Analysis of Waste Matrix Material Experiments Mixed with Highly Enriched Uranium on the Thermal Energy Region

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The basic characteristics of waste materials such as silicon dioxide, aluminum and iron fueled with highly enriched uranium and moderated and reflected by polyethylene were investigated. These critical mass experiments were performed at the Los Alamos Criticality Experiments Facility (LACEF) on the Planet critical assembly. The primary intention of these experiments is to provide supplementary data that can be used to validate and improve criticality data for the Yucca Mountain and the Hanford Storage Waste Tanks Projects. The secondary intention of these experiments is to reduce the H/U ratio and increase the waste material/U ratio from previously published experiments. These experiments were designed to supply data for interlaced waste material/Fuel/Moderator systems on the thermal region. The experiments contained silicon dioxide (SiO_2), aluminum (Al) and iron (Fe) mixed with 93.23% enriched uranium and moderated and reflected by polyethylene. A base case experiment was also performed with polyethylene-only. This analysis systematically examines uncertainties associated with the critical experiments as they affect the calculated multiplication factor. The systematic analysis is separated into uncertainties due to mass measurements, uncertainties due to fabrication and uncertainties due to composition. Each type of uncertainty is analyzed individually and a total combined uncertainty is derived. The SiO_2-HEU experiment had a measured k_{eff} of 0.993, the Al-HEU experiment had a measured k_{eff} of 0.990, the Fe-HEU had a measured k_{eff} of 1.000 and the polyethylene-HEU had a measured k_{eff} of 1.0025. The calculated k_{eff} values tend to agree well with the experimental values. The sensitivity analysis of these critical experiments yielded a total combined uncertainty on the measured k_{eff} of ± 0.0024 for SiO_2, of ± 0.0028 for Al, of ± 0.0026 for Fe, of ± 0.0020 for polyethylene.

KEYWORDS: aluminum, assembly, critical mass, HEU, iron, polyethylene, silicon dioxide.

1. Introduction

Many theoretical studies have been performed to study whether fissile uranium from spent fuel or from low-level waste facilities can be concentrated through hydrochemical processes to permit nuclear criticality. Long after closure of geological repositories for fissile material, geological processes may change disposal geometries and spacing, and water may enter the repository. One preliminary study suggested that concentrations of special nuclear material in geometries that constitute nuclear criticality safety concerns are plausible. Therefore, it was determined that further analysis was needed to determine the geological process that might lead to potential nuclear criticality. Neutron moderation and reflection by water was assumed in the analysis, since water exclusion could not be guaranteed for millions of years. For this to occur water would be required to enter the repository, gain access to the fissile material, and then dissolve, transport and selectively deposit the fissile material in a location of significant quantity and concentration. Consequently, if by some chance this were to occur, criticality could potentially result. Other variables that were part of the analysis included composition of soil (SiO_2), aluminum canisters and iron scraps.[1]

