

## Sulfate attack of Algerian cement-based material with crushed limestone filler cured at different temperatures

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### Abstract

The cement production industry is one of the main consumers of energy and raw materials. Over the last years, a great effort has been made in order to substitute clinker for less energy demanding materials. In many countries, it is popular to use limestone as admixture material to improve the consistency of concrete. Nevertheless, the construction industry needs durable materials with improved properties. Following this objective, this work is a part of an ongoing project developed in the materials laboratory of LABMAT/ENSET, Oran, Algeria. This paper presents data on engineering properties such as compressive strength and visual change of mortar specimens incorporating limestone filler, subjected to severe sulfate attack and cured at: (i) 5° C and (ii) 23 °C. Specimens with 4 replacement levels of limestone filler (0%, 10%, 20%, and 30% of cement by mass) were immersed in magnesium sulfate solutions with 5% of concentration for periods of up to 360 days. In order to identify the products formed by sulfate attack, microstructural analyses, such as XRD and SEM, were also performed on paste and mortar samples with similar replacement levels of limestone filler.

The test results demonstrated that mortar and paste samples incorporating higher replacement levels of limestone filler were more susceptible to sulfate attack at low temperatures. On top of that, the deterioration was strongly associated with thaumasite, gypsum, and brucite formation in the magnesium sulfate solution.

**Key Words:** Crushed limestone fillers, Microstructure, Reduction in compressive strength (RCS), X-ray, Thaumasite, MgSO<sub>4</sub> solution

## Introduction

In the last decades, environmental challenges arose due to the necessity of reducing worldwide levels of CO<sub>2</sub> emissions, energy consumption, and the use of natural raw materials. The application of the Kyoto Protocol further promoted an expressive increase in research efforts to find viable alternatives to minimize the pollution effects of the main productive processes.

To attain this aim, cement manufacturers are blending or intergrinding mineral additives such as slag, natural pozzolans, sand, and limestone. The reduced cost of limestone is mainly due to the energy savings by substitution of a portion of the calcined clinker with a small amount of limestone and the presence of limestone deposits near cement kilns, leading to reduced transportation costs.

The use of Portland cement containing limestone is common practice in European countries. The recent European Standard EN 197 identifies 2 types of Portland limestone cements (PLCs): Type II/A-L containing 6%-20%, and Type II/B-L containing 21%-35%. In addition, the inclusion of 5% of filler material that can be calcareous is accepted in all cements. During the 1990s, Latin American countries also moved in this direction and the use of limestone filler in Portland cements (PCs) was standardized. After many years of discussion, in 2004, the ASTM C150 (2004) standard specification for Portland cement was modified to allow the incorporation of up to a 5% mass fraction of limestone in ordinary Portland cements.

Furthermore, results on the sulfate performance of cement mortars containing limestone filler show different trends. Some authors (Zelic et al., 1999) concluded that limestone filler could increase the sulfate resistance of cement, while other researchers (Gonzalez et al., 1998; Xiaojian et al., 2008) found a decrease of sulfate resistance of this cement depending on the replacement level and clinker composition.

Sulfate attack is a generic name for a set of complex and overlapping chemical and physical processes caused by reactions of numerous cement components with sulfates originating from external or internal sources (Skalny et al., 1976). External sulfate attack is caused by sulfates from ground water, soils, solid industrial waste and fertilizers, atmospheric SO<sub>2</sub>, or liquid industrial wastes. The ready availability of these sulfates causes damage to concrete, depending on its concentration and solubility, the transport of water, and environmental conditions (Skalny et al., 1976; Torres et al., 2003; Senhadji et al., 2005). External sulfate attack on concrete is itself a well-documented phenomenon (Cohen et al., 1991; Brown et al., 1998), but the influence of the new materials that are being used for the production of blended cement has not yet been completely comprehended.

Many studies have shown that the ingress of sulfates into a concrete structure by ground water containing soluble sulfates or soils laden with sulfates leads to several expansive reactions and the alteration of the microstructure (Crammond and Halliwell, 1995). Ettringite is a common mineral formed during this process and may lead to the expansion and cracking of concrete. Besides ettringite, gypsum is also formed in the system, which is dependent upon the sulfate concentration. Formation of gypsum softens the cement paste and therefore leads to the deterioration of the paste microstructure. Thaumasite is formed in the system when limestone is present in the concrete and is subjected to sulfate attack at low temperatures (5 °C) in an environment with high moisture (Crammond and Halliwell, 1997; Hartshorn et al., 1999). The formation of thaumasite leads to the complete destruction of the cement paste and loss of cohesiveness (Sarkar et al., 1998). The occurrences of thaumasite in concrete fields in colder environments have been reported in the UK, Italy, Germany, and Canada (Bickley et al., 1994; Hartshorn et al., 1999). However, recent papers show that thaumasite can be formed in the warmer climatic conditions of the USA (Hartshorn et al., 1999; Brown et al., 2000).

