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Identification of a Secondary Crystallized Phase formed during Nuclear Glasses Leaching – Effect of the Leached Glass Composition

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Silicate glass leaching in a unrenewed aqueous solution leads to gel formation at the glass water interface. This amorphous hydrated layer sometimes behaves like a diffusion barrier: the glass alteration rate decreases according to an inverse square root of time equation. In the case of Mg-containing glasses, the alteration rate usually remains quasi constant and seems to be controlled by the growth of secondary crystallized phases. These phases consume elements from solution. They can sustain gel dissolution inducing a decrease of its passivating properties. Long-term behaviour modelling of Mg-containing glasses designed for the confinement of fission products (AVM glasses, Atelier de vitrification de Marcoule, France) first requires a precise identification of these Mg-rich phases. Experimental investigations were performed on several glass samples using X-ray diffraction (XRD), 27Al and 29Si MAS NMR and elemental chemical analysis. Aluminous hectorite Na0.45(Mg2.28Li0.11Al0.39Fe0.06M0.16)(Si3.21Al0.79)O10(OH)2, with M being a divalent cation, was identified.

25 glasses were required for a full representation in term variations of AVM glasses composition range. Their residual alteration rates were proved to significantly depend on glass composition since a one order of magnitude difference was measured between the less and the most altered glasses. Nevertheless, the same crystallized phase was evidenced whatever the glass composition and whatever the initial composition of the solution (pure water or Mg-rich groundwater). Only a shift of the (060) peak between 1.521 and 1.530 Å was evidenced. It can be attributed to slight composition variations. Relations between the glass magnesium fraction, the amount of precipitated hectorite, the residual rate, and the measured pH are not obvious. However, hectorite precipitation was proved to depend on pH, being favoured between pH50°C 9 and 9.5. Moreover, the higher the amount of precipitated hectorite, the lower the solution pH. Self regulation of hectorite precipitation evidences that all mechanisms involved are strongly interdependent.