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Chemical Weathering Rate of Basaltic Glass as a Function of Temperature, pH, Organic Acids and Solution Composition

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Basaltic glass plays a major role in the global flux and cycling of numerous metals and nutrients because of its widespread occurrence on the ocean floor and volcanic terrains, its emission during volcanic eruptions, and its relatively rapid dissolution rate. Despite the importance of basaltic glass in the surface environment (0-50 ºC), experimental data on the effect of pH, organic acids, and solution composition on its dissolution rate are scarce. The overall objective of this study is to create a model that can accurately predict the dissolution rates of volcanic glasses of variable composition in the Earth's surface environment. We report here some of the findings for basaltic glass comparing the experimental conditions with natural systems. The basaltic glass used in the experiments came from the Stapafell mountain, Southwest Iceland. It is similar in composition to a typical mid ocean ridge basalt (MORB). All the experiments were performed under controlled conditions in mixed-flow reactors. The pH dependence of the basaltic glass dissolution rate mimics that of aluminium oxide mineral solubility: it exhibits a sharp increase with decreasing pH at acidic conditions and a more gentle increase with increasing pH at neutral to basic conditions. The minimum rate is around the pH of 6. The pH of water in the vicinity of erupting volcanoes can be as low as 2. The pH of most surface waters is in the range of 4 to 8, but the pH of springs issuing from basaltic glass can be well above 10 (Gislason et al., 1996). The temperature dependence of the dissolution rate, 6 to 150 ºC, can be described by the Arrhenius equation. The overall activation energy of the dissolution of the glass is 45 kJ/mole, making the dissolution rate of basaltic glass less temperature dependent than that of most aluminium silicates. At conditions far from equilibrium and constant pH in mildly acidic solution the dissolution rate increased with increasing oxalic and acetic acid concentration. At pH 3 and 25 ºC, the dissolution rate tripled by increasing the oxalic acid concentration from 0 to 1 mmole/kg. The organic acid concentration range was similar to the concentration of dissolved organic carbon in soil waters (0 to 1 mmole/kg). At conditions far from equilibrium and pH 3, basaltic glass dissolution rates were 1) independent of aqueous silica activity, 2) they decreased substantially with increasing aqueous aluminium activity. The Al concentration ranged from 0 to 50 ppm and the Si concentration ranged from 0 to 15 ppm Si. The Al and Si concentration of surface waters in the vicinity of erupting volcanoes (pH 2.55) can be as high 160 ppm and 155 ppm respectively. The stoichiometry of the glass dissolution far from equilibrium changed with time. Initially, at pH 4 and 25 ºC, the dissolution was non-stoichiometric, in decreasing order; K, Na, Ca, Mg, Al, Fe and finally Si. After about 0.1% of the mass of the glass had dissolved, dissolution rates of these elements became near stoichiometric. At pH 2 and 50 ºC, dissolution was near stoichiometric with a slight preferential dissolution of Si while 3% to 40% of the mass of the glass dissolved. Regression of the experimental results (Oelkers et al., 1999) are consistent with (1) where, k+ designates a rate constant, s refers to the glass-solution interfacial surface area, a i stands for the activity of the subscribed aqueous species, KAl signifies the equilibrium constant for the Al/proton exchange reaction, A refers to the chemical affinity of the K- Na - Ca - Mg- depleted layer dissolution reaction, R is the gas constant, T absolute temperature, and n is a stoichiometric coefficient of ~ 1/3. Note that the pre-exponential part of this equation is identical to that proposed for describing alkali feldspar dissolution rates (Oelkers et al., 1994). Use of Equation (1) together with an Arrhenius relationship yields dissolution rates as a function of temperature, and taking account of solution speciation calculations permits description of rates as a function of pH, and the aqueous activities of Si, Al, and organic acids.

$$r = k_{+} \frac{\left(\dfrac{a^{3}_{H^{+}}}{a^{n}_{Al^{+3}}} \right)^{n}}{1 + K^{\bullet}_{8} \left(\dfrac{a^{3}_{H^{+}}}{a^{n}_{Al^{+3}}} \right)^{n}} \left(1 - \exp\left(- A/\sigma RT\right)\right) \qquad (1)$$

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