8-12-2001

Nuclear Criticality Analyses of Separations Processes for the Transmutation Fuel Cycle

William Culbreth
University of Nevada, Las Vegas, william.culbreth@unlv.edu

Pang Tao
University of Nevada, Las Vegas, pang@physics.unlv.edu

Project Title:

Nuclear Criticality Analyses of Separations Processes for the Transmutation Fuel Cycle

August 12, 2001

Principal Investigators: William Culbreth, Ph.D.
Department of Mechanical Engineering
University of Nevada, Las Vegas
4505 Maryland Parkway, Las Vegas, NV 89154-4027
(702) 895-3426, culbreti@clark.nscee.edu, fax: (702) 895-3936
http://culbreth.me.unlv.edu

Tao Pang, Ph.D., Co-Principal Investigator
Department of Physics, Box 454002
University of Nevada, Las Vegas
(702) 895-4454, fax: (702) 895-0804
pang@nevada.edu

Collaborator (UNLV): Paige Russell, M.S. (Doctoral Student)
Department of Mechanical Engineering
Department of Physics Student (to be determined)

Denis Beller, Ph.D., UNLV/AAA University Liaison
Harry Reid Center for Environmental Studies
(702) 895-2023, beller@lanl.gov

Collaborators (DOE): George Vandegrift, Ph.D.
Senior Chemist, Group Leader
Separation Science and Technology
Chemical Technology Division
Argonne National Laboratory
(630) 252-4513, fax: (630) 972-4513
Vandergrift@cmt.anl.gov

James Laidler, Ph.D.
Director, Chemical Technology Division
Argonne National Laboratory
(630) 252-4479, fax: (630) 252-5528
Laidler@cmt.anl.gov

AAA Research Area: **Separations**

Requested First Year Funding: $ 110,123 (out years comparable)

Note: ANL employees do not require funding from UNLV to participate in this project.

Nuclear Criticality Analyses of Separations Processes for the Transmutation Fuel Cycle

Abstract

To mitigate the waste created by conventional fission reactors, spent nuclear fuel must be mechanically separated from its cladding. For the development of fuel processing technology to support the Advanced Accelerator Applications (AAA) Program, aqueous and pyrochemical processes will be used to further separate technetium and iodine, uranium and the higher actinides (see Figure 1 for an example of the process layout)[1]. The higher actinides, including plutonium, americium, curium, and neptunium will be separated from the waste to facilitate their fabrication into new fuel for placement in a transmuter. High-energy neutrons generated by spallation in the transmuter break down these actinides and long-lived fission products through activation and fission to produce stable elements or radionuclides with short half lives.

During the separation process, concentrated quantities of fissionable plutonium and americium pose a potential nuclear criticality risk. At each stage in the process, an assessment of the effective neutron multiplication factor, k_{eff}, will be necessary to prevent the possibility of sustained fission[2]. We propose to perform nuclear criticality analyses in support of the development of fuel separation processes for AAA. This project has been identified as a critical R&D need of the Chemical Technology Division at the Argonne National Laboratory (ANL-CTD).

