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PHYSICS OF REACTOR DESIGN

Plutonium-Water Critical Assemblies

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The experiments described in this paper were carried out to provide data on which the design of storage vessels for plutonium solution could be based, and to check the validity of theoretical methods of critical size calculations on aqueous solutions of fissile material. Preliminary calculations showed that the variation of critical mass in aqueous solution should exhibit a minimum at a hydrogen to plutonium atomic ratio of about 600; experiments were therefore planned to measure the critical mass as a function of concentration in this region. Three concentrations were chosen, one at the expected minimum, and two at concentrations bracketing this value; the H/Pu^{239} ratios finally achieved were 397, 655 and 892.

In order that the experiment should use a small amount of fissile material, it was decided to use a water-reflected system, and the geometry of a cylinder reflected at the sides only was chosen as the most convenient shape. The solution was obtained in the form of plutonium nitrate dissolved in nitric acid, the free acidity of the solution was kept as low as possible in order to make the hydrogen concentration in the solution as near as possible to that in the reflector. The acidity used in the experiments was in fact 1.7N, a value considered necessary to avoid hydrolysis.

APPARATUS

Tank

Because of the health hazard associated with the manipulation of plutonium compounds the whole apparatus was enclosed in a dry-box and remotely controlled (Fig. 1). The reactor tank consisted of a stainless-steel cylinder 30.5 cm in diameter, 61 cm high, with walls 1.59 mm thick. The base of the tank was 6.35 mm thick and was dished so that the solution drained into a small cavity. The lid of the tank was provided with a gasket and all tubes and probes passing through it were fitted with glands; this enabled the solution to be transferred to and from the tank by suction. The solution was added via a 1 mm bore stainless steel tube; this diameter was chosen to give a small hold-up of solution in the tube and also served to restrict the maximum rate of addition to about 40 cm³/min. The solution was extracted via a 3.2-mm bore tube which reached to the base of the cavity in the base of the tank; the solution could thus be extracted at a much greater rate than it could be added.

The depth of the solution was measured in two ways; a polythene float to which a long calibrated

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needle was attached, or a calibrated electric probe accurate to about 0.5 mm. Depth measurement was necessary because of the irregular shape of the tank base, and to enable the reflector height to be set to the correct value.

The solution tank was surrounded by a aluminium-sprayed steel tank open at the top and 76.3-cm diameter. Water could be admitted to the tank by a pipe from the main supply, the tank could be emptied quickly by a magnetically operated dump valve. Depth of water was measured by a calibrated gauge glass.

Neutron-Detectors

The neutron density in the system was monitored by six BF_3 proportional chambers 28 cm long by 1-cm diameter filled with BF_3 gas to 40 cm Hg pressure; these were screwed into re-entrant stainless-steel tubes in the base of the system. One chamber filled with natural BF_3 gas was placed in a tube along the axis of the central tank; two filled with natural BF_3 and two filled with enriched BF_3 were placed equally spaced in a ring just outside the solution tank in the reflector, and one enriched chamber was placed in the reflector at a radius of 40.6 cm. This arrangement of chambers was made so that the neutron density could be measured over many decades.

Pulses from the counter were detected and counted in a conventional way; in addition the output of the central counter was fed to a rate-meter, the output of which was connected to a simple trigger circuit which could energize the safety devices. The tank and counting equipment are shown in Figs. 2 and 3.

Safety Controls

In the event of the reactor becoming super-critical unexpectedly, it could be made sub-critical by (1) dropping a cadmium tube inside the core, (2) dropping a cadmium tube which surrounded the core, (3) dumping the water from the reflector.

Solution Handling Equipment

The quantity of solution in the tank was obtained by weighing each addition of solution. The solution was transferred from one vessel to another via a flexible polyvinyl chloride (P.V.C.) tube and the use of a suction pump. This technique was found to be very reliable, and transfers were reproducible to within a few tenths of a cm³. The solution to be added was transferred to an 800 cm³ vessel which was then weighed on a balance; a flexible tube connected to the tank was fitted with a stainless-steel "needle" which

could be pushed through the rubber cap of the vessel into a depression at the base. The solution was then transferred to the tank by use of a suction pump, and the vessel reweighed to find the mass of solution added. The pump could be remotely controlled from behind a concrete wall about 1 metre thick for added safety. Hold-up of solution in the transfer pipes was negligible.

THEORY OF THE METHOD

It can be shown on simple one-group diffusion theory that for an unreflected cylinder of fissile material not too far from critical size

$$\frac{W^2}{I} \propto W_c^2 - W^2 \tag{1}$$

where W is the mass of plutonium in the system; W_c is the critical mass of plutonium in the system; and I is the neutron density at the centre of the system.