In Algeria, as in many other countries, crushed limestone is the main source of aggregates used in concrete.

However, such production is associated with high percentages of fines that make these aggregates unacceptable in the concrete design mix. As a result, over 20% of such products cannot be used and become hazardous to the environment.

In the present study, thus, the effect of replacement levels (0%, 10%, 20%, and 30% by mass) of limestone filler on magnesium sulfate attack at low and ambient temperatures was investigated. The experimental study was developed to obtain conclusive data on the negative or positive effects of limestone filler in sulfate solutions and of both curing temperatures on the formation of expansive products.

## Materials and Methods

### Materials

The mortar and paste mixtures were prepared at the laboratory of the Civil Engineering Department, ENSET, Oran, Algeria, using the following materials.

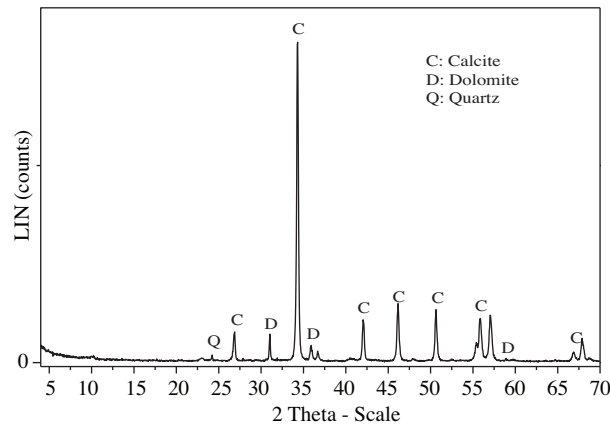
*Cement* The cement used was a blended Portland cement type (CEM I 42.5R), manufactured by the Zahana Cement Company, located in the west of Algeria. The factory cement conforms to the Algerian standard NA 442 (2003), which was mainly based on the European EN 197-1. Its physical properties and chemical composition data are presented in Table 1.

**Table 1.** XRF<sup>a</sup> chemical composition and physical data for the cement and limestone used.

Chemical composition	Cement	Limestone
SiO <sub>2</sub> , %	21.35	7.89
Al <sub>2</sub> O <sub>3</sub> , %	459	2.58
Fe <sub>2</sub> O <sub>3</sub> , %	5.52	1.13
CaO, %	63.89	45.45
MgO, %	1.37	1.72
SO <sub>3</sub> , %	2.72	0.21
K <sub>2</sub> O, %	0.41	0.95
Na <sub>2</sub> O, %	0.13	0.0
CL	0.0	0.0
LOI	0.48	42.48
Mineralogical compound (Bogue calculation)		
C <sub>2</sub> S, %	25.69	–
C <sub>3</sub> S, %	47.15	–
C <sub>3</sub> A, %	2.84	–
C <sub>4</sub> AF, %	16.7	–
Calcite content (CaCO <sub>3</sub> , %)	–	86.98
Setting time (min)	Initial set	103
	Final set	225
Specific gravity (kg/m <sup>3</sup> )	3100	2670
Fineness (cm <sup>2</sup> /g)	3500	3200

a: X-ray florescence (XRF) spectrometry

*Limestone* The limestone was obtained from limestone quarries in the city of Oran. It was ground to a specific surface area of 3200 cm<sup>2</sup>/g, with a maximum nominal size of 0.080 mm. Its content of CaCO<sub>3</sub> was about 86.98%. The X-ray diffractogram of limestone is shown in Figure 1. Its chemical composition is given in Table 1. The chemical composition of the materials was determined by X-ray fluorescence (XRF).



**Figure 1.** X-ray diffractogram of crushed limestone.

*Sand* Local well-graded crushed sand obtained from a quarry in Kristel (Oran) was used. Its physical properties were as follows: relative specific density, 2650 kg/m<sup>3</sup>; water absorption, 1.5%; bulk density, 1500 kg/m<sup>3</sup>; and maximum grain size, 3 mm.

### Specimen preparation, curing, and testing

Four different mortar mixtures were prepared to compare the effect of adding limestone filler as a cement replacement material. The first was the control, without limestone, and the other 3 mortar mixtures were made by replacing cement with 10%, 20%, and 30% limestone by mass. The letter in each mix designation representing the mix is (LS), and the numbers 0, 10, 20, and 30 represent the percentages of the replacement of Portland cement by limestone filler at 0%, 10%, 20%, and 30% by weight of cementitious material, respectively.

