

DRAFT

WSRC-TR-95-0036

**CONSEQUENCES OF THE BOWMAN-VENNERI
NUCLEAR EXCURSION THESIS ON THE
PROSPECTS FOR PLACING VITRIFIED
PLUTONIUM CANISTERS IN GEOLOGIC
REPOSITORIES (U)**

MARCH 9, 1995

**UNCLASSIFIED
DOES NOT CONTAIN
UNCLASSIFIED CONTROLLED
NUCLEAR INFORMATION**

ADC/Reviewing Official: Richard W. Bony

Date: 3/21/95

Prepared for the U.S. Department of Energy under Contract DE-AC09-89SR18035

Westinghouse Savannah River Company
Savannah River Site
Aiken, SC 29808

9504180436 950407
PDR WASTE PDR
WM-11

Attachment 3

DRAFT

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represent that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

3/9/95

DRAFT

WSRC-TR-95-0036

Consequences of the Bowman-Venneri Nuclear Excursion Thesis on the Prospects for Placing Vitrified Plutonium Canisters in Geologic Repositories

P. B. Parks, T. G. Williamson, and M. L. Hyder

INTRODUCTION

Bowman and Venneri of the Los Alamos National Laboratory have prepared a paper entitled "Underground Autocatalytic Criticality from Plutonium and Other Fissile Material" and are circulating drafts for comment and critical review.¹ The basic thrust of the paper is to point out that

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

Obviously, this paper will have a significant effect on the several DOE programs that aim to dispose of fissile material in underground repositories, whether of the mined geologic type (tunnels and drifts as at Yucca Mountain) or of the deep borehole type.

This report examines the relevance of the Bowman-Venneri thesis to the ongoing efforts of the newly created DOE Office of Fissile Materials Disposition. The program of that office is built around disposition options for unirradiated plutonium and enriched uranium materials declared excess to the security needs of the United States. The consensus option for enriched uranium disposal appears to be to make it into low-enriched nuclear reactor fuel and to burn it in the nation's commercial power reactors. The options for plutonium disposal are more complicated but generally fit within one of three options, two with variant sub-options.²

- Store the plutonium in metal, oxide, or some other stabilized form for the indefinite future.
- Dispose of the plutonium in such a way that its disposal form meets the "spent fuel standard" by being as inaccessible for weapons use as the plutonium in spent nuclear fuel from commercial power reactors:
 - 1) Make mixed-oxide (plutonium-uranium) fuel and burn it in power reactors. Dispose of the spent fuel in a geologic repository.
 - 2) Immobilize the excess plutonium in some medium (glass, ceramic, etc.) with or without radioactive high-level wastes to form canisters for disposal in a geologic repository.
 - 3) Dispose of the plutonium directly in metal, oxide, or immobilized forms by burying it in deep boreholes.
- Dispose of the plutonium in such a way that goes beyond the "spent fuel standard" by nearly completely consuming it in:

DRAFT

WSRC-TR-95-0036

- 1) Accelerator - subcritical reactor combinations where the plutonium would be fissioned nearly to extinction.
- 2) Deep burn nuclear reactors, with or without spent fuel reprocessing cycles, where the plutonium would be fissioned nearly to extinction.

The disposal of the plutonium by any of the sub-options that meet the "spent fuel standard" are brought into question by the Bowman-Venneri thesis, because these sub-options involve burial of either spent mixed-oxide fuel or unirradiated plutonium directly into the ground. Such buried disposal forms are susceptible to the creation of single event nuclear yield in the "hundreds of tons" range (as shown in the body of Reference 1). If the buried canisters are closely spaced, the excursion can spread domino-fashion throughout most or all of the forms, multiplying the total yield.

This report is confined to the effect of the Bowman-Venneri thesis upon the sub-option of the "spent fuel standard" option that involves disposal of unirradiated plutonium in canisters of borosilicate glass within the confines of a mined geologic repository or a deep borehole. The extension of this study to the other "immobilization forms" (ceramics, metals, concrete, etc.) for Pu disposal should not be difficult but is not undertaken here.

SUMMARY

The first requirement for any thesis to have an impact on a program is technical acceptance. To be accepted, the Bowman-Venneri thesis will have to pass tests that can be summarized in four questions:

- Is the criticality physics correct?
- Is the excursion yield physics correct?
- Is the occurrence of the autocatalytic event a reasonably probable event?
- Is the consequence of an event unacceptable?

Two of the tests appear to already have been passed.

