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Nuclear Criticality, Shielding, and Thermal Analyses of Separations Processes for the Transmutation Fuel Cycle

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BACKGROUND

The first step in any transmutation strategy is the separation of radionuclides in used nuclear fuel. The current separation strategy supporting the Advanced Fuel Cycle Initiative (AFCI) program is based on the use of a solvent extraction separation process to separate the actinides, fission products, and uranium from used commercial nuclear fuel, and on the use of pyrochemical separation technologies to process used transmuter fuels. To separate the fission products and transuranic elements from the uranium in used fuel, the national program is developing a new solvent extraction process, the Uranium Extraction Plus, or UREX+, process based on the traditional solvent extraction reprocessing technologies.

Preparing fuel for possible burn up in light water reactors, fast reactors, or accelerator-driven systems involves various chemical processes to partition the transuranics (neptunium, americium, plutonium, and curium) from the fission products. This results in waste streams that are highly radioactive and require radiation shielding for safety. These transuranic elements pose varied criticality, thermal, and radiation risks during storage and handling. Additionally, the radioactive decay of strontium and cesium waste products of the UREX+ technique produce roughly half of the thermal products and gamma radiation emissions in spent fuel. These radioisotopes require storage for approximately 300 years before heat and radiation hazards decrease to a safe level.
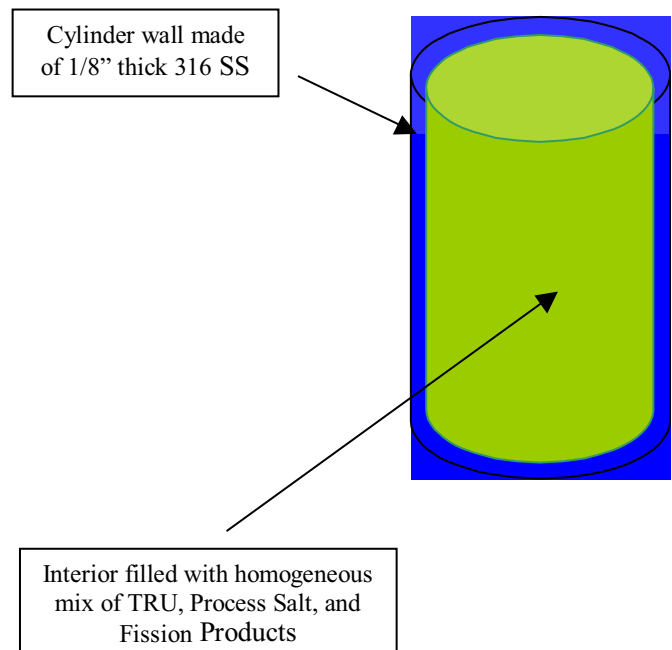
As the volume of waste requiring treatment increases, a higher probability exists that fissionable isotopes of plutonium, neptunium, and curium can accumulate and form a critical mass. Criticality concerns warrant an assessment of the effective neutron multiplication factor, or k_{eff}, to prevent a possible sustained fission reaction. Maintaining k_{eff} below a safe level (<0.95) prevents criticality events. This parameter can be computed for any combination of fuel and geometry using Monte Carlo neutron transport codes. Monte Carlo simulations establish the best means of examining the criticality safety of the proposed separation processes, and allow engineers to develop proper safety measures for the reprocessing and fabrication of actinide fuels.

Candidate storage containers also require analysis to assess the need for radiation shielding. Since minor actinides generate significant amounts of heat through radioactive decay, proposed containment measures must be designed to avoid excessive temperatures. Radioactive decay also generates heat that can lead to melting of the fuel during storage and handling.

RESEARCH OBJECTIVES AND METHODS

The primary goal of this research program was to provide the nuclear and thermal modeling support for the development of this new separation process. The assessments of nuclear criticality, radiation for shielding, and thermal analyses of wastes in the Cs/Sr, Pu/Np, and Cm/Am waste streams will assist in designing the UREX+ process. This project was identified as a critical R&D need of the Chemical Technology Division (CTD) at the Argonne National Laboratory (ANL) as safety concerns associated with criticality, shielding, and heat buildup must be addressed prior to further development of the UREX+ process.

UNLV students used nuclear analysis codes to perform assessments of k_{eff} at different points in the separation processes that have been identified by the project collaborators at ANL-CTD. They also worked on problems to assess the need for radiation shielding and to develop software to assess the possibility of excessive temperatures due to radioactive decay in separated wastes. ANL-CTD has provided sample fuel process geometries and compositions for calculation of k_{eff} as a function of the relative concentrations of process salt, transuranics, and fission products.

Cylinder wall made of 1/8" thick 316 SS

Interior filled with homogeneous mix of TRU, Process Salt, and Fission Products

Cylindrical problem with TRU, process salt, and fission products.

