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SUMMARY OF EXPERIMENTAL AND CALCULATIONAL RESULTS FROM THE JOINT USDOE/PNC CRITICALITY DATA DEVELOPMENT PROGRAM*

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ABSTRACT

A joint exchange program on the subject of criticality data development was completed in August 1988. The program was between the U.S. Department of Energy (DOE) and the Power Reactor and Nuclear Fuel Development Corporation (PNC) of Japan.[1] The primary purpose of the program was to perform critical experiments to allow validation of computer codes and cross-section libraries used for safety analyses for facilities that recycle nuclear fuel. The experiments spanned the range of neutron spectra from the very under-moderated condition (H/Pu ratio=22) to the very over-moderated condition (H/Pu ratio=2220). Based on computations performed at the Oak Ridge National Laboratory (ORNL), the average calculated *k*-effective for these experiments is 1.004. Calculated *k*-effectives ranged from 0.991 to 1.021, and the standard deviation of these results is 0.006. PNC staff have obtained similar calculational results in their validation studies. The good agreement between experiment and calculation gives confidence that the calculational methods can be applied to similar plant conditions.

INTRODUCTION

In the United States, licensing regulations require that computational methods be validated to ensure that adequate safety margins are used. The larger the extrapolation from experimental conditions, the larger the safety margin required. Facilities that recycle fast breeder reactor fuel encompass criticality conditions that are much different from conditions experienced in thermal reactor fuel recycling. In particular, these facilities process mixed Pu+U solids and solutions with a high plutonium content. However, prior to 1982, data from less than 100 critical experiments conducted with mixed Pu+U solutions were available for validation studies. To enhance facility safety and efficiency, critical experiments were conducted in this program to enlarge the data base of benchmark experiments.

The critical experiments were performed at the Hanford Critical Mass Laboratory (CML) from 1985 to 1988. As indicated in Table I, the experiments were categorized by subtask number, according to the fuel used in the experiment and facility equipment or vessel to which they applied. The computational method used in the validation studies is discussed in the following section. The critical experiments and the calculational analyses are reviewed in the next four sections. Further information regarding the experiments and validation studies are provided in the various references cited.

CALCULATIONAL METHOD

The SCALE code system was developed by and is maintained at ORNL to provide users with a consistent system that is used in design and safety studies for facilities that handle nuclear materials.[2] The validation calculations were performed with various versions of the SCALE code system (SCALE 3.1 and SCALE 4.0) using the KENO-V.a code to determine the effective multiplication factor, *k*-effective. The one-dimensional transport code XSDRNPM was used to examine experimental uncertainties and calculate neutron spectra and reaction rates. The SCALE 27 energy-group cross-section library was used in all calculations.[3] This library was derived from Evaluated Nuclear Data File B -Version IV. The *k*-effective calculations were performed on a CRAY-XMP computer.

SOLVENT EXTRACTION EQUIPMENT

In evaluating nonreactor systems for licensing and operational controls, water moderation is often assumed to result in conditions in which a system is most reactive. However, past studies have indicated that some organic mixtures may be better moderators than water.[4] This topic is of particular importance to the criticality safety of fuel processing plants where fissile material is dissolved in organic solutions during the solvent extraction process. In the past, it has been assumed that the codes and libraries validated with water-moderated critical experiments were adequate when performing design and licensing studies of organic-moderated systems. Rather than relying solely on the water-moderated experiments and speculating on the reactivity of organic-moderated systems, a set of experiments was conducted (Subtask 220) with mixed oxide (MOX) fuel pin arrays moderated by an organic mixture typical of the Purex process.[5] The organic mixture consisted of 32 vol % tributylphosphate and 68 vol % normal paraffin hydrocarbon. The experiments used five lattice pitches to yield results from near optimally moderated to very under-moderated configurations. The range of H/Pu ratios spanned by these experiments was 22

to 244. These experiments were deliberately designed to correspond to conditions used in previous experiments performed in 1978 with the same MOX fuel pins immersed in water.[6] The critical sizes of the assemblies are given in Table II. A photograph of the assembly used in the experiments conducted in 1985 (CML experiment 063) is given in Fig. 1. The photograph shows the assembly with the MOX fuel pins loaded into the 0.968-cm polypropylene plates.

