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*The Myth of Nuclear Explosions at
Waste Disposal Sites*

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CONTENTS

ABSTRACT	1
EXECUTIVE SUMMARY	1
I. INTRODUCTION: THE MYTH	2
II. PLUTONIUM PRODUCTION CONSIDERATIONS	3
III. CRITICAL MASS CONSIDERATIONS	4
IV. CRITICAL MASS OF A MIXTURE OF PLUTONIUM, SOIL, AND WATER	6
V. THE EFFECT OF THE RESONANCE AT 3200°C	8
VI. CHARACTERISTICS OF A POSTULATED POWER EXCURSION	10
VII. THEORETICAL MODEL FOR POWER EXCURSIONS	12
VIII. APPLICATION TO A PLUTONIUM-WATER-SOIL MIXTURE	15
IX. CONCLUSIONS	16
ACKNOWLEDGMENTS	17
REFERENCES	17

APOLOGIA

This study endeavors to show that an alleged event (postulated to be a nuclear explosion in a waste disposal site) did not, in fact, happen. To show or prove the negative is difficult at best, and because the alleged event is said to have occurred in a quite inaccessible spot with nothing but hearsay testimony available, the task would seem to be nearly impossible. However, reasonable assumptions can be made, and physically and mathematically rigorous analyses can be applied to conditions at the hypothetical site.

THE MYTH OF NUCLEAR EXPLOSIONS AT WASTE DISPOSAL SITES

by

William R. Stratton

ABSTRACT

Approximately 25 years ago, an event is said to have occurred in the plains immediately west of the southern Ural mountains of the Soviet Union that is being disputed to this very day. One person says it was an explosion of nuclear wastes buried in a waste disposal site; other people say it was an above-ground test of an atomic weapon; still others suspect that an alleged contaminated area (of unknown size or even existence) is the result of a series of careless procedures.

Since the event, a number of articles about the disposal-site explosion hypothesis written by a Soviet exile living in the United Kingdom have been published. Although the Soviet scientist's training and background are in the biological sciences and his knowledge of nuclear physics or chemistry is limited, people who oppose the use of nuclear energy seem to want to believe what he says without question. The work of this Soviet biologist has received wide exposure both in the United Kingdom and the United States.

This report presents arguments against the disposal-site explosion hypothesis. Included are discussions of the amounts of plutonium that would be in a disposal site, the amounts of plutonium that would be needed to reach criticality in a soil-water-plutonium mixture, and experiments and theoretical calculations on the behavior of such mixtures. Our quantitative analyses show that the postulated nuclear explosion is so improbable that it is essentially impossible and can be found only in the never-never land of an active imagination.

EXECUTIVE SUMMARY

The problems associated with producing a violent nuclear explosion in a disposal pit containing a mixture of plutonium, soil, and water are examined. The postulated situation is that waste solutions from a chemical processing plant contain some concentration of plutonium that is assumed to be discarded. The following matters are discussed.

The amounts of plutonium that are produced in reactors designed for that purpose are estimated for

“units” of 1000 megawatt years. Although the amounts of plutonium produced per reactor are relatively large, the amount that could have been discarded is relatively small if reasonably efficient chemical processing techniques were used. It is most unlikely that hundreds of kilograms could have been discarded.

The amount of plutonium required to produce a critical geometry in a mixture of soil and water and with water reflection is estimated. Generally, depending on moderation and reflection, either very large amounts of plutonium or a large volume fraction of water is required.

I conclude that the amount of plutonium required for criticality is more than could have accumulated in a waste stream; therefore, it is most unlikely that a critical geometry ever existed.

Nuclear power excursions* have been created in solutions both in the United States and France. These transient experiments are analyzed by use of a rigorously correct computer program. The quality of agreement between experiment and theory is examined and found satisfactory. Magnitudes of energy releases are presented and discussed. The difficulty (near impossibility) of creating an explosion in a solution reactor is illustrated by reference to experiments and also is analyzed theoretically.

The same theory, with minimum modifications, is then applied to postulated critical mixtures of soil, water, and plutonium. Magnitudes of energy releases associated with reasonable and even unreasonable physical and chemical situations are derived and discussed. The conclusion reached is that even if a critical situation were created and reactivity added at a most rapid rate, an explosion of more than trivial magnitude (equivalent to that of a few ounces of black powder) could not be created.

I. INTRODUCTION: THE MYTH

The earliest references to an event or events near Kyshtym in the Soviet Union in 1958 suggested a "catastrophic accident" (Ref. 1) and an "atomic explosion" (Ref. 2), and several reports³ written by émigrés and released by the CIA** mention explosions of one sort or another. However, the first suggestion (to our knowledge) that the event or events in question might have been a nuclear (not chemical) explosion associated

*A power excursion is the rise and subsequent fall of fission power that results from the addition of reactivity to a fissile system that is already close to the critical state. The reactivity can be in the form of a control rod motion, addition of fissile material (U-235, Pu-239, U-233), change of neutron reflector, neutron moderator, etc.

**In 1977, in response to the Freedom of Information Act requests, the CIA released censored reports of interviews of persons leaving the Soviet Union. Several of those interviewed mentioned explosions in connection with an event or events near Kyshtym in the late 1950s. All such reports are hearsay, as is Medvedev's information on the same subject obtained from different sources.

with nuclear wastes was made by Zhores Medvedev,⁴ a professional biologist. His colorful remarks included "nuclear reactions had led to overheating," "an enormous explosion, like a violent volcano," "radioactive dust and materials high up into the sky," "strong winds blew the radioactive clouds hundreds of miles," etc. In Refs. 5 and 6, the hypothesis of a nuclear explosion in a chemical waste disposal site was asserted again. Medvedev's strongest statements, however, were made in his book,⁷ published in 1979, and in his own review⁸ of his book. He discusses, in a very qualitative fashion, the production of plutonium in a reactor designed for that purpose, the chemical dissolution of the fuel, separation of plutonium and uranium from the fission products, and the final storage of those fission products. Without knowledge of actual practices (admitted to be the case), he postulates practices, conditions, and events to fulfill his earlier adamant insistence that a nuclear explosion occurred in a waste disposal site.

He mentions, as supporting evidence, the so-called Z-9 trench in the Hanford, Washington, reservation area in the U.S.^{9,10} He correctly states that low-level liquid wastes ("lean solutions"), containing very little plutonium and few fission products, were disposed of in this covered trench in the Hanford reservation soil. He incorrectly asserts that (1) the trench contained approximately 100 kg of plutonium, (2) a near-disaster was barely averted, and (3) this amount of plutonium is sufficient for nearly a hundred atomic bombs. His allegations relative to these points result either from inadequate review of the matter (the complete file is readily available) or from failure to understand the physics and chemistry involved because of his specialized biological training and background. The facts of the case show that the original analysis of the criticality of the trench was in error; the *upper limit* of plutonium in the ground was about 25 kg and may have been very much less.^{9,10} His reference to 100 atomic bombs is deemed by this author to be a clear exaggeration to support his earlier dogmatic assumption of nuclear explosions in a Soviet waste disposal site.

However, because of the number of reports (Refs. 1-3) that mention an explosion (or explosions) and because of Medvedev's adamant insistence that there was an explosion, we think that something of this sort could have occurred and may have been a part of whatever did occur in 1958 near Kyshtym. We do not attempt to prove what kind of explosion might have occurred or

even that one did happen. In this report, the requirements that must be imposed on a ground disposal site to cause it to explode "like a violent volcano" are presented and discussed. The plausibility of Medvedev's postulates will be discussed as may be appropriate, and implications, if any, to the environment will be addressed. We note in passing that there is no firsthand evidence of an "explosion."

To clarify the problem and define the issues to be addressed, we paraphrase Medvedev's assumptions and postulates and add comments as may be appropriate.

