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Criticality Safety Data Applicable to Processing Liquid-Metal Fast Breeder Reactor Fuel

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Critical-experiment data are presented on a heterogeneous lattice of fuel rods comprised of uranium and plutonium oxides, clad with stainless steel, and moderated with (U + Pu) nitrate solution, a condition not unlike that encountered in a fuel-element dissolver operation. The effect of a soluble neutron absorber (gadolinium nitrate) on the criticality of this type of system was also examined for its possible use as a method of criticality prevention and control during the dissolution step. The results provide data for code validation, an essential requirement on complex systems such as this, if the calculations are to be utilized to prescribe subsequent control limits under similar or related conditions in fuel processing. Experiments indicate (for the very limited data presented) that a heterogeneous system composed of these fuel rods in water can have a larger buckling than the fuel in the dissolved state. The question is, whether a fuel rod of a size different from that used in these experiments, immersed in fissile-bearing solutions, might have a still higher buckling (and smaller critical size) than the highest achievable buckling for fuel rods of optimum diameter and spacing in water. This important consideration regarding the criticality safety aspects of dissolvers must be examined in each case. The results of calculations of these systems with the KENO Monte Carlo code utilizing ENDF/B-III cross sections are presented.

INTRODUCTION

The processing of reactor fuels requires the dissolution of fuel elements in a dissolver vessel. During this processing step, the partly dissolved fuel elements are immersed in fissile-material-bearing solutions. Experimental criticality data are required to validate criticality calculations for the complex conditions encountered during the dissolution cycle—which involves a heterogeneous system moderated with solution.

The application of neutron absorbers for criticality prevention in fuel cycle operations can permit the safe handling of larger quantities of material with reduced probability of criticality. If, however, such soluble neutron poisons are to be considered as either a primary or secondary means of criticality control, their use must be based on a firm knowledge of the effects of the absorber.

The objectives of the experiments[1,2] reported herein were (a) to provide data on the criticality of fuel rods contained in fissile solutions of (U + Pu) nitrates and (b) to determine the effectiveness of a soluble neutron absorber (gadolinium nitrate) as a method of criticality prevention on such systems.

EXPERIMENTAL DESCRIPTION

The fuel assembly used in the criticality experiments is shown schematically in Fig. 1. It was composed of a lattice of 301 fuel rods positioned in a 55.5-cm-i.d. Type 304L stainless-steel vessel. The thickness of the walls of this vessel was 0.079 cm, of the flat bottom 1.429 cm, and of the top 1.270 cm. The vessel was, in turn,

[1] R C LLOYD, *Trans. Am. Nucl. Soc.*, **18**, 165 (1974).

[2] R C LLOYD and E D CLAYTON, *Trans. Am. Nucl. Soc.* **19**, 183 (1974).

