Evidence of two layers alteration gel in nuclear glasses containing Zr

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Abstract

Vitrification of high-level radioactive waste in borosilicate glasses has received a great attention in several countries since decades. Glass leaching by water in geological repository is an important phenomenon that needs to be understand to better constrain the long-term evolution of the glasses used to store these wastes.

We will present structural features of the evolution of nuclear glasses under forcing conditions. During the alteration by water, an amorphous gel is formed at the surface of the glass. The durability of the gel and its properties depend on the structural role played by different elements. New generations of spent fuels require higher content of Zr4+ in glasses. The modifications of the Zr4+ environment in the gel has been investigated as a function of increasing ZrO2content from 1 to 8 mol% in simplified 5-oxide glass compositions at various pH using Zr L-2,L-3-edge and K-edge XANES and by Zr K-edge EXAFS.

In glasses and alteration gels, Zr may adopt three coordination numbers: [6]Z, [7]Zr or [8]Zr.

The structural evolution of the gel around Zr4+ in two distinct layers illustrates the molecularscale alteration of the glass when in contact with water.

Keywords: nuclear glasses, Zirconium, X, ray absorption spectroscopy

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