One would expect this formulation to be modified for a system with a side reflector, and far from critical; however, in our experience it appears to be adequate over a wide range. As the experiment proceeds, W is increased, and if W^2/I is plotted as a function of W^2, a nearly linear variation is obtained which intersects the W^2 axis at the value of the critical mass.

Several corrections are needed for this formula in our geometry.

Effects of Fissile Counter Length

The flux variation given above applies for a small detector at the centre of the system, in fact, to avoid the difficulties of mechanical movement and the use of very small counters, long counters fixed in the axial direction were used. If we neglect the effect of neutrons incident on that part of the counter which is not submerged in the liquid, then a simple correction to Equation 1 may be deduced. If we assume that the first harmonic of the neutron distribution predominates throughout, then the shape of the axial flux distribution is constant and the total number of neutrons entering the counter is just proportional to the height of solution. Then Equation 1 becomes

$$\frac{W^3}{R} \propto W_c^2 - W^2 \tag{2}$$

where R is the counting rate in the counter.

Unequal Length of Counter and Liquid Depth

The counter has a fixed length; thus for small depths of solution some of the counter is not immersed, and for large depths the counter will not record the neutron flux over the whole depth of solution. We define a function $\alpha(h) = n/n'$ where n = number of neutrons entering the counter; n' = number of neutrons entering the counter if it were the same length as the liquid depth.

Theoretical estimates showed that $\alpha(h)$ could take values up to about 2 for practical cases; a separate experiment was performed to deduce values of $\alpha(h)$ for use in this experiment. This was done by arranging the counter along the axis of a water tank in which fast neutron sources were placed above and below the liquid, in this way it was hoped to obtain an approxi-
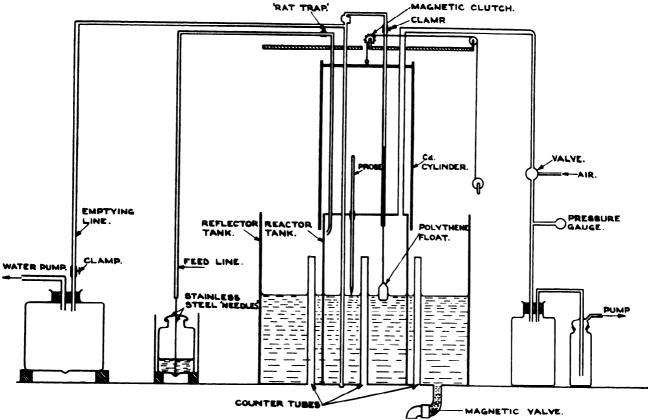

Figure 1. Reactor tank and accessories

mately uniform distribution of thermal neutron sources in the water. Values of counting rate were taken with and without a cadmium sleeve covering that portion of the counter which protruded from the water, and $\alpha(h)$ deduced as a function of liquid depth. We note here that $\alpha(h)$ need not be known with great accuracy; the introduction of this quantity merely reduces the curvature of the approach to critical graph and thus enables extrapolations to the value of W_c to be made with greater safety during the assembly. Theoretical estimates of $\alpha(h)$ were used for liquid depths greater than the counter length; in this region the theory should give reliable results.

Values of $\alpha(h)$ are shown in Fig. 4.

The final curve to be plotted in the approach is thus

$$\frac{W^{-3}\alpha(h)}{R} \propto W_c^{-2} - W^{-2} \qquad (3)$$

Selection of Counter Sensitivity

In order to be able to follow the variation of neutron flux in the system from the lowest possible value of W it is necessary to calculate the large neutron-source strength existing in the solution (spontaneous fissions, etc.) and thus provide counters of the necessary sensitivity, or a separate source to give a flux of sufficient intensity. Calculations showed that the major source of neutrons in the solution was from (α,n) reactions between the oxygen present and Puα-particles. These calculations showed that this should provide a suffi-
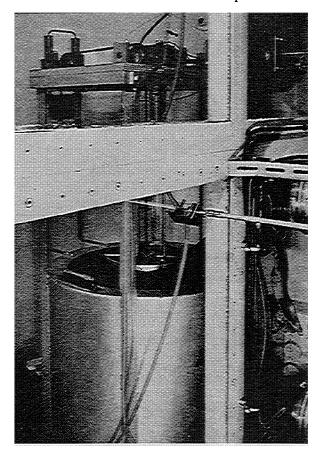
ciently large source (about 11 neutrons/ml/cm^3 in a 50 mg Pu/litre solution) to make an external source unnecessary, and on this basis the counters referred to above were chosen. Agreement between the predicted and observed fluxes was satisfactory.

