# SON68 nuclear glass alteration : Gold implanted tracer validation

**Christelle CHELLY**

## Introduction

R7T7 Nuclear waste glass results from spent nuclear fuel which has been reprocessed and vitrified. A French inactive substitute named SON68 is used in laboratory to study glass behaviour in conditions of deep geological disposal, in contact with groundwater. Glass can be partly dissolved by water, and consequently, elements are released into the liquid.
In this study, our aim was to measure the concentration of glass elements which had been dissolved, especially boron, a glass network former, not retained in alteration products, in order to determine indirectly the alteration layer thickness. ICP-AES (Inductively Coupled Plasma Atomic Emission Spectrometry) and ICP-MS (ICP Mass Spectrometry) methods were used to accomplish this. These sensitive analyses permitted trace concentrations to be quantified in liquid samples.
Currently, a new method is being developed using tracers. Two chosen elements were implanted into the glass at different depths; this served to measure glass alteration with the more direct SIMS (Secondary Ion Mass Spectrometry) analytical technique.
The two tested tracers were gold, for surface tracing, and carbon, which was implanted deeper, as an internal reference. Carbon had already been validated in a previous study but the gold tracer had yet to be studied.

## Experimental Conditions

Samples were 1X1X0.2 $cm^3$ glass beads, polished on the two main faces.
Static tests were designed to observe the glass alteration; all experiments were ran at 90°C.
Reactors were 60mL Savillex™ PTFE containers, in which SON68 glass sample was placed in contact with 30mL of pure water. Tests were performed at 14, 24 and 48 hours. After leaching, solutions were filtered and acidified to pH 1 using nitric acid at 2M concentration.
A pH 1 solution was also prepared using only nitric acid and pure water, and used as a blank.
Leachates were analyzed with ICP-AES to determine the concentration of boron, a SON 68 constitutive element present at higher concentration, and with ICP-MS to measure gold concentrations present at trace levels.

## Results and discussion

From these concentrations, the normalised mass loss for boron could then be calculated using the following formula:

$$NL_B = \frac{C_B}{x_B \cdot S/V}$$ with CB as the boron concentration (mg/$cm^3$) in the

the solution, xB is the boron mass fraction within the glass, and S/V is the ratio between the glass reactive surface and the leachate volume.
The NLB value (expressed in gglass/$m^2$) allows for the altered glass quantity to be measured. Thus, the alteration thickness (expressed in nm) could be calculated using the glass density ρ.
As for gold, its implantation profile was used to estimate the altered glass thickness.
These were hypothetical calculations, especially for gold, assuming the elements were totally dissolved in the considered glass thickness.
For both elements, the altered thickness increased with leaching duration, as shown in Figure 1; however the loss of boron was much faster than gold's. The boron loss was almost linear, while the gold loss remained steady from 24 hours onward.

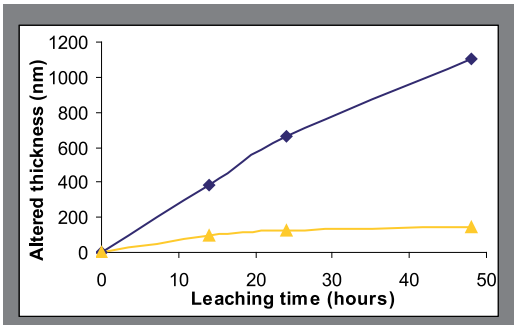


Figure 1: Evolution of glass alteration : comparison between a usual tracer (Boron) and Gold implanted.
Blue curve: Boron thickness alteration in glass
Yellow curve: Gold thickness alteration in glass

## Conclusion

In spite of the very small gold concentration within glass, this element was not soluble enough to be dissolved as quickly as boron. A consequence of this was: during leaching, gold would reach the glass surface and accumulate upon it. This element could possibly be used to quantify glass alteration. However, another type of analysis should be made, such as a SIMS analysis for example, to ascertain with the carbon tracer, more determinedly how the gold profile would evolve with leaching.

Institut de Physique Nucléaire de Lyon