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Experimental study of microstructural controls on the weathering of carbonate rocks

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Interactions between water and carbonate rocks are involved in various issues such as CO_2 storage, karst development and coastal weathering. However, the effect of pore structure on the weathering of carbonate rocks remains a key issue. In this study, we developed an integrated multiscale experimental workflow to investigate microstructural controls on the dissolution processes in limestone.

Two different carbonate formations are considered: Euville crinoidal limestone (φ=12-18%, k=10-150 mD) and Lavoux oolitic limestone (φ=20-30%, k=100-300 mD). Both formations have similar mineralogy (99% calcite) and a bimodal porosity distribution, but their microstructure are significantly different. Despite their higher porosity and permeability, Lavoux samples show a larger fraction of micropores.

CO_2-saturated water was injected in 80 – 100 mm long samples under 30 bar confining pressure and room temperature at a flowrate of 140 cm^3/h for 5 days (coreflood experiments). Samples petrophysical properties were measured, and CT scanner imaging, SEM observation and BET analysis were done, before and after the coreflooding experiments. Micro-coreflood experiments using chromatography columns were also performed on limestone powders. For both coreflood and micro-coreflood experiments, chemical concentrations of dissolved species were determined using ICP-AES and a thermodynamic modeling software (JChess) was used to estimate outlet fluids saturation indexes.

CT Scanner imaging shows that the dissolution pattern of Lavoux limestone is dominated by the development of localized dissolution channels ("wormholes"); whereas the dissolution of Euville limestone appears more diffuse. Mean porosity increased by around 4% for Lavoux samples and 2-3% for Euville samples, while permeability was multiplied by at least 10 and 35 respectively. In terms of dissolution kinetics, Lavoux samples are characterized by higher mean dissolution rates. Microstructural characterizations based on BET analysis and SEM observations, combined with micro-coreflood experiments performed on limestone powder, show that the higher dissolution kinetics of Lavoux limestone is mainly related to a larger specific surface area leading to a higher reactivity. Moreover, BET measurements revealed similar specific surface areas for the bulk rock and powder samples for both limestones suggesting that the specific surface area of the microporous grains is the key parameter that controls the dissolution pattern.