# Neutron diffraction study and RMC modelling of borosilicate matrix glasses

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Borosilicate glasses are of significant current interest as suitable materials for isolating host media for radioactive waste materials [1,2]. We have undertaken a systematic structure investigation on a newly synthesized borosilicate based matrix glass system of general formula of mole% $(65\text{-}x)SiO_2$*25 $Na_2O$*5BaO*5$B_2O_3$*x$ZrO_2$ ($0 \leq x \leq 5$) and $CeO_2$ for simulation of radioactive $PuO_2$. As Ce and Pu coordination in complex oxide environments is similar, we expect that the Pu coordination will be properly simulated by $CeO_2$ addition in the host glasses. The samples were prepared by melting in platinum crucible at 1300-1450 ºC, working in atmospheric conditions.

Neutron diffraction measurements were performed at the 10 MW Budapest research reactor using the 'PSD' and 'MTEST' neutron diffractometers [3]. Despite of the great hydrolytic stability of the samples, the first few experiments revealed their tendency to superficially adsorb $H_2O$. Atmospheric humidity caused a surface swelling of the air-kept samples, and the hydrogen contained by the hydrolysed layer produced great incoherent scattering. The samples were dried at 120 C for 4 hours under vacuum conditions, which proved to be completely sufficient to obtain neutron diffraction pattern adequate for data treatment.

For data evaluation both the traditional direct Fourier-transformation, and the reverse Monte Carlo (RMC) simulation methods were applied. As a result, we have obtained the partial atomic pair correlation functions for these multi-component glasses, making possible to determine first neighbour atomic distances and coordination numbers. For RMC starting model a disordered atomic configuration was build up. The convergence of the RMC calculation was good inspite of the extremely high number of simulated parameters, and the final fit matched very well the experimental structure factors. We could successfully calculate the different atomic distances of the glass network, i.e. Si-O and B-O distances at 1.4 and 1.7 Å, respectively, the discrete values of Si-B, Si-Na, B-Na, O-O, O-Na and Ba-Na distances around 2.6 Å, and the further neighbours up to 4.5 Å.

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[1] W.G.Ramsey, N.E.Bibler and T.K.Meaker,"Composition and durabilities of glasses for immobilization of plutonium and uranium", Proc. WM95, University of Arizona, 1995.

[2] Donald, I.W., Metcalf, B.L. and Taylor, R.N.j., *Journal of Material and Science*, **32**, 5851 (1997)

[3] http://www.bnc.hu/