PHASE SEPARATION AND CRYSTALLIZATION OF COMPLEX BOROSILICATE MELTS FOR GLASS-CERAMIC WASTE FORMS

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ABSTRACT:

A three–year project is proposed to study the development and properties of multi-phase materials that form when baseline borosilicate glass melts loaded with molybdenum-rich spent nuclear fuel (SNF) waste are cooled from above their liquidus or consolute temperatures through the glass transition temperature. Specifically, the effects of compositional variations around a baseline molybdenum-waste loaded borosilicate composition, developed by PNNL, on the precipitation of molybdate and lanthanide phases will be determined. Mechanisms for the growth of these phases, including the role of liquid-liquid phase separation, will be developed, and applied to determine the effects of quench rate on the formation and growth of desirable crystalline phases, like powellite ($CaMoO_4$) and oxyapatite ($Ca_2Nd_8Si_6O_{26}$). In particular, fast-quench experiments will be done to characterize the onset of the formation of initial phases (crystalline and phase-separated liquids) and the evolution of these phases with time and decreasing temperatures into the final phase assemblages in slowly cooled melts will be described. The tools developed to characterize the microstructural development of the baseline materials will then be applied to studies of modified compositions, with the goal of understanding how to maximize the formation of the desirable crystalline phases. These modified compositions will be selected on consultation with partners from the national labs. Finally, the effects of the development of these new phases on the properties of quenched glass-ceramic materials, including chemical durability, thermal expansion coefficient, and glass transition temperature, will be described.