



Effect of mixed vs single brine composition on salt weathering in porous carbonate building stones for different environmental conditions



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ABSTRACT

The influence of different factors, brine composition, environmental conditions and pore structure, on salt weathering of porous carbonate building stones is investigated. The main goal of this study is to highlight the differences in weathering between single salts and a mixture of different salts. Two kinds of weathering mechanism are studied, the crystallization pressure and the deliquescence/recrystallization process. To achieve this purpose wetting and drying cycles and cycles varying relative humidity on samples containing salts were performed. The effect of total salt content with different salts and mixtures is simulated by weathering tests with solution of different compositions (NaCl, 6% in weight, Na₂SO₄ · 10H₂O, 6% in weight, CaSO₄ at saturation, and a mixture of them at total concentration of 6.2%). Weathering tests were performed at room temperature and 50 °C and under low and high relative humidity.

The results show that the final weathering depends on a complex combination of three parameters: (i) environmental conditions, (ii) salt composition and (iii) petrophysical properties of stones, and general conclusions on the impact of each parameter can be drawn. An important result is that salt mixtures have a lesser impact on weathering during crystallization cycles: single salt brines, especially sodium sulphate, produce more weathering than complex brine compositions during wetting and drying cycles. Conversely during variations on RH, samples with salt mixtures suffer more damage than samples weathered with a brine of single salt composition.

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1. Introduction

Salt crystallization is one of the most dangerous weathering agents in monumental stones. Salts are always present in any environment, natural or anthropogenic and under any climatic conditions. The composition of these salt is very variable (Goudie and Viles, 1997) and depends on many different factors such as proximity to sea; actual and historical air, soil and water pollution; mineralogical and chemical composition of affected materials, i.e. not only the studied material but also other materials such as mortars, cements, and metals in contact with it. The behaviour of salts depends very much on the environmental conditions, mainly temperature and relative humidity (RH) that control the stability of phases present in the stone. For example, natural cycles of RH variations can cross the equilibrium curve between thenardite (anhydrous sodium sulphate) and mirabilite (sodium sulphate decahydrate) which is probably one of the most important causes of damage in natural rocks or in building materials (Steiger and Asmussen, 2008). Other salts such as gypsum can dehydrate to less hydrate or anhydrous salts but this reaction occurs on a geological

time scale and therefore, phase changes of this species do not contribute to building stone weathering. Nevertheless, the importance of gypsum as a deterioration factor over time is recognized (Charola et al., 2007).

Material properties are also important, not all stones react in the same way under the action of one weathering agent. Benavente et al. (2007) summarized the factors controlling salt weathering in porous materials as pore size and porosity, nature of the salt, transport properties of the solution in pores and strength and the material's resistance to crystallization pressure.

Laboratory studies in salt crystallization impact on construction materials began more than a century ago. In 1824 the “Annales des Mines” (Anonymous, 1924) published a note about the work done by M. Brard to determine if construction materials are vulnerable to frost action. To simulate the effect of water solidification, stones, bricks and other materials were saturated with different salt solutions and it was found that the Glauber salt (sodium sulphate) produced the “best” results. Research concerning monumental stone durability began in the 1920s and 1930s to identify the role played by soluble salts on the degradation of building materials (West, 2000).

The most studied salt is sodium sulphate even if other salts have been tested as well. An extended bibliographic review of crystallization tests with sodium sulphate can be found in Steiger and Asmussen (2008). Other salts used in laboratory tests are sodium chloride

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(Rodríguez-Navarro and Doehne, 1999; Desarnaud et al., 2014), magnesium sulphate (Espinosa-Marzal and Scherer, 2013; Ruiz-Agudo, 2007), gypsum (Menéndez and David, 2013), etc. In nature and monuments a combination of different salts is usually present.

The weathering caused by salt crystallization in nature is a complex process also involving the action of different aqueous salt mixtures together with frost. McGreevy (1982) performed frost cycles in samples saturated by saline solutions of sodium chloride, sodium sulphate and magnesium sulphate and found that freezing of NaCl solutions is more destructive than freezing of sodium sulphate solutions.

Only recently has the behaviour of salt mixtures been investigated in laboratory. Sawdy and Heritage (2007) studied crystallization of binary salt mixtures in non-porous media. The authors combined nitromagnesite ($\text{Mg}(\text{NO}_3)_2 \cdot \text{H}_2\text{O}$) and halite (NaCl) with nitratite (NaNO_3), niter (KNO_3) and gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$). The obtained salt mixtures were exposed to different relative humidity, below and above their equilibrium RH at room temperature, and the mass variation was registered. After 2 h under RH above the equilibrium, all samples had undergone some degree of deliquescence but none were completely dissolved. They concluded that in comparison to single salts deliquescence/dissolution rate increases, deliquescence began when the RH was above the lowest individual equilibrium RH of salts in the mixture and the evaporation rate decreases.

López-Acevedo et al. (1997) investigated the salt weathering of two calcareous stones with different salts and salt mixture compositions: sodium, potassium and magnesium sulphates. Their experiment consisted in a continuous capillarity rise of salt brines without any control of environmental conditions. They observed differences in the weathering patterns as a function of the employed salt with efflorescences for sodium and magnesium sulphates and subflorescences and crusts for potassium sulphate.

