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Underground Supercriticality from Plutonium and Other Fissile Material

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Abstract

Several widely endorsed solutions to the 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 consequences of such supercritical excursions could range from modest energy releases to the generation of explosive nuclear yields of up to a few hundred tons of high explosive equivalent from a single event. Without water, 50-100 kg of fissile material is required to reach autocatalytic criticality. Amounts as small as 2 kilograms 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. 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. 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 50-cm diameter and two-meters long borosilicate log would contain about 100 kg of plutonium. A single log of this material would be substantially subcritical owing to geometry and the deliberate addition of neutron poison. Other storage forms of the w-Pu would also 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.

Subcriticality could also be enhanced by the inclusion of neutron poisons and by choosing geometry and composition such that only fast neutrons could be effective in propagating a chain reaction. Even without poisons, w-Pu in these amounts and in these configurations 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 g/cm^3 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 for that radius are supercritical; those below are subcritical. Obviously material cannot be implanted underground as supercritical

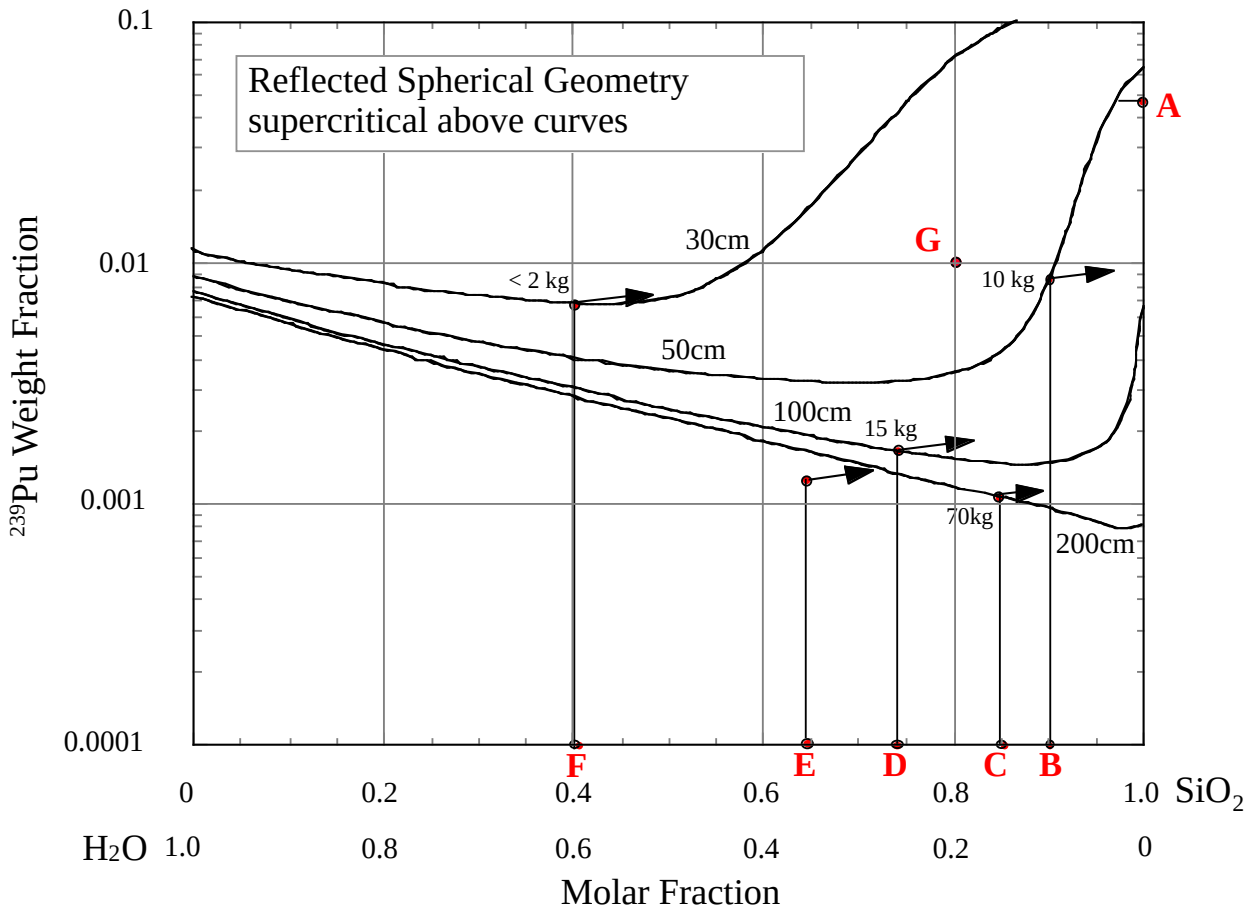

Figure 1. Examples of positive and negative feedback following underground criticality. *Criticality curves are given for spherical volumes of radius 30, 50, 100, and 200 cm for mixtures of ^{239}Pu, water and SiO_2 reflected by SiO_2. The ordinate is the weight fraction of ^{239}Pu in the sphere. A 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*