The experimental results (shown in Table II) indicate that the systems were slightly more reactive when moderated and reflected by water rather than by the organic. However, as shown by the results of experiments 003R and 067 and experiments 013 and 068R, isotopic changes in the MOX fuel between 1978 and 1985 was the cause of some of the difference between the number of fuel pins needed for criticality for the two solution types. A direct comparison of these two moderators can be made using the experimental results from two sets of experiments. The water-moderated assembly was found to be more reactive in the 0.761-cm pitch configuration (CML experiment 067 vs 065). In the 1.537-cm pitch configuration, the reactivity of both systems was found to be the same, within the ±1 fuel pin experimental uncertainty (CML experiment 068R vs 061).

The results of the benchmark calculations are given in Table II.[7] Over the range of energy spectra, calculated *k*-effectives compared well with the experimental results (i.e., *k*-effective=1). The average calculated *k*-effective was 1.002 (σ=0.003) for the organic-moderated assemblies and 1.009 (σ=0.003) for the water-moderated assemblies. These calculations were made using an explicit model of each MOX fuel pin, excluding the aluminum supports that were too far from the fissile material to significantly perturb the results. Similar *k*-effective results were obtained when cell-averaged cross sections were used.

WASTE STORAGE

The purpose of the experiments was to provide benchmark data for dilute Pu+U aqueous solutions where the criticality control parameter, limiting critical concentration, is appropriate. The limiting critical concentration for ^{239}Pu in nitric acid is 7.3 g/*l*.[8] In fuel cycle facilities, plutonium is mixed with acid and often with uranium. In the presence of uranium, the limiting critical concentration can be expected to be higher, which would allow more economical subcritical limits to be used.

A series of critical experiments was conducted (Subtask 110) in a large, stainless steel cylinder (68.68-cm ID) that was water reflected.[9] The experimental assembly is shown in Fig. 2 with the large cylinder shown on the right side of the water reflector tank. A summary of the measurement results from these experiments is given in Table III. The last two experiments were conducted to irradiate solid-state track recorders (SSTR) for neutron spectra studies.[10]

The results of the *k*-effective calculations are also shown in Table III.[11] The calculational bias was determined to be 1.1%, independent of the Pu/Pu+U ratio. Experimental uncertainties do not appear to be the sole cause of the bias, since these uncertainties were found to affect calculated *k*-effective by only ±0.001. Nor should the calculational bias be attributable to the calculational model, since it included all significant structural materials. Discrepancies were found between the measured reaction rate ratios and the ratios calculated by the XSDRNPM code. The cause of the disagreements were traced to the differences in calculated ^{238}U reaction rates, rather than to differences in the thermal spectra. However, it is possible that the disagreement is within the experimental uncertainty of the SSTR measurements. After adjusting for the calculational bias, it was determined that although the limiting critical concentration increased with higher fractional amounts of natural uranium, the increase only amounted to 2%, for the case where uranium comprises 80% of the heavy metal in solution.

FISSILE SOLUTION STORAGE

The Subtask 120 experiments surveyed the basic criticality conditions relating to simple physical configurations.[12] Instead of performing criticality experiments on actual equipment, a generic data base was developed. The experiments provide a basic understanding of the reactivity effect of such parameters as Pu/Pu+U ratio, heavy metal concentration, and neutron reflection. The experiments were conducted in a small, stainless steel cylinder (35.39-cm ID) with three reflection conditions (*bare*, water, and concrete). The small cylinder is shown on the left side of the water reflector tank in Fig. 2. A variable thickness slab tank with two reflection conditions (*bare* and water) was also used. A photograph of the slab tank is shown in Fig. 3. The experimental and *k*-effective calculational results are given in Table IV. The *k*-effective results ranged from 0.992 to 1.021, with an average of 1.007.[13] Sensitivity studies indicated that the larger experimental uncertainty, in terms of calculated *k*-effective, for the concrete-reflected experiments (±0.004) compared to the other experiments (±0.002), was due to the uncertainty in the concrete gap and the elemental constituents (not just water) in the concrete. The unreflected slab experiments calculated relatively low (1.000) and it is suspected that the reason may be incomplete accounting for neutron room return.

