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BENCHMARK CHARACTERIZATION FOR THE DILUTED HIGHLY ENRICHED URANIUM EXPERIMENTS WITH WASTE MATRIX MATERIALS

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ABSTRACT

Critical experiments are carried out in order to validate, improve, and benchmark the extensive data calculations available. A series of such experiments was performed at the Los Alamos Criticality Experiments Facility (LACEF). These experiments were performed to provide criticality safety data for waste matrix materials. These critical experiments were fueled with highly enriched uranium (HEU), moderated with polyethylene, and mixed with silicon dioxide (SiO_2), aluminum (Al), magnesium oxide (MgO) and gadolinium (Gd). The uncertainties affecting the experiment were divided into three broad categories: mass measurement, geometry, and material composition. Each category is considered in turn and then the total experimental uncertainty is derived. These uncertainties were calculated using neutronic codes (MCNP, KENO, DANTSYS). All four experiments had a measured k_{eff} of 1.000. The benchmark of these critical experiments yielded uncertainties in the measured k_{eff} of ± 0.0023 for SiO_2, ± 0.0024 for Al, ± 0.0026 for MgO, and ± 0.0024 for Gd. These experiments were judged to be of benchmark quality.

1. INTRODUCTION

Critical experiments fueled with highly enriched uranium (HEU), moderated with polyethylene, and mixed with silicon oxide (SiO_2), aluminum (Al), magnesium oxide (MgO) and gadolinium (Gd) were performed at the Los Alamos Critical Experiments Facility. The objective of these experiments was to provide additional criticality data to support the National Spent Fuel Program. This paper examines the uncertainties associated with these critical experiments. A more detailed description has been prepared to be included in the International Handbook of Evaluated Criticality Safety Benchmark Experiments.[1]

This set of experiments approximates the postulated worst-case criticality scenario for a long-term geologic repository involving high-level waste, primarily spent nuclear fuel. It has been postulated that over long periods of time the fissile material could possibly be concentrated into thin slabs of metal by the presence of organic material. Interstitial slabs of water and other matrix materials would separate the thin slabs of fissile material. The matrix materials could either be present in the repository (SiO_2), or introduced as part of a high level waste stream (Al or MgO), or added to increase criticality safety margins (Gd). The HEU foils experimentally approximate the organically concentrated thin slabs of fissile material. The polyethylene slabs experimentally approximate the effect of interstitial water. The matrix materials are examined separately in the four different experimental configurations.

This paper looks at the exact content of the assembly, the mass and compositions of the constituents, and the geometry uncertainties (tolerances). It provides realistic uncertainties and a confidence level useful to users working in code qualification. The benefit of the benchmarks is to provide data or information to help qualify codes and cross sections used in criticality assessments. In addition, when the benchmark calculations and models used in the validation of the analysis have realistic uncertainties, the biases and associated uncertainties will best reflect the true accuracy of the criticality calculations. Knowledge of the uncertainties provides a solid foundation for setting appropriate safety margins.

2. DESCRIPTION OF THE EXPERIMENT

These four experiments were performed using the Planet universal critical assembly at the Los Alamos Critical Experiments Facility. The experiment consisted of placing HEU foils interspersed with the waste matrix materials (SiO_2 plates, Al plates, MgO powder or Gd plates) in a column stack. The uranium foils were moderated and reflected by polyethylene square plates. A unit consisted of one polyethylene plate with a recess in its top side that contained the waste material being examined and a smaller recess in its lower side for two HEU foils.

The experimental arrangement is depicted in Figure 1. As Figure 1 illustrates, the stack is divided into two parts. The bottom half of the stack rests on an aluminum support plate that is 1-inch thick. The top half of the experiment rests on 0.75-inch thick polyethylene plate. Criticality is achieved by decreasing the gap between the top and bottom portions of the stack. To disassemble the configuration, the bottom stack is dropped to its initial position. There are no other control or safety rods inside the assembly.[2]

Figure 1. The HEU waste matrix experiments mounted on the Planet assembly.