1. The discarded solutions contained significant concentrations of plutonium (this is an implicit assumption, not explicitly stated, but necessary for his thesis). Comment: the objective of the industrial activity was production and extraction of plutonium metal; the product is too expensive to handle carelessly.
2. The liquid wastes from a Soviet chemical processing plant were discharged into a pit in the ground, instead of into storage tanks. Comment: the use of tanks is accepted practice for storage of high-level waste throughout the world. I know of no reason whatsoever for a different practice in the Soviet Union.
3. The plutonium precipitated on soil particles in a thin layer of soil near the surface. Comment: this behavior of plutonium is in accord with existing data; plutonium will precipitate from solution in a near-insoluble form close to the surface of the ground.
4. A great many kilograms of plutonium accumulated in the disposal pit. Comment: because of his admitted lack of nuclear expertise, his estimates are very qualitative. The amount that could be produced and the mass actually required for criticality are calculated in this report for various conditions.
5. Given a large mass of plutonium in the postulated disposal pit, he hypothesized a supercritical condition was created by inflow of water. Comment: again, because of his lack of knowledge of criticality physics, the matter is not discussed quantitatively. Criticality is one of the major topics of this report (see Secs. III and IV).
6. The supercritical condition caused by the plutonium water moderation and reflection created "an

enormous explosion, like a violent volcano." Comment: the possibility of this condition occurring is examined quantitatively below.

The plan of this report is to examine the several requirements needed to produce a near-critical system and set the stage for a nuclear reaction in a water-moderated mixture of soil and plutonium. Realistic conditions and assumptions will be taken; where possible, reference will be made to known practice and history in the United States, France, or the United Kingdom. Reasonableness and/or probability will be judged as data are developed. The analysis of conceptual nuclear power excursions in a waste solution storage site in the ground will be examined in three stages. First, an estimate will be made as to the possible plutonium production rate and the fraction that might have been discarded; second, the criticality characteristics of plutonium, plutonium solutions, and, in particular, characteristics of plutonium solutions in a rectangular basin in soil will be examined; and third, the nuclear power characteristics of a supercritical, soil-water-plutonium mixture will be studied and the possibility of explosions evaluated.

II. PLUTONIUM PRODUCTION CONSIDERATIONS

The amount of plutonium potentially available to have accumulated in a disposal pit is basic to this analysis, of course. This amount is not available, but its order of magnitude can be estimated fairly readily. The thermal power of the early World War II plutonium production reactors (using natural uranium metal as fuel) in the United States was a few hundred megawatts, but later, with the availability of slightly enriched uranium and the advantage of operating experience, the power level was increased over a period of years to several thousand megawatts. It is reasonable to assume a comparable history in the Soviet Union. Thus, a "unit" of 1000 MW for a year (300 days, to allow for refueling and downtime) is convenient, and independent assumptions can be made in regard to how many units per reactor and how many reactors for how many years at one site need be considered.

Given the assumptions of 1000 MW for 300 days, 0.67 plutonium atom created per fission,^{11,12} and the

equivalence of 1 W and 3×10^{10} fissions/s, the plutonium production rate would be 205 kg/year. The plutonium is created within the uranium fuel, and this fuel must be dissolved in acids (usually nitric). Separation of the fission products, uranium, and plutonium is accomplished by wet chemistry techniques. Depending on the process and management philosophy, less than 100% of the theoretically available plutonium is generally recovered. Losses occur, first because the cladding hulls are not fully dissolved, and second because the wet chemistry separation process itself creates losses. Thus, if the hull dissolution and wet chemistry were 99% effective (reasonable), the amount lost would be about 2 kg/year. If the recovery were only 95% effective (very poor), about 10 kg might not be recovered. Some operations in the United States recover as much as 99.9% of the plutonium available in solution.

Thus, if an average of several thousands of megawatts were available for several years (we note that in the 1950s the Soviet Union must have been increasing their plutonium production capacity both in numbers of reactors and in fission power of each reactor), it is conceivable that several tens of kilograms could have been discarded in solutions containing fission products. It is safe to conclude that hundreds of kilograms would be an unreasonably large amount to have been discarded. As will be developed later, tens of kilograms is not sufficient plutonium to create the postulated critical system.

III. CRITICAL MASS CONSIDERATIONS

The mass of a fissile material (Pu-239, U-233, U-235) needed to form a critical* system or critical assembly is known accurately for a very large number of different material densities, geometrical arrangements, diluents, poisons, and structural materials by virtue of hundreds of experiments¹³⁻¹⁵ performed during the past 37 years. These experiments are correlated and tied together by use of rigorously correct neutron transport computer programs¹⁶ that have been used in many laboratories throughout the world. The agreement between ex-

*A critical mass of fissile material is that amount of U-235, U-233, or Pu-239 along with diluents, poisons, and reflectors in a defined geometry that will just sustain a constant fission rate (any power level) or neutron population. An experimental arrangement of materials is often called a "critical assembly" and is brought to the critical state by remote control and operated at a very low, near-zero, power.

perimental and theoretical results is extraordinary, and for reasonably definable geometrical situations, the results of theory are regarded nearly as well as those of an experiment. For poorly defined mixtures or geometries, bounding assumptions can be (and are) made to assure a conservative result, that is, overestimate criticality.

Calculations of this sort make use of tabular sets of neutron cross sections. For a given material, say Pu-239, the neutron cross sections¹⁷ must reflect the known experimental phenomena such as neutron capture with fission, capture without fission (as caused by a poison), and scattering with a reduction in energy of the neutron. Because neutrons are born with high energy following fission, the various interactions must be modeled from several million electron volts down to thermal energy (about 1/40 eV). The cross sections used in the computations for this study are from the Hansen-Roach 16-group set,¹⁸ that is, a compilation that divides the energy region from multimillion electron volts to thermal energy into 16 regions. Each isotope of each chemical element assumed to be in a computational model must have the appropriate cross sections defined. The excellence of the combined use of these cross sections and (numerical) neutron transport equations is tested by results of studies that compute a wide variety of critical conditions. In the applications of the functions used, as mentioned above, the comparison of theoretical critical radius or mass to the experimental value is surprisingly good.¹⁴

The neutron cross sections for capture, fission, and scatter for many elements in and near the thermal energy (room temperature) region are characterized by a proportionality to the reciprocal of the neutron velocity, such as

$$\text{cross section} \propto \frac{1}{v} . \quad (1)$$

Because the neutron kinetic energy is proportional to the square of the velocity, the cross section is then proportional to the reciprocal of the square root of the energy, such as

$$\text{cross section} \propto \frac{1}{\sqrt{E}} , \quad (2)$$

and this is the functional form commonly found in graphical presentations of cross sections.¹⁷

A noteworthy exception to this general rule [Eq. (2)] is the near-thermal fission cross section of Pu-239. The exceptional character of this isotope is caused by a

resonance* in the fission cross section at 0.3 eV or at about 3200°C. The magnitude of the cross section at this effective temperature is larger than at thermal temperatures. This resonance seriously perturbs the $1/v$ character of the neutron cross section; at thermal energies, the fission cross section is decreasing as the neutron energy is increasing but less rapidly than $1/v$. The general character of the cross section of plutonium for these energies is shown in Fig. 1.