Along with slab tanks, annular tanks are used to store fissile solutions in a manner that is floor-space efficient while utilizing favorable geometry for criticality control. To provide a data base to validate the computational methods for mixed Pu+U solutions in annular geometry, eighteen critical experiments were performed in Subtask 140.[14] The approach-to-criticality was made by adding fissile solution to the annulus of the tank. Various inserts and bottles (containing fissile solution) were located in the inner region of the vessel. Six inserts were used -- four made of concrete with various amounts of B_4C, and two made of plastic with cadmium covers. A photograph of the annular tank and an inner bottle is shown in Fig. 4. The experimental and *k*-effective results for these experiments are given in Table V.[15] The average calculated *k*-effective was 1.007,

with a standard deviation of 0.005. All of the significant structural materials were included in the calculational model. Full water reflection, radially and below the vessel, simplified the modeling. No trends in the calculational bias with any parameter were identified.

FUEL DISSOLUTION

Finally, experiments were conducted in Subtask 320 to address the criticality conditions present in a nuclear fuel dissolver.[16] A stainless steel cylindrical vessel was fabricated so that 996 MOX fuel pins could be surrounded by mixed Pu+U solutions. The same MOX fuel pins used in the Subtask 220 experiments (19.78 wt % plutonium) were arranged in a cylindrical configuration (1.40-cm square pitch) with an effective diameter of 49.8 cm. A polyethylene spacer was placed at the bottom of the vessel, in the region below the MOX fuel, to effectively exclude fissile solution from this region. A photograph showing the inner portion of the vessel, prior to installation and welding of the outer solution containment cylinder (53.2-cm diam), is given in Fig. 5. The photograph shows the empty, thin-walled guide tubes that were required to isolate the MOX fuel pins from the plutonium-bearing solution.

This configuration presents a very complicated analytical problem for the computational system. The SCALE code uses the NITAWL code (Nordheim integral method) to prepare problem-dependent cross sections. The Nordheim method assumes that the moderator outside the fuel lump is a 1/E slowing down medium with no absorption. Similarly, the resonance self-shielding calculation for those nuclides in the solution does not properly account for the neutron flux entering the solution. Therefore, the assumptions in the Nordheim method are not consistent with the neutronic conditions present when fissile, resonance, or high-absorption nuclides are present in the solution surrounding the fuel lumps.

Calculations were performed with an earlier version of SCALE (SCALE-3.1) that processed solution cross sections assuming the solution was an infinite slab with an infinite, non-absorbing medium on either side.[17] The more recent version of SCALE (SCALE-4.0) now uses an annular model to treat the nuclides in solution so that a Dancoff factor can be applied. However, it must be recognized that the inherent assumptions and limitations of the Nordheim method are still present. The calculational k-effective results for the critical experiments from the two cross-section treatments are given in Table VI, along with the experimental data. When SCALE-3.1 was used, a negative bias in calculated k-effective of up to 2% develops as Pu+U was added to the moderator. The negative bias did not change when gadolinium was added. The calculational results using SCALE-4.0 show that the annular treatment yields higher k-effectives, with no apparent trend with increasing fissile or gadolinium concentration. Therefore, the shortcomings of the Nordheim method are not serious enough to significantly reduce the calculational accuracy for these experiments. However, it is possible that other configurations will more clearly emphasize the limitations of the NITAWL code. Therefore, future work should be directed at clarifying when more sophisticated cross-section processing codes (e.g., ROLAIDS[18]) are necessary.

CONCLUSIONS

The critical experiments conducted in the joint USDOE/PNC criticality program substantially added to the data base of quality benchmark data for validating computational methods. The average k-effective calculated for all experiments is 1.004. The calculated k-effectives were found to have a normal distribution. Using the calculational biases, experimental uncertainties, Monte Carlo statistical uncertainties, and appropriate safety margins, subcritical limits can be determined for specific applications.

The experiments conducted in this program have significantly enlarged the data base available for computer code and cross-section library validation. The validation studies indicate that the SCALE computational system can accurately model the systems that were investigated. These experiments and studies should lead to improvements in the safety and efficiency of nuclear fuel recycle plants.