3. DESCRIPTION OF MATERIAL DATA

The moderator and reflector for this experiment were constructed from high-density polyethylene. The average density of these plates was 0.961 g/cm^3. The dimensions for these moderating and reflector plates were 39.116 by 39.116 cm in length and width. The thickness of the moderating plates was 1.905 cm, and the thickness of the reflector plate was 2.54 cm. The Highly Enriched Uranium (93.23 wt%) foils used in these experiments were 22.86 by 22.86 cm square and 0.00762-cm-thick before they were laminated. The lamination material was polyethylene. The final laminated foils had dimensions of approximately 25.4 by 25.4 cm and were 0.02286-cm-thick.

The HEU-SiO_2 experiment was critical with 15 units. Each unit is composed of a polyethylene moderating plate with the waste material embedded in the recess of the plate and two highly enriched uranium foils laminated in polyethylene. The HEU-Al experiment needed 18 units to become critical. The total mass of the Al was 16.6 kg. The HEU-MgO experiment consisted of 20 repeating units, and the total MgO mass in this experiment was 9.453 kg. The HEU-Gd experiment consisted of 14 repeating units, and the total Gd mass was 111.6 g. All the experiments were reflected by 4 inches of top polyethylene at the top and bottom. The critical data for the benchmark modeling is presented in Table I. Figure 2 illustrates the four materials used for the waste matrix experiments.

Table I. Material Data for the Waste Matrix Materials.

Material	Dimensions (inches)	Density (g/cm^3)	Mass (g)	Composition (%)
SiO_2	9 x 9 x 0.25	2.21	372.15	98.9
Al	9 x 9 x 0.25	2.782	923.44	97.38
MgO	9.01x9.01x0.265	1.397	472.685	98.38
Gd	2 x 2 x 0.015	8.107	7.971	99.887
Moderator	15.4x15.4x0.75	0.961	2421.13	99.999
Reflector	15.4 x 15.4 x 1.0	0.959	3723.71	99.999
Bare Foil	9 x 9 x 0.003	17.438	69.438	93.23 in ^{235}U
Lamination	10 x 10 x 0.003	1.081	11.634	99.999

