*c. 3*

# UNDERGROUND AUTOCATALYTIC CRITICALITY FROM PLUTONIUM AND OTHER FISSILE MATERIAL



Photograph: by Chris J. Lindberg

## Los Alamos
### NATIONAL LABORATORY

# Underground Autocatalytic Criticality from Plutonium and Other Fissile Material

## C. D. Bowman and F. Venneri

## Abstract

*Several widely endorsed solutions to the intermediate and long-term disposition of weapons plutonium and other waste fissile nuclear material involve placement of batches of the material underground in subcritical concentrations. It is pointed out here that such concentrated subcritical fissile material underground might reach criticality that is autocatalytic or self-enhancing. This criticality could come about upon dispersion into the surrounding medium by either natural or unnatural processes, or by the fissile material being carried to other sites where it can collect into different autocatalytic critical configurations . Underground, where the material is confined and there is an abundance of moderating medium around it, the results of such supercritical excursions could range from modest energy releases to the generation of explosive nuclear yields of up to a few hundred tons from a single event. Without water, 50-100 kg of fissile material is required to reach autocatalytic criticality. Amounts as small as a kilogram can reach autocatalytic criticality with water present. In varying degrees, all categories of waste containing fissile actinide appear to be susceptible to these criticality excursions, including vitrified weapons plutonium, research reactor and DOE spent fuel, commercial and MOX spent fuel.*

## Introduction

The long term disposition of thermally fissile material (TFM) is currently the focus of much national and international attention. These materials include excess weapons plutonium (w-Pu) and highly enriched uranium (HEU) from the reduction in nuclear weapons stockpiles in the U. S. and Russia, naval reactor spent fuel which contains a high concentration of $^{235}$U, spent fuel from research reactors containing HEU, spent fuel from commercial reactors containing plutonium (c-Pu) and other heavy elements such as neptunium, which also are potentially useful as nuclear weapons materials. Recent prominently publicized studies[1] considering the long-term disposition of w-Pu have identified several options all of which end up with the material in permanent storage deep underground. These studies strongly influence current U. S. Government policy[2]. The purpose of this report is to show that underground storage as presently recommended could lead to underground autocatalytic criticality and the uncontrolled dispersal of the TFM with significant nuclear energy release and possibly nuclear explosions in the few hundred ton range.

The weapons plutonium portion of the TFM is perhaps of greatest current concern and for this reason $^{239}$Pu is used for the most part in this paper to illustrate the criticality risks of underground TFM. The actual concentration would vary with the storage situation. For the option of vitrification of the plutonium followed by storage in deep boreholes, the National Academy of Sciences study on plutonium disposition[3] considers a concentration of up to 10% by weight so that a borosilicate cylindrical log 50-cm in diameter and two meters long would contain about 100 kg of plutonium. A single log of this material would be substantially subcritical owing to geometry and neutron poison. Other storage forms of the w-Pu also would contain substantial amounts of fissile material. MOX spent fuel assemblies for w-Pu destruction would contain 18 kg each[3] and several of these might be stored together. If w-Pu were simply vitrified with high level waste at 2% mass fraction, the w-Pu mass would be 44 kg each[3] in logs of 3-m length and 0.6-m diameter.

In order to keep the costs of preparation for storage and for actual emplacement underground of TFM low, and to make the repository storage site small, there would be pressure to store the TFM in concentrated but still safely subcritical amounts. Subcriticality would also be enhanced by the inclusion of neutron poisons and by choosing geometry and com-

position such that only fast neutrons could be effective in propagating a chain reaction.

Even without poisons, w-Pu in these amount and in these configuration would be subcritical. The reason is that the neutrons do not have a chance to moderate in the rock before leaving the w-Pu and cannot find the w-Pu after moderation. However, once containment has been breached and the TFM is free to disperse in the underground matrix containing good moderators such as water and rock in various proportions, critical configurations are possible which may have positive or negative feedback features.

## Feedback positive and negative

Many factors influence criticality such as amounts of fissile material, water, other moderating material, poison, the configuration, and resonance behavior. Poisons can be very important but their physical properties such as solubility and boiling point will in general be substantially different from those of the fissile material. In the view of the authors, poisons may not be a reliable means of preventing criticality over the long term. The effects of resonances can be significant only if there are large amounts of $^{238}$U or $^{232}$Th present, which is often not the case for TFM. The relative concentrations of fissile material, water, and other moderator such as rock are the most important factors and these can be analyzed for positive or negative feedback on criticality using Figs. 1 and 2.

Fig.1 shows the criticality conditions for several volumes of different radii with almost any mixture of $^{239}$Pu, water and $SiO_2$ surrounded by a $SiO_2$ reflector. The calculations were done using the MCNP[4] code. $SiO_2$ at a density of 2.2 approximates to a reasonable degree the nuclear properties of rock. The figure gives the mass fraction of plutonium on the ordinate and mole fraction of water and $SiO_2$ on the abscissa. Therefore for point G in the figure, Pu makes up 1 % of the sphere mass. The remainder of the material in the sphere expressed in mole fraction is 20 % water and 80 % $SiO_2$. A system lying on the left ordinate has no $SiO_2$ in it. A system on the right ordinate would have no water. A system with mass fraction of plutonium of one (not shown on the figure) would be pure plutonium.

The curves show critical homogenous mixtures for $SiO_2$-reflected spheres with radii of 25, 50, 100, and 200 cm. Mixtures of a given radius which lie above the curve are supercritical; those below are subcritical. Obviously material
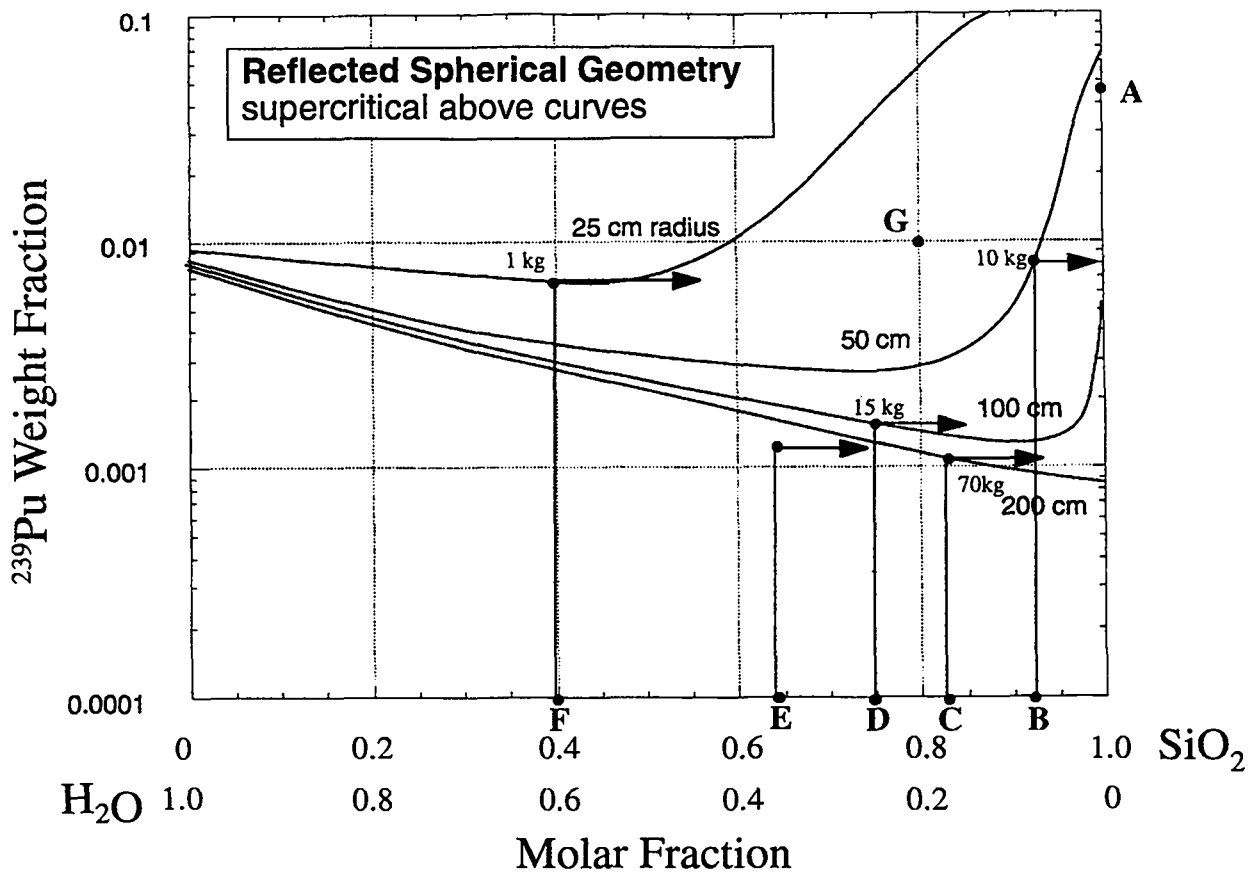
**Figure 1. Examples of positive and negative feedback following underground criticality.** *Criticality curves are given for spherical volumes of radius 25, 50, 100, and 200 cm for mixtures of $^{239}Pu$, water and $SiO_2$ reflected by $SiO_2$. The ordinate is the weight fraction of $^{239}Pu$ in the sphere. A fraction of 1.0 means the system is pure plutonium. The abscissa is the molar fraction of water or $SiO_2$ in the rest of the volume. A system lying on the left hand ordinate contains no $SiO_2$. A system lying on the right ordinate contains no water. Systems of a given radius with composition lying above the line for that radius are supercritical; those lying below are subcritical. Therefore only systems lying below the line can be placed in the repository. Systems which reach criticality where the slope of these curves are negative have positive feedback and are therefore autocatalytic. Systems which reach criticality where the slope of the curve is positive will have negative feedback*

