Towards mechanistic understanding of how citrate impacts dissolution of aluminosilicate materials

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Rising CO_2 levels call for quick action. One method for decreasing CO_2 in air and water is mineralisation as carbonate phases. A promising approach is geologic carbon capture and immobilisation in basaltic rocks, such as the CarbFix method. A key step in the overall mineralisation process is release of divalent cations by dissolution or leaching. Faster release means faster CO_2 uptake.

A dissolution rate quantifies the *average* rate of release in a system. It is determined from macroscopic experiments, where the effects of many microscopic processes combine to produce the overall result. Our aim is to understand the *individual*, molecular level mechanisms that work together to make the macroscopic dissolution rate. For example, organisms produce organic compounds that control basalt dissolution. Part of the process results from complexing Al in solution [1], which decreases dissolved Al activity, thus promoting further dissolution. However, the potential impact of organic compounds on release rate, as a result of adsorption onto solid surfaces, has received less attention. Just as organic molecules can complex a cation in solution, they would have high affinity for complexing with that cation on a solid surface.

We studied the dissolution of basaltic glass and synthetic CaAlSi glass, with and without citrate, in adsorption and flowthrough dissolution experiments, with atomic force microscopy (AFM) and X-ray photoelectron spectroscopy (XPS), and with density functional theory (DFT) (Beltrami et al., Merino-Diez et al., Andersson et al.: abstracts, Goldschmidt 2025). Our results indicate strong adsorption of citrate on glass particle surfaces, implying that at least part of the drive toward dissolution results from weakened bonds in the vicinity of the adsorption site, promoting Al release to solution. Gaining insight into how organic compounds act will offer insight for understanding what makes organic molecules efficient catalysts in rock weathering and could show the way toward a compound for optimising CO_2 mineralisation.

[1] Oelkers, E. H., & Gislason, S. R. (2001). *Geochim. Cosmochim. Acta*, 65, 3671-3681.

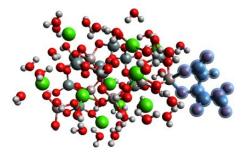


Fig. 1: A CaAlSi glass cluster, with a citrate molecule bound to Al, highlighted in blue

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