3D characterisation of the micro-geometry changes of a single fracture in the course of dissolution

Catherine NOIRIEL1)2), Philippe GOUZE1), Benoît MADE2), Céline BRUDERER1), Richard LEPROVOST1) and Thierry POIDRAS1)

1) UMR 5568, Université de Montpellier 2, place E. Bataillon, 34095 Montpellier cedex 5, France
2) CIG, Ecole des Mines de Paris, 35, rue Saint Honoré, 77305 Fontainebleau cedex 5, France

Abstract: Carbonate dissolution of tensile fractures during percolation by CO2-enriched fluid is studied using synchrotron X-Ray Computed Micro Tomography as a tool to quantify the changes of the fracture walls geometry and extend. The experimental dissolution of a pure-calcite sample confirms the process of homogeneous chemical “erosion” classically implemented in the numerical simulators, but with a reactive surface coefficient much larger than unity. In opposition, the dissolution of a non-pure calcite sample, representative of the average micritic carbonate composition, displays very heterogeneous dissolution patterns associated with an increase of the fractal dimension. The initial topographic surfaces of the fracture walls evolve rapidly toward “non-topographic” interfaces displaying multiple overhangs due the preferential dissolution of the carbonate grains. In this case the classical 2D-profilometric methods fail. The specific surface coefficient increases strongly, more than 5 times the planar surface, and probably faster than the reactive surface

INTRODUCTION

Limestone aquifers constitute a major reserve of fresh water throughout the world. The fractures, which are the principal path for water flow and potential pollution, can be strongly modified by geochemical transfers because of the high reactivity of the carbonates in continental environments. Experimental, modelling and theoretical studies of single fracture flow and transport put forward the strong control of the fracture aperture, surface roughness and aperture tortuosity (due either to twisting of the median plan or to the distribution of the contact points) on the flow and dispersion (Adler and Thovert, 1999). Although theoretical basis exists (Lasaga, 1998), it is only recently that quantitative modelling and experimental measurements of the fracture aperture spatial changes bound by chemical reactions were realized (Dijk and Berkowitz, 1998), producing important constraints for modelling flow and dissolution patterns. However, large uncertainties still remain on the relation between the macroscopic evolution of the aperture and the microscopic (grain scale) morphological evolution. Furthermore, assuming that (i) flow and transport can be modelled using aperture and roughness as input parameters and (ii) thermodynamic and kinetic coefficients of reactions are known, the reaction-transport-coupled modelling of the aperture change requires the knowledge of the spatial and temporal evolution of the reactive surfaces. This parameter acts as a multiplicative coefficient of the rock-fluid transfers so that it is a key parameter, yet difficult to measure

The aim of this presentation is to report the results of reactive percolation experiments in a single fracture for which X-Ray computed tomography is used to determine the structural changes in fracture morphology in course of dissolution.

EXPERIMENTAL SETUP AND DATA ACQUISITION

The experiment focusses on the percolation of acidic fluid in tensile rough fissures sub-collinear to the core axis. The cores are 12mm long and 6 mm diameter. Dissolution is obtained by flowing, at a constant rate of 2.8 10^-9 m^3.s^-1, pure water enriched with carbon dioxide at partial pressure of 0.1 MPa (pCO2 = 4 ±0.1). Output pH is 5.1 ± 0.15. As the mini-cores were rigidified with epoxy, the effect of confining pressure (equal to the max pressure at the inlet) variation was negligible. The pressure at the outlet was maintained above 0.1 MPa in order to avoid CO2 degassing in the mini-core.

At the initial state and for different stages of dissolution, we record the sample structure using non invasive X-ray computed micro-tomography CMT. This technique is based on the X-ray attenuation measurements of thousand radiographies taken at different angles of the sample. We used the ID19 beam of European Synchrotron Radiation Facility which allows us to observe digital images of the evolution of the geometrical characteristics of the sample over four scales with a resolution of 4.91µm. Then, fracture walls are extracted from 2048 X 2048 x 4096 tri-dimensional images.

