

## How do silica coatings affect mineral weathering rates?

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A long-standing problem in water-rock kinetics has consisted in the attempt to link together mineral dissolution and precipitation rates determined in laboratory experiments to the rates measured in the field. Discrepancies of orders of magnitude have actually been reported. Among the possible explanations for such outstanding inconsistencies is the potential role of secondary phases on the transport of reactants from and to the reactive surface. Such an effect could be encountered during carbonation reactions, where amorphous silica is a by-product of the process. Testing this assumption requires to perform specific experiments at an intermediate scale of study in which both the chemistry of the fluid and the mineralogy of the solid phase(s) are free to evolve. We thus studied the carbonation of wollastonite or of olivine powders as a function of time in batch reactors, at conditions relevant to geologic CO<sub>2</sub> sequestration.

Wollastonite carbonation was found to reach completion within a couple of days. Comparing the measured extents of carbonation as a function of time to the ones obtained by kinetic modeling indicates that the thick (up to 100 μm) pseudomorphic amorphous silica coating that forms on the wollastonite grains during the reaction does not affect their dissolution rates significantly. Consistently with these macroscopic measurements, TEM investigations of the silica layer revealed that it is extensively fractured and has a high porosity (up to 30%). Besides, combining diffusion modeling and [Ca] profiles across the silica/wollastonite interface measured by scanning TEM allowed to determine that the silica phase was not formed by a solid-state mechanism.

In the case of olivine carbonation, no secondary phases other than a thin (≤ 50 nm) non-porous amorphous silica layer was observed after 45 days of experiment. The fluid composition was measured to be roughly constant from the 5<sup>th</sup> to the 45<sup>th</sup> day of reaction, close to saturation with respect to amorphous silica, and with a [Mg<sup>2+</sup>]/[SiO<sub>2</sub>(aq)] ratio slightly above 2 (corresponding to a stoichiometric release).

Likely models to explain such surprising differences from one mineral to another, involving a link between the chemistry of the silica gel and its microstructure, will be discussed.

## The relative roles of source vs differentiation in determining the characteristics of arc magmas

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Mt Pelée (Martinique) and the Quill (Statia) are typical active arc-front composite volcanoes separated by ~300km along the Lesser Antilles. Differentiation trends from basaltic andesite to dacite are observed at both volcanoes. Removal of plagioclase and amphibole at both centres can largely explain major and trace element trends. Amphibole is a common phase in plutonic blocks from the volcanoes, although it is rare in the volcanic rocks, suggesting that geochemical trends are imparted by a higher pressure mineral assemblage than that erupted. Variations in isotopic composition that correlate with indices of differentiation indicate that crustal contamination accompanied fractionation (AFC). Careful evaluation of differentiation trends allows parental compositions to be constrained which, in turn, can be related to the contributions of components at source. Although similar processes are invoked to occur at both centres the resultant trends are distinct from each other and do not converge on a common parent. This observation may reflect a) derivation of distinct parental magmas from a common primary magma due to different deep, overprinted, differentiation mechanisms, or b) derivation of distinct primary/parent magmas from different mantle sources. The overlap in major element compositions of parental compositions makes (a) less likely.

The parent/primary magmas at each centre, therefore, reflect differences in source process – such as wedge composition % melting, or addition of fluid, sediment or melt. Simple one stage mantle-sediment mixing cannot produce the observed Pb isotope characteristics or the relative LILE enrichment. Sediment addition (or sediment melt addition) needs to be coupled with fluid addition to explain the relative enrichment in LILE. Because this enrichment is not accompanied by particularly radiogenic Pb or Sr, the fluid is thought to extract labile elements from the altered basaltic crust rather than sediment. Such a model would generate more incompatible element enriched magmas at Mt Pelee compared with the Quill. Subsequent crustal contamination might be expected to affect these magmas less than the more depleted Quill magmas, whereas the opposite is seen.