

# Underground Supercriticality from Plutonium and Other Fissile Material

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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 results of such supercritical excursions could range from modest energy releases to the several hundred gigajoules range from a single event. Without water, 50–100 kg of fissile material is required to reach autocatalytic criticality. Amounts as small as 2 kg 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 materials (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 also contains a high concentration of U-235, spent fuel from research reactors containing HEU, spent fuel from commercial reactors containing plutonium (c-Pu) and other transuranic elements such as neptunium.

Recent studies<sup>1</sup> 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.<sup>2</sup> The purpose of this report is to show that

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underground storage, as presently recommended, could lead to underground supercriticality and the uncontrolled dispersal of the TFM, with significant nuclear energy release and possibly nuclear explosions in the several hundred gigajoule range.

The weapons plutonium portion of the TFM is perhaps of greatest current concern and for this reason Pu-239 is used for the most part in this paper to illustrate the criticality risks of underground TFM. In order to lower the costs of preparation and for actual emplacement of TFM, and to make the underground repository site small, the TFM would be stored in concentrated but still safely subcritical amounts. For the option of vitrification of the plutonium followed by storage in deep boreholes, a National Academy of Sciences study<sup>3</sup> considers weight concentrations up to 10 percent; a 60-cm diameter, three-meters long borosilicate log at 5 percent concentration would contain about 90 kg of plutonium. A single log of this material would be substantially subcritical. MOX spent fuel assemblies for w-Pu destruction would contain 18 kg each<sup>3</sup> and several might be stored together.

Storage subcriticality would be enhanced by the inclusion of neutron poisons, but even without poisons, w-Pu in these amounts and in these configurations would be subcritical. The reason is that the neutrons cannot moderate before leaving the w-Pu and cannot find the w-Pu after moderation. However, once containment has been compromised and the TFM is no longer prevented from dispersing 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. A system with positive feedback would have the feature that power generation would lead to changes in the system which would cause the rate of power generation to rise even higher. Such a system also is sometimes referred to as autocatalytic.

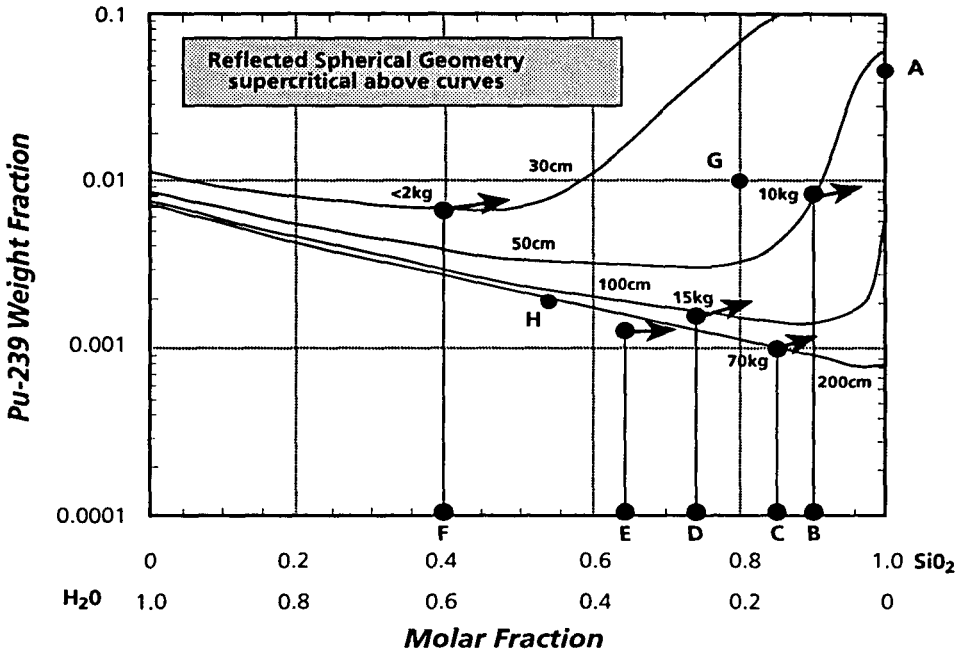
## FEEDBACK: POSITIVE AND NEGATIVE

Many factors influence criticality such as amounts of fissile material, water, other moderating material, poison, configuration, and resonance behavior. The effects of resonances can be significant if there are large amounts of resonance absorbers present, such as U-238 or Th-232, which is often not the case for TFM. Poisons in general, whether resonance absorbers or not, can be very important in reducing the criticality risks of underground stored TFM, but their physical properties such as solubility and boiling point will in general be different from those of the fissile material with which they must bind. In the view of the authors, poisons may not be a reliable means of preventing critical-

ity over the long term. The relative concentrations of fissile material, water, and other moderator such as rock are the most important factors and they can be analyzed for positive or negative criticality feedback using figure 1.

Figure 1 shows the criticality conditions for spherical volumes of different radii with mixtures of Pu-239, water and  $\text{SiO}_2$  surrounded by a  $\text{SiO}_2$  reflector. The calculations were done using the MCNP<sup>4</sup> code.  $\text{SiO}_2$  at a density of  $2.2 \text{ g/cm}^3$  approximates to a reasonable degree the nuclear properties of rock. The figure gives the mass fraction of plutonium on the ordinate and mole fractions of water and  $\text{SiO}_2$  on the abscissa. Therefore for point G in the figure, Pu makes up 1 percent of the sphere mass. The remainder of the material in the sphere in mole fractions is 20 percent water and 80 percent  $\text{SiO}_2$ . A system lying on the left ordinate has no  $\text{SiO}_2$  in it. A system on the right ordinate would have no water. The curves therefore show critical compositions as water is exchanged for  $\text{SiO}_2$  or vice versa for systems of different volume. 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 in supercritical configurations, 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 underground systems have a natural tendency to revert to subcriticality by means of negative feedback. The main feedback mechanisms illustrated in the figure is water ejection and we make the point that this feedback can be positive as well as negative. If criticality is reached, energy is generated and the water starts to be driven away and the system may move into subcriticality or supercriticality. Since in a practical situation the water usually would not be replaced by  $\text{SiO}_2$ , the figure is not useful for quantitatively predicting the course of supercriticality. With regard to water ejection (indicated in figure 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 still higher criticality (positive feedback) and 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.* Water is not replaced by void upon expulsion in the figure, therefore the curves should be viewed as indicative of initial condition behavior when criticality is reached. Likewise, the arrows only qualitatively indicate the trend of the criticality excursion. While the figure is 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.