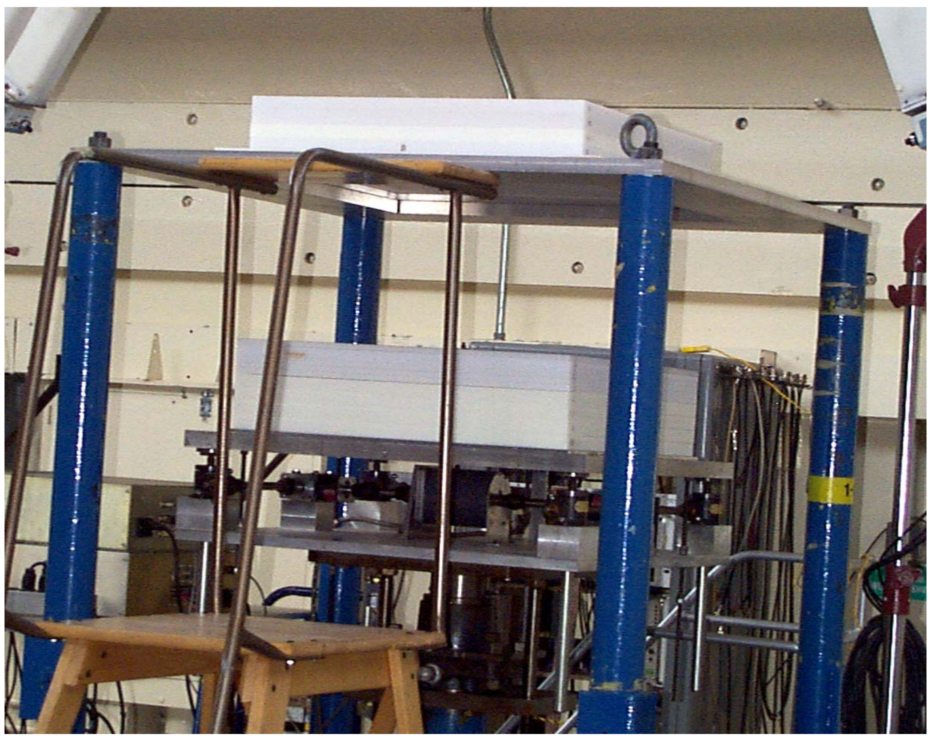
A series of experiments were designed and performed at the Los Alamos Critical Experiment Facility to approximate the aforementioned worst-case criticality scenario for a long-term geologic repository involving high-level waste, primarily spent nuclear fuel. These experiments performed included some of the waste materials (SiO_2, Al and Fe),[2,3,4] and one base case experiment where the waste matrix material was replaced by polyethylene.[5] This experiment is referred as the polyethylene-only experiment. Highly Enriched Uranium (HEU) foils were used to experimentally approximate the uniformly distributed fissile material and to simulate a homogenous solution system. A hydrogenous moderating material in the form of polyethylene slabs experimentally approximated the effect of flooding. These experiments were performed to provide validation data which could be of interest to those investigating criticality scenarios for geologic repositories or criticality scenarios. These experiments were of high quality and were documented according to International Criticality Safety Benchmark Evaluation Project (ICSBEP) guidelines.

Three factors are important in the analysis of waste materials. The ability of the waste to either absorb neutrons (Fe), the ability to scatter neutrons (SiO_2 and Al) and the ability to moderated neutrons (polyethylene). Since cross sections vary as a function of the neutron energy, criticality analyses in the postulated flooding scenarios required data for thermal systems. Thus this set of experiments was performed on the thermal region for the postulated wet condition scenario. Under these wet conditions, uranium can be potentially transported with the advecting fluids and admixed in the soil (SiO_2) with the waste materials (Al and Fe). Regrettably, few experiments satisfactorily represent the characteristics of waste materials with ^{235}U. Hence, the ability of the codes and data libraries to predict the correct k_{eff} for these systems is not entirely known. The lack of experimental data has left nuclear criticality analysts to rely almost solely on the results provided by neutronic codes.[6]

2. Description Of The Experiment

The experiments were performed using the Planet Critical assembly at the Los Alamos Critical Experiments Facility (LACEF). The Planet Critical assembly is a vertical lift machine residing in CASA I (Critical Assembly and Storage Area). The Planet Critical assembly consists of a movable platform powered by a hydraulic lift and jack screws. The maximum speed of the movable platform is adjustable for each experiment to limit the insertion rate of reactivity to less than 0.05 $/sec. To disassemble the configuration, the movable platform is dropped to its initial position. Figure 1 shows a picture of the critical assembly and experimental arrangement.

Figure 1: Planet Critical Assembly with 2x2 Waste Matrix Experiment.



The experiments consisted of placing HEU foils interspersed with the waste materials (SiO_2 plates, Al plates or Fe plates) in a short and fat 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 four waste material plates being examined and four HEU foils on top of the waste material plates.

