For both reactor design and safety in nuclear-fuel processing, one needs to know masses, concentrations, and dimensions of critical plutonium systems. Behavior is similar to that of uranium, but different enough that extrapolation is difficult and experiments are necessary. Experiments at a Hanford facility have examined criticality in water solutions and plutonium assemblies.

PLUTONIUM CRITICALITY EXPERIMENTS

By E. D. Clayton

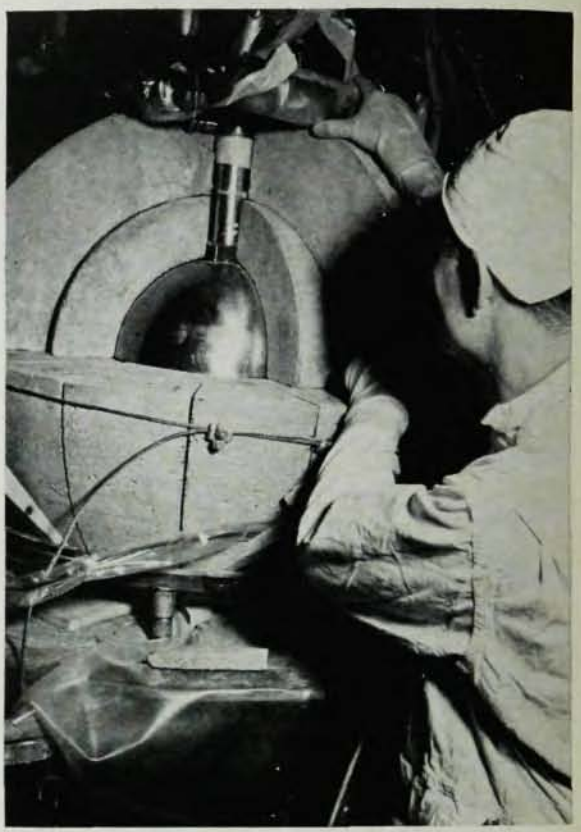

Fig. 1. Experimenter places concrete layer around vessel that holds plutonium-nitrate solution in criticality experiment.

Plutonium, the man-made element of atomic number 94, has established itself mainly as the fissile component of nuclear weapons. It is always made in the operation of a uranium-fueled reactor and has been manufactured in large quantities since its introduction in World War II. It has long been considered inevitable that plutonium will become an important reactor fuel. It offers hazards and puzzles to reactor physicists, and for two reasons in particular they must know the conditions under which assemblies containing it become critical. First, they must design processing systems that will not have accidental and dangerous chain reactions. Second, they need data for reactor design.

To determine these data (and extend some that were determined more than a decade ago), calculations to determine criticality parameters and experiments like that shown in Fig. 1 are being carried out in the Critical Mass Laboratory at the Battelle Institute's Pacific Northwest Laboratory at Hanford, Wash.

Why study plutonium?

In uranium-fueled reactors fissile plutonium is made because ^{238}U readily captures neutrons to become ^{239}U. The ^{239}U undergoes two successive beta decays (half-lives are 23 minutes and 2.3 days) to become first ^{239}Np and then ^{239}Pu.

Early criticality measurements with plutonium were made at the Los Alamos Scientific Laboratory, but with large-scale production of the element, more data were needed. Therefore, during 1950-51 a series of criticality experiments at Hanford, a major plutonium-producing site, were performed to determine safe limits for handling the material. F. Kruesi and his colleagues made measurements to find the minimal critical mass of a light-water-reflected, plutonium-nitrate solution in water with plutonium concentrates in the range of 25-125 g Pu/liter.

With the development of new processing methods, still other data were needed on solutions and compounds outside the ranges previously studied. So our facility was constructed, and experiments started in it in mid-1961. More than 500 experi-

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ments have been performed, about two-thirds with plutonium-nitrate solutions and the others with PuO_2-plastic mixtures in solids.

Critical mass, volume, size

Critical masses and minimal critical dimensions of ^{239}Pu for a few simple shapes are given in Table 1. Variation of critical mass with volume for ^{239}Pu-water mixtures is complex, as can be seen from Fig. 2. The curves show radius of the critical sphere and mass of plutonium contained in it as the plutonium is mixed with increasing amounts of water. The upper curve is for bare, homogeneous plutonium-water spheres. The lower one is for plutonium-water spheres surrounded by an infinite reflector of pure water.