cannot be implanted underground as supercritical material, so all initial subcritical arrangements of a given radius are located below the criticality curve. Over time one must expect that the relative concentrations of water, rock and fissile material could change and some of these changes could lead to criticality. It is widely believed that upon reaching criticality all systems will revert to subcriticality by natural means of negative feed back. One of the principal purposes of this paper is to show that the feedback can be either negative or positive and to outline means to distinguish between these two possibilities. The main feedback mechanisms illustrated in the figure are water ejection and TFM dispersal. With regard to water ejection, indicated in Fig.1 by a horizontal move to the right, *if the system reaches criticality at a point where the curves have a negative slope, the system will have positive feedback and could therefore be autocatalytic, if the system reaches criticality at a point where the curves show a positive slope, the system will have negative feedback and will be self-limiting or self-termi-nating.* While the figure is only valid for spherical geometry, the criteria illustrated are relevant to many critical shapes. We illustrate these criteria by examining six conditions labeled in the figure as A through F.

*Case A (negative feedback)*

Nearly all TFM would be emplaced as dry material and therefore will initially lie along the right ($SiO_2$) ordinate. If we consider the curve for 50 cm, we see that the mass fraction of $^{239}Pu$ mixed with $SiO_2$ must be less than about 0.065 or the system would be critical upon implantation. The case A examined here is for a loading of a weight fraction of 0.044, corresponding to about 50 kilograms of $^{239}Pu$, which is well below the dry critical mass of 80 kg of $^{239}Pu$ for a radius of 50 cm. If water enters this system, it will move horizontally to the left until it reaches wet criticality at a water mole fraction of about 2 % (0.6 % mass fraction). The system will generate fission energy, with the power density depending on the ra-dius, until water near the center is converted to steam which then drives the water out of the system. If the water expulsion is complete, the system returns to its starting point on the ordi-nate. The next incursion of water will cause the same process and this could continue indefinitely so long as the 50 kg of TFM remains within the 50-cm radius. This phenomenon is similar to that of the Oklo system. The Oklo natural reactor[5] in Gabon, Africa is frequently cited as an underground critical system which operated for about one million years. This was a

deposit of high grade uranium which existed at a time when the natural isotopic composition of uranium was about 3.7% $^{235}U$, before it decayed to the present 0.7%. Criticality was initiated when water entered the system. The fission heat boiled the water away taking the system to subcriticality until the next incursion of water. The average power of that system was 20 kilowatts. Oklo is a good example of the effects of negative reactivity feedback.

### Case B (negative feedback)

Water in small amounts in the rock can give rise to smaller critical masses than either water or rock alone. From Fig. 1 for the 50-cm radius case, we find the critical mass of $^{239}Pu$ with water alone is 4 kg; with rock only the amount is 80 kg; for 20% mole fraction of water in rock the critical mass is only 3 kg. If some of the TFM should leave its original implantation site and began to migrate to a new volume with 50-cm radius and containing 8 % mole fraction of water ( 2.4 % mass fraction), the system would start at point B and move vertically in the figure. Mechanisms for migration could be as simple as water carrying plutonium oxide particles and depositing them somewhere else. When a plutonium mass fraction of 0.008 had accumulated, corresponding to a mass of about 9 kg in the 50-cm radius, the configuration would become critical and start generating heat. The expulsion of the water by fission heat would however move the system to the right and therefore to subcriticality. As long as no further dispersion of the $^{239}Pu$ occurred, the system could move horizontally into and out of criticality indefinitely following repeated incursions of water.

### Case C (positive feedback)

Although water is generally known to be a better moderator than rock, the infinite medium TFM density to maintain subcriticality in rock is smaller than in water. This is a consequence of the capture cross sections for the rock which is about 0.255 barns per molecule compared to 0.66 barns per molecule for water, the different molecular densities for these two materials, and the fact that the energy loss per collision is not a relevant parameter for a large volume calculation. If some TFM from one or more original implantation sites should migrate to a 200-cm radius volume where the mole fraction of water was 15 %, the system would start at point C and move vertically until the system went critical at a mass concentration of 0.001. This corresponds to about 70 kg of $^{239}Pu$ in the 2-m radius 70-ton sphere. When this system becomes critical and the heat begins to drive the water out, the system also moves to the right but in doing so it drives itself to *higher* criticality reaching its highest criticality in a dry supercritical state. The authors are not aware that such a situation and consequence have been recognized before.

### Case D (positive feedback)

The accumulation of plutonium in a sphere of 100-cm radius with water present as a 23 % mole fraction will stop when criticality is reached at 15 kg. This system is also in a region of negative slope on the criticality curve so that the system autocatalytically drives itself to the right. This case has the interesting feature that expulsion of the water eventually can take the system subcritical after the mole fraction of water has decreased to about 4 %.

### Case E (positive feedback)

This case illustrates another autocatalytic condition which is simply approached somewhat differently. It illustrates the deposition of plutonium in wet media to overmoderated subcritical concentrations of fissile material followed by drying of the system. As the system dries, it reaches autocatalytic criticality. For the 200-cm radius system (large systems) this type of supercriticality can happen with any ratio of water to rock since the slope is always negative. For the smaller systems this danger is present for a wide range of water concentrations. Examples of this case are not restricted to underground phenomena. The Chernobyl disaster is an example of this Case E condition. This reactor was well moderated by graphite. Water was present for heat removal and, in the presence of the large amount of graphite in this very large reactor, the moderator function of the water was not important and from the neutron economy perspective it served mainly as a poison. Malfunction of the control system led to "drying out" by uncontrollable boiling and the system became autocatalytic and destroyed itself.

### Case F (positive feedback)

The critical curve for a 25-cm radius is interesting primarily for the small amount of material which could become autocatalytic. For the point F at a mole fraction of 0.4, the system is autocatalytic at a plutonium mass of less than 1 kg. Nearly the same mass of plutonium has negative feedback for a molar fraction of 0.5. At a mole fraction of 0.2, where the mass of water and $SiO_2$ in the sphere are about equal, the system is distinctly autocatalytic with respect to water ejection with even smaller amounts of plutonium. As the radius under consideration becomes smaller, the region of negative slope (autocatalytic condition) becomes smaller and the magnitude of the slope decreases as well. For radii smaller than about 20 cm, the slope of the criticality curve is always positive and the feedback always negative regardless of the rock-to-water ratio.