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Table I

Organization of Criticality Experiments by Application

Subtask	Fuel Conditions	Application
220	MOX moderated by organic/water	Solvent extraction
110	Dilute Pu+U nitrate	Waste storage
120	Pu+U nitrate in cylinders/slabs	Solution storage
140	Pu+U nitrate in annular tank	Solution storage
320	MOX moderated by Pu+U+Gd nitrate	Dissolver

Table II

Experimental and Calculational Results for Subtask 220

	Water Moderator				Organic Moderator			
Pitch (cm)	CML Experiment Number	Date	Critical Number of Fuel Pins	Calculated *k*-effective[a]	CML Experiment Number	Date	Critical Number of Fuel Pins	Calculated *k*-effective[a]
0.761	067	7-17-85	1046.9	0.994	065	6-5-85	1054.8	1.001
0.767	003R	1-24-78	1036.8	0.991				
0.968	021	11-3-78	571.9	1.001	063	5-21-85	599.2	1.006
1.242	043	1-9-79	293.9	1.005	062	5-15-85	301.8	1.000
1.537	013	10-9-78	196.7	0.998				
1.537	068R	7-25-85	199.7	1.003	061	5-14-85	199.5	0.999
1.935	032	12-14-78	165.1	1.004	060	5-8-85	165.3	1.005

[a]KENO *k*-effective statistics $\pm\sigma<0.002$.

Table III

Experimental and Calculational Results from Subtask 110

CML Experiment Number	Critical Height (cm)	Pu/Pu+U	Pu (g/*l*)	Calculated *k*-effective[a]
058	76.80	0.52	11.88	1.008
059	83.14	0.52	11.73	1.009
061	81.72	0.23	12.19	1.010
061TR	81.09	0.23	12.15	[b]
061TRR	80.10	0.23	12.15	[b]

[a]KENO *k*-effective statistics $\pm\sigma<0.002$.
[b]Experiment for SSTR irradiation.

Table IV

Experimental and Calculational Results for Subtask 120

CML Experiment Number	Vessel	Reflector	Pu/Pu+U	Pu (g/l)	Critical Dimensions (cm)[a]	Calculated k-effective[b]
046	Cylinder	Water	0.53	59	23.82	1.002
046R	Cylinder	Water	0.53	59	24.06	1.004
047	Cylinder	Concrete	0.52	59	24.88	1.008
051	Cylinder	Bare	0.53	59	34.93	0.996
049A	Slab	Bare	0.52	173	18.10/78.74	0.999
050	Slab	Bare	0.52	174	18.10/71.88	1.000
054	Slab	Water	0.52	118	12.19/60.62	1.000
055	Slab	Water	0.52	60	12.19/83.41	1.006
056	Slab	Bare	0.52	61	19.05/45.31	1.004
056A	Slab	Bare	0.52	61	17.78/60.15	0.998
065	Cylinder	Bare	0.40	42	44.46	1.006
066	Cylinder	Water	0.40	42	28.11	1.006
067	Cylinder	Concrete	0.40	42	29.36	1.017
068	Cylinder	Concrete	0.41	119	27.03	1.021
069	Cylinder	Water	0.41	119	25.26	1.004
070	Cylinder	Bare	0.41	119	41.08	1.007
077	Cylinder	Bare	0.40	173	57.97	1.018
078	Cylinder	Water	0.40	173	28.93	1.007
083	Cylinder	Concrete	0.40	173	30.60	1.016
063	Slab	Bare	0.39	41	19.81/54.70	0.998
064	Slab	Water	0.40	42	13.97/80.14	1.003
071	Slab	Water	0.41	119	13.97/47.44	1.015
072	Slab	Water	0.41	119	12.70/67.16	1.012
074	Slab	Bare	0.41	119	18.03/70.13	0.992
075	Slab	Bare	0.40	173	19.05/85.38	1.004
076	Slab	Water	0.40	173	13.46/74.19	1.014
103	Cylinder	Water	0.23	76	28.43	1.006
104	Cylinder	Bare	0.23	76	50.78	1.017
105	Cylinder	Concrete	0.23	77	30.01	1.013
084	Cylinder	Concrete	0.22	102	32.62	1.015
085	Cylinder	Bare	0.22	102	67.55	1.010
086	Cylinder	Water	0.22	102	31.28	1.010
101	Slab	Water	0.23	75	13.97/70.86	1.011
102	Slab	Bare	0.23	75	19.56/63.70	1.003
089	Slab	Bare	0.22	103	19.81/78.87	1.001
090	Slab	Water	0.22	103	13.97/88.96	1.005

[a]Slab: solution thickness/height, Cylinder: solution height.
[b]KENO k-effective statistics $\pm\sigma<0.003$.

