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Nuclear Criticality Analyses of Separations Processes for the Transmutation Fuel Cycle

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Task 11

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GOALS AND BACKGROUND

The separation and partitioning of used commercial reactor fuel is a vital component of any reprocessing or transmutation strategy. To process the high actinide fuels required for a transmutation effort, the Chemical Technology Division (CMT) at Argonne National Laboratory (ANL) is developing a pyrochemical separations process. Currently, this work is being done via small experiments. While this is more than sufficient to develop the technologies required to process actinide-bearing fuels, it does not allow for the direct investigation of criticality concerns that would be present in larger systems. As the volume of waste to be treated increases, a higher probability exists that fissionable isotopes of plutonium, americium, and curium can accumulate, forming a critical mass. These criticality events can be avoided by ensuring the effective neutron multiplication factor, k_{eff}, remains below a safe level. Monte Carlo simulations to evaluate k_{eff} are the best way to examine the criticality safety of the proposed separation processes, and will allow engineers to develop proper safety measures for the reprocessing and fabrication of high actinide fuels.

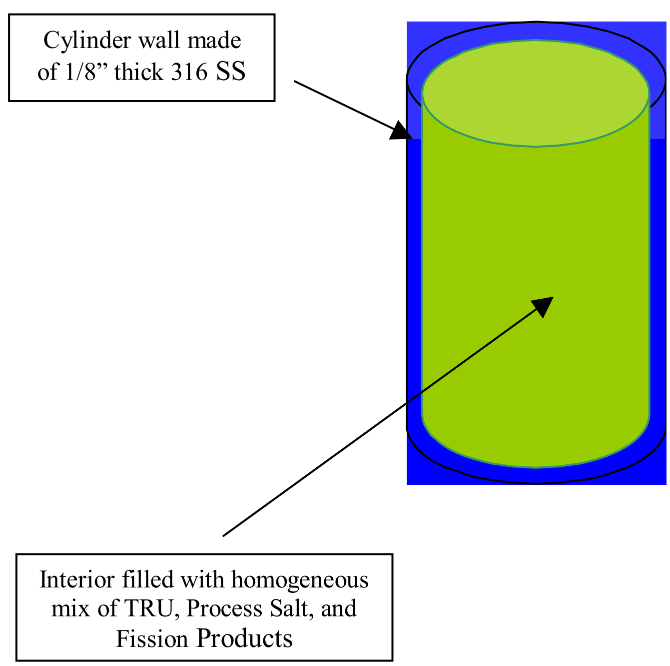
A related problem for handling high actinide fuels is the heat generated by the decay of the higher actinides. In particular, the presence of curium in transuranic wastes poses a significant problem. To minimize the impact of curium on the fabrication of actinide-bearing fuels, the process engineers and chemists would like to remove the curium from the fuel. Curium, however, not only poses a criticality concern, but also includes isotopes that generate a great deal of decay heat. This heat generation creates safety problems with regards to handling and storing curium. The decay heat can cause samples to melt very quickly if excessive quantities of curium are present. SCALE, a Monte Carlo code, simulates the scattering and absorption of neutrons. This technique permits assessment of what quantities of curium will result in a critical mass. This information is then combined with thermal transfer models to examine the decay heat issue, and evaluate thermally and critically safe storage configurations for curium-rich waste streams.

RESEARCH OBJECTIVES AND METHODS

The primary objective set out by the UNLV research team was to build expertise in criticality safety at UNLV, and then to use that expertise to analyze problems and develop tools for the CMT group at ANL in order to support the development of these new separations and partitioning technologies. For the first year, the UNLV research team set forth the following goals: (1) train the UNLV students to use the specialized codes necessary to examine criticality safety; (2) develop models, using these codes, to assess neutron multiplication factors for geometries and material concentrations of interest to the chemists and chemical engineers developing the technologies; (3) develop software to incorporate criticality estimates into the existing ANL models for the pyrochemical treatment processes; and, (4) to bring the students to meet the chemists and engineers at ANL to identify, understand, and verify research objectives and goals established through collaboration with ANL. Additionally, the UNLV research team was requested to examine not only the criticality concerns, but also the thermal concerns associated with the storage of separated curium.