- Independent calculations at SRS, reported in the Appendix of this paper, have confirmed the criticality physics.
- The yields possible in the autocatalytic excursions appear to be reasonable, and have been confirmed at LANL.

The last test cannot be resolved by technical discussion alone, for, at heart, it is a regulatory/political question. In our opinion, the consequences will be perceived as politically unacceptable. The initial response to the Bowman-Venneri paper appears to substantiate this.

For these reasons, this paper concentrates on the probability of the Bowman and Venneri type of criticality excursion. It is argued here, qualitatively, that the event is sufficiently probable over a long time period that it must be taken into account when trying to estimate the programmatic risk of any proposed action to dispose of excess plutonium.

The Bowman-Venneri nuclear excursion cannot occur as long as the canister shell remains intact (i.e., for hundreds or thousands of years) and as long as sufficient nuclear poisons remain with the fissile material. After canister degradation, the probability of a Bowman-Venneri type of nuclear excursion per year is probably small. However, three observations concerning the nuclear supercriticalities and the repository design temper whatever comfort we might draw from the small yearly probabilities of initiator formation.

DRAFT

WSRC-TR-95-0036

1. Only near canister mixing of soil and plutonium is necessary to initiate a nuclear eruption involving a single borosilicate glass canister if nuclear poisons are not present in significant amounts. Removal of the neutron poisons by leaching from the Pu-glass, combined with only a modest amount of soil mixing can initiate the eruption. Indeed, putting the Pu in glass provides a "head start" for initiation.
2. The vulnerability to criticality events persists nearly indefinitely because Pu-239 decays to U-235, which has a half-life of 7×10^8 years and which is almost equally fissionable. Thus, even if the annual probability of an event is 10^{-6} per year or less, a period much greater than 10^6 years must be considered.
3. A possibility of the spread of the supercriticality blast throughout the entire repository (if the canisters are reasonably closely spaced) would exist.

If the Bowman-Venneri type of excursion occurs, the result can be a yield of ~0.4 kilotons (and a vaporized cavity of ~9 meters diameter) with a possible extension to ~150 kilotons yield if all of an assumed 50 MT of excess plutonium became involved in the excursion. The overall probabilities of such a scenario have not been shown to be small enough to satisfy regulators, let alone the public.

To counter this threat, repository and canister designers would have to adopt one or more of the following design requirements.

1. The concentration of Pu in each canister must be small enough that leaching of the neutron poisons naturally occurring in borosilicate glass (B and Li), combined with modest fissile material spreading into the repository soil, cannot create the conditions for a large nuclear eruption.
2. The canisters must be widely dispersed to prevent reinforcement of a supercriticality should one occur.
3. Some neutron poisons, to be added to the Pu-glass during fabrication, must be found that remain with the plutonium and its uranium daughter products for essentially the life of the Pu-239 and U-235, respectively, or the habitable life of Earth, whichever comes first. A single poison is not sufficient because plutonium and uranium solubilities differ by a factor of about 300.
4. A repository design would have to be devised that could absolutely exclude water for the habitable life of the Earth.

The first two conditions would increase the cost of a mined geologic repository or borehole. The third condition appears to be very difficult to certify, especially over the long geologic times involved. The need for two different poisons lessens confidence in this approach. The fourth requirement does not seem technically or economically feasible in an underground environment.

Faced with the costly choices to keep the canister content of Pu small, and to spread the canisters widely, the repository designer might decide to accept the risk of the Bowman-Venneri type excursions. He could, in principle, resort to the discipline of Probabilistic Risk Assessment (PRA) in an attempt to gauge that risk. However, a PRA (or Performance Assessment) does not appear to be credible as a means for establishing confidence in the magnitude of the risk when attempted for a length of time of 1-2 billion years.

DRAFT

WSRC-TR-95-0036

For these reasons, the prospects for disposal of vitrified plutonium in a mined geologic repository or a borehole appear to be brought into serious question.

None of the storage or disposal options that go beyond the "spent fuel standard" are affected by the Bowman-Venneri thesis. Moreover, the MOX fuel option remains unassailed if chemical processing is assumed instead of direct fuel disposal in underground geologic repositories. (Direct fuel disposal is envisioned in the present MOX option of the plutonium disposition program.) However, the probability of Bowman-Venneri type blasts from degradation of spent mixed oxide fuel has to be much smaller than from degraded vitrified plutonium logs, as argued in the Discussion.