The importance of this resonance is the effect it might have on the criticality or reactivity of a water-moderated plutonium system should it become critical and increase in temperature. If all neutrons were at exactly the thermal energy corresponding to the temperature of the material and if all fissions were caused by these neutrons, the system could become more reactive as the temperature increased and could be at a maximum when the temperature reached 3200°C. This behavior of neutrons is not the case, of course, but if the other materials (for example, hydrogen) have cross sections strictly proportional to $1/v$, the effect of the resonance can be enhanced somewhat.¹⁹ That this resonance might influence significantly the supercriticality characteristics of the postulated mixture of soil, water, and plutonium was first brought to my attention by Freeman Dyson in 1980.²⁰

We have examined this matter carefully and found that at least two factors diminish the effect of the resonance in creating, as postulated by Dyson in Ref. 20,

*A resonance in a neutron capture (or fission) cross section may be compared to a person pushing and adding energy to a second person sitting on a swing. For ease in visualization, we imagine a swing from a high support so its period is long relative to the pusher's agility. If the person pushing does so at or nearly at the obvious time, she is "in resonance" with the swing. If she pushes too frequently (too agile), or too infrequently (too lazy), or if in a direction different from the plane defined by the motion of the swing (intoxicated), she is "out of resonance." If she attempts to "stick," that is, to join the person on the swing, it is much easier and with much higher probability of success if she is "in resonance" and acting in the same plane as the motion of the swing. If the swing is the plutonium atom and the pusher is the neutron, it is reasonable that the probability of sticking is greatest if the contact is made at the resonance frequency. In the neutron and the plutonium atom interaction, "frequency" and "angle" are "just right" at 3200°C. The combined properties of the two nuclei, as they join to become (momentarily) Pu-240 in an excited state, determine the characteristics of the resonance. The capture cross section at this energy is very large but decreases sharply at lower energies but especially rapidly at higher energies.

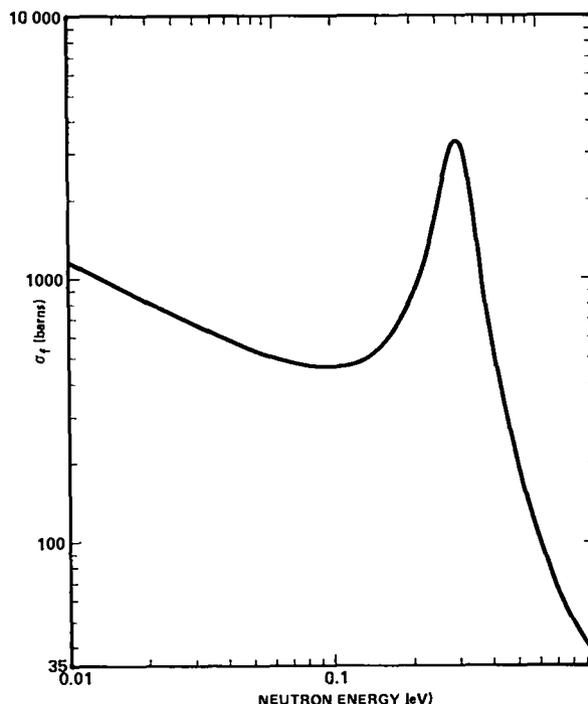


Fig. 1. Absorption cross section (fission plus capture) of Pu-239 as a function of incident neutron energy. Room temperature, about 68°F or 20°C, is equivalent to 1/40 eV, whereas the Pu-239 resonance at 0.3 eV is equivalent to about 3200°C.

an autocatalytic power excursion, that is, one during which the reactivity increases as the power increases. These are as follows: (1) not all fissions are caused by neutrons at exact equilibrium with the thermal energy of the material. The equilibrium character of neutrons at a given temperature is described by what is known as a Maxwellian distribution with some neutrons at what are effectively higher and lower temperatures. As an example, if the peak of the neutron energy distribution is at 0.3 eV but if a quarter of the neutrons are at a higher energy, where the cross section is lower by a factor of 100, the effect of the resonance is decreased. Thus, the distribution of neutrons can lessen the effect of the resonance. And (2) to moderate the neutrons, water must occupy a large fraction of the volume, more in fact than may be available in sandy soil.²¹ If water is present, the character of the postulated power excursion will be dominated by the response of water to the deposited fission energy, and the very high temperatures will never be achieved as long as water is present in any quantity. The reactivity changes in this case would be dominated by thermal expansion, bubble formation, and boiling, all of which will be discussed in more detail below.

2. The composition of the soil was simplified to consist of SiO₂ (89%), Al₂O₃ (6.6%), and FeO (4.4%). The full density is 2.43 g/cm³ (Ref. 10).
3. The plutonium in the soil was concentrated near the surface as has been observed.¹⁰ The plutonium density distribution function is given in Table I. This distribution is in reasonable accord with actual observations of plutonium concentrations in soil that has absorbed solutions containing very low levels of plutonium. Apparently, soil particles selectively absorb and hold plutonium very tightly on their surfaces, an interesting property per se.
4. Plutonium was assumed to be mixed in as a metal powder or an oxide powder at several different concentrations. Each of these concentrations was assigned to the position of the density function 1.0 for the top layer with concentrations dropping off as specified in assumption 3 above.
5. The isotopic concentration was 95% Pu-239 and 5% Pu-240, again arbitrary but reasonable.
6. No neutron poisons were assumed to be present. This assumption is very conservative and maximizes the calculated reactivity and minimizes the mass required to achieve criticality. Any real solution would have neutron absorbers in at least the acids and some of the fission products.
7. Two water volume fractions were assumed as given in Table II. The amount of water for Case I is not unreasonable,²¹ but that for Case II is extraordinary and is quite unrealistically high. It would be much more like a thick liquid than wet sand.

TABLE I. Relative Density of Plutonium in Soil as a Function of Depth

Soil Depth (cm)	Relative Density of Plutonium
0-2	1.0
2-8	0.45
8-15	0.15
15-23	0.04
23-30	0.015
30-50	0.0

TABLE II. Volume Fractions of Soil and Water Assumed for This Study

Material	Volume Fraction	
	Case I	Case II
Soil	0.7	0.4
Water	0.3	0.6

8. The computation of reactivity used a transport code, ONEDANT²⁴ with Hansen-Roach 16-group cross sections.¹⁸ ONEDANT was used in planar (slab) geometry with a buckling correction²⁵ to simulate the finite size of the disposal pit. The validity of this method of computation has been discussed above.

Given these assumptions and conditions, the critical concentration of plutonium was computed for several areas (within the postulated disposal site conditions) and thicknesses of water reflection. The relative distribution of plutonium, as a function of depth as shown in Table I, was held constant. These criticality data are assembled in Table III.

The major characteristic of these postulated, critical mixtures of sand, water, and plutonium is the thin, slab-like layer of plutonium near the surface. For this geometry, the required mass for criticality increases very rapidly with a very slight diminution of the concentration and increase in area to compensate. This effect is evident, for example, for the four critical masses in Case II with a 10-cm water reflection. The concentration decreases only by 20%, but the mass required for criticality increases by a factor of 130, from 4.7 to 610 kg.

The critical amounts of plutonium for the full 9-m × 18-m slab are extraordinary, ranging from 600 to 4000 kg. These are very large amounts, and to assume that this quantity would be discarded suggests either extreme naiveté or a determination to confuse and befog a technical matter.

The whole area of the postulated disposal site need not be used to create a critical system, however. Smaller areas can be made critical with increased water reflection, water volume fraction, and plutonium concentration (note that some plutonium densities are extraordinarily large in Table III). We comment that Case II, 60% water by volume, is sufficiently unrealistic that it can be discarded on this basis alone but, nevertheless, is discussed further below to complete our argument. The

TABLE III. Computed Critical Concentrations and Masses for Two Volume Fractions of Water, Various Areas, and Amounts of Water Reflection

Area (m ²)	Water Reflection (cm)	Plutonium Concentration (g/l)	Critical Mass (kg)
Case I: 30% Water			
4	0	565	140
10	0	530	325
10	1	400	245
10	5	194	120
10	10	150	93
162	0	430	4300
162	1	340	3400
162	5	143	1430
162	10	114	1140
Case II: 60% Water			
1	10	76	4.7
4	10	65	16.1
10	0	155	96
10	1	121	75
10	5	77	48
10	10	64	40
162	0	136	1360
162	1	114	1140
162	5	71	710
162	10	61	610

critical masses for the smaller areas of Case I range from 93 to 325 kg. Thus, a critical system for a fraction of the area of the pit can be postulated, but the mass of plutonium required is still very large, and it is unlikely that such large, expensive, and precious amounts would knowingly or accidentally be discarded.