**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 Pu-239, water and SiO<sub>2</sub> reflected by SiO<sub>2</sub>. The ordinate is the weight fraction of Pu-239 in the sphere. A fraction of 1.0 means the system is pure plutonium. The abscissa is the molar fraction of water or SiO<sub>2</sub> at densities 1 and 2.2 respectively in the rest of the system. Water can be contained in the rock pores of solid rock, up to the porosity fraction characteristic of the rock under consideration (4-15 percent in volume) or in the cracks and voids surrounding highly fractured material or loose gravel. Dry configurations lie on the right hand ordinate axis. Systems with compositions lying above the criticality line for a given radius are supercritical; those lying below the line are subcritical. Systems which reach criticality where the slope of these curves are negative have positive feedback. Systems which reach criticality where the slope of the curve is positive have negative feedback. The figure only provides indications as to the behavior of the systems upon becoming critical. The arrows qualitatively give the general direction in which the systems will evolve when they reach criticality.

**Case A - Water Ingress in TFM + Rock (Negative Feedback)**

Nearly all TFM would be emplaced as dry material and therefore will initially lie along the right (SiO<sub>2</sub>) ordinate. If we consider the 50-cm spherical geometry, we see that the mass fraction of Pu-239 mixed with SiO<sub>2</sub> must be less than 0.065, for the system to be subcritical. The case A has a Pu-239 loading (weight fraction) of 0.044, about 50 kg, smaller than the dry critical mass for that geometry. If water enters this system (see figure 1A), the composition of the system in figure 1 will move to the left, until wet criticality is reached at a

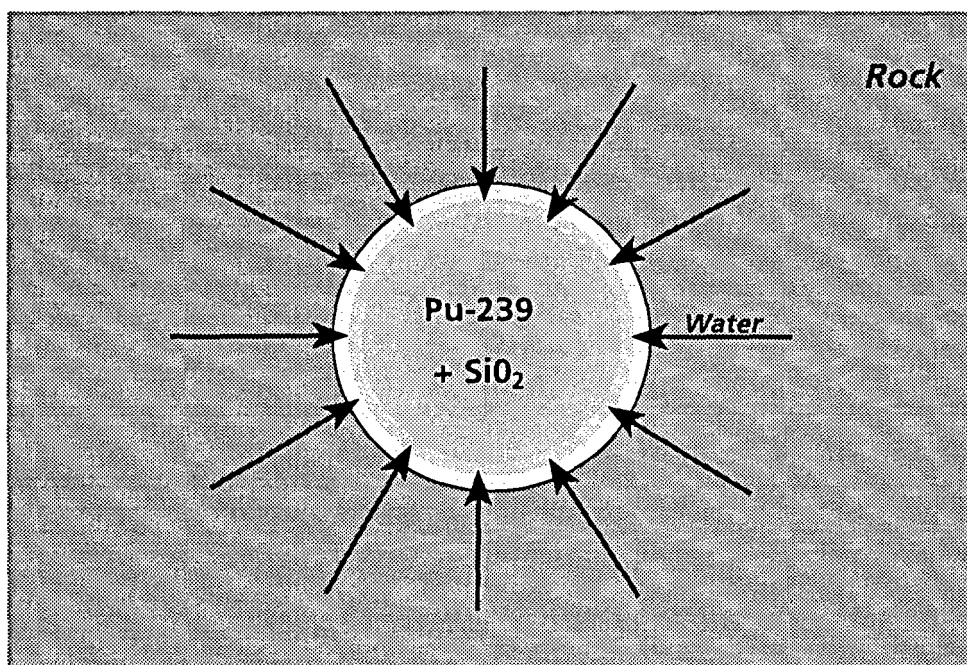
water mole fraction of about 2 percent (0.6 percent mass fraction). The system will generate fission energy at some level until the water that produced the critical state is converted to steam, is otherwise driven out of the system, or leaves of its own accord. If water elimination is complete, the system returns to its starting point on the right ordinate axis. The next incursion of water would 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 might be similar to that of the Oklo natural reactor<sup>5</sup> in Gabon, an underground critical system which operated for about one million years.

### **Case B - TFM Migration to Wet Rock (Negative Feedback)**

There are conditions in which a mixture of water and rock of a given volume can give rise to smaller critical masses than either water or rock alone. This is the case for example of the 50-cm radius system shown in figure 1. There we find the critical mass of Pu-239 with water alone is 4.6 kg; with rock only the amount is 80 kg; for 30 percent 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 percent mole fraction of water ( 3.2 percent 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 figure 1B/C). When a plutonium mass fraction of 0.009 had accumulated (10 kg) in the 50-cm radius, the configuration would 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 Pu-239 occurred, the system could move into and out of criticality indefinitely following repeated incursions of water.

### **Case C - TFM Migration to Wet Rock (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, and the different molecular densities for these two materials, and the fact that neutron energy loss per collision is not important for large volume systems unless strong resonance absorbers are present. If some TFM from the original implantation sites should migrate to a 200-cm radius volume where the mole fraction of water was 15 percent, the system would start at point C and move vertically until the system reached criticality at a Pu-239 mass concentration of 0.001 (70 kg of Pu-239 in the 2-m radius, 70-ton sphere).

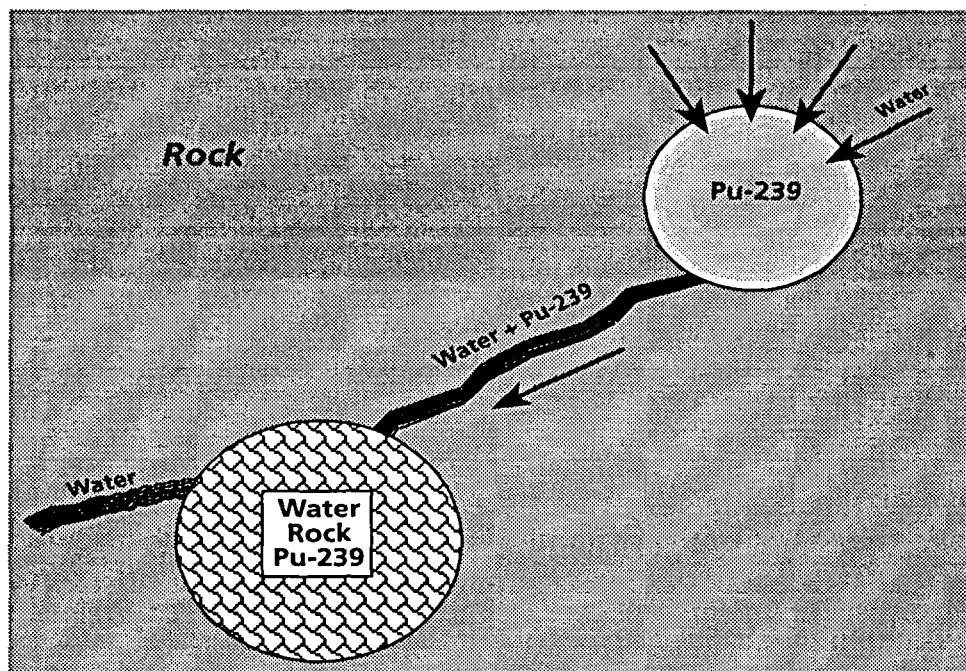


**Figure 1A: Water ingress in Pu-239+SiO<sub>2</sub> mixture.** Criticality reached under these conditions always has negative feedback. Water can however boil locally and disperse some of the plutonium into the surrounding rock on its way out. This dispersal mechanism could eventually lead to dry supercriticality.