Though not a part of the plutonium disposition program, it should be mentioned that the disposal of defense high level waste in borosilicate glass logs in mined repositories is unaffected by the Bowman-Venneri thesis. The amount of fissile material in these logs is too small to be susceptible to eruptions.

DISCUSSION

1. Background

The Plutonium Immobilization program, now being conducted by the DOE Office of Fissile Materials Disposition, envisions the disposal of excess weapons plutonium (assumed 50 MT for planning purposes) by incorporating the fissile material in some long-lived medium suitable for dispersal in a geologic repository. The repository could be either a mined set of tunnels and caverns of the type being designed and explored at Yucca Mountain in Nevada, or one or more deep boreholes sunk several kilometers beneath the earth's surface.

Several different media that could incorporate the plutonium in reasonably stable canisters are being investigated, including:

- Borosilicate Glass
- Synroc
- Phosphate Glass
- Metallic Alloy
- UETAP
- High Silica Glass
- Monazite
- etc.

To date, the leading candidates appear to be borosilicate glass, the same medium in which the defense high level waste will be incorporated for disposal, and Synroc. This discussion is confined to the borosilicate glass medium, although extension to other glass media, as well as Synroc, will be readily apparent. Two useful publications on glass properties and the use of glass for plutonium disposition are listed in References 3 and 4.

Bowman and Venneri have recently put forward a study which shows that, over a period of geologic time, plutonium disposal in repositories is vulnerable to nuclear eruptions caused by autocatalytic (positive feedback) supercritical excursions.¹ In the scenarios envisioned by Bowman and Venneri, after canister degradation in the repository, the plutonium would become mixed with the surrounding soils, principally SiO₂ or any other such light compounds, by natural forces, such as dissolution in water and consequent soil binding, or by inadvertent human intrusion or sabotage.

In such a dry configuration, large autocatalytic criticality excursions are predicted to occur if sufficient plutonium (50-100 or more kgs) is dispersed in repository soils over a thickness of around 1-5 meters.¹ Such underground supercritical systems, by themselves, could be expected to have yields in the "hundreds of tons" range, or higher, depending on the geometries. If the canisters were sufficiently closely spaced, the vaporization of repository medium could extend to adjacent canisters, involving them in the event, and thus reinforcing this massive energy release. Even if the probabilities, which have not been quantified, are very low, the subsequent consequences could be high, perhaps raising the risks to unacceptable levels.

2. On the Probability of the Bowman and Venneri Nuclear Eruption

The following scenario is introduced to show that an excursion of the Bowman-Venneri type is not impossible. Every assertion about k_{eff} is backed up with specific k_{eff} calculations that are reported in the Appendix of this paper. Assume that a steel canister, of the type produced in the SRS Defense Waste Production Facility (2 ft diameter and 10 ft length), containing a Pu-borosilicate glass mixture is deposited in a mined geologic repository or a borehole. In the course of geologic time, the outer steel shell will corrode away exposing the Pu-glass contents to the leaching effects of ground water. In underground repositories, over such long periods, no technology is known that can prevent the ultimate corrosion and destruction of the protective canister shell. (When considering geologic time periods, the existence of a man-made, engineered protective container becomes irrelevant.)

Boron, one of the neutron absorbing elements in borosilicate glass, will be leached out of the Pu-glass rubble relatively quickly.³ Lithium is also present, and depending on the repository chemistry, may or may not form precipitates, which may or may not stay reasonably close to the original canister site. Most lithium compounds are quite soluble in water. A soil reflected canister containing 134 kgs of weapons plutonium (metal) will experience a rise of k_{eff} from about 0.3 to about 0.9 if the poisons are effectively removed. This amount of plutonium is within the range of Pu loadings per canister discussed as feasible in the recent report by the National Academy of Sciences on plutonium disposal. Once the boron and lithium have been removed, the presence of water with only an H/Pu atom ratio of ~10 (2 wt% H₂O) would drive the system critical. (Complete removal of the boron and lithium is not necessary. A higher water concentration would still cause a criticality.) Use of other poisons might delay but can not be shown to prevent the above criticality scenario (as discussed in Section 6).