Mortars and pastes based on the materials mentioned above were mixed and cast as 50 × 50 × 50 mm<sup>3</sup> prisms for compressive strength measurement, meeting the requirements of ASTM C 305 (1995). The ratio of water to cementitious material (w/cm) was 0.5 and the ratio of sand to binder mass was maintained equal at 2.75 by weight. All of the specimens were compacted on a vibrating table. The tops of all specimens were leveled and finished with a steel trowel after compaction. The specimens were then covered with a plastic sheet to avoid any loss of water.

Immediately after casting, the specimens were air-cured in the laboratory for 24 h, removed from the mold, weighed, and cured in saturated limewater until they achieved 30 ± 3 MPa in compressive strength, for 27 days at room temperature (23 ± 2 °C). After 27 days of cure time under those conditions, excluding the control specimens, which were continuously immersed in water, all others were transferred into a 5% MgSO<sub>4</sub> solution. The samples were cured at: (i) 5 °C (laboratory refrigerator, ± 2 °C) and (ii) 23 °C (laboratory environment, ±3 °C). In both cases, the MgSO<sub>4</sub> solution was changed every 3 months.

The fluidities of mortars were determined by flow table tests according to the ASTM C 230-90 (1990) American standard. The mortar sample spread diameter, measured before and after 15 strokes (1 stroke per second), represents the consistency of different mortars.

Prior to the compressive strength tests, cubic mortar specimens were thoroughly visually investigated, and signs of deterioration, e.g. cracking, spalling, and delamination, were checked. The compressive strength of the mortar cubes was measured after 28, 150, 240, and 360 days of 5% magnesium sulfate exposure. Companion

mortar specimens cured in tap water for the same period were also tested. The strength deterioration was evaluated by the reduction in compressive strength (RCS), as expressed in Eq. (1):

$$\text{Reduction in compressive strength (\%)} \\ \text{RCS} = [(CS_{cw} - CS_{cs}) / CS_{cw}] \times 100 \quad (1)$$

Where:

$CS_{cw}$  (in MPa) is the average strength of the specimens cured in tap water, and

$CS_{cs}$  (in MPa) is the average strength of the specimens immersed in 5% magnesium sulfate solutions.

Three mortar specimens from the tap water and magnesium sulfate solutions were tested in each test period, respectively, and then the values were averaged.

XRD observations on paste samples with or without limestone filler were carried out to identify the products formed by sulfate attack after 360 days of immersion in 5% magnesium sulfate solution at 5 °C. Cement pastes were chosen instead of mortars or concretes in order to eliminate the parallel effect of the calcareous sand and/or aggregates on the products' formation. The XRD analysis was carried out with a Siemens D-5000 X-ray diffractometer with Cu  $K\alpha$  radiation and  $2\theta$  scanning, ranging between 4° and 70° of  $2\theta$ , with a step size of 0.04° and a measuring time of 2 s/step. A voltage of 40 kV and current of 30 Ma were used.

To complete the investigation of deterioration products' formation, a study using a scanning electron microscope (SEM) was necessary. The demolition products were identified by means of an SEM, a JEOL JSM 6380LV model. In order to determine the properties of the mortar specimens from a microscale view, SEM photos were taken at the end of the 360-day testing period.

## Results and Discussion

The flow values were between 87% and 68% (Table 2). The inclusion of 10%, 20%, and 30% limestone filler reduced the workability, on average, by 2%, 6%, and 10%, respectively. The addition of limestone filler seems to reduce the consistency of mortar mixtures. This may be explained by the difference of the chemical composition and fineness between the limestone and the Portland cement used.

**Table 2.** Compressive strength values of mortar specimens cured in tap water and flow of normal mortar.

Sample	Compressive strength, MPa) (at room temperature, 23 ± 2 °C)			Flow of normal mortar (%)	Density (kg/m <sup>3</sup> )
	2 days	7 days	28 days		
LS0	15.6	31.7	46.3	86.5	2339
LS10	13.3	29.8	44.3	84.5	2315
LS20	9.3	18.9	37.3	81.0	2296
LS30	1.3	17.1	32.0	77.5	2286

