

Title: CRITICALITY CHARACTERISTICS OF MIXTURES OF PLUTONIUM, SILICON DIOXIDE, NEVADA TUFF, AND WATER

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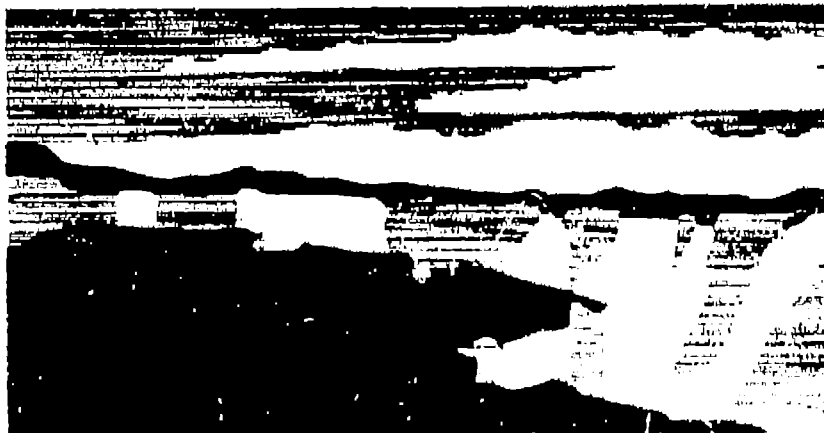
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**CRITICALITY CHARACTERISTICS OF MIXTURES OF PLUTONIUM,
SILICON DIOXIDE, NEVADA TUFF, AND WATER**

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ABSTRACT

The major objective of this study has been to examine the possibility of a nuclear explosion should 50 to 100 kg of plutonium be mixed with SiO₂, vitrified, placed within a heavy steel container, and buried in the material known as Nevada tuff. To accomplish this objective, we have created a survey of critical states or configurations of mixtures of plutonium, SiO₂, tuff, and water and examined these data to determine those configurations that might be unstable or autocatalytic. We have identified regions of criticality instability with the possibility of autocatalytic power behavior. Autocatalytic behavior is possible but improbable, for a very limited range of wet systems.

1. INTRODUCTION

The nuclear criticality characteristic of mixtures of plutonium, silicon dioxide and water (Part A) or plutonium, silicon dioxide, Nevada Yucca Mountain tuff, and water (Part B), have become of interest due to the appearance of recent papers on the subject.¹⁻³ These papers postulate that if excess weapon-grade plutonium is vitrified into a silicate log and buried underground, a self-sustaining neutron chain reaction may develop given sufficient time and interaction with the burial medium. Moreover, given specific geologic actions resulting in postulated configurations, the referenced papers state that nuclear explosions could occur with kiloton yields¹ or yields equivalent to hundreds of tons of TNT.³

The objectives of this paper are as follows:

1. To establish the parameters (density, moderation, reflection, poison, and dilution with non-fissile material) that determine when the mixtures of Pu-239, SiO₂, and water (Part A) and mixtures of Pu-239, Nevada tuff, and

water (Part B) are critical, i.e., are capable of sustaining a continuing neutron chain reaction without change of fission power.

2. To examine these data to find those configurations that may be unstable or "autocatalytic" (the critical state in which an increase in fission power leads to a further increase of reactivity and, hence, greater fission power); and

3. To establish the restrictions or constraints on these data that are required by critically physics, the amount of plutonium postulated to be stored, and possible water content in the Nevada tuff.

Part A of this paper will concentrate on the pure SiO₂ diluent, while in Part B, criticality computations have been completed using the actual elemental composition of the Nevada Yucca Mountain tuff.⁴ Tuff contains elements that are neutron poisons, which place additional constraints on the postulated situations in Refs. 1, 2, 3.

The calculations presented in this paper assume homogenous spherical distributions with pure Pu-239 as the fissile material. This is a conservative assumption because the critical masses for pure Pu-239 are substantially lower than those for U-235 and also lower than for weapons-grade plutonium. In addition any significant heterogeneity of the plutonium is expected to reduce the reactivity of the system significantly (even enough to preclude criticality in some cases).