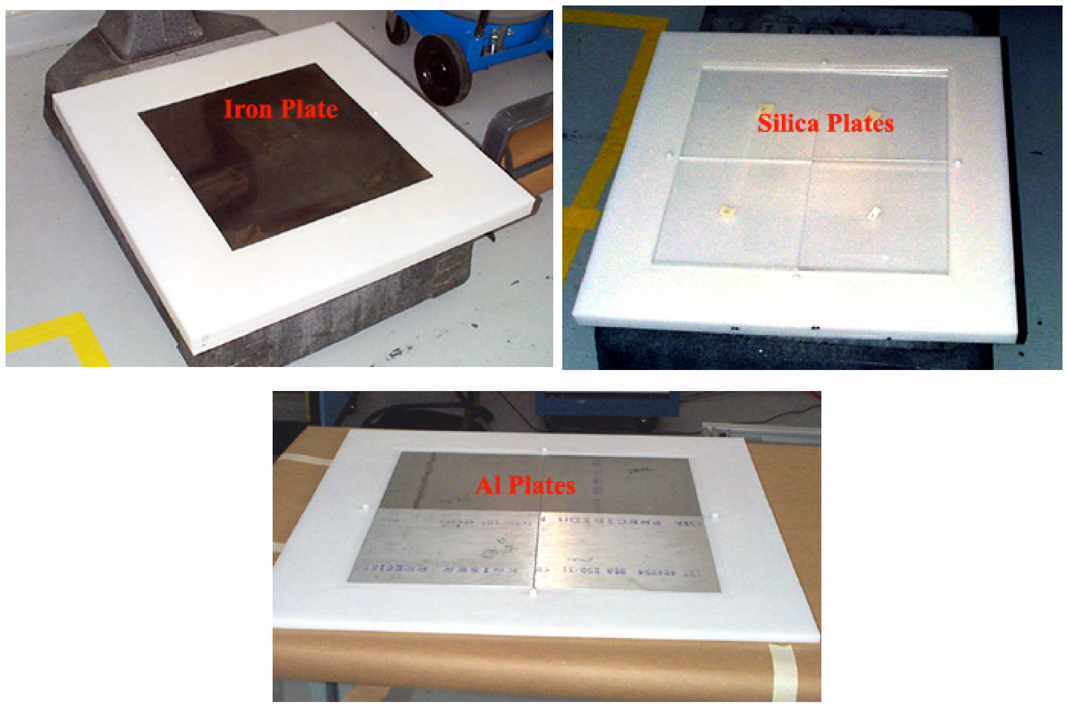
The approach to critical for this type of configuration was in some cases non-conservative. Thus, special care was taken during the approach to critical, since the reactivity worth of each unit was greater than the previous one. The approach to critical was performed by, first, hand stacking the units following the half-way rule and the ¾ rules. The ¾ critical mass rule states that hand stacking of multiplying systems shall never exceed ¾ of the extrapolated (or predicted) critical mass. While the half-way rule states that the size of each step shall not exceed one-half the increment to predicted delayed critical. In all the experiments, the more conservative rule was taken when adding an additional unit. Once the hand-stacking limit was reached, typically 8-14 units, the operators split the stack. The bottom part of the stack, which contained approximately half of the critical mass, was placed on the movable platform of the Planet Critical assembly. The top part of the stack was placed on the top platform and typically contained 2 or 3 units. The lower portion of the stack, which contained a Pu-Be source, was then raised remotely.

3. Description Of Material And Data

The four experiments were fueled with highly enriched uranium foils. The nominal dimension of the bare HEU foils were 22.86 by 22.86 cm square and 0.00762-cm-thick. These foils were laminated with plastic (polyethylene) to minimize the amount of oxidation and airborne contamination. The average mass and density for HEU foils were 68.74 ± 2.03 grams and 17.26 ± 0.51 g/cm^3, respectively. The foils were 93.23 wt.% enriched in U-235.