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 is water ejection. With regard to water ejection (indicated in Fig.1 by a move to the right): *if the system reaches criticality at a point where the pertinent curve has a negative slope, the system will move towards higher criticality (positive feedback) and could therefore be autocatalytic; if the system reaches criticality at a point where the curve shows a positive slope, the system will move towards lower reactivity (negative feedback) and will be self-limiting or self-terminating.* While the figure is only valid for spherical geometry, the criteria and behaviors illustrated are valid for many different shapes. We illustrate these criteria by examining six conditions labeled in the figure as A through F.

Case A - Water ingress in TFM + rock systems (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 50-cm spherical geometry, we see from Fig.1 that the mass fraction of ^{239}Pu mixed with SiO_2 must be less than about 0.065, for the system to be subcritical 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 for that geometry. If water enters this system (as shown in figure 1A), the composition of the system in Fig. 1 will move to the left, until it reaches wet criticality (the 50-cm curve) at a water mole fraction of about 2 % (0.6 % mass fraction). The system will generate fission energy, until the water is converted to steam and is thereby driven out of the system. If the water expulsion is complete, the system returns to its starting point on the right ordinate axis. 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 is frequently cited as an underground criti-

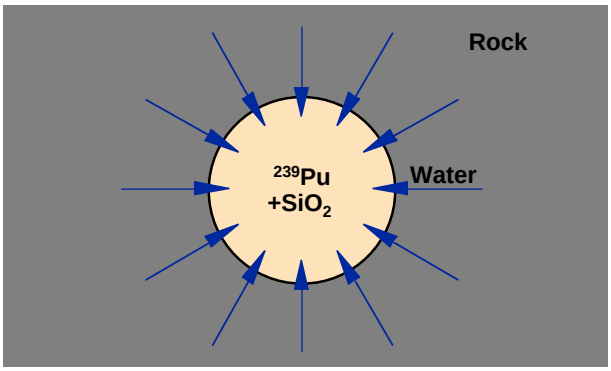

Figure 1A. *Water ingress in $^{239}Pu+SiO_2$ mixture. Criticality reached under these conditions has always negative feedback. Water can however boil locally and disperse some of the plutonium into the surrounding rock on its way out. This dispersal mechanism could eventually lead to dry supercriticality.*

cal 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 - TFM migration to wet rock systems (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 4.6 kg; with rock only the amount is 80 kg; for 30% mole fraction of water in rock the critical mass is only 3.4 kg. If some of the TFM should leave its original implantation site and migrate to a new volume with 50-cm radius and containing 10 % mole fraction of water (3.2 % 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 (see Fig.1B/C). Water-driven plutonium migration from waste sites has been observed (ref.). When a plutonium mass fraction of 0.009 had accumulated, corresponding to a mass of about 10 kg in the 50-cm radius, the configuration would reach the criticality curve for the 50-cm geometry and become critical, 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 addition or dispersion of the ^{239}Pu occurred, the system could move horizontally into and out of criticality indefinitely following repeated incursions of water.

Case C - TFM migration to wet rock systems (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 neutron energy loss per collision is not a relevant parameter for large-volume systems. 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 reached criticality upon hitting the 200-cm curve in Fig.1, at a ^{239}Pu 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 composition, as represented in Fig.1 also moves to the right into the supercritical region, and in doing so the system drives itself to *a higher* criticality, reaching its highest criticality in a dry supercritical state.

Case D - TFM migration to wet rock systems (positive feedback)

The accumulation of plutonium in a sphere of 100-cm radius with water present as a 27 % mole fraction, started at point D in Fig.1, will stop when criticality is reached at 15 kg. At that point, this system is in a region of negative slope on the criticality curve for the100-cm geometry, so that by expelling water, the system autocatalytically drives itself to the right into the supercritical region. 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 15 % of the original concentration. The reactivity evolution of this system is illustrated in figure 1D, which shows the positive feedback excursion as water warms up and expands and ultimately boils off leaving the system dry. Larger fractions of excess water with respect to the optimum moderation case (bottom of the curve) would have larger reactivity increases.