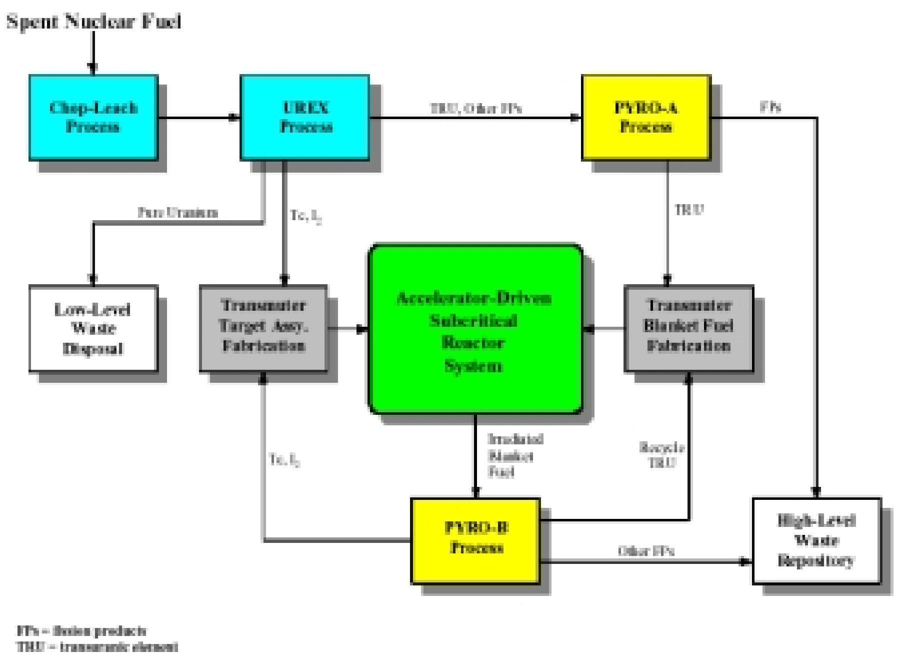

Figure 1. Chemical Separation Process for the AAA System (http://www.cmt.anl.gov/research_highlights/default.htm)

Work Proposed for Academic Year 2001-2002, Goals, and Expected Results

UNLV students will use nuclear analysis codes SCALE 4.4 and KENO VI and/or MCNP to perform assessments of k_{eff} at different points in separation processes that have been identified by ANL-CTD. 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 UNLV students will work under the supervision of Dr. Culbreth in the Department of Mechanical Engineering and Dr. Pang in the Department of Physics.

The proposed work will begin this fall by training students in the use of SCALE 4.4 or MCNP. Students unfamiliar with criticality codes will take the MEG 455/655 course titled: "Fundamentals of Nuclear Engineering," this fall semester at UNLV where KENO is introduced. The initial work on the project will also include:

1. Review of the literature and standards on criticality safety during fuel separation and processing of actinides.
2. Schedule visits for UNLV students and faculty to ANL-East to acquire information on the separation process, to review the steps in the process that may lead to criticality problems, and to learn the use of the CMT Excel program used to simulate the separation process.

Dr. Laidler and Dr. Vandegrift at ANL have sent data for a hypothetical batch-type pyrochemical process. The configuration of TRU, FP, and molten salt will be analyzed to ensure criticality safety according to the following plan:

1. SCALE 4.4/KENO VI input files will be prepared based on the isotopic composition of the transuranic material (TRU), the fission products (FP), and the molten process salt (PS). The geometry of the process vessel is in the shape of a cylinder with a thin 316 SS wall.
2. Conduct parametric studies of k_{eff} as a function of the process vessel aspect ratio and the ratio of (TRU + FP) to process salt. The maximum mass of TRU allowable per batch will be computed. 10 different values of each independent variable will be used.
3. Compute the variation of k_{eff} with fission product concentration in the process mixture.
4. Compute the effect of the key neutron absorbers within the fission products on k_{eff}. A plot of the key neutron absorbers among the fission products has already been computed as shown in Figure 2.
5. Report the results of the criticality runs through tables and curvefits of k_{eff}.
6. Work with the ANL-CMT scientists to provide additional criticality data as required for their work on the separation processes for ATW.
7. ANL-CMT has developed a Microsoft Excel program to model the separation processes. UNLV will work with ANL-CMT to incorporate criticality safety predictions into their Excel code. Excel or Visual Basic programs will be written to spawn KENO or MCNP runs based on the process mix and the vessel geometry and composition. The KENO or MCNP runs will return k_{eff} to the Excel simulation to ensure that the geometry and composition are allowable.

