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Comments on the Draft Paper "Underground Supercriticality from Plutonium and Other Fissile Material," Written by C.D. Bowman and F. Venneri (LANL)

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In response to a request from the Director of the Los Alamos National Laboratory, several members of the staff of the Lawrence Livermore National Laboratory participated in a technical review of a draft paper by C. D. Bowman and F. Venneri dealing with the potential for nuclear criticality in the geologic disposal of fissile materials. This review consisted of a consideration of the technical issues raised in the draft paper, and did not include discussions with the authors.

INTRODUCTION

Criticality safety in geologic disposal is an important topic which is provided for in Federal regulations and which has received attention from the technical community since the 1970's. In our view it is a subject that is best dealt with by a careful, risk-based analysis of possible scenarios, taking account of the actual details of proposed designs, rather than ad hoc calculations applying to hypothetical, idealized configurations. When the results of such a risk-based analysis are fed back to produce improvements in the designs, overall criticality safety is enhanced. In our judgment this is what is needed, and when properly done, the risk posed by the potential criticality of commercial reactor spent fuel and surplus weapons grade plutonium will be shown to be sufficiently low that it will not be of concern.

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Unfortunately, the draft paper by Bowman and Venneri does not take this approach and hence, suffers from a number of significant errors and shortcomings such as the following:

- It blurs the important distinctions among the various fissile-containing materials under consideration for geologic disposal. These distinctions relate to the individual criticality potential and to the current status of planning and decision-making for each of the materials.
- It makes statements and assumptions that are inconsistent with the known properties and behavior of actual waste forms and real rock.
- It fails to present plausible explanations of a process or processes that could transform the actual material as emplaced into something close enough to the hypothetical configurations modeled to make them relevant.
- It underestimates the capability of engineering, coupled with hydrogeochemical understanding, to make design choices that will control long-term criticality potential. These choices include dilution of fissile material, a range of possible added neutron absorbers, highly durable ceramic waste forms, and carefully designed physical layout in geologic disposal.
- It fails to take account of many of the effects of the strong coupling between the nuclear processes and the materials behavior in a system that has achieved criticality. These effects nearly all act in the direction of terminating autocatalytic action and rendering the system subcritical.
- It ignores the disparity between the slow rates of geologic processes and the rapid speeds of assembly needed to achieve prompt criticality and significant neutron multiplication before disassembly. It therefore vastly overestimates the potential fission energy release in the unlikely event of criticality.

Perhaps this draft paper has had the positive effect of drawing attention to a topic that was in need of it. However, nuclear criticality safety requires attention of a more careful and thoughtful nature than was given in this draft.

Because of the serious technical errors and shortcomings of the draft paper, we do not believe it would make a useful contribution to the literature in the field of criticality safety in geologic disposal of fissile materials.

BACKGROUND

In response to a request from the Director of the Los Alamos National Laboratory, several members of the staff of the Lawrence Livermore National Laboratory performed a technical review of the subject draft paper.[1] The results of that review are summarized here.

The topic of nuclear criticality safety of fissile materials in geologic disposal is an important one, and it deserves serious and careful study. Its importance has been recognized since consideration was first given to geologic disposal of commercial reactor spent fuel in the 1970's.[2,3,4,5,6,7,8,9,10,11,12,13] This recognition is reflected in the Federal regulations governing geologic disposal of spent fuel and high level radioactive waste, promulgated by the U.S. Nuclear Regulatory Commission in 1983.[14]

DISTINGUISHING AMONG FISSILE-CONTAINING MATERIALS

In discussing this topic, it is important at the outset to distinguish clearly among the several materials containing significant concentrations of fissile nuclides that are under consideration for geologic disposal, using two criteria:

(i) their fissile content, and hence, their potential in principle to achieve criticality, and

(ii) the stage of consideration, decision-making and planning associated with their disposal.