A chugging type of criticality event could occur. The system k_{eff} would probably not exceed delayed critical. Water could be driven out by steam formation to temporarily shut down the criticality until sufficient water trickled back in. However, this repeated churning of steam and heated water over aeons though the Pu-glass rubble could accelerate the leaching of plutonium and its equally fissile uranium daughter products and could spread these materials into the soil surrounding the canister. Bowman and Venneri argue that the repeated steam bursts would fracture the rock in the immediate vicinity which would further the possibility of Pu/U being driven into the adjacent medium. The dispersal of Pu/U into the surrounding soil would be accompanied by an increasing dry system k_{eff} as the dispersal progressed. That is, between the wet system criticality excursions, the minimum k_{eff} would sooner or later exceed unity. When a Pu/U dispersal of only some 30-50 cm into the surrounding soil occurred, the calculations listed in the Appendix show that the dry k_{eff} could exceed prompt critical and cause a Bowman-Venneri type of excursion. Bowman and Venneri estimate the yield at about 3 tons/kg of fissile material. Hence, the yield of a single canister event would be about 0.4 kilotons.

Any collection of Pu in this amount (134 kgs) will be vulnerable to a criticality excursion if buried underground as a single unit. The Pu-glass wastefrom has the misfortune to have provided the excursion a "head start" because the principal component of the glass is SiO₂.

We have shown that the Bowman-Venneri type of excursion is possible. We have not shown that it is probable. Indeed, there is insufficient data available to prove conclusively just what the probabilities of the above scenario are. From this point on, we can only argue qualitatively concerning the probabilities. But to address this issue, we must first determine the period of time over which we must be concerned about such a criticality event.

3. The Time Required for Nuclear Blast Prevention

The definition of a time for which a nuclear eruption (and by inference criticality) must be prevented within a geologic repository of fissile waste transcends the bounds of purely technical discussion. The thought of allowing 400 tons of nearly instantaneous energy release within the confines of a geologic nuclear waste repository appears to be politically daunting. As argued in Reference 1, the blast generated from multiple canisters could be much larger than that from a criticality involving a single canister.

Such eruptions would not be some "ho-hum" affair that could be dismissed easily as having little or no consequence. It might be argued that such eruptions and their effects would be entirely confined within the repository and not cause any threat to humans or any other biota on the surface.* After all, many nuclear detonations have been set off in the underground test chambers of the Nevada Test Site on which Yucca Mountain is partially sited.

However, such energy releases are not supposed to occur in nuclear waste repositories, particularly if significant fission product inventories remain. Venting of blast created fission products would be a concern. The effect on the performance of the repository would have to be painstakingly researched. An overwhelming case would have to be constructed before the technical community could be convinced that these large eruptions in the midst of the repository were harmless. Even then, politicians and regulators would have a difficult time facing the intense public scrutiny that would surely follow the announcement of such a conclusion.

If the above argument holds, then selecting some very long, but arbitrary time for the prevention of such nuclear excursions is a meaningless exercise. Why should generations living near the repository after, say, 10,000 years be protected any less than generations born before 10,000 years? incidentally, the EPA selected criterion of 10,000 years of protection is based on the prevention of harmful fission product release to the accessible environment from high-level-waste burial, not the prevention of a critical nuclear excursion.

In connection with this line of inquiry, the half-life of Pu-239, the principal fissile component in weapons plutonium, is approximately 24,000 years. However, the daughter product of Pu-239 decay is U-235, which is almost as fissile as Pu-239. The half-life of U-235 is $\sim 7 \times 10^8$ years. In the Appendix, the progression of k_{∞} is shown with time as Pu-239 and Pu-240 decay to U-235 and U-236, respectively. The effect on k_{∞} is <6% over 100,000 years. Hence, if the above argument prevails, criticality prevention must be maintained for a few billions of years (assuming mankind and Earth last that long), unless some arbitrary regulatory limit is imposed.

* Note: As Bowman and Venneri point out, the nuclear eruptions are not explosions in the classic sense because the energy releases occur over some milliseconds of time, too slow for the formation of shock waves. This does not mean that this is not a violent event. The use of gunpowder as a propellant is a case in point.

On the other hand, the conclusion that criticality must be prevented for such a large time is certainly arguable. Just as income or debt anticipated many years in the future ought to be discounted, the importance of preventing a possible disaster far enough in the future could be discounted; it could be lost in the noise of what is going on continuously. According to the World Almanac, there have in this century been four earthquakes and four cases of storm and/or flood that have each killed >100,000 people; tens of millions have been killed by war, famine, or pestilence; there have been some truly massive volcanic eruptions, which did immense damage and had the potential for massive fatalities; and there was a meteorite strike comparable in its effects to those of a rather large thermonuclear weapon. All of these exceed the likely effects of a partially contained fission explosion in a, presumably, lightly populated area.