The conclusion of this section is that to create a critical system of plutonium in soil would be most expensive in the mass of plutonium, requiring high concentrations or unrealistic amounts of water. Existence of such a critical system is very unlikely.

V. THE EFFECT OF THE RESONANCE AT 3200°C

The importance of this resonance was mentioned above. Briefly, if all neutrons causing fissions were in

exact equilibrium with the material temperature, the reactivity could rise (after some initial heating) to a maximum at the resonance. The resonance is illustrated in Fig. 1 for more energetic neutrons.

This effect could create what is called an autocatalytic power excursion, because as temperature rises, the reactivity rises, the reactor period shortens, and the power increases more rapidly. Such a self-feeding process could increase until the resonance temperature was reached.

However, as mentioned above, the neutrons are born as fast neutrons with energies in the million-electron-volt range. The amount of moderation (as with hydrogen) is important in calculating the reactivity because fission cross sections are larger at thermal or near-thermal temperatures. Hence, the reactivity at each temperature must reflect the neutron energy distribution at that temperature. Some neutrons would cause fissions at high energies and some while being reduced in energy, but the dominant effect on reactivity would derive both from the distribution in energy of the thermal neutron population and from the existence of absorbers such as hydrogen.

One can visualize the effect of the distribution of neutrons by imagining, for example, the temperature (energy) to be 460°C (0.1 eV). The peak of the distribution would be at this point: neutrons at lower temperatures would have a higher cross section, and neutrons at higher temperatures (closer to the resonance) also would have a higher cross section. For this condition, the effect of the neutron distribution is to raise the effective cross section (see footnote on p. 5). However, if the equilibrium temperature were just at the maximum of the resonance, neutrons at both higher and lower temperatures would see lower cross sections. Thus, the effect of the distribution would be to wash out the influence of the resonance to some extent. The modified cross section, with allowance for a Maxwellian distribution in neutron energy, is illustrated in Fig. 3. The effective maximum is shifted to a lower energy, because the cross section (Fig. 1) decreases very sharply above the resonance at 3200°C.

A convenient way to characterize the moderation and the distribution is to express the moderation (and absorption) by the ratio of hydrogen to plutonium atoms (H/Pu). An appropriate reactivity parameter is the reproduction factor for the infinite, unbounded (in dimension) system, the so-called k_{∞} . This parameter depends only on neutron temperature and H/Pu ratio and not on the assembly size or density or boundary

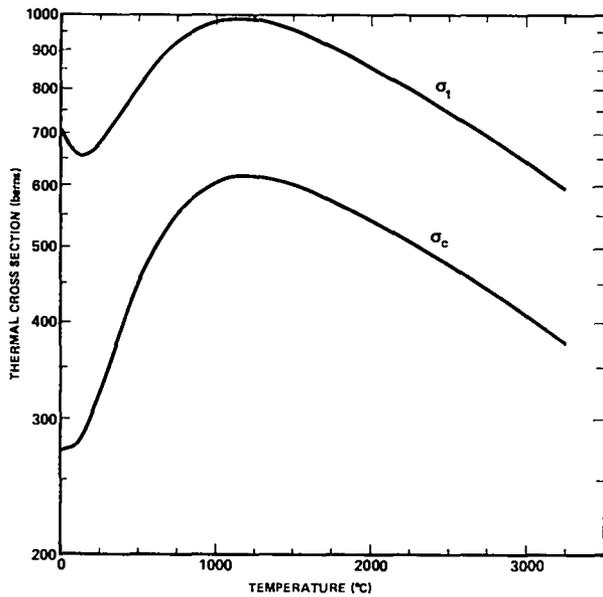


Fig. 3. Absorption (σ_a) and total (σ_t) cross sections for plutonium with allowance for a Maxwellian distribution of neutrons about energy E. These curves can be compared with that in Fig. 1, in which the neutrons are monoenergetic.

conditions. The result of these computations is presented in Table IV and illustrated in Fig. 4.*

Table IV or Fig. 4 shows that, to have a positive temperature coefficient (that is, reactivity at room temperature increasing as temperature increases), the ratio of hydrogen atoms to plutonium atoms (H/Pu) must be greater than 1000. For H/Pu = 800, the assembly

*These data were contributed by Gordon Hansen, Los Alamos National Laboratory, 1981.

(mixture of materials) must get to about 300°C before the coefficient becomes mildly positive, whereas, for H/Pu less than 500, there is no region for which the reactivity increases with temperature.

To relate these data to the imaginary assembly at hand, the H/Pu atom ratios can be calculated for the plutonium and water densities bounding or close to those presented in Table III and for the soil depths and density functions presented in Table I. These data are given in Table V.

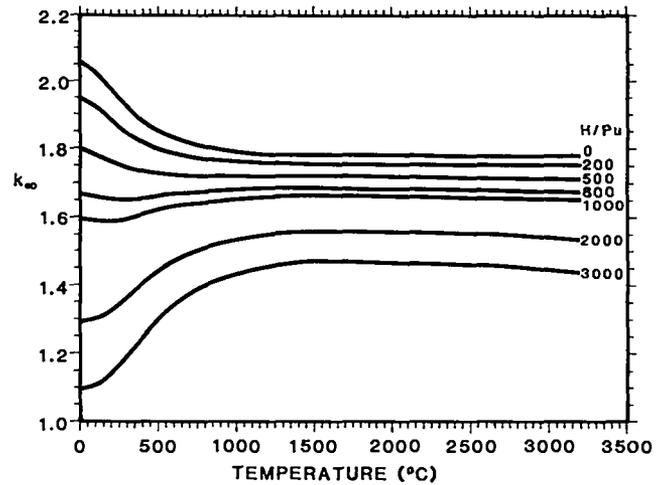


Fig. 4. The neutron reproduction factor (k_{∞}) for an unbounded system (size not limited in any direction) consisting of Pu-239 and water versus the equilibrium temperature of the mixture. Various amounts of water are defined by the hydrogen atom/plutonium atom ratio, and cross sections are taken from Figs. 1 and 3. The apparent dominant effect of the Pu-239 resonance in Fig. 1 is diminished by the neutron energy distribution and the effect of hydrogen absorption. See also comments in Ref. 19.

TABLE IV. Infinite Reproduction Factors (k_{∞}) for Mixtures of Plutonium and Water

H/Pu	Temperature (°C)								
	17	130	300	540	890	1470	2050	2630	3210
0	2.060	2.015	1.922	1.840	1.795	1.781	1.778	1.776	1.778
200	1.945	1.912	1.847	1.792	1.763	1.756	1.753	1.750	1.750
500	1.795	1.776	1.745	1.726	1.718	1.720	1.717	1.713	1.711
800	1.667	1.658	1.653	1.664	1.675	1.685	1.682	1.678	1.673
1000	1.591	1.588	1.597	1.625	1.648	1.663	1.660	1.655	1.648
2000	1.296	1.310	1.366	1.455	1.523	1.559	1.556	1.550	1.536
3000	1.093	1.115	1.193	1.317	1.416	1.468	1.465	1.458	1.438

TABLE V. Hydrogen-to-Plutonium Atom Ratios for Two Volume Fractions of Water, for Four Plutonium Concentrations, and as a Function of Depth in Soil

Soil Depth	Density Function	H/Pu for Given Plutonium Densities					
		Case I: 30% Water			Case II: 60% Water		
		100 g/l	200 g/l	500 g/l	20 g/l	100 g/l	200 g/l
0-2	1	76	38	155	760	50	76
2-8	0.45	173	86	344	1730	115	173
8-15	0.15	530	265	103	5310	354	530
15-23	0.04	1990	990	387	19900	1320	5300
23-30	0.015	5300	2650	1033	53000	3530	5300

From this table and Table IV, it can be seen that the influence of the resonance at 3200°C will be small at most. The H/Pu ratio rises sharply with increasing depth of soil, but the temperature rise of these regions of such an imaginary fissioning system would drop off rapidly with depth simply because the relative plutonium atom density and hence fissions decreases rapidly.