### Compressive strength of mortars

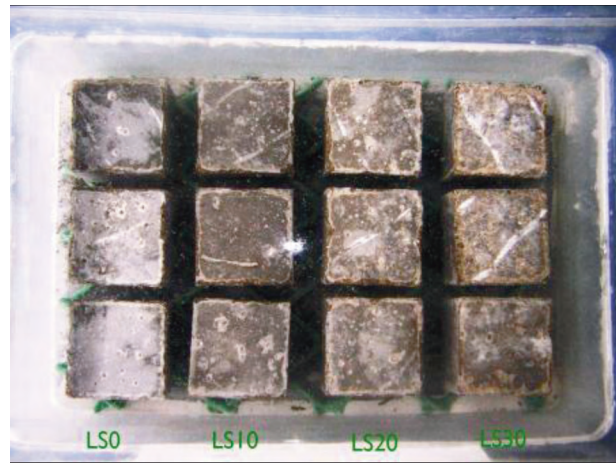
The results for compressive strength development of 50 mm cube specimens with a w/cm of 0.5 cured in tap water at the ages of 2, 7, and 28 days and the normal mortar flow of the tested samples are summarized in Table 2. These results clearly show the decreased values of compressive strength with the increased replacement levels of limestone filler. It is believed that these are also partially associated with the change of the water to Portland cement ratio with respect to the hydration process.

### Visual inspection

The specimens were visually examined on a regular basis. Photos of specimens stored in the magnesium sulfate solution for 360 days are presented in Figures 2 and 3. The samples stored at 5 °C showed the first signs of deterioration after 5 months of exposure, while the specimens stored at 23 °C did not show any clear evidence of sulfate attack for up to 9 months. The discussion below concerns the samples stored at 5 °C. In all cases, the first sign of attack was the deterioration of the corners, followed by cracking along the edges. Progressively, expansion and spalling took place on the surface of the specimens.



**Figure 2.** Mortar cubes (50 mL) exposed to 5% magnesium sulfate solution for 360 days at 5 °C.



**Figure 3.** Mortar cubes (50 mL) exposed to 5% magnesium sulfate solution for 360 days at 23 °C.

After 5 months of immersion in a solution of 5%  $\text{MgSO}_4$  at 5 °C, all 4 surfaces of the specimens developed a white cover that was friable and started to peel off, leaving the aggregates uncovered and reducing the connectivity of the paste. As can be seen from Figure 2, it was clearly observed that the degree of surface deterioration was different with the replacement levels of limestone filler. The extent of surface deterioration after 360 days of exposure had a tendency to increase with the increased replacement level of the limestone filler. The LS30 mortar sample displayed many wide cracks, spalling, and substantial material loss at the faces as well as at the corners and edges, while the LS0 mortar sample only exhibited cracks around the corners and edges.

The specimens preserved in a solution of 5%  $\text{MgSO}_4$  at 23 °C for a year did not present any surface damage, except for some deterioration at corners (Figure 3). Both mortars, with and without limestone addition, had the same behavior under the same preservation conditions, and it seems that the damage proceeded independently of limestone addition. Consequently, the specimens' surface damage appears to have been inversely related to the temperature of conservation. It is noteworthy that the specimens that were preserved at 23 °C had approximately the same damage that specimens preserved in a solution of 5%  $\text{MgSO}_4$  at 5 °C had at the age of 3 months, which was also limited only to one surface of the mortars.

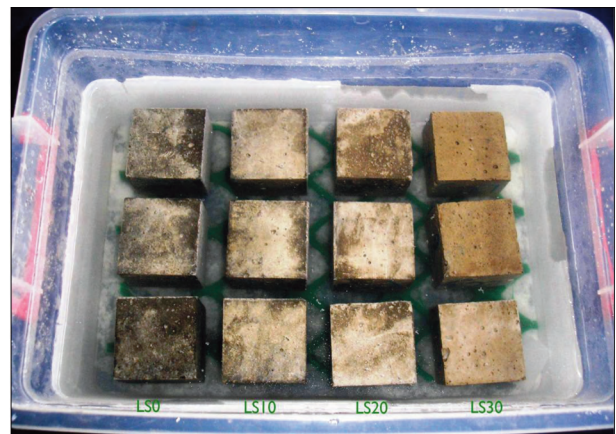
The mortars were sectioned so that not only the surface but all of the mass of the mortar could be examined macroscopically and the extent of the damage could be measured. This test showed that the damage, if present, was limited to the surface layer for all mortars and all temperatures.

Photos of specimens stored in tap water for 12 months at 5 °C and 23 °C are presented respectively in

Figures 4 and 5. After the initial 30-day curing under water, a little white precipitate was present in the sample containers, particularly on the top surface of the specimens. This material is attributed to the portlandite ( $\text{Ca}(\text{OH})_2$ ) leaching out from the mortar and a little carbonate in the water. With the increasing curing under water, the precipitate accumulated continuously, and a white powdery coating formed on the whole top surface and partial side surface of the control specimens when immersed for 1 year. However, storage in water appeared to be benign; no evidence of cracking or spalling was detected on the control samples during the whole experiment.