Case E - Drying-out of TFM deposits in rock (positive feedback)

This case illustrates another autocatalytic condition which is approached somewhat differently. It illustrates the deposition of plutonium in wet media to overmoderated subcritical concentrations of fissile material followed by drying of

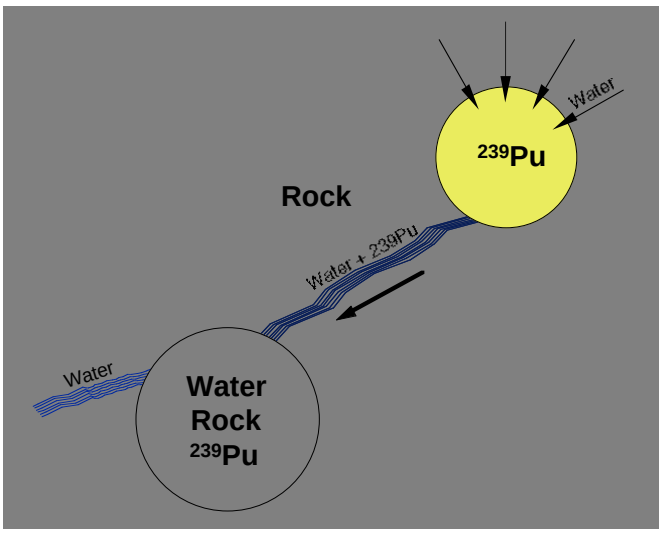

Figure 1 B/C. *Plutonium migration to wet rock systems. Water comes into the plutonium emplacement and carries off some of it in particle form. Eventually plutonium from many separate emplacements could collect in a wet rock volume. Criticality can be reached in this volume after enough plutonium is collected. The criticality reached under these conditions can have negative or positive feedback, as explained in the text, depending on the geometry and material composition of the collection volume*

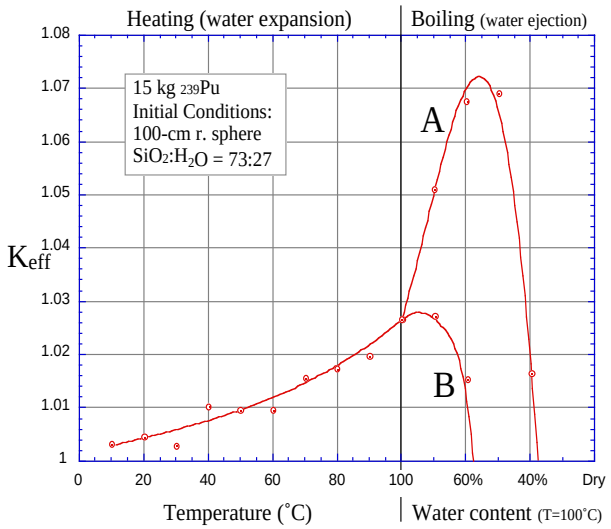

Figure 1D. *The positive feedback excursion for case D in Fig.1. As the criticality curve is reached for the 100-cm geometry, heat starts to be generated at Keff > 1. As a result of the heat generation, water warms up and expands. Rock will also expand, albeit to a much smaller degree. The overall effect of the reduced water density in the critical geometry is to increase the reactivity of the system, and therefore to drive the heat production to higher levels, until the boiling point of water is reached. Water boil-off can happen in many different ways. Two scenarios are considered in the figure: in curve A boiling is modeled as a uniform density decrease in the spherical volume (this leads to the highest reactivity level); in case B boiling is modeled by water being pushed out from the center, leaving empty space behind (this leads to the lowest reactivity increase). Realistic boiling scenarios are highly dependent on local parameters and would fall between these two extremes.*

the system. As the system dries, it reaches autocatalytic criticality. For the 200-cm radius system (large systems) this type of supercriticality can occur with almost any ratio of water to rock since the slope of the 200-cm criticality curve in Fig. 1 is always negative except for the very driest systems (water molar fraction less than 2%). Examples of this case are not restricted to underground phenomena. The Chernobyl disaster (ref.) is an example of 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 was mainly a poison. Malfunction of the control system led to "drying out" by uncontrollable boiling and the system became autocatalytic and destroyed itself.