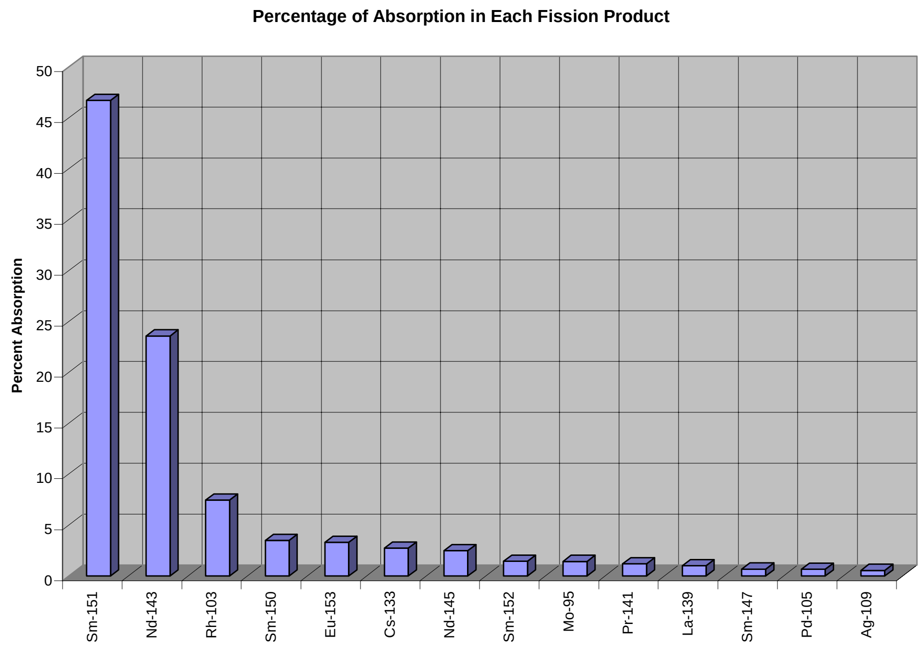

Figure 2. Strong Neutron Absorbers within the Fission Products

Background and Rationale

<u>Fuel Separation Processes</u>

The aqueous UREX process has been proposed for the separation of used light water reactor (LWR) fuel for AAA use. The UREX process is similar to the PUREX solvent extraction process to separate uranium, iodine, technetium, and transuranics (TRU) in commercial spent fuel. The transuranics include neptunium, americium, plutonium, and curium. As shown in figure 1, UREX raffinate contains TRU's as well as various fission products (FP). The fission products contain strong neutron absorbers, including samarium and europium, that help to suppress the ability of the TRU's to sustain a chain reaction. To prepare fuel for a transmuter, a pyrochemical process, PYRO-A, is used to partition the transuranics from the fission products. PYRO-A produces separated transuranics in molten salts. The mass of TRU that can be accommodated in the UREX raffinate and in the partitioned PYRO-A product must be limited to prevent the chance of a criticality event. A third pyrochemical process, PYRO-B, is under development to treat transmuter wastes. In PYRO-B, transuranics will again be separated from iodine and technetium for recycling.

Criticality during processing depends on the relative concentrations of constituents (process salt, uranium, TRU, and fission products) and on fuel geometry (container size, shape) and process temperature. These characteristics can be represented in nuclear criticality codes that can then be used to perform parametric analyses of the processes.

Nuclear Criticality Codes

The SCALE 4.4 code from ORNL and/or MCNP from LANL will be used to assess the effective neutron multiplication factor, k_{eff}. Criticality safety can be assured by maintaining subcritical ($k_{eff} < 1$) quantities of TRU in any vessel or pipe during reprocessing. Due to uncertainties in the computer codes used to model criticality, the NRC requires a conservative factor of $k_{eff} < 0.95$.

KENO VI within SCALE and MCNP are both Monte Carlo codes that simulate neutron trajectories through nuclear fuel, matrix material, and support structures. Neutron absorption, scattering, and fission are modeled in the code. A statistical distribution of neutron histories are used to assess the ratio of the number of neutrons in each generation to the previous generation. This ratio represents k_{eff}. Through previous work on the DOE Yucca Mountain Project, Dr. Culbreth and his students have acquired considerable experience in the use of KENO and SCALE to model criticality events in spent fuel containers and in geologic formations[3-8].

Both Monte Carlo codes require a complete description of the geometry of the material containing the fissionable material and matrix material. This includes the geometry of structural steel and container walls that can reflect neutrons and any surrounding material that can moderate or thermalize neutrons produced by fission. The concentration of TRU, fission products, and process salts will need to be defined in the input files for the computer codes. Finally, the number density of each element or radioisotope within the TRU, FP, and salts will be defined. Monte Carlo codes are compute-intensive and can require a great deal of computer time for complex geometries. Fortunately, high-speed workstations are now available to accommodate KENO and MCNP. Both codes are available on UNIX platforms and for the Microsoft Windows operating system. The output of the codes includes k_{eff} and the distribution of neutron flux within fuel, matrix material (process salt), and the surrounding structure. If required, ORIGIN-S or similar software can be used to predict the production rate of fission products during a criticality event within process raffinate.

The Chemical Technology Division at ANL has been preparing a Microsoft Excel simulation of the separation process for process design. As the pyrochemical treatment processes are developed for AAA, small-scale demonstrations of each process will need to be scaled up to the final prototype. Criticality concerns will likely limit the amount of TRU that can be handled at each stage of the process and will be an important component of the design. We propose involving students in the development of computer codes to integrate criticality assessment into the ANL Excel simulation code. Through the use of Excel, Visual Basic, and KENO VI or MCNP, the Excel code can be modified to use k_{eff} as a constraint during simulations of possible pyrochemical processes.

