The Effect of Phosphorus on Molybdenum Solubility in Boroaluminosilicate Glasses

Scott Kroeker*†1 and Arun Krishnamurthy
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¹Department of Chemistry, University of Manitoba – Winnipeg, Manitoba, Canada

Abstract

Despite the widespread acceptance of borosilicate glasses as a matrix for nuclear waste immobilization, the low solubility of certain fission products demands further research into the optimization of these wasteforms. For example, Mo loading above its solubility threshold leads to phase separation of water-soluble molybdates which sequester radioactive alkali and alkaline earth cations like Cs-137 and Sr-90. One promising approach has been to make use of the high cationic field strength of phosphorus as a network former to compete with molybdenum for oxygens. Indeed, phosphate glasses can dissolve high amounts of Mo by incorporation into phosphate chains, however at the expense of chemical durability. We have explored the phosphosilicate system in an attempt to take advantage of both networks to tune the glass properties, with boron and aluminum serving as additional network formers to mitigate the crystallization tendency. The compositions are modeled on SON68 and slow-cooled to mimic industrial nuclear waste processing conditions. The effect of this substitution on the glass structure with and without added Mo is studied using multinuclear magnetic resonance spectroscopy. NMR shows that molybdate and phosphate crystallization are inhibited in P-bearing aluminoborosilicate glasses. The structural origin of these effects appears to be related to increased Mo reticulation at the expense of B and Al connectivity.

Keywords: borate, phosphate, aluminosilicate, molybdenum, NMR, nuclear waste

^{*}Speaker

[†]Corresponding author: scott.kroeker@umanitoba.ca