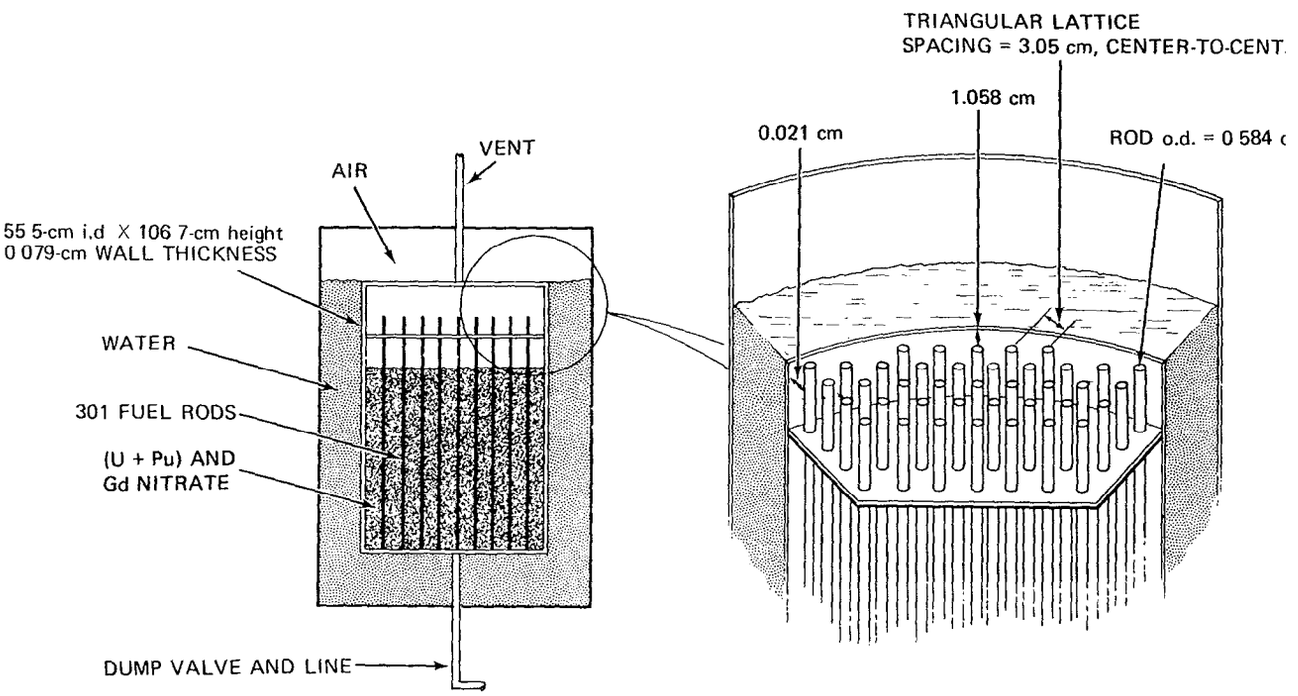


Fig. 1. UO_2-PuO_2 fuel rods in fissile solution containing gadolinium.

contained within a larger 101-cm-i.d. cylindrical vessel providing for water reflection as shown. The water reflector below the inner vessel was 20 cm thick. The water level was held at the same height as the top of the inner vessel.

Figure 2 shows the unit with the fully loaded lattice positioned therein. The experimental vessel was connected to a solution-handling system within a large hood located in the critical assembly room. Criticality was determined as a function of solution height within the inner vessel.

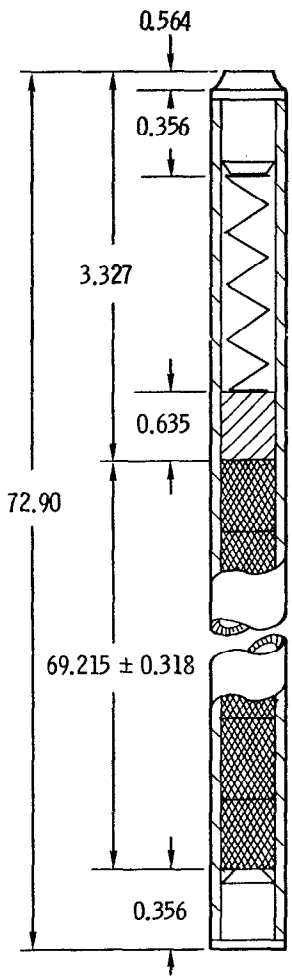
During the first series of critical experiments, the (U + Pu) nitrate solutions were varied over a concentration range of 1 to 255 g (U + Pu)/liter. The fuel rods were composed of a mixture of PuO_2-U(NAT)O_2 (25.2 wt% plutonium) clad in stainless steel. These rods were 0.495 cm o.d. and 69.22 cm long. A more detailed description is given in Table I.