The curves show three effects. First, beginning with plutonium metal at the density of the alpha phase (19.6 g/cm³), critical mass and volume increase as water is added. The increases in critical mass and volume occur because decrease in plutonium density leads to increased neutron leakage.

Second, with further dilution, moderation by the water becomes increasingly important; probability of slow-neutron-induced fission increases. Although critical radius becomes larger, critical mass decreases; in other words, critical plutonium concentration falls more rapidly than critical volume increases. Ultimately one reaches a condition of optimal moderation and minimal critical mass (~ 520 g of plutonium at a concentration of ~ 32 g Pu/liter).

Finally, both critical size and mass increase once more because of excess neutron capture in water (principally in hydrogen) and become infinite as concentration reaches 8 g Pu/liter (H/Pu ratio of 3312).

Table 1. Critical values for ^{239}Pu[1-4]

Density[a]	Mass, sphere (kg)	Diameter, infinite cylinder (cm)	Thickness, infinite slab (cm)
Water-reflected metal			
α	5.6	~ 4.3	~ 0.6
δ	7.6	~ 5.3	~ 0.7
Unreflected metal			
α	10	~ 6.1	~ 2.8
δ	16.3	~ 7.6	~ 3.5

Minimum critical mass of homogeneous mixture in water

Water-reflected sphere 520 g Pu

Unreflected sphere 900 g Pu

Smallest critical concentration in water[b]

8.0 ± 0.3 g Pu/liter

[a] Density of the α phase is 19.6 g/m³; density of the δ phase is 15.6 g/cm³.

[b] This is the smallest concentration that can be made critical in an infinite volume of aqueous mixture.

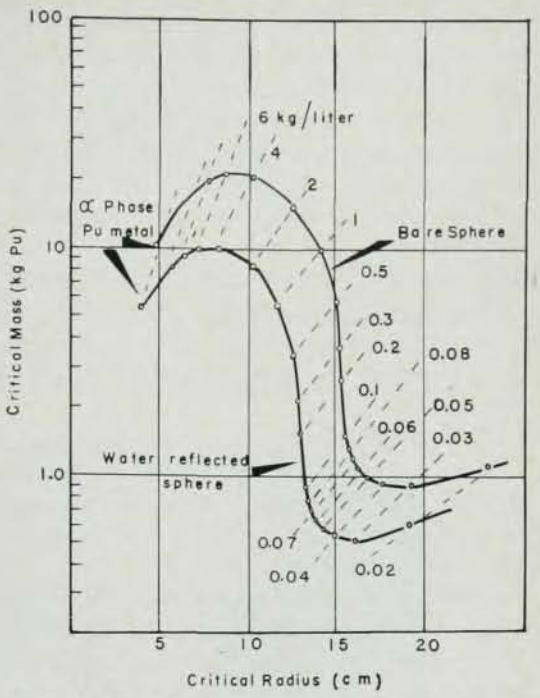

Fig. 2. Estimated mass and radius of critical plutonium-water spheres. Upper curve is for bare spheres and lower for water-reflected ones. Labeled dotted lines show densities, starting at 19.6 kg/liter of α-phase metal. Bottommost knee is principal region of existing critical data and steepest portion is now being explored.

Criticality in ^{239}Pu and ^{233}U

We know less about criticality conditions for ^{239}Pu than for ^{235}U. Nevertheless, one can make a few simple comparisons between the two based on neutron capture cross sections.

Thermal-neutron-induced fission in plutonium will release on the average 2.91 fast neutrons ($\nu = 2.91$), whereas for ^{235}U the number is 2.47. The effective 2200-m/sec absorption cross section of plutonium for neutrons with a Maxwellian distribution at room temperature is 1107 barns, 676 barns for uranium. But since fewer fissions occur for each neutron absorbed in plutonium than in uranium, the net yields of neutron released per neutron captured in fuel are nearly equal (2.09 for plutonium, 2.07 for uranium). Some of the neutrons absorbed merely produce ^{240}Pu and ^{236}U.