The computations in this paper have been completed by use of the ONEDANT neutron transport computer program,⁵ that solves the one dimensional, time independent, multigroup discrete ordinates form of the Boltzman transport equation.

II. NUCLEAR CRITICALITY-SIMPLE SYSTEMS

To introduce the discussion of nuclear criticality parameters for a three-component system, it is useful to begin with illustrations of the criticality properties of a simple two-component system that is more familiar than mixtures of plutonium, SiO_2 or tuff, and water. Thus, in Fig. 1, the critical mass of plutonium metal mixed with water is illustrated as a function of the density of plutonium in the water.⁶ The mixture is idealized to be only metal and water; actual solutions of plutonium compounds would differ but little. Both water-reflected (20-cm thickness) and unreflected (bare) systems are shown. The general characteristics illustrated are similar for nearly all diluents of fissile material: U-235, U-233, Pu-239. The initial effect of the diluent (water in this case) is to reduce the density of plutonium, thus allowing greater neutron leakage and requiring a larger mass of fissile material to achieve the critical state. A maximum critical mass (about 9 kg for the reflected case) can be seen at a plutonium density of 6 g/cm³ (kg/l) where the moderating property of hydrogen starts to dominate the loss of neutrons due to leakage. As still more diluent is added, the moderating power of hydrogen causes the critical mass to decrease to a minimum value (about 500 grams at a plutonium density of 0.03 kg/l for the reflected case) at which point a balance is reached between the moderating and absorptive properties of the diluent. With more dilution, the absorption cross-section of hydrogen dominates, and the mass required for criticality increases until finally an asymptote is reached, which establishes the limiting plutonium density for dilution; both the mass and volume are unbounded. The limiting value of the plutonium density is an important property that will be useful in later discussions. For plutonium-water mixtures, the asymptote is at about 0.007 kg/l and, obviously, both the bare and reflected cases converge to the same value. These data are very well established by experiment⁷ and the computational scheme is well established by comparing calculational results with experimental results.⁶

The presence of water has a significant effect on the critical mass. It is this sharp dependence of the Pu-239 critical mass on the moderator content (in this instance, the hydrogen content) of the system that makes the analyses of criticality phenomena so complex. A wide variety of results are possible. For example, as seen in Fig. 1, the critical mass of a plutonium metal sphere with a water reflector is about 5.2 kg, while the critical mass of a plutonium solution with the same water reflector is about 0.5 kg. Under more unusual and extraordinary circumstances (non-absorbing diluents and cryogenic temperatures), the critical mass might be significantly lower.

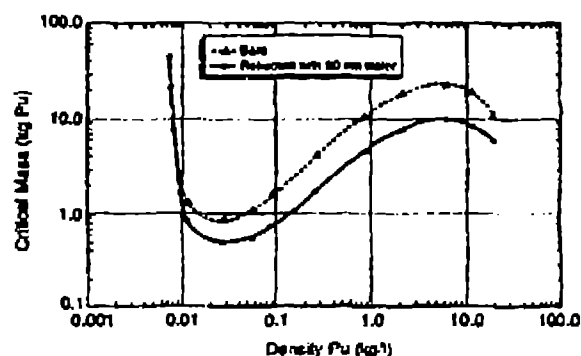


Figure 1. Critical mass of plutonium metal as a function of plutonium density when mixed with water.

This representation is useful in understanding the influence of various parameters and in establishing conditions of nuclear criticality stability or possible instability. For example, if a slightly supercritical plutonium-water mixture should be created at a density just a little greater than the low-density asymptote (choose about 10 kg and a density of about 0.008 kg/l) the solution would heat and lose water, thus decreasing the volume and increasing the plutonium density. Given only these two effects, the system would now be at a very high reactivity and power would increase. This condition is often referred to as "unstable" or "autocatalytic." However, any decrease of volume caused by water loss would be balanced by boiling and expansion, thus reducing reactivity. The net result is boiling, just maintaining criticality. This would continue until the system is dry enough to be subcritical. This illustrates the complexity of competing effects which may produce a variety of results. Another example, although not an unstable condition, is the region in Fig. 1 between densities 1.0 and 0.08 kg/l. If water, which is a diluent, moderator, and absorber, is added, a supercritical state is created. However, this is not autocatalytic like the first example; thermal expansion, possibly with boiling, would reduce reactivity to return the system to subcriticality; little change of composition would occur.

