b) were composed mainly of coarse-grained (800 μm to 3.5 mm) calcite crystals, whereas fine-grained (~ <15 μm) calcite crystals were the essential constituent of white and grey limestones (Figure 1c, 1d). Unlike white limestone samples, grey ones contain high amounts of diatoms composed of skeletal shells originating from many kinds of unicellular algae (Figure 1d) [24].

The analysis of chemical composition conducted by SEM-EDS revealed that all marbles and limestones were mainly composed of CaO. In addition, MgO content of Mugla marble and SiO_2 content of grey limestone, were significantly higher than those of Marmara marble and white limestone (Table 1). High content of SiO_2 in grey limestone may be due to the presence of diatoms.

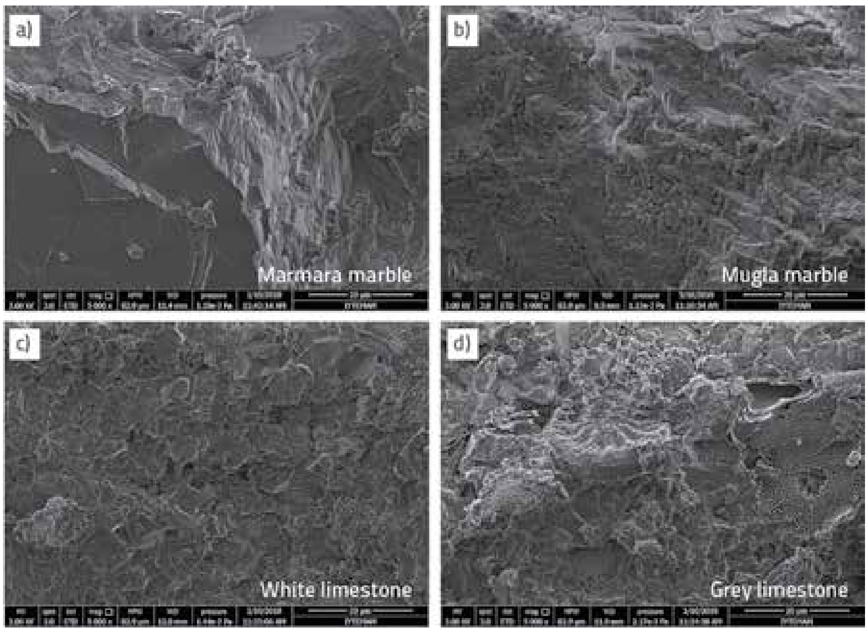

Figure 1. Coarse-grained calcite crystals of marbles (a, b) and fine-grained calcite crystals of limestones (c, d)

Table 1. Major oxide compositions of marbles and limestones

Samples	Major oxides [%]							
	CaO	MgO	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	Na_2O	K_2O
Marmara marble	96.7 ±0.2	2.3 ±1.6	ND	ND	ND	ND	ND	ND
Mugla marble	81.6 ±0.4	10.1 ±0.7	2.4 ±0.4	2.2 ±0.4	0.8 ±0.8	0.2 ±0.3	2.2 ±0.5	0.5 ±0.3
White limestone	84.4 ±2.5	2.8 ±0.6	5.2 ±0.6	2.6 ±0.4	2.4 ±0.7	0.5 ±0.3	1.5 ±0.9	0.6 ±0.1
Grey limestone	58.6 ±0.8	2.8 ±0.3	26.5 ±3.5	6.6 ±0.0	2.7 ±0.7	0.5 ±0.4	1.2 ±0.6	1.1 ±0.1
ND - Not detected								

It was established by XRD analyses that calcite was the principal mineral component of all limestone and marble samples (Figure 2). Other minerals detected by these analyses were dolomite for Mugla marble, and amorphous silica minerals originating from diatoms for grey limestone.

Calcination of marbles (Figures 3.a, 3.b) and limestones (Figures 3.c, 3.d) was found to start at 670 °C and end at 836 °C as a result of TGA. Depending on these results, marble and limestone samples were calcined at 850 °C to produce quicklime for mortar preparation. XRD patterns of quicklimes revealed that calcium oxide was the major mineral for all samples (Table 2). In addition to calcium oxide, the analysis revealed magnesium oxide due to magnesium carbonate for Mugla marble, and dicalcium silicate (Ca_2SiO_4) due to the reaction of diatoms with CaO for grey limestone. This result confirms that the production of hydraulic lime from limestone containing diatoms at relatively low temperature was possible [25].