TEMPERATURE EFFECTS

When the first assembly had been made, it was clear that sufficient plutonium was available to make a critical assembly at the second concentration, the effective reproduction constant k_E could thus be made as near to unity as we pleased. It was decided to use this facility to attempt to measure the variation in reactivity of the system with temperature; since the neutron flux in the system varies inversely as $(1 - k_E)$ a small change in k_E for $k_E \sim 1$ should produce a large change in the observed neutron flux. The reflector was therefore fitted with a 2 kw immersion heater.

EXPERIMENTAL METHOD

Approach to Critical

The experiments were performed in the following way. In the first run plutonium was added cautiously in batches of 800 ml until it became clear that the approach-to-critical curve was being followed. The procedure was then:

1. The safe quantity of solution to be added was deduced by an inspection of the approach-to-critical
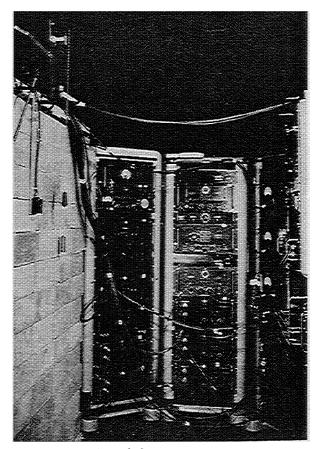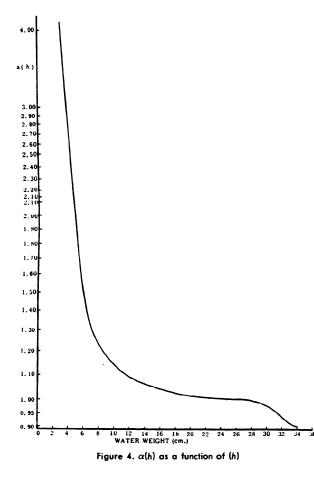

Figure 2. Reactor in position

Figure 3. Counting equipment

curve. This was done by taking one-quarter of the difference in predicted critical mass and mass already added, or 800 ml, whichever was the smaller.

2. The amount thus decided was transferred to the transfer vessel from stock and weighed. The weighed bottle was then connected to the tank by the flexible tube, and the reflector height raised to correspond to the expected height of solution after the addition. The safety rod and tube were checked, and the next operation performed from behind the concrete safety-wall.

3. Plutonium was then transferred to the tank, this operation took about 30 min and rough checks of neutron density were made periodically to ensure that it was not rising faster than expected.

4. When all the solution had been added, a short time was allowed to ensure that the system was not diverging, and the depth of solution was then measured. The depth of the water reflector was adjusted if necessary, the neutron density was measured by taking two consecutive readings of the counter. The empty transfer vessel was reweighed and the mass of plutonium added, determined; the value of $W^3\alpha/R$ was computed and plotted, and a fresh estimate of W_c made. At several points measurements were made with the Cd-cylinder between the reflector and solution and also with the reflector removed, these were used to make estimates of the critical masses with fast reflection only, and with no reflection at all.

Figure 4. $\alpha(h)$ as a function of (h)

5. The procedure was then repeated for the next addition.

Measurement of Temperature Effect

At two points during the second assembly (multiplications of about 30 and 100), the amount of plutonium in the tank was held constant while the temperature was varied. It was difficult to make an accurate record of the time variation of temperature due to the large thermal capacity of the system, the temperature and the count rate in all counters was thus recorded in a cycle in which the temperature was brought to a maximum and the tank then allowed to cool. The maximum difference in count rate and the maximum temperature difference in the reflector were thus recorded. There was no provision for measuring the temperature of the plutonium solution directly, but theoretical considerations showed that due to the relatively small thermal capacity of the plutonium solution the temperature rise in it should have been the same as that in the reflector to within 10%. The temperature of the reflector was observed with a mercury-in-glass thermometer.

To check the magnitude of temperature effects due to the counters themselves, a separate experiment was performed in which a counter was heated in a beam of thermal neutrons from the Harwell pile. To provide a further check on the effect, a run was made during the progress of the third assembly at the same solution height; in this case the multiplication was very low, and any change in counting rate was therefore due to counter temperature effects.

The change in density with temperature of the plutonium solution used in these experiments was measured separately over the range of temperatures employed.

