

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

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**Abstract:** Carbonate dissolution of tensile fractures during percolation by CO<sub>2</sub>-enriched fluid is studied using synchrotronic 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

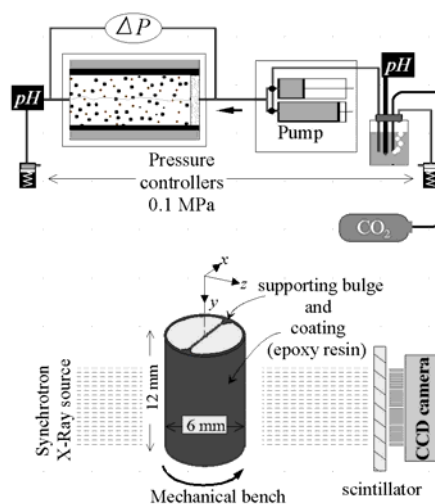
The aim of this presentation is to report the results of reactive percolation experiments in a single fracture for which X-Ray computed tomography is used to determine the structural changes in fracture morphology in course of dissolution.

## 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  $2.8 \cdot 10^{-9} \text{ m}^3 \cdot \text{s}^{-1}$ , pure water enriched with carbon dioxide at partial pressure of 0.1 MPa ( $pH = 4 \pm 0.1$ ). Output  $pH$  is  $5.1 \pm 0.15$ . 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 CO<sub>2</sub> 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  $4.91 \mu\text{m}$ . Then, fracture walls are extracted from  $2048 \times 2048 \times 2048$  tri-dimensional images.



**Fig.1 Schematic illustration of (top) the dissolution reactor, (bottom) the mini-core characteristics and the tomographic apparatus**

## RESULTS AND DISCUSSION

Two samples of micritic limestone of same origin are studied. The first one is a pure calcite ( $\phi = 2.2 \%$ ) with a fracture of aperture  $a = 50 \pm 8 \mu\text{m}$  and the second is composed of 85 w% calcite, 10 w% dolomite, 5 w% clay ( $\phi = 5.6 \%$ ) with a fracture of  $20 \pm 7 \mu\text{m}$ . 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  $5 \mu\text{m}$  to 1cm. The fractal dimension  $\mathcal{D}$ , computed coherently using 1D and 2D (Fourier Transform) power spectrum method is  $2.5 \pm 0.1$  without noticeable anisotropy for both the samples. At the initial stage, the reactive surface coefficient  $S_R$  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,  $S_R$  is kept constant and  $S_S$  is embedded in an “efficient” kinetic reaction rate (Lasaga, 1998) which doesn’t take into account  $S_R$  and  $S_S$  changes during dissolution. The total surface of each of the fracture walls is computed from the CMT images by:

$$S_z = \iint_{x,y} \sqrt{\left(\frac{\partial z}{\partial x} + 1\right)^2 + \left(\frac{\partial z}{\partial y} + 1\right)^2} dx dy$$

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  $\mathcal{D}$ , the specific surface and reactive surface coefficients (figure 3) are all invariant. The fracture walls remain TS.



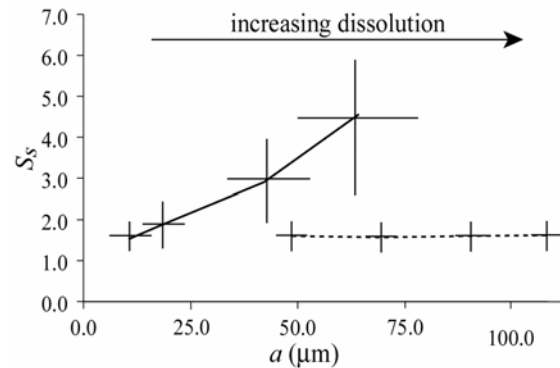
**Fig.2** A cross-section of the fracture. From left to right: (1) 100% carbonate before dissolution, (2) 100% carbonate after dissolution, (3) 85% carbonate before dissolution, (4) 85% carbonate after dissolution

In contrast, the dissolution of the second sample is noticeably heterogeneous (figure 2). The fracture walls cannot be described as TS anymore; the surfaces are folded with multiple overhangs. An analysis of 2D-projected digital images of the fracture walls would be biased. The surfaces were then analysed using isomorphic 2D and 3D-box-counting method (BCM). All those methods give similar results: a linear relation between the logarithm of the number of boxes of dimension  $l$  and the logarithm of  $l$ . This indicates that the self-affine fractal characteristic of the surface remains. A theoretical value of the fractal dimension of the surfaces can be calculated from the slope  $s$  of the former linear trend. A value of  $\mathcal{D}_{BCM} = 2.2 \pm 0.1$  was calculated for the fracture surfaces at the initial stage. Dissolution induces an increase toward  $\mathcal{D}_{BCM} = 2.4 \pm 0.1$  for the final stage of dissolution. Albeit the noticeable, but expected (Schmittbuhl *et al.*, 1995), difference between the dimension measured by spectral methods and the BCM, the results reveal a quantifiable increase of the fractal dimension. As shown on figure 3, the dissolution is also characterised by a large increase of the specific surface coefficient (from 1.5 to more than 4.5) together with a noticeable decorrelation between the aperture  $a$  and  $S_S$  as dissolution increases. This data spreading is to be related with the increasing chemical and structural heterogeneity of the fracture walls while dissolution progresses.

## 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 tool to quantify the fracture surface changes bounded by dissolution, with a resolution close to the grain size and over a large field of view ( $2048^3$  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  $\mu\text{m}$ ,  $S_S$  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  $(\lambda c)^{2-\mathcal{D}}$ , then, if we assume that this characteristic persists until the average grain size of characteristic length  $\lambda = 2\mu\text{m}$  (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 deeply 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  $S_R$  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.

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