

# Incompatibility stone-mortar Influence of pH on the sensitivity of rocks to saline alterations

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**Abstract**—This paper discusses incompatibilities between stones and mortars. The aim is to understand the evolution of rocks of acid nature, sandstones in this case, in the presence of mortars of an alkaline nature, such as cement mortars used for jointing or coatings. Accelerated aging tests combining alkaline alteration and salt alteration were simultaneously carried out in order to study the synergy between these two processes of degradation. The degradation process is follow-up by observing the induced alteration patterns, the identification of the newly appeared mineral phases and the evolution of the ultrasonic propagation speed of the materials tested. The obtained results allow showing that the pH-salt crystallization synergy is particularly aggressive towards the studied materials and that the sustainability of the rocks subjected to circulations of basic solutions due to the proximity of mortars of alkaline nature is seriously reduced.

**Keywords**— durability, incompatibilities, mortar, pH, stone, salts.

## I. INTRODUCTION

The aim of this work is to study the behavior of monument stones through an approach based on accelerated cycles of aging in a research theme aiming to study the influence of the chemical nature of mortars on the durability of implemented stones. The juxtaposition in a masonry of materials with different mechanical and chemical qualities can be an additional source of disorders and degradations. The problem of compatibility between waxes stones and mortars is essential [1]. It is indeed necessary to define the behavior of the "stone-mortar" assembly with regard to the factors of alteration in order to improve the durability of the structures. Among the agents of alteration, the soluble salts, which pass through the masonry by means of capillary ascents or rainwater, play a preponderant role in the degradation of the stones used. In fact, accelerated aging tests were carried out to analyze the behavior of materials in the laboratory. The aim of this study is to subject rock samples to the simultaneous action of salts and alkaline solution to see to what extent the alkaline mortars can weaken the stones with which they are associated. The

building stones studied are red vosgian sandstone, beige vosgian sandstone, coal sandstone and molière sandstone.

## II. MATERIALS STUDIES

The studied sandstones, of varied provenances, are used in many historical monuments and in particular in the construction of the famous Strasbourg Cathedral with respect to the vosgian sandstones in particular. Coal sandstone comes from the coal basin of the Cevennes. It constitutes the bulk of the waste rock from the coalfields dating from the Stephanian. The molière sandstone comes from the Permian lands of the Montredon-Labessonnié region in the Tarn. The Beige and Red Vosgian sandstones come from the Formation of the "Sandstone at lower Voltzia" of the upper Buntsandstein of the Triassic of the Vosges.

The characterization of the studied materials (Fig. 1) was determined by various analytical techniques including optical microscopy (Figs. 2, 3, 4, and 5) for the petrographic aspect. The characterization of the porous space was done by mercury porosimetry.



Fig. 1: Macroscopic aspect of sandstone materials, reflecting the differences in mineralogical composition and matrix that gives the rocks their specific color, 1- molière sandstone, 2-coal sandstone, 3- beige vosgian sandstone, 4- red vosgian sandstone.

The apparent volumetric mass, the compressive strength and the propagation velocity of the ultrasound defined the mechanical cohesion of the materials. The characteristics of the studied stones are grouped in Table 1.