When this system becomes critical and the heat begins to drive the water out, the system composition, as represented in figure 1, also moves to the right into the supercritical region, and in doing so the system drives itself to a *higher* criticality and a larger energy release.

#### **Case D - TFM Migration to Wet Rock (Positive Feedback)**

The accumulation of plutonium in a sphere of 100-cm radius with water present in the 27 percent mole fraction, started at point D in figure 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 the 100-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 percent of the original concentration. Starting with the initial, barely critical, configuration in figure 1, the reactivity evolution of this system, consistently evaluated for water evaporating and eventually leaving the volume, is illustrated in figure 1D, which shows the

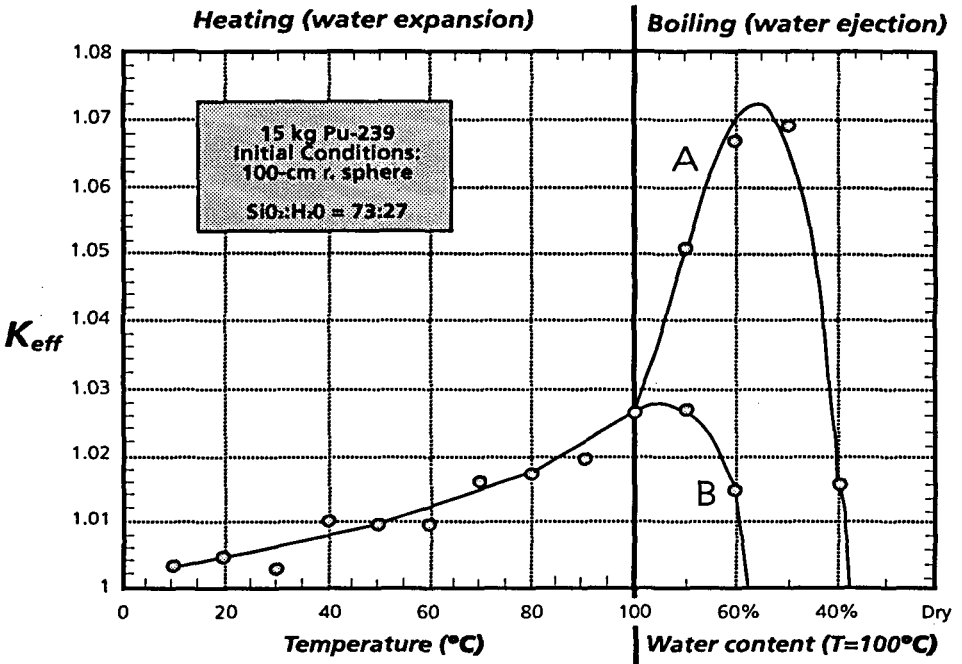


**Figure 1B/C: Plutonium migration to wet rock systems.** Water comes into the plutonium emplacement and carries off some of it in particle form. Eventually plutonium from many separate emplacements could collect in a wet rock volume. Criticality can be reached in this volume after enough plutonium is 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.

positive feedback excursion as water warms up and expands and ultimately boils off leaving the system dry. A  $k_{eff} = 1$  system is critical. Larger fractions of excess water with respect to the optimum moderation case (bottom of the curve) would cause larger reactivity increases upon expulsion.

### Case E - Drying-Out of TFM Deposits (Positive Feedback)

This case illustrates the deposition of plutonium in wet media to overmoderated subcritical concentrations followed by drying of the system. As the system dries, it reaches criticality. For the 200-cm radius system (large systems) this type of supercriticality can occur with positive feedback at almost any ratio of water to rock, since the slope of the criticality curve is always negative except for the very driest systems (water molar fraction less than 2percent). The Chernobyl disaster is an example of Case E condition. This reactor was well moderated by graphite. Water was present for heat removal and its moderator



**Figure 1D: Positive feedback excursion for (case D in figure 1).** As the criticality curve is reached for the 100-cm geometry, heat starts to be generated at  $k_{eff} > 1$ . 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 an 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.

function was not important. From the neutron economy perspective water was mainly a poison. Malfunction of the control system led to “drying out” by 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 point F at a SiO<sub>2</sub> mole fraction of 0.4, the system becomes autocatalytic for 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<sub>2</sub> molar frac-

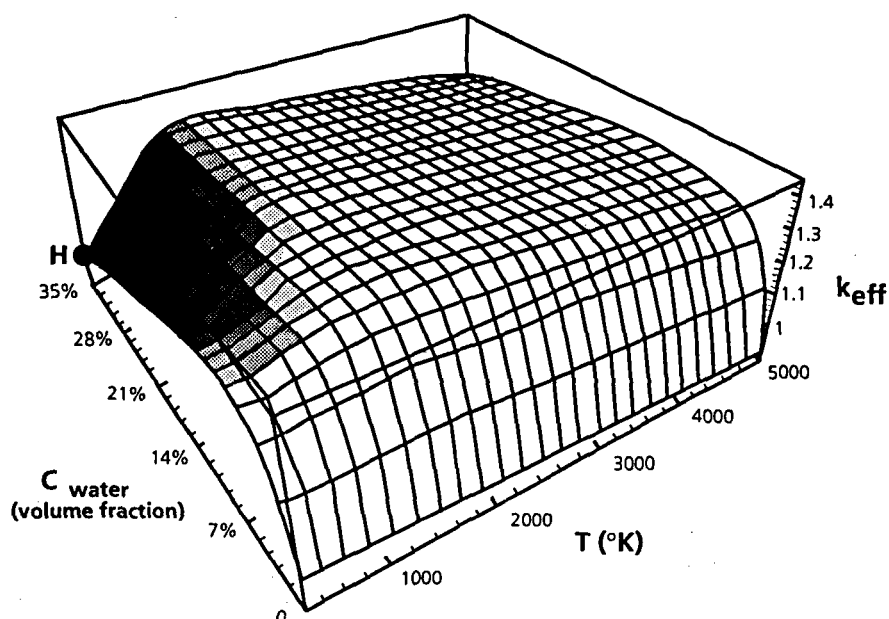


tion of 0.5. At a  $\text{SiO}_2$  mole fraction of 0.2, where the mass of water and  $\text{SiO}_2$  in the sphere are about equal, the system is distinctly autocatalytic with respect to water ejection and criticality is reached for even smaller amounts of plutonium. As the radius under consideration 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  $\text{SiO}_2$ -to- $\text{H}_2\text{O}$  ratio.