RESULTS AND DISCUSSION

Two samples of micritic limestone of same origin are studied. The first one is a pure calcite (ϕ = 2.2 %) with a fracture of aperture a = 50 ± 8 µm and the second is composed of 85 w% calcite, 10 w% dolomite, 5 w% clay (ϕ = 5.6 %) with a fracture of 20 ± 7 µm. For both samples, the initial fracture surfaces are topographic surfaces (TS), that is, to any x-y coordinate corresponds a single elevation z. In this case fracture walls can be analyzed by extracting the x-y projection of the CMT images, producing profilometry-equivalent data set to be characterized by classical statistical and spectral surface analysis. Results display ubiquitous self-affine behaviour all over the measured scale range, from 5µm to 1cm. The fractal dimension D, computed coherently using 1D and 2D (Fourier Transform) power spectrum method is 2.5 ± 0.1 without noticeable anisotropy for both the samples. At the initial stage, the reactive surface coefficient SR is estimated as the product of the volume fraction of the mineral involved in the calcite reaction (1 for sample 1 and .85 for sample...
2), by the specific surface coefficient \( S_S \). The later is the ratio of
the total surface of the fracture walls to its planar projection, so
that \( S_S = 1 \) for a planar surface. It is assumed here that the fracture
intersects the rock randomly. Usually, for numerical modelling,
\( S_S \) is kept constant and \( S_S \) is embedded in an “efficient” kinetic
reaction rate (Lasaga, 1998) which doesn’t take into account \( S_S \)
and \( S_S \) changes during dissolution. The total surface of each of
the fracture walls is computed from the CMT images by:

\[
S_S = \iiint_{x,y} \left( \frac{\partial z}{\partial x} + 1 \right)^2 + \left( \frac{\partial z}{\partial y} + 1 \right)^2 \, dxdy
\]

were \( z(x,y) \) are the fracture wall elevations.

The dissolution of the 100% carbonate sample is characterised by
a uniform translation of the fractures walls surfaces along the \( z \)
axis. As a consequence, the fractal dimension \( D \), the specific
surface and reactive surface coefficients (figure 3) are all
invariant. The fracture walls remain TS.

The dissolution of the 100% carbonate sample is characterised by
a uniform translation of the fractures walls surfaces along the \( z \)
axis. As a consequence, the fractal dimension \( D \), the specific
surface and reactive surface coefficients (figure 3) are all
invariant. The fracture walls remain TS.

CONCLUSIONS

- The remarkable performances of the synchrotronic X-ray CMT
(compared to diverging source X-ray methods and NMRI) has
proved to be an efficient toolsto quantify the fracture surface
changes bounded by dissolution, with a resolution close to the
grain size and over a large field of view (20482 voxels or four
measurement scales). It is presently the leading method for
monitoring dynamic processes at this scale.

- The dissolution of a monophasic carbonate is characterised by
a process of chemical erosion. The properties of the surface are
preserved. Though, it must be noticed that the reactive surface
coefficient, here equal to the specific surface coefficient, is
certainly much larger than 1. We found that with a resolution
limit of 4.91 m, \( S_S \) is close to 1.5 and that the fracture surfaces
were self-affine fractal all over four orders of measurement. By
definition the specific surface coefficient \( S_S \) is proportional to
\( (\lambda_a)^{D-2} \); then, if we assume that this characteristic persists until
the average grain size of characteristic length \( \lambda = 2 \mu m \) (evaluated
from SEM observations) one obtain a value of \( S_S = 2.4 \).

Fig.3 Specific surface as a function of the aperture. Plain and
dashed curves correspond to the 85% and the 100%
carbonate samples respectively. Crosses denote the data
dispersion.

- The dissolution of the non-pure carbonate sample displays
deply different behaviour characterised by a very heterogeneous
dissolution leading to the creation of non topographic surfaces.
In this case, profilometry methods fail to capture the real structures
and the extension of the surfaces which is proved here to increase
strongly. Using the same relation as above, one can estimate that
a value of \( S_S \) larger than 9 is reached at the end of the dissolution
experiment. However, because of the high heterogeneity of the
dissolution, it is difficult to relate \( S_S \) to \( S_S \) which is certainly non-
linear because of the preferential dissolution of the carbonated
phase. The ubiquitous increase of \( S_S \) with \( a \) cannot persist
indefinitely. From now it is important to consider dissolution over
larger time in order to explore if this relation reaches an
asymptotic regime or if cyclic behaviour takes place.

Acknowledgements : Many thanks to. ESRF-ID19 Team.
Research partly funded by the European Commission-RDG-
(EVK1-CT-2001-00091 “ALIANCE”).

REFERENCES

Adler, P.M. and J.F. Thovert, Fractures and fractures networks,
Dijkstra, P.E. and B. Berkowitz, Precipitation and dissolution of
reactive solutes in fractures, Water Resour. Res. 34(3),.457-
Lasaga, A.C., Kinetic theory in the earth sciences, Princeton
Schmittbuhl, J., J.P. Vilotte and S. Roux, Reliability of self-affine