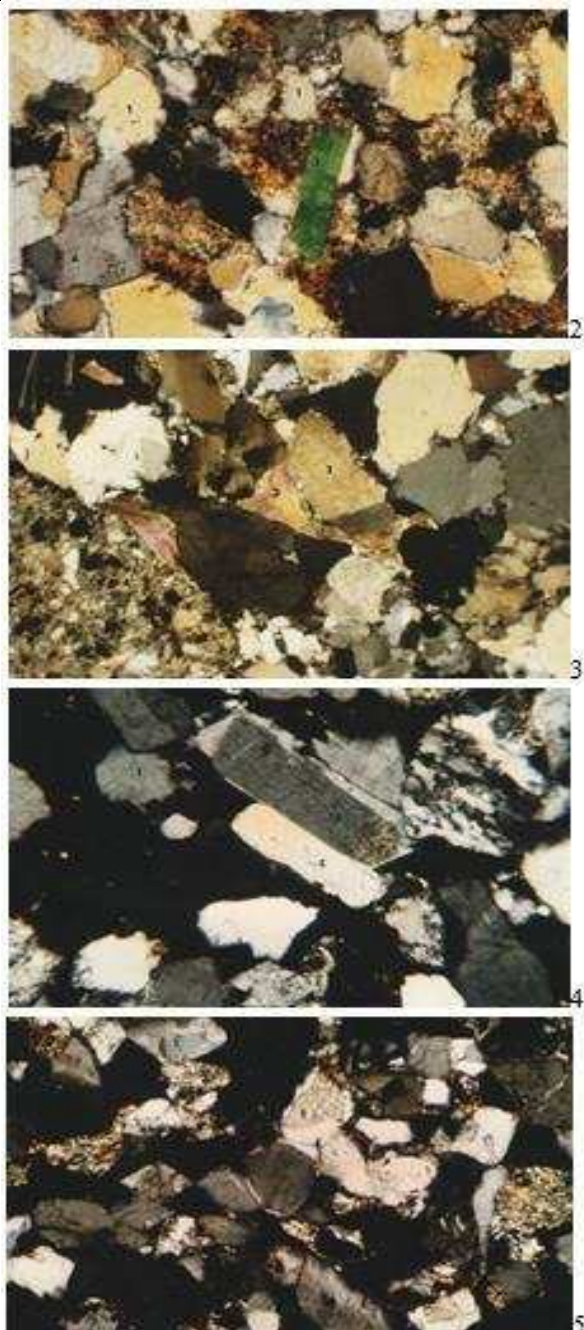


Fig. 2-3-4-5: Optical microscopy of sandstones. The molière sandstone (1) presents aggregates of microcrystalline quartz, detrital quartz, and muscovite flakes bathed in a clay-ferruginous matrix ; Coal sandstone (2) shows fragments of detrital and polycrystalline quartz, muscovite flakes and a clay matrix ; Beige vosgian sandstone (3) shows fragments of detrital quartz, quartz fed by secondary silica, fragments of metamorphic rock and muscovite ; The red vosgian sandstone (4) has elements of secondary feeding detrital quartz, aggregates of micrograiny quartz and a hematite rich matrix.

Table.1: Main characteristics of studied sandstone

	coal sandstone	molière sandstone	beige vosgian sandstone	red vosgian sandstone
Mercury porosity	13.6 %	13.9 %	3.6 %	6.5 %
Apparent volumetric mass ( g/cm <sup>3</sup> )	2,3	2,22	1,82	1,99
Mechanical resistance ( MPa)	⊥ au lit : 60	⊥ au lit : 60	⊥ au lit : 24	⊥ au lit : 37
Ultrasonic propagation speed (m/s)	2760	2870	1840	1890

### III. EXPERIMENTAL PROCEDURE

The experimental protocol adopted is as follows: sandstone samples are subdivided into two batches. The first batch is first subjected to an alkaline bath for 28 days at 60 °C in a saturated solution based on calcium hydroxide imparting a pH of 12.6 similar to that of the solutions passing through the alkaline mortars .

In a second step, samples of control rock and those of the batch treated at pH 12.6 are then subjected to an accelerated aging test by salt alteration according to the experimental protocol recommended by RILEM [1]. The salt used is sodium sulfate. The immersion solution is prepared from 14% sodium sulfate decahydrate with a concentration of 14 g of salt for 86 g of water. Samples are subjected to repeated cycles of consecutive imbibition and drying. Samples are immersed in the saline solution for 2 hours at room temperature. The second phase of the cycle consists of desiccation for at least 16 hours under conditions of high relative humidity at the beginning of drying. This phase makes it possible to reach the saturation of the solution and then the crystallization of the salts.

### IV. RESULTS

The observation of thin plates of rocks of the treated batch at pH 12.6 shows the development of an important microcracking of the quartz grains constituting the sandstones. The microcracking revealed is generalized to all the materials studied (Figs. 6-7-8-9-10-11-12-13). Such disturbances have already been noted [2] in sandstones, granites and mortars, and attributed to the synergy of several processes: thermal effects (variations in temperature), hydric (successions of humidification-drying cycles, chemicals (dissolutions) and mechanical (saline crystallizations).





