08-034

Designing More Versatile Nuclear-Waste Glasses Based on Structural Insights from Nuclear Magnetic Resonance Spectroscopy

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Crystallization control is a key aspect of many industrial applications of glasses and glassceramics. In glasses used for nuclear waste disposal, preventing crystallization is important to retain radioactive species within a durable glass matrix for long-term immobilization. When the complexity of such materials makes it impossible to prevent crystallization, strategies for selective crystallization have been proposed to ensure that the devitrification products are not water-soluble and do not contain radioactive ions. Despite significant research efforts, limiting waste loading remains necessary to ensure that radioactive species do not leach out of the glasses over long periods of time. We have been exploring the role of cation field strength in mixed network-forming glasses including boron, silicon, aluminum and phosphorus to compete effectively for oxygen against poorly soluble ions, forcing them into high-coordinate disordered environments which disfavour nucleation. A similar principle can be applied with network modifiers to improve glass homogeneity. Such compositional tuning is capable of increasing the retention of molybdenum and sulfur in the glassy phase by up to a factor of four. Central to this approach is the structural understanding obtained by nuclear magnetic resonance (NMR) spectroscopy, which is used both to quantify the devitrification products and to characterize the structural chemistry of these complex glasses, identifying specific bonding sites in the base glasses which can accommodate the problem ions, thereby facilitating the design of multicomponent materials best suited for their incorporation. In favourable cases, NMR can also be used to spectroscopically observe crystallization and infer structural changes at the elevated temperatures found in radioactive glass canisters, providing results more relevant to geologic repositories. While these studies focus on preventing nucleation in complex glasses, the principles may also prove valuable in understanding crystallization in glass ceramics