Microstructural characteristics of calcined marbles and limestones showed that the quicklimes formed at this temperature have mostly nano-porous structures (pore sizes < 1μm) due to removal of carbon dioxide from calcite minerals (Figure 4). This porous structure can make calcium oxide crystals

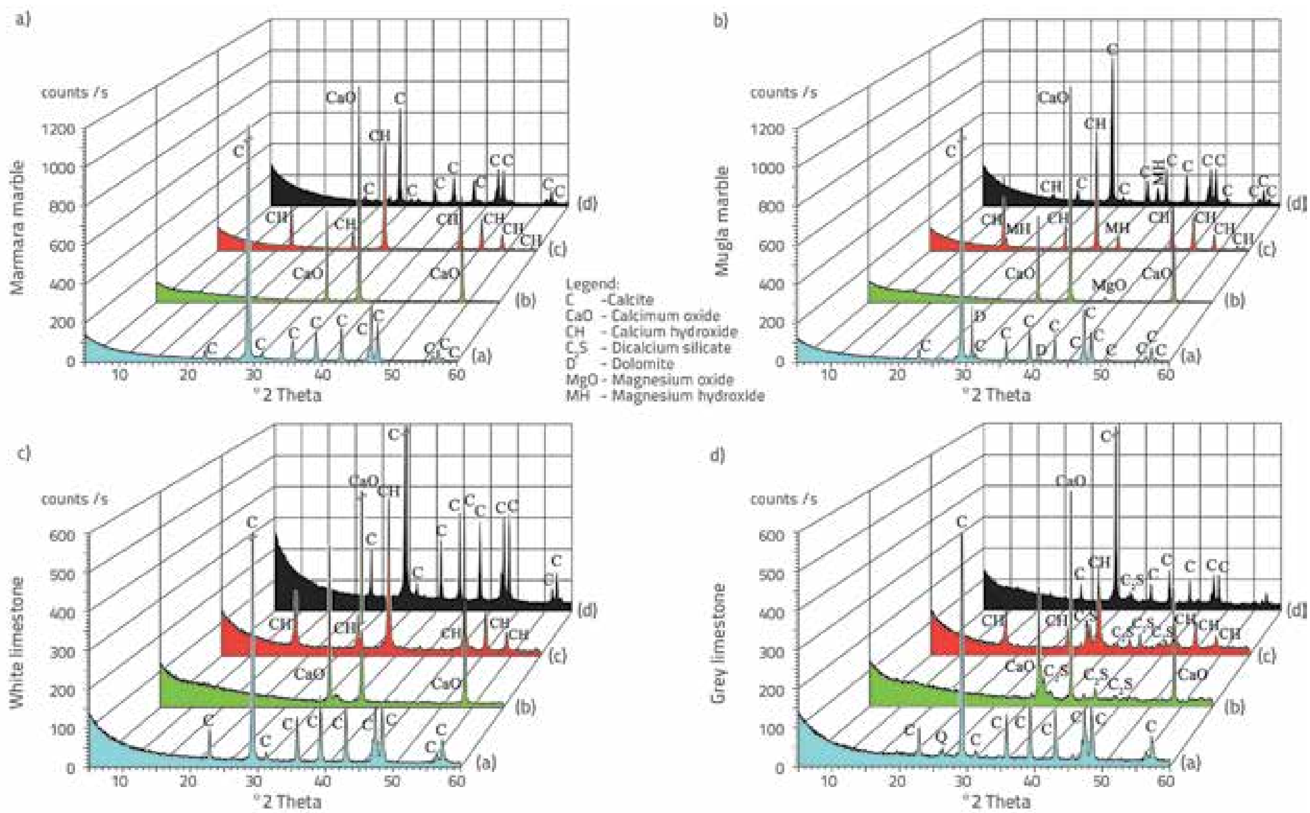

Figure 2. XRD patterns of: a) uncalcined; b) calcined; c) slaked; d) carbonated limes produced from marbles and limestones