The conclusion of this section of the study is that the resonance in the fission cross section of plutonium at 3200°C would have little, if any, significant influence on the characteristics of a postulated power excursion in the assembly of materials as postulated. This conclusion makes the proposition that a severe nuclear power excursion could occur on a waste disposal site in soil even more improbable, if not practically impossible. Nevertheless, for completeness, the characteristics of power excursions will be reviewed and applied as appropriate to the imaginary plutonium-sand-water system.

VI. CHARACTERISTICS OF A POSTULATED POWER EXCURSION

The preceding discussion developed characteristics that would be required for a critical mixture of plutonium, soil, and water. The dominant characteristics are the large amounts of water needed (volume fractions of 0.3 and 0.6 water were used as examples) and the very large mass of plutonium required.* The large amount of water immediately suggests that the fissioning characteristics of the mixture will be comparable to the fissioning characteristics of water solutions of uranium

*A critical slab can be created with lesser amounts of water, but the amount of plutonium required increases astronomically.

or plutonium. To provide background for an analysis of postulated nuclear power excursions in the plutonium-water-soil mixture, we will examine briefly the characteristics of solutions.

The importance of understanding solution criticality and power excursion behavior in water reactions arises from the use of solutions in spent-fuel chemical processing plants. Generally, spent fuel is dissolved in strong acids (primarily nitric), and the separation of plutonium, uranium, and fission products is completed by solution chemistry techniques. The importance of control of criticality to the safety of operating personnel and to integrity of equipment is obvious. Indeed, in the history of nuclear criticality accidents and incidents,²³ about a dozen have occurred with solutions in processing plants or during the performance of solution experiments. Two of these accidents resulted in fatalities, but the characteristics of all were such that no physical damage to equipment resulted. Personnel safety requirements make the need for experiments and precise calculations obvious.

As mentioned above, a very large number of criticality experiments with solutions have been completed. These serve to provide validation points for theory, and, indeed, the correspondence between theory and experiment is extraordinary, suggesting that the various parameters are well understood. Safety in processing plants is controlled in part by assuring that solutions of a given concentration are allowed to be only in containers of a "safe" size or in a container that has fixed poisons. However, several of the solution accidents occurred because a solution reached a container of the wrong size or one with insufficient poison, the solution became supercritical, and a power excursion developed.

Because of this history and because of expectations of large-scale chemical processing, the French Commissariat à l'Énergie Atomique (CEA) authorized a series of experiments in the late 1960s to investigate power excursions in uranium solutions. The objective was to establish the level of risk in order to design additional protection if needed. The experiments performed were straightforward and applicable to the problems for which they were designed and, unintentionally, also applicable to the plutonium-soil-water mixture postulated by Zhores Medvedev. The series of experiments was designated "Consequences Radiologiques d'un Accident de Criticité" (CRAC) (Ref. 26).

The CRAC experiments followed a series of experiments in the United States known as the Kinetics Experiments in Water Boilers (KEWB) (Ref. 27). This latter program was started in the mid-1950s and extended into the early 1960s. In this case, the solution was in the form of a small reactor, fully equipped with cooling coils and control rods. The KEWB reactors were used both for various steady-state experiments and for transient or excursion experiments. The transient or excursion experiments were initiated by control rod motion and were contained, as opposed to the CRAC experiments, which were initiated by pouring uranyl nitrate solution into an open tank already containing a near-critical volume of solution. In spite of the differences of geometry, containment, and mode of initiation, the early stages and most significant part of the power excursion were comparable in behavior. The same statement could be made for a sudden change of reactivity from a different cause, say by addition of a reflector.

If a subcritical volume of solution is made supercritical, the response of the system can be described easily, at least qualitatively. Upon becoming supercritical, the system fission power begins to rise, the rate depending on the amount of reactivity added; this rate can be very slow or very fast, for example, equal to a doubling of power once or twice per millisecond in extreme cases. This rate is many, many times faster than would occur by action of natural processes. This power rise would continue until solution expansion mechanisms would allow increased leakage of neutrons, consequent reduction of reactivity, and reduction of power to a low level. This first rise and fall is often called the power spike or the transient. The peak power attained during this spike is higher than the power during any following fluctuation and is proportional to a measure of the rate of adding reactivity or to the amount added. Typical

CRAC experimental fission rate traces are reproduced in Figs. 5 and 6. The oscillatory behavior following the first power spike is supported by the rate of pouring solution and by the existence of delayed neutrons, whose precursor nuclei were created during the spike. Figure 7 illustrates the solution temperature measured during one of the experiments; the time at which boiling occurs is several hundred seconds after the start of the experiment, long after the quenching of the power spike.

The mechanisms that limit the rise of power in the first spike and control the subsequent power level are three in number and are naturally occurring in any solution reactor. Thermal expansion is first to be evident; as the

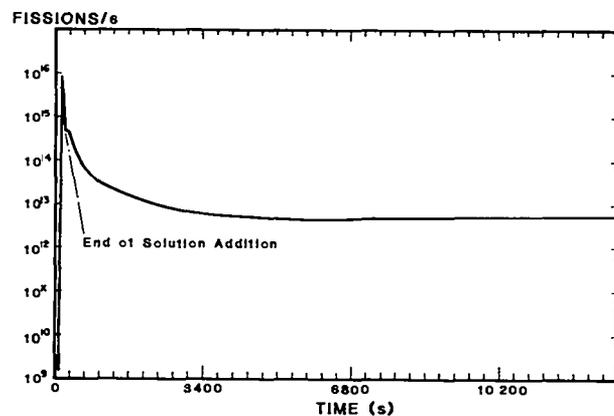


Fig. 5. Relative fission rate versus time for CRAC experiment number 04. The CRAC program,²⁶ "Consequences Radiologiques d'un Accident de Criticité," consisted of experiments during which a solution of enriched uranium nitrate was poured into a large diameter cylinder to heights well in excess of the critical height. The result was a fission power excursion, an example of which is illustrated in this figure.

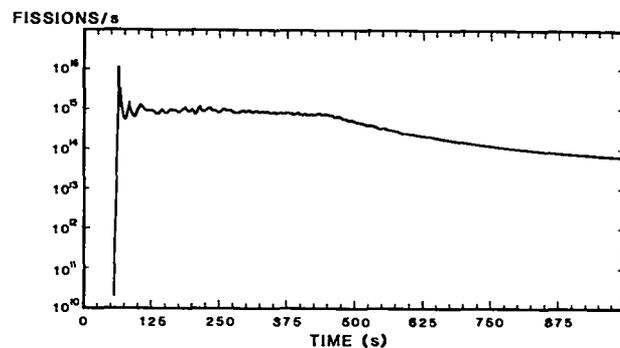


Fig. 6. Relative fission rate versus time for CRAC experiment number 12.

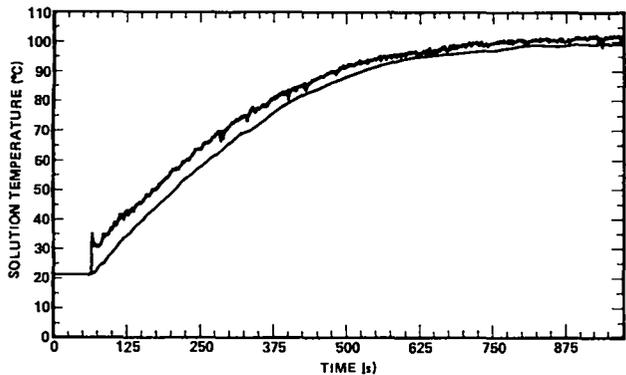


Fig. 7. Temperature of the solution versus time for CRAC experiment number 12. The upper curve was near the center, while the lower was at the boundary of the container. Note that boiling temperatures were not achieved for 725 s after the start of the experiment.

temperature rises, the solution expands and even a small amount is significant for increasing neutron leakage. The second is bubble formation resulting from fission products passing through water and creating hydrogen, oxygen, and steam. These tiny bubbles also cause the solution to expand in volume and increase neutron leakage. Finally, and much later, boiling temperatures are reached, and, usually, the onset of boiling reduces reactivity and power substantially more.