III. PART A: CRITICALITY DATA: PLUTONIUM, SILICON DIOXIDE, AND WATER

A. Nuclear criticality--other materials

As was mentioned above, the general characteristics of Fig. 1 can be seen when other materials are used as a diluent. Examples are illustrated in Fig. 2 in which the critical masses of plutonium diluted with SiO_2 and reflected by 100 cm SiO_2 , and plutonium diluted with and reflected by 20 cm of water are presented. The critical mass data for

U-235 diluted with and reflected by graphite³ are added for comparison.

The general characteristics of these data are qualitatively similar, but the maximum and minimum critical masses differ by large factors as do the low-density asymptotes caused by water, graphite, and SiO₂. The importance of the asymptote is emphasized because its existence is independent of the material but its value (in terms of the fissile material density) is very dependent on the material and its density and helps to limit the possibilities of underground critical states for the problem being investigated. The calculations describing U-235 diluted with graphite are included to illustrate the wide range of criticality possibilities.

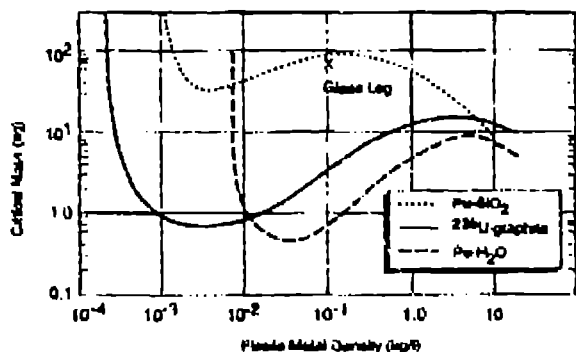


Figure 2. Critical masses of Pu-H₂O mixtures reflected by water, Pu-SiO₂ mixtures reflected by graphite.

No experimental critical data are available to assure the correctness of the calculations for the Plutonium-SiO₂ system, except for the end point of unreflected plutonium metal. There are no integral tests of the silicon cross sections. In addition, for dry silicon and plutonium mixtures, many fissions take place primarily in the intermediate energy range (between neutron energies of 0.1 eV and 100 keV), where no integral tests for the plutonium cross sections exist. Thus, for these dry systems, calculations should be treated with some caution. However, because of the abundance of other experimental critical data for both fast and thermal plutonium systems,⁶ the calculations for those thermal and fast Plutonium-SiO₂ systems are expected to be reliable.

H. Critical data for Pu-SiO₂-H₂O

In Fig. 3, the reflected critical-mass data for Plutonium-H₂O and Plutonium-SiO₂ are reproduced from Fig. 2 along with the addition of critical data for Plutonium-SiO₂ in which water has been mixed in equal amounts in both the core and the 100 cm thick reflector. The model for adding water assumes that the volume of water displaces an equal volume of SiO₂ and plutonium.

For this model, the densities of the plutonium and SiO₂ decrease with the addition of water. This is the three-component material postulated in Refs. 1, 2, 3 that we wish to study in detail. As seen in Fig. 3, the top curve describes dry Pu-SiO₂ mixtures reflected by 100-cm-thick dry SiO₂. The bottom curve is plutonium mixed with and reflected by a 20-cm thick layer of water, while the intermediate curves are Pu-SiO₂ cores mixed with successively greater weight fractions of water and reflected by 100-cm-thick SiO₂ with the same weight fraction of water. The powerful moderating property of water is evident. The atom ratio of silicon to plutonium for each of the "wet" curves is indicated in Fig. 3 and this same ratio is held constant as more water is added. Thus, in this model, plutonium density decreases steadily as water is added.

