2002

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

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Task 11 Continuation Proposal

Project Title:

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

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AFCI Research Area: Separations

Requested Funds: $113,940 (Year Three)

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

Abstract: The remediation of nuclear waste created by conventional fission reactors will rely upon the separation of the waste products for further treatment. The UREX+ process (Figure 1) now under review will involve the use of an aqueous chemical process to separate out depleted uranium resulting in a product containing minor actinides, fission products, cesium, strontium, technetium, and iodine. The radioactive decay of strontium and cesium produces roughly half of the thermal and gamma production in spent fuel and the relatively short halflife of isotopes of both of these elements requires storage for about 300 years before heat and radiation decreases to safe levels.

A waste stream composed of plutonium and neptunium will be stored before fabrication into modified LWR fuel. The DIAMEX/SANEX process will result in a waste stream predominately composed of americium and curium. Both of these elements pose a thermal, radiation, and criticality risk during storage and handling before

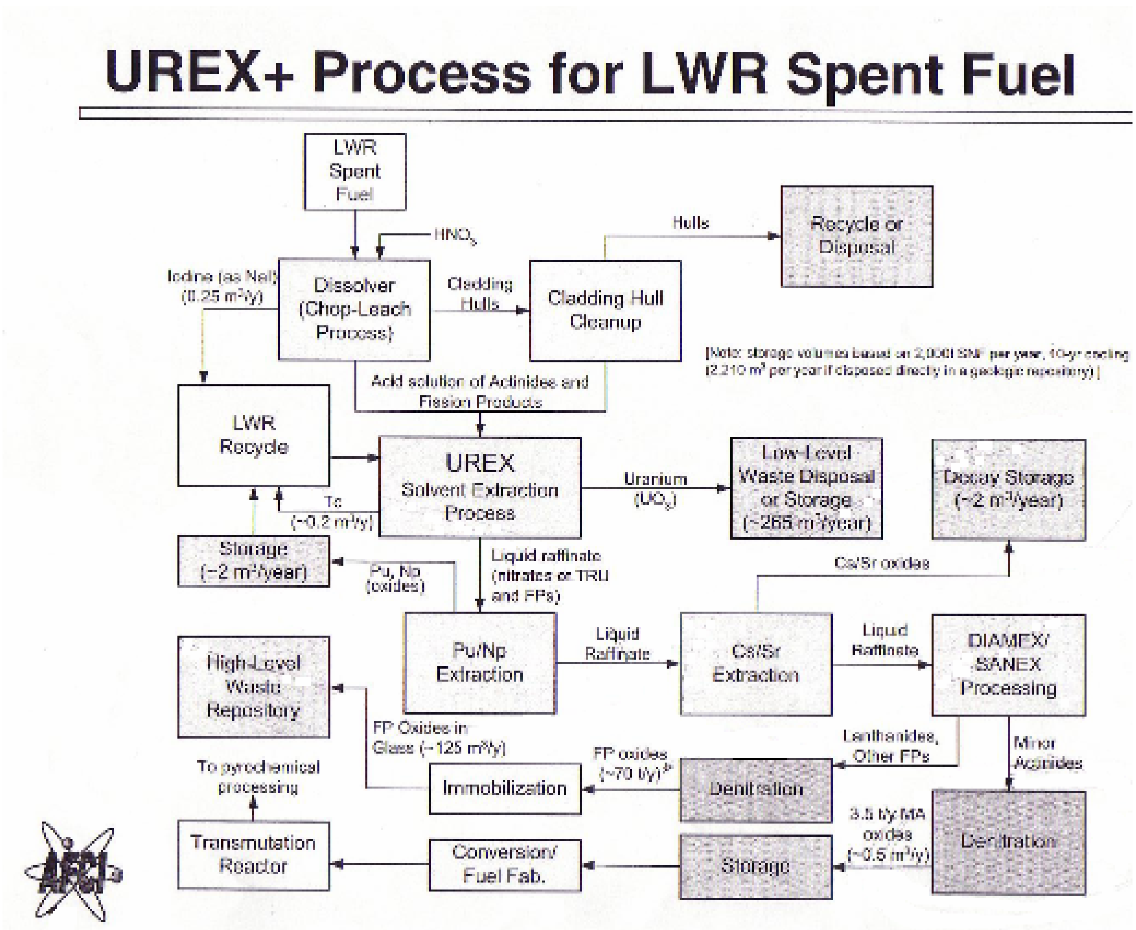


Fig. 1 Outline of the UREX+ Process for the Advanced Fuel Cycles Initiative

fabrication into a fuel suitable for a fast neutron reactor.

In the Pu/Np, and Cm/Am waste streams, products must be stored for intermediate periods of time before fuel fabrication. For the Cs/Sr waste stream, radiation shielding and thermal analyses are needed to design storage containers capable of safely isolating these radioisotopes for extended periods of time.

