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What can dissolution anisotropy teach us about crystal reactivity?

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Providing experimental constraints on mineral dissolution and precipitation kinetics has mobilized geoscientists for over 50 years. The early development of reactive transport codes to model fluid-rock interactions has maintained a sustained interest in this work, with the ultimate need for the formulation of so-called 'kinetic rate laws', intended to feed these codes. In that respect, the classical strategy to determine the rate-controlling parameters of mineral reactivity has long been derived from powder dissolution experiments. While the practical benefits of such a strategy are not to be proven, getting insight into the molecular-scale mechanisms of crystal dissolution thanks to this approach is more questionable, because it implicitly assumes that crystal reactivity is an isotropic process, for which the elementary flux can simply be averaged over all the different crystallographic planes exposed to the fluid.

In this presentation, we will provide our latest developments associated to the measurement and stochastic modeling of the dissolution anisotropy of crystals at the atomic-scale, with a specific focus on carbonate and silicate minerals. The key message of our work is that the anisotropic nature of mineral reactivity has far-reaching consequences with respect to the conventional treatment of dissolution kinetics. This statement will be illustrated following various directions, including the questioning of (i) the unicity of dissolution rate-Gibbs free energy relationships for a given mineral; (ii) the "rate constant" concept; or (iii) the existence of a single elementary rate-limiting step associated to crystal dissolution. Finally, we will show the extent to which, beyond representing a detrimental complicating factor, the recognition of the anisotropy of crystalline materials reactivity offers an invaluable opportunity to refine our understanding of fluid-solid reaction mechanisms, while showing promise as a means to relate surface microstructural features resulting from dissolution to reaction conditions.