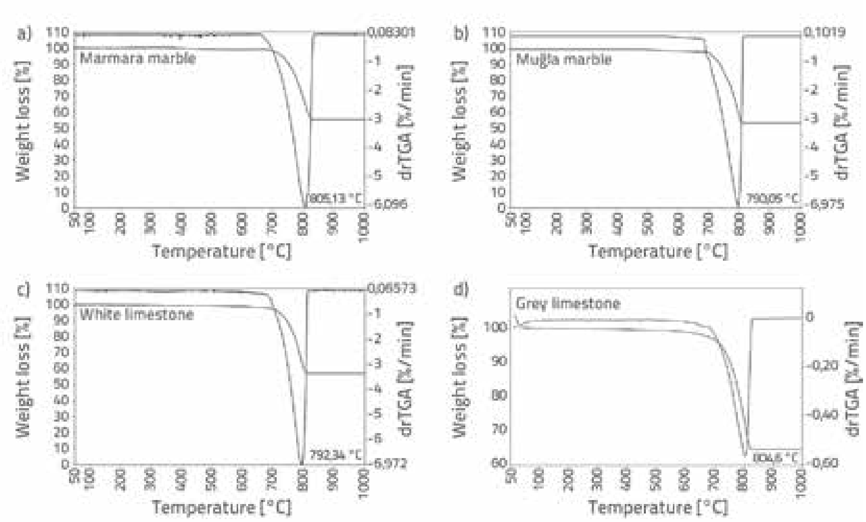

Figure 3. TGA-drTGA graphs of: a) Marmara marble; b) Muğla marble; c) white limestone; d) grey limestone

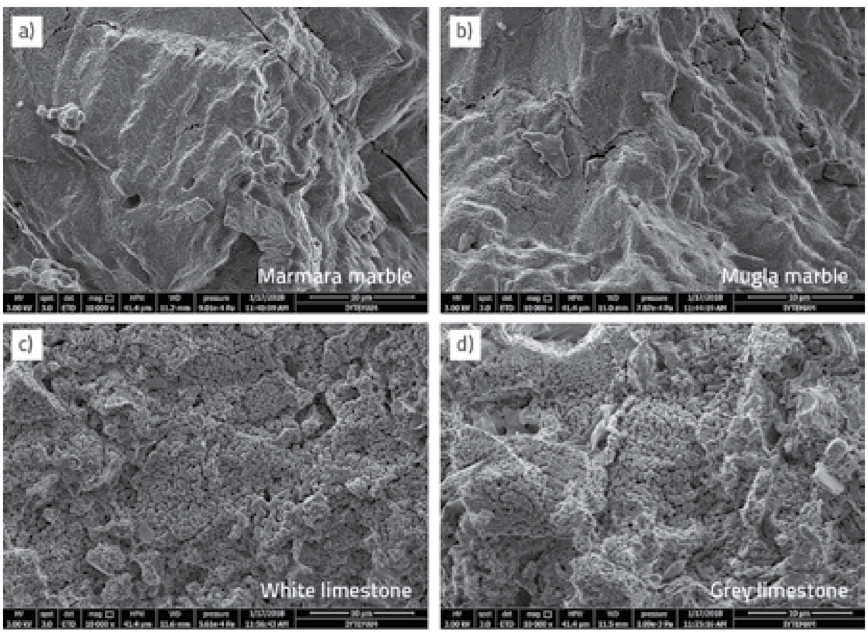

Figure 4. Porous structures of calcined limes (quicklime) of marbles (a,b) and limestones (c,d)

more reactive with water [26]. In addition to nano-porous calcium oxide minerals, formation of calcium silicate was also indicated for grey limestones as a result of reaction between diatoms and calcium oxide (Eq. 3) (Figure 4d).

$$2CaCO_3(s) + SiO_2(s) \rightarrow Ca_2SiO_4(s) + 2CO_2(g) \quad (3)$$

Afterwards, calcined marbles and limestones were slaked with distilled water and dried in a desiccator. Mineralogical composition of slaked limes determined by XRD revealed that portlandite ($Ca(OH)_2$) was the common major mineral in all samples (Figure 2). In addition to portlandite, magnesium hydroxide for Mugla marble (Figure 2.b) and dicalcium silicate for grey limestone samples were also observed on XRD patterns (Figure 2.d).