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. Candidate storage containers must also be analyzed to assess the need for radiation shielding. The minor actinides generate significant amounts of heat through radioactive decay and proposed storage must be designed to avoid excessive temperatures.

In the continuation of our work with ANL, we will conduct assessments of nuclear criticality, radiation transport for shielding, and thermal analyses of wastes in the Cs/Sr, Pu/Np, and Cm/Am waste streams to assist in the design of the UREX+ process. This project has been identified as a critical R&D need of the Chemical Technology Division at the Argonne National.

Work Proposed for Academic Year 2003-2004, Goals and Expected Results:

UNLV students will use 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 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_{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 and Dr. Beller in the Department of Mechanical Engineering.

1. Analyze the Cesium/Strontium Waste Stream.
2. Analyze the Plutonium/Neptunium Waste Stream.
3. Analyze the Americium/Curium Waste Stream.

The proposed work will continue this fall by training new students in the use of SCALE 4.4a and MCNPX. Students unfamiliar with criticality codes will take the MEG 455/655 course titled: "Fundamentals of Nuclear Engineering," this fall semester at UNLV where KENO and MCNP4B is introduced. We will continue work this summer with an emphasis on the cesium/strontium waste stream analysis. Analysis of Pu/Np and Am/Cm will proceed during the academic year.

Cesium/Strontium Waste Stream

The first study proposed in the timeline shown in Table 1 is based on a request by ANL to study the possibility of storing Cs/Sr in oxide or chloride form in commercial dry casks. The procedure that will be used in the study is described below.

Strontium and cesium will be separated from spent nuclear fuel and must be safely stored through approximately 10 halflives (30 years) until emitted radiation reaches safe levels. In oxide or chloride form, it is proposed that these substances be placed in commercially-available dry casks used for the above-ground storage of spent nuclear fuel. Two problems must be addressed to see if this method of storage is feasible: 1) the decay heat generated by Cs and Sr must be within the design specifications of the container, and 2) the radiation dose at the outside wall of the container must be within acceptable limits. The following procedure is proposed to validate this storage method for strontium and cesium oxides and chlorides.

Procedure

1. Generate the inventory of Cs and Sr isotopes expected from spent nuclear fuel.
    a. The design basis fuel is 50,000 MWd/MTIHM, 4.26% initial enrichment, 10 year old fuel.
    b. RADDB or ORIGEN-S may be used to determine the inventory.
    c. Assess the inventory of decay products from Cs and Sr as a function of time.
2. Obtain the design specifications for a sample dry cask (Duke Engineering, Sierra Nuclear, NUHOMS, NAC, etc.)
3. Tailor a loading of Cs_2O and SrO (or Cs and Sr chlorides) to fill a container to the same thermal load as spent fuel.
    a. Determine the maximum centerline temperature within the waste.
    b. Fill with an inert substance with good thermal conductivity (SiC) to decrease the thermal load and increase heat transfer.
    c. Ensure that the melting temperature of the Sr and Cs ceramics are not exceeded.
    d. Compute the internal temperature distribution of the Cs/Sr mix assuming an upper surface covered with argon gas.
    e. Compute the expected temperature distribution as a function of Cs/Sr mix porosity.
    f. Compute the wall temperature distribution and compare to the case of spent fuel.
4. Calculate the radiation dose on the outside wall of the container and compare to the dose expected if the container were filled with the design spent fuel.
    a. Ensure that the gamma dose outside the container is within acceptable limits.
5. Report on the following:
    a. Thermal loading.
    b. Internal temperature distribution.

c. Gamma production.
d. Combined radiation dose at the outer wall.
e. Amount of Sr/Cs that can be safely contained in a dry cask.

Plutonium/Neptunium Waste Stream

In the UREX+ process shown in figure 1, plutonium and neptunium will be separated from other components of spent fuel and will be stored in oxide form. These actinides will later be formed into mixed oxide fuel to be burned up in conventional light water reactors. The mixed Pu/Np oxides pose criticality concerns and the critical mass must be identified for varying fuel burnup, fuel age, and initial enrichment. A significant amount of decay heat is also generated within the mixed oxide and analyses of the temperature distribution in proposed storage containers will be conducted to ensure safe storage. Radiation shielding analyses will also be conducted on the containers, if necessary.

Americium/Curium Waste Stream

The DIAMEX/SANEX process will result in the separation of americium, curium, and other minor actinides (up to berkelium) that pose criticality and thermal concerns. These actinides will be fabricated into an oxide form and ultimately will be made into fuel to be burned up in a fast neutron spectrum. On this project, we will investigate critical mass, shielding, and heat transfer concerns for proposed storage container designs.