The CRAC experimental program²⁶ consisted of a large number of power excursions (about 40) in two cylindrical vessels of diameters 30 and 80 cm. The uranium in the water-uranyl nitrate solutions was highly enriched, about 93% U-235, and solutions of various concentrations were used. Generally, at least three important conclusions can be derived from these experiments that are significant to the study at hand. First, the reactivity quenching mechanisms, thermal expansion, bubble formation, and (later) boiling are invariably present; the first two limit the magnitude of the first power spike. Second, the power excursions are not destructive; some splashing of solution occurred in nearly every experiment, but the most violent of these splashes was only a bit more than two meters high. The most violent experiment created a hydrostatic pressure*

*The hydrostatic pressure is a liquid-phase pressure, the characteristics of which are a very steep pressure-volume relationship. That is, a very small increase in volume (or decrease of density) reduces pressure dramatically. This is different from a pneumatic (compressed gas) pressure, which, for the same magnitude, stores much more energy. The hydrostatic pressure generally will create little motion or damage.

high enough to bend the aluminum supports of the tank. These measured (and computed below) hydrostatic pressures are strictly proportional to the peak power in the first spike raised to the two-thirds power as is illustrated in Fig. 8. And third, for the more severe transients, the first power spike invariably dominates the experiment in terms of power, pressure, splashing, etc. Boiling of solution never occurred until long after the first power excursion, and boiling did not contribute to the early splashing of any of the experiments. A general conclusion is that to create a highly destructive power excursion in a solution system with a free (air) surface is most difficult if not impossible.

Because of these experiments with solutions of fissile materials and because of the required conditions for criticality for the postulated soil-water-plutonium mixture (almost a thick or dense solution), a reasonable conclusion is that not only is a critical system most unlikely, but that a severely violent (that is, highly explosive) excursion is nearly impossible. Nevertheless, a theoretical model will be applied to the problem for completeness.

VII. THEORETICAL MODEL FOR POWER EXCURSIONS*

The theoretical model used to analyze the CRAC and KEWB experiments is part of a computer program designed to integrate the neutron kinetics equations with the addition of a linear response to energy deposition, rise in temperature, or appearance of pressure. Many such programs exist for applications in the field of reactor physics, dynamics, and kinetics. One of the earliest such programs was the Los Alamos RTS code,²⁸ which was used in the late 1950s and early 1960s to elucidate questions in regard to delayed neutrons, criticality accidents, and reactor dynamics. The particular formulation used in this study is the "Mackin Program" (Ref. 25) created for the specific purpose of studying the extensive KEWB and CRAC series of experiments mentioned earlier. Discussion herein will be brief because complete descriptions of the code, testing, and applications can be found in Ref. 25.

In all such computations, a basic assumption made is that the rate of change of reactor power (rate of change

*The theoretical model used for analyses in this study and the computer calculations performed to illustrate the physical effects are the work of Harry M. Forehand, Los Alamos National Laboratory.

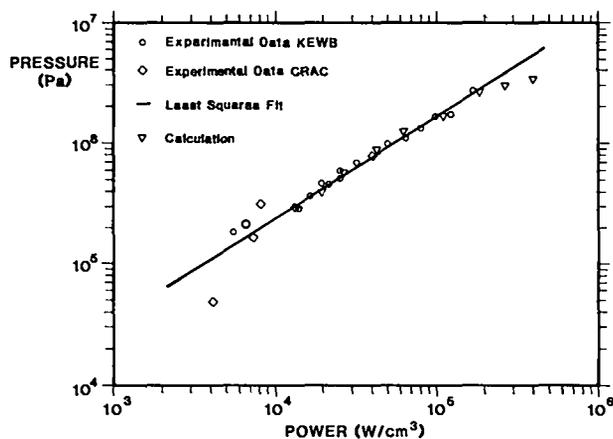


Fig. 8. Hydrostatic pressure developed in the initial power spike versus the peak specific fission power. The experimental data show a relationship: pressure proportional to (specific power)^{2/3}. The theory verifies this relationship but also shows that, for more extreme conditions, the pressure increases even less rapidly as peak power increases.

of neutron flux) is proportional to the existing reactor power (neutron flux). The proportionality factor is the reactivity in excess of the critical condition or state. For studies of the solution experiments, the complete set of delayed neutrons is incorporated as an integral part of the program. The feedback from deposited fission energy is associated with the reactivity proportionality factor; that is, the reactivity is a function of temperature, density, and state of solution (microbubbles, boiling, etc.). In abbreviated form, if

$$\begin{aligned}
 E(t) &= \text{fission energy deposited (a function of time),} \\
 n(t) &= dE/dt = \text{reactor power, a rate of change of} \\
 &\quad \text{energy (also a function of time), and} \\
 \tau &= 1/\omega = \text{reactor period or rate of change of} \\
 &\quad \text{reactor power,}
 \end{aligned}$$

then the following functional relationship holds:*

$$\frac{d}{dt} n(t) = \omega n(t) = \omega \frac{dE}{dt} \quad (4)$$

If

*If additional investigations are pursued, it is important to note that nomenclature is not universally the same. In this discussion, that chosen is taken to be the same as in Ref. 25 for convenience to the reader. Other references may have a different notation. I apologize for the double use of $\rho(t)$ both for density and for reactivity.

$\rho(t)$ = reactivity as a function of time,
 β = the fraction of neutrons that are delayed in time,
 ℓ = the neutron generation time or the time for the neutron density or fission power to rise by a factor of $e = 2.718$,
 $C_i(t)$ = concentration of delayed neutron precursors, and
 λ_i = decay constant,

then

$$\frac{d}{dt} n(t) = \left[\rho(t) - \beta \right] \frac{n(t)}{\ell} + \sum_{i=1}^6 \lambda_i C_i \quad (5)$$

$$\frac{d}{dt} C_i(t) = \beta_i n(t) - \lambda_i C_i(t) \quad (6)$$

and the reactivity $\rho(t)$ can be expressed by

$$\rho(t) = \alpha_0 T(t) + \alpha_1 T^2(t) + \phi V(t) \quad (7)$$

in which $T(t)$ is temperature, $V(t)$ is volume, α_0 and α_1 are the linear and quadratic components of the temperature coefficient of reactivity, and ϕ is the void coefficient of reactivity.

The temperature (or temperature rise) is computed directly from the deposition of fission energy and known heat capacities. The microbubble volume $V_m(t)$ is given by the functional relationship

$$V_m(t) = \begin{cases} 0 & \text{for } E < E_c \\ \frac{1}{2} \nu [(E^2(t) - E_c^2)] & \text{for } E(t) \geq E_c \end{cases} \quad (8)$$

in which,

ν = constant relating energy density to radiolytic bubble volume,* and
 E_c = threshold energy for production of microbubbles.

*A comparable model has been used that relates fission energy density to pressure resulting from the radiolytic gases created by fission particles. This is also described in Ref. 25.

This set of differential equations has been programmed for a fast digital computer and tested against experimental data such as those from the CRAC and KEWB series of experiments. Pertinent parameters are the density of U-235 in the solution (this affects the neutron lifetime), the reactivity insertion rate (for example, addition of solution or motion of control rods), past history of operation (affects delayed neutrons), and a volume of solution large enough that container walls are, at most, a minor perturbation. Observables that should be matched (for example, from Figs. 5, 6, and 7) are the reactor period, the peak specific power, the energy release and temperature rise for the first spike, and prediction of the hydrostatic pressures created, if any. Prediction of the onset of boiling should follow without difficulty.