In its simplest form the reproduction factor k for a system of ^{239}Pu or ^{235}U and moderator that is critical on thermal neutrons can be written $k = \eta\, f\, P$. (k is ratio of number of neutrons in one generation to the corresponding number in the preceding generation; η is the number of neutrons produced per neutron absorbed in fissile material; f is the probability that a neutron will be absorbed in fissile material—the number of neutrons absorbed in fissile material divided by the total number absorbed; P is the probability that neutrons

do not escape from the system through leakage. In terms of nuclear concentrations and cross sections $f = N_x\sigma_x/(N_x\sigma_x + N_m\sigma_m)$ where N_x and N_m are concentrations of fissile and moderator nuclei and the σ's are appropriate microscopic capture cross sections.)

The minimal water-reflected critical mass for a homogeneous ^{235}U-water mixture is 820 g,[1] and the corresponding concentration is ~ 52 g/liter. That ^{239}Pu has a smaller minimal critical mass than ^{235}U comes principally from its higher neutron capture, not from its higher neutron yield at fission ν.

One might suppose that for equal concentrations the critical mass of plutonium will always be smaller than that of uranium. (Since the average absorption cross section is larger, f will be larger.) But this is not true.

The capture-to-fission ratio α varies with neutron energy, and the variation differs between ^{239}Pu and ^{235}U. The factor η, therefore, varies with energy [$\eta = \nu/(1 + \alpha)$]. For example, plutonium has a large resonance capture cross section for 0.3-eV neutrons, and at this energy η is only ~ 1.7. The neutron energy spectrum depends on the ratio H/X of hydrogen to fissile atoms.

Monte Carlo calculations by L. L. Carter of k as a function of H/X for infinite homogeneous systems of ^{239}Pu and ^{235}U in water are shown in Fig. 3, and η and f for the same systems are shown in Fig. 4. When averaged over neutron energy η differs from its 2200-m/sec value; it varies smoothly with H/X within wide limits and is somewhat insensitive to resonance peaks in capture cross sections.

Variation of η with energy is one reason simple theory fails to give correct results at higher plutonium concentrations and neutron energies. This is also why attempts to predict critical data for ^{239}Pu in terms of known critical ^{235}U masses generally have not been successful.

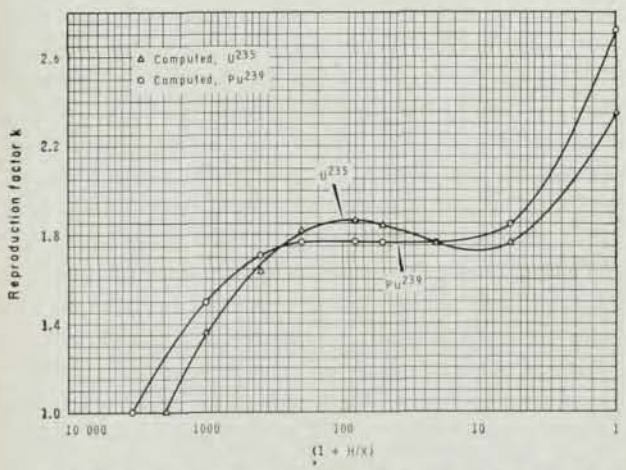

Fig. 3. Monte Carlo values of reproduction factor k in homogeneous ^{239}Pu- and ^{235}U-water mixtures. (H/X is atomic ratio of hydrogen to fuel.)

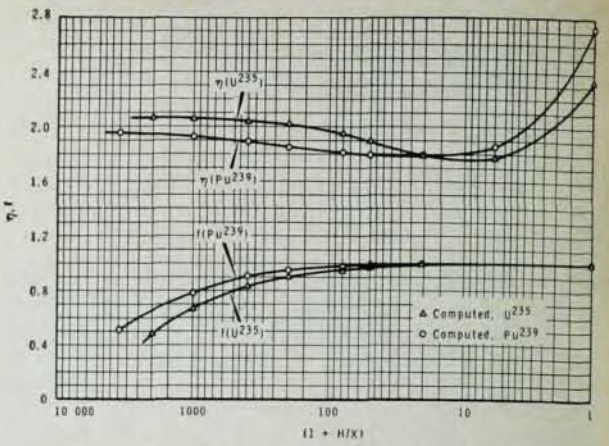

Fig. 4. Monte Carlo values of fuel absorption probability f and neutron-production ratio η for ^{239}Pu and ^{235}U in homogeneous water mixtures.