Funding Profile:

Academic Year:	2001-2003	2003-2004
Total (K$)	$110,123	$113,940

Background and Rationale:

Fuel Separation Processes

The aqueous UREX+ process has been proposed for the separation of used light water reactor (LWR) fuel for transmutation and remediation. 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 possible burnup in LWR's or a fast reactor, various chemical processes are used to partition the transuranics from the fission products. This results in waste streams of separated Pu/Np and Am/Cm. Strontium and cesium are also both separated from other fission products for further treatment or storage.

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. All waste streams involve highly radioactive material that requires radiation shielding for safety. Radioactive decay also results in the generation of heat that can lead to melting of the fuel during storage and handling. Safety concerns associated with criticality, shielding, and heat buildup must be addressed as the UREX+ process is further developed.

Nuclear Criticality and Radiation Transport 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 radiation transport 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[5-9].

Laboratory and University Contacts

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

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.[6-11]

Dr. Denis Beller recently joined the UNLV Mechanical Engineering Department as a Research Professor. He is a former employee of the Los Alamos National Laboratory

and he has extensive experience with the AFCI (and former AAA) programs. Dr. Beller has his Ph.D. in Nuclear Engineering and will be working with the students on criticality and shielding assessments using MCNPX.

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 ATW spent fuel.

Proposed Work

Research Objectives and Goals:

The goal of the proposed continuation project is to assist ANL in the development of the UREX+ process by providing assessments of criticality, radiation shielding requirements, and thermal problems for the following waste streams:

- Cs/Sr
- Pu/Np
- Am/Cm

The objectives of the proposed work include:

- Educate students in the use of RADDB, MCNPX, SCALE 4.4a, and specialized heat transfer software to support criticality, shielding, and thermal assessments.
- Train students in the rudiments of nuclear engineering. This includes metriculating new students through the UNLV nuclear engineering course, MEG 455/655.
- Analyze each waste stream to determine keff, radiation levels, and temperature distributions during handling and storage.
- Analyze the buildup of new radionuclides in separated wastes as a result of radioactive decay.
- Create reports on the assessment for each waste stream for submission to ANL.

Technical Impact:

Development of the UREX+ process will have long-term advantages in the nation's effort to transmute nuclear waste into short halflived radioisotopes. The proposed research work will involve criticality assessments, radiation shielding analyses, and heat transfer studies necessary to design the waste separation, handling, and storage processes for the three waste streams.

Research Approach:

Industry standard codes (SCALE 4.4 or MCNP) will be used to carry out the nuclear criticality simulations. UNLV will write finite difference codes to assess heat transfer in proposed waste containers or utilize commercial codes. All work will be conducted in collaboration with ANL/CMT.

Expected Technical Results:

The results of the research include:

- Radiation shielding assessments for each waste stream, including estimation of dose and recommended shielding thicknesses.
- k_{eff} results for specific geometries, component concentrations, and radionuclide content.
- Temperature distributions in waste storage containers for each waste stream.

Fig. 2 Nevada Radiation Computational Cluster at UNLV

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 Mechanical Engineering. The necessary computational resources will be available at UNLV including use of our Nevada Radiation Computational Cluster composed of 20 parallel computers for the use of MCNPX. 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 AFCI User Labs:

Due to the computational requirements for the proposed work, one additional computer workstation is requested for the students working on the project. The project purchased two 1.8 GHz computers in 2001 for student work. The new computer will be used for students to run MCNPX, SCALE, and MathCad for thermal analyses. Additional software will be purchased for heat transfer analyses. The SCALE and MCNPX software and associated cross section libraries are available from the Radiation Sciences Information computing Center at Oak Ridge National Laboratory without

charge to universities. UNLV is also a member of the beta test team for MCNPX through the Los Alamos National Laboratory.

Project Timeline:

Timeline Narrative:

Elizabeth Bakker is trained in the use of two nuclear criticality and radiation transport codes, SCALE 4.4a and MCNPX. She will assist in the training of other students early in the project. The timeline is shown in figure 1 and will start with studies of thermal and shielding problems in the storage of cesium/strontium mixtures. Topics to follow include Np/Pu and Am/Cm waste streams. An initial report on Cs/Sr will be forwarded to ANL early in the summer of 2003 to assist them in decision-making regarding the separations process. Some flexibility in the timeline should be allowed to accommodate requests for additional data from ANL.