Case F - Small volume systems (positive feedback)

The criticality curve for a 30-cm radius geometry is interesting primarily for the small amount of material which could lead to autocatalytic criticality. For the point F at a SiO_2 mole fraction of 0.4, the system becomes autocatalytic upon reaching the 30-cm curve in Fig. 1, at a plutonium mass of less than 2 kg. Nearly the same mass of plutonium on the other hand will have a negative feedback at criticality for a SiO_2 molar fraction of 0.5. At a SiO_2 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 and criticality is reached for 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 25 cm, the slope of the criticality curve (not shown) is always positive and the feedback always negative regardless of the SiO_2-to-water molar ratio.

Case A (extended) - Dispersion of TFM in dry rock

Starting with the configuration previously examined as case A, plutonium can be dispersed in the surrounding rock matrix by repeated sequences of water ingress, negative-feedback criticality and water-steam expulsions (see Fig.2). In the presence of substantial concentrations of plutonium, as a consequence of plutonium dispersion, the system can become repeatedly critical with ever less water present. If sufficient ^{239}Pu is available in a particular emplacement, the dispersion of plutonium to larger radii can take the system to dry criticality with positive feedback at the location of emplacement. The situation is further illustrated in Fig. 3, 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 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 K or with a radius greater than 200 cm (point J). 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 ordi-

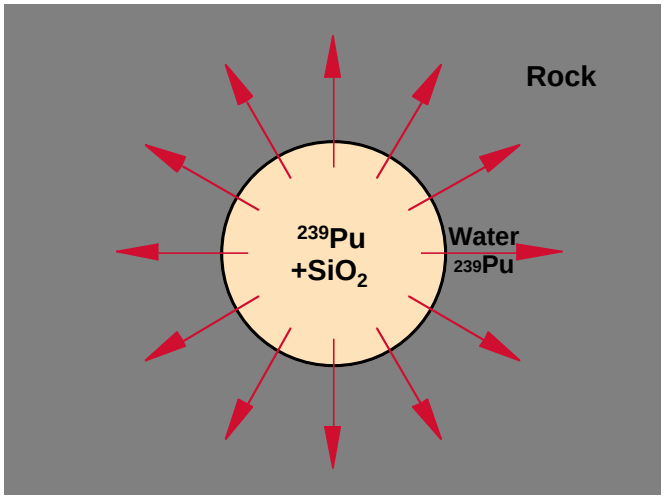

Figure 2. *Plutonium can be dispersed in the surrounding rock matrix by repeated sequences of water ingress, negative-feedback criticality and water-steam expulsions. If large amounts of plutonium are present in the emplacement, the dispersion of the plutonium in the rock could lead to better moderated geometries and therefore to criticality in the presence of smaller water amounts and eventually in dry situations. The criticality can then be self-enhancing or autocatalytic, if a mechanism for rapid plutonium dispersion in the rock can be triggered, such as plutonium vaporization.*

nate 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 K 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 3%. 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 taken by the repeated excursions down to point K where it could become dry autocatalytic.

Gradual dispersion and the associated incremental increase in k_{eff} would lead to a slow approach to criticality. However the plutonium could be dispersed suddenly by natural mechanisms such as volcanic action, earthquakes, or more modest geologic shifts, and by man-made events, such as well drilling, mineral exploration, or attempts at recovery of the buried material.

Another way of describing the situation illustrated in Fig. 3 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.