The (U + Pu) nitrate solution was added incrementally to the loaded lattice assembly and criticality was determined through variation of the solution height. The relative plutonium-to-uranium content of the solution varied slightly during the course of experiments, as noted in Table II, and the plutonium concentration ranged from 0.2 to 78 g Pu/liter. The ^{240}Pu content of the plutonium was 5.7 wt%. The critical height of the solution with 0.2 g Pu/liter was 52.73 cm; the smallest

TABLE I

Fuel Rod Description

Dimensions (cm)			
	i.d.	o.d.	Length
Fuel column	---	0.495	69.22
Cladding (Type 316 stainless steel)	0.513	0.584	72.90
Lower endcap	---	---	0.356
Upper end spacer	---	---	0.635
Upper air gap	---	---	1.773
Upper endcap	---	---	0.563
Fuel Composition			
25.2 wt% Pu			
Fuel per Pin			
PuO_2-U(NAT)O_2: 138.4 ± 1.23 g Pu: 30.75 ± 0.03 g U: 91.16 ± 1.03 g O: 16.49 ± 0.17 g			
Fuel Density			
10.35 ± 0.09 g/cm^3 (93.34 ± 0.79% theoretical)			
Isotopic Composition of Plutonium in Pins			
^{238}Pu: 0.04 ± 0.01 at.% ^{239}Pu: 86.19 ± 0.06 at.% ^{240}Pu: 11.88 ± 0.06 at.% ^{241}Pu: 1.73 ± 0.01 at.% ^{242}Pu: 0.16 ± 0.01 at.%			



The second series of experiments was performed to determine the effectiveness of soluble neutron poison (gadolinium nitrate) on the criticality of the same heterogeneous assembly of fuel rods and (U + Pu) nitrate solution as was used with the unpoisoned fissile solution. This arrangement simulates the simplest approach to a possible dissolver configuration wherein, during the dissolution process, a heterogeneous system of solids in fissile solution (the partly dissolved fuel) would prevail. The (U + Pu) nitrate solution, containing various amounts of gadolinium, was added to the loaded lattice assembly—with the resultant variation in critical solution height. Gadolinium was chosen as a neutron poison because of its high neutron cross section, high solubility, and compatibility in the separation process. The gadolinium was added to the solution in the form of Gd_2O_3; the free acid of the solution readily formed $Gd(NO_3)_3$ when mixed by pumping.

RESULTS AND ANALYSIS

The Unpoisoned Assembly

The critical experiment data from the series of unpoisoned experiments are summarized in Table II, which also includes the chemical analysis of the solutions used and the calculated criticality factors.

The criticality factors were computed for each experiment through application of the well-known KENO Monte Carlo code[3] utilizing ENDF/B-III cross-section data. The 17 epithermal energy groups were obtained through application of the

[3]G. E WHITESIDES and N. R CROSS, "KENO—A Multigroup Monte Carlo Criticality Program," CTC-5, Oak Ridge Computer Technology Center, Union Carbide Corporation (1969).

TABLE II

Criticality of PuO_2-UO_2 Rods in Solution

Experiment	Critical Height[a] (cm)	Chemical Composition of Solution					k_{eff} (EGGNIT-KENO)
		Plutonium Concentration[b,c] (g/liter)	Uranium Concentration[c] (g/liter)	Acid Molarity	Total NO_3 (g/liter)	Specific Gravity	
115	19.205	77.6	180.0	3.25	375.0	1.463	1.021 ± 0.008
091	18.740	62.2	151.2	2.77	313.7	1.384	1.018 ± 0.009
093	18.839	55.3	134.4	2.61	288.0	1.347	1.017 ± 0.009
094	18.867	46.4	112.6	2.24	244.7	1.287	1.036 ± 0.009
095	19.035	39.5	95.9	1.95	210.8	1.252	1.029 ± 0.009
096	19.515	30.8	74.6	1.62	170.5	1.197	1.029 ± 0.008
097	20.317	25.3	61.2	1.54	152.8	1.167	1.014 ± 0.008
098	21.316	20.3	49.1	1.41	133.9	1.138	1.026 ± 0.008
099	22.883	15.3	37.5	1.35	118.5	1.111	1.020 ± 0.007
100	24.727	11.8	29.0	1.29	107.1	1.094	1.017 ± 0.007
101	52.730	0.2	0.7	0.3	18.9	1.003	1.000 ± 0.007

[a]A total of 301 pins on 3.05-cm center-to-center spacing on triangular pitch, tank i.d. = 55.49 cm. Fuel column covered by solution.