RESEARCH ACCOMPLISHMENTS

Through the meetings between the UNLV research team and the CMT at ANL, the modelers were able to gain significant insights with respect to the geometry of the equipment used in fuel separation along with scenarios that can lead to potential criticality events. Being able to actually see the equipment used allowed the modelers to better understand the complexity of working with radioactive and fissionable substances through glove boxes, which in turn



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

allowed them to develop more useful alternative approaches that would minimize criticality risk while still being useable by the researchers and engineers at the CMT in these difficult conditions. Results from the criticality simulations were submitted as a report to ANL entitled, "Assessment of Criticality Safety for Cylindrical Containers to be used in the Processing of Spent Fuel". These results were subsequently integrated into the excel model developed by the CMT researchers to support the development of their experiments and flowsheets. The UNLV code package assesses the maximum mass of transuranic material that can be safely accumulated in a pyrochemical cell. This code was based on models developed by the students using the SCALE code to assess the safety of the pyrochemical cell as transuranics are accumulated.

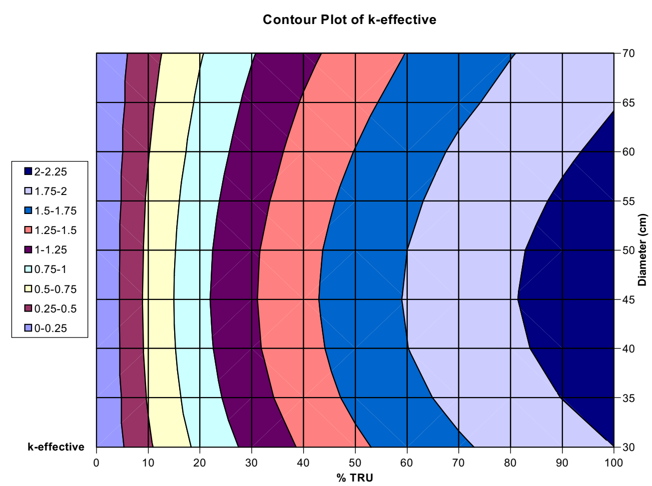
During this effort, the group's collaborators at ANL came to focus on the particular problems associated with separating and handing curium. As a result, ANL requested that the UNLV team re-direct their research to examine the potential for curium samples to "go" critical. Ms. Elizabeth Bakker (UNLV undergraduate Mechanical Engineering student) analyzed the problem using SCALE. This work was documented extensively in the "Fission and Thermal effects in Curium Separated from Spent Nuclear Fuel" report that was submitted to ANL. This report documents the thermal hazard involved with separating curium from spent nuclear fuel, the fissionable isotopes produced, and the maximum mass of curium that can be safely stored. This work also included parametric studies to determine k_{eff} values for specific geometries, component concentrations, radionuclide content, and fuel configurations.

FUTURE WORK AND GOALS

The UNLV research team is working with their collaborators in the CMT at Argonne to develop and scope out additional criticality problems for analysis. One issue already identified relates to the criticality safety of the pyrochemical cells. The UNLV research team will also begin the evaluation of various candidate separations processes for efficiency, safety, reliability, and cost. These evaluations, along with the criticality safety analyses, are a vital component of the safety case for these processes, and are a necessary step in designing activities for process equipment and facilities that must begin in the not too distant future.

HIGHLIGHTS

- Ms. Bakker, Mr. Lowe, (Students, UNLV-ME) presented papers on this work at the American Nuclear Society Student Conference, as well as posters at the ANS Conference, both held in Reno, NV, in November 2001.
- Mr. Jason Viggato, a graduate student employed on this project, completed his graduate studies and went to work for Bechtel-SAIC as part of the Yucca Mountain Project, analyzing thermal and criticality problems for the proposed national nuclear waste repository (putting his training from the UNLV project to work).
- Two reports, "Fission and Thermal Effects in Curium Separated from Spent Nuclear Fuel" and "Assessment of Criticality Safety for Cylindrical Containers to be used in the Processing of Spent Nuclear Fuel," were prepared and delivered to the team's collaborators at Argonne National Laboratory.



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

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