### TEMPERATURE COEFFICIENT OF REACTIVITY FOR Pu-239

Pu-239 has a large resonance centered at 0.3 eV ( $\sim 3000$  K). Since the neutron spectrum is not completely thermal, the effects of this resonance are apparent even at room temperature. Especially important is the effect of the resonance on the reactivity of wet, overmoderated systems. The spectrum averaged fission cross section for Pu-239 doubles as the temperature approaches the resonance energy and many neutrons that would otherwise have leaked out or been absorbed parasitically in water are absorbed by Pu-239 leading to more fission and greater reactivity as the temperature increases and the neutron spectrum becomes harder. The effect is especially dramatic for large fractions of water. The strong positive temperature feedback associated with the Pu-239 resonance carries criticality past the delayed critical stage (driven by delayed neutrons and characterized by slow transients) into the prompt critical stage. Figure 2 shows the reactivity for a 2-m diameter sphere containing Pu-239,  $\text{SiO}_2$ , and water (initial condition is point H in figure 1) as a function of temperature for various amounts of water present.

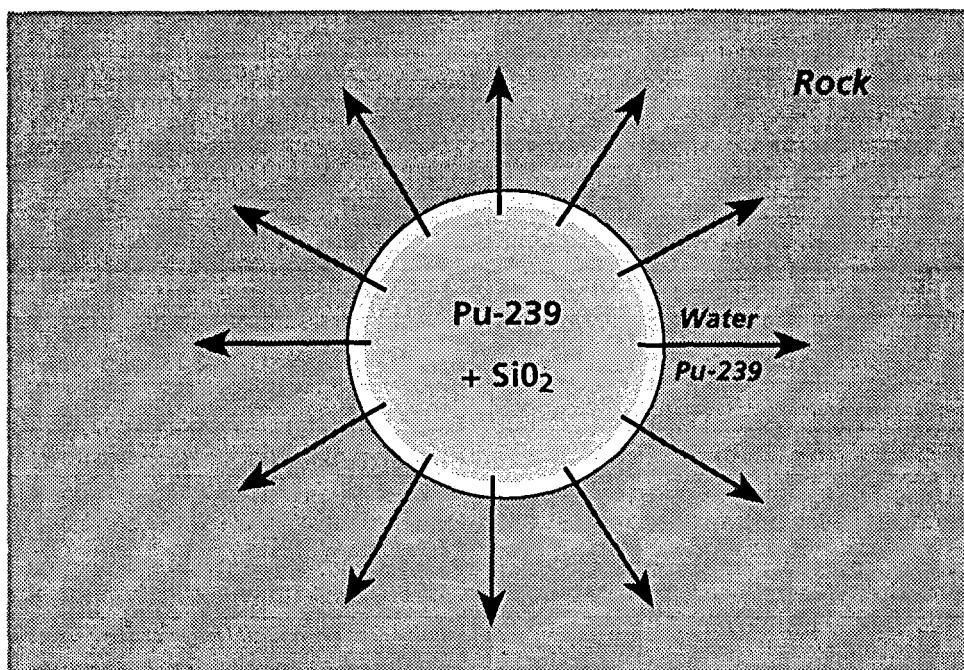
If criticality were reached at point H, the system would increase in temperature where the upward slope is steepest. However, since the thermal expansion of the water is greater than that for the rock, the water content would also be reduced until the boiling point of water is reached. At this point the water would start to be ejected faster, with the ejection rate ultimately limited by the rock porosity and heat transfer. For  $k_{eff}$  in the range of 1.2, the energy generation rate would be very fast, and the temperature would quickly reach the boiling point of  $\text{SiO}_2$  (2500 K), before all the water is ejected. From that point on, the water/ $\text{SiO}_2$  ratio would remain locked as the temperature rises and the system would remain highly supercritical to temperatures well above 5000 K (0.45 eV).



**Figure 2: Plutonium reactivity changes.** The reactivity ( $k_{eff}$ ) for a 2-m radius sphere of Pu-239 in  $\text{SiO}_2$  and water is plotted as a function of water content and temperature. There are 120 kg of Pu-239 in the sphere, and water is replaced by void along the water content axis, so that the  $k_{eff}$  values can be used in dynamic calculations of reactivity excursions. Point H corresponds to the critical composition H in figure 1. The reactivity changes associated with the prominent 0.3eV Pu-239 fission resonance are especially noticeable in highly overmoderated systems, where water constitutes a considerable poison. For dry systems in which plutonium and rock do not move with respect to one another, the resonance has little effect and the temperature coefficient becomes slightly negative. The calculations were performed using the MCNP4 code.