Fig.6-7: Observation of thin blade of the coal sandstone before (6) and after (7) treatment with pH 12.6



Fig. 8-9: Observation of thin blade of the red vosgian sandstone before (8) and after (9) treatment with pH 12.6

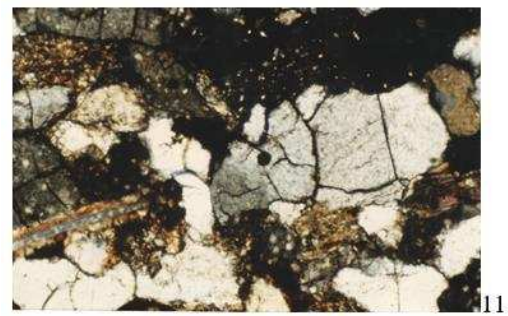
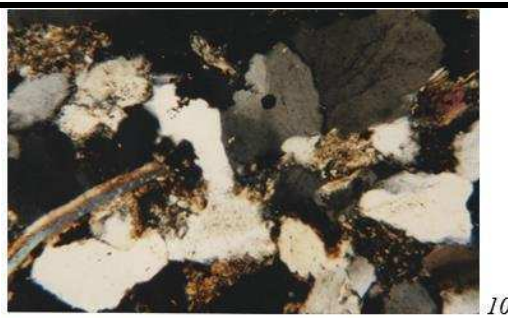


Fig. 10-11: Observation of thin blade of the molière sandstone before (10) and after (11) treatment with pH 12.6

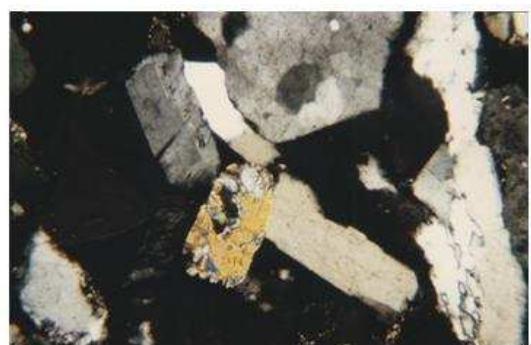


Fig. 12-13: Observation of thin blade of the beige vosgian sandstone before (12) and after (13) treatment with pH 12.6

Following the accelerated aging test by the salts, the two batches of samples studied show, over the course of the cycles, a high granular disintegration leading to the complete degradation of the test tubes (Fig. 14). The results obtained confirm the abundant data in the literature concerning the problem of the saline alteration of the stones used in the historical monuments [3], [4], [5], [6].

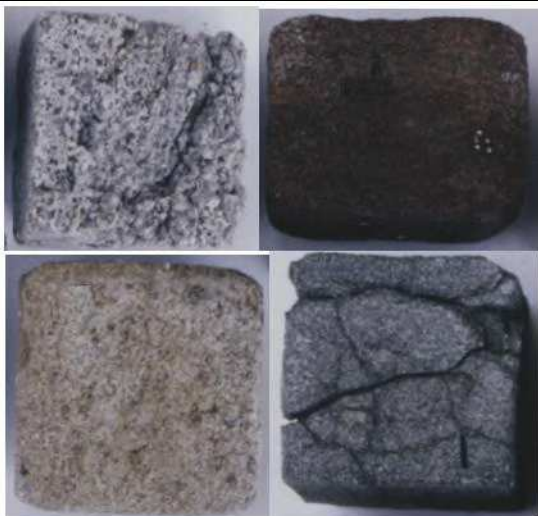


Fig.14: Illustration of facies of alteration of sandstone under the effect of saline crystallizations.

The alteration process is monitored by measurements of the propagation speed of the ultrasound. This measure, which has the advantage of being non-destructive, reflects the degree of cohesion and compactness of the materials [7]. Regardless of the studied material, ultrasonic propagation velocity measurements could not be carried out during last cycles, whereas the rock maintained a state of cohesion allowing the test to continue. The attenuation of the waves at this stage is such that no readable signal can cross these materials. The degradation of the specimens following saline crystallization is accompanied by the progressive fall in the propagation speed of the sound.

The results are plotted in the figures (15-16-17-18) where  $\Delta V(\%)$  represents the variation of the ultrasonic velocity as a function of the saline alteration cycles.

$$\Delta V (\%) = 100 (V_c - V_x) / V_c \text{ with:}$$

$V_c$  = ultrasonic propagation velocity measured through the samples of the control batch;

$V_x$  = ultrasonic propagation velocity measured through the materials after x cycles.