The glass plates used in the SiO_2 experiments were 22.86 by 22.86-cm in width and length. The thickness of the glass plates was 0.635 ± 0.0245-cm. The average mass and density of these plates were 736.59 ± 5.62 grams and 2.23 ± 0.02 g/cm^3. The total SiO_2 mass in the experiment was 64,979.84 grams which yielded a Si/U ratio of 42. The aluminum plates had nominal dimensions of 22.86 by 22.86-cm in width and length. The thickness of the Al plates was 0.635-cm. The average mass and density of the aluminum plates were 921.34 ± 2.58 grams and 2.75 ± 0.01 g/cm^3. The total amount of Al used in the experiment was 95,680.23 grams which yielded an Al/U ratio of 120. The low carbon-steel plates were 45.72 by 45.72-cm in width and length. The thickness of these plates was 0.0381-cm. The average mass and density of the Fe plates were 617.16 ± 4.69 grams and 7.74 ± 0.06 g/cm^3. The total amount of Fe used in the experiment was 5,5554.4 grams, which yielded an Fe/U ratio of 9.38. In the polyethylene only experiment, polyethylene plate inserts were used to fill the space of the recess (instead of the waste matrix material). This polyethylene plate insert was 22.86 x 22.86 cm square and 0.635 cm thick. Figure 2 shows a picture of the three materials used for the waste materials experiments.

Figure 2: Waste Materials Embedded in the Moderating Plates



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. Two different sizes of moderating plates were used in this experiment (see Figure 1). The dimensions for the bottom moderating and reflector plates were 66.04 by 66.04 cm in length and width. The thickness of the moderating plates was 1.05 cm, and the thickness of the reflector plate was 2.54 cm. The bottom moderating plates that compose the bottom stack rest on the movable platform of the Planet Critical assembly. The second set of moderating plates that constitute the upper part of the stack rest on the on the top platform of the assembly. These plates are larger than the bottom ones, because they hold the entire weight of the top stack. The dimensions of the top moderating plates were 75.184 by 75.184 cm in length and width. The thickness of the second set of moderating plates was also 1.05 cm.

4. Method Of Analysis

The experiments were reported with enough detail to permit the development of a very comprehensive model. The calculations analyzed in this study were performed using the MCNP code. 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.

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 calculated 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 combined standard uncertainty, σ_{keff}, is defined as the square root of the quadratic sum of the effects of the individual standard uncertainties on the experimental parameters. As stated above each individual change in k_{eff} is the effect from the variation of a parameter (i.e., mass of fuel, geometric effect or impurity) that is equal to the standard uncertainty of that parameter. All these individual changes were taken into account when calculating the total or combined standard uncertainty. Thus, the combined standard uncertainty is defined as:

$$\sigma_{k_{eff}}^2 = \sum_{i=1}^{N} \frac{S.U._{p_i}^2}{\delta_{p_i}^2} \left[\left(k_{eff}^i - k_{eff}^r \right)^2 + 2\sigma_{MC}^2 \right] \qquad (2)$$

Where ($k^i_{eff} – k^r_{eff}$) represents a change in k_{eff} induced by change in δ_{pi} on parameter p_i, $S.U._i$ is the standard uncertainty of the parameter p_i and N is the number of parameters being included on the analyses. The parameter σ^i_{MC} is the statistical uncertainty on the k_{eff} calculated by the Monte Carlo code. If this value is sufficiently small, then the effect of the σ_{MC} will be negligible. The parameter variation, δ_{pi}, was kept small enough to maintain the linear-dependence assumption of k_{eff} on the parameter.