Computed curves for k_∞ (k for an infinite homogeneous volume) imply there is a range of concentrations over which critical masses of ^{235}U are less than those of ^{239}Pu at the same concentrations. It is interesting to note that ^{239}Pu has a smaller minimal critical concentration, smaller minimal critical mass (at optimal moderation), and smaller critical mass in the pure metal, although k for ^{235}U exceeds that for ^{239}Pu throughout the H/X range from ~ 20 to 300. The explanation is purely the different variations in the capture-to-fission ratio α with energy.

Plutonium isotopes

Isotopes of plutonium other than ^{239}Pu are also of interest, and it is interesting to speculate as to the possibility of criticality. A few percent of ^{240}Pu is normally present in all plutonium since it is formed in nuclear reactors from radiative neutron capture in ^{239}Pu.

This heavier isotope adversely affects criticality. Capture of thermal neutrons in ^{240}Pu results almost entirely in formation of ^{241}Pu; little fissioning is caused by slow neutrons. But at high neutron energies fission becomes important. In this respect ^{240}Pu is like ^{238}U. The effect of ^{240}Pu on the criticality of ^{239}Pu depends on H/Pu ratio, or degree of moderation. For dilute solutions (for example, 30 g Pu/liter), the critical concentration of ^{239}Pu will be increased by about 3 percent for each added percent of ^{240}Pu.[2]

Calculations show that ^{240}Pu has its greatest effect on critical mass for H/Pu ratios of 20-60. In this range of H/Pu ratios, the neutron energy spec-

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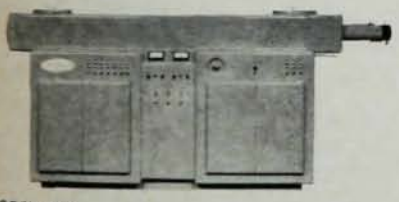

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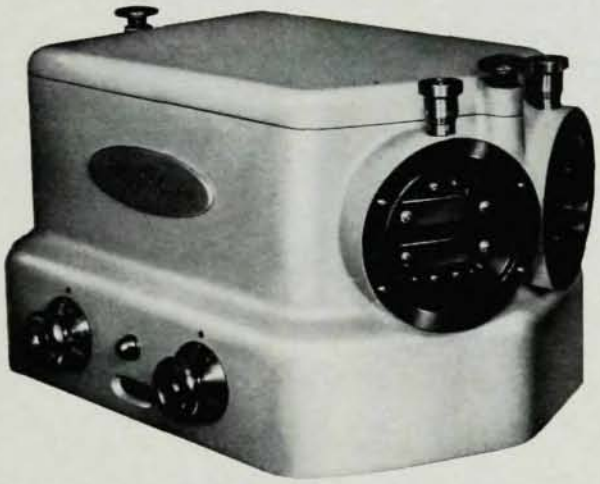

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trum is of intermediate neutron energy, being between that for a predominantly thermal reactor and a fast reactor.

The isotopes ^{240}Pu and ^{238}Pu are like ^{238}U in that all three nuclei have even numbers of protons and neutrons. A large quantity of ^{238}U will not chain react by itself because of energy losses from inelastic scattering of fission neutrons in the uranium. The fissionability of a nucleus with charge Z and mass number A is determined by the ratio of Z^2/A, and ^{240}Pu and ^{238}Pu have larger values of this parameter than ^{238}U—hence these nuclei have lower threshold energies for induced fission.

Carter's Monte Carlo calculations for k_∞, in infinite metal systems of three uranium and four plutonium isotopes are shown in Table 2. So far, criticality has been achieved with three of these nuclides; ^{233}U, ^{235}U, and ^{239}Pu. The values of k_∞ show that criticality is also possible with the even-even nuclei ^{238}Pu and ^{240}Pu, as well as with ^{241}Pu. It is important to note, however, that these results are very sensitive to cross sections, and cross sections are not well known between 0.05 and 5 MeV where most fissions and absorptions occur in fast systems.