[b]Impurities measured in the solution included gadolinium and iron in amounts corresponding to the following weight ratios: $Gd/Pu = 2.4 \times 10^{-4}$, $Fe/Pu = 1.66 \times 10^{-2}$. (Chromium and nickel were also present in the same ratio to iron as found in Type 304L stainless steel.)

[c]Isotopic composition of the (U + Pu) solution:

^{238}Pu: 0.018 ± 0.0008 wt%
^{239}Pu: 93.843 ± 0.015 wt%
^{240}Pu: 5.700 ± 0.013 wt%
^{241}Pu: 0.360 ± 0.004 wt%
^{242}Pu: 0.079 ± 0.0004 wt%
^{235}U: 0.659 wt%
^{236}U: 0.012 wt%
^{238}U: 99.329 wt%.

ETOG code[4] and the GAM portions of the EGGNIT code.[5] The single thermal group was obtained via application of the FLANGE code[6] followed by the THERMOS code.[7] These homogenized group constants were used in KENO, assuming a cylindrical vessel with a cross-sectional area equal to that of 301 hexagonal cells. Ten thousand neutron histories were used in the KENO calculations.

The computed criticality factors were found to be slightly above unity, with the average value of k_{eff} being 1.021. This result is in conformity with previous calculations on homogeneous, hydrogen or aqueous-moderated, plutonium-bearing assemblies wherein calculations with ENDF/B cross sections have tended to overpredict the criticality factor by ~2% (Refs. 8 through 14).

Critical bucklings deduced from experiments[14,15] by use of the EGGNIT code are plotted in Fig. 3. In one case, bucklings are plotted against the plutonium concentration (plutonium contained in rods and solution) averaged over the lattice cells within the vessel. A triangular lattice assembly, with rods positioned in water on a

[4]D E. KUSNER, R. A DANNELS, and S. KELLMAN, "ETOG-1: A FORTRAN-IV Program to Process Data from the ENDF/B File to the MUFT, GAM, and ANISN Formats," WCAP-3845-1 (ENDF-114), Westinghouse Electric Corporation (1969).

[5]C R. RICHEY, "EGGNIT: A Multigroup Cross Section Code," BNWL-1203, Pacific Northwest Laboratories (1969).

[6]H. C. HONECK and D R FINCH, "FLANGE-II (Version 71-1): A Code to Process Thermal Neutron Data from an ENDF/B Tape," DP-1278 (ENDF-152), Savannah River Laboratory (1971)

[7]C L. BENNETT and W. L PURCELL, "BRT-1: Battelle-Revised-THERMOS," BNWL-1434, Pacific Northwest Laboratories (1970).

[8]R. C. LLOYD, S. R. BIERMAN, and E. D. CLAYTON, *Nucl. Sci. Eng.*, **55**, 51 (1974).

[9]S. R. BIERMAN and E. D CLAYTON, *Nucl. Sci. Eng.*, **55**, 58 (1974).

[10]S. R. BIERMAN, E. D. CLAYTON, and L. E. HANSEN, *Nucl. Sci. Eng.*, **50**, 115 (1973).

[11]R. C LLOYD and E. D CLAYTON, *Nucl. Sci. Eng.*, **52**, 73 (1973).

[12]R. C. LLOYD, E. D. CLAYTON, L E. HANSEN, and S. R. BIERMAN, *Nucl. Technol.*, **18**, 225 (1973).

[13]R. J. HALL and S R BIERMAN, *Trans. Am. Nucl. Soc.*, **18**, 1964 (1974).

[14]R. C. LLOYD and E. D. CLAYTON, *Trans. Am. Nucl. Soc.*, **17**, 269 (1973).