SEM images indicated that slaked limes of Marmara and Mugla marbles had a microcrystalline structure mainly consisting of portlandite crystals that form a network with grains of irregular shape and average size in the sub-micrometer range (Figure 5.a). On the other hand, slaked limes of white and grey limestones were composed of the cluster of very fine particles of less than 1 μm in diameter and of amorphous appearance. Due to the formation of calcium silicate, weathered textured diatomite was also present in slaked grey limestone (Figure 5.b).

Mineralogical, hydraulic and microstructural characteristics of carbonated limes were investigated on samples aged for one month.

Calcite mineral was indicated on the XRD patterns of all carbonated limes (Figure 2). However, calcium silicate peaks observed in the grey lime disappeared after carbonation of lime (Figure 2.d). This can be explained by the formation of amorphous calcium silicate hydrates that cannot be determined by the XRD analysis during the carbonation reaction [9].

SEM-EDS analysis revealed that carbonated limes produced from marbles (Figure 6a) and white limestones were composed of micritic calcite crystals. Well-developed and well packed crystals ranged approximately between 2 and 5 μm in size. On the other hand, carbonated lime produced from grey limestone was composed of micritic calcite crystals connected with fibrous structures rich in silicon oxide (Figure 6b), which points to the calcium silicate formation [25].

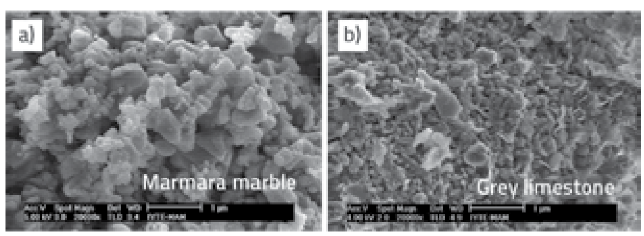

Figure 5. Microcrystalline and amorphous structures of slaked limes of: a) Marmara marble; b) grey limestone

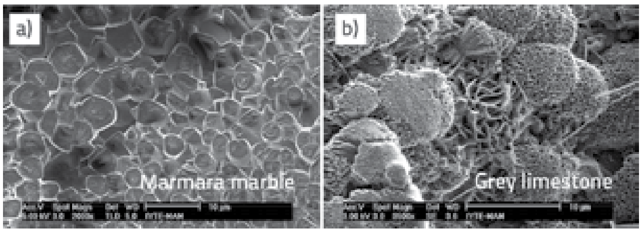

Figure 6. a) Micritic calcite crystals in carbonated lime of Marmara marble; b) calcium silicate formation in carbonated lime of grey limestone

Chemical compositions identified by SEM-EDS were used to calculate hydraulic index (H.I.) and cementation index (C.I) values of carbonated limes in order to describe their hydraulic properties. H.I. values lower than 0.1, and C.I. values lower than 0.3 point to the non-hydraulic character of lime, whereas H.I. values higher than 0.4 and C.I. values ranging between 0.7 and 1.1 exhibit a highly hydraulic character [21]. Hydraulic and cementation indices of the studied samples show that only carbonated lime of grey limestone is of highly hydraulic nature (Table 2).

Table 2. Hydraulic and cementation indices of carbonated limes

Sample	Hydraulic Index (H.I.)	Cementation Index (C.I.)
Lime of Marmara marble	0.00	0.00
Lime of Mugla marble	0.06	0.11
Lime of White limestone	0.12	0.22
Lime of Grey limestone	0.58	1.35

3.2. Microstructural and mechanical characteristics of lime mortars prepared by hot lime mix method

The effects of different limes on microstructural characteristics of mortars were investigated on fully carbonated samples (aged for 140 months) to observe possible long-term influences of the hot lime mix method.

SEM analysis conducted on fully carbonated mortars revealed that they were composed of a superficial layer and an inner part that may be differentiated by the difference in their pore and crystal structures (Figure 7). Superficial layers were outer surfaces of mortars which were directly exposed to CO_2 of the atmosphere and formed due to rapid carbonation. These layers 1 mm in thickness had a denser and low porous structure in which crystals were well-packed compared to the inner parts of mortar [27].

