

Hybrid Process Development – Chlorination

Christine T. Snyder, Javier Figueroa, and Leonard Leibowitz

Chemical Engineering Division, Argonne National Laboratory



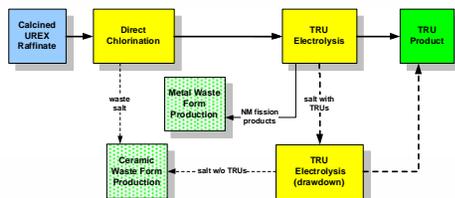
Introduction

The Department of Energy (DOE) Advanced Fuel Cycle Initiative (AFCI) was launched to address pressing nuclear issues facing the United States. The goals of the AFCI Program are to:

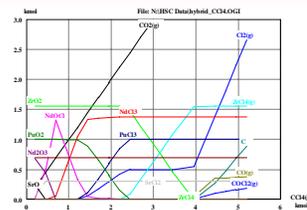
- ❖ Reduce the long-term radiological impact of waste
- ❖ Enable development of a simpler, cheaper repository
- ❖ Improve long-term prospects for nuclear power

The long-term hazards in spent nuclear fuel are posed by 1% of the content, which includes plutonium, neptunium, americium, and curium. These transuranic elements will fission and release large amounts of energy. However, before the transuranics are subjected to transmutation, they must be partitioned from the spent fuel.

Hybrid separations processes are being developed to address the AFCI spent nuclear fuel treatment goals and meet the separations criteria.



The development of the direct chlorination method represents an initial step in the advancement of a process for the treatment of uranium extraction (UREX) raffinate.



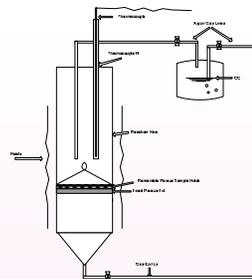
The experiments this year mark the initial evaluation of chlorinating methods which will eventually be adapted for use in a glovebox, where actinide oxides found in spent nuclear fuel will be converted to chlorides.

Experimental Approach

Our approach to developing a direct chlorination method began with:

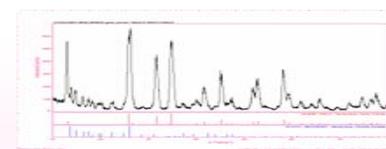
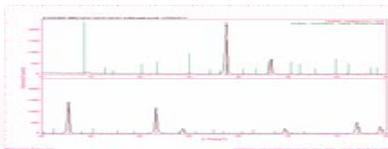
- 1) Fabricating a quartz reaction vessel to withstand the range of temperatures needed for the reaction (500°C-800°C)
- 2) Choosing a surrogate oxide for use in place of an actinide oxide
Neodymium oxide and cerium oxide
- 3) Selecting a chlorinating agent
Carbon tetrachloride

Argon gas saturated in carbon tetrachloride will flow into the reaction vessel and pass through a porous frit containing the rare-earth oxide.



Experimental Results

Neodymium oxide and cerium oxide were successfully reacted with carbon tetrachloride and converted to neodymium chloride and cerium chloride.



Conclusions

The oxide-to-chloride conversion process was demonstrated using rare-earth oxides as surrogates for the transuranic oxides. Process development will continue, with the goal being to optimize process operating conditions, maximize conversion rate, and minimize the amount of chlorinating agent required for conversion. Multicomponent chlorination tests to validate process kinetics will also be completed. Tests with actinide oxides will follow the multicomponent chlorination studies.