The value of k_∞ determines whether criticality is possible, and critical size depends on both k_∞ and neutron leakage. Leakage depends sensitively on total cross section (scattering plus absorption). Although k_∞ for ^{235}U and ^{239}Pu differs by only ~ 18 percent, the critical mass of pure ^{235}U exceeds that for ^{239}Pu by a factor slightly greater than four after correcting for differences in density. Thus, although differences in k_∞ may be small, difference in critical mass can be large in these all-metal systems. Monte Carlo calculations by Carter give the critical mass for a bare sphere of ^{238}Pu (density 19.6 g/cm^3) as 5.2 kg, which is about half the value of ^{239}Pu metal. Critical mass for ^{240}Pu bare metal sphere is estimated to be ~ 80 kg.

Table 2. k_∞, fissions and absorptions in infinite metal systems[a]

Nuclide	*Fissions*	*Absorptions*	k_∞[b]
^{233}U	3740	3884	2.56 ± 0.01
^{235}U	7199	7918	2.35 ± 0.01
^{238}U	440	4013	0.31 ± 0.03
^{238}Pu	3232	3938	2.66 ± 0.02
^{239}Pu	5439	5939	2.77 ± 0.01
^{240}Pu	3050	4021	2.50 ± 0.07
^{241}Pu	3205	3944	2.62 ± 0.03

[a] From Monte Carlo calculations of L. L. Carter.

[b] $k = \nu_{av} \cdot \frac{\text{fissions}}{\text{absorptions}}$

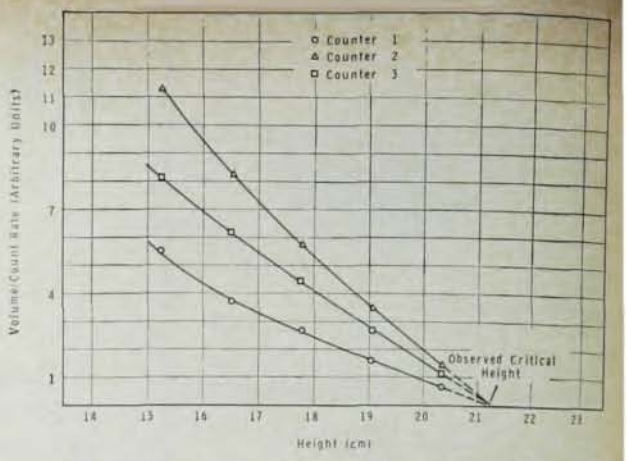

Fig. 5. Critical-approach curves plot reciprocal counting rate (× volume) against amount of material in assembly. Here amount of material was represented by height of a Lucite-reflected rectangular prism with 22.9-cm square base. Core material was PuO_2-polystyrene compacts with 1.12 g Pu/cm^3 and an overall H/Pu atomic ratio of 15.

Critical experiments

In our critical experiments, the fissile material (plutonium in various forms and concentrations) necessary for a self-sustaining chain reaction is carefully assembled in prescribed geometries under precisely measured and controlled conditions. As more fissile material is added, k approaches unity, and criticality is achieved.

During the construction of a critical assembly, it is important that the assembly size never becomes so large that criticality can occur on prompt neutrons alone. As long as all neutrons released in fission (both prompt and delayed) are required for criticality, the system is said to be "delayed critical". Then its kinetic behavior is determined by the mean emission life of the delayed neutrons (those that are released only after decay of certain fission products). Changes in k from unity result in exponential changes in neutron flux at a rate depending on average neutron lifetime in the system.

On the other hand, if k were to become so great that $(k\text{-}1)/k$ were equal to β (the fraction of neutrons delayed) or greater, the system would become critical because of prompt neutrons alone. Delayed neutrons would no longer be controlling. Since the time from birth to capture is small compared with mean life of the delayed-neutron emitters, neutron flux would increase at a rapid and uncontrollable rate.

During the approach to criticality, source-neutron multiplication is a guide to fuel additions. Multiplication (the number of neutrons freed in the future by one neutron free at present) can be expressed by

$$M = 1 + k + k^2 + k^3 \ldots k^n.$$

When k is less than unity, the expression reduces to $M = 1/(1 - k)$. Since M is proportional to

counting rate, one can predict criticality by plotting the reciprocal of the observed counting rate as a function of the amount of material in the assembly. As k approaches unity, the reciprocal of the counting rate approaches zero. Typical multiplication curves from a critical experiment are shown in Fig. 5.