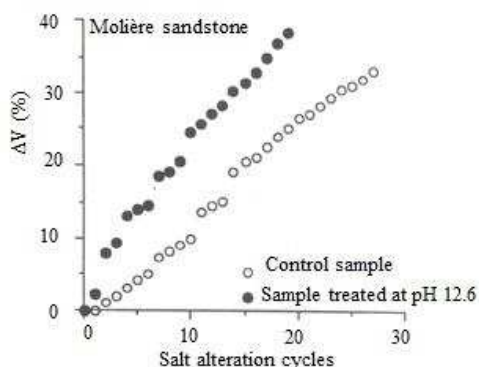


Fig.15: Variation according to salt alteration cycles of rate of loss of ultrasonic speed of the control and treated samples at pH 12.6 for 28 days at 60°C.

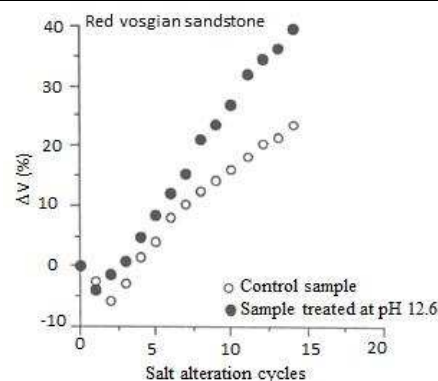


Fig.16: Variation according to salt alteration cycles of rate of loss of ultrasonic speed of the control and treated samples at pH 12.6 for 28 days at 60°C.

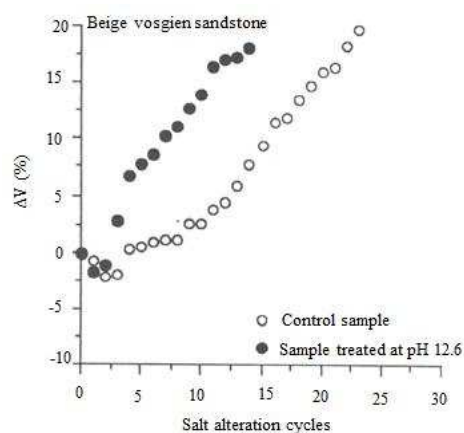


Fig. 17: Variation according to salt alteration cycles of rate of loss of ultrasonic speed of the control and treated samples at pH 12.6 for 28 days at 60°C.

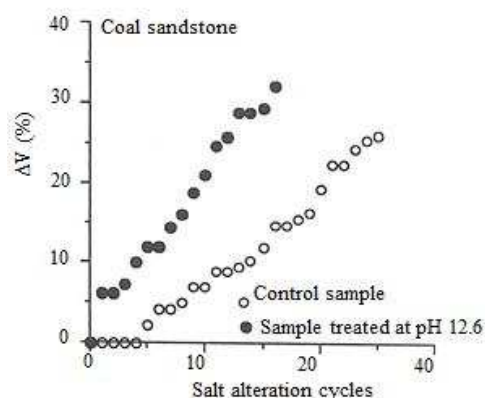


Fig.18: Variation according to salt alteration cycles of rate of loss of ultrasonic speed of the control and treated samples at pH 12.6 for 28 days at 60°C.

It should be noted that the batch treated with the basic pH shows a reduction in the propagation speed of the sound that is greater than the control samples, for all classes of rocks combined. These results can be attributed to the microcrack network revealed in quartz crystals following the alkaline attack. The play of the dilation and the



growth of salts in this microporosity (fig.19) further weakens the rocks by accentuating the observed disorders.