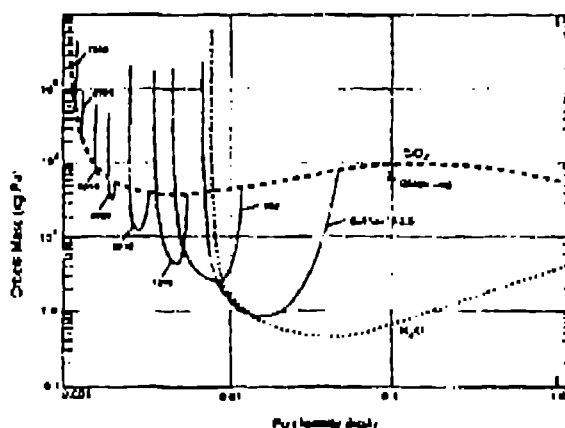


Figure 3. Critical data for mixtures of plutonium, SiO₂, and water plotted as a function of plutonium density.

For relatively low atom ratios of silicon to plutonium, e. g., 183, 752, 1570, the initial addition of water decreases the critical mass and critical volume very sharply, or, one can say that this three-component system has significantly greater reactivity. For each silicon to plutonium ratio, however, a minimum critical mass is reached as water is added, comparable to the minima seen in the two phase mixtures. For still larger weight fractions of water, the hydrogen in the water becomes a poison (a neutron absorber) and a water (hydrogen) asymptote is seen; its location is a unique function of the plutonium density, the atom ratio of silicon to plutonium, and water content. As examples, for Si/Pu=752 the asymptotic plutonium density is 0.0045 kg/l; for Si/Pu=2812, it is 0.0025 kg/l. For very large Si/Pu ratios, greater than 6000, the addition of water (hydrogen) acts invariably as poison, and the critical mass increases regardless of the amount of water introduced.

The data presented in Fig. 3 can be examined in another way that is useful in understanding the constraints on the problem at hand. Figure 4 shows the critical masses of plutonium taken from Fig. 3 as a function of the weight percent of water; the dry critical mass from Fig. 3 is the starting point on the left ordinate and the water content increases to the right. The critical mass decreases with additional water moderation (except for very high Si/Pu atom ratios) until a minimum is reached. With additional water content, the critical mass increases until an asymptotic value is achieved. For each Si/Pu atom ratio, the weight percent water at the low-plutonium-density asymptote is readily seen or can be estimated with reasonable confidence. For example, for Si/Pu=2812 it is 11.2% and for 5513 it is 3.0%. Thus, for a fixed Si/Pu ratio the water content must be less than the asymptote or criticality cannot occur.

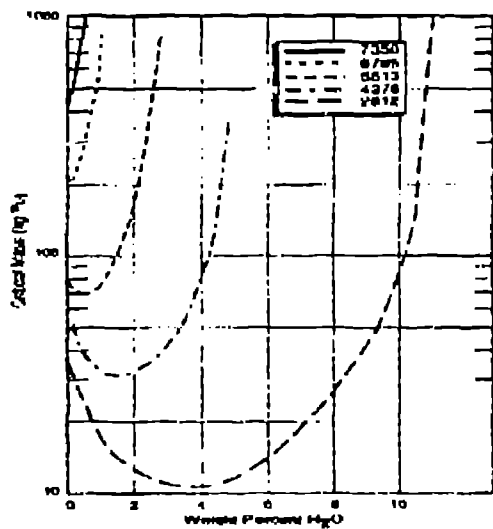


Figure 4. Critical mass of plutonium as a function of the weight percent of water for various Si/Pu atom ratios.

C. Constraints on the possible critical configurations

The set of necessary data for mixtures of plutonium, SiO₂, and water is complete, and constraints can be applied that limit the number of cases that need to be examined in more detail. One constraint on the critical configurations is the water-poisoning asymptote (Fig. 4) for each Si/Pu ratio. For example, if the water concentration is more than 3 wt%, criticality is not possible for ratios of Si/Pu greater than 5513, regardless of how much plutonium is involved. This constraint, based on the water induced asymptote, is illustrated in Fig. 5; for a given Si/Pu ratio, the water content must be less than the uppermost line on the figure for criticality to be achieved. For some mixtures of plutonium, SiO₂, and water, a configuration that is found

just on or very near this line on Fig. 5 can be unstable and possibly could be "autocatalytic."

