
Development of Waste Processing Methods for MCFR Fuel Salt

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ABSTRACT:

The molten chloride fast reactor (MCFR) is a promising new reactor design that has benefits of low-pressure operation, strong temperature driven negative reactivity feedback, and perpetual access to the fuel for monitoring and controlling its composition. Use of a fast neutron spectrum enables burning of transuranic actinides (TRU) coupled with tolerance for a high concentration of salt-soluble fission products. However, end of life salt will necessarily be produced to bleed out fission products that are neither fissile nor fertile. Based on the assumption that the salt will be initially fueled with expensive high assay low enriched uranium (HALEU) and that the Cl⁻ ions will be enriched in costly ³⁷Cl, the spent fuel salt will contain high economic value based on inventory of ²³⁵U, TRU, and ³⁷Cl. If left untreated, the spent fuel salt will be extremely difficult to dispose of safely due to its tendency to absorb water, dissolve in water, and become extremely corrosive to metals. It is likely a necessity that the spent fuel salt be converted into a durable waste form suitable for permanent geologic disposal.

The objectives of this project are to develop methods for recovering U and TRU from MCFR spent fuel salt, dechlorinating the salt, and converting the residual waste into glass waste forms that will be durable and safe to permanently dispose. If successful, this will result in closing the nuclear fuel cycle for the MCFR and dramatically improving the fuel economics relative to direct disposal of the salt. Need for long term storage of spent fuel salt will be eliminated along with nuclear criticality and risk of environmental contamination. Reuse of fissile actinides in the spent fuel salt would also dramatically reduce the proliferation risk associated with storing the salt.

To accomplish these objectives, a diverse team of experts in molten salts, actinide processing, electrochemical processing, machine learning, and experimental and computational waste form development has teamed up with the leading company working to develop the MCFR, TerraPower, LLC. Based on their design that uses a eutectic NaCl-UCl₃ or NaCl-KCl-UCl₃ salt, TerraPower will calculate burnup for spent fuel salt with composition of soluble fission products and actinides. Surrogate salts will then be synthesized for testing actinide recovery, dechlorination, and waste form synthesis.

Two processes will be tested for actinide recovery using surrogate salts (UCl₃ will be included in select experiments). One such process is designed to volatilize U via reaction with Cl₂ to form highly volatile UCl₅ and UCl₆. A condenser will be designed and tested to recover the volatile U compounds in a form that can be reduced back to UCl₃ and returned to the MCFR. The other process is designed to electrochemically plate out metallic U/TRU onto cathodes that can be rechlorinated and returned to the reactor. Similar technology has been used in electrorefining of metallic spent fuel in molten LiCl-KCl but has never been adapted to MCFR fuel salt. A key difference between the two salts is that the MCFR salt requires a high U concentration to depress the salt melting temperature. Eutectic NaCl-UCl₃ contains 33 mol% UCl₃ with an overall mass of the salt is 52% U. Simply extracting U from the salt can quickly cause the salt to freeze unless it is heated to extremely high and corrosion-inducing temperatures. For

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this project, we are proposing to maintain molten salt mixtures throughout U/TRU extraction from the salt by a replacement reaction that keeps the melting temperature of the salt low. Gd reactive anodes will be used to replace UCl_3 with GdCl_3 . A eutectic NaCl-UCl_3 should morph into a eutectic NaCl-GdCl_3 . Most electrochemical experiments will be performed with TbCl_3 as a surrogate for UCl_3 , and select verification tests will be done with UCl_3 .

Two processes will also be tested for dechlorinating the salt post-actinide removal. In one process, $\text{SiO}_2\text{-Al}_2\text{O}_3\text{-P}_2\text{O}_5$ (SAP)-based precursor composites will be mixed with simulated waste salt to release $\text{Cl}_{2(g)}$ upon heating, as the metal cations from the salt to partition into chemically durable crystalline and glassy phases. This process has been reported for dechlorinating LiCl salt waste generated from electrolytic reduction of oxide spent fuel but has never been applied to MCFR fuel salt. Additionally, the design was empirically based, and an understanding how chloride in the targeted interacts with specific reactive linkages (e.g., P-O-P) is still needed. Therefore, first principles-based calculations will be used in tandem with experiments to understand how dechlorination reactions develop, and this information will be used to adapt the SAP-based process to MCFR salt. Another dechlorination process will also be tested in which MCFR salt is reacted with organic acids (e.g., oxalic acid) to partition the metal cations into oxides and release $\text{HCl}_{(g)}$. The benefit of this second process is that the resulting dechlorinated product can be introduced into a chemically durable borosilicate glass; however, it is limited to waste streams that are compatible with a high moderator content. Going into this project, it is unknown whether actinide removal can be sufficiently efficient to make it possible to run a high moderator content process. Thus, it is essential to develop two dechlorination process options. Included in the project scope will be criticality calculations to determine the actinide concentration limits that will allow for the use of moderators in the dechlorination process.

The dechlorinated products from both processes will be converted into ceramic and glass waste forms via vitrification (e.g., aluminosilicates, apatites, borosilicates). Machine learning and molecular dynamics will be used to inform experimentalists on how to select precursors to develop chemically durable waste form compositions depending on the starting dechlorinated product. Waste forms samples will then be synthesized and characterized for structure, stability, and leach resistance.

In order to comply with storage, transportation, and waste acceptance requirements, existing regulatory frameworks will be assessed compared to the expected composition and properties of salt waste and glass waste forms generated. Salt-specific properties such as affinity for water will be considered when outlining storage packaging/environment requirements. Heat decay will also be considered for its impact on storage, disposal, and transportation. Importance of dechlorination will be assessed via its impact on radiolytic gas generation that could contain long-lived radioactive ^{36}Cl , which is particularly harmful to the environment. New cask designs may be required, given existing casks were designed for solid, clad light water reactor fuel.

There are several outcomes expected from this project. First, it will develop one or more processes to recover valuable fissile U/TRU that can be returned to the fuel salt. Second, it will develop one or more processes to dechlorinate the fuel salt for the dual purpose of reusing valuable ^{37}Cl and minimizing the potential contamination of the environment with long-lived ^{36}Cl . Third, it will develop chemically durable waste forms for MCFR fuel salt that can be safely disposed of in geologic repositories. And fourth, it will generate data on dechlorinated salt waste forms and actinide processing in molten salts that will be published and will advance the science and technology of molten salt processing of metallic and oxide spent fuel.