On one end of the spectrum is commercial reactor spent fuel,[15,16] which has low concentrations (less than 2 percent by weight, on the average) of fissile material, and which is destined according to long-standing federal policy and statute to be disposed of in a geologic repository. On the other end of the spectrum are surplus weapons grade plutonium and naval and research reactor Highly Enriched Uranium (HEU) spent fuel. Unless (or until) it is diluted, surplus weapons grade plutonium is over 90 percent fissile Pu-239 by weight. Its disposition is in the very early stages of the decision-making process.[17] It has not yet been determined whether this plutonium will be used in nuclear reactors to generate energy or will be placed in some form in geologic disposal. If it is used in light water reactors, mixed oxide (MOX) spent fuel will result. If it is to be placed in disposal, it has not been decided whether it would go into a mined repository or into one or more deep boreholes. The waste form, its fissile concentration, added neutron absorbers, and the physical layout are still open choices as well. The destiny of the naval and research reactor HEU spent fuels is also still undecided. Direct disposal of these fuels offers particular criticality challenges because of their high fissile content and because there would be less waste form design flexibility than for plutonium. HEU spent fuel is discussed in a risk-based criticality study recently published.[13] Careful treatment of criticality safety requires that the various materials be dealt with individually and that it be made clear which design options are available.

CRITICALITY POTENTIAL OF COMMERCIAL REACTOR SPENT FUEL

Turning first to commercial light water reactor spent fuel, it must be noted that its actual physical configuration and composition[15,16] as well as its planned repository emplacement are very different from the hypothetical models of critical configurations discussed in the draft paper. Commercial reactor spent fuel consists of uranium oxide pellets in long assemblies of (mostly) zirconium alloy tubes. For average spent fuel, more than 93 weight percent of the pellets consists of $^{238}UO_2$. The remainder is composed of $^{235}UO_2$ (about 1 weight percent), oxides of plutonium isotopes (less than 1 weight percent), fission products and their oxides (less than 4 weight percent), and other actinide oxides (less than 1 weight percent). The spent fuel assemblies are to be inserted and sealed inside rather substantial containers incorporating added neutron absorbers.[18,19] No silica is found in commercial spent fuel. The regulations require that water not be present in the packages in amounts that could compromise the waste package long-term containment.[14] Current designs envision that these large packages would be emplaced horizontally in open drifts (tunnels) in a potential repository in the unsaturated zone about 200 meters above the water table inside Yucca Mountain, if this site is found suitable (site suitability is also a decision yet to be made). The rock there is Topopah Spring tuff, as mentioned in the draft paper. It is a welded, devitrified, porous tuff composed of oxides of a variety of chemical elements in addition to silicon. The pores are about 2/3 filled with water under ambient conditions.[20]

In the draft paper, on the other hand, all the actual modeling was done on hypothetical, spherical, homogeneous mixtures of Pu-239, silica, and water, imbedded apparently with no void space in an infinite, dry silica medium. If such modeling is to have relevance to geologic disposal of commercial reactor spent fuel, it must be shown, at least plausibly, how the actual material and configuration could transform to something approaching the hypothetical models. This was not done in the draft paper, and thus its applicability to commercial reactor spent fuel disposal has not been demonstrated. This is not to say that criticality is completely impossible with commercial reactor spent fuel in geologic disposal after a very long time and under some particular conditions. However, the likelihood is extremely small, and in our view can be made even smaller (and thus acceptably small) by appropriate design choices. In our judgment, the most productive means of studying such low-probability events is by means of a careful, risk-based assessment that takes account of the actual characteristics of the system studied, and feeds results back to produce design improvements and greater safety. Some of this work has been done; more is needed.

Some of the characteristics that would help to prevent the occurrence of criticality for commercial reactor spent fuel in a potential Yucca Mountain repository are as follows:

- The fraction of fissile nuclides is small. Note that the Oklo natural fission reactors[21] achieved criticality when the U-235 content was 3.68 weight percent, whereas commercial spent fuel would have an average of about 2 weight percent (after plutonium decay to U-235). For comparison, the minimum enrichment required for a pure, infinite, homogeneous UO_2-light water system to achieve criticality is 0.96 weight percent U-235.[22] Mixed oxide (MOX) spent fuel, if produced, would have somewhat higher fissile content than current commercial reactor spent fuel.