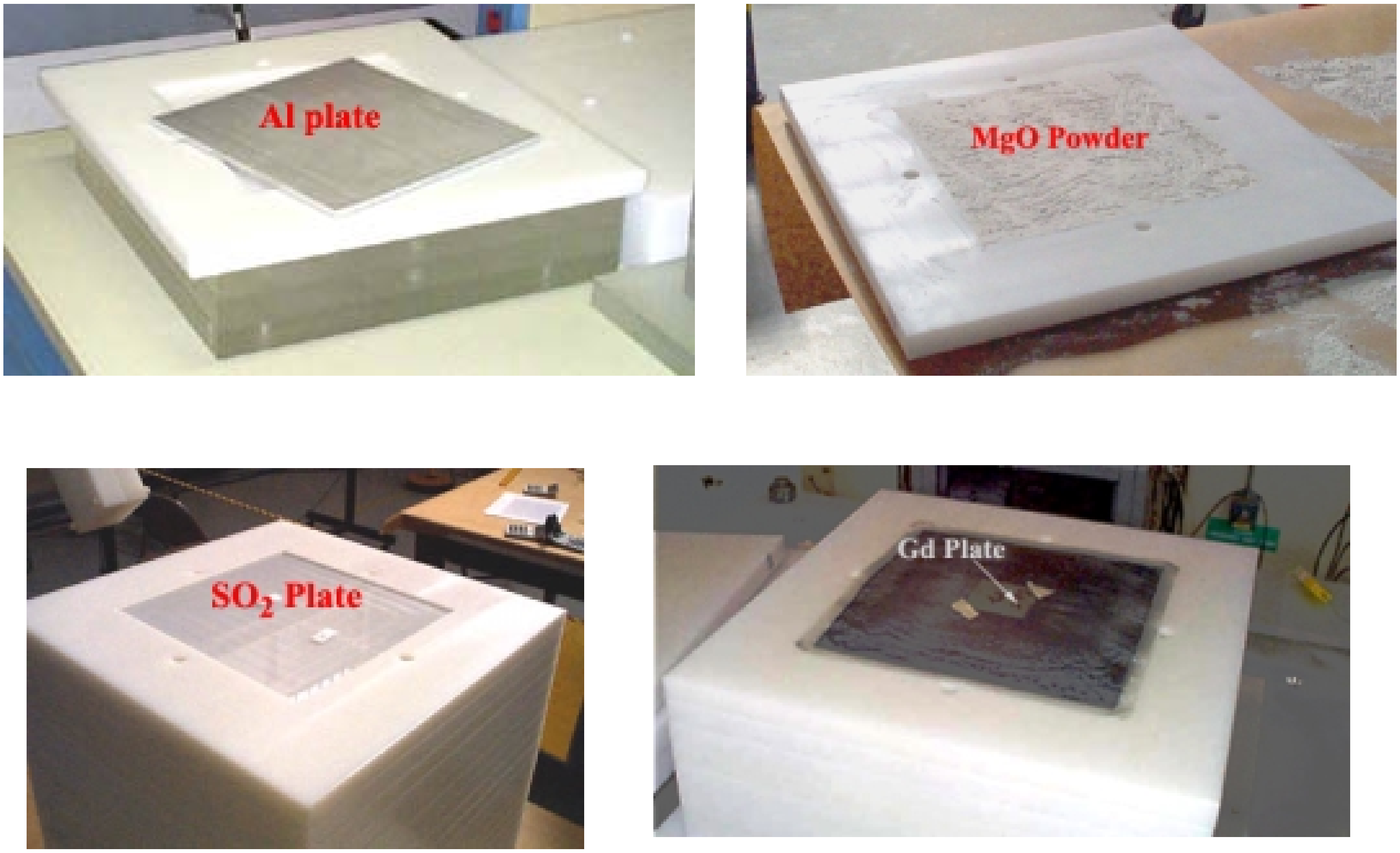

Figure 2. Waste Matrix Materials Embedded in the Polyethylene Plates.

4. METHODOLOGY

The reactivity effects of many of the uncertainties discussed below were quantified using a MCNP model and a DANTSYS model of the benchmark. The MCNP analysis was performed by employing a detailed three-dimensional model with continuous-energy cross sections from ENDF/B-VI neutron data. The MCNP calculations had 6,000,000 active histories. A total of 5,000 histories per generation was used and 1,250 generations of neutrons. The first 50 generations were skipped to obtain a well-distributed neutron source. DANTSYS three-dimensional and one-dimensional (infinite slab geometry) codes were used with the Hansen-Roach 16-group cross section library. The calculations with the THREEDANT code were performed with the quadrature varying from S_2 to S_8, P_0 scattering, and the convergence criterion set to 1 x10^{-3}. However, the use of the THREEDANT code was limited due to the size of the problems. The problems tended to exceed the large core memory available.

The individual effect of the parameter being analyzed on the k_{eff} of the system was done by varying one parameter at a time. First a reference k was obtained, $k_{eff}(r)$, using the reference values of the experiment. Then a parameter, r_i, is perturbed while all other parameters are kept at their reference value, and a new k is calculated based on the perturbation. The change in k (Δk_{eff}) is then calculate for ± the standard uncertainty (S.U.). Thus the change in k_{eff} is defined as:

$$\Delta k_{eff} = \frac{\left[\left| k_{eff}(r) - k_{eff}(r + S.U.) \right| + \left| k_{eff}(r - S.U.) - k_{eff}(r) \right| \right]}{2} \qquad (1)$$

Where $k_{eff}(r)$ is the reference case, k_{eff} (r + S.U.) is the perturbed case in the positive direction of the standard uncertainty, and k_{eff} (r - S.U.) is the perturbed case in the negative direction.

The type of uncertainties examined in the benchmarks were of type A or type B. Type A uncertainties refer to observations based on a finite number of measurements. Therefore, a type A evaluation is defined as the experimental standard deviation of the mean. The experimental standard deviation of the mean is defined as the positive square root of the variance, s(ā), divided by the square root of the number of measurements, namely s(ā)/√n.

The type B evaluation of uncertainty refers to uncertainties other than statistical analysis. It is a scientific judgment based on all the information available. When a parameter is given as p ± Δp, such is the case in tolerances, then the ± Δp appears to refer to the upper or lower bound of the quantity. In such cases the standard uncertainty is obtained by $\Delta p/3$, if the distribution is normal, $\Delta p/\sqrt{3}$, if the value is equally probable everywhere within the interval, or $\Delta p/\sqrt{6}$, if the value follows a triangular distribution.