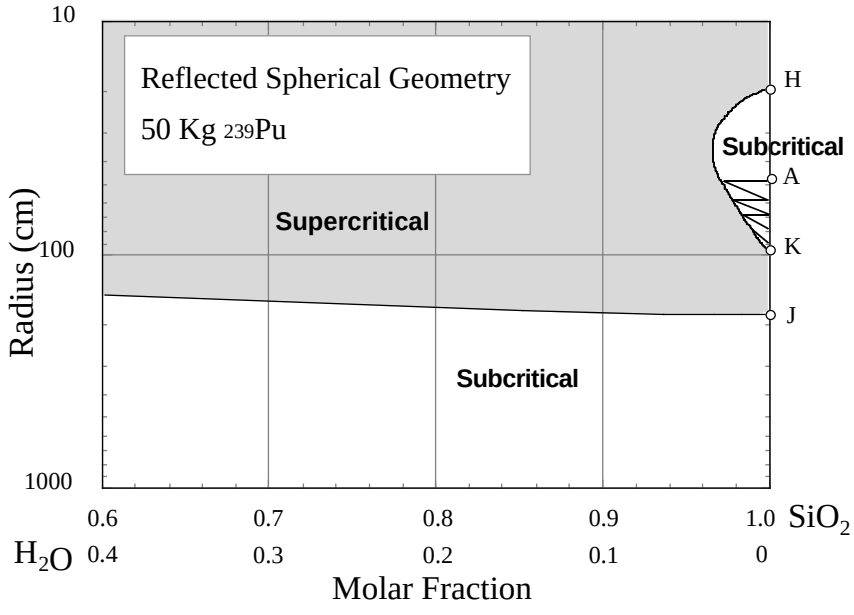

Figure 3. Criticality conditions for 50 kg of ^{239}Pu in a sphere surrounded by SiO_2 reflector. *The ordinate is the radius of the volume containing the plutonium. The abscissa is the molar fraction of water and SiO_2 in the sphere. The shaded region identifies regions of supercriticality. The other regions are the subcritical regions and represent those subcritical mixtures which could be placed underground.*

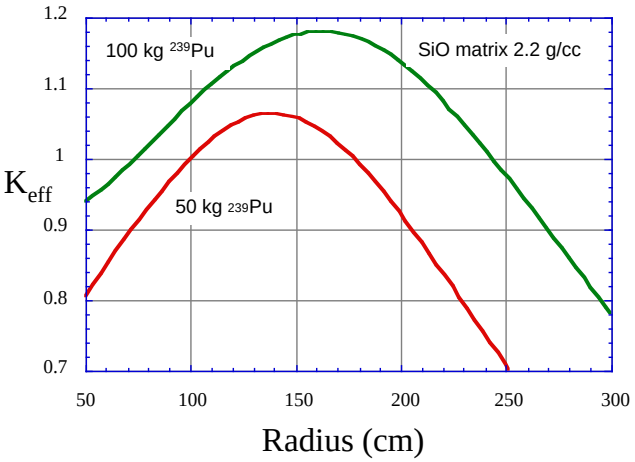

Figure 4. The change in k_{eff} with dispersion. *The situation shown in Fig. 3 is presented in more detail here where curves of k_{eff} vs radius of dispersion in spherical geometry are given for masses of 50 and 100 kg of ^{239}Pu in SiO_2 of density 2.2 g/cm^3. It is assumed that the plutonium loading 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.*

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 disperse further, 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 other factors don't act to cause a more benign event.

Estimates of Explosive Yield

The usually negative feedback mechanisms present in nuclear reactors (fuel temperature coefficient, moderator temperature coefficient) are relatively small or even positive (in the case of the ^{239}Pu fuel temperature coefficient), so that the previously described situations might indeed "run away" under the impact of their much larger positive reactivity feedbacks, reaching high levels of criticality and releasing 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.

Once a system has become prompt critical, the fission energy yield will increase approximately exponentially until the en-

ergy 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. 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 completes a phase change as the pressure nears 30 GPa (0.3 megabars). The phase change is associated with an increase in density by a factor of about two[6]. 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 $1.3x10^{12}$ 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 comparable even though the temperature is much lower in the rock. The conditions under which the system reaches subcriticality have been confirmed by Monte Carlo calculations[7] to be close to those estimated in Appendix B analytically and therefore the nuclear yields estimated here should be valid within a factor of two.

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 very small compared to the leakage from the large extended surfaces. For this situation, as is shown in Appendix B, the final yield per kilogram of TFM can be substantially higher than for the sphere. In addition, the leakage does not increase as the density decreases. Therefore only the temperature rise and the associated neutron spectrum hardening 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 mainly the difference in the mass of rock participating. For the slab the same amount of fissile material would be distributed in a rock mass that is much larger than for the 100-cm sphere.

Real plutonium, real water, real rock and the real world

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 is very efficient for dry systems, where neutrons lose only 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.

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 would significantly lessen its negative effects on the plutonium reactivity[7]. The reason is that the fraction of energy loss by a neutron per collision with hydrogen is large so that the neutron will rapidly be removed from the 1.05-eV ^{240}Pu resonance region and the probability of it escaping capture in the resonance in between collisions would be high.