- Added neutron absorbers would be present, and would have to be physically separated from the fissile material to allow criticality. Absorbers with a range of solubilities are available, including the lanthanides and hafnium, as well as boron and others.

- A moderator would be required to produce criticality with commercial reactor spent fuel,[23] but the site is unsaturated, and the current water infiltration rate is low. (These hydrological conditions, however, cannot be assured over extremely long time scales, of the order of the half-life of U-235).

- It would be difficult to reconcentrate the fissile materials to achieve a critical mass after geochemical transport, if the fissile materials are the mobile species.

- It would be difficult to achieve a shape with low enough surface-to-volume ratio to achieve criticality in the presence of gravity, which would promote slumping into a configuration with higher neutron leakage, if the drifts remain open, with essentially flat floors.

- Neutrons would be absorbed parasitically by U-238, and this does not result in fission. This will be particularly important if the self-shielding that was provided by the lumped-fuel configuration is lost. Alteration and transport of uranium would produce a more nearly homogeneous fuel-moderator configuration.

A careful, risk-based study of possible scenarios coupled with neutron transport calculations for bounding but realistic configurations is needed. Measurements and geochemical modeling to develop increased understanding of the geochemical behavior of the fissile elements as well as the complete range of potential added neutron absorbers are also needed. It is our view that if this work is carried out, and the results are fed back into design, the geologic disposal of commercial reactor spent fuel can be shown to be sufficiently safe from nuclear criticality that the issue will cease to be of concern from a comparative risk viewpoint.

CRITICALITY POTENTIAL OF SURPLUS WEAPONS GRADE PLUTONIUM IN GEOLOGIC DISPOSAL

As noted above, the potential geologic disposal of surplus plutonium is at a much earlier stage of consideration and decision-making than is that of commercial reactor spent fuel.[17] Accordingly, the emplacement designs are only in the conceptual stage, and there remain several options. One option is the choice of the waste form. Candidate waste forms include plutonium metal, plutonium oxide, or plutonium incorporated in another metal, in glass, or in a crystalline ceramic. These candidates have different properties and durability. One of the crystalline ceramics under consideration is based on a naturally-occurring uranium-containing mineral (zirconolite). Specimens of this mineral have been found that have ages in the range of billions of years.[24] This ceramic could be expected to have very high long-term durability as a waste-form. Zirconolite can incorporate a variety of neutron absorbers, including lanthanides and hafnium, in addition to its intrinsic titanium, all chemically bound in its crystal structure on the atomic scale. In fact, some hafnium (up to 4 percent by weight of zirconium) is naturally present in zirconium ores and would be present in the zirconium used to make zirconolite. Therefore, neutron absorption by hafnium would be obtained without special effort. Zirconolite does not contain silica.

Another open issue is the question of the concentration of plutonium in the waste form. Concentrations ranging from one to 12 weight percent are under consideration. At the one percent level, the ceramic waste form in any amount would be subcritical in the presence of any amount of water, even without additional neutron absorbers. Waste form designs incorporating depleted uranium, so as to dilute the U-235 that will result from plutonium decay, are also under consideration.

As was pointed out in connection with commercial reactor spent fuel, in order for the models in the subject paper to have relevance, there must be plausible means of transforming the initial emplacement configuration into the hypothetical configurations modeled. As was also true for spent fuel, the draft paper did not elucidate this for plutonium.

The draft paper appears to proceed on the basis that homogeneous mixing of geologically disposed material with the host rock would be inevitable, simply given enough time, and the only question is which mechanism will do it soonest. This notion is apparently based on a misapplication of thermodynamics, and is simply false. As an illustration, witness the inhomogeneity of the earth's continental crust on both fine and coarse scales at its mean age of about 1.5 billion years.[25]

Even if one starts with plutonium in a borosilicate glass waste form, which consists largely of silica, it is not a simple matter to arrive at something approaching a plutonium-silica-water system. In the case of borosilicate glass, it is actually observed experimentally that as the glass is attacked by water, boron and silica are removed stoichiometrically,[26] rather than selectively in proportion to their solubilities, as assumed in the draft paper. Boron does remain in aqueous solution, and can be transported away from the glass if there is sufficient flow. The silica reprecipitates in alteration phases (mainly clays) on or near the original glass. This is dictated by the high silica concentration in the water, resulting from its contact with the surrounding silicate rocks. The less soluble elements in the glass (including other added neutron absorbers beside boron, in this case) also remain. Criticality would likely thus continue to be controlled by these absorbers. There is no experimental basis for expecting reacted glass to take on a "spongy character." In actual fact, it is reduced to a mushy mass of hydrous alteration minerals.

