Glasses for the conditioning of fission products and minor actinides: solubility limits and implications

Lionel Campayo*†[1], Sophie Schuller , Damien Perret , and Isabelle Giboire

[1]CEA Marcoule, DES, ISEC, DE2D – CEA Marcoule 30207 Bagnols sur Ceze – France

Abstract

The management of nuclear wastes is the subject of much research. For the most harmful, France has chosen to store them in a deep geological disposal after their conditioning in matrices with high confinement performance. This is the case for solutions of fission products and minor actinides resulting from the reprocessing of spent fuels that are currently vitrified in aluminoborosilicate glass (R7T7 glass). This principle for the confinement of radionuclides at a molecular scale involves suitable formulations and conditions for producing such a glass in order to guarantee a homogeneous material for which a model of release in a disposal situation can be associated.
In this presentation, we will endeavor to describe the specific behavior of certain fission products during the production of a R7T7 type glass with particular attention paid to their solubility limit. The solubility limit is indeed an important parameter that must be taken into account both during the step of chemical reactivity between the calcinate and the glass frit during the production of the material as well as after extinction of any reactivity.

Three emblematic cases will be considered here: minor actinides and rare earths, molybdenum and iodine.

In the case of rare earths and minor actinides, exceeding the solubility limits can be the source of the emergence of crystallizations in the glass such as apatites (1) or cerianites. By concentrating the activity of alpha emitters in their crystal lattice, these phases are likely to amorphize and mechanically constrain the glass, which can cause a fracturing phenomenon. In addition, the accumulation of alpha particles in the vitreous network can result in the emergence of gas bubbles in the glass and the mobility of helium must then be studied (2, 3).

For molybdenum, an incorporation rate that would result in exceeding its solubility limit at the production temperature will lead to a liquid-liquid phase separation responsible for the appearance of alkaline and/or alkaline earth molybdates after cooling (4). By grouping together, these alkaline molybdates can be at the origin of the formation of typical microstructures called "yellow phase" which can carry cesium and present a labile character in contact with a vector of dissemination like water.

For its part, iodine is an element that is little retained in the vitreous network of R7T7 type glasses due to its volatility at the temperatures at which these matrices are produced.

*Speaker

†Corresponding author: lionel.campayo@cea.fr

The same is true for other fission products such as technetium. However, iodine has been the subject of particular attention because of its role in the safety studies of potential disposal sites. Its solubility in glass depends both on the nature of the alkalis and on their overall content (5). Research is currently being carried out on both glass and glass-ceramics and on new forming conditions to promote its confinement (6, 7).

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