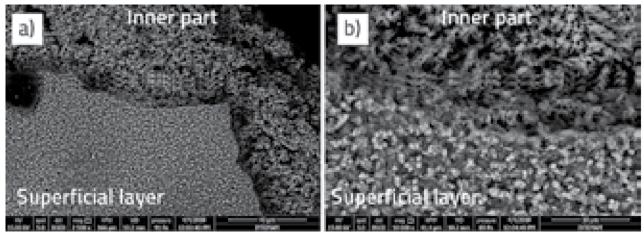

Figure 7. SEM images showing crystal structure and porosity of inner part and superficial layers of mortars aged for 140 months

Within the inner parts, calcite crystals were more apparent in aerial lime mortars prepared by using marbles and white limestone (Figures 8.a, 8.b). These crystals were generally elongated particles measuring <2μm in size, with rather sharp finishes. Their characteristic feature is the spongy like texture of their surfaces. However, calcite crystals lost their sharpness in lime mortar obtained by using grey limestone containing diatoms (Figures 8.c, 8d.). Rather, this mortar was mainly comprised of amorphous particles pointing to its hydraulic properties. Despite the difference in their crystal structure, all mortars had porous microstructures and their pores were homogenously dispersed. Pore sizes varied between 1 and 10 μm in mortar made of grey limestone, and between 1 and 5 μm in the rest. The porous microstructure of all mortars and the spongy texture of calcite crystals of aerial lime mortars may be the indicators of hot lime mix method in which water vapour was generated by heat released during mixing. Porous microstructures of hot lime mortars were also noted in recent studies [13].

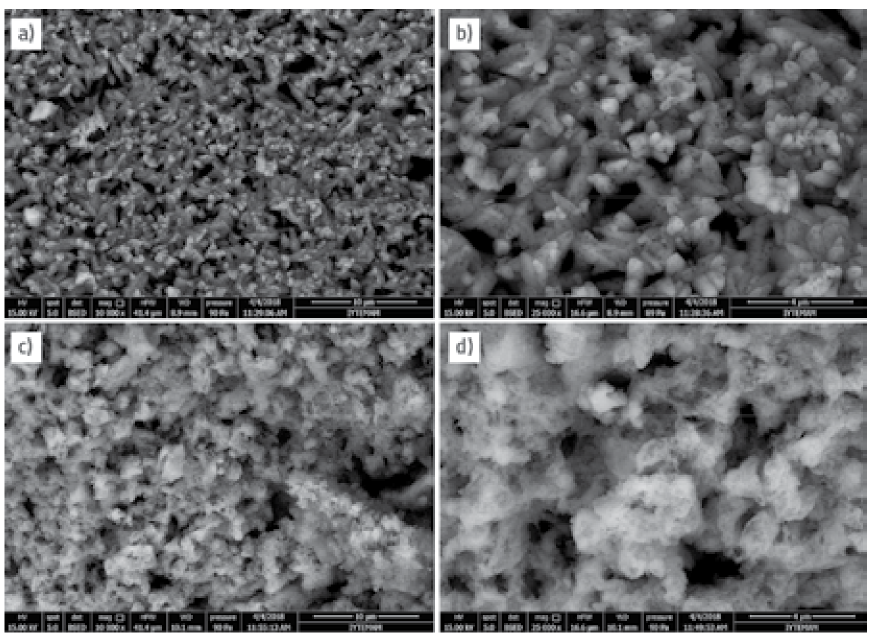

Figure 8. Porous microstructures of lime mortars (a, c) and spongy microstructures of calcite crystals (b, d)

