

## GLASS ALTERATION

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Every year about one km<sup>3</sup> of volcanic glass is formed on Earth, mostly at mid ocean ridges but some during explosive eruptions in the terrestrial environment. Chemical alteration of natural and synthetic glass constitute two main processes; dissolution of the parent glass and precipitation of secondary minerals and biofilms. Here we focus on the dissolution process. Far-from-equilibrium dissolution rates of natural glasses and silicate minerals decrease with increasing Si:O ratio; glass dissolution rates are faster and the difference increases with rising Si:O ratio. Rapid quenching of glasses results in less polymerisation and less ordering of Si–O bonds than in minerals, making glass less resistant to dissolution [1]. We have measured the far-from-equilibrium dissolution rates of volcanic glasses of variable composition, at variable solution composition, from 6°C to 300 °C [2-6]. These experiments suggest that all natural aluminosilicate volcanic glasses have similar dissolution mechanisms consisting of the sequential removal of metals from the glass structure via proton exchange reactions. The overall dissolution rate within this mechanism is governed by the detachment of tetrahedral Si atoms that have been partially liberated from the glass structure through the removal of adjoining Al. Thus, organic and inorganic ligands that complex aquatic Al<sup>3+</sup> will enhance the dissolution rates considerably at low pH (1-7).

These rate expressions have been used to model volcanic glass-soil-water interactions, glass-water-CO<sub>2</sub> interactions in laboratory and field scale experiments and basaltic glass-water-gas-mixtures interactions associated with industrial carbon capture and storage. Organic ligands accelerate the glass dissolution rates in soil, initial dissolution of pristine volcanic ash is accelerated by F<sup>-</sup> and SO<sub>4</sub><sup>2-</sup>, derived from surface coatings, and the same inorganic ligands enhance glass dissolution rates and the efficiency of mineral carbon storage during impure carbon capture and storage [e.g. 5,7-9]. [1] Wolff-Boenisch et al. (2006) *GCA*. [2] Oelkers & Gislason (2001) *GCA*. [3] Gislason & Oelkers (2003) *GCA*, [4] Wolff-Boenisch et al. (2004a,b) *GCA*, [5] Flaathen et al. (2009) *CG*, [6] Declercq et al. (2012) *GCA*. [7] Sigfusson et al. (2008) *Geoderma*. [8] Gysi & Stefansson (2011) *GCA*. [9] Snæbjörnsdóttir et al. (2017) *Goldschmidt 2017*.