RESULTS

Critical Mass

The results obtained are set out in Figs. 5, 6 and 7. It was found that the curves obtained with each of the six counters were almost linear for values of $(W/W_c)^2 \gtrsim 0.5$; the accuracy of curve fittings and convergence of the approach curve was such that the critical mass could be deduced to within about 1%. Estimates of the fast-reflected system could be made in solutions I and II, an estimate of the bare system could only be made for solution I. No estimate of the fast-reflected or bare system is possible in solution III.

Table I

Quantity	Solution		
	I	II	III
Concentration (mg Pu ml)	62.86	38.63	28.55
Observed critical mass in grams Pu for reflected cylinder	1182	920	944
Observed critical mass in grams Pu for fast reflection only	1350	1200	—
Observed critical mass in gm Pu for bare system	1700	2200	
H/Pu atomic ratio	397	655	892

Table II. Estimated Effect on Critical Mass in gm Pu

Disturbance	Solution		
	I	II	III
Counter tube displacement	2	—	—
Absorption in Pu^{240}	60	67	126

Table III

	Run 1	Run 2
Temperature rise (°C)	6 ± 0.6	9 ± 0.9
Observed change in counting rate	1.075 ± 0.01	1.44 ± 0.02
= counting rate at ambient/maximum temperature reached		
Observed change in counting rate due to temperature effect in counters	1.018 ± 0.006	1.027 ± 0.009
Net observed change	1.057 ± 0.01	1.413 ± 0.02

Table IV

		Water parameters	
σ (239)	1113 b	Thermal transport mfp	0.48 cm
σ (240)	1000 b	Fast transport mfp	3.2 cm
σ (N)	1.78 b	(slowing down length)^2	33 cm^2
σ (H)	0.34 b	(diffusion length)^2	7.129 cm^2

The chemical composition of the solution used is given in Table I; with these values, the results observed for critical mass variation can be summarized as follows. Effects due to absorption of neutrons in the counters and impurities were negligible; other effects are summarized in Table II. The plutonium used contained about 3% of Pu^{240} for which the absorption cross section was taken to be 1000 barns.

Temperature Effect

The results obtained on solution II are summarized in Table III.

CONSTANTS ADOPTED

In order to determine values of η, values for the nuclear constants of water, nitrogen, plutonium-239 and plutonium-240 had to be used. The values adopted are given in Table IV. The characteristic lengths quoted for water were adjusted in the core according to the observed hydrogen densities for the three solutions studied.

ANALYSIS OF RESULTS

From the concentration and quoted cross sections, the thermal utilization and characteristic lengths in the core were obtained. The observed values for the critical size and a two-group calculation gave values of K^2 for the three systems, and so k_∞ was deduced. The equation $k_\infty = \eta f$ then gave the values of η.

The results are given in the following Table V:

Table V

Solution	$1/f$	k_∞	η
I	1.165	1.853	2.16
II	1.251	1.728	2.16
III	1.329	1.610	2.14

DISCUSSION OF RESULTS

These values of η are about 10% higher than would be predicted from the published cross sections and ν value for plutonium. Epithermal fissions have been neglected in the calculations, and if due allowance is made for this a reduction of about 5% is obtained. However, there is considerable discrepancy between the experimental and theoretical slowing-down length in water, and if the latter value is used in the analysis, values of about 2.0 are obtained. In view of the existing uncertainties in the cross sections of Pu^{240} and more particularly as these results have been used in safety calculations for chemical plants, it has been considered advisable to retain the higher values of η in further work on these solutions.

As a guide to critical size problems in plutonium solutions, calculations have been made using a two-group theory and a value of η of 2.22.[1] This value is chosen to allow a margin of error in the safe direction. Two types of systems have been considered: the hypothetical case of Pu^{239} suspended in water without change of hydrogen density, and the more realistic one of plutonium nitrate in $5N$ nitric acid solution.

ANALYSIS OF TEMPERATURE EFFECTS

From the observed ratio of count rate for the subcritical reactor at two temperatures 9° apart, a value was calculated for the corresponding change in reactivity. This was based on the following theory:

