Hydrothermal alteration of a simulated nuclear waste glass: effects of a thermal gradient and of a chemical barrier

CHRISTOPHE POINSSOT\(^1\)(\(^2\)), BRUNO GOFFÉ\(^1\), MARIE-CLAUDE MAGONTHIER\(^2\)
and PIERRE TOULHOAT\(^2\)

\(^1\) Ecole normale supérieure, Département de Géologie, URA 1316 du CNRS,
24 rue Lhomond, 75005 Paris, France
\(^2\) Commissariat à l’Energie Atomique, CEA-Fontenay, DCC/DESD/SES/Section de Géochimie,
BP n°6, 92265 Fontenay-aux-Roses Cedex, France

Abstract: The disposal of nuclear waste will create a thermal gradient in the near-field environment, the magnitude of which depends on the cooling delay before disposal, and on the geological environment. For anticipated conditions, such a disposal is expected to have a gradient of 100 to 200°C for 100 to 1000 years during the thermal period of disposal (P.A.G.I.S., 1988). We studied the influence of this thermal gradient on the alteration processes in terms of mass transfer and alteration phases.

A simulated French nuclear waste glass (containing 8 cations) was enclosed in a gold tube filled with deionized water and put in a cold-seal vessel submitted to thermal contrasts of 320-280°C and 250-220°C between 95 to 145 days at 130 bars. The new solid phases and their spatial distribution along the tube were studied by SEM observations coupled to EDS analyses, and X-ray microdiffraction. A mineralogical zonation implying mass transfer through the tube was systematically observed whatever the experimental temperature. It allowed us to demonstrate the mass transport of elements either towards the hot or the cold point of the system; indeed, zirconium and calcium moved towards the hot extremity whereas elements of the transition series and aluminium preferentially migrated to the cold one. Moreover, the absolute temperature did not affect the trend of this mass transport and only influenced the nature of the secondary phases: the lowest temperatures favoured amorphous products and phyllosilicates vs. well-crystallized phases.

In a similar experiment, the influence of a chemical barrier made of kaolinite was studied. The presence of kaolinite successfully prevented the migration of heavy elements and calcium from the glass, by inducing crystallization of oxides trapping zirconium and calcium, and by favouring precipitation of hydrated amorphous products sealing the pores of the clay.

These experiments clearly demonstrate that a thermal gradient might have important consequences for the performance of the near-field of a long-term nuclear waste disposal facility. Thermally induced mass transport might prevent the release of the radionuclides from the very-near field environment and thus be involved in the confinement properties of the disposal system. Lastly, the presence of an aluminium-rich environment increases the efficiency of the retention by favouring the crystallization of oxides that trap the heaviest elements.

Key-words: thermal gradient, nuclear waste, glass corrosion, mass transfer, hydrothermal alteration.

Introduction

The development of the nuclear industry has produced different types of nuclear wastes. High-level wastes are considered to be best disposed of deep underground in order to be removed from the biosphere. Disposing of such wastes at a depth of several hundred metres raises