**Figure 4.** Mortar cubes (50 mL) cured in tap water for 360 days at 5 °C.



**Figure 5.** Mortar cubes (50 mL) cured in tap water for 360 days at 23 °C.

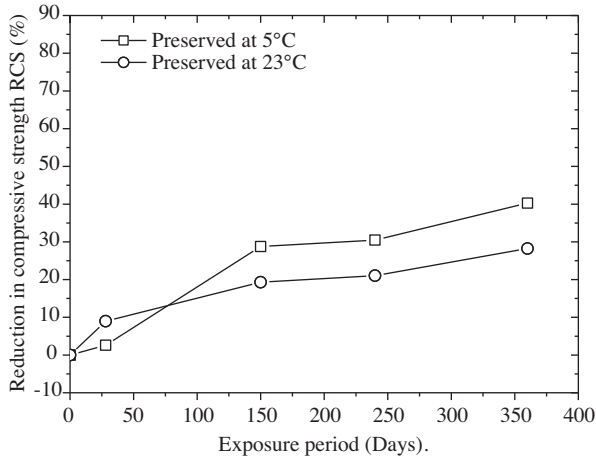
### Reduction in compressive strength (RCS)

After 27 days of pre-curing, in order to characterize the change of the compressive strength of mortar specimens incorporating limestone filler that was due to sulfate attack, the compressive strength values after storage in tap water and sulfate solutions were measured after up to 1 year of exposure. The reduction in the compressive strength values of mortar specimens with limestone filler of 0%, 10%, 20%, and 30% replacement levels immersed in sulfate solutions under various conditions, as well as that of the control samples (stored in water), is represented graphically versus time in Figures 6–10, respectively.

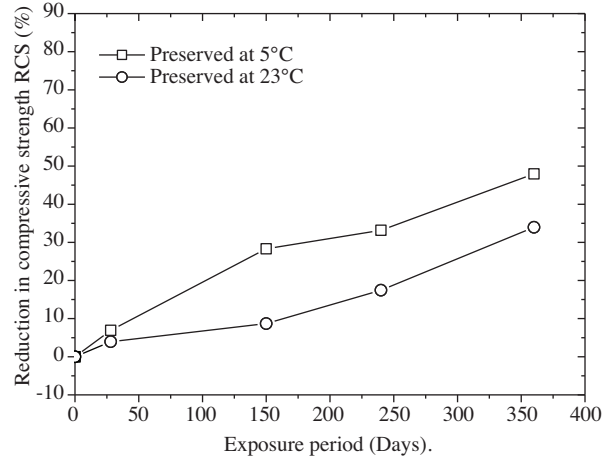
The compressive strength decreased with the period of exposure. Figure 6 shows the RCS of mortar specimens LS0 immersed in a magnesium sulfate solution and stored at 5 °C and 23 °C. For the LS0 mortar specimens, the RCS was almost linear after 28 days of exposure. This shows that the compressive strength of the LS0 cement mortar specimens decreased from 1% to 22% and from 4% to 15% within 12 months of exposure to the sulfate solution cured at 5 °C and 23 °C, respectively. The strength reduction in LS10, the cement mortar specimens with 10% limestone, was almost similar and did not vary much, with the RCS of LS10 being in the range of 3%-24% for specimens stored at 5 °C and in the range of 2%-17% for the mortars preserved at 23 °C (Figure 7).

Figures 8 and 9 show the compressive strength in blended cement mortar specimens LS20 and LS30. The results exhibited an initially marginally negative compressive strength loss/gain, which was more pronounced for the LS20 and LS30 mortar mixtures conserved at 5 °C after 28 days. This phenomenon is attributable to

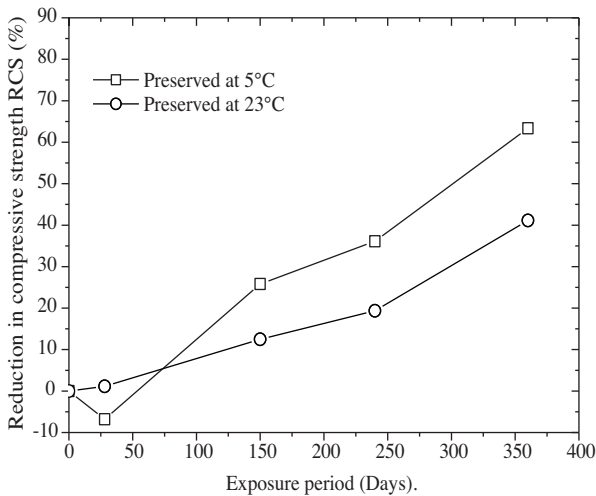
the filling up of pores by the expansive reaction products, thereby densifying the hardened mortar mix and increasing the weight and strength (Montemor et al., 2000).