Table V

Experimental and Calculational Results for Subtask 140

CML Experiment Number	Insert Description[a]	Pu/Pu+U	Pu (g/l)	Critical Height (cm)	Calculated *k*-effective[b]
052	Part 26/B-1	0.52	173	31.32	1.008
052R	Part 26/B-1	0.52	173	31.06	1.008
053	Part 26/B-1	0.52	113	29.24	1.012
057	Part 26/B-1	0.52	61	31.05	1.003
062	Part 26/B-1	0.52	226	34.49	1.007
087	Part 26	0.22	102	48.55	1.003
087S	Part 26	0.22	103	48.99	1.006
091	Part 24/B-2	0.22	103	27.67	1.001
092	Part 25/B-2	0.22	106	37.19	1.005
093	Part 27/B-2	0.22	108	51.10	1.011
094	B-2	0.22	108	32.86	1.011
095	Part 26/B-3	0.97	196	27.51	1.015
096	Part 26/B-3	0.97	110	25.69	1.014
097	Part 26/B-3	0.96	58	28.94	1.012
098	Part 26/B-2	0.23	73	39.58	1.011
099	Part 29/B-2	0.23	74	79.18	1.015
100	Part 28	0.23	74	104.62	1.015
108	Part 26/B-2	0.23	47	45.09	1.005

[a] Part 24: Annular concrete insert with 0 wt % B_4C.
Part 25: Annular concrete insert with 1 wt % B_4C.
Part 26: Annular concrete insert with 2 wt % B_4C.
Part 27: Annular concrete insert with 6 wt % B_4C.
Part 28: Solid Cd-covered polyethylene insert.
Part 29: Annular Cd-covered polyethylene insert.
B-1, B-2, B-3 are bottles containing Pu+U nitrate.
[b]KENO *k*-effective statistics $\pm\sigma<0.003$.

Table VI

Experimental and Calculational Results for Subtask 320

CML Experiment Number	Pu (g/l)	U (g/l)	Gd (g/l)	Critical Height (cm)	Calculated *k*-effective[a]	
					SCALE-3.1	SCALE-4.0
106	0.88	2.7	0.0	18.41	0.997	1.001
106R	0.88	2.7	0.0	18.55	1.001	1.002
110	22.63	77.84	0.0	18.90	0.987	1.001
110R	22.63	77.84	0.0	19.03	0.991	0.999
109	47.50	163.06	0.0	20.01	0.985	1.001
107	73.95	254.09	0.0	21.34	0.983	0.997
111	103.70	353.98	0.0	23.87	0.981	1.000
112	103.18	360.62	0.49	30.28	0.979	0.999
113	102.23	359.69	0.98	38.78	0.986	1.000
114	102.65	359.59	1.47	51.06	0.980	0.996
115	102.68	359.55	1.97	73.08	0.980	0.998
116	103.61	362.45	2.16	90.27	0.980	0.997
117[b]	83.30	286.57	0.0	27.42	1.017	1.010

[a]KENO *k*-effective statistics $\pm\sigma<0.003$.
[b]Fuel pins were removed in this experiment.

