

# Aqueous durability of nuclear waste glasses in high-pH environments

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Experiments have been carried out to investigate the aqueous durability of nuclear waste glasses in high-pH environments. These experiments focused on two main areas; 1) the dissolution behaviour of UK simulant waste glasses and the ISG reference glass, and 2) the unique role of Ca and Mg in the formation of alteration layers at high-pH.

The former area has been investigated through long-term durability experiments, up to 2 years in duration, conducted on three relevant glasses; MW-25%, a waste-loaded simulant of UK High-Level Waste vitrified product, International Simple Glass (ISG), a reference glass used for comparison with borosilicate nuclear waste base glasses, and G73, a waste-loaded simulant of a potential glass composition for vitrification of Intermediate-Level Waste (ILW). These experiments utilised the MCC-1 protocol, at 50 °C in saturated Ca(OH)<sub>2</sub> solution (pH<sub>50</sub> 11.6). Alteration layers and crystalline precipitates were observed for each glass at each time interval, the structures and compositions of which are elucidated here through analysis by SEM-EDX, TEM with electron diffraction and geochemical modelling using PHREEQC. The formation of these layers is related to the observed dissolution behaviour of the three glasses.

The role of Ca and Mg in forming surface alteration layers on borosilicate glasses under high-pH conditions was investigated through PCT-B type durability experiments at 50 °C. These glasses contained varying CaO and MgO contents, ranging from 0 to 10 mol.%, with Ca(OH)<sub>2</sub> and KOH/KCl solutions (pH<sub>50</sub> 11.6) used as leachants. Aqueous analytical data up to 112 days show greater dissolution of all glasses in KOH/KCl than in Ca(OH)<sub>2</sub>, and a decrease in the residual rate of dissolution with increasing CaO and MgO contents. SEM-EDX of altered samples suggests that an alteration layer is only formed in the presence of Ca or Mg. The presence of Ca or Mg at high-pH, whether in solution or within the glass matrix, leads to a reduced level of glass dissolution due to the formation of a protective alteration layer on the glass surface, which impedes further leaching.