Radioactive iodine conditioning in silver phosphate glasses

Anne-Lise Chabauty*[1,2], Lionel Campayo[2], Francois Mear[3], Lionel Montagne[4], and Olivier Pinet[2]

[1]Unité de Catalyse et de Chimie du Solide (UCCS) – Ecole Centrale de Lille, Université Lille I - Sciences et technologies, CNRS : UMR8181, Ecole Nationale Supérieure de Chimie de Lille (ENSCL), Université d'Artois – Cité Scientifique - Bâtiment C3 59655 Villeneuve d'Ascq Cedex, France

[2]CEA, DEN, DE2D/SEVT/LDMC – CEA Marcoule – BP 17171, 30207 Bagnols-sur-Cèze, France

[3]Unité de Catalyse et de Chimie du Solide (UCCS) – Ecole Centrale de Lille, Université Lille I - Sciences et technologies, CNRS : UMR8181, Ecole Nationale Supérieure de Chimie de Lille (ENSCL), Université d'Artois, Université dÁrtois – Cité Scientifique - Bâtiment C3 59655 Villeneuve d'Ascq Cedex, France

[4]Unité de Catalyse et de Chimie du Solide - UMR 8181 (UCCS) – Université d'Artois, Université de Lille, Sciences et Technologies, Ecole Centrale de Lille, Ecole Nationale Supérieure de Chimie de Lille (ENSCL), Centre National de la Recherche Scientifique : UMR8181 – Cité Scientifique - Bâtiment C3 59655 Villeneuve dÁscq Cedex, France

Abstract

In France, high level waste generated by nuclear industry is vitrified in alumino-borosilicate glasses at temperatures ranging from 1050◦C up to 1300◦C. However, radioactive iodine is highly volatile in such conditions. Thus, it can't be efficiently confined by this way. Therefore, in the hypothesis of an iodine management lying on its conditioning, it is necessary to consider other matrices. Particularly, such a matrix development should take into account the constraints of long term storage. Especially, considering the very long half-life of 129-I (more than 15 million years) and its high mobility in geological formations, the chemical durability of these matrices will have to be high.

At the end of the nineties, Japanese teams suggested that silver phosphate glasses could be used as potential conditioning matrices *(Fujihira 1999),* as they can incorporate high amounts of iodine, and can be synthetized at low temperature (between 450 and 650◦C). However, their chemical durability could be too low for them to be deemed acceptable matrices for radioactive iodine conditioning. To improve their resistance to alteration in aqueous medium, some recent works showed that crosslinking reagents, such as Al2O3, could be used (*Lesmesle 2013*). Their role is to create new bonds and thus, to strengthen the glass network. However, alumina was found to have a low solubility in these glasses, leading to a limited enhancement in their chemical durability. Therefore, in the current study, it was decided to investigate new crosslinking reagents, like Nb2O5 and Bi2O3.

The synthetized glasses were analyzed by NMR spectroscopy. This showed that the insertion of bismuth and niobium in the glass network is responsible for a decrease in the number of Q20 units, while the number of Q11 units increases. This betrays a gradual substitution of P-O-P-O-P bonds by P-O-(Nb, Bi)-O-P bonds. Moreover, leaching tests, carried out in pure water at 50 ◦C far from equilibrium (S/V=0.1 cm-1) as well as in near saturation conditions

*Speaker

(S/V=80 cm-1), showed that this insertion is followed by a significant drop in the dissolution rate for some of these glasses.

Keywords: nuclear waste, iodine, silver phosphate glasses, chemical durability