For other waste forms, the plutonium would not initially be mixed with silica. In order to achieve the configurations described in the paper, the host rock would have to participate, the actual character of the rock would have to be compatible with the final compositions envisioned, and it would be necessary to have a mechanism to perform this mixing.

There are several problems associated with these requirements. First, the draft paper hypothesizes a situation in which Pu, H_2O, and SiO_2 are homogeneously distributed over a scale from tenths of a meter to several meters. This is impossible for the rock at the potential Yucca Mountain site. The rock units there are very heterogeneous on scales of millimeters to meters.[27] This is a result of the fact that these rocks were deposited by volcanic processes of vast proportion. The eruptions over 12 million years ago broke apart the preexisting rock units, incorporated fragments of them in the newly erupted material, and deposited them as complex mixtures of rock fragments, volcanic glass, and lava. Subsequent alteration of the rocks took place unevenly, thus resulting in a highly varied rock mass.

Second, the paper suggests that it is possible to simulate rock, for neutron transport purposes, by any arbitrary ratio of silica and water. However, the porosity of the rock at Yucca Mountain is limited to a maximum of about 20 volume percent.[20] Even if the rock were saturated with water, the equivalent mole fraction of water would not exceed 0.24. Thus, most of figure 1 [in the Bowman and Venneri paper, this issue], including much of the so-called "autocatalytic" region, could not be realized with intact Yucca Mountain rock, and thus any speculation about criticality behavior in this region is irrelevant. One might suggest that crushed backfill would have more porosity and greater homogeneity. However, it has not been decided whether backfill will be used.

Third, with regard to the actual character of the Yucca Mountain rock units, many of the minerals that occur have complex chemistries, containing significant amounts of other chemical elements beside silicon and oxygen, including stronger neutron absorbers as well as hydrogen, predominantly a neutron moderator. These occur inhomogeneously scattered throughout the rock and would have important effects on the neutron economy. In the case of the deep borehole disposal concept under consideration for weapons grade plutonium, the brines that would be encountered are rich in chloride, which is also a good neutron absorber.

The notion of a homogeneous distribution of plutonium in an arbitrary mixture of only SiO_2 and water with spherical geometry is thus at wide variance with the properties of the actual rock under consideration.

With regard to mixing processes, the paper envisions the transport of plutonium in rock, both from higher to lower concentrations as well as the reverse. The mechanisms for this purported transport, supposedly bringing about uniform concentrations in spherically-shaped regions of rock, are not elucidated in detail. Phrases such as "water-steam expulsions," "driven by fission heat through the rock," "water carrying plutonium oxide particles and depositing them somewhere else," and "earthquakes or more modest geologic shifts" are used. The paper seems to be suggesting several means of transporting plutonium and mixing it with rock, roughly falling into the categories of aqueous, thermal, and mechanical processes. The known aqueous processes include: (1) dissolution followed by transport and sorption or precipitation, and (2) dissolution followed by colloid formation, transport, and deposition. The thermal processes suggested apparently include: (1) heating, evaporation, vapor transport, and vapor deposition, (2) entrained aerosols transported by steam bursts, and (3) dissolution in magma resulting from volcanic activity, followed by transport and/or liquid-phase mixing. The draft paper suggests that mechanical transport and/or mixing could arise either from tectonic or human causes. These potential transport and mixing processes are discussed in order.