This philosophical question ultimately turns on the argument as to the responsibility of the present generation to avoid taking any action that might ultimately add to the risks facing future generations. This is a political question that must be dealt with by regulators and politicians, to which the technical community can contribute, but not decide.

It seems likely that the political/regulatory community will impose the condition that excursions of the Bowman-Venneri type must not occur in the repository for the life of humanity or perhaps 1-2 billion years. In effect, then, the yearly probability of such an event will have to be integrated over an extremely long time. Even if the yearly probability remains very small, say $\sim 10^{-9}$ per year, the overall probability could be as high as unity over 10^9 years (assuming a constant probability).

Of course, we don't know the yearly probability at this time. However, we might consider recourse to the discipline of Probabilistic Risk Assessment (PRA) to determine it, and the attendant risk of a criticality excursion.

4. On the Use of PRA Techniques to Determine the Criticality Excursion Risks in a Repository

A commonly accepted technique for dealing with low probability - high consequence accidents is to perform a probabilistic risk assessment (PRA) in an attempt to discover whether the risk (probability x consequence) is acceptable. PRA is an excellent tool for predicting accident frequencies in the area of once in a million years; although at this frequency, the uncertainties may be fairly large but tolerable. PRA is well suited where equipment, procedures, or human actions provide barriers to prevent possible untoward scenarios from occurring. However, one must be able to construct with a high degree of certainty each of the possible scenarios and the probability of failure of each of the barriers. It is very unlikely that all of the possible scenarios have been identified. It is even more unlikely that probability of failure of each barrier over 10^9 years can even be remotely determined. Uncertainties in values applied to the individual "components" would be so large that even if an assessment could be made, the combined uncertainties would render a final answer virtually useless.

Thus, the technical community will be faced with a untenable situation. It will be asked to pass technical judgement on the probability of Bowman-Venneri type of excursion, over a period of about a billion years. However, the only tool the community will have to perform this analysis will be inapplicable. Therefore, any estimate of the probability, high or low, will be unsupported by any objective analysis and can only be subjective, at best.

5. On the Loading and Spacing of Pu Containing Canisters in the Repository

The more plutonium that is placed in a single canister, the fewer will be the number of canisters. If the criticality excursion problem in the repository did not exist, then the upper limit of plutonium that could be placed within a single canister would be set either by criticality consideration during canister filling or by the properties of the Pu-glass system.

On the basis of very early calculations, the boron and lithium in the borosilicate glass itself could prevent criticality during fabrication if the percentage of weapons Pu in the glass did not exceed ~15 wt%.⁵ Pu-glass with apparently acceptable properties in which the Pu percentage was about 7-10 wt% has already been demonstrated.⁶ The canister size has not been chosen, but if one used the glass canisters the size of Defense High Level Waste (DHLW) canisters as an example (10 ft length, 2 ft diameter), about 1,680 kgs of Pu-glass could be contained in each. If 8 wt% is chosen as an upper limit of the percentage of Pu that could be placed in a single large canister of the DHLW type, then ~134 kgs of weapons grade (or other) Pu could be contained. Just this type of canister, and loading, was considered in the NAS report on plutonium dispositions as within the possible range.

In Reference 5, a DHLW type canister containing 23 kgs of weapon's Pu (~1.4 wt%) was postulated for costing purposes. On the basis of a range of 23-134 kgs Pu being selected for inclusion in DHLW type canisters, the total number of canisters would be somewhere between 373 and 2,174 to dispose of 50 MT of plutonium.

The disposal of glass canisters in a geologic repository is costly. A measure of this is the repository fees contemplated for DOE-owned spent fuels, which are in the range of \$250,000 to \$700,000 per canister. These costs are strongly affected by the number of canisters and the canister spacing, as the overall cost of preparing and qualifying the repository must be apportioned among all the different types of canisters emplaced. It can be estimated that the repository fees for glass canisters would be in the range of \$100 million and \$1.5 billion, depending on canister loading and spacing, and there would be a strong incentive to aim for the lower number. This economic factor strongly encourages close spacing of the canisters, say a few meters apart.

The upper end of the range of Pu concentrations in a single canister is sufficient to fulfill the Bowman and Venneri condition for criticality initiation as shown in Section 3. At the lower end, the Pu content of three or four of the 2,174 canisters would have to combine to trigger the eruption. If the spacing were only a few meters, the fireball or neutron field from a 400 ton nuclear eruption (~9 meters in diameter¹) could encompass other adjacent canisters, which would cause the inclusion of the other canisters in the eruption and lead, domino fashion, to a very large (~150 kilotons) total energy release, if the other canisters were in a similar state of leaching.