Let H_c = extrapolated critical height of solution; h_c = extrapolated height of solution; R_c = extrapolated radius of tank; S = neutron source density; D = diffusion coefficient for thermal neutrons; λ_z = axial extrapolation length; λ_r = radial extrapolation length; C = count rate in centre counter; d = solution density; k_∞ = reproduction constant; L = diffusion

length in core; and L_s = slowing down length in core. Suffixes 0 and 1 refer to the initial and final temperatures respectively, and Δk_∞ is the change in k_∞ due to this temperature rise. Then

$$\frac{C_0}{h_0} = \frac{S_0}{D_0} H_{0e}^2 \frac{h_{0e}^2}{H_{0e}^2 - h_{0e}^2}$$

$$\frac{C_1}{h_1} = \frac{S_1}{D_1} H_{1e}^2 \frac{h_{1e}^2}{H_{1e}^2 - h_{1e}^2}$$

$$\frac{\pi^2}{H_{0e}^2} + \frac{2.405^2}{R_{0e}^2} = -\frac{1}{2}\left(\frac{1}{L_0^2} + \frac{1}{L_{s0}^2}\right)$$
$$+ \frac{1}{2}\sqrt{\left(\frac{1}{L_0^2} + \frac{1}{L_{s0}^2}\right)^2 + \frac{4(k_\infty - 1)}{L_0^2 L_{s0}^2}}$$

$$\frac{\pi^2}{H_{1e}^2} + \frac{2.405^2}{R_{1e}^2} = -\frac{1}{2}\left(\frac{1}{L_1^2} + \frac{1}{L_{s1}^2}\right)$$
$$+ \frac{1}{2}\sqrt{\left(\frac{1}{L_1^2} + \frac{1}{L_{s1}^2}\right)^2 + \frac{4(k_\infty + \Delta k_\infty - 1)}{L_1^2 L_{s1}^2}}$$

In these equations h_{e0}/H_{e0} is known, $\delta = d_0/d_1$ is known, and R_1/R_0 can be obtained by a knowledge of thermal expansion of the tank. This implies that h_0/h_1 is known. Furthermore $\delta = S_0/S_1 = D_1/D_0$. Knowing the solution density changes in core and reflector it is possible to find L_1^2, L_{s1}^2 and λr_1 and λz_1. From this, it is possible to express $\Delta k_\infty/k_\infty$ in terms of the other parameters and for solution II the following value was obtained:

$$\frac{\Delta k_\infty}{k_\infty} = -0.00846\% \pm 0.003\%/°C.$$

DISCUSSION OF TEMPERATURE EFFECTS

$\Delta k_\infty/k_\infty$ as observed is the sum of the fractional changes in thermal utilization and η. The expected change in f can be deduced from figures given in a companion paper by Egelstaff. Assuming that the neutron spectrum in the core was pure Maxwellian corresponding to 20°C, the expected change is

$$\frac{\Delta f}{f} = 0.0197\%/°C$$

This, with the experimental reactivity change, would give for the change in η
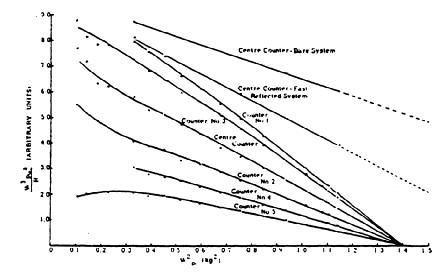
$$\frac{\Delta \eta}{\eta} = -0.0282\%/°C$$

This is in disagreement with the experimental values for the cross sections of Pu^{239} as a function of energy.[2] These suggest that η is constant up to 0.5 ev within the experimental uncertainty of about $3\frac{1}{2}\%$. As discussed before, the Pu^{240} cross section is an uncertainty, and more so the temperature variation of this cross section. The Pu^{240} concentration is low in these experiments and so its effect is unlikely to explain the discrepancy.
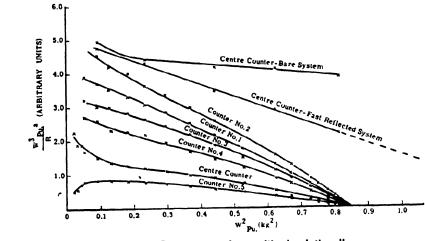
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2. Egelstaff, P. A., *The Slow Neutron Cross Sections of Pu*^{239}, NRDC52 (1954).

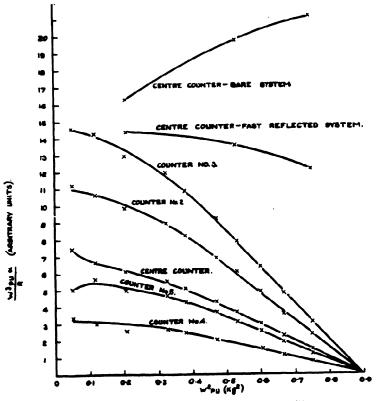Figure 5. The approach to critical solution I

Figure 6. The approach to critical solution II

Figure 7. The approach to critical solution III