First, transport of plutonium by aqueous dissolution is limited by its maximum solubility in water, and deposition by sorption is limited by its maximum sorption coefficient. Typical Yucca Mountain host rock has about 15 percent porosity, and the pores are about 67 percent saturated with water.[20] According to a consensus of specialists,[28] the maximum sorption coefficient (Kd) for Pu in devitrified tuff under expected oxidizing conditions is 200 l/kg, and its maximum solubility under the expected conditions is 10^{-6} mol/l (there remains some uncertainty in variation of solubility with temperature and with organic complexing species). An upper limit for the mass fraction of Pu present as dissolved and sorbed material is obtained by assuming 100 percent water saturation: one cubic meter of rock at sorption equilibrium would contain about 3.6×10^{-5} kg Pu in solution and about 0.11 kg Pu sorbed on the solid. The mole fraction of water is about 0.18. Note that the amount of Pu in solution is entirely negligible in comparison with the sorbed component. The resulting mass fraction of Pu can be calculated to be about 4.8×10^{-5}, compared to about 1×10^{-3} required for criticality (from the lowest curve in figure 1 of the draft paper. A higher concentration would be required in actual rock because of natural neutron absorbers).

Not only is this concentration of Pu subcritical when uniformly distributed in a sphere of less than 2 meters radius, the time required for it to be emplaced by flowing vadose-zone groundwater at Yucca Mountain is quite long. The current estimate for the site-average infiltration rate is 1.4 mm/year, or 1.4 l/year into each square meter of surface area. If percolating water were to encounter Pu, dissolve it, and carry it downward into a cube 1 meter on a side, about 160,000 years would be needed for enough water to pass through the cube to satisfy its sorption capacity for Pu. In a heterogeneous system of fractured rock, localized flow of 10 to 100 times the average has been observed in underground openings, so it is conceivable that as little as 1,600 years could be required to deposit this (subcritical) mass by transport in aqueous solution.

Clearly, if the current values of the solubility, sorption, and infiltration parameters are correct, there would have to be some process other than sorption to allow enough Pu to accumulate for the system to reach criticality. Suppose there is a chemical reaction that precipitates Pu from aqueous solution uniformly within a 0.5-m radius sphere. From figure 1 of the paper, the mass fraction required for criticality with a water mole fraction of 0.18 is about 0.003, corresponding to about 3.8 kg of Pu. The amount of saturated aqueous solution required to deposit this much Pu is 1.6 x10^7 liters. At the average infiltration rate, this would require 1.1 x 10^7 years; even with a 100-fold flow concentration due to heterogeneity, the time required is more than 100,000 years, or about four half-lives of Pu-239. It thus appears that it is simply not possible to create a critical configuration using plutonium by transport in solution, given the limited amount of water available at Yucca Mountain.

If the time under consideration were sufficiently far in the future (a multiple of 24,000 years), the plutonium would have essentially completely decayed to U-235. Uranium has a higher solubility (a central tendency of about $10^{-4.5}$ mol/liter has been noted),[28] but it also has a lower maximum sorption coefficient (5 l/kg).[28] The two parameters nearly compensate, and the mass fraction resulting from sorption would be similar to that obtained for Pu. If a hypothetical precipitation reaction were invoked, the higher U solubility would permit an accumulation rate of U considerably more rapid than that for Pu. However, the required concentration to achieve criticality would also be somewhat higher. In this hypothetical case, the longer half-life of U-235 would allow accumulation of a sufficient mass, but it should be borne in mind that precipitation of uranium does not normally occur unless reducing conditions are present,[29] and these are not observed naturally at Yucca Mountain.

Plutonium has been found in colloids when glass is attacked by small amounts of water.[30] Detailed work on transport of these colloids through rock remains to be done. However, colloids can only move through fractures, and this would not likely lead to anything near a uniform distribution of Pu on the scale suggested in the paper, because the spacing between fractures in the intact rock is approximately 0.3 meter on the average.[20] Crushed backfill would provide interstitial space for possible colloid deposition. However, as noted above, a decision has not been made about its use. If used, it would likely be emplaced in a graded configuration to provide a capillary barrier to water penetration of the emplaced fissile material. If the site remained unsaturated and this barrier continued to operate as designed, water would not be available to form or transport colloids. In the case of deep borehole disposal, free void volume would be minimized by design, and the granite found at a few kilometers of depth is observed to have very low permeability and widely separated fractures.