Criticality is finally achieved by withdrawal of a small neutron-absorbing rod or insertion of a little fuel (fuel-bearing control rod) by remote methods. With k slightly greater than unity, neutron flux will increase exponentially until the rod is repositioned to give $k = 1$. The power level is then controlled by subsequent rod adjustments.

Figure 1 shows a typical critical assembly unit for a critical experiment with plutonium-containing liquids. The sphere is being covered with 4 inches of concrete, which reflects some of the neutrons that would otherwise escape back into the plutonium solution. Since concrete is standard structural material, its effect on criticality is of practical interest.

Plutonium-nitrate solutions of various concentrations are pumped into the sphere through a line at the bottom. If the concentration is adjusted properly, a self-sustaining chain reaction will occur. Control is by absorbing rods that enter the solution from above through the sphere neck.

We conduct critical experiments with plutonium-containing solids using a remote-control split-table machine in one of the critical-assembly room hoods.

Figure 6 shows PuO_2-polystyrene cubes being hand-stacked on the table halves before an experiment. With table faces in the fully open position

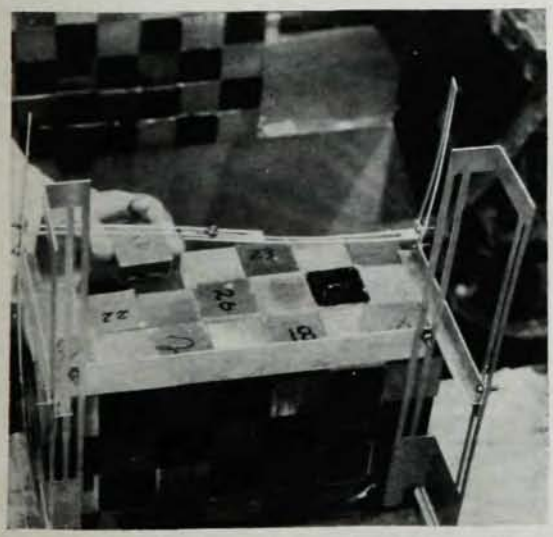

Fig. 6. Hand-stacking of PuO_2-polystyrene cubes on remotely-operated split-table machine.

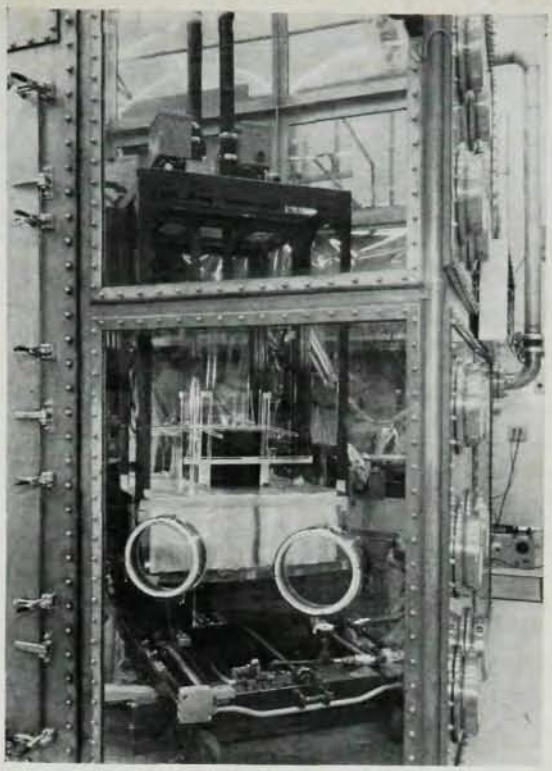

Fig. 7. Split-table machine with fuel assembled and faces together. Control drives visible at top

material is added; after personnel have left the area and the room is sealed, the faces are brought together by remote methods. Multiplication is then observed; the faces are opened, and further additions are made. When the material is separated (faces open) the assembly is well subcritical. We continue the procedure until criticality is achieved on closing the faces and inserting a small fuel-bearing rod.

