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## Alternative High-Performance Ceramic Waste Forms

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**Program:** Separation and Waste Forms

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### ABSTRACT

We propose *to advance fundamental understanding of crystalline ceramic waste forms in terms of nucleation-growth and structural evolution and to demonstrate their viability as alternative waste forms.*

Major scientific and technical objectives of the proposed project are:

1. To study phase equilibrium and assemblage in the selected family of ceramic waste forms for combined Cs-Sr/Lanthanide/Transition metals high molybdenum waste streams. We will study both how the individual phases form and perform when processed individually and also how they form when processed together in multi-phase systems.
2. To evaluate melt and sintering processes to produce dense morphology that is critical to high performance in terms of elemental releases
3. To characterize the waste forms in terms of structure and chemical durability
4. To establish nucleation and growth kinetics and mechanisms of the waste forms, especially as a function of processing methods
5. To design and demonstrate a multi-phase ceramic waste forms with performance superior to the baseline borosilicate glasses.

The waste form consists of a ceramic host system that will form chemically durable titanate and aluminate phases. Targeted additives are  $\text{Al}_2\text{O}_3$  (3-5 weight%),  $\text{CaO}$  (7-10 weight%), and  $\text{TiO}_2$  (40-60 weight%) and their addition needs to be minimized to maximize the waste loading. The alkali and alkaline earth elements tend to partition to aluminotitanate phases (e.g., hollandite or  $(\text{Ba,Cs,Rb})(\text{Al,Ti})_2\text{Ti}_6\text{O}_{16}$ ), the lanthanides to aluminate perovskites (e.g.,  $\text{LaAlO}_3$ ), and strontium to a titanate perovskite ( $\text{SrTiO}_3$ ) which will form powellite ( $(\text{Ca,Sr,Cd})\text{MoO}_4$ , calzirite ( $\text{CaZrO}_3$ ), and other minor phases with an addition of  $\text{CaO}$ . Target waste loading range is 30-60 weight %. Unique challenges of the proposed project and our proposed mitigation plans are as follows:

- Molybdenum in the waste stream poses difficulty in phase formation in the ceramic waste form. Our mitigation strategy is to use a combined waste stream and maximize molybdenum loading without hindering phase assemblage and waste form performance.
- As a waste form, crystal structures of hollandite and powellite allow structural variability. At the same time, the variability needs to be controlled to achieve the most optimum microstructure and phase assemblage and best performance. Our mitigation plan is to advance fundamental understanding and development of the structure-property relationship, using novel processing as well as characterization.

Major outcomes of the proposed project are scientific understanding of the underpinning structure-property relationship in designing alternative waste forms, specific formulations and processing conditions required to produce those waste forms, technical presentations in national/international conferences, publishing in peer-reviewed journals, training of next-generation of engineers and scientists (one post-doc, one full-time



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graduate student, and 4-6 undergraduate students) for the nuclear energy industries. On the scientific front, our project seeks understanding of the structure-property relationships within these advanced waste forms with the objective of tailoring them for optimum performance. The results of the project will lead to *better understanding of these waste forms* and also *reveal new formulations, improved processing routes, and solutions for significantly increasing the performance*. Programmatically, our project directly supports the DOE's program on Fuel Cycle R&D and the need identified under Separation and Waste Forms campaign, "*Development of alternative waste forms that have the potential of significantly increased waste loading (of fission products) and durability over borosilicate glass.*"

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