Neither is it likely that thermal processes could move Pu through rock to form a spherical distribution. Solid state diffusion is ruled out by the low homologous temperatures and the low solid solubility of Pu in rock-forming silicate minerals, owing to its large ionic radius and high charge. Vapor transport is ruled out by the very low vapor pressure of PuO_2 up to very high temperatures.[31] The mechanism suggested in the paper for selectively heating Pu to thousands of degrees while the surrounding rock is heated by only a few degrees is untenable on the grounds that the time scale of heating for a criticality achieved by slow geologic processes would be much too slow to allow such a temperature differential in the presence of the conductive and radiative heat transfer rates from the Pu to the rock. Furthermore, since fractures in rock are rough and circuitous, a gaseous Pu-containing molecule would soon collide with the rock surface, resulting in condensation. Smooth, radial cracks distributed with spherical symmetry do not exist in real rock. Invoking steam-transported aerosols is also questionable, since it is not clear how the plutonium aerosols would form, particularly with a ceramic wasteform, or if they did form, how they would be homogeneously distributed in rock, since the aerosols could only move through fractures, which, as noted above, are widely spaced. In addition, the flow of gases through fractured media tends to occur along preferential paths, since the flow resistance is very sensitive to the width of the fracture openings. This process would thus tend to spread the plutonium into planar, rather than spherical distributions, which would have greater neutron leakage.

Dissolution of Pu in a magma resulting from volcanic processes at Yucca Mountain is judged to be a low probability event, based on a review of available evidence about volcanism in the area.[32] The probability of occurrence of a magmatic event at Yucca Mountain is estimated to be about 10^{-4} over 10,000 years and about 10^{-2} over one million years. If a magmatic event occurred, the next questions would be whether the packages would be contacted, and if so, whether the waste form would dissolve in the magma. If zirconolite were used as the waste form, it might not dissolve, since its melting point is much higher than common magma temperatures, and zirconolite crystals could be stable in contact with siliceous melts, depending on their composition.

Earthquakes can result in the shift of one block of rock relative to another and can mix a very narrow layer between, but neither of these effects involves mixing one chemical species homogeneously with another over a distance of the order of a meter.

Unwittingly drilling into a geologic repository in the course of exploring for minerals is a human intrusion scenario that has been considered. The probability that this will occur at some far distant time in the future is not possible to quantify accurately. What has been done is to ensure that the potential repository site at Yucca Mountain is not one that would appear particularly attractive for exploration compared to other sites, in terms of its geologic makeup. If drilling occurred at the site, the probability that the drill bit would intersect a waste package would be small. The economic incentive to use small bit diameters to reduce drilling costs would limit the in-situ diameter of material mixed during the drilling process. It is true that wet drilling produces a considerable volume of drilling mud that is brought to the surface, but the degree of mixing of plutonium with siliceous material would again depend on the waste form. Small pellets of zirconolite, if used, could remain essentially intact.

Our view is that sufficient design flexibility remains in the surplus weapons grade plutonium disposition program that criticality can be prevented with a high degree of confidence. As in the case of commercial reactor spent fuel disposal, a careful, risk-based study coupled with design is needed. More fundamental understanding of the geochemical behavior of the fissile elements and potential neutron absorbers, including solubility, sorption, complexation, and colloid behavior is also needed.

AUTOCATALYTIC BEHAVIOR

As we have explained above, the likelihood of achieving nuclear criticality in geologic disposal of commercial reactor spent fuel and surplus weapons grade plutonium can be made extremely small. However, it is important to examine the processes that could take place in the unlikely event that it actually occurred. The draft paper has suggested the possibility of autocatalytic behavior. The term autocatalytic in this context refers to any arrangement in which the motions of material that are produced by the nuclear chain reaction will act, at least for a time, to increase the effective neutron number. (The effective neutron number is equal to the effective neutron multiplication factor, k_{eff}, minus one). Autocatalytic methods in nuclear explosives design were discussed in the Los Alamos Primer in 1943.[33]

