Irradiation effects on the leaching of nuclear waste glasses: Understanding and modeling of leaching mechanisms

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Abstract

In France, borosilicate glass, known as R7T7, is used to contain high-level, long-lived nuclear waste from spent fuel reprocessing (1). The glass canisters containing these wastes are destined to be stored in deep geological repository. After several hundred years, groundwater is expected to come in contact with the glass, which will have been self-irradiated by the radioactive elements it contains. Alteration of the glass by water, whose structure and properties will have been modified by the effects of it self-irradiation (2), represents the most important source of radionuclide dispersion in the environment.
Insofar as the alteration phenomenon takes place on a time scale that is inaccessible, modeling enables to simulate the very long-term glass behavior. In this case, the GRAAL 2 model (3), which will be used during this Ph.D. study, allows the prediction of the glass constituent elements releases in solution. Nevertheless, GRAAL 2 does not allow the distinction between the alteration of a pristine glass and a radioactive glass yet.

In order to better understand the alteration mechanisms of nuclear glasses and to adapt the GRAAL 2 parameters to radioactive glasses, this work focuses on the study of the alteration mechanisms of a simplified borosilicate glass, called ISG (International Simple Glass). Its simple chemical composition (SiO2, B2O3, Na2O, Al2O3, CaO, ZrO2) exacerbates some phenomena, such as leaching and irradiation-induced structural modifications, and makes characterization and modeling easier (4). In order to simulate the various irradiation sources of a radioactive glass, we will study two types of samples in parallel:

- Radioactive curium-doped ISG glass, which has accumulated electronic and nuclear doses for several years;

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- Non-radioactive ISG glasses, which will be irradiated using external beams under different scenarios (electrons, He, Au, Xe, Si, Ne...) to simulate the effect of self-irradiation.

The methodology chosen to study the effects of irradiation on the structure and properties of the two types of glasses as well as their behavior under alteration by water will be presented and supplemented by the first results obtained. The stored energy of each sample depending on the irradiation scenario will be quantified using calorimetric studies carried out by DSC (Differential Scanning Calorimetry) (5), and different leaching protocols will be used to favor some alteration mechanisms over the others and thus independently measure the impact of irradiation on each of them.

Keywords: ISG, Leaching, Irradiation, modeling, curium