Si isotopes to investigate mechanisms and kinetics of glass alteration

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Whatever their nature and composition, basaltic, nuclear or historic glasses are subject to alteration when in contact with water. It is thus crucial to understand mechanisms of glass alteration and to determine their associated kinetics to implement them into models aiming at predicting for instance the impact of basaltic glass alteration on ocean budget, the durability of radioactive waste in geological storage or the conservation of cultural heritage artefacts. However, recent studies [1,2,3] revealed that the respective contributions of diffusion, dissolution, condensation and precipitation are still a matter for debate. In this work, the alteration of a historic glass was investigated. Alteration experiments were performed using a dynamic device, at 30°C, at pH of 8 and 9 during 1 month with a solution doped in ^{29}Si to discriminate between the silicon from glass (mainly ^{28}Si) and from solution. The results demonstrate that the alteration layer is not a glass skeleton as it is reorganized by hydrolysis and precipitation reactions. This process is progressively achieved from the internal to the external part of the altered layer as the progress of these reactions is constrained by the extent of the modifier cations release by interdiffusion. The proposed mechanism is therefore a dissolution / precipitation process driven by interdiffusion. The key role of interdiffusion contrasts with a strictly coupled dissolution / precipitation process established for minerals and potentially for other glass types. This discrepancy will be discussed by the light of differences of glass composition and experimental conditions that control kinetics of ion exchange and hydrolysis.

[1] Hellmann et al. (2015), Nature Mat. 14, 307-311. [2] Geisler et al. (2015), Geochim. Cosmochim. Acta 158, 112-129. [3] Gin et al. (2015), Nature Comm. 6, 6360, 8pp.