RESEARCH ACCOMPLISHMENTS

An investigation and analysis of criticality and thermal effects for the safe storage of curium was completed. The assessment involved determining k_{eff} as a function of fuel burnup, initial enrichment, and time since irradiation. Additionally, since curium generates a substantial quantity of decay heat, an analysis was completed to determine the mass of curium that will lead to temperatures high enough to melt the metal. A spherical geometry was used in the analysis. Heat removal from the sphere was assumed to be a combination of natural convection and radiation heat transfer. This heat transfer analysis was also modified and resulted in an analysis that utilized a more sophisticated and suitable cylindrical container. A report developed for Dr. Laidler at ANL described the in-depth investigation regarding the criticality and thermal properties of curium. The heat transfer spreadsheet was used for other combinations of minor actinides as indicated by ANL.

Investigators also investigated the properties of the other minor actinides, including plutonium and americium. A study was conducted on the values of k_{eff} and ultimate temperature for varying combinations of plutonium, americium, and the remaining minor actinides.

UNLV students used nuclear analysis codes SCALE 4.4 and/or MCNPX to perform assessments of k_{eff} at different points in separation processes that have been identified by ANL-CTD. They also worked on problems to assess the needs for radiation shielding and develop software to assess the possibility of excessive temperatures due to radioactive decay in separated wastes. ANL-CTD has provided sample fuel process geometry and composition for calculation of k_{eff} as a function of the relative concentrations of process salt, TRU actinides, and fission products. The research team analyzed the cesium/strontium waste stream, the plutonium/neptunium waste stream, and the americium/curium waste stream.

TASK 11 PROFILE

Start Date: August 2001
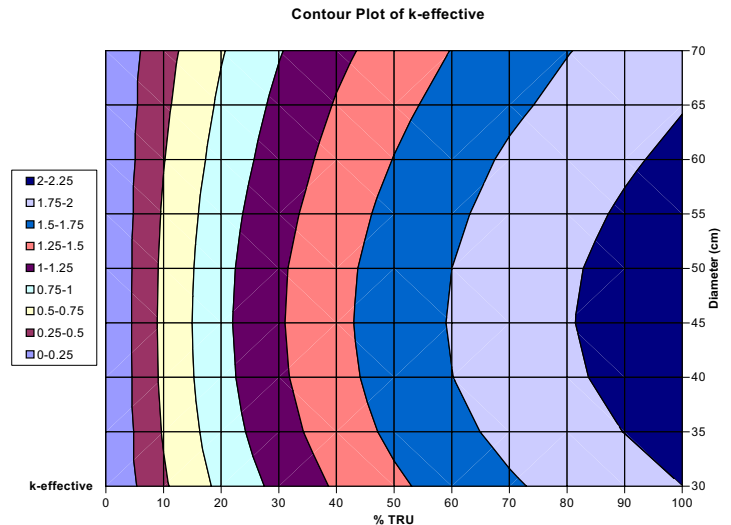Completion Date: August 2004

Thesis Generated:
Srinivasa Elizabeth Bakker, M.S., Mechanical Engineering Department, "Criticality and Thermal Analysis of Separated Actinides in Transmutation," November 2003 .

Reports:
B. Culbreth, E. Bakker, and J. Viggato, "The Fission Properties of Curium Separated from Spent Nuclear Fuel," Department of Mechanical Engineering, University of Nevada, Las Vegas, April 2002.
B. Culbreth, D. Lowe, and J. Viggato, "Assessment of Criticality Safety for Cylindrical Containers to be Used In the Processing of Spent Fuel," Department of Mechanical Engineering, University of Nevada, Las Vegas, October 28, 2001.

Cm Metal Cylinder		Criticality		Heat Transfer	
		rad (cm)	mass (kg)	rad (cm)	mass (kg)
	bare	4.6	82.52	1.1	1.129
	water	2.7	16.69	3.3	30.48
Cm_2O_3 Cylinder		Criticality		Heat Transfer	
		rad (cm)	mass (kg)	rad (cm)	mass (kg)
	bare	5.4	125.59	1.55	2.74
	water	3	21.53	2.25	8.37
Am_2O_3 Cylinder		Criticality		Heat Transfer	
		rad (cm)	mass (kg)	rad (cm)	mass (kg)
	bare	11.2	1032.28	1.6	3.01
	water	10.5	850.58	1.9	5.04
Am_2O_3+Cm_2O_3		Criticality		Heat Transfer	
Cylinder		rad (cm)	mass (kg)	rad (cm)	mass (kg)
	bare	10	737.9	1.4	2.02
	water	9.3	593.54	2	5.88

Recommendations for Cm and Am oxide inventories based on criticality and melting temperature.

Contour Plot of k-effective

Legend:
- 2-2.25
- 1.75-2
- 1.5-1.75
- 1.25-1.5
- 1-1.25
- 0.75-1
- 0.5-0.75
- 0.25-0.5
- 0-0.25

k-effective

Contour Plot of the Effective Neutron Multiplication Factor as a function of cylinder diameter and % TRU in the mixture.

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