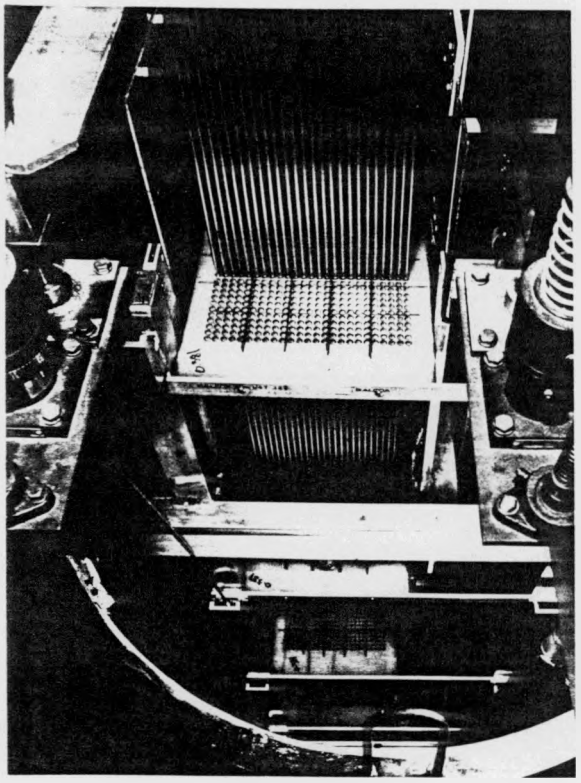

Fig. 1. Experimental assembly 063 with moderator removed.

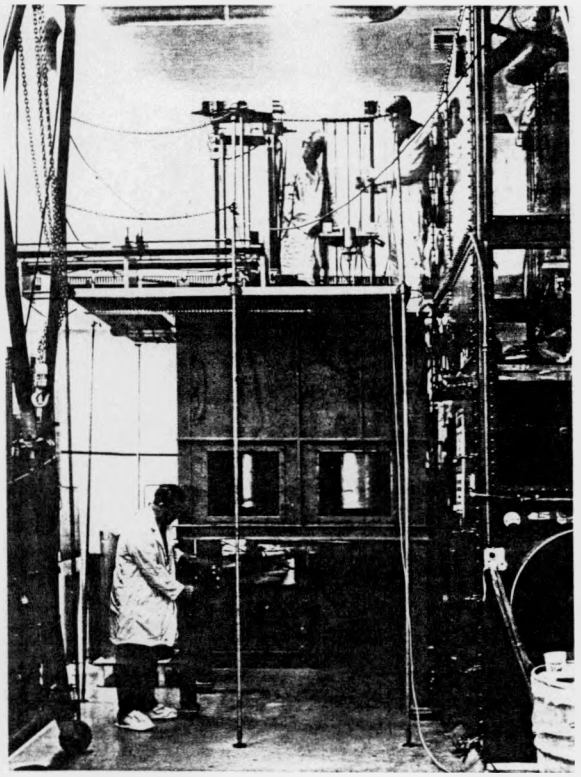

Fig. 2. Water reflector containment tank.

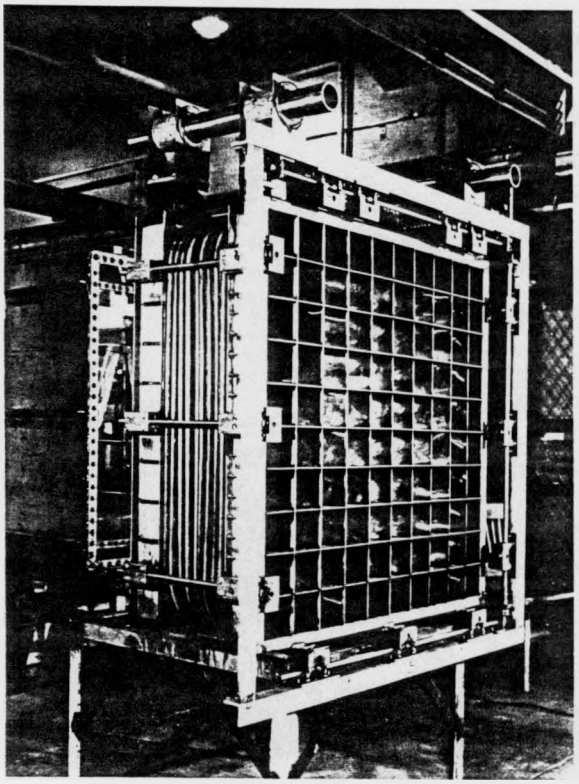

Fig. 3. Variable thickness slab tank.

Fig. 4. Annular vessel with bottle.

Fig. 5. Inner assembly (996 guide tubes) of boiler-tube type vessel.