## Change of dissolution rate of rhyolitic glass during 52,000 years

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Numerous dissolution experiments for various silicate minerals and glasses have been conducted so far to elucidate the dissolution rate and mechanism. A dissolution rate is generally fast in the beginning of the reaction and becomes slower as the reaction time passes; the decrease of rate gradually becomes smaller. In many cases the dissolution rate is determined at reasonable elapsed time even if the rate does not completely reach a constant value. However, in order to extrapolate laboratory values to predict long term weathering rates of rocks, it is important to know whether the dissolution rate constantly decreases or finally becomes constant.

In order to investigate the change of dissolution rate of glass during long term weathering, dissolution experiments were conducted using three glass-dominated rhyolites with different weathering time. The eruption ages of the three rhyolites are 1.1-, 26-, and 52-ka and the intensity of weathering is proportional to the ages. The chemical compositions of the rhyolites at the time of eruption are similar to one another (Yokoyama and Banfield, 2002). All lavas are composed of about 90% of glass and the older lava has thicker alteration (hydration) layer on glass (Taniguchi, 1980). The amount of clay minerals (mainly consisting of allophane and halloysite) increases with increasing weathering time. The lavas are commonly very porous (porosity=30-37%; Oguchi et al., 1999). The specific surface areas (hereafter referred to as SA) of the lavas measured by BET were 0.29, 1.7, and 7.6 m2/g for the 1.1-, 26-, and 52-ka lavas, respectively. The increase in SA with age of lava is mostly attributable to the increase in amount of clays.

In the dissolution experiments, a cylindrical block sample (diameter=2.5 cm, height=8 cm) cut from each lava was inserted in a flow type reactor. Input solution was deionized water at 20 degree C (flow rate=30 ml/day) and the concentrations of dissolved ions in output solution were measured by ICP. The obtained dissolution rates after 300 days' immersion were 4.0E-18, 3.9E-18, 1.7E-18 mol(Si)/cm2/sec for the 1.1-, 26-, and 52-ka lavas, respectively. Silicon can be dissolved from any of glass, plagioclase, quartz, and clay minerals. In order to estimate the dissolution rate of glass, it is necessary to know the contributions from these phases. Because the lavas predominantly consist of glass, it is assumed that the total SA is dominated by the fractions of the glass and clay minerals. In order to measure the SA of the glass and clays, the lavas were roughly crushed, ultrasonificated in water, the glass and clays were separated, and the SA of the glass and clays were measured by BET. Then, the dissolution rate of the glass can be written as:

Rate glass = (total rate \* total surface area - rate clays \* clays surface area) / glass surface area.

Because an experimental dissolution rate of glass generally decreases as reaction proceeds, it is reasonable to assume the following relations:

(rate young glass) is equal to or greater than (rate old glass),

(rate old glass) is equal to or greater than (rate clays).

On the basis of the above assumptions, the estimated dissolution rates of the glass are 4E-18, 4E-18, 4 to 2E-18 mol(Si)/cm2/sec for the 1.1-, 26-, and 52-ka lavas, respectively. Although the thickness of hydration layer on glass is different among the three lavas, the estimated dissolution rates are similar to one another. This result indicates that the dissolution rate of glass in nature does not decrease significantly during long term weathering.