Carbonation process can last for many years depending on the characteristics and dimensions of mortar and also on atmospheric conditions [18, 28]. In this study, carbonation degrees of lime mortars were semi-quantitatively compared using height ratios of the intense diffraction peaks of CaCO3 (29.4 2θ) and Ca(OH)2 (34.0 2θ) on their XRD patterns at 3, 6, 9, 12 and 140 months [29]. $CaCO_3$/$Ca(OH)_2$ ratios were found to be around 2 for lime mortars of marbles (Figures 9.a, 9.b) and white limestone (Figure 9c) whereas they were over 4 for lime mortar of grey limestone (Figure 9d) at 6 months (Figure 10.a). This ratio calculated for 140 months aged samples amounted to 20 in the first case, and 62 in the second case (Figure 10.a). The higher carbonation degree of lime mortar obtained from grey limestone can be explained by the fact that $Ca(OH)_2$ was consumed by both the CO_2 of air and the hydraulic reaction of amorphous particles of siliceous origin. Hydraulic characteristics of lime mortars were determined by thermal analysis at the 140th month since reliable results can only be obtained from samples in which carbonation is almost completed. For this purpose, weight losses between 200 and 600 °C, due to loss of chemically bound water of hydraulic products, and between 600 and 900 °C due to release of CO_2 of $CaCO_3$, were calculated. CO_2/ chemically bound water ratios between 1-10 are determinant for hydraulic lime mortars [30, 31]. Thermal analysis results revealed that lime mortar from grey limestone possessed hydraulic characteristic with the chemically bound water/CO_2 ratio of 3.92, whilst the rest were non-hydraulic. Hydraulic character of lime mortar obtained from grey limestone could be attributed to the presence of dicalcium silicates originating from diatoms found in grey limestone.

Mechanical properties of mortars were defined by their uniaxial compressive strengths. For this purpose, uniaxial compressive strengths of mortars were determined at 3, 6, 12 and 140 months. During application of compression, detachment of superficial layers of mortar can initially be observed, which is generally accepted as "the outer carbonated part" of the mortar [27]. Carbonation is the essential process that improves mechanical properties of lime mortars. Uniaxial compressive strength measurements of all samples revealed the enhancing effect of carbonation.

Uniaxial compressive strengths ranged between 0.8 and 1.3 MPa at 3 months, and between 1.5 and 2.4 MPa at 12 months for non-hydraulic lime mortars, whereas 3.9 MPa at 3 months and 7.2 MPa for hydraulic lime mortar (Figure 10.b). These values approximately correspond to ranges given in recent studies on lime mortar production [13, 14, 17, 18, 27, 28, 32, 33]. At 140 months, compressive strengths remained between 1.8-2.6 MPa for non-hydraulic lime mortars while it increased to 12.3 MPa for hydraulic lime mortar. The compressive strength