5. Results and Analyses

The above delayed critical state ($k_{eff} > 1$) or subcritical state of the assembly ($k_{eff} < 1$) for the experimental configurations were attained by closing the gap between upper and lower stacks in Figure 1. The SiO_2 and Al experiments were slightly subcritical while the Fe and polyethylene experiments were supercritical (excess reactivity of 13 cents and 44.8 cents, respectively). To determine the k_{eff} in the Fe and polyethylene experiments, several reactor periods were measured and converted into reactivity through the Inhour equation. This reactivity, in turn, was converted to k_{eff}. For the SiO_2 and Al experiments, the experimental multiplication factors are reported as less than 1. The k_{eff} in these systems was estimated by the following formula:

$$k_{eff} = \left[\frac{M_U}{M_{\exp}}\right]^{1/3} \tag{3}$$

where M_u is the amount of uranium mass in the experiment, M_{exp} is the extrapolated critical mass from the 1/M curve. This relationship is particularly accurate for systems that have a multiplication higher than 100. The multiplication for the Silica experiment was 142 and for the Aluminum was 105.

The experimental k_{eff} for these four experiments and the calculated k_{eff} values is presented in Table 1. Table 1 shows that calculated and the measured k_{eff} for the Al, SiO_2 and Fe experiment compare very well. On the other hand, the experimental and calculated value for polyethylene-only experiment tend to disagree. This might be attributed to the S(α, β) scattering kernel of hydrogen that takes into account the binding energy of the molecule. Table 1 also presents worth calculations for the waste materials. The waste materials were replaced by void in the calculations to show the importance of these materials in the experiment.

Table 1: Material Data for the Waste Matrix Materials.

Material	**Experimental k_{eff}**	**Calculated k_{eff}**	**Worth Calculations for Material in ($)[a]**
SiO_2	0.9931	1.0012 ± 0.00034	-$17
Al	0.9904	0.9983 ± 0.00034	-$10
Fe	1.0001	0.9990 ± 0.00034	+$4
Poly	1.0025	1.0099 ± 0.00034	-$71.5

(a) Waste Material replaced by void in calculation.

The SiO_2, Al, Fe and polyethylene experiments were carefully performed and measurements of all significant parameters were recorded. Thus, the uncertainty analyses can be performed on all the components of the experiment. The uncertainties affecting the experiment have been divided into three broad categories. They are: 1) mass measurement, 2) fabrication, 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 total combined uncertainty is presented in Table 2.

Table 2: Summary of Total Uncertainties.

	Total for SiO_2	**Total for Al**	**Total for Fe**	**Total Polyethylene**
Total Uncertainty: Quadratic Total	± 0.0024	± 0.0028	± 0.0026	± 0.0023

6. Conclusion

These experiments were designed to supply data for interlaced waste material/Fuel/Moderator systems on the thermal region. All these experiments were performed at the Los Alamos Critical Experiments Facility to understand the characteristics of materials most likely to be found during the disposal of spent fuel and low level waste materials as well as in the Hanford Waste Storage Tanks. These waste materials could either be present in the repository (SiO_2), be introduced as part of a high level waste stream (Al or Fe) or be part of the moderating medium (polyethylene). The HEU foils experimentally approximate the uniformly distributed fissile material, and the polyethylene plates experimentally approximate the effect of interstitial water or flooding. The waste materials are examined separately in three different experimental configurations plus a polyethylene-only configuration. Detailed models were prepared for these experiments in order to assess the effect of the known uncertainties in the k_{eff} of the system.

The experiments analyzed contained silicon dioxide, aluminum, iron and polyethylene mixed with 93.23% enriched uranium and moderated and reflected by polyethylene. A base case experiment was also performed with polyethylene-only. The systematic analysis was separated into uncertainties due to mass measurements, uncertainties due to fabrication and uncertainties due to composition. Each type of uncertainty was analyzed individually and a total combined uncertainty was derived. The sensitivity analysis of these critical experiments yielded a total combined uncertainty on the measured k_{eff} of ± 0.0024 for SiO_2, of ± 0.0028 for Al, of ± 0.0026 for Fe and of ± 0.0023 for polyethylene.

8. Acknowledgment

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