Laboratory and University Contacts

The proposed project relies on the collaboration of two UNLV faculty as co-principal investigators: Dr. William Culbreth from the Department of Mechanical Engineering and Dr. Tao Pang from the Department of Physics.

Dr. Culbreth received his B.S. in Physics from the California State Polytechnic University, his M.S. in Nuclear Engineering and Ph.D. in Mechanical Engineering from the University of California, Santa Barbara. From 1981 to 1985, he taught nuclear engineering and fluid mechanics courses at the U.S. Naval Postgraduate School and spent several summers at the Naval Research Laboratory in Washington, D.C. as an ONR/ASEE Research Fellow. Since 1985, he has been a faculty member at the University of Nevada, Las Vegas. He taught mechanical and nuclear engineering courses at UNLV. His research has included criticality calculations for the Yucca Mountain Project using KENO and SCALE 4.1, radiation transport studies for the Nevada Test Site, and simulations of the Oklo natural reactors.[3-8]

Dr. Tao Pang received his B.S. in Physics from Fudan University and his Ph.D. in Condensed Matter Theory from the University of Minnesota. Dr. Pang has been a faculty member of the Department of Physics at UNLV since 1991. His research interests include the theory of quantum many-body systems, molecular dynamics, and quantum Monte Carlo simulations of condensed matter systems[9-13].

The Argonne National Laboratory collaborators are Dr. George Vandegrift and Dr. James Laidler. Dr. Laidler is the Director of ANL's Chemical Technology Division and Dr. Vandegrift serves as the group leader for Separation Science and Technology. ANL is developing the technology necessary to separate technetium, iodine, and transuranic wastes from high-level spent nuclear fuel. The separation technology is needed to fabricate and fuel for accelerator-based transmuters and to recycle AAA spent fuel.

Proposed Work

Research Objectives and Goals

For the first year of the proposed research, our goal is to provide student and faculty support for CMT activities at the Argonne National Laboratory in nuclear criticality assessment. The specific tasks include:

- Train UNLV students in the use of SCALE and/or MCNP for the assessment of nuclear criticality.
- Assess neutron multiplication factors for geometries and material concentrations as defined by the collaborating team from ANL-CMT for the AAA project.
- Provide software, as required, to incorporate criticality estimates into the existing ANL Excel model of the pyrochemical treatment process to be used for AAA.

Proposals for subsequent years of support will be submitted if continued work is required by ANL-CMT.

Technical Impact

Monte Carlo simulations for criticality safety will be required during the development of the pyrochemical processes employed in the fabrication of AAA fuel. The development of new fuel separation and processing technology will be necessary if AAA is to succeed.

Research Approach

Industry standard codes (SCALE 4.4 or MCNP) will be used to carry out the nuclear criticality simulations. Support code will be written in Excel or Visual Basic to integrate Monte Carlo neutron transport codes with existing ANL-CMT code.

Expected Technical Results

The results of the research include:

- The development of criticality modeling software for integration with existing ANL-CMT code.
- k_{eff} results for specific geometries, component concentrations, and radionuclide content.
- Parametric studies of k_{eff} for selected geometries and fuel configuration.

Capabilities at the University and at Argonne National Laboratory

The proposal is for a numerical project involving the Chemical Technology Division at the Argonne National Laboratory and the UNLV Departments of Physics and Mechanical Engineering. The necessary computational resources will be available at UNLV with the acquisition of two high-speed computer workstations. If additional computing resources are required, the National Supercomputing Center for Energy and the Environment at UNLV has multiprocessor Silicon Graphics and SUN computers available for faculty and student use.

Equipment Requested for AAA User Labs

Due to the computational requirements for the proposed work, two computer workstations are requested for the students working on the project (one for the students in physics and one for mechanical engineering). The computer workstations will be equipped with microprocessors with at least 1 GHz of speed and at least 1 gigabyte of random access memory. The SCALE and MCNP software and associated cross section libraries are available from the Radiation Safety Information Computational Center at Oak Ridge National Laboratory without charge to universities.

Project Timeline

Timeline Narrative

To begin the project, students will be trained in the use of SCALE or MCNP in the fall semester. Early criticality assessments will be completed by the faculty with students doing the majority of the simulations by December. Student and faculty visits to the Argonne National Laboratory to discuss problems with ANL-CMT collaborators are scheduled for December and May/June. Code development will take place from December to the end of the project.