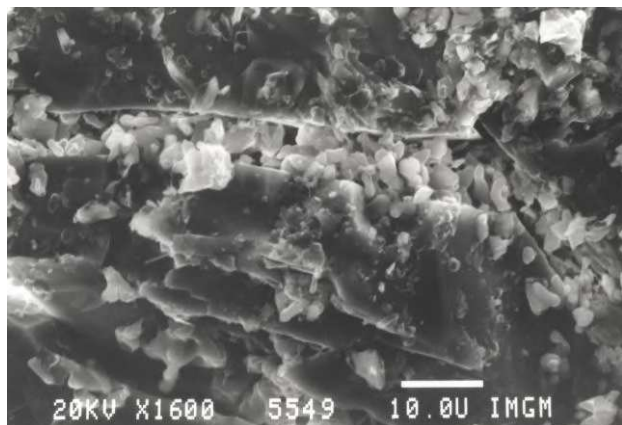


Fig. 19: SEM observation of crystallizations of mirabilite ( $Na_2SO_4 \cdot 10 H_2O$ ) in the cracks of quartz crystals previously induced

To these mechanical factors are added particularly interesting chemical mechanisms to the quartz: certain salts (NaCl, Na<sub>2</sub>SO<sub>4</sub> ...) are able to generate chemical alteration of the quartz grains by sequences of dissolution-precipitation [8], as well as the presence of salts leads to an increase in the dissolution rate of silica [9].

It also appears that the mineralogical nature of saline crystallizations varies according to the nature of the batch of samples. If, in the batch of control samples, the crystallization of the mirabilite is observed, the batch treated at pH 12.6 also shows the appearance of ettringite (FIG. 20), calcium sulfate and hydrated aluminum, of formula:  $Ca_6Al_2(SO_4)_3(OH)_{12} \cdot 26H_2O$ . The calcium introduced during the alkaline treatment reacts in the presence of the sodium sulphate of the saline immersion solution to give ettringite, known salts for its swelling properties [10], [11].

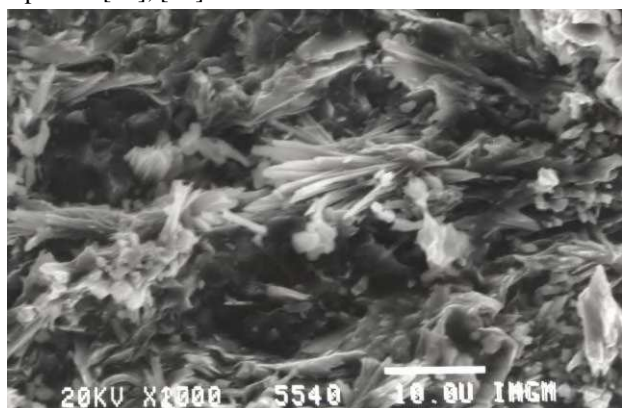


Fig.20: Observation in the SEM of crystallizations of ettringite ( $Ca_6Al_2(SO_4)_3(OH)_{12} \cdot 26H_2O$ ) in the samples treated simultaneously in pH 12.6 then in the cycles of ageing by salt alteration.

The simultaneous presence of these two salts further accelerates degradation processes and reduces the number of cycles tolerated by the materials (Table 2).

Table.2: Number of saline alteration cycles performed as a function of the initial state of the materials

	Number of cycles	
	Samples treated at pH 12.6	control samples
Coal sandstone	24	35
Molière sandstone	24	34
Beige vosgian sandstone	22	30
Red vosgian sandstone	15	19

## V. CONCLUSION

In order to follow the evolution of sandstone rocks in the presence of a basic mortar, simulations of alkaline attack were carried out in order to simulate the potential role of the portlandite of these mortars. These tests made it possible to identify the nature of the reaction mechanisms involved, namely the alkali-silica reactions. These reactions lead to a profound disorganization of the microstructure of the sandstone rocks, in particular by the embrittlement of the major constituent elements of the sandstones, namely crystals of quartz. The pre-existing defects in these minerals are then revealed and accentuated following dissolution reactions. The consequences of the disorders due to the proximity of basic media also have repercussions on the resistance of the sandstones to the conventional alteration agents such as salt crystallizations. Indeed, the synergy pH-crystallizations saline was particularly harmful in the case of the sandstones studied. By creating an important porosity by revelation and accentuation of microcracks within the quartz, the basic solutions multiply the risks of alteration by alternating dissolutions and saline crystallizations. The durability of these materials is considerably reduced. The enrichment of the environment with calcium ions constitutes an additional degradation factor: in combination with the elements dissolved in rock and sulfur, calcium promotes the crystallization of ettringite known for its swelling properties.

Thus, the proximity of mortar joints in a structure can, by modifying the chemical equilibria, cause a weakening of the intrinsic structure of the sandstones and thereby their durability in a global manner.

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