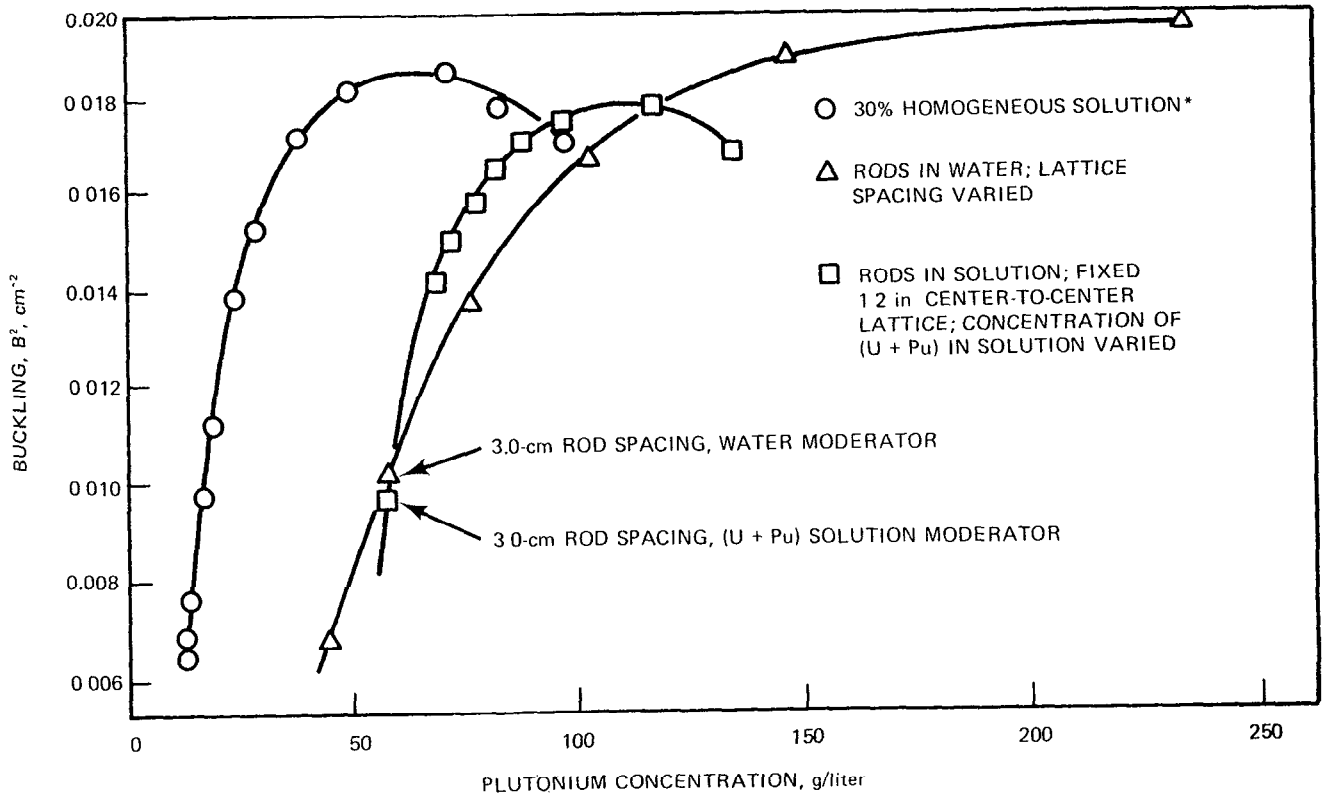
[15]C. L BROWN and C A ROGERS, *Trans. Am. Nucl. Soc* , **13**, 663 (1970).