Task	9/01	10/01	11/01	12/01	1/02	2/02	3/02	4/02	5/02	6/02	7/02	8/02
Train Students in use of Monte Carlo Codes												
Student and Faculty Visits to ANL/CMT												
Simulation of Criticality for ANL Designs												
Integrate Criticality Codes into Excel Model												

Expected Technical Results

The results of the research will include computer simulations of criticality in the pyrochemical process used to treat AAA fuel. Computer code will also be developed to incorporate criticality predictions into existing ANL-CMT computer models.

Milestones

The project milestones include training students in the use of neutron transport codes, simulation of criticality in ANL designs, and preparation of code to enhance existing ANL code.

Deliverables

The products of the research will include quarterly reports that detail progress on the research, copies of publications, and summaries of collaboration with the national laboratory. A final annual report will be submitted by the 12th month of the project. Student work, including copies of theses, dissertations, and senior design reports will also be included with quarterly progress reports.

SCALE or MCNP simulation results will be submitted directly to the collaborators at ANL-CMT. All computer codes prepared to predict criticality within the ANL-CMT Excel pyrochemical code will be presented to the ANL collaborators, along with appropriate documentation.

Related Publications

1. U. S. Department of Energy, "A Roadmap for Developing Accelerator Transmutation of Waste (ATW) Technology," DOE/RW-0519, October 1999.

2. Duderstadt, J., and Hamilton, L., Nuclear Reactor Analysis, John Wiley & Sons, 1976.

3. Culbreth, W., and Viggato, J., “Determination of the Depth and Pressure within the Oklo Natural Reactors,” Proceedings of RPS2000, Spokane, Washington, September 2000.

4. Culbreth, W., and Steeps, L., “Nuclear Criticality at the Oklo Natural Reactors,” Proceedings of the International Conference on Nuclear Engineering, San Diego, CA, May, 1998.

5. Culbreth, W. G., and Zielinski, P. R., "Long-Term Effects of Poison and Fuel Matrix Corrosion on Criticality," Proceedings of the Fifth Annual International High-Level Radioactive Waste Management Conference, 634-641 (1994).

6. Zielinski, P. R., and Culbreth, W. G., "Calculation of k_{eff} for Vitrified Plutonium Waste Packages," Proceedings of the Fifth Annual International High-Level Radioactive Waste Management Conference, 679-683 (1994)

7. Culbreth, W. G., and Zielinski, P., "Analysis of the Criticality of a Spent Fuel Waste Package using Mathcad for Windows," Ninth ICMCM, Berkeley, CA, July 1993.

8. Culbreth, W. G., and Zielinski, P. R., "The Effect of Fuel Burnup and Dispersed Water Intrusion on the Criticality of Spent High-Level Nuclear Fuel in a Geologic Repository," Scientific Basis for Nuclear Waste Management XVII, Materials Research Society, 333, 445-454 (1993).

9. T. Pang, *An Introduction to Computational Physics*, Cambridge University Press, New York, 1997, pp 393.

10. S. Pearson, T. Pang, and C. Chen, “Critical Temperature of Trapped Hard-Sphere Bose Gases,” Physical Review A, 58, 4796 (1998).

11. S. Pearson, T. Pang, and C. Chen, “Bose_Einstein Condensation in Two Dimensions: A Quantum Monte Carlo Simulation,” Physical Review A, 58, 4811 (1998).

12. S. Pearson, T. Pang, and C. Chen, “Bose-Einstein Condensation in One-Dimensional Power-Law Traps: A Path-Integral Monte Carlo Simulation,” Physical Review A, 58, 1485 (1998).

13. T. Pang, “Properties of Ionic Hydrogen Clusters: A Quantum Monte Carlo Study,” Chemical Physics Letters, 228, 555 (1994).

Dear Bill,

I have reviewed your proposal, "Nuclear Criticality Analyses for the Transmuter Fuel Fabrication and Reprocessing Process," and find it to be an excellent complement to the work that is being done at Argonne National Laboratory on separations process development. The proposed work fills a niche that has not been included in our work to date, and is extremely important because we are entering a program phase in which the various candidate separations processes will be evaluated for efficiency, safety, reliability and cost. Criticality safety analyses will be an essential part of these evaluations and certainly an absolute mandatory requirement for process equipment and facility design activities that will begin in the not too distant future. I sincerely hope that it will be possible to support this work on a continuing basis and I speak for the Chemical Technology Division in saying that we look forward to working with you and your students.

Best regards,

Jim

James J. Laidler PhD.
Chemical Technology Division
Argonne National Laboratory
9700 South Cass Avenue
Argonne, IL 60439-4837