According to Bowman and Venneri, this domino-like involvement would come about from direct heating of the neighboring thermally fissionable material or from neutron induced fissioning.¹ The eruption would, within milliseconds, create a plasma of vaporized plutonium, glass, and rock out to a diameter of about nine meters. If the canister spacing were about four meters, or less, the nearest neighboring canisters would be exposed to the fireball and nearly instantaneously vaporized. The rock would suffer a phase change and increase in density about a factor of two. This would create free space into which the vaporized material could rapidly expand under the driving force created by the extreme plasma temperature. The rock vapor and the Pu-glass vapor of the nearest neighbor canister would begin to mix. This system would experience an increasing k_{eff} , and when k_{eff} exceeded delayed critical, would undergo a new autocatalytic, criticality eruption. This direct heating scenario could spread, domino-fashion, throughout the repository. If the total energy release were simply additive (not a given) then the total could amount to about 150 kilotons (assuming 400 tons from each 134 kg Pu canister).

6. On the Use of Neutron Poisons to Prevent Bowman-Venneri Excursions

Obviously, a possible step in the prevention of excursions of the Bowman and Venneri type would be to include neutron poisons in the Pu-glass waste forms. In connection with the question of nuclear poisons, it is worth noting that uranium is potentially more soluble than, and hence, more mobile than the plutonium. Thus, at least two types of poisons would be

necessary, one to stay with the plutonium and the other to stay with the more mobile uranium daughter products. Gadolinium could be used for the plutonium and depleted uranium for the uranium daughters. The added U-238 would have to be many times the concentration of U-235 to be effective as a neutron poison. This would leave much less room for the incorporation of the plutonium into the glass. The use of gadolinium (or another rare earth) as a poison does not ensure it will stay with the plutonium; the coefficient of solubility, K_{sp} , for gadolinium hydroxide is about 10^{-22} moles⁴/liters⁴; that of Pu (IV) hydroxide is about 10^{-55} moles⁵/liters⁵.

It does not appear to be possible to devise laboratory tests that will demonstrate that neutron poisons, such as gadolinium, will stay with the plutonium over a period of, say, 10^9 years. Practical leaching tests have uncertainties in the measured result of some one to five percent. However, differences in leach rates of a magnitude similar to the test uncertainty could result in significant separation over geological time of a poison material and a fissile material. In criticality studies, the burden of proof is always on the proponent of a mitigating action. It is difficult to see how leaching tests can provide the required proof of non-separation over aeons.

If one wishes to address the poison-fissile material subject correctly, one should not speak of the fissile material and poison as separating. In reality, the more leachable and soluble material (presumably the poison) would have a broader distribution than the fissile material. That is, the breadth of the fissile material distribution could be spread, over geologic time, more narrowly than the poison. See Figure 1. This would facilitate the formation of the initiating criticality event. If a Bowman-Venneri type of autocatalytic excursion occurred, the fireball could encompass more of the region containing the spread out poison. If it did so appreciably, the excursion could be terminated prematurely and the yield decreased. However, the modeling of the poison/fissile material spreading, the nuclear excursion initiation, and the fireball spreading would be difficult to accomplish with any certainty. The lack of certainty would not argue in favor of taking credit for the poison remaining within the fireball region.

7. Consequences of the Bowman-Venneri Thesis on Pu-Glass Disposal in a Mined Geologic Repository

It is not impossible to dispose of Pu-glass canisters in a mined geologic repository with a high degree of safety if one is willing to pay the price. Reduction of Pu loading per canister will greatly reduce the probability of initiation of the nuclear eruptions of the positive feedback, autocatalytic type described by Bowman and Venneri. Wide spacing of the canisters in a single repository would delay, but not absolutely prevent, the collection of Pu in sufficient quantity at one place in the soil of the repository necessary for a nuclear eruption, and would prevent the domino-like involvement of the whole repository in the excursion. Both of these measures will simply increase the cost of repository disposal.

Consider the canister spacing alone. To prevent the domino effect of one blast causing all the canisters in the repository to become involved, the spacing would have to be much wider than the fireball diameter of the original blast (~9 meters by Bowman and Venneri). Let us choose 25 meters for the spacing. Storage of 373 canisters, each containing 134 kgs would then require ~0.23 km² of repository area. The use of a smaller canister loading would increase the size of the required repository area by the inverse of the loading. For instance, at