The specific fission power, as a function of time, is a primary output of the Mackin Code; from this power density and the derived energy density, the temperature rise, bubble volume, expansion, etc., are computed. Examples of this output are illustrated in Fig. 9. At zero time, reactivity insertion rates of 10, 5, 1.05, 0.5, or 0.1 $\$/s^*$ were imposed on the computational model, and the computed specific power rose exponentially until the reactivity quenching mechanisms were strong enough to stop the rise. The internal pressure was a maximum at this time, as can be deduced from Figs. 8 and 9. The traces on Fig. 9 can be compared, qualitatively, to the shape of the early portion of the power traces in Figs. 5 and 6.

A quantitative comparison of Mackin computations to KEWB and CRAC experimental data is shown in Figs. 10 through 13. Figures 10 and 12 compare the calculated and experimental peak specific power for both series, whereas Figs. 11 and 13 illustrate the fission energy density deposited in the power spike to the time of peak specific power. Generally, the theoretical prediction of peak specific power is very good and not significantly different from the apparent fluctuations in the experimental data. The integrated specific power, or energy density deposited in the power spike to the time of peak specific power or for the complete spike, is less precise for relatively long period transients but becomes more precise for the short period experiments. These latter

*The dollar (\$) was proposed as a unit of reactivity in 1945. It is the interval of reactivity between delayed critical, where all neutrons are needed including those delayed in time, and prompt critical, where only prompt neutrons are needed to maintain a constant fission rate. A rate of 0.1 $\$/s$ is a very rapid rate of change of reactivity for a reactor.

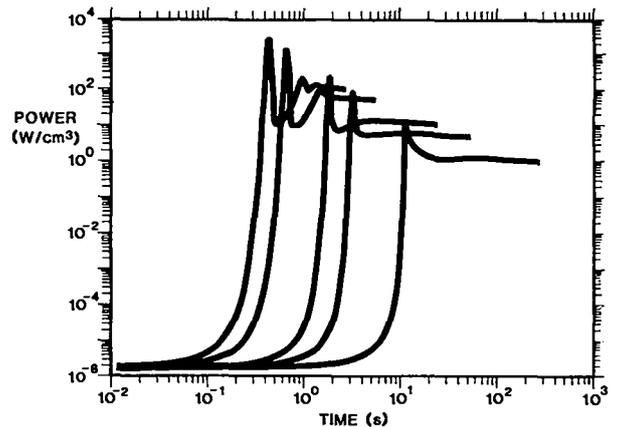


Fig. 9. Specific fission power versus time as computed by the Mackin Code. The reactivity insertion rates were 10, 5, 1.05, 0.5, and 0.1 $\$/s$.

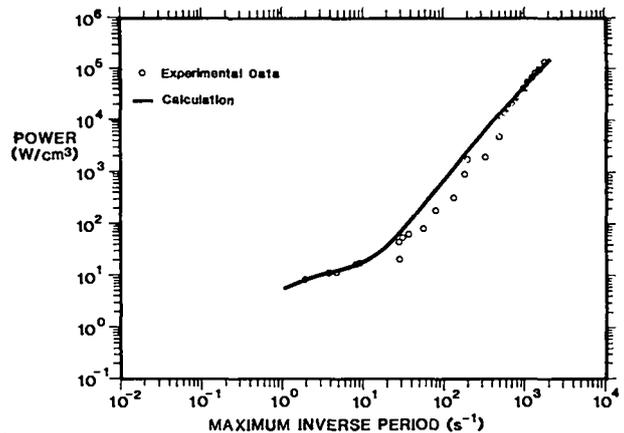


Fig. 10. Comparison of the results of the Mackin Code²⁵ with KEWB program experimental data. Peak specific power is plotted against the inverse period. The inverse period is proportional to the rate at which reactivity is added.

experiments are much more important if prediction of violent effects is the objective of the study.

The experimental peak hydrostatic pressures measured in these experiments are shown in Fig. 8. These momentary hydrostatic pressures are proportional to the specific power raised to the two-thirds power, as was mentioned earlier. The Mackin Code does not compute pressures from basic principles, but this has been done (in an independent study) by application of a version of a dynamics program,^{25,29} which can compute the production of hydrostatic pressures and the action of these pressures to expand the system and hence reduce

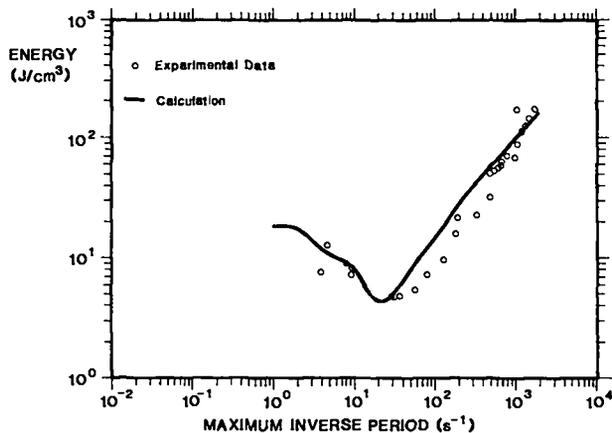


Fig. 11. Comparison of results of theory with experimental data. Specific energy density generated to the time of peak specific power is plotted against the inverse period. Experiments are from the KEWB program.

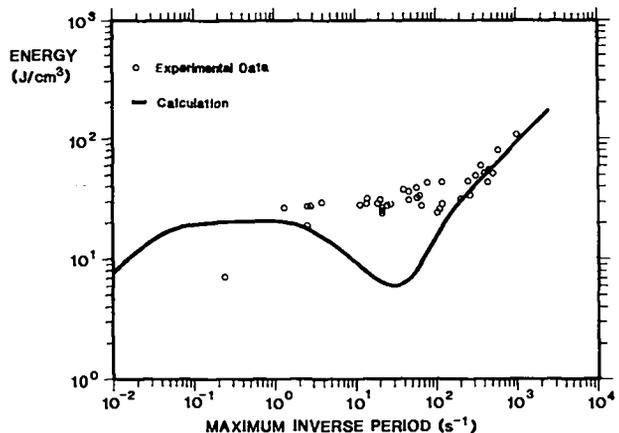


Fig. 13. Comparison of theory and experiment. Specific energy density at the time of peak power is plotted against the inverse period. Data are from the CRAC series of experiments.

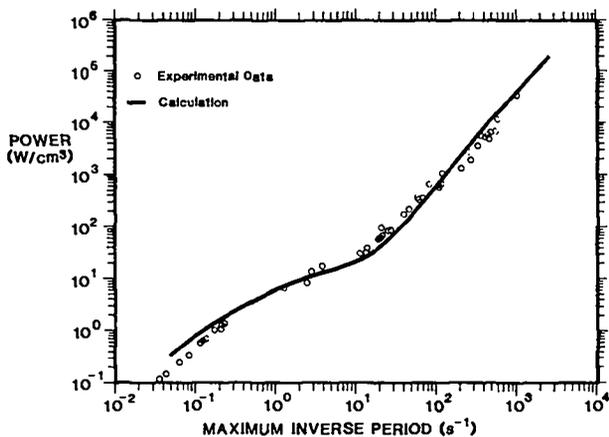


Fig. 12. Comparison of theory and experiment. Peak specific power for CRAC experiments is plotted against the inverse period.

pressures. The significance of this latter program is to demonstrate the correspondence between peak specific power and peak pressures. Specific power is predicted accurately by the Mackin Code and, therefore, pressures are known well.

The computer program generally overpredicts the energy release for the complete power spike (not illustrated); this result can be regarded as a conservatism in the case of predictions of different systems in which "magnitude and/or violence" of the excursion is an important factor.

VIII. APPLICATION TO A PLUTONIUM-WATER-SOIL MIXTURE

To apply the Mackin Code to the plutonium-water-soil mixture requires a few more assumptions. The most important of these are listed below.