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 is chemically different, has a much lower absorption cross section and 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 spontaneous supercriticality event[7]. After 25,000 years weapons-grade plutonium will be functionally equivalent to ^{239}Pu. In the same period of time the plutonium contained in LWR once-through 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.

The non thermally fissile isotope of plutonium ^{242}Pu, on the other hand, has a much longer half-life, and could play a role in keeping plutonium safely subcritical at all times[7], but its concentration is significant only in highly recycled or deep-burn plutonium fuels.

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[7]. Considering the composition

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
Mg0	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)

of Nevada tuff (Topopah Spring tuff), the overall thermal cross-section for neutron absorption is about 50% larger than for pure SiO_2, which indicates that 50% larger quantities of TFM would be needed for the criticality curves near the *right side* of figure 1 when SiO_2 is replaced by the Nevada tuff[7].

Pure H_2O was also used for water. Here instead, the effect of various poisons dissolved in the water, most notably chlorine, would be to increase the TFM concentration for the critical curves of Fig.1 near the *left side* of the graph[7].

Finally, the use of the homogeneous spherical geometry allows description by the simple specification of one parameter and is amenable to analytical treatment. While more realistic models might be necessary to arrive at more precise estimates, none of the simplifications assumed affects the qualitative content and conclusions of the foregoing discussion[7].

Application to specific TFM 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[3] would involve the emplacement of plutonium 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 planar 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 larger specific yields than possible for smaller geometries. Migration could also reposition the TFM into autocatalytic overmoderated concentrations.

Destruction by underground nuclear detonation

A Russian group[8] 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 dispersal*, the autocatalytic behavior could start, and the yield from the event might therefore 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 of once-through LWR spent fuel will approximate the isotopic composition of w-Pu and the criticality risk for stored material will become larger as more time passes (deep-burn spent fuel would not exhibit this behavior). Water infiltration could further reduce the effectiveness of the ^{240}Pu poison. Eventually the amount of commercial plutonium (c-Pu) turned into weapons-grade plutonium (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 and if the uranium is largely removed, 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 rearranged, spontaneous criticality with positive feedback is possible and 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 occur if the TFM disperses or 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 few-kilogram range behaving autocatalytically in some wet scenarios.

3. The dry autocatalytic feature of buried w-Pu, HEU, naval spent fuel, and spent research reactor fuel could give rise to sequential ignitions when this nuclear material is stored in an extended array of large individual emplacements (more than 50 kg per emplacement).

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

5. The maximum yields for the rock sphere containing 100 kg of TFM and for the infinite rock slab 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 for rock.

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 or after the ^{240}Pu decays away.

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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, table 3-3 on p.68 (March, 1994)

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Acknowledgments

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Appendix A. The approach to criticality

The intention here is only to show that prompt criticality is possible rather than to treat the subject comprehensively. We will describe a possible scenario for TFM encased in glass logs. 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 be thoroughly cracked already at emplacement as a consequence of the cooling process and has no structural integrity in itself, being held together by the canister[A1]. The water will penetrate throughout the vitrified mass and dissolution of the contents of the contained will start. Some solubilities at 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 large volume available for water and the mass will be critical upon water ingress.

The result of the heat production associated with criticality would be warming of the water and eventually (depending on the reactivity excess introduced by water ingress) steam generation and a possibly violent expulsion of water and steam from the vitrified mass, especially for systems that do not contain significant fractions of resonance absorbers such as ^{238}U. This negative-feedback criticality would be repeated periodically as water reentered the system. The consequences are almost certainly further cracking of the vitrified material perhaps eventually converting it to powder, cracking and 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 by the periodic steam bursts. At this point, different scenarios could follow. The plutonium might be carried with the water to the bottom of the emplacement cavity or elsewhere, possibly leading to the wet criticality scenarios described for cases C, D, E, F in Fig. 1. If, on the other hand, enough plutonium is present at the emplacement site (over 50 kg) and it disperses in the surrounding cracks and rubble, "dry" criticality could ensue (case A in Fig. 1).

"Dry" (undermoderated) criticality

We call this class of critical excursions *"dry criticality"* because the plutonium-rock volume contains no free water, although water can still be locked into the rock, and the feedback mechanism is the movement of the plutonium.

A last steam burst might carry enough additional plutonium into the surrounding rock to establish undermoderated prompt supercriticality. Supercriticality might also occur at any time as a result of the sudden collapse of the glass cylinder onto itself following erosion of a substantial portion of its volume. At this point the criticality excursion could proceed towards explosive energy release under its positive feedback mechanism (plutonium dispersal in the rock).