A second constraint is found by further examination of the functions in Fig. 4. Referring to Fig. 4, the dry critical mass for a Si/Pu ratio of 5513 is about 84 kg. The minimum of this function is about 70 kg at 0.5 wt% water. At higher concentration of water, the critical mass increases continuously up to the asymptote corresponding to about 3 wt%. Between 0.5 wt% and 3 wt% water, an unstable or possibly autocatalytic region exists. Autocatalysis is impossible between 0.0 wt% and 0.5 wt% water; it is possible only between 0.5 wt% and 3.0 wt%. The minimum of each of Si/Pu functions in Fig. 4 is plotted on Fig. 5 as the lower curve. The unstable or possibly autocatalytic region lies only between these two curves on Fig. 5. The lower limit function represents the water content for no instability.

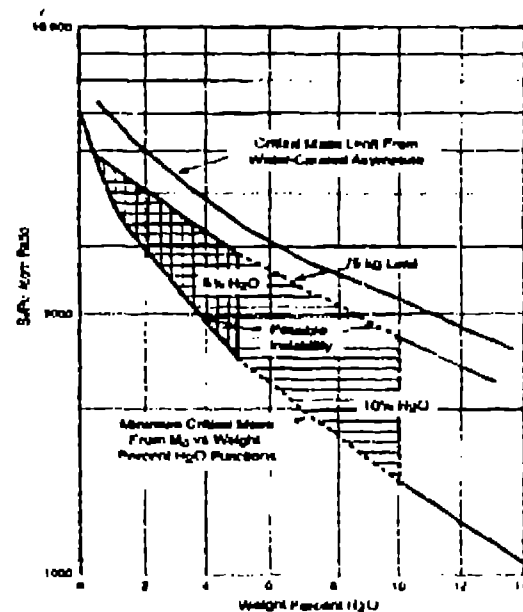


Figure 5. Limiting values of water content as a function of Si/Pu ratio.

The third constraint is, of course, the available mass of plutonium, which has been taken to be 75 kg. If this amount is assumed to move into a wet stratum of SiO₂, the Si/Pu ratio will slowly decrease. The following examples are instructive. If the water content were, e.g., 0.5 wt% (see Fig. 4), criticality is possible for Si/Pu atom ratios of 7350 or 6785, but the required masses are 1000 kg and 300 kg, respectively; only 75 kg are available. If the water concentration were 1.0 wt%, 75 kg could be critical at a Si/Pu ratio of 5513. Proceeding to lower Si/Pu ratios at 75 kg on Fig. 4, this mass could be critical for 4.0 wt%

water at $Si/Pu=4376$ and 10 wt% at $Si/Pu=2812$. The locus of these points is plotted on Fig. 5 as the middle function. The region of autocatalysis is now restricted to the region between the lowest and the middle function of Fig. 5.

The final constraint to be discussed in this section is the assumed amount of water in the SiO_2 surrounding the Pu- SiO_2 mixture. The only guidelines on water content of the soil or rock are the actual measurements and analyses by the geologists. A reasonable upper limit is 5 wt%. Both 5.0 wt% and 10.0 wt% water content will be considered to be conservative. These constraints are illustrated by vertical lines on Fig. 5.

The full picture of all these constraints can be seen on Fig. 5 as triangles shaded on the semi-log plot. The defining corners for 75 kg and 5 wt% water are as follows: $Si/Pu=3800$, % $H_2O=0.5$; $Si/Pu=3900$, % $H_2O=5.0$; and $Si/Pu=2400$, % $H_2O=5.0$. If the limiting amount of water is 10.0 wt%, the defining points on Fig. 5 are $Si/Pu=3800$, % $H_2O=0.5$; $Si/Pu=2800$, % $H_2O=10.0$; and $Si/Pu=1600$, % $H_2O=10.0$. Configurations outside these limits are of no interest for the reason just developed.