Cardell et al. (2008) carried out capillary rise crystallization tests with different sulphate solutions (Ca, Mg, Na, K) and with binary mixtures of them in a limestone. They observed differences in the weathering pattern depending on the salt composition. Calcium sulphate did not produce any apparent weathering, magnesium sulphate caused a hard crust on the lower part of the wet stone, potassium sulphate created a hard film on the surface of the sample and sodium sulphate caused granular disintegration and efflorescences. Mixed solutions only produced a thin crust on the surface. They found that single salt solutions cause more damage than do mixed brines.

De Clercq et al. (2013) performed crystallization experiments in two limestones with two different brine compositions, a first one composed of sodium sulphate and nitrate and a second one composed of sodium and potassium sulphate. The RH conditions were sustained at 50% and 85%. They found different mineralogical compositions of efflorescences depending not only on the nature of the solution but also on the type of stone, mainly on their porosity and evaporation rate. They concluded that crystallization behaviour of mixed salt solutions in porous materials is more complicated than in bulk solutions.

Lindstrom et al. (2015) studied the behaviour of salt mixtures of NaNO_3 and Na_2SO_4 in sandstones with comparison to simple salt behaviours. Samples previously saturated with salts were exposed to wetting and drying cycles with pure water. Among many other results they found that NaNO_3 is the least damaging salt and Na_2SO_4 the most damaging one. Tests with salt mixture were not in accord with previously obtained results in solution droplets. The authors suggested that more research should be done.

Investigation of the behaviour of salt mixtures equilibrium models provides an extremely useful tool for predicting crystallization behaviour as a function of brine composition and environmental conditions. The results can contribute to define the safe ranges of climatic conditions for the optimal conservation of objects (Steiger and Heritage, 2012).

To summarize it has been recognized that salt crystallization weathering on stone with salt mixtures is different from weathering

when only one salt is concerned. Cardell et al. (2008) reported that salt mixture produces less damage than single salts. To test the effect of salt mixture versus single salt on salt crystallization weathering a mixture of three salts: NaCl, Na_2SO_4 and CaSO_4 were used to perform accelerated ageing tests. The obtained results were compared to those obtained for the same kind of experiments but performed with a single salt. Other parameters considered in this paper are the temperature and relative humidity. Salt cycles were performed under two different RH conditions (high and low) and two different temperatures (20 °C and 50 °C). Low relative humidity values are different for the two temperatures due to experimental limits, 45% at room temperature corresponding to the room RH and 5% at 50 °C, relative humidity in the oven.

The two principal mechanisms of salt weathering are crystallization pressure and phase transitions. Crystallization pressure plays an important role during brine evaporation and precipitation of salts. To test this mechanism wetting and drying cycles were performed under environmental conditions given above. To test the effect of salt phase transitions RH variation cycles were performed at 50 °C and room temperature. The brine concentrations for all the tests were always around 6%, except for calcium sulphate (0.2%). The obtained results allow a better understanding of salt weathering phenomena under real conditions where salt mixtures are present.

2. Materials

Three different calcareous stones used in the construction of monuments in the NW of France, but also in other regions and countries were tested in this study. The selected stones are Lutetian limestone (“roche fine” variety), Vernon chalk and Caen limestone (“demi-ferme” variety). These stones were chosen due to their similar mineralogical composition (calcite) with different microstructure and pore size distribution.

Lutetian limestone (Fig. 1a) used in this study is a middle Lutetian limestone from the Saint Maximin quarry (Rocamat company) known as “Roche Fine”. The Lutetian formation has different stratigraphic levels with very different properties (Angeli et al., 2007; Fronteau et al., 2010; Vazquez et al., 2015). The selected variety is a homogenous yellowish limestone without macroscopic fossils and with brownish spots (ferruginous oxides). It is a detritic limestone formed by accumulation of calcite grains and foraminifers (Moreau, 2008). Macroscopically, numerous small pores can be observed (Noel, 1970).

Vernon stone (Fig. 1b) is a hard white Santonian chalk, probably equivalent to the Newhaven Formation in Great Britain. The selected samples correspond to the “Gros lien” level and come from the Vernon quarry (TERH company) in Vernon. It is a white stone with small black dots (manganese oxides) and flint nodules. The flint nodules can reach several tens of centimetres but in the studied samples their maximum size was only few centimetres. This stone underwent a complex diagenetic history through a dolomitization and dedolomitization process (Blanc, 1990).

Caen stone (Fig. 1c) samples come from the Cintheaux quarry (Societe des Carrieres de la Plaine de Caen) and correspond to the “demi-ferme” variety. It is a middle Bathonian light yellow limestone composed of lime sand (bioclasts and pellets) with calcite cement (Blows et al., 2003). Small pyrite crystals are locally present, producing an oxidation halo of rusty staining (Palmer, 2008).

Mineralogical analysis obtained by X-ray diffraction showed that the main mineral is calcite for the all three rocks with some quartz (~10%) in the Lutetian limestone.

Porosity of the studied rocks is given in Table 1, with mean, maximum and minimum values as well as standard deviation for 16 samples. It can be seen that Lutetian limestone has the highest porosity which is quite uniform for all the samples. Vernon Chalk has an average porosity of 25% but porosity varies very much from one sample to another. This variability can be explained by the presence of flint nodules. Caen limestone has the lower porosity with an intermediate variability compared to the other investigated rocks.

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