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


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## Task 11

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

W. Culbreth and D. Beller

## 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.

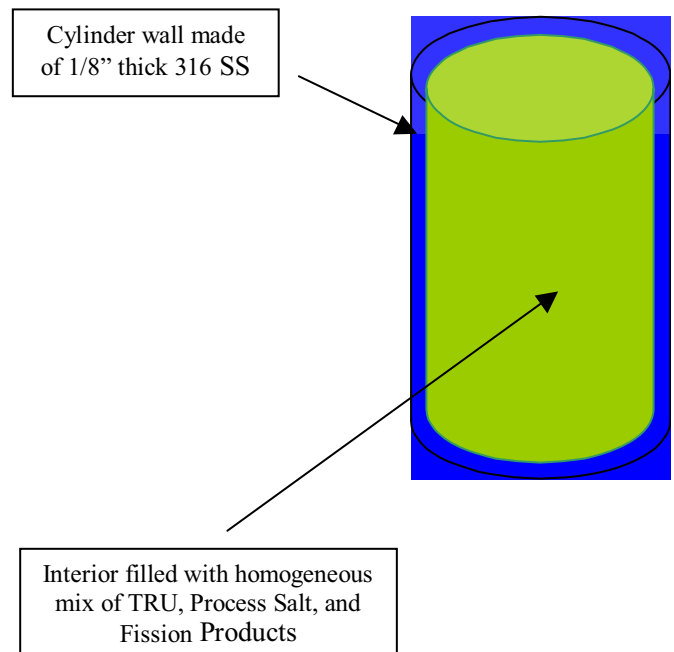
As the volume of waste requiring treatment increases, a higher probability exists that fissionable isotopes of plutonium, neptunium, and curium can accumulate and form a critical mass. Criticality concerns warrant an assessment of the effective neutron multiplication factor, or  $k_{\text{eff}}$ , to prevent a possible sustained fission reaction. Maintaining  $k_{\text{eff}}$  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 is 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 has been 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  $k_{\text{eff}}$  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  $k_{\text{eff}}$  as a function of the relative concentrations of process salt, transuranics, 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  $k_{\text{eff}}$  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 will be used for other combinations of minor actinides as indicated by ANL.

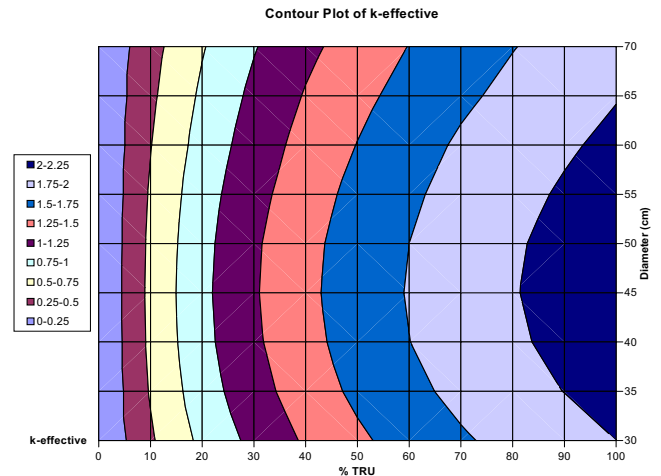
Investigators initiated preparatory efforts to investigate and analyze the properties of the other minor actinides, including plutonium and americium. A study was begun on the values of  $k_{\text{eff}}$  and ultimate temperature for varying combinations of plutonium, americium, and the remaining minor actinides.



*Elizabeth Bakker presenting her research at the ANS Student Conference, Berkeley, CA, April 2-6, 2003.*

## HIGHLIGHTS

- ◆ “Critical and Thermal Investigation of Curium and Other Minor Actinides for Safe Storage and Disposal” presented at the ANS Student Conference, Berkeley, CA, April 2-6, 2003.



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

## FUTURE WORK

UNLV students will use nuclear analysis codes SCALE 4.4 and/or MCNPX to perform assessments of  $k_{\text{eff}}$  at different points in separation processes that have been identified by ANL-CTD. They will also work 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  $k_{\text{eff}}$  as a function of the relative concentrations of process salt, TRU actinides, and fission products. The research team will analyze the cesium/strontium waste stream, the plutonium/neptunium waste stream, and the americium/curium waste stream.

### Research Staff

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