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)*

A possible fast mechanism for plutonium dispersion in the rock matrix could be vaporization due to the heat of fission. 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 (located on the rock surfaces) and its immediate vicinity. If the ratio of plutonium mass to the mass of rock is about 1/50, then 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. 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 moderator temperature coefficient (calculated to be $-1 \times 10^{-5}/^{0}C$ 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 entire plutonium mass could vaporize after only a few-degree increase in rock temperature.

"Wet" (overmoderated) criticality

We call this class of critical excursions *"wet criticality"* because the plutonium-rock volume contains large amounts of free water, and the feedback mechanism is the expansion and ejection of the water.

The transition to high k_{eff} for the overmoderated wet systems (cases C, D, E, F in Fig. 1) is a simpler process, with the reduction of water concentration first by warming and then by conversion to steam. Three feedback mechanisms can be at work during this process: the spectral shift for ^{239}Pu neutron cross-section, the rock thermal expansion, and the water thermal expansion and eventually ejection by boiling. In the temperature region of interest ^{239}Pu has a mild positive temperature reactivity coefficient. For water-overmoderated systems, rock expansion will reduce the reactivity, while water expansion will increase it. Rock however expands a lot less than water and therefore the overall effect is of a positive temperature feedback through water warming and eventually conversion to steam. For small geometries, at some point, water expulsion will start reducing the neutron moderation so that the feedback becomes negative, and the transient ends before the system reaches the dry state. Substantial yields can be reached nevertheless, depending on the rate of escape of the water. If the system is large (sphere radius greater than 200 cm), sufficient neutron moderation can be achieved via the dry rock and a high level of supercriticality can be sustained into the dry state.

References

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Appendix B. Energy Release (Yield) Calculations

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 TFM 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 much larger than for above ground scenarios. To gain some understanding of the order of magnitude involved, we evaluate these yields in some idealized conditions. The exponential time constant for fission power generation P is referred to as α and the appropriate expression is

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

The quantity α is generally time dependent and is given by

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

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

$$\tau = 1/\Sigma_a v \quad (3)$$

where v is the thermal neutron velocity and Σ_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 α 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 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 completes a phase change as the pressure approaches about 30 GPa (0.3 megabars). The phase change is associated with an 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 much 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 in to simulate 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

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

Σ_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

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

where S is the source intensity in neutrons per second, D is the thermal neutron diffusion constant given by $1/3\Sigma_s$ and Σ_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 thermal neutron will travel before absorption. In terms of defined parameters[B1] it is

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

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

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

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 Σ_a and Σ_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

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

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 large dot 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 ρ_0 and a_0 are the initial density and radius. Substituting for a in 8 to turn it into a temperature-density relation-

ship gives

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

This curve of density vs. T (temperature) is shown in the left part of Fig. B1, 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 of density vs. T 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 1.2x10^{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 0.1x10^{12} joules. The total energy generated in the expansion phase by the time the system passes to subcriticality is therefore 1.3x10^{12} joules or about 0.3 kilotons.

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 ρ 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 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. The system grows until the capture in the rock is too large compared to the fission from the amount of fissionable material available to sustain criticality and the system becomes subcritical again. Since very little of the available TFM has been consumed during the excursion, the system might become critical again following rearrangement of the material, as described in the paper..

For the spherical geometry just explored, the energy generation was terminated by the loss of neutrons into 4π solid angle, by the increase in temperature and finally by rock accretion. Perhaps a more probable rearrangement of large quantities of emplaced thermally fissile material would be in a slab-like geometry of rock and water, such as might be found in strata or the water table. The corresponding reactor physics problem is the infinite slab. For such a system with thickness