## DISPERSION OF TFM IN DRY ROCK

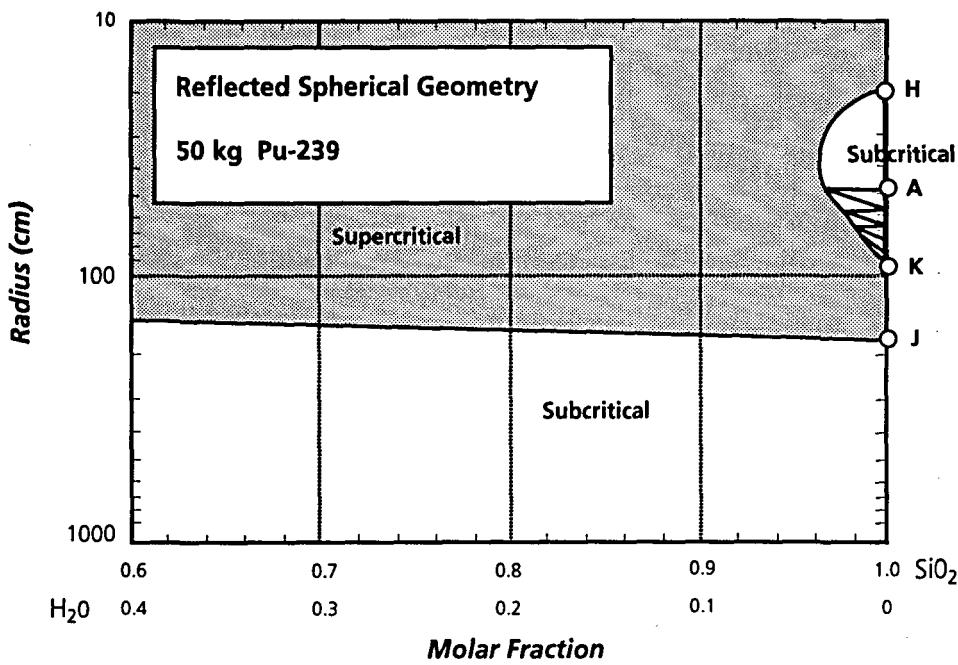
Starting with the previously examined configuration A in figure 1, plutonium can be dispersed through the rock matrix by repeated sequences of water ingress, negative-feedback criticality and water-steam expulsion (see figure 3). As a consequence of plutonium dispersion during these cycles, the system can become repeatedly critical with ever less water present. If sufficient Pu-239 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 figure 4, where the radius of the spherical critical mass for 50 kg of Pu-239 is shown for various molar fractions of water and rock represented by  $\text{SiO}_2$ , with water at density 1.0 and  $\text{SiO}_2$  at density 2.2. The shaded area of the curve is the region of supercriticality for which emplacement is impossible. The unshaded area rep-



**Figure 3: Plutonium dispersion.** Plutonium can be dispersed in the surrounding rock matrix by repeated ingress of water followed by 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.

resents subcritical configurations for which emplacement is possible. Most emplacements would likely be as dry material (along the right ordinate). In that case, the radius of containment for 50 kg of Pu-239 is between 20 and 100 cm (points H and K) or with a radius greater than 200 cm (point J). A horizontal movement in the figure represents water ingress or ejection, a vertical movement represents TFM dispersal or concentration to different radii. As in the case of figure 1, evaluation of dynamic behavior from figure 4 following the attainment of criticality is necessarily qualitative since water usually cannot be exchanged for rock or vice-versa in practical situations.

The farther into the supercriticality region, the greater is  $k_{eff}$ . Therefore the region on the right ordinate between points J and K represents a region of supercriticality with maximum value halfway in between. If a system reaches point K and conditions are such that the plutonium is dispersed by fission



**Figure 4: Criticality conditions for 50 kg of Pu-239 in a sphere surrounded by  $\text{SiO}_2$  reflector.** The ordinate is the radius of the volume containing the plutonium. The abscissa is the molar fraction of water and  $\text{SiO}_2$  in the sphere at the densities of 1.0 and 2.2 respectively. The shaded region identifies regions of supercriticality. The other regions are the subcritical regions and represent those subcritical mixtures which could be placed underground. As was the case with figure 1, this figure only provides qualitative indications on the behavior of the systems after reaching criticality.

heat through the rock, the system is dry autocatalytic. It is worth noting that any dry system initially in a configuration between H and K, when exposed to water could work its way down to K (the zigzag line in the "ear" in figure 4). An initially dry system is shown there with a radius of 50 cm (point A). The incursion of water would move the system to the left until it became critical (3 percent water). Upon the generation of fission heat, the system would expel 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 dispersion radius of the plutonium would grow. Then the return to dryness would not follow a horizontal line, but a line with a slight downward slope. If the plutonium were not carried away (perhaps to one of the conditions illustrated in figure 1), and remained in the immediate surroundings, the system would eventually be taken down by repeated excursions to point K where it could become dry autocatalytic.

The process could be accelerated by natural events such as volcanic action, earthquakes or more modest geologic shifts, and by man-made intervention such as well drilling, mineral exploration, or attempts at recovery of the buried material.

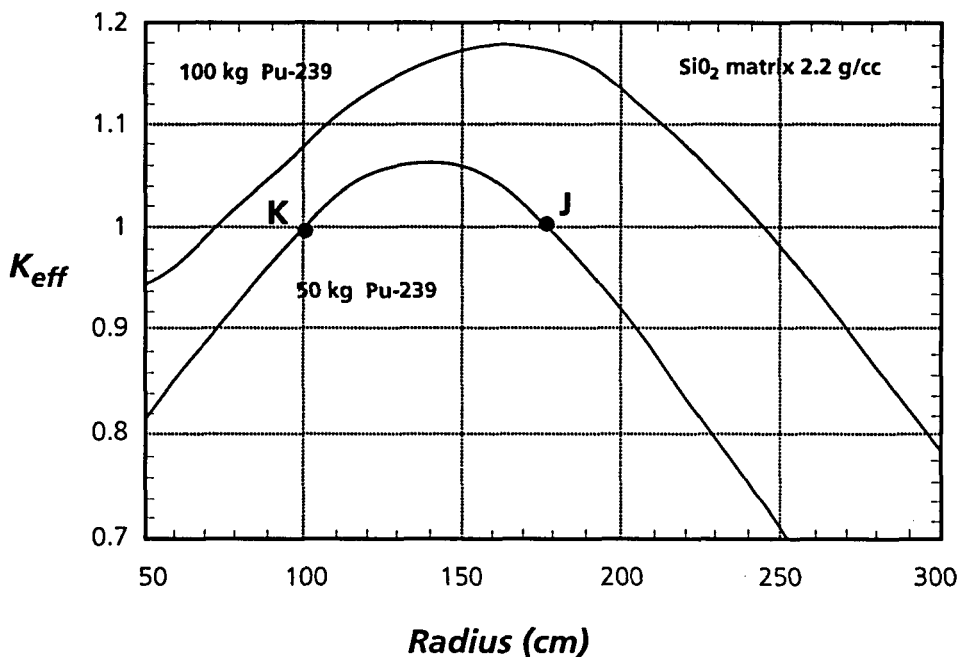
Another way of describing the effects of TFM dispersal is that after dispersion in the surrounding  $\text{SiO}_2$ , the neutrons can reach more nearly thermal energies, for which reaction cross sections are higher than in the original undermoderated system, and the same mass can become supercritical. The effect is illustrated in figure 5 where the value for  $k_{\text{eff}}$  is given for Pu-239 in  $\text{SiO}_2$  as a function of radius for 50 and 100 kg of Pu-239. The curves describe highly idealized cases, with spatially uniform Pu-239 concentrations, but they illustrate the point that Pu-239 dispersal can increase its reactivity. For the 50 kg case,  $k_{\text{eff}}$  has a minimum at a radius of about 50 cm and a maximum at 150 cm. The curve could represent a system that passes through criticality and starts generating significant energy, possibly driven through the delayed critical phase by a positive temperature coefficient.