IV. PART B: CRITICALITY DATA--PLUTONIUM, NEVADA TUFF, AND WATER

A. Nuclear criticality--Nevada tuff

The tuff composition used in these calculations is taken from Ref. 4. This composition differs from SiO_2 in that SiO_2 content is only 77% by weight with the difference made up primarily by oxides of aluminum, potassium, sodium, and with a number of minor constituents, some of which are neutron poisons.

In Fig. 6, the calculated reflected critical masses for plutonium mixed with Nevada tuff are shown as a function of plutonium density along with the comparable critical masses for plutonium mixed with pure SiO_2 . The model used for mixing plutonium and tuff assumes that the volume occupied by the plutonium displaces a comparable volume of tuff. Significant differences can be noticed immediately. For instance, the low-density asymptote is increased from 0.001 to 0.003 kg/l. Thus, the number of highly diluted cases requiring consideration is further restricted. In addition, the moderating/absorbing effects that decrease the critical mass from 95.7 kg at a plutonium density of 0.184 kg/l to 35.1 kg at a plutonium density of 0.048 kg/l very nearly disappear when the diluent has the composition of the tuff. With tuff as a diluent, the maximum and minimum critical masses are 112.6 kg and 85.6 kg. This increase in the minimum critical mass when the Nevada tuff is the diluent eliminates the need to

consider the "dry critical" problem that was postulated in Ref. 1; 75 kg of plutonium would never become critical in dry tuff. Including Pu-240 or allowing for the decay of Pu-239 to U-235 would increase the critical masses even more.

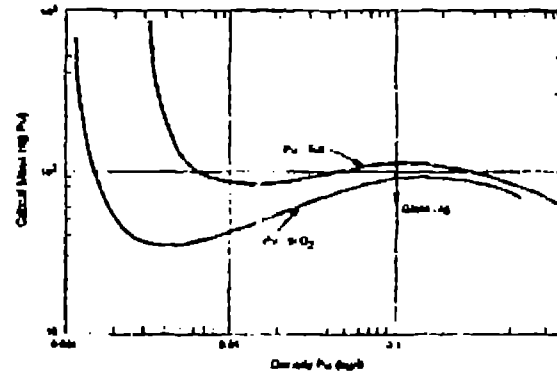


Figure 6. Reflected critical mass of plutonium metal mixed with tuff or SiO_2 vs plutonium density.

B. Critical data for Pu-tuff- H_2O

The critical data for a Pu-Tuff- H_2O are illustrated in Fig. 7 as a function of the weight percent of water. The increased effect of the neutron absorbers in the tuff is clearly seen when comparing the minimum critical masses and low density asymptotes presented in Fig. 7 to those in Fig. 4. Note that to compare these data with the data in Fig. 4, the Si/Pu ratios for the tuff are multiplied by 1/0.77 and denoted by Si/Pu^* .

C. Constraints on the possible critical configurations

Referring again to Fig. 7, the minimum of each function is the lowest water concentration above which instability or autocatalysis is possible. A lower water concentration cannot lead to instability. The locus of these points is plotted on Fig. 8 as the lower line, providing a constraint on the amount of water and Si/Pu^* ratios to be considered.

The limitation set by the mass loading of 75 kg can be read from the functions for each of the Si/Pu^* atom ratios on Fig. 7. The locus of these points is plotted on Fig. 8 as the middle line, comparable to Fig. 5 for SiO_2 .

The final constraint is the amount of water that would actually be found in the tuff. This has been estimated to be 5 wt%.⁹ However, twice this amount will be considered to assure conservatism.