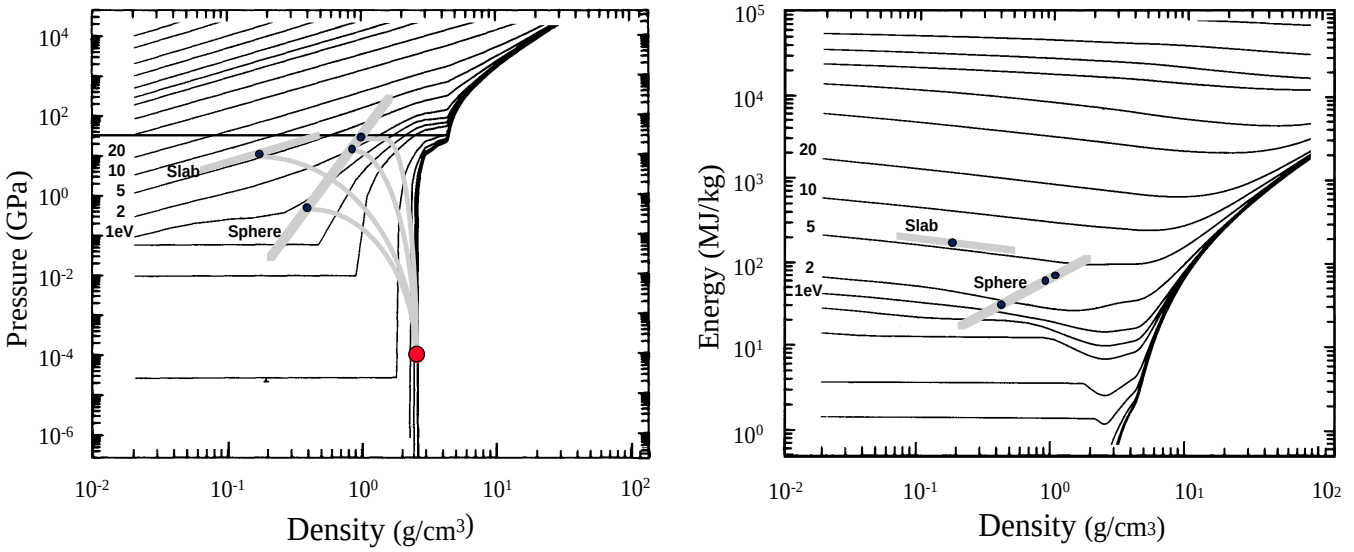

Figure B1. Equation of state for rock (Westerly Granite). *The figure on the left shows the pressure in gigapascals (GPa) as a function of the density of SiO_2. The different curves correspond to the different temperatures given in Kelvin on the right. 1 eV corresponds to 11600 degrees. The temperature in eV for a few of the curves is given just inside the ordinate scale. A phase change to twice the density is completed 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 large dot) and moves upwards 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 left 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.*

2a, the same analysis for probability of absorption gives[6] .

$$P(a) = 1 - \cosh(d/L)/\cosh[(a+d)/L] \quad (10)$$

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

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

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 $4\pi/3$, compared to 4π for the sphere. Once supercritical, the slab 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 the right-hand side of Fig. B1, the energy density is 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.

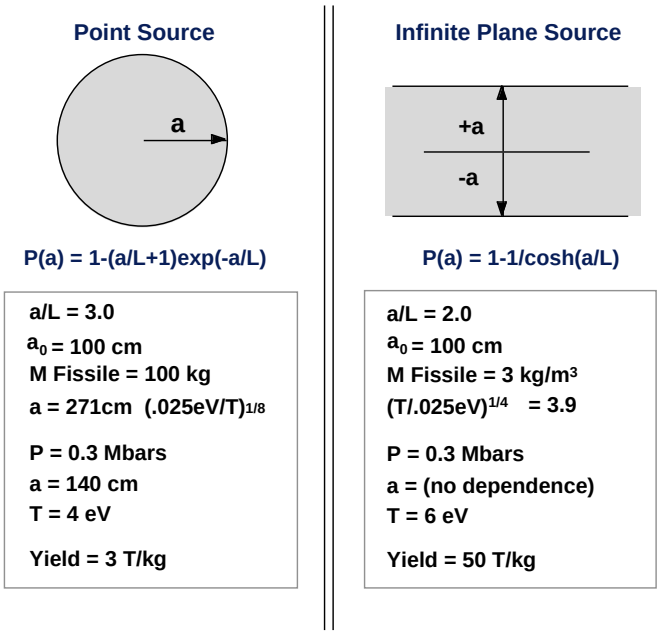

Figure B2. Underground yield estimation results. *The yield is estimated for spherical, cylindrical, and slab geometries. The amount of fissile material is: (1) 100 kg for a sphere of one meter radius, and (2) 3 kg per cubic meter for an infinite slab of two meter thickness. The yield reached at first subcriticality as a result of expansion is given here. The hot system then might accrete additional mass from the surrounding rock, cool, and become critical again. This accretion process could continue 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.*

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