The possibility of autocatalytic behavior in the plutonium-high silica soil-water system was suggested by Clayton[34] in 1979, based on calculations by Carter[35] in 1973 pertaining to the criticality potential of the enclosed trench 216-Z-9 at Hanford. These were static neutron transport calculations for a series of assumed configurations having diminishing water contents, much like the calculations reported in the draft paper by Bowman and Venneri. While such calculations can indicate the initial slope of the k_{eff} vs. water loss curve for a given composition, they are not adequate to simulate the dynamic course of events that would actually occur in such a critical system. The reason for this is that the salient feature of such systems is the strong coupling between the nuclear processes that take place and the mechanics of the materials involved. This coupling occurs because the energy released by fission produces a temperature rise in the material, which in turn produces changes in the materials, most of which tend to reduce the nuclear reactivity of the system.

In order to accurately simulate the behavior of such a system, one must take account of the effects of the released energy and the temperature increase on both the materials behavior and the nuclear processes. Because of the strong coupling between them, this requires an iterative calculation that takes account of feedback.

Which effects come into play will depend on the details of the configuration. The effects can be viewed as a series of hurdles tending to render the system subcritical. Under certain circumstances, a system can be made autocatalytic for a short time, but it will soon encounter an insurmountable hurdle which will shut down the chain reaction unless very special efforts are made. These hurdles were some of the major challenges faced by the Manhattan Project in developing both the nuclear reactor and the atomic bomb.[36]

In a system composed of rock, water, and the fissile-containing materials under consideration for geologic disposal, the sequence of effects on the materials could include the following:

- increased thermal vibrations of the atomic cores,
- thermal expansion of both the water and the solid materials (by differing amounts),
- hydrothermal convection if the rock were saturated,
- dissolution and transport of minerals if the rock were saturated,
- evaporation of water below the boiling point if the rock were unsaturated,
- boiling of water at the boiling point,
- solid state phase transitions in the rock (with accompanying volume changes),
- melting of the rock (with accompanying volume changes) in the unlikely event that the previous processes did not terminate the criticality and the solidus temperature were reached,
- changes of shape or collapse of the geologic media under the effect of gravity,

- dissolution of inert rock in magma, resulting in dilution of the fissile material, and
- vaporization of rock and perhaps fissile material, in the unlikely event that sufficiently high temperatures were reached.

The accompanying effects on the nuclear processes as these materials effects occurred would include the following:

- increase in the average energy of the thermalized neutrons, with consequent changes in the various effective neutron interaction cross sections,
- changes in the ratios of moderator and absorbers to fissile material,
- changes in neutron leakage from the system resulting from volume changes, and
- Doppler broadening of resonance absorption peaks resulting from increased thermal vibrations of the atomic cores.

After extended fissioning, depletion of fissile material and accumulation of fission products would also be significant.

Unless these effects are accounted for, it is not possible to project the detailed course of events. Since the draft paper by Bowman and Venneri does not accurately do this, its projections are not defensible.

THE POTENTIAL FOR AN AUTOCATALYTIC EXPLOSION

Even if initially autocatalytic configurations such as those described by Bowman and Venneri were actually achieved, which appears to be very improbable, we find no merit in the proposition that they could lead to an explosive event. While we have not done a detailed quantitative analysis beyond confirming the essential validity of their static criticality calculations, we find that there are several assumptions in their analysis that are unjustified and are inappropriately applied. The major source of error in their reasoning is in the estimates of the time scales involved. In order to have an explosion, the generation of energy must take place on a time scale which is faster than the disassembly of the critical mass. Bowman and Venneri use a k_{eff} of 1.1 to derive a time scale of milliseconds for energy release, which might be short enough to allow an explosive event. However, a k_{eff} of 1.1 is reached only after the autocatalysis of the assembly, and hence cannot cause the autocatalysis. The appropriate time scale for the start of autocatalysis is that associated with a k_{eff} that is only slightly above critical (1.0). At this degree of criticality the time scale of energy release will be orders of magnitude longer than that associated with a k_{eff} of 1.1 and incompatible with the generation of an explosion. The references to the mechanisms that they believe will autocatalytically increase criticality are vague; however, all seem to involve phenomena that will take place on time scales that are orders of magnitude longer than the short time scale required for an explosive release of energy. Without a physical mechanism capable of increasing k_{eff} on a time scale shorter than the disassembly time, no explosion is possible. There are a variety of mechanisms, depending upon the particular case, that will cause disassembly long before the autocatalytic effects can operate. Some of these were listed in the previous section. The draft paper's treatment of this crucial issue of time scales is vague and incomplete to the point that a detailed analysis of the paper's proposals is not possible. In summary, we find no credibility in the authors' arguments for the cases they have presented. There must be a more detailed and definitive explanation of the processes that they believe could result in an autocatalytic explosion before a detailed analysis of their proposals could be made.