Thus, the constraints for a Pu-tuff- H_2O can be defined similarly to those for a Pu- SiO_2 - H_2O for a 5 wt% water

and 75 kg, the coordinates of the small triangle are: $Si/Pu^* = 2700$ and 0.4 wt%; $Si/Pu^* = 1850$, 5 wt%; and $Si/Pu^* = 1450$, 5 wt%. For 10 wt% water and 75 kg, the coordinates are: $Si/Pu^* = 2700$, 0.4 wt%; $Si/Pu^* = 1500$, 10.0 wt%; and $Si/Pu^* = 1100$, 10.0 wt%. Configurations that lie outside these boundaries are not of interest because autocatalytic behavior is not possible.

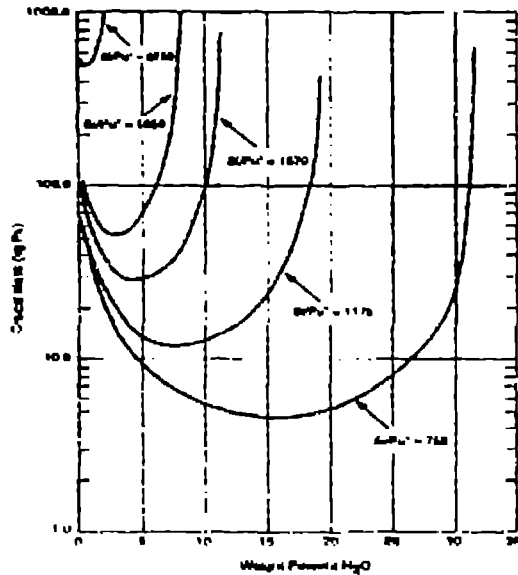


Figure 7. Reflected critical mass of plutonium metal mixed with tuff and water vs weight percent water.

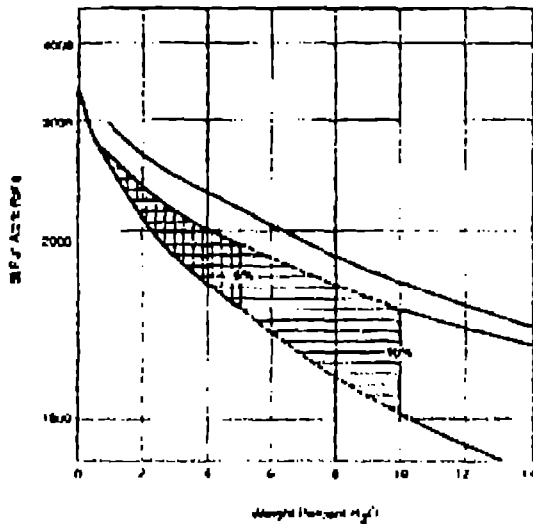


Figure 8. Limiting values of water content as a function of Si/Pu ratio. Note that the Si/Pu ratio in tuff is multiplied by 10.77 and denoted as Si/Pu^* .

V. REACTIVITY CALCULATIONS FOR Pu-TUFF- H_2O

The previous section defined the possibilities for critical conditions, given certain constraints. This section will examine the possibilities for supercritical configurations that might be created and developed as a result of movement and mixing of plutonium, tuff, and water. As an example, if a critical state is achieved at a water concentration of 10.0 wt%, plutonium mass of 108 kg, and at a Si/Pu^* of 1570, the resulting fission power could heat the volume and transport water out of the volume. This would lead to a reduction in the water content of the mixture and an increase in the reactivity of the system.

The cases of most interest are the data for a Si/Pu^* atom ratio of approximately 1500. These data are illustrated in Fig. 9. The masses of plutonium and the initial water are as follows: Case (1) 404.7 kg and 11.2 wt% water; Case (2) 108.3 kg and 10.0 wt%; Case (3) 63.8 kg and 9.0 wt% water; Case (4) 45.5 kg and 8.0 wt% water; Case (5) 36.4 kg and 7.0 wt% water. These data cluster in the upper right hand corner of the permissible triangle on Fig. 8. This "corner" (10 wt% water, 75 kg of plutonium, and Si/Pu^* ratio of 1500) is the condition that will lead to the highest k_{eff} and the largest integral of k_{eff} as a function of weight percent of water. The reactivity functions of Fig. 9 are almost precisely at the correct spot to examine the most severe limiting case. Case (1) on Fig. 9 has too much plutonium and water; Case (2) exceeds the mass limit but has the correct limiting amount of water. The other three are all within permissible constraints. The maximum k_{eff} for 75 kg is about 1.06 (by interpolation) when approximately three-fourths of the water is forced out of the system (see Fig 9).