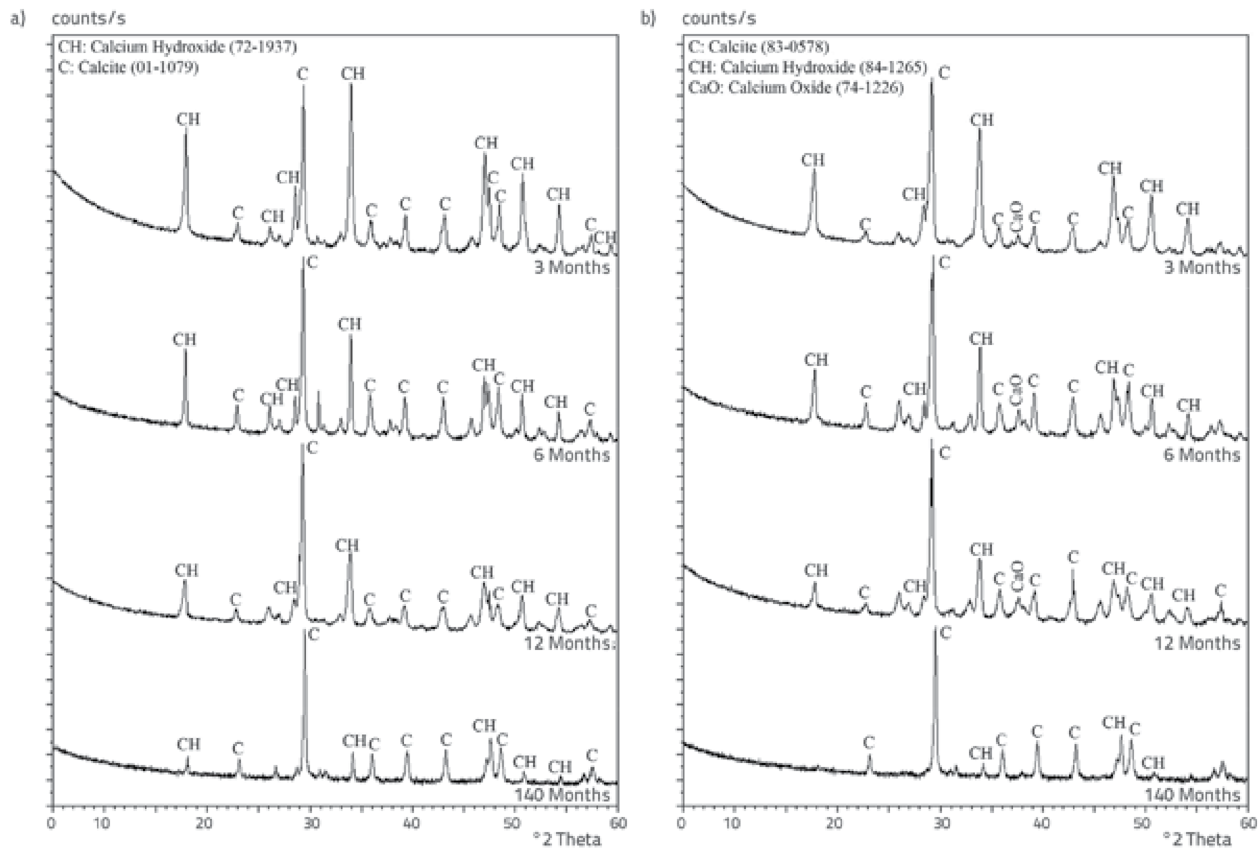

Figure 9. XRD patterns of mortars prepared by: a) Marmara marble; b) Mugla marble; c) white limestone; d) grey limestone after 3, 6, 12 and 140 months of carbonation (first part)

