Influence of glass composition on vapor hydration of nuclear waste glasses

SATHYA NARAYANASAMY¹, PATRICK JOLLIVET¹, NICOLE GODON¹, FRÉDÉRIC ANGELI¹, ABDESSELAM ABDELOUAS²

¹CEA Marcoule, DEN, MAR, DE2D, SEVT/LCLT, bât 208, BP17171, 30207 Bagnols sur Cèze cedex, France

²SUBATECH, CNRS-IN2P3, IMT Atlantique-Université de Nantes, 4 rue Alfred Kastler, BP20722, 44307 Nantes cedex 03, France

In the context of confinement of high-level activity radioactive waste in a vitreous matrix, the behaviour of glass under exposure to vapor phase was investigated. In this framework, three inactive complex Mg-borosilicate glasses (>20 oxides) and three simplified (Na/Mg-Na/Ca-Na)-alumino-borosilicate glasses (4/5 oxides) were altered in vapor phase (50°C and 95% relative humidity) for 180 days and 557 days.

The glass response is strongly composition dependent. The significant effect of glass stoichiometry, especially the molar ratio of Al₂O₃/MgO, was highlighted by this result. A higher concentration of MgO than Al₂O₃ in two of the glasses resulted in 10-20 times faster alteration than the other two glasses that contained a lower concentration of MgO than Al₂O₃. In the former case, the alteration rate seemed to be driven by the precipitation of Mg-smectites. extensive The precipitation of these phases also seemed to affect the morphology of the altered layer. Network-hydrolysis seemed to be the rate-limiting vapor hydration mechanism of the four more durable glasses until 180 days.