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Getting AnCers: Metallothermic Molten Salt Synthesis and Reaction Thermodynamics of Actinide Ceramic Fuels

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ABSTRACT:

Generation IV reactor designers have identified uranium mononitride (UN) and uranium monocarbide (UC) alongside other actinide nitrides as promising non-oxide ceramic reactor fuels. Their favorable nuclear and thermophysical characteristics offer improved efficiency over standard uranium dioxide fuels, potentially yielding benefits in the cost of manufacture and operation. While UC is currently incorporated into many TRISO fuel schemes, it suffers from oxygen contamination, stoichiometry control, and challenges in large-scale production. UN is considered particularly promising due to a low swelling rate, low fission gas releases, high actinide density, and very favorable heat conductivity. Nonetheless, synthesis of high quality material remains a challenge, with high temperatures, high pressures, and long durations over successive reactions necessary to produce high yields. Moreover, the low efficiency of ^{15}N nitridation in the current synthesis increases costs significantly. A low-temperature, high-yield, short-duration reaction that directly synthesizes UN and UC could reduce the cost of these advanced fuels greatly.

In this proposal, a new method of synthesizing actinide (An) ceramics (Cer) by the molten salt flux method will be described. Molten salts with eutectic points below 1000 ^{o}C made of mixed chlorides or fluorides are proposed for molten salt reactors, as they can incorporate An as soluble An-halide species. With the proper chemical reducing agents and soluble anions, however, precipitation of AnCers can be promoted from such a molten salt (MS). Nitridation reactions of this kind can be so exothermic as to be self-propagating, requiring a large mass of salt as a heat sink. Similar to hydrothermal and solvothermal synthesis methods, molten salt synthesis (MSS) is a solution-based method, which can enable larger particle sizes and more efficient use of isotope-enriched materials than traditional methods. Reactive metal (metallothermic) reduction will be used to shift equilibrium away from starting materials. This proposal will examine the conditions that promote synthesis of AnCers, evaluate the utility of the metallothermic reduction of An oxides to nitrides and carbides, and measure the thermodynamics of these reactions by Differential Scanning Calorimetry (DSC) and temperature-dependent powder X-Ray Diffraction (pXRD).