Figure 7 shows the material assembled with faces closed. Some control- and safety-rod drives can be seen. Plutonium concentration in the cubes is 1.12 g Pu/cm^3, and the H/Pu atomic ratio for the assembly is 15. For criticality in the approximate form of a cube as shown, the quantity of plutonium is ~34 kg (30.5 × 30.5 × 31.8 cm). If the cube is reflected on all sides with plastic, critical dimensions are reduced and critical mass becomes 11.4 kg (core dimensions 22.9 × 22.9 × 19.2 cm).

This fuel composition makes a system in which the neutron spectrum is neither fast nor thermal; most fissions are produced by intermediate-energy neutrons. Another Monte Carlo calculation (by C. R. Richey) has given a rough estimate of the fractions of fissions caused by neutrons of different energies in the base system:

Energy (eV)	*Fissions (%)*
<0.4	12
0.4 — 10	20.7
10 — 1000	42.8
>1000	24.5

Neutron lifetime has been determined by S. R. Bierman with pulsed-neutron-source experiments. The lifetime is obtained from the measured ratio β_{eff}/l of the effective delayed neutron fraction to the neutron lifetime. The lifetime is defined as the average time a neutron lives in a finite system after being released in fission, with neutron loss from absorption and leakage. Measured lifetime was ~ 1.4 μsec in the unreflected cube and ~ 14 μsec in the reflected cube. In the reflected cube, neutron lifetime is the weighted average of core lifetime and the average time spent by neutrons in the reflector before returning to the core. (Lifetime of a thermal neutron in reflector material alone would be ~ 200 μsec.)

Value of the plutonium needed for the experiments is high (~ $1 million in this case). But the plutonium is not harmed in zero-power experiments, and its further use is not restricted. The PuO_2-plastic cubes are used to cover the density range of plutonium precipitates and polymers that can form in chemical separation plants.

Safety

Many automatic safety devices are built into our criticality system. With a loss of electric power, the table is opened by an air motor. If air pressure is not available, the table cannot be closed. Opening a door to the room automatically shuts the system down. Loss of power causes control and safety rods to move and shut the assembly down. Should criticality be approached too rapidly, causing a too rapid change of fission rate, the system shuts down automatically. If the neutron level rises above preset values, the system also shuts down. Moreover, operating personnel in the control room can take action at any time to shut down any critical assembly.

In general, criticality is of concern wherever fissile materials are handled in significant quantities (for example, in chemical plants for reprocessing spent fuel). There is the possibility that enough material may accumulate to constitute a critical mass and produce a chain reaction.

In nuclear safety we are concerned with prevention of accidental chain reactions in nonreactor environments. One must consider various in-plant factors that affect criticality of an individual unit or quantity of fissile material. Allowances must be made for neutron reflectors, interactions between associated vessels, and units in related operations (storage arrays, interconnected pipes, operations in adjoining hoods).

Many methods have been used for criticality control, and combinations of several methods are often used. These are some of the methods that have been used:

Process vessels are designed to be safe by geometry; that is, neutron leakage is great enough to prevent criticality *regardless of how much fissile material is present.*

Operating limits are imposed on material compositions such as density of fissile material in solution and quantity of diluent or moderator that can be present.

Amount of material handled in a single operation is limited to a subcritical amount; that is, a safe mass limit or batch control is applied.

Restrictions are applied to amount and form of material permitted in a storage array and on spatial distributions of the material.

Soluble or fixed neutron absorbers can be used to increase the mass limit or make a vessel safe by geometry.

In current practice, nuclear safety is based mainly on results of experiments together with interpretation and extrapolation of experimental data. But application of critical data to practical problems in processing plants often involves assumptions to cover prevailing conditions not duplicated in the original experiments. Then the usual procedure is to compare theory and experiment for situations measured in the laboratory. When satisfactory correlations have been effected and the calculational method verified, the method is then used to extend the data and provide answers to safety problems not yet studied experimentally.

First the reactor physicist must be able to apply theory and compute accurately the values measured in the laboratory. Doing so involves basic study of the physical theory of criticality as related to plutonium systems and neutron interactions.

If criticality conditions are precisely known, equipment can be specifically designed to prevent nuclear accidents, and methods can be devised for safe handling of plutonium. Programs like ours contribute to the good overall safety record of nuclear energy.

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