Geochemical constraints on the alteration processes of building materials

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Water is the principal vector for dammages of limestone monuments and buildings by its action at two levels: a physical level because it induces gelifraction and a chemical level through material dissolution.

Many studies on the durability of materials address the problem of substrate protection from a physical point of view: on one hand because they are based on visual and/or physical observations and, on the other hand, because the accelerated ageing tests look at the physical degradation factors linked to volume variations, such as salt cristallizations and/or frost-thaw cycles.

The present study addresses the problem from a dynamic chemical point of view, i.e. by following transfers of elements between the substrate (limestone) and the external medium. In order to get a more thorough and quantitative view on small time scales and better constraints on the processes, geochemical tracers (major elements, trace elements and strontium isotopes) have been used in various leaching experiments as they are characteristic of the substrate - be it treated or not by a mineralizing product used as waterproofing agent.

Sampling and analysis

The whole preparation is done in a class 100 clean lab: samples are cleaned with subboiled RP ethanol in ultrasonic waves equipment. Then, they are dried in an oven at 40°C.

The protective treatment by the waterproofing agent is always applied after cleaning. In our study, it is conducted by complete immersion of the sample in the product. After two successive immersions in the product, samples are again dried at 40°C.

Up to 7-days long leaching experiments were performed with Milli-Q water. The 6 sides of the samples were in contact with water. Leach aliquots were taken after 1, 3 and 7 days for analyses. Solutions were filtered on 0.45 µm f cellulose acetate filter and stored at 4°C until analysis. Major elements measurements were conducted by capillary electrophoresis, trace elements measurements by ICP-MS and Sr isotopic compositions by mass spectrometry after chemical purification.

As for the artificially ageing program, priority was given to nature-like simulation tests: we have reproduced in a climatic room the natural temperature and relative humidity variations only with a faster frequency than in our natural regional context.

Results and discussion

The non-treated sample leaches show significant alkali-earth but low alkali contents which represent the limestone composition of the substrate. On the other hand, treated sample leaches present important alkali and low alkali-earth contents, which correspond to the product composition, and indicate the 'barrier-like' efficiency of the treatment. Exchanges between the material and its environment are reduced and can be quantified by an apparent minimum retention factor of 7. Moreover, the comparison of these concentrations with those obtained on artificially aged samples emphasizes the alteration of the substrate.

Sr isotopic compositions in non-treated sample leaches are very homogeneous (0.70855 to 0.70869 - average 0.70863) and represent the substrate endmember. In treated sample leaches, Sr isotopic compositions are more radiogenic because of the silicate characteristics of the product (0.71101). The decrease of these values with the leaching time shows an increasing participation of Sr coming from the substrate. We have calculated the Sr proportions coming from the two endmembers (Fig. 1) and therefore corrected the apparent retention factor of 7 in order to get a real retention factor of 10 (for the Sr coming from the substrate) after 7 days of leaching.

Data for treated samples have been plotted in a two-component mixture diagram (Fig. 2): the points of non-artificially-aged samples are situated on a line between the 'substrate' and 'product' endmembers.

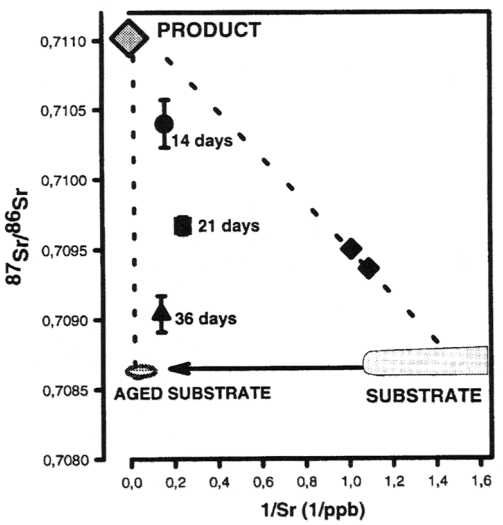

FIG. 1. [Sr] and Sr isotopic composition evolution in leaches during leaching time in treated samples. Origin of leached Sr during leaching time.

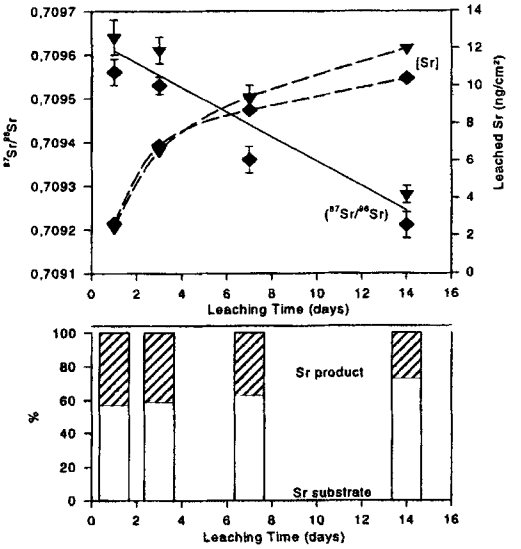

FIG. 2. Two-component mixture diagram. Artificially aged treated samples (7 days of leaching).

Note that the substrate endmember also shifts with time of ageing.

Conclusion

Combined data of element contents and Sr isotopes in leaches show the barrier efficiency of the product and lead to a calculated real retention coefficient of 10. Comparison of data with artificially-aged substrate show the persistence of the waterproofing efficacy of the product with time.

The use of original methods - trace element geochemistry and Sr isotopes- thus allows to quantify dissolution processes in conditions close to natural ones.