5. BENCHMARK OF THE EXPERIMENT

The uncertainties affecting the experiment have been divided into three broad categories. They are: 1) mass measurement, 2) geometry, and 3) material composition. Each category is considered in turn and then the combined experimental uncertainty is presented. Each uncertainty estimate is one standard deviation.

The first category includes the material mass uncertainty, which is calculated by changes in density. The uncertainties in the mass of the fuel, in the mass of the waste matrix material (SiO_2, Al, MgO and Gd), in the mass of the polyethylene moderator plates, and in the mass of the lamination were considered. The uncertainty in the ^{235}U enrichment was also investigated under this category. The second category includes the geometry uncertainties of the different components. The geometry uncertainties examined include the change in volume of the waste matrix material plates, the moderator plates, the fuel, the lamination, and the filling of the magnesium oxide powder. The third category examined was the uncertainty in the material composition and impurities. Other uncertainties analyzed were room-return neutrons.

Each uncertainty estimate is one standard deviation. A summary of the uncertainties is collected in Table II.[3,4]

Table II. Summary of Uncertainties

Source of Uncertainty	**Standard Uncertainty in Δk_{eff} for *SiO_2***	**Standard Uncertainty in Δk_{eff} for *Al***	**Standard Uncertainty in Δk_{eff} for *MgO***	**Standard Uncertainty in Δk_{eff} *Gd***
Material Mass				
HEU Mass	± 0.0008	± 0.0007	± 0.0006	± 0.0011

Enrichment in ^{235}U (wt %)	± 0.0002	± 0.0001	± 0.0002	± 0.0001
Polyethylene Plate Mass	± 0.0011	± 0.0004	± 0.0002	± 0.0003
Polyethylene Lamination Mass	± 0.0010	± 0.0001	± 0.0001	± 0.0004
Material Plate Mass	± 0.0001	± 0.0008	± 0.0005	± 0.0008
Geometry Dimensions				
HEU	± 0.0003	± 0.0002	± 0.0001	± 0.0003
Polyethylene Plates	± 0.0011	± 0.0006	± 0.0012	± 0.0004
Polyethylene Large Plate 48	± 0.0002	± 0.0001	± 0.0002	± 0.0001
Polyethylene Lamination Mass	± 0.0001	± 0.0002	± 0.0003	± 0.0002
Material	± 0.0001	± 0.0003	± 0.0002	± 0.0019
Axial Air Gap	± 0.0036	± 0.0027	± 0.0016	± 0.0020
Material Composition				
Impurity in Lamination	- 0.0001	- 0.0001	- 0.0006	- 0.0004
Impurity in Polyethylene Plates	- 0.0002	- 0.0002	- 0.0005	- 0.0003
Composition of Material	- 0.0001	+ 0.0007	+ 0.0006	+ 0.0002
Additional Calculations				
Support plates and Room return	<0.0010[a]	<0.0010[a]	<0.0010[a]	<0.0010[a]
Total Uncertainty Quadratic Total:	± 0.0042	± 0.0027	± 0.0026	± 0.0024

(a) 0.0010 is estimated as the standard uncertainty from support plates and room return.

6. CONCLUSION

Four critical experiments were performed at the Los Alamos Critical Experiments Facility to understand the characteristics of waste matrix materials. These matrix materials could either be present in the repository (SiO_2), or be introduced as part of a high level waste stream (Al or MgO), or added to increase criticality safety margins (Gd). The HEU foils experimentally approximate the concentrated thin slabs of fissile material, and the polyethylene slabs experimentally approximate the effect of interstitial water. The matrix materials are examined separately in the four different experimental configurations. These four waste matrix materials analyzed were SiO_2, Al, MgO and Gd.

Models were prepared for these experiments in order to assess whether these experiments were of benchmark quality.

All sensitivity calculations were performed using MCNP and DANTSYS with ENDF/B-VI or Hansen-Roach cross section libraries. The uncertainty analysis was prepared following the ICSBP guidelines. The uncertainties in the expected value of k_{eff} of the benchmark models arise from neutron return from surroundings, uncertainties in material measurements (primarily HEU and polyethylene density, mass, and composition), machining tolerances of components, and others. Individually, the effects are small, and taken together they may be compensating. These critical experiments are acceptable as benchmarks.

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8. REFERENCES

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