TABLE III

Criticality of Rods in Gadolinium-Poisoned Solution

Experiment Number	Critical Height (cm)	Gadolinium (g/liter)	k_{eff} (KENO-ENDF/B-III)
115	19.205	0.02	1.021 ± 0.008
116	23.066	0.258	1.019 ± 0.008
117	28.227	0.515	1.010 ± 0.007
119	45.753	1.040	1.004 ± 0.007
120	64.506	1.280	1.000 ± 0.006
122	68.862	1.338	0.998 ± 0.005

Chemical Composition of Solution		Isotopic Composition of Plutonium and Uranium in Solution (wt%)			
		Plutonium		Uranium	
Pu	77.63 g/liter	239	93.846	234	0.006
U	180.0 g/liter	240	5.715	235	0.665
H^+	3.4 g/liter	241	0.367	236	0.012
NO_3	384 g/liter	242	0.057	238	99.317
Fe	0.8 g/liter	238	0.015		
H_2O	817 g/liter				
Specific gravity	1.463 g/cm^3				



*These data describe an aqueous solution of the nitrates of plutonium and uranium having a Pu:(U + Pu) weight ratio of 0 3.

Fig. 3. Effective concentration of plutonium versus buckling for (U+Pu) systems.

3.048-cm spacing, contained plutonium at 55.3 g Pu/liter. Bucklings are also given for the rods in water and for a homogeneous aqueous solution of the nitrates of plutonium and uranium having a Pu:(Pu + U) weight ratio equal to 0.3, a "30% solution." The rods in water show the maximum buckling to be ~0.020 cm^{-2}. The interesting information here is that the buckling of the rods in solution peaks at ~0.018 cm^{-2}, which is less than for either the rods alone in water or the homogeneous solution. Though this result comes from only one set of data, it is the kind of information needed to ascertain that a condition does not occur during the dissolution process that results in a greater buckling than for either the initial or final states.

The Poisoned Assembly

The critical experiment data for the poisoned solutions, together with their chemical makeup, are presented in Table III. The gadolinium proved to be very effective in this system. In the absence of the gadolinium, the lattice-solution assembly became critical at a solution height of only 19.2 cm, whereas, with 1.3 g Gd/liter, the critical solution height was increased to 68.9 cm. The hydrogen-to-plutonium ratio for the fuel pin-solution combination was ~86.

In the case of the gadolinium-poisoned solutions, the average value of the computed criticality factor is ~1.009. The results are conservative in that the critical system would be computed as being supercritical; that is, the computed value of k_{eff} will be found to be larger than the true value. Thus, if criticality is based on the computed k_{eff}, the computed mass will be less than the actual critical value. There appears, however, to be a trend toward computed values of k_{eff} becoming closer to unity as the system contains more gadolinium.

The gadolinium remained fixed within the solution, there being no evidence of any precipitation during the course of the measurements. Previous tests have shown no indication of change in gadolinium content over a one-month-long period.[16]

CONCLUSIONS

These criticality experiments provide data on a heterogeneous lattice assembly moderated by fissile-material-bearing solutions, a condition not unlike that encountered in a dissolver operation. The experiments indicate that a higher buckling can be achieved with a heterogeneous system composed of these particular fuel rods in water than for the homogeneous solution. The question as to whether fuel rods of a size different from that used in these experiments might have a higher buckling (equivalent to a smaller critical size) than the highest achievable buckling for fuel rods of optimum diameter and arrangement in water has not been answered in these experiments.

The criticality aspects of a heterogeneous mixed-oxide system moderated with fissile-material-bearing (U + Pu) solutions must be ascertained on an individual basis for the particular dissolver operation in question.

Using the KENO Monte Carlo code with ENDF/B cross-section data, the average value of the computed criticality factor, over a range of plutonium solution concentrations, was 1.021. For the case of the gadolinium-poisoned solutions, the value was somewhat less, 1.009.

The gadolinium was found to be an effective soluble poison for possible use as a method of criticality prevention and control in these types of systems. During the course of the experiments, the gadolinium appeared to remain stable within the solutions.

ACKNOWLEDGMENT

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[16]R. C. LLOYD, E. D CLAYTON, and L. E. HANSEN, *Nucl. Sci. Eng.*, **48**, 300 (1972).