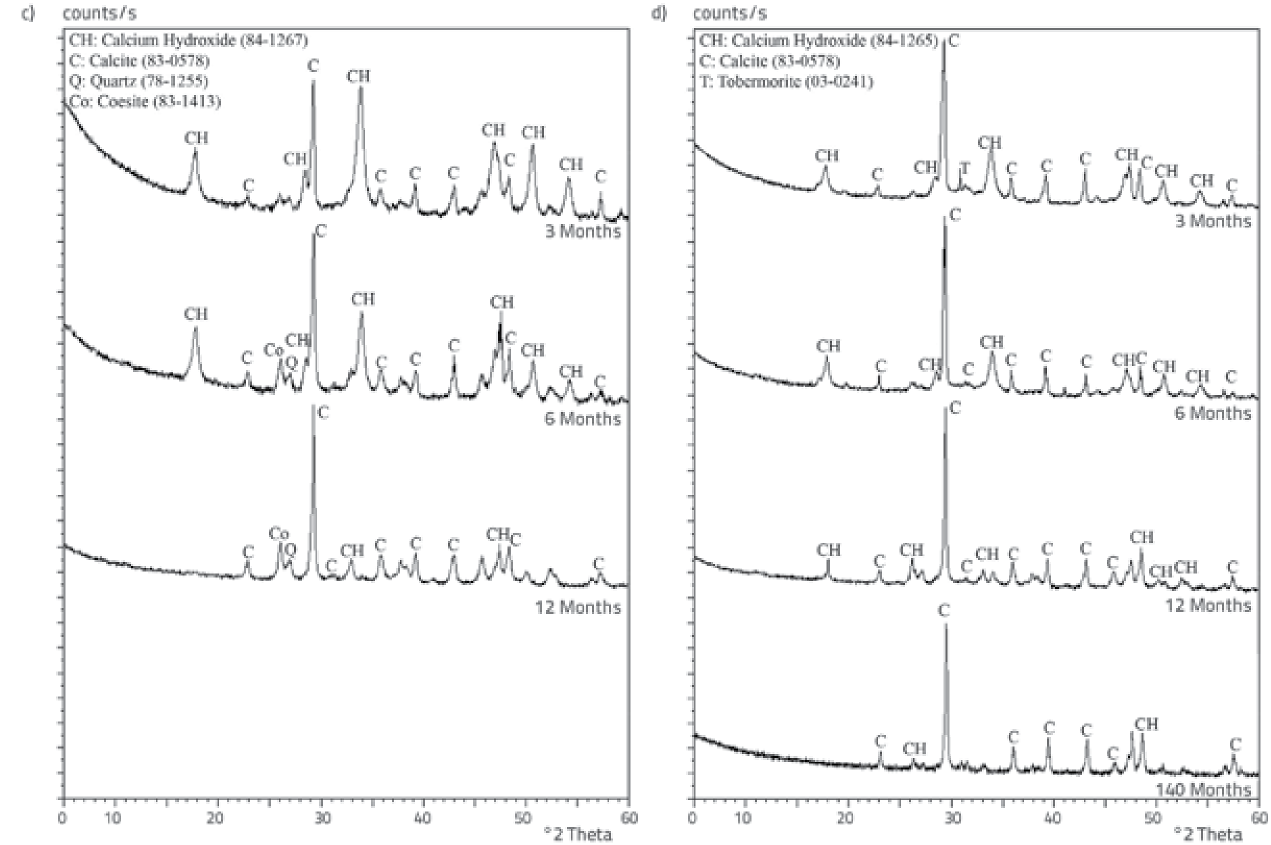

Figure 9. XRD patterns of mortars prepared by: a) Marmara marble; b) Mugla marble; c) white limestone; d) grey limestone after 3, 6, 12 and 140 months of carbonation (second part)

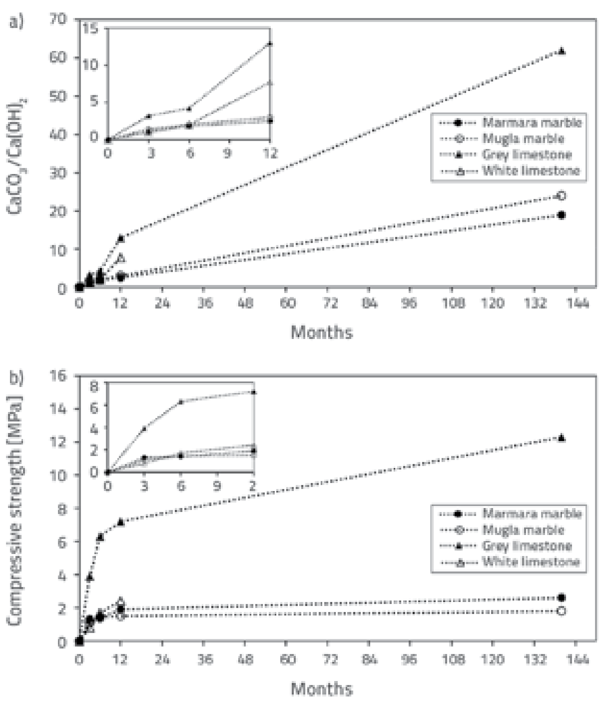

Figure 10. Variation of: a) carbonation ratio; b) compressive strength of lime mortars

values of non-hydraulic lime mortars remained in close range with each other at all time intervals. This reveals that the differences between their chemical compositions regarding low amounts of magnesium oxide and silicon oxides (nearly 10 %) did not influence their mechanical properties significantly.

Compressive strength measurements carried out at the end of 140 months were far beyond researches intended for production of new lime mortars since the majority of them were generally concluded at the end of two years. Therefore, it would be better to compare compressive strengths of 140 months aged lime mortar samples with lime mortars of historical structures. Compressive strength values of 140 months aged lime mortars were found to be moderately below the values of historic lime mortars [25, 34, 35]. This difference may depend on the use of pozzolanic aggregates in the historic lime mortars which ensured their hydraulic characteristics.

4. Conclusion

In this study, lime was produced using two different marbles, white limestone and grey limestone containing diatoms, so that it can be used in mortar preparation by the hot lime mix method. Mortars based on calcined grey limestone containing high amounts of diatoms exhibited relatively high compressive strength compared to other mortars due to formation of

dicalcium silicate at relatively low temperature (850°C). All lime mortars have porous microstructures consisting of calcite crystals of spongy like texture in aerial limes, whereas amorphous particles refer to hydraulic properties in mortars produced from grey limestone. The porous microstructure of all mortars and the spongy texture of calcite crystals of aerial lime mortars may be the indicators of hot lime mix method in which water vapour is generated as a result of heat released during mixing.
This study shows that characteristics of limestones used for the production of limes, and the preparation method, directly affect hydraulic, mechanical and microstructural properties of mortars. Thus, the studies concerning historic mortar characterization and production of intervention mortars should take into account properties of raw materials of limes, as well as the properties of other ingredients of mortar.

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