Once in the dry critical state, the Pu-239 is expected to disperse further, possibly as a result of vaporization. Nearly all of the heat of fission would be deposited within a few microns from the place of origin by the fission fragments and therefore Pu-239 kernels would be expected to preferentially vaporize, leaving  $\text{SiO}_2$  behind. Because the preferential dispersion of Pu-239 in  $\text{SiO}_2$  is characterized by positive feedback, the configuration could drive itself to an accelerated energy release.

## 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 and would have no structural integrity in itself, being held together by the canister<sup>6</sup>. The water will penetrate throughout the vitrified mass and dissolution of the contents of the container will start. Some solubilities at 300 K are listed in table 1.

The  $\text{B}_2\text{O}_3$  in the borosilicate glass is 100 times more soluble in water than the  $\text{SiO}_2$ , and  $\text{SiO}_2$  and  $\text{B}_2\text{O}_3$  are several orders of magnitude more soluble than  $\text{PuO}_2$ . The  $\text{B}_2\text{O}_3$ , which makes up about 15 percent of the mass of the glass will be leached away much faster than the  $\text{SiO}_2$ . The heat from the plutonium will warm the water entering the vitrified mass bringing it to an unsaturated condition regardless of its initial concentration of silicates and



**Figure 5: The change in  $k_{eff}$  with dispersion in dry systems.** The situation shown in figure 4, along the K to J path, 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 Pu-239 in dry  $\text{SiO}_2$  of density  $2.2 \text{ g/cm}^3$ . It is assumed that the plutonium loading is uniform in the sphere and the temperature is taken to be  $25^\circ\text{C}$ . 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.

therefore allowing the log dissolution process to continue. After a significant portion of the  $\text{B}_2\text{O}_3$  (and  $\text{SiO}_2$ ) has been leached away, the vitrified mass might take on a spongy character with large volume available for water and the mass can become critical upon water ingress.

The result of the heat production associated with criticality would be warming of the water and eventually (depending on the rate of reactivity excess introduced by water ingress) perhaps steam generation and expulsion of water from the vitrified mass, especially for systems that do not contain significant fractions of resonance absorbers such as U-238. This negative-feedback criticality would be repeated over time as water reentered the system. The consequences would include further cracking of the vitrified material perhaps eventually converting it to sand, and a more rapid dispersion of the plutonium-bearing mass.

If enough plutonium is present at the emplacement site (over 50 kg) and it disperses in the surrounding cracks and rubble following the intermittent water induced criticality, "dry" criticality with positive feedback could ensue at the location of emplacement.

**Table 1:** Solubilities of relevant materials in water at 300 K.

Material	Solubility (Mol/liter)	Reference
PuO <sub>2</sub>	10 <sup>-7</sup>	Michaels <sup>7</sup> and Wilson <sup>8</sup>
UO <sub>2</sub>	10 <sup>-4.5</sup>	Michaels <sup>7</sup> and Wilson <sup>8</sup>
SiO <sub>2</sub>	~10 <sup>-3</sup>	Comparing solubility of UO <sub>2</sub> in water and silicate-saturated water
B <sub>2</sub> O <sub>3</sub>	10 <sup>-k0.8</sup>	CRC Handbook (1.0 grams/100 ml)

If sufficient care was taken in the emplacement strategy so that individual concentrations of TFM would not exceed the minimum mass capable of "dry" supercriticality, once the undissolved TFM oxide is freed by the disappearance of the boron and part of the glass, it might still be carried out of the vitrified mass by water as a colloidal suspension or in solution and initiate any of the overmoderated "wet" scenarios already described.

While the average rainfall at a repository site could be small, it would be likely that most of the water would move through the repository along major cracks of the rock following rains. There is ample evidence for this in the existence of springs and the frozen ice one sees on cracks of the face of deep cuts through rock. If much of the water flowed along particular channels in the rock, most of the emplaced waste might be almost dry and free of dissolution. But the waste inadvertently emplaced in such a stream might be compromised in much shorter times than designed for according to the average water percolation rate.

Colloidal and particulate transport<sup>10</sup> is the most likely mechanism by which plutonium (which as an oxide has very low solubility in water and decays faster than uranium) can be carried away from its position of emplacement into a different location. Overmoderated criticality is most likely in the near field, where enough water might be present to give rise to one of the supercritical configurations previously discussed. Short and continued periods of localized high water flows in the repository would enable this transport mechanism.

Pu-239 eventually decays to U-235 and large amounts of highly enriched uranium spent fuel might be present in a repository as well. Because of the much higher solubility of uranium oxide in water, in addition to colloidal transport, solution in water will be available to U-235 as a mechanism for leaving the position of original emplacement and collecting at a different loca-

tion. Solution transport would not be limited to the near field. Again water would be the prime mover in this transport mechanism, but now the flows need not be large and fast to cause the movement. Water containing uranium in solution would flow through the repository and possibly converge into a few focal points if impermeable layers were present below the repository rock. The uranium would stay in solution in the water until enough reducing rock is encountered that could produce precipitation and accretion of the uranium as pitchblende<sup>10</sup>. Such rocks, containing for instance significant amounts of iron oxides, are not infrequent in many geological settings.

The process rates and probability for a sufficient amount of U-235 to deposit in an autocatalytic critical configuration would be dependent on the amounts of TFM originally available and on the geochemistry of the site. Given however the very large amounts of highly enriched TFM that are planned for underground storage, the relatively high solubility of uranium in water and the very long time required for the decay of the U-235 into non fissionable isotopes ( $7 \times 10^8$  years half-life), the possibility of such events might not be dismissed as an invalid concern.

## ENERGY RELEASE

The usually negative feedback mechanisms present in nuclear reactors (fuel temperature coefficient, moderator temperature coefficient, expansion reactivity coefficient) are relatively small or not available in the case of underground stored TFM once critical conditions are reached, so that the previously described situations might indeed "run away" under the impact of the large positive reactivity feedbacks already discussed, reaching high levels of criticality and releasing significant energy before the excursions are terminated.

Some qualitative arguments could shed light on the magnitude of the actual energy release. Once the system has become prompt critical, the fission energy produced 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 with poor absorbing properties, and the disassembly mechanism is hampered by the presence of the rock, so that the final energy release to be expected could be much larger than for above ground scenarios.



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. Its behavior is crucial to the eventual magnitude of the yield<sup>9</sup>. The generation of explosive yields in fact will be limited by the degree of supercriticality reached and by the compression strength and inertia of the surrounding rock.

