Contribution of zeolite-seeded experiments to the understanding of resumption of glass alteration

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The study of the long-term behavior of nuclear glass packages is a major issue for countries that have chosen to reprocess the spent nuclear fuel.

In contact with water, glass undergoes both dissolution and irreversible transformation into more stable phases. Formation of a passivating layer causes the reduction of the alteration rate until persistence of a slow residual rate. However, a resumption of alteration (also called "stage 3")—i.e. a new increase of the glass corrosion rate—can occur [1, 2]. This phenomenon is associated with the precipitation of zeolites, mainly in alkaline environments at relatively high temperatures [3]. Understanding the origin and the consequences of this phenomenon is necessary for the prediction of glass long-term behavior. Achieving this goal is complicated by the potentially long latency period preceding the resumption of alteration. In this study, we show that this latency period, likely corresponding to the nucleation step of zeolites, is greatly reduced through the development of a new tool: seeding, i.e. the addition of zeolite seeds.

By accelerating the kinetics of glass alteration, seeding enables the study of new temperature–pH pairs for which the demonstration of a hypothetical resumption of alteration would have been difficult at typical laboratory time scales. At $pH_{90°C}$ = 10.4 and 10.1, seeding effects are less severe than at more alkaline pH. There are no significant consequences of the presence of zeolites on the glass durability at $pH_{90°C}$ = 9. In this study, seeding demonstrates its capacity to extend the predictive ability of laboratory experiments.

[1] Vienna *et al.* (2013) *Int. J. Appl. Glass Sci.* **4**, 283-294. [2] Fournier *et al.* (2014) *J. Nucl. Mater.* **448**, 348-363. [3] Ribet and Gin (2004) *J. Nucl. Mater.* **324**, 152-164.