CONSEQUENCES OF CRITICALITY IN A GEOLOGIC DISPOSAL FACILITY

In the very unlikely event that a criticality should occur in geologic disposal of commercial reactor spent fuel or surplus weapons grade plutonium, its effects would be to produce heat energy, fission products, and actinides. The significance of these must be judged by comparison with the amount of radioactive decay heat normally emitted by the emplaced material, as well as the initially emplaced radionuclide inventories.

Although quantitative analysis of these effects would require examination of detailed scenarios, it is possible to project that these effects would not represent a serious threat, given the criticality conditions that could be produced by geologic means. The eighteen known fossil natural nuclear reactors in the Franceville Basin near Oklo, Gabon, Africa, provide qualitative precedents for the scale of what might be expected.[21] Temperatures in the reactor zones there apparently ranged from 160 °C to 350 °C during the reactions. These temperatures are within the design range of a geologic repository. The reactions are thought to have been terminated by silica dissolution, causing the collapse of the natural roofs of the reactors. This restructured the natural reactors and squeezed out the water moderator. Interestingly, this is one of the processes that Bowman and Venneri suggested would produce criticality. At

Oklo, this process apparently had the opposite effect. Over the following two billion years, most of the actinides were retained in the reactor zones. Some of the fission products underwent migration from the reactor zones, but many of them were retained in the halo regions around them. Quantitative comparison with geologic disposal of fissile materials is difficult because of differing or unknown geochemical conditions. Nevertheless, the scale of the effects observed in the only known instances of natural criticalities is instructive and is illustrative of what natural processes can achieve.

CONCLUSIONS

We conclude that the draft paper by Bowman and Venneri has failed to note the important differences among the several fissile-containing materials under consideration for geologic disposal. It has not demonstrated that the hypothetical models used are relevant to the disposal of commercial reactor spent fuel, which bears little resemblance in composition or configuration to the models it discusses.

In the case of surplus weapons grade plutonium disposal, the draft paper does not present plausible means for achieving the homogeneous mixtures modeled, and the models are inconsistent with the characteristics of actual rock. The draft paper fails to consider the possibility of selecting durable wasteforms or of selecting neutron absorbers on the basis of their projected geochemical behavior relative to that of the fissile elements to ensure long-term criticality control. Nor does the draft paper consider the possibilities of controlling the initial plutonium concentration at low levels or of arranging the physical layout in a configuration that would render the possibility of criticality remote. Since these design choices are yet to be made, there is ample opportunity to preclude criticality with a high degree of confidence.

The draft paper fails to take account of the strong coupling between the nuclear processes and the materials behavior in a system that has achieved criticality. The effects of this coupling are to terminate autocatalytic action and to render the system subcritical.

The draft paper also ignores the disparity between the slow rates of geologic processes and the rapid speeds of assembly needed to achieve prompt criticality and significant neutron multiplication before disassembly. It therefore vastly overestimates the potential fission energy release in the unlikely event of criticality.

It is our view that careful, risk-based analysis of possible scenarios and neutron transport calculations for bounding but realistic configurations are needed. Development of increased understanding of the geochemical behavior of the fissile elements and of potential neutron absorbers to be added is also needed. If this work is properly carried out, and the results are fed back into design, we believe that the geologic disposal of commercial reactor spent fuel and surplus weapons grade plutonium can be performed in a manner that is sufficiently safe from nuclear criticality.

Because of the serious technical errors and deficiencies in the draft paper by Bowman and Venneri, we do not believe it would make a useful contribution to the literature in the field of criticality safety in geologic disposal of fissile materials.

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