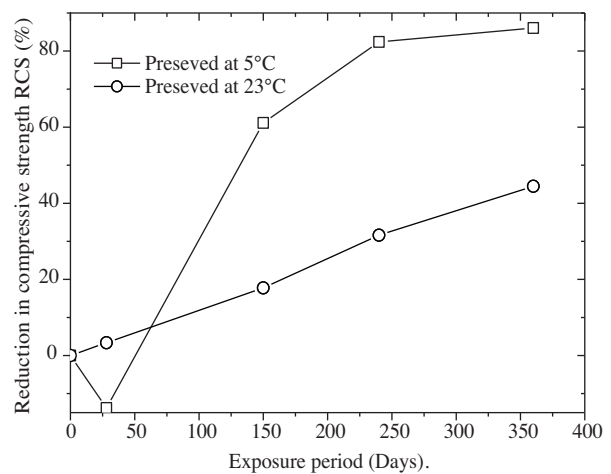
**Figure 6.** Reduction in the compressive strength of LS0 mortar specimens exposed to 5% magnesium sulfate solution at 5 °C and 23 °C.



**Figure 7.** Reduction in the compressive strength of LS10 mortar specimens exposed to 5% magnesium sulfate solution at 5 °C and 23 °C.



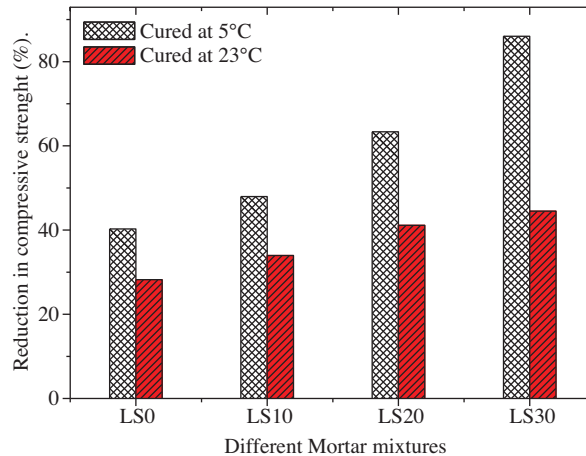
**Figure 8.** Reduction in the compressive strength of LS20 mortar specimens exposed to 5% magnesium sulfate solution at 5 °C and 23 °C.



**Figure 9.** Reduction in the compressive strength of LS30 mortar specimens exposed to 5% magnesium sulfate solution at 5 °C and 23 °C.

These results also emphasize that the incorporation of 20% and 30% limestone filler leads to an increased strength loss compared to that of the LS0 mortar specimens, regardless of the conservation temperature. The negative effect of the higher replacement level of limestone filler was observed more remarkably. With respect to the strength loss of mortar specimens after an exposure of 360 days (Figure 10), the worst performance was observed in all mortar specimens exposed to the magnesium sulfate solution at 5 °C, compared to the same mortar specimens exposed to the magnesium sulfate solution preserved at 23 °C.





**Figure 10.** Reduction in the compressive strength of LS0, LS10, LS20, and LS30 mortar specimens exposed to 5% magnesium sulfate solution at 5 °C and 23 °C after 360 days.

As can be seen from Figures 6-10, a decrease of storage temperature indicated a more pronounced deterioration of all mortar specimens with or without limestone compared to the mortar specimens immersed in magnesium sulfate solution at 23 °C.

