Passivation of basaltic glass by surface layers : analogy with nuclear glass

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Context

The behaviour of basaltic glasses over geological timescales raises challenging scientific issues when calculating the chemical mass balance of the oceans, or when attempting to assess potential CO2 sequestration by silicate rocks because current rate laws still cannot accurately predict the long term alteration of natural or synthetic glasses. Comparaison between calculations and field observation shows discrepancies. That could be explained by the formation of surface layers between pristine glass and bulk solution during alteration. Scientists face similar difficulties when they trying to demonstrate the safety of the highly radioactive wastes glass disposal.

Experiments & Results

For a better comprehension of the processes controlling the basaltic glass durability we performed isotopically tagged alteration experiments of model basaltic glass coupons in a closed system at 90°C in a solution initially saturated with amorphous $^{29}SiO_2$. These conditions allows the system to bypass the first transient stages of glass corrosion and to focus on the long-term rate-limiting mechanisms.

Despite the fact that the solution was initially saturated, the glass still corrodes. The formation of an alteration layer and secondary phases is evidenced by TOF-SIMS and TEM analysis. Mobiles species display local gradient well anticorrelated with that of H. This pattern is generally attributed to ion exchange and fast hydrolysis of weakly bounded species. Evidences on the alteration layer formation mechanisms are provided by following the isotopic $^{29}Si/^{28}Si$ ratio with solids caracterisation and solution analysis by MC-ICP-MS.

The results of this study are compared to those provided by the same test operated by Gin et al[1] on a six oxide borosilicate glass used as a reference material by the nuclear glass community. Some differences linked to the glass structure and composition are clearly established.

[1] Gin, S. et al. (2015) *Nat. Commun.* **6:**6360