1. The specific heat of the water-soil-plutonium mixture was adjusted for the volume fraction of water and the presence of soil and plutonium.
2. The reacting zone was assumed (arbitrarily) to be a 75-cm radius, 50-cm plug in the (also assumed) larger rectangular disposal site. An assumption of size was necessary to relate change of temperature and volume to reactivity.
3. The expansion was constrained to be only in the upward direction. This would overemphasize the upward motion, if any.
4. The reactivity insertion rate was related to the forced flow of water through a 4-in. pipe. This flow would be 0.155 m³/s and result in an increase of reactivity for the whole pit of 1.05 \$/s. Fractions and multiples of this rate were taken arbitrarily. A realistic rate of change of reactivity from water flooding might be a few cents per second.
5. The effect of the resonance in the plutonium cross section at 3200°C (as shown in Figs. 1 and 3) was

imposed upon the computation in addition to whatever reactivity insertion rate was chosen. That is, given an assumed value of H/Pu and some calculated change of temperature, the change implied by Fig. 4 was imposed upon the computation of reactivity as the calculation proceeded. In fact, exaggerated values of H/Pu were chosen to emphasize the influence of resonance.

6. The Mackin Code is a "point" kinetics computation. This, in essence, means that the properties of the critical system are assumed to be uniform throughout. This is very conservative for this problem as a space-dependent model would reflect the existence of the much higher plutonium concentrations near the surface. A high surface concentration would lead to easy and quick expansion of this layer; hence, no explosion whatsoever. A space-dependent model exists, but this was deemed to be unnecessary for the study.

Given the modifications listed above, the Mackin Code was applied to imaginary power excursions in the plutonium-soil-water system. Three ratios of hydrogen to plutonium were chosen, 1,000, 2,000, and 3,000, even though these are unrealistically conservative in relation to the amount of water required. Reactivity insertion rates up to 10 \$/s (equivalent to ten 4-in. pipes ejecting water at the maximum rate) were assumed. The most significant results of these calculations are illustrated in Fig. 14, in which the peak specific power (watts per cubic centimeter) is plotted against the maximum reciprocal period attained during the power excursion. Data for the three H/Pu ratios are plotted along with a collection of experimental data for both the KEWB and CRAC series of experiments. The only change from the experimental data is to increase the peak specific power as the H/Pu ratio increases. These calculated peak power densities should then be transferred to Fig. 8, which shows this momentary hydrostatic pressure as a function of peak specific power. The peak hydrostatic pressures calculated this way are only small multiples of an atmosphere and quite incapable of causing a disturbance more violent than a moderate splash. Because the momentary pressure increases only as the peak specific fission power raised to the two-thirds power, it is apparent that a physically violent excursion is improbable to the point of being impossible. Even the exaggerated cases never suggest "an enormous explosion, like a violent volcano."

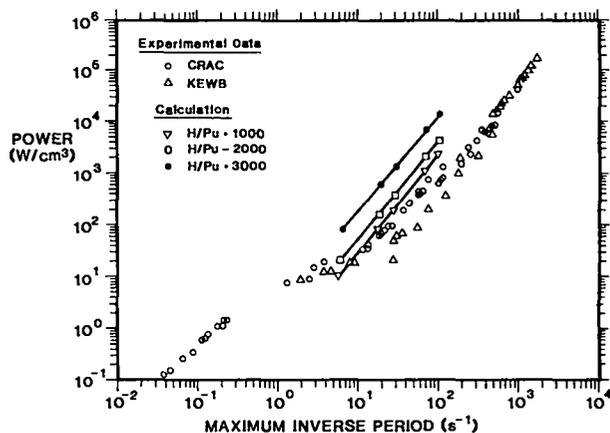


Fig. 14. Comparison of theory and experiment with the effect of the Pu-239 resonance included. Peak specific power is plotted against the inverse period. Data for both the KEWB and CRAC experiments are illustrated, whereas the solid lines show results for excursions calculated to occur in the postulated soil-water-plutonium mixtures. The effect of the resonance is to raise the peak power by a factor of 10 or 20. From the data in Fig. 8, the hydrostatic pressure will be raised by only a factor of 4 to 7 for impossibly large amounts of water (high H/Pu). Pressures are at most a couple of atmospheres, far too low to cause a violent eruption.

A somewhat hidden conservatism in this last step of the investigation should be emphasized. This is the effect of the experimentally observed density gradient of plutonium in soil that follows when a solution is disposed of in the ground. The plutonium is selectively deposited at and near the surface; very little penetrates more than a few centimeters. This effect means that fission densities created during an excursion would be largest near the surface, and hence, pressures would be dissipated easily and quickly. Any inertial confinement and subsequent explosion would be negligible.

IX. CONCLUSIONS

The physical problems associated with a postulated fission-product acid-uranium-plutonium disposal site in soil have been examined. The materials have been simplified to include only SiO₂, FeO, Al₂O₃, water, and plutonium. Any neutron-capturing (poison) material has been ignored. This is a conservatism that may be large but that cannot be made quantitative.

This imaginary solution was assumed to be deposited in a 30-cm-thick layer of soil in a concentration gradient

similar to that actually observed in experiments, which is preferentially near the surface. Critical masses in a postulated soil pit 9 m × 18 m with about 50% water by volume and with water reflection were in the hundreds and thousands of kilograms. Smaller volumes could have smaller critical masses but with large volume fractions of water or extraordinarily high concentrations of plutonium. Critical masses with substantially less water moderation are possible only with very much greater masses of plutonium. To postulate these masses to be discarded is most unlikely and uneconomic, given that the object of the activity is to create and chemically purify plutonium.

Because the postulated disposal system has very significant amounts of water, the characteristics of fission power transients in solutions have been investigated and are reported. Only with significant effort can a solution be forced to cause damage or violence (splashing) during a power transient. From the experiments themselves, it is evident that creating an "explosive" excursion in a solution is most difficult, and its happening by accident is essentially impossible.

A neutron-point kinetics computer program (Mackin Code) was used to investigate the properties of a supercritical soil-water-plutonium mixture even though such a mixture is most unlikely. The code predicts that for even extraordinary flooding rates, the power excursion would not produce a violent physical effect. This is the case even for exaggerated emphasis of the resonance at 3200°C in the fission cross section of Pu-239.

Finally, we would like to comment on Medvedev's⁸ postulate that spring floods caused a drastic increase in the proportion of water in the water-soil-plutonium mixture. First, the earlier arguments about masses of plutonium and the influence of the H/Pu ratio are unaltered as is the discussion about the experiments and calculations of the solutions excursions. Only the addition of water as moderator and reflector in the theoretical study would change. In the calculations, the reference rate was 0.155 m³/s from a 4-in. pipe; rates ten times this, or 1.55 m³/s, also were assumed. From the results, it is apparent that a reactivity insertion rate ten times this, or 15.5 m³/s, could be assumed without the calculations showing pressures sufficient to cause significant damage or violence during the excursion. A rate of 155 m³/s is equal to $1,668 \frac{3,474}{1,668} \text{ ft}^3/\text{s}$, an extraordinary rate and quite impossible to achieve short of a good-sized river.

Thus, it is shown that a violent nuclear explosion in a soil-based, chemical solution disposal site is so difficult to

achieve that it may be taken to be impossible. Earlier comments said to have originated with Sir John Hill in 1977* in the United Kingdom relating to the impossibility of such an explosion were timely and correct.

ACKNOWLEDGMENTS

Gordon E. Hansen and Harry M. Forehand, Jr., contributed a great deal to the technical content of this report. The discussion of the resonance in the plutonium cross section was developed by Hansen, whereas the availability of the Mackin Code, as created by Forehand, made the analysis of postulated power excursions fairly painless. I thank both gentlemen for their contributions.

However, responsibility for the conclusion of the study, namely, the essential impossibility of a nuclear explosion in a waste disposal site, as postulated, is mine.

Finally, I would also like to thank the following people for their contributions to this report: Linda Randolph, Jane Sherwood, Jan Harris, Kyle Wheeler, and Richard Miller.

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