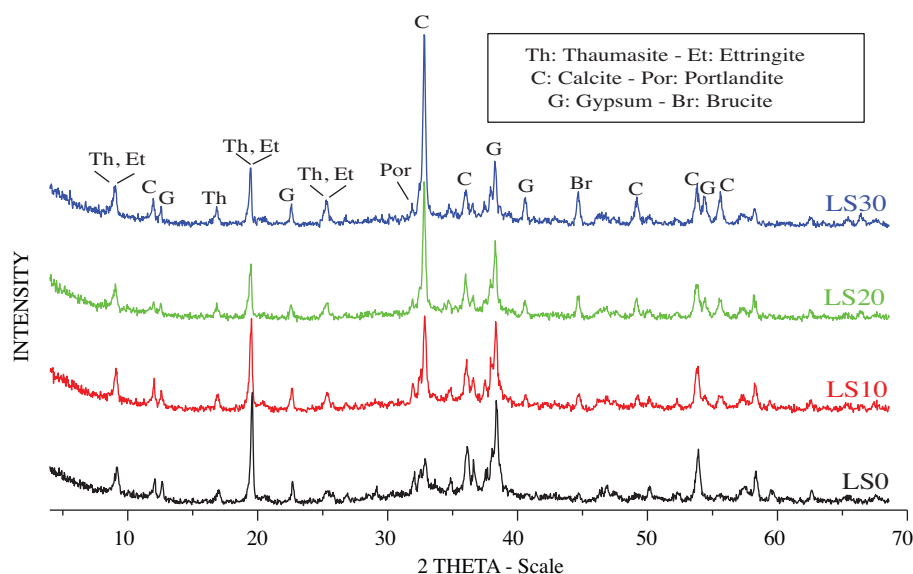
Furthermore, it was confirmed that the values of strength loss depended greatly on the replacement levels of limestone filler, especially at later exposure periods. These results imply the negative effect of limestone filler on the compressive strength of mortar specimens under sulfate attack orienting from magnesium sulfate solutions, and are in agreement with those reported elsewhere (Justnes et al., 2003; Pipilikaki et al., 2009).

Many researchers (Hartshorn et al., 2001; Torres et al., 2003; Tsvilis et al., 2003; Irassar et al., 2005) have reported that the sulfate deterioration in cement systems incorporating significant levels of limestone filler at ambient temperature as well as low temperature is associated with the formation of deterioration products such as thaumasite, ettringite, gypsum, M-S-H, and brucite. Torres et al. (2003) have reported that thaumasite was formed in cement mortars containing 5% limestone replacement, as well as in a higher replacement of limestone (15%-30%), especially when those mortars were exposed to a magnesium sulfate solution at a low temperature (5 °C).

### Microstructural analysis

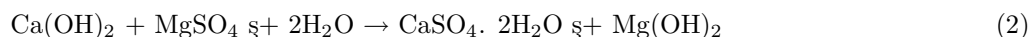
*X-ray diffraction* Measurements were carried out on samples from the deteriorated parts of the specimens. Figure 11 shows the XRD patterns for the powdered samples from the paste specimens exposed to magnesium sulfate solution at 5 °C. These patterns indicated the presence of ettringite (Et), thaumasite (Th), gypsum (G), portlandite (Por), calcite (C), and brucite (Br), which are the expected products formed by cement hydration and magnesium sulfate attack. The relatively strong calcite peaks detected in the LS10, LS20, and LS30 paste samples were due to the replacement of cement by limestone filler. The gypsum peaks were very strong, especially in the LS30 paste sample, compared to the peaks of other products formed.

There were very weak peaks for portlandite ( $\text{Ca}(\text{OH})_2$ ) in the XRD patterns of the LS20 and LS30 paste samples. Portlandite most probably had reacted with magnesium sulfate to form gypsum and brucite, both present in the degradation products.



**Figure 11.** XRD patterns of the damaged surface of the mortars with 0%, 10%, 20%, and 30% limestone preserved at 5 °C for 12 months.

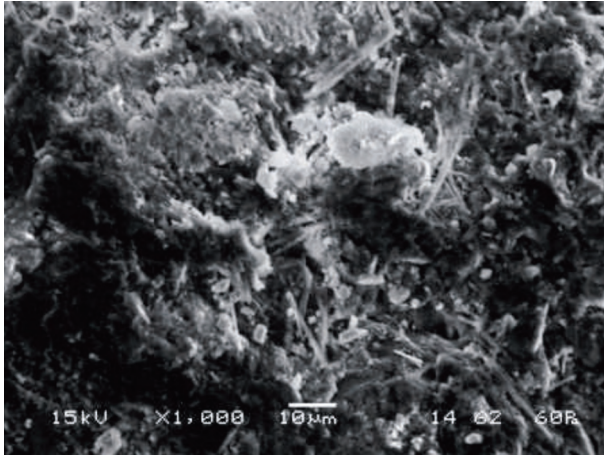
Furthermore, for the LS20 and LS30 paste samples, the ettringite and/or thaumasite peaks were very intense compared to those in the LS0 paste sample. Ettringite and thaumasite have similar crystal structures, and their XRD patterns show similarities at the 2 major peaks around  $9.1^\circ$  and  $16.0^\circ$   $2\theta$  (Soroka and Stern, 1976; Hartshorn et al., 1999). This suggests that, in the case of cement systems incorporating high levels of limestone filler under a magnesium sulfate environment, one of the primary causes for sulfate damage may be thaumasite formation, but not gypsum formation. Although thaumasite formation is frequently regarded as a low-temperature phenomenon, it can also be detected in paste specimens exposed at ambient conditions (Brown et al., 2002; Hartshorn et al., 2002). The coexistence of gypsum and brucite ( $\text{Mg}(\text{OH})_2$ ), in combination with the absence of portlandite, leads to the conclusion that portlandite, most probably, had reacted with magnesium sulfate to form gypsum and brucite, according to Eq. (2):