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  $\alpha$  and the appropriate expression is

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

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

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

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

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

where  $v$  is the thermal neutron velocity and  $\Sigma_a$  is the macroscopic absorption cross section for the medium. The value of  $\tau$  is in the range 0.1-1 milliseconds. For  $k_{eff} = 1.1$ , which appears to be a typical value for these supercritical systems, the value for  $\alpha$  in inverse microseconds is  $\alpha = 1/1000$  to  $1/100$  so that the yield will increase by the factor  $e$  in 1-10 millisecond. The time for sound to move one meter in  $\text{SiO}_2$  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 or both. The expansion of the system is limited by the confining pressures. Three pressures may be relevant in underground scenarios: the lithostatic pressure, given by the material overburden, the crushing pressure or compressive strength, given by the yield strength of the rock, and the phase-change pressure at which rock contracts as a result of a phase change. A yield calculation based on the lithostatic pressure is appropriate for producing a spherical volume in a medium which can sustain no shearing force such as a liquid, but it would not be applicable to rock which will withstand a shearing force. It is probably impossible to produce a sphere-like volume in a few milliseconds without doing work against the structural strength of the rock. The compres-

sive strength of the rock or the crushing pressure is therefore the determining parameter in yield calculations, along with inertial and frictional effects. The excursion is probably too slow to allow the buildup of pressure to the level required for phase change in rock. We illustrate this view by considering liquid, sand, and rock successively.

### **Liquid**

In a liquid for which a shearing force is absent, the lithostatic pressure is appropriate for calculating work done for a slow release of energy. For a faster release of energy, the inertia of the mass moved and the energy required for accelerating this mass can require a pressure which can be substantially larger than the lithostatic pressure.

### **Sand**

Sand cannot sustain significant shear stress and so it is much like water. However in this case there will be frictional forces as the sand particles grind against each other during the volume expansion. This pressure will depend on the rate of expansion. Therefore for sand the relevant pressure will have lithostatic, inertial, and frictional components.

### **Rock**

Spherical-like volumes cannot be created in welded rock without first crushing the rock. Therefore for rock one must deal with the pressures associated with crushing, friction, and inertia in addition to the lithostatic pressure. Since the crushing force is much larger than the lithostatic force, the lithostatic force can be neglected. The compressive strength for rock is defined as that pressure at which a cylindrical sample compressed axially and unconfined on its circular surface gives way. This pressure is about 50 MPa for tuff and about 150 MPa for granite. Of course the tuff rock in situ is in fact supported to some degree by the surrounding rock and in that case the compressional strength is considerably larger. If the lateral pressure equals the unconfined compressional strength, the compressional strength is increased by a factor of about five. Since the lateral pressure is at least as large as the unconfined compressional strength of 50 MPa, we use as the compressional strength 250 MPa for tuff.

To reach this pressure the rock must be vaporized and the temperature of the gas depends on the density. If we assume that the rock has expanded to a volume with a radius of two meters before the system becomes sub critical, the

density has decreased from 2.2 to 0.275. From the equation-of-state in LA-UR 4022A, we find a temperature of about 0.8 eV and an energy density in the gas of 25 megajoules per kilogram. The energy content of the 9000 kg of gas is about 60 tons of High-explosive equivalent.

It also takes energy to crush the rock and thereby to make space for the gas expansion. Assuming a 2-meter radius volume is to be yielded up by the surrounding rock and the rock is crushed at the compressive strength of 250 MPa, in response to an estimated 1.0 percent volume reduction in the crushing process, the actual crushed volume must be 100 times larger than the 2-meter volume. Its crushing radius is therefore 2 meters  $\times 100^{1/3} = 9.3$  meters. If a 1-meter high by 1-m<sup>2</sup> block of rock must be compressed by 1 percent (.01 meters) before it crushes, the energy required to crush the block would be 250 MPa  $\times 1 \text{ m}^2 \times 0.01 = 2.5 \text{ MJ}$ . The volume to be crushed contained in a 9.3-meter radius sphere would be 3300 m<sup>3</sup>. The work done in crushing the rock would be 2.5 MJ  $\times 3300 \text{ m}^3 = 2$  tons of HE equivalent. Note that if the rock crushed after yielding by only 0.3 percent rather than the 1 percent used above, the work done per m<sup>3</sup> would be lower by a factor of 3. However the volume required to be crushed would be 3 times larger so the work done in crushing the rock and creating the sphere is independent of the yield point for crushing of the rock.

The kinetic energy carried by the moving rock outside of the two meter radius depends on its effective mass. At the outer boundary of the rubble  $R = 9.3 \text{ m}$ , the rock is at rest. Therefore there is a velocity gradient with a cubic dependence through the rubble. If  $a$  is the radius of the gas ball surface,  $R$  is the outer radius of the rubble and  $v_0$  is the velocity at the surface of the gas ball, we take the velocity profile in the rubble to be:

$$v(r) = v_0 [1 - y]^3 \text{ where } y = (r - a) / (R - a) \quad (4)$$

The kinetic energy  $dE$  carried by a shell of thickness  $dr$  in the rubble is:

$$dE = (1/2) \rho 4\pi r^2 v(r) dr \quad (5)$$

This integrates to

$$KE = (1/2) \rho (4\pi/3) (R - a)^3 v_0^2 \times 0.0735 \quad (6)$$

The equation says that the problem is equivalent to the 0.0735 of mass in the rubble shell moving with the velocity  $v_0$  of the inner surface of the rubble. For

$R = 9.3$  meters as estimated above,  $\rho = 2.2$ ,  $a = 2$  meters, and  $v_o = 500$  m/s, the kinetic energy carried is about 15 tons. The total energy then is:

Crushing energy	2 tons
Kinetic energy	15 tons
Gas ball energy	<u>60 tons</u>
<b>Total</b>	<b>77 tons</b>

Energy generation doesn't stop when the system  $k_{eff}$  passes downward through  $k_{eff}=1$ . The power in fact is a maximum there. The energy will probably double or triple in a confined system before fission stops completely. If it doubled, the energy would increase to 150 tons and most of this would be in the gas. Most of this energy would be expended in vaporizing more rock or in producing more rubble until the pressure drops to the crushing pressure of 250 MPa. If all of the energy went into vaporizing the rock and the pressure stabilized at 250 MPa, the 150 tons of energy would vaporize about  $2 \times 10^5$  kg of rock according to the equation-of-state. It would occupy a volume of  $90 \text{ m}^3$  corresponding to a radius of 2.8 meters with a pressure of about 2500 atmospheres. This pressure may be compared with the 66 atmosphere lithostatic pressure at 300-meter depth. Even though the prompt kinetic yield, estimated at 15 tons, is much larger than that calculated from the lithostatic pressure, it is only about 1/4 of the total yield. This prompt kinetic yield will likely produce little surface damage but it could have significant effects on neighboring waste packages. Serious risk also would come from the release of this gas into the atmosphere and its accompanying 100 kg of plutonium and the prompt radiation associated with the fission product from the 150 tons of energy generation.