Table 1 Timeline for the Proposed Project Continuation

Task	2003					2004						
	Aug	Sep	Oct	Nov	Dec	Jan	Feb	Mar	Apr	May	Jun	Jul
A. Cesium/Strontium Waste Stream												
1. Research above-ground container design												
2. Prepare initial model results												
3. Develop database of Cs,Sr properties												
4. Develop thermal model												
5. Develop shielding MCNPX model												
6. Write final report												
B. Plutonium/Americium Waste Stream												
1. Develop database for Pu/Am and oxides												
2. Prepare thermal model												
3. Prepare shielding model												
4. Prepare criticality model												
5. Write final report												
C. Neptunium/Curium Waste Stream												
1. Develop database for Np/Cm and oxides												
2. Prepare thermal model												
3. Prepare shielding model												
4. Prepare criticality model												
5. Write final report												

Expected Technical Results:

The results of the research will include criticality, shielding, and thermal analyses of separated radioactive wastes in the UREX+ process used to treat spent nuclear fuel. Results to be reported will be incorporated into graphs and tables to assist designers in the fuel separation process. Studies will include problems that may occur in the handling,

storage, and long-term storage of radioactive wastes that generate significant radiation and decay heat.

Milestones:

Milestones are indicated as completion of items on the timeline. These include reports on predicted shielding requirements, critical masses of actinides, and temperature buildup due to decay heat generation.

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 12^{th} 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 MCNPX simulation results will be submitted directly to the collaborators at ANL-CMT. All computer codes prepared to predict criticality within the ANL-CMT Excel or MathCad codes will be presented to the ANL collaborators, along with appropriate documentation.

Review of Work in 2001/2003

1. Elizabeth Bakker, an undergraduate mechanical engineering student, completed her Bachelor of Science degree in May 2002. She worked extensively on this project. She is now a graduate student working on a thesis based on criticality assessments in separated actinides.
2. In the Spring of 2002, Elizabeth and Danny Lowe attended an MCNPX course taught on the UNLV campus by Laurie Waters and her colleagues. MCNPX was used for some of the criticality work on the project and is now in use for shielding assessments in Cs/Sr wastes.
3. In November, 2001 Elizabeth, Danny, and Jason Viggato, a doctoral student in mechanical engineering, presented

Calculation of k-effective for a Cylindrical Container with TRU and Process Salt

Bill Culbreth
University of Nevada, Las Vegas

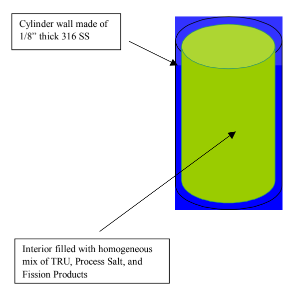
This program calculates the effective neutron multiplication factor, k-effective, for a cylindrical container with a 1/8 inch thick wall made from 316 SS. The mixture inside the container is assumed to be composed of TRU with Process Salt. The nominal diameter may be varied, and the height will be computed to preserve the original container volume. SCALE 4.4a was used to generate the results. A least-squares regression fit o fthe data was used to estimate k-effective as a function of cylinder diameter and %TRU.

A. Input

Please change the two values included below to recalculate the value of k-effective:

Enter the Cylinder Diameter (cm):	50
Enter the percent (%) TRU:	40

Nominal Volume (cm^3) is:	147262.15
The Resulting Container Height (cm) is:	75



B. Results

k-eff:	**1.403**
Standard Error:	3.05E-002
Maximum Error:	5.82E-002

Maximum KENO Uncertainty:	0.0031	
Maximum Possible Value of k-eff:	1.464	
Limit of k-eff for Safety:	0.95	
Is this Safe from a Criticality Event?	SAFE	**UNSAFE**

C. Calculations

Order	Coefficient	I	J	
0	-9.78E-002	0	0	-0.09783378
1	0.6884337	0	1	0.27537349
2	-0.0522729	0	2	-0.00836366
3	0.6726148	1	0	0.48043915
4	2.8150107	1	1	0.80428877
5	-2.1761193	1	2	-0.24869935
6	-0.507453	2	0	-0.25890461
7	-2.0879773	2	1	-0.42611782
8	1.6187672	2	2	0.13214426
			Sum:	0.65232646
			k-eff:	1.40289329

Fig. 3 Example of Criticality Assessment For ANL Container in Year 1 Work

papers related to this project at the ANS Student Conference held in Reno, Nevada. Jason won "best presentation" award for his section.

4. In January, 2002, Dr. Culbreth and the students visited Drs. Laidler and Vandergrift at ANL in Chicago.
5. Several reports and summaries were presented to Dr. Laidler based on work assessing criticality and thermal concerns in curium, americium, plutonium, and in the molten salt process[2-5].
6. Elizabeth Bakker accepted a summer appointment at ANL in Chicago with Dr. Laidler to expedite the research. She is also partially supported from the project this summer to continue the initial analysis of Cs/Sr storage.

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11. 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).

Copy of Letter from Dr. Laidler Regarding the Initial Proposal

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.
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