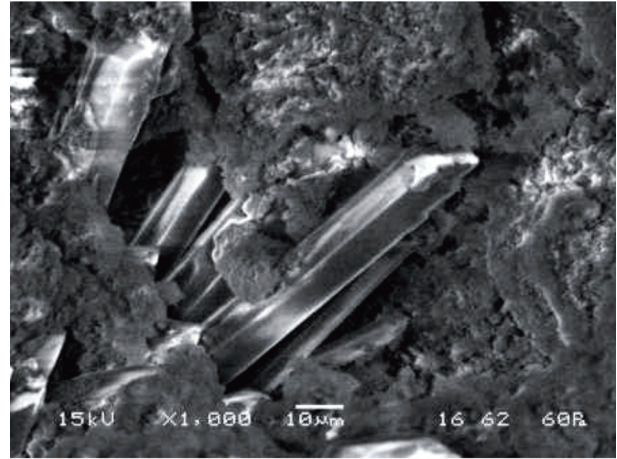
The very low solubility of brucite favors the consumption of calcium hydroxide. This leads to a pH reduction and, as a result, C-S-H becomes more susceptible to sulfate attack. Other researchers have also reported that portlandite is a reactant rather than a product of thaumasite formation (Hartshorn et al., 1999). To obtain further evidence to corroborate these observations from XRD, further investigation was made using SEM.

*SEM examination* The damaged mortar of LS30 exposed to  $\text{MgSO}_4$  solutions at 5 °C was selected as an example to observe the microstructure of deterioration products, and the SEM images are shown in Figure 12. No portlandite ( $\text{Ca}(\text{OH})_2$ ) crystal existed in the sulfate-attacked mortar, and many club-shaped or needle-like crystals were embedded irregularly in the pulpy material with a very open microstructure. Figure 12a shows a large number of needle-like thaumasite crystals, smaller than  $0.5 \mu\text{m}$  in diameter and  $3\text{-}4 \mu\text{m}$  in length, and the existence of different morphologies of the C-S-H gel in the specimen covering the mortar surface before sulfation. In the pores or surface cracks, there were many club-shaped gypsum crystals, which had much larger sizes than the thaumasite, as shown in Figure 12b. The formation of both thaumasite and gypsum led to the

cracking, spalling, and decomposition of the limestone filler cement mortar exposed to a sulfate environment. The combination of XRD and SEM led to the positive identification of the deterioration products' formation.



(a) Needle-like thaumasite crystals in the deteriorated mortar.



(b) Club-shaped gypsum crystals in the deteriorated mortar.

**Figure 12.** SEM photos of the deteriorated mortar (LS30) after exposure to 5% magnesium sulfate solutions at 5 °C for 360 days.

## Conclusion

Based on the experimental results obtained from this study, the following conclusions can be drawn:

- Cements with 10%, 20%, and 30% limestone needed more water in the mortar mixture and cement paste than the reference mix. However, the increase in water was not so high. The reason for the change in water demand could be explained by the difference of the chemical composition and fineness between the limestone and the Portland cement used.
- The compressive strength of mortar decreases with an increase in crushed limestone content.
- From the visual observations, it was clear that mortars immersed in a solution of  $MgSO_4$  at 5 °C for 360 days, with 10%, 20%, and 30% limestone addition, had severe surface damage; inspections indicated that the samples were suffering severely from magnesium sulfate attack.
- The deterioration of Portland limestone cement mortar is higher than that of Portland cement mortar. The higher the content of limestone, the more serious the deterioration of concrete, and, also, the lower the temperature, the earlier the deterioration of mortar.
- Thaumasite formation is accompanied by the formation of brucite and secondary gypsum, while calcium hydroxide is a reactant, rather than a product of the sulfate attack reaction.

In light of the present findings and today's knowledge, the Portland limestone cement is allowed according to the Algerian standard NA 442 2003 and the new European standard, with up to 6%-20% (CEM II/A-L) and 21%-35% (CEM II/B-L) limestone, respectively; its use should be restricted to environments in which magnesium sulfate is not present.

## Acknowledgments

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