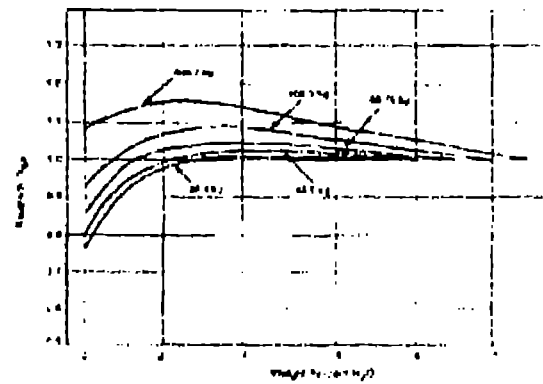


Figure 9. Reactivity (k_{eff}) vs weight percent for $Si/Pu^* = 1570$ (tuff) and 404.7, 108.3, 63.8, 45.5, and 36.42 kg of plutonium.

The intent of this section has been to examine the criticality aspects of the unstable regions defined in the previous section for a Pu-tuff-H₂O system. The postulated explosion aspects, discussed in Ref. 3, will be examined in a companion paper on dynamics¹⁰ given the restrictions and constraints developed in this study.

VI. SUMMARY AND CONCLUSIONS

This paper has examined the static criticality aspects of mixtures of plutonium and SiO₂; plutonium, SiO₂, and water; plutonium and Nevada tuff; and plutonium, Nevada tuff, and water.

A conservative approach was taken by assuming the plutonium was pure Pu-239, homogeneously distributed within the moderator/absorber material in a spherical form. Any deviation from these criteria, e.g., substitution of weapons-grade plutonium (about 5 wt% Pu-240, 95 wt% Pu-239) for pure Pu-239 results in lower neutron multiplication factors and higher critical masses than those calculated in this work. Inclusion of elements found in the Nevada tuff also lowers the calculated neutron multiplication factors. All reactivity calculations have included the assumption of uniform, fine-grain mixing of all components throughout.

The general characteristics of fissile systems diluted with a moderator/absorber material were summarized. No qualitative features of the plots of the critical masses of such systems versus the fissile density are generally similar. All exhibit a maximum critical mass with small amounts of diluent, followed by a minimum caused by the moderating effect of the diluent, and finally a low-density unbounded critical mass asymptote caused by the neutron absorbing effect or neutron capture of the diluent. Configurations near the asymptote can display instability and possible autocatalytic behavior. The quantitative features, such as the values for the critical masses, the locations of the maximum and minimum critical masses, and the location of the asymptote depend on the fissile material and the moderator/absorber materials present.

Several cases were examined for the potential of autocatalytic behavior. We obtained the following results:

a) The system consisting of pure Pu-239 with pure SiO₂ as a diluent exhibits the classic dependence of the critical mass on the fissile material density. Thus, a critical system is possible, but very improbable; autocatalytic behavior is judged to be possible only if the assumptions regarded as impossible are accepted. When the Nevada tuff is the diluent, nuclear criticality and autocatalytic behavior are not possible for log loadings of less than approximately

84 kg of Pu-239. If weapons-grade plutonium (5 wt% Pu-240) is considered, the requirements are much more stringent.

b) For processes that add water, criticality is possible for both SiO₂ and Nevada tuff diluents. However, no autocatalytic behavior is expected because the system has sufficient time to respond to reactivity increases by ejection of water, which reduces reactivity.

c) For plutonium entering a wet stratum of SiO₂ or Nevada tuff, criticality and autocatalytic behavior is possible, but Si/Pu ratios for which this can occur are constrained by the total amount of plutonium and by the water content of the strata. These constraints limit the insertion of reactivity when water is removed from the system. The behavior of these wet systems will be studied in a companion paper on dynamics.¹⁰

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