Since a pressure exceeding the lithostatic pressure by about 50 percent is known to be sufficient to widen existing fissures in the rock, the 40 times larger gas pressure should be adequate to open up significant channels. If a channel 10 cm in diameter were opened up along an existing fault line, the volume of a "pipe" of this size 300 meters to the surface would be about  $2 \text{ m}^3$ . This may be compared with the  $90 \text{ m}^3$  estimated above at the 2500 atmosphere pressure. The contained radioactivity including the plutonium could escape or move far from its original site elsewhere in the repository.

It is useful to note that the cavity remaining after a conventional nuclear explosive is created by the strong shock driving the rock away from the center. Most of the energy is carried away into the surrounding rock by the shock wave. By contrast, in the absence of shock for the systems considered here, the gas ball contains 80 percent of the energy generated. The 150-ton spontaneous

shockless explosion discussed here would generate much more gas by mass than the conventional underground nuclear explosion and the containment of the 250 MPa pressure must be maintained much longer than that from the smaller amount of gas generated in a nuclear weapons test. Therefore we expect that the confinement of the high pressure gas created by a spontaneous criticality event would be more difficult than that for underground nuclear weapons tests.

## REAL PLUTONIUM, REAL WATER, REAL ROCK, REAL GEOMETRY

In the previous analysis, pure Pu-239 was used as representative of TFM. While this is a good approximation for high enrichment uranium fuel (almost entirely U-235), in reality, plutonium almost always is accompanied by a significant component of Pu-240. 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 significant 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 Pu-240 resonance and therefore would significantly lessen its negative effects on the plutonium reactivity<sup>10</sup>. 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 Pu-240 resonance region and the probability of it escaping capture in the resonance would be high.

The half-life for Pu-240 is 6,600 years whereas the Pu-239 decay rate is about four times slower, and the daughter product of Pu-240 is U-236, 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<sup>11</sup>. After 25,000 years weapons-grade plutonium will be functionally equivalent to pure Pu-239. In the same period of time the plutonium contained in once-through spent fuel will be transformed by decay into weapons-grade plutonium.

The non thermally fissile isotope of plutonium Pu-242, on the other hand, has a much longer half-life, and could help in keeping plutonium safely subcritical essentially at all times, but its concentration is significant only in highly recycled or deep-burned waste fuels.

Another simplification introduced in the paper is the use of pure  $\text{SiO}_2$  as representative of rock. While the moderating properties of most rocks are very similar, most rock constituents have lower cross-sections than silicon. The effects of trace elements with high cross sections, such as rare earths, are usually negligible<sup>11</sup>. Considering, for instance, the composition of Nevada tuff, the overall thermal cross-section for neutron absorption is about 50 percent larger than for pure  $\text{SiO}_2$ , which indicates that 50 percent larger quantities of TFM would be needed for the criticality curves near the *right side* of figure 1 when  $\text{SiO}_2$  is replaced by the Nevada tuff.

Pure  $\text{H}_2\text{O}$  was also used for water. Here the effect of various poisons dissolved in the water, most notably chlorine, would be to increase the TFM concentration for the critical curves of figure 1 near the *left side* of the graph.

Considerable amounts of water (up to 15 percent in volume) can be lodged in the pores of structural rock, such as tuff. If the rock is fractured, much larger amounts of water can be present, as in the case of gravel and water table strata.

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<sup>11</sup>. While the spherical geometry allows criticality to be reached with a minimum amount of TFM, larger quantities of emplaced TFM could rearrange in a slab-like geometry of rock and water, such as might be found in strata or the water table. If the waste were to rearrange itself into an extended slab geometry, characterized by large length- and width- to thickness ratios, then the final energy release per kilogram of TFM could be substantially higher than for the sphere<sup>11</sup>. In these configurations, in fact, the leakage does not increase as the density decreases and therefore only the temperature rise and the associated neutron spectrum hardening will terminate the energy generation from an autocatalytic critical excursion.

## SUMMARY

It should be clear that we have only discussed possible mechanisms leading to unstable behavior in TFM stored underground in the most general terms. It was not the intention of the paper to discuss these mechanisms in relation to any specific geological setting. However, we believe that the possibility of these supercriticality mechanisms occurring in a particular geologic setting should be taken into account in the determination of the potential emplacement strategy.

As long as canisters containing thermally fissile material maintain their integrity, underground autocatalytic criticality is not a concern. After the canisters have been breached and eventually destroyed and the fissile material is free to move, spontaneous criticality with positive feedback is possible and explosions of significant yield can occur. The main points can be summarized as follows:

- ♦ 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.
- ♦ Both wet and dry autocatalytic conditions are possible, with TFM quantities as small as a few-kilograms behaving autocatalytically in some wet scenarios.
- ♦ The role of the 0.3 eV resonance in Pu-239, with its large temperature feedback, in initiating the positive feedback excursion is emphasized.
- ♦ The autocatalytic feature of buried w-Pu, HEU, naval spent fuel, and spent research reactor fuel could give rise to sequential ignitions if this nuclear material were to be stored in extended arrays of large individual TFM emplacements.
- ♦ Energy release in the several hundred gigajoule range is possible for spherical configurations containing about 100 kg of TFM. Larger yields per kg of TFM are possible in extended slab geometries.
- ♦ The role of the 1 eV resonance in Pu-240 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.

## NOTES AND REFERENCES

1. "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, 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.

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).
4. The MCNP 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. Cowan, G.A. "A Natural Fission Reactor," *Scientific American* 235, 36-47 (July 1976).
6. P. B. Parks, Savannah River Site, Private Communication, December 1994.
7. 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.
8. M. Wilson, et al, "Total System Performance Assessment for Yucca Mountain - Second Iteration" (TSPA-1993), SAND93-2675, Sandia National Laboratory, April 1994.
9. "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).
10. Petersen, P and Olander, D. (University of California Berkeley, Nuclear Engineering dept.) private communication.
11. Bowman, C.D. and Venneri, F. "Underground Supercriticality from Plutonium and Other Fissile Material" Los Alamos LA-UR 94-4022A.