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X-ray Absorption Studies of Strontium in Borosilicate Waste Glasses

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Beamline: X23A2

Introduction: XANES and EXAFS data were collected and analyzed to characterize the strontium environments in borosilicate glass formulations to be used for immobilization of nuclear wastes. Similar to Mn, Sr can become a major constituent in some radioactive wastes, due to the use of Sr-compounds in waste pretreatment processes.

Methods and Materials: X-ray absorption fluorescence data, that included the Sr K-absorption edge near 16107eV, were collected for two powdered crystalline standards, SrCO₃ and haradaite (SrVSi₂O₇), as well as for 13 powdered borosilicate glasses. The glass samples were synthesized in crucible and joule-heated melter environments, where melt redox conditions were varied. Glass SrO concentrations range from approximately 2 to 30 wt.% as determined from X-ray fluorescence techniques.

Results: Sr XANES data are virtually identical for all glasses investigated, and indicate divalent strontium. Average Sr-O distances and coordination numbers (n) for the glasses are smaller than those observed for haradaite as shown by the Sr RDFs (Fig. 1). EXAFS analysis for SrCO₃ and haradaite indicate average Sr-O distances at 2.59 and 2.68 Å, respectively, which probably indicate that EXAFS is sensing only the closest 4 to 5 atoms in the coordination sphere around Sr in these crystal structures. EXAFS analysis results for all glasses indicate average Sr-O distances near 2.53Å.

Conclusions: XANES and EXAFS findings for the standards and glasses indicate Sr⁺⁺ that is most likely in positionally disordered environments, where EXAFS senses the inner atoms of the coordination sphere. EXAFS fitting results show that the Sr environments in these glasses have smaller Sr-O nearest neighbor distances than those for haradaite and are statistically invariant with respect to glass composition, to different melt environments, as well as to various redox conditions.

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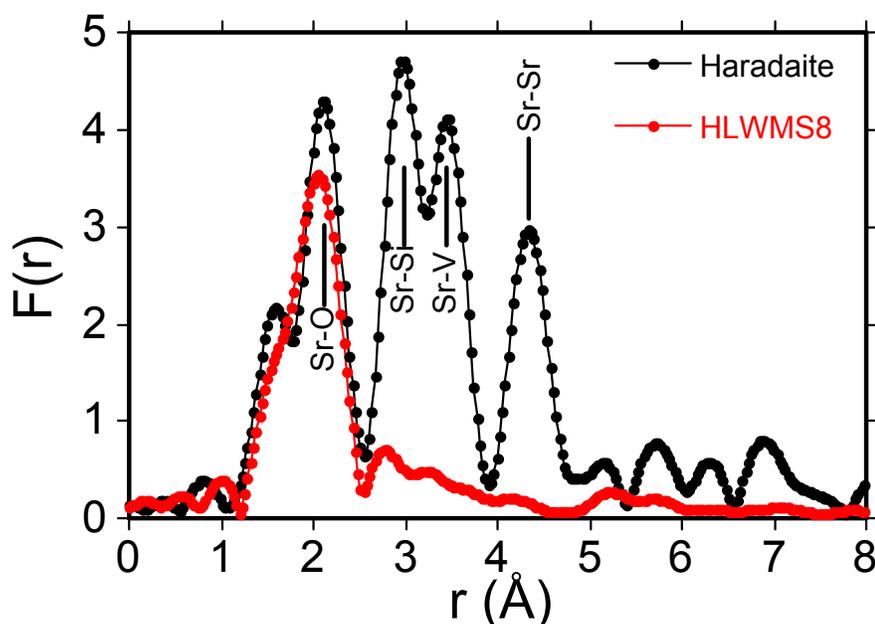


Fig. 1. Sr RDFs of crystalline SrVSi₂O₇ (haradaite) and a representative borosilicate glass (HLWMS8). Pair correlations for the haradaite RDF peaks are indicated.