### Case A (revisited)

In the presence of substantial concentrations of plutonium (case A), the plutonium can be dispersed by repeated water-steam expulsions, and the system can become critical with less water present. If sufficient $^{239}Pu$ is present, the dispersion of plutonium can take the system to dry criticality at a larger plutonium radius, at the location of emplacement. The situation is more clearly illustrated in Fig.2, where the radius of the spherical critical mass for 50 kg of $^{239}Pu$ is shown for various molar fractions of water and rock represented by $SiO_2$. The shaded area of the curve is the region of supercriticality and therefore denotes mixtures for which emplacement is impossible. The unshaded area represents subcritical regions where emplacement can be made. As a practical matter it would seem that most emplacements would be as dry material and therefore would be made along the right ordinate. In that case, the radius of containment for 50 kg of $^{239}Pu$ must be between 20 and 100 cm as shown by the points H and J or with a radius greater than 200 cm. The 'ear" on the right ordinate is larger for smaller amounts of fissile material. The relevant transitions for this situation are either horizontal (water ingress or ejection) or downward in the figure (TFM dispersal to larger radii and lower concentrations).

The farther the systems move into the supercriticality region, the greater $k_{eff}$. Therefore the region on the right ordinate between points J and K represents a region of supercriticality with the maximum value about halfway in between. If a system reaches point K and conditions are such that the plutonium is driven by fission heat through the rock, the system is dry autocatalytic. Any dry system emplaced with a configuration between H and J when exposed to water *could work its*

way down to K. This is illustrated by the zigzag line in the "ear" lying on the ordinate. An initially dry system is shown with a radius of 50 cm (point A). The incursion of water would move the system to the left until it became critical at about a water mole fraction of 5 %. Upon the generation of fission heat, the system would expel some or all of the water and move perhaps all of the way back to the ordinate. Incursion of water will start the process again. This could go on indefinitely, but it seems likely that eventually the plutonium would be spread by these criticality excursions and that the effective radius of the plutonium would grow. In that case the return to dryness would not be exactly horizontal, but would exhibit a slight downward slope. If the plutonium were not carried away (perhaps to one of the conditions illustrated in Fig. 1), the system would eventually be carried by these repeated excursions down to point K where it could become dry autocatalytic. Incremental dispersion and the associated incremental increase in $k_{eff}$ leads to a slow approach to criticality by several routes. However the plutonium could be dispersed suddenly. These mechanisms could be natural events such as volcanic action, earthquakes, or more modest geologic shifts. They could be man-made events also, such as well drilling, mineral exploration, or attempts at recovery of the buried material. High concentrations of nuclear material might be attractive sites for acts of malicious human intent as well.

Another way of describing the situation illustrated in Fig. 2 is that after TFM dispersal in the surrounding moderating medium ($SiO_2$) the neutrons can reach more nearly thermal energies, for which their reaction cross sections are much higher than those in the original undermoderated system, and the same mass can therefore become supercritical. This situation is illustrated in Fig. 3 which shows an inner sphere of $SiO_2$ with density of 2.2 containing 75 kg of [239]Pu immersed in an infinite medium of $SiO_2$. When the [239]Pu is confined to the 50-cm radius, the system is comfortably subcritical at $k_{eff}$ = 0.85. However if the same 75 kg of material were spread uniformly into the surrounding $SiO_2$, it would pass through criticality at about 100 cm and reach a maximum $k_{eff}$ of 1.12 at about 150 cm. This behavior is illustrated further in Fig. 4 where the value for $k_{eff}$ is given for [239]Pu in $SiO_2$ in a spherical geometry as a function of radius for two masses of [239]Pu. The value of $k_{eff}$ is approximately at its minimum at a radius of about 50 cm and reaches its maximum at about 150 cm. Once the system passes through criticality and starts to generate significant energy, the [239]Pu is expected to further disperse, most probably as a result of vaporization, as described in Appendix A. Because the system is characterized by positive feedback, it could drive itself by dispersion to an accelerated energy release with significant nuclear yield. The situation is in marked contrast to criticality accidents on the earth's surface which are terminated by explosive dispersion of the material after the yield has reached about a kilogram of high explosive equivalent if
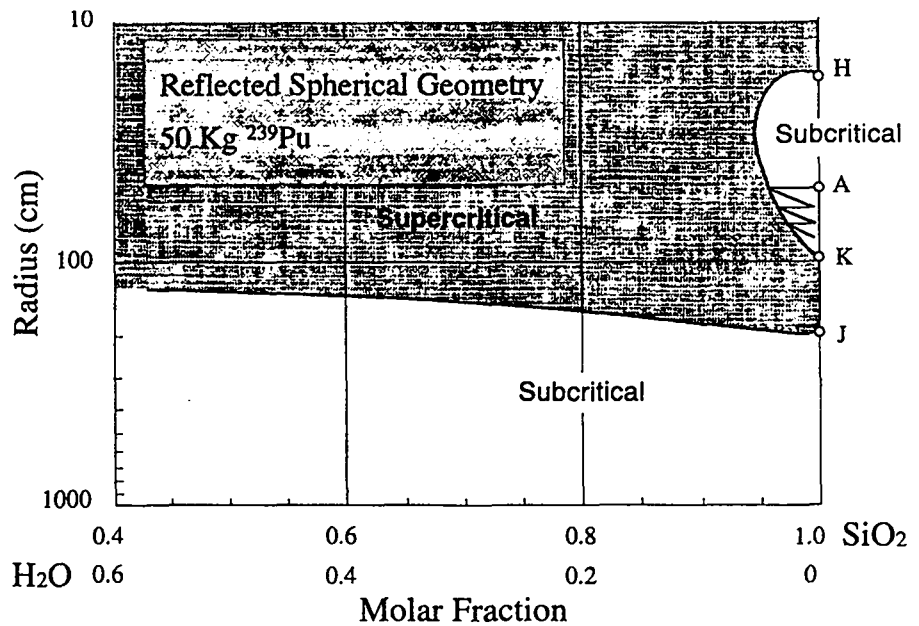


Figure 2. *Criticality conditions for 50 kg of* [239]*Pu in a sphere surrounded by* $SiO_2$ *reflector. The ordinate is the radius of the volume containing the plutonium. The abscissa is the molar apportionment of the water and* $SiO_2$ *in the sphere. The shaded region identifies regions of supercriticality. The other regions are the subcritical regions and represent those subcritical mixtures which could be placed underground. Systems which undergo rearrangements taking them to criticality can have positive or negative feedback as described in the text.*
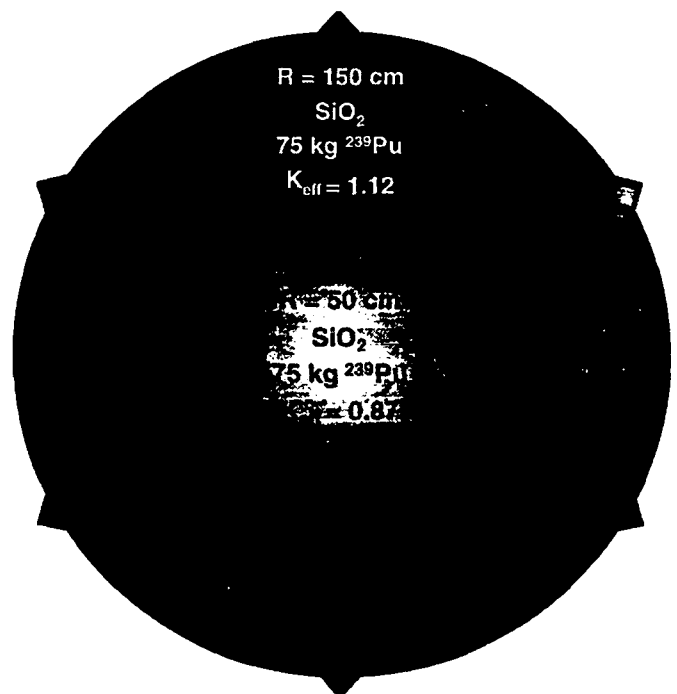


Figure 3. *The increase in* $k_{eff}$ *with dispersion. Initially 75 kilograms of* [239]*Pu stored underground in a one meter diameter sphere of* $SiO_2$ *would be comfortably subcritical with* $k_{eff}$ *= 0.87. As the plutonium is dispersed into the surrounding rock,* $k_{eff}$ *grows and when it has dispersed with uniform density to 150 cm radius,* $k_{eff}$ *has risen to 1.12.*

other factors don't act to cause a more benign event. Nuclear excursions are self-terminating above ground but can be self-enhancing underground.

The transition to high $k_{eff}$ for the overmoderated wet systems (cases C, D, E, F in Fig. 1) is a much simpler process, with the reduction of water concentration first by warming and then by conversion to steam.

The usual negative feedback mechanisms present in nuclear reactors (fuel temperature coefficient, moderator temperature coefficient) are either unavailable or relatively small, so the previously described situations might indeed "run away" with appreciable probability and release significant energy. This energy release can be rapid, and the generation of nuclear yields in the hundreds of tons of high explosive equivalent cannot be ruled out.

### Estimates of Explosive Yield

Once a system has become prompt critical, the fission energy yield will increase approximately exponentially until the energy generation is terminated by some physical change in the system. Above ground, this mechanism could be disassembly resulting from the nuclear energy generation after it had reached a level of a kilogram or so of high explosive. Underground however, the fissile material is confined and surrounded by rather good moderating material and this disassembly mechanism is not available.

The course of the confined explosion is determined by the characteristics of the surrounding medium and is described in some detail in Appendix B. The fission chain reaction can be terminated either because of expansion of the system or from an increase in temperature or both together. The surrounding rock first acts to confine the excursion as the energy builds up. If the rock is taken to be solid $SiO_2$, tuff, or granite, the rock is rather stiff until the pressure nears 30 GPa (0.3 megabars). As this pressure is approached, these materials undergo a phase change with an associated increase in density by a factor of about two. The compression of the surrounding rock provides expansion space for the supercritical mixture.

About 10% of the energy generated goes into compression of the rock. The main portion is spent in heating the gaseous rock. For systems containing about 100 kg of TFM with 1/v dependence of absorption cross sections, the temperature of the gas, when it becomes subcritical, is about 4 eV. The total energy generated in the expansion phase by the time the system passes to subcriticality is $1.3 \times 10^{12}$ joules or about 0.3 kilotons. If the energy generation rate is slow enough, the system might expand before reaching the 30 GPa level in which case the yield could be substantially lower as explained in Appendix B. In a fission nuclear weapon the temperature generated is much higher but the mass much smaller than the tons of rock in the sphere, so the yields can be roughly of the same size even though the temperature is lower in the rock. The conditions under which the system reaches subcriticality have been confirmed by Monte Carlo calculations to be close to those estimated in Appendix B analytically and therefore we believe that the nuclear yields projected here are confirmed to about a factor of two[6].

If other batches of fissile material were buried within about 4-5 meters of the original site, the other batches would probably be vaporized and dispersed by the heat or fission neutrons released by the first explosion and could therefore be driven supercritical with probable subsequent explosions. Even though the average concentration of the fissile material might be small in an underground storage facility with the fissile
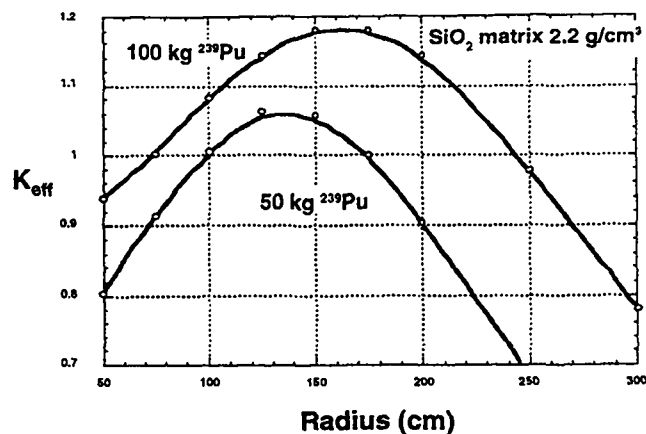


*Figure 4. The change in $k_{eff}$ with dispersion. The situation shown in Fig. 3 is presented in more detail here where curves of $k_{eff}$ vs radius of dispersion in spherical geometry are given for masses of 50 and 100 kg of $^{239}Pu$ in $SiO_2$ of density 2.2 g/ cm³. It is assumed that the plutonium loading is uniform in the sphere and the temperature is taken to be 25 degrees Celsius. It is important to note that these curves have a positive slope as the system first passes through criticality. Once criticality is reached, the fission heat generated could vaporize the plutonium and lead to further dispersion and higher criticality. Such a system therefore would be autocatalytic.*

material well separated at discrete sites, supercriticality at one site could spread throughout the storage facility .

An additional case of importance arises if the waste were to rearrange itself into an extended slab geometry. Such geometries are characterized by large length- and width- to thickness ratios, such that neutron leakage from the lateral areas is small compared to the leakage from the large extended surfaces. The analytical approximation for these geometries is the infinite plane source. For this situation there is only leakage from the two extended surfaces of the source, and the final yield per kilogram of TFM can be substantially higher than for the sphere. In addition, as is shown in Appendix B, the leakage does not increase as the density decreases. Therefore only the temperature rise and the associated neutron spectral shift will increase the leakage and terminate the energy generation from an autocatalytic critical excursion. For an extended rock slab of thickness 2 meters and for a fissile material density of 3 kg of $^{239}Pu$ per cubic meter, the temperature rises to about 6 eV with energy generation of about 50 tons per kilogram of TFM. The reason that the energy generation per kilogram of fissile material can be so much larger for the plane than for the sphere (3 tons per kilogram of TFM), even though the temperatures are nearly the same (4 vs. 6 eV), is the difference in the mass of rock participating. For the 1-meter radius sphere containing 100 kg of TFM, the rock mass contained is about 8 m³ whereas for the slab the same amount of fissile material is distributed in a rock mass that is about ten times larger.

### Weapons-grade plutonium and real rock

In the previous analysis, pure $^{239}Pu$ was used as representative of TFM. While this is a good approximation for high enrichment uranium fuel (almost entirely $^{235}U$), in reality, plutonium almost always is accompanied by a significant component of $^{240}Pu$. This isotope exhibits a resonance at 1.05 eV which is more than 100,000 b high at its peak. It operates as a trap to neutrons moderating down to thermal energy. The trap

*Table 1. Composition of Several Underground Media*

| Compound | Westerly Granite | Sandstone | Nevada Alluvium | Limestone |
|---|---|---|---|---|
| $SiO_2$ | 73.9 | 78.3 | 71.6 | 5.2 |
| $Al_2O_3$ | 14.9 | 4.8 | 12.1 | 0.8 |
| $H_2O$ | 0.0 | 0.0 | 4.0 | 0.8 |
| $K_2O$ | 4.5 | 1.3 | 3.5 | 0.3 |
| CaO | 3.3 | 5.5 | 2.4 | 42.6 |
| MgO | 0.0 | 1.2 | 0.0 | 2.7 |
| FeO | 2.0 | 1.4 | 0.0 | 0.5 |
| $CO_2$ | 0.0 | 5.0 | 0.0 | 41.5 |
| Total | 98.6* | 97.2* | 93.6* | 94.4* |

\* Plus lesser amounts of other oxides
\# F. J. Pettijohn, "Sedimentary Rocks," Published by Harper Brothers (1948)

is very efficient for dry systems, where neutrons lose a small fraction of their energy for each collision and therefore cannot easily bypass the resonance on their way to thermalization. The effect of the resonance is to significantly reduce the possibility for undermoderated criticality for w-Pu in dry systems. However the half-life for $^{240}$Pu is 6,600 years whereas the $^{239}$Pu decay rate is about four times slower, and the daughter product of $^{240}$Pu is $^{236}$U, which does not exhibit the same resonance behavior. Therefore the longer weapons-grade plutonium remains in permanent storage, the higher will be the risk of a dry spontaneous supercriticality event. After 25,000 years weapons-grade plutonium will be functionally equivalent to $^{239}$Pu. In the same period of time the plutonium contained in spent fuel (spent fuel standard isotopic composition) will be transformed by decay into weapons-grade plutonium. Decay eventually will convert w-Pu and c-Pu into almost pure TFM.

Small quantities of water or other hydrogen-bearing materials in the rock medium could lead to a bypass of the $^{240}$Pu resonance and therefore significantly lessen its negative effects on the plutonium reactivity. The fraction of energy loss by a neutron in hydrogen in fact is very large so that the probability of a neutron escaping capture in the 1.05-eV $^{240}$Pu resonance in between hydrogen collisions can be very high.

Another simplification introduced in the paper is the use of pure $SiO_2$ as representative of rock. In reality rock can have widely different compositions, as shown in Table 1. While the moderation properties of most rocks are very similar, some rock constituents have lower cross-sections than silicon, such as calcium or carbon, while other possible constituents have higher cross-sections, such as sodium and potassium. The effects of trace elements with high cross sections, such as rare earths, are usually negligible. Considering the composition of Nevada tuff (Topopah Spring tuff), the overall thermal cross-section for neutron absorption is about 50% larger than for pure $SiO_2$, which takes us to the conclusion that 50% larger quantities of TFM would be needed when $SiO_2$ is replaced by the Topopah Spring tuff to reach the same conditions.

**Application to specific W-Pu disposition options**

Spontaneous supercriticality could be a significant concern for any of the plutonium and other TFM disposition proposals now under consideration which require permanent, unattended underground storage.

*Deep borehole storage*

This proposal[6] would involve the emplacement of plu-

tonium probably with vitrification into boreholes of about 50-cm diameter at a depth of 2000 to 4000 meters so that a linear array of casks would be placed one above the other. They would be separated by suitable filler material. Perhaps 50 tons of w-Pu would be placed in one such hole. If the casks were two meters long and each contained 100 kg of w-Pu, 500 such casks would be required. We have seen that yields of up to about several hundred tons could conceivably result from the autocatalytic explosion of 100 kg of buried w-Pu in this form. The resulting heat would vaporize all material within about an 8- to 10-meter diameter, possibly initiating explosions of the same size in the w-Pu above or below the first explosion, with further coupling possible.

*Geologic storage of w-Pu, HEU, naval spent fuel and research reactor spent fuel*

Geologic storage of these materials as canisters of concentrated material in a rectangular planer array would carry a risk of spontaneous explosion. The first spontaneous explosion could propagate to other emplacements in the array, if the emplacements are not well separated. If the average density of fissile material were high enough, the infinite slab geometry could be approached, and the autocatalytic criticality for this case could result in much larger yields per kilogram than possible for isolated events.

*Destruction by underground nuclear detonation*

A Russian group[7] has proposed the destruction of the nuclear weapon stockpile by placing a number of weapons underground and destroying them with a nuclear explosion. The practical implementation of this would probably include several tons of w-Pu for each nuclear detonation. Instead of the w-Pu being trapped in fused rock as suggested by the Russian group, an alternative prospect would be that the plutonium would be vaporized and dispersed into the surrounding medium. If criticality were reached in one or more locations *after this dispersal*, the autocatalytic behavior could be initiated. The yield from the event therefore might be much larger than anticipated.

*Geologic storage of reactor spent fuel*

From the spontaneous criticality perspective, underground storage of commercial spent fuel can be done safely over the short term for either a thermal or a fast spectrum because the amount of $^{238}$U poison present in the fuel is so large and, if the system is dry, because of the presence of $^{240}$Pu. The plutonium and uranium however could separate over time since the uranium solubility in an oxidizing environment is about 300 times higher than that for plutonium. Furthermore, the $^{240}$Pu has a 6,600 year half life and in two half lives the plutonium isotopic composition will approximate the isotopic composition of w-Pu and the criticality risk for stored material will become larger as more time passes. Water infiltration could further reduce the effectiveness of the $^{240}$Pu poison and the mass of TFM that could become autocatalytic. Eventually the amount of c-Pu turned into w-Pu would be much larger than the 50-100 tons of w-Pu presently requiring disposition. Therefore in the long term, after the canister integrity is lost, reactor spent fuel could be subject to the autocatalytic criticality scenarios described in the paper.

**Summary**

As long as the canisters containing thermally fissile material maintain their integrity, underground criticality is not a concern. When the canisters have been breached and the

fissile material loosened and rearranged, spontaneous criticality with positive feedback is possible. If the feedback is rapid, explosions of significant nuclear yield can occur. The main points can be summarized as follows:

1. Criticality underground is not always characterized by negative feedback; situations with positive feedback can readily be reached if the TFM migrates from its original emplacement to a new geometry or location.

2. Both wet and dry autocatalytic conditions are possible, with TFM quantities in the kilogram range behaving autocatalitically in some wet scenarios.

3. The autocatalytic feature of buried w-Pu, HEU, naval spent fuel, and spent research reactor fuel could give rise to sequential ignitions when this concentrated nuclear material is stored in an extended array.

4. A simple means of roughly estimating the yield from autocatalytic phenomena is described which does not require detailed information on the explosion time history and material equation of state.

5. The maximum yields for the sphere containing 100 kg of TFM and for the infinite slab geometries were estimated respectively at 3 and 50 tons per kg of TFM, with the yield for the slab being relatively insensitive to the equation of state.

6. The role of the 1 eV resonance in $^{240}$Pu as a temporary (6,600 year half-life) barrier to commercial spent fuel spontaneous supercriticality is pointed out. The barrier largely disappears if water is present in the storage medium.

## Acknowledgments

The authors acknowledge the helpful criticism generated in an internal review of this work at the Los Alamos National Laboratory.

## References

1. For example, *"Management and Disposition of Excess Weapons Plutonium,"* Committee on International Security and Arms Control, National Academy of Sciences, National Academy Press, Washington, DC (January 24, 1994); "Dismantling the Bomb and Managing the Nuclear Materials," OTA-0-572 Washington, DC, US. Government Printing Office (September 1993); "Limiting the Spread of Nuclear Weapons," B. G. Chow and K. A. Solomon, National Defense Research Institute, RAND Corporation, (1994)

2. For example, two panels presented testimony on April 19, 1994 at the hearing on the Disposition of Plutonium and Highly Enriched Uranium before the Military Application of Nuclear Energy Panel, Committee on Armed Services, U. S. House of Representatives. An external panel consisting of Richard Garwin, Brian Chow, and Thomas Cochran presented the perspectives from these studies and an internal panel consisting of Charles Curtis from the Department of Energy, Harold Smith from the Department of Defense, and Robert Einhorn from the Department of State echoed the results from the first panel.

3. *"Management and Disposition of Excess Weapons Plutonium,"* Committee on International Security and Arms Control, National Academy of Sciences, National Academy Press, Washington, DC, p.247-248 and table 3-3 on p.68 (March, 1994)

4. This code has been used world wide for many years for neutron and gamma ray transport calculations. A manual is available, *"MCNP-A General Monte Carlo Code for Neutron and Photon Transport,"* Los Alamos National Laboratory Report LA-7396-M, Rev. 2. (1991)

5. G. A. Cowan, *"A Natural Fission Reactor,"* Scientific American 235, 36-47 (July 1976)

6. *"T-4 Handbook of Material Properties Data Bases, Vol. 1c, Equations of State"*, edited by K. S. Holian, Los Alamos National Laboratory Report LA-10160-MS (1984).

7. Y. A. Trutnev and A. K. Chernyschev (Arzamas-16), presented at the *"Fourth International Workshop on Nuclear Warhead Elimination and Nonproliferation,"* Washington, DC, February 26, 1992.

## Appendix A. The approach to criticality for undermoderated systems

The intention here is only to show that prompt criticality is possible rather than to treat the subject comprehensively. After sufficient time has passed for the emplacement canister to lose its integrity, water will enter the canister and start the dissolution of the vitrified material. The glass will already be thoroughly cracked at emplacement as a consequence of the cooling process and has no structural integrity in itself, it having been held together by the canister[A1]. The water will penetrate throughout the vitrified mass and dissolution of the contents of the contained will commence. Some solubilities at about 300 K are listed in table A1.

The $B_2O_3$ in the borosilicate glass is 100 times more soluble than the $SiO_2$, and $SiO_2$ and $B_2O_3$ are several orders of magnitude more soluble that $PuO_2$. Therefore the water will move through the cracks preferentially removing the boron first and then the $SiO_2$.

The solubilities for water if it were silicate-saturated would be lower than the above solubilities if the vitrified plutonium were at the same temperature as the surrounding rock. However the decay heat of the plutonium will warm the water entering the vitrified mass bringing the silicate to an unsaturated condition. The $B_2O_3$, which makes up about 15 % of the mass of the glass will be leached away much faster than the $SiO_2$ and the preferential leaching of the $B_2O_3$ will destroy the structural integrity of the glass. After a significant portion of the $B_2O_3$ (and $SiO_2$) has been leached away, the vitrified mass might take on a spongy character with substantial volume available for water and the mass will reach criticality upon water ingress.

A rise in temperature averaged over the mass of the sphere would cause a larger rise in the water temperature at the center, since the power density in a uniformly loaded sphere depends strongly on the radius. The result would be steam generation starting near the center and a possibly violent expulsion of a water and steam mixture to the outside of the vitrified mass, especially for systems that do not contain signifi-

cant fractions of resonance absorbers such as $^{238}U$. This would be repeated periodically as water reentered the system. The consequences are almost certainly further cracking of the vitrified material perhaps eventually taking it to powder, loss of integrity of the surrounding rock by the repeated steam bursts possibly converting it to rubble, and more rapid dissolution of the plutonium-bearing mass.

Once the undissolved $PuO_2$ is freed by the disappearance of the boron and part of the glass, it might be carried out of the vitrified mass and into the cracks and rubble by the steam bursts. Some of the plutonium might be carried with the water to the bottom of the glass-containing cavity or elsewhere possibly leading to the criticality described for cases C, D, E, F in Fig. 1. Some of it could however disperse in the cracks and rubble surrounding the original site.

A last steam burst might carry enough additional plutonium into the rock to establish dry prompt supercriticality. A much larger reactivity increment might also occur at any time as a result of sudden collapse of the glass cylinder onto itself following erosion of a substantial portion of its volume. At this point the autocatalytic criticality could proceed towards explosive energy release, if the feedback mechanism is fast enough. Vaporization could mobilize the plutonium to the point that such releases are possible.

The plutonium should not be uniformly distributed in the rock, but rather dispersed in many cracks. Only about 1/20 of the energy of the fission is carried by the neutrons and this component is deposited in the rock in the moderation process. The bulk of the energy is deposited by the fission products in the plutonium and its immediate vicinity. The ratio of plutonium mass to the mass of rock is about 1/50. The heating rate for the plutonium could then be up to 50 x 20 = 1000 times faster than for the rock and lead to plutonium vaporization well before the surrounding rock is heated appreciably. We therefore believe that plutonium vaporization in these circumstances is possible. Depending on the degree of vaporization and the velocity with which the plutonium disperses through the cracks in the surrounding rock matrix, the small negative temperature coefficient (calculated to be $-1 \times 10^{-5}/$ $^0C$ for the relevant conditions) might not be able to counterbalance the reactivity increase due to the dispersion of the plutonium in the rock. From Fig. 4, the value for $k_{eff}$ would increase by about 0.0025 per cm of spreading. If each degree of heating in the rock corresponds to a plutonium temperature increase of 1000 K, then the plutonium volume could vaporize after only a few-degree increase in rock temperature. A two-fold increase in volume corresponds to an increase of up to 20% for the dispersion radius of plutonium in the rock. For a sphere of 50 cm in radius, this would cause $k_{eff}$ to go up from slightly above 1 to 1.025, even including the effects of the negative temperature coefficient

In order for the generated energy to be explosive in nature, the time scale of the driving mechanism (increase in $k_{eff}$) would have to be comparable to the energy generation time scale. The energy generation constant (e-folding time)

for these systems is on the order of one millisecond, so that a plutonium expansion velocity in $SiO_2$ of about 100 m/s could bring about a $k_{eff}$ of 1.1 in within very few e-folding times for the case of 100 kg of $^{239}Pu$. These values for the expansion velocity might indeed be possible in the presence of such pronounced overall positive reactivity feedbacks.

## References

A1. P. B. Parks, Savannah River Site, Private Communication, December 1994

A2. G. E. Michaels, "Potential Benefits of Waste Transmutation to the U. S. High Level Waste Repository," International Conference on Accelerator-Driven Transmutation Technologies and Applications, Las Vegas, Nevada, July 1994

A3. M. Wilson, et al, "Total System Performance Assessment for Yucca Mountain - Second Iteration" (TSPA-1993), SAND93-2675, Sandia National Laboratory, April 1994.

## Appendix B.    Explosive Yield

Once the system has become prompt critical, the fission energy yield will increase approximately exponentially until the energy generation is terminated by some physical change in the system. Above ground, this mechanism could be disassembly resulting from the nuclear energy generation after it had reached a level of a kilogram or so of high explosive. Underground however, the fissile material is confined and surrounded by rather good moderating material, and a disassembly mechanism leading to quick shutdown in principle is not available. Other shutdown mechanisms will have to be invoked to terminate the event and the final yields to be expected could be substantially larger than for above ground scenarios. To gain some understanding of the order of magnitude involved, we will attempt to simply evaluate these yields in some idealized yet relevant conditions. The exponential time constant for fission power generation P is referred to as $\alpha$ and the appropriate expression is

$$P = P_0 e^{(\alpha t)}. \tag{1}$$

The quantity $\alpha$ is generally time dependent and is given by

$$\alpha = (k_{eff} - 1)/\tau \tag{2}$$

where $k_{eff}$ is the neutron multiplication factor and $\tau$ is the lifetime for neutrons in the system. For thermal systems, this time can be shown to be given by

$$\tau = 1/\Sigma_a v \tag{3}$$

where v is the thermal neutron velocity and $\Sigma_a$ is the macroscopic absorption cross section[B1] for the medium. For 100 kg of $^{239}Pu$ spread uniformly throughout a one-meter-radius sphere of $SiO_2$, we find $\tau \approx$ 100 microseconds. For $k_{eff} = 1.1$, which appears to be a typical value for these supercritical systems, the value for $\alpha$ in inverse microseconds is $\alpha = 1/$ 1000 so that the yield will increase by the factor e in 1 millisecond. The time for sound to move one meter in $SiO_2$ is about 200 microseconds so that a one

*Table A1. Solubilities of relevant materials in water at 300 K.*

| Material | Solubility (Mol/liter) | Reference |
|---|---|---|
| $PuO_2$ | $10^{-7}$ | Michaels[A2] and Wilson[A3] |
| $UO_2$ | $10^{-4.5}$ | Michaels[A2] and Wilson[A3] |
| $SiO_2$ | $\sim 10^{-3}$ | Derived from comparing solubility of $UO_2$ in water and silicate-saturated water |
| $B_2O_3$ | $10^{-0.8}$ | CRC Handbook (1.01 grams per 100 ml) |

meter radius system can adjust itself fairly well to the increasing energy deposition by fission without shock effects.

The course of the confined explosion is determined by the characteristics of the surrounding medium. The fission chain reaction can be terminated either because of expansion of the system or from an increase in temperature. After the excursion is launched, the termination of the excursion may be summarized briefly. The surrounding rock first acts to confine the excursion as the energy builds up. If the rock is taken to be solid $SiO_2$, tuff, or granite, the rock is rather stiff until the pressure approaches about 30 GPa (0.3 megabars). As this pressure is approached, these materials undergo a phase change with an associated increase in density by a factor of about two[6].

The compression of the surrounding rock provides expansion space for the supercritical mixture. Eventually the expansion of the system and the increase in the temperature together cause the neutron leakage to increase and the system to cross into subcriticality. In a fission nuclear weapon the temperature generated is much higher but the mass much smaller than the tons of rock in the sphere, so the yields can be comparable even though the temperature is lower in the rock.

The yield estimation is based on a model assuming an initial uniform distribution of fissile material in $SiO_2$ at a density of 2.2 with radius a surrounded by rock of the same density and composition. $SiO_2$ is a fairly good approximation since it is a major fraction of the material in soil or rock of various types and since the lighter elements have very small neutron capture cross sections with the exceptions of sodium, chlorine, boron, lithium and nitrogen. These more absorptive elements are found only in "mines" such as salt deposits which have not been selected for geologic storage of high-level waste. The compositions of several underground media are given in Table I. Note that limestone is primarily $CaCO_3$ which has lower neutron absorption cross section than $SiO_2$, and is a better moderator. Therefore supercriticality is more readily reached than for the media consisting primarily of $SiO_2$. On the other hand, the absorption cross section of Topopah Spring Tuff (Nevada rock) is about 50 % higher than that of $SiO_2$ and the moderation properties of this rock and $SiO_2$ are nearly the same. For simplicity, $SiO_2$ is used as a surrogate in this calculation for the various types of rock which might make up the storage medium.

For the fissile material quantities and densities and for the normal density of $SiO_2$, the likelihood of losing a neutron by leakage after thermalization is small. However the slowing down range for a neutron in $SiO_2$ at normal density is about 100 cm so that for a 200-cm diameter sphere, there is a substantial loss of fast neutrons before they slow down. Each fission in $^{239}Pu$ produces 2.88 neutrons. In order to sustain a chain reaction, one of these must be spent in causing fission of the next $^{239}Pu$ nucleus, an additional 0.35 must be spent because the ratio of capture to fission is 0.35. In addition a few percent of these neutrons are lost to absorption in the $SiO_2$ so that altogether about 50% of the fission neutrons remain and could be lost by leakage while still maintaining criticality. The leakage of fast neutrons depends much less on the temperature and density of the system than does the loss of moderated neutrons for which the competition between absorption and leakage is important. Assuming that half of the neutrons are fast and leak out anyway, the system should become subcritical when the leakage of thermal neutrons becomes significant (larger than 20%). We analyze the system as a thermal neutron diffusion problem by assuming that all neutrons start from the center of the sphere and by then calculating the probability of 20 % leakage. The number of neutrons n(r) that are absorbed

in the spherical shell of volume dV at radius r when the flux at r is $\Phi(r)$ is given by

$$n(r)dr = \Sigma_a \Phi(r)dV. \qquad (4)$$

$\Sigma_a$ is the macroscopic absorption cross section for the material in the sphere. The volume element dV is $4\pi r^2 dr$ and the flux in the sphere from a point source of neutrons is given[B1] by

$$\Phi(r) = (S/4\pi Dr)e^{-r/L} \qquad (5)$$

where S is the source intensity in neutrons per second, D is the thermal neutron diffusion constant given by $1/3\Sigma_s$ and $\Sigma_s$ is the macroscopic scattering cross section. The parameter L is a nuclear engineering term called the diffusion length and is related to the distance a neutron will travel before absorption. In terms of defined parameters[B1] it is

$$L = 1/(3\Sigma_a\Sigma_s)^{1/2} \qquad (6)$$

Substituting in (5) and (4) and dividing n(r) by S to convert it to the probability P(r), we have after integrating from 0 to a the probability of absorption of the thermal neutron in the sphere

$$P(a) = 1 - (a/L + 1)e^{-(a/L)} \qquad (7)$$

The parameter L depends on the scattering cross section in the sphere which is independent of the temperature and on the absorption cross sections which have a temperature dependence in eV of $(.025\ eV/T)^{1/2}$ where 0.025 eV is the starting temperature of the medium in electron volts. The absorption cross section for the TFM is that for $^{239}Pu$ at thermal energy. Both $\Sigma_a$ and $\Sigma_s$ depend on the atomic density of the sphere which changes with the radius a as the sphere of constant mass expands. The problem then is to find the radius at which the system becomes subcritical. Setting P(a) = 0.80 in (7) gives a/L = 3. If the sphere contains 100 kg of TFM in $SiO_2$ at a density of 2.2, we find using 6 combined with a/L = 3 that

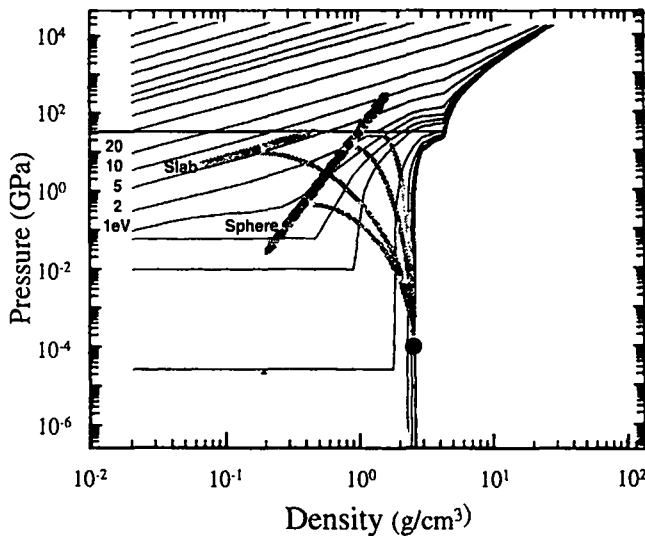$$a = 271\ cm\ (0.025eV/T)^{1/8}. \qquad (8)$$

Therefore for a sphere of vaporized TFM and $SiO_2$ which expands owing to fission heating to the temperature T in eV conserving $SiO_2$ and TFM mass, 8 gives the radius at which it becomes subcritical. We must consider the equation of state of $SiO_2$ to estimate the temperature. The equation of state also will provide the energy density in the sphere at that temperature so that the fission yield when the system enters subcriticality can be obtained by multiplying the energy per unit mass by the total mass in the sphere.

The phase diagram for granite is shown[6] in Fig.B1. The phase diagram for $SiO_2$ and tuff are similar. The left figure gives the pressure vs. density for several temperatures. The curve on the right shows the energy density as a function of pressure for various temperatures. The temperatures in eV are shown for a few of the curves in the left hand side of both upper and lower figures. Upon reaching supercriticality underground, the system starts out at ambient temperature at a density of 2.2. This is shown as the circle in the left figure. The system builds up energy so that the TFM is vaporized with the rock following soon and the pressure builds. The TFM would disperse further into the rock upon vaporization. For the purposes of this calculation it is assumed that the radius of the volume containing TFM grows to a sphere with 100 cm radius which, as is shown in Fig.4, is substantially prompt supercritical. If the pressure reaches 30 GPa (0.3 megabars),

the rock completes a phase change to stiskovite which has a density higher by a factor of two than the original rock. Above 30 GPa the rock becomes quite stiff. The sphere containing TFM at a temperature of about 0.3 eV becomes a mixed vapor of TFM and rock at lower than original density. Since the rock outside of the gas is "infinite" in extent, the surrounding rock can continue to yield as the fission energy builds creating more volume. The pressure underground does not increase above the 30 GPa pressure because of the extra space provided by the phase change unless energy multiplication is very much higher and faster than established here for these large low TFM-density systems. The radius of the spherical system containing fissile material simply grows larger.

As the volume and temperature increase, the density decreases and the system moves to the left along the constant-pressure horizontal line at 30 GPa pressure. As the system expands, the sphere becomes less absorptive and this effect is further enhanced by the decreasing cross sections with increasing temperature. Expression (8) gives the relationship between temperature and radius (density) at which the system passes into subcriticality. This point can be obtained in a density-temperature relationship by substituting for the radius a using the simple relationship between density and radius $\rho = \rho_0(a_0/a)^3$ where $\rho_0$ and $a_0$ are the initial density and radius. Substituting for a in 8 to turn it into a temperature-density relationship gives

$$\rho/\rho_0 = (100/271)^3 (T/0.025 \text{ eV})^{3/8} \qquad (9)$$

This curve of density вs. T is shown in the left part of Fig. 5, labeled "Sphere". The curve crosses the 30 GPa pressure line at 4 eV where the density is 0.8 g/cm$^3$ and a = 140 cm. This is the point at which the systems becomes subcritical. This estimate from thermal neutron diffusion theory was confirmed by a Monte Carlo calculation for $^{233}$U, for which $k_{eff}$= 0.99± 0.03 was found. The same curve is located in the figure to the right and also labeled Sphere. The energy density is found to be 100 MJ/kg. Multiplying by the mass in the sphere gives $1.2 \times 10^{12}$ joules. The energy stored in the compressed mass pushed out by the increase in sphere radius from 100 to 140 cm is smaller[6] at $0.1 \times 10^{12}$ joules. The total energy generated in the expansion phase by the time the system passes to subcriticality is therefore $1.3 \times 10^{12}$ joules or about 0.3 kilotons.

While this is an upper limit value to the actual yield, the yield estimate does not depend strongly on the details of the phase change. As long as the rate of energy generation is large enough that the system reaches the horizontal line at 30 GPa before reaching the 4 eV temperature, the yield will be the same. However if the energy generation is too slow, the system might never reach the horizontal line. It would instead intercept the $\rho$ vs. T curve (Eq. 9) at a lower temperature as illustrated by the other path-lines in Fig. B1. If it intercepted at a temperature of 0.5 eV, the yield would be lower by a factor of ten (30 tons).

The e-folding time for multiplication in this system is about one millisecond so that nearly all of the energy is generated in about 3 milliseconds. The system next enters another slower energy generation phase during which the hot plasma
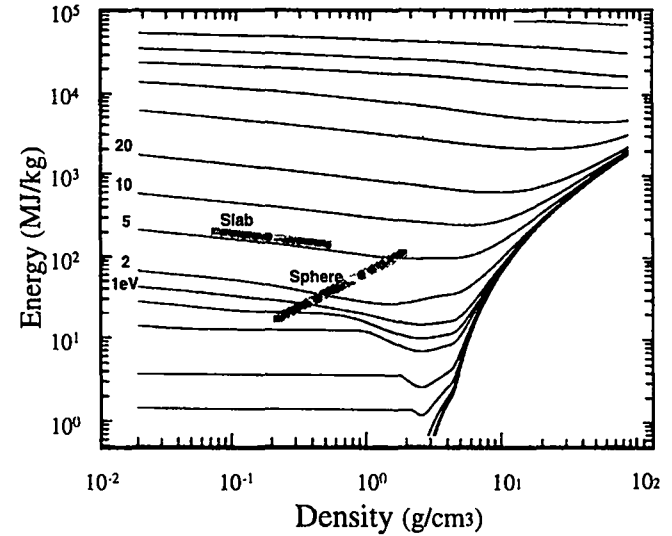


Figure B1. Equation of state for rock (Westerly Granite). The figure on the left shows the pressure in gigapascals (GPa's) as a function of the density of $SiO_2$. The different curves correspond to the different temperatures given in Kelvin on the right. 1 eV corresponds to 11600 degrees. The temperature in eV for a few of the curves is given just inside the ordinate scale. A phase change to twice the density occurs in the $SiO_2$ at a pressure of 30 GPa (0.3 megabars). This pressure is shown as the horizontal line. A cool supercritical system starts at its normal density of 2.2 (indicated by the circle) and moves vertically while expanding until it reaches the horizontal line. The change in density at this pressure of the surrounding rock, which is mainly $SiO_2$, creates space to accommodate the growing nuclear energy yield so that the pressure cannot rise higher. The system then moves horizontally to the left along the constant pressure line increasing in temperature as the system expands until the system becomes subcritical from the combination of expansion and temperature increase. The line shown labeled Sphere crosses the horizontal line indicating the density at which a sphere of $SiO_2$ of original radius 100 cm and containing 100 kg mass of fissile material would become subcritical. The figure on the right shows the energy density in the material also as a function of mass density. The corresponding point in the right-hand figure is the point of crossing in the upper figure and gives the energy density at subcriticality. For the sphere described above, energy generation stops at 4 eV when the system radius has grown to 140 cm. The energy density in the system is 100 megajoules per kilogram of $SiO_2$. The total fission energy reached at subcriticality is about 0.3 kilotons. The yield from the infinite slab geometry obtained using these curves is described in the text.

accretes rock from the spherical surface into the plasma increasing its density and cooling it with the result that the capture and the fission cross sections for the materials in the plasma increase. Leakage therefore decreases and the system returns to criticality and increases its temperature entering a quasi-steady state where the system remains critical and the energy is taken up by accretion of more rock to keep the system critical. The system grows until the capture in the rock is too large compared to the fission from the fixed amount of fissionable material to sustain criticality and the system becomes subcritical permanently.

For the spherical geometry just explored, the energy generation was terminated by the loss of neutrons into $4\pi$ solid angle, by the increase in temperature and finally by rock accretion. Probably the most likely means of practical storage of a much larger amount of weapons material such as 50 tons would be in a rectangular array of concentrated fissile material batches. The corresponding reactor physics problem pertinent after dispersion or coupling spreads the weapons material sufficiently is the infinite slab reactor. For such a system with thickness 2a in a vacuum, the same analysis for probability of absorption gives[6].
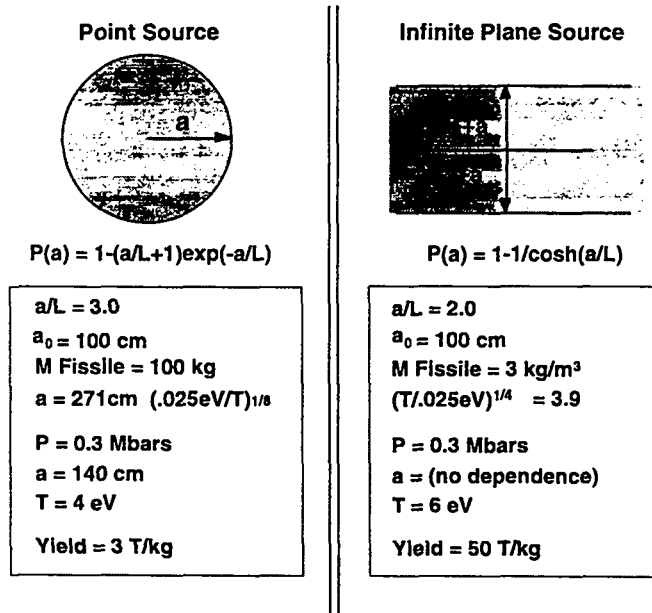
$$P(a) = 1 - \cosh(d/L)/\cosh[(a+d)/L] \qquad (10)$$

where d is the straight line extrapolation distance of the flux outside of the boundary a to a flux of zero. If this distance is taken to be 10 cm for a value of a = 100 cm, a value for a/L = 2 is found (compared to 3 for the sphere) for a leakage probability of 0.20 (same as for the sphere). The value of d is not very important as long as it is small compared to a. Using (6) in the same way as was followed to get expression (8) involving a and T, the result

$$(T/0.025 \text{ eV})^{1/4} = 3.9 \qquad (11)$$

is found for an infinite slab of thickness 2a = 2 meters and for a fissile material density of 3 kg of $^{239}$Pu per cubic meter. This interesting relationship (11) exhibits *no dependence on a* and shows that the infinite slab never goes subcritical because of expansion. The reason is that the solid angle for loss of neutrons from the infinite slab and the absorption probability don't change with the thickness of the slab so long as mass in the slab is conserved. In that case the slab thickness doesn't influence $k_{eff}$. The leakage solid angle also is smaller than for the sphere, roughly $4\pi/3$ for this case compared to $4\pi$ for the sphere. Once supercritical, the system only goes subcritical if the temperature gets too high. Solving 11 for the temperature at which the systems becomes subcritical gives T = 6 eV. This is shown as the constant temperature line Slab, in the left side of Fig. B1.

As shown from the corresponding line in Fig. B1 (right-hand side), the energy density os nearly independent of the mass density and is about 200 MJ per kg of rock. With the temperature rising to 6 eV, the energy generation would be about 50 tons per kilogram of TFM at which point the system would become subcritical because of reduction in the cross sections from the temperature increase alone. As in the case of the sphere, the expansion phase might be followed by an accretion phase which returned the system to supercriticality and the yield might increase further. The parameters and yields for energy generation are summarized in Fig.B2.

### References

B1. See for example, *"Introduction to Nuclear Engineering,"* by John R. Lamarsh, Addison-Wesley Publishing Company (1983).

| Point Source | Infinite Plane Source |
|---|---|



| $P(a) = 1-(a/L+1)\exp(-a/L)$ | $P(a) = 1-1/\cosh(a/L)$ |
|---|---|

| | |
|---|---|
| a/L = 3.0 | a/L = 2.0 |
| $a_0$ = 100 cm | $a_0$ = 100 cm |
| M Fissile = 100 kg | M Fissile = 3 kg/m³ |
| a = 271cm $(.025eV/T)_{1/8}$ | $(T/.025eV)^{1/4}$ = 3.9 |
| P = 0.3 Mbars | P = 0.3 Mbars |
| a = 140 cm | a = (no dependence) |
| T = 4 eV | T = 6 eV |
| Yield = 3 T/kg | Yield = 50 T/kg |

*Figure B2. Underground yield estimation results. The yield is estimated for spherical, cylindrical, and slab geometries. The amount of fissile material is: (1) 100 kg for a sphere of one meter radius, and (2) 3 kg per cubic meter for an infinite slab of two meter thickness. The base yield is the yield reached at first subcriticality as a result of expansion. The hot system then accretes additional mass from the surrounding rock, cools, and becomes critical again. This accretion process continues until the system becomes subcritical because of increased neutron capture from the additional mass competing with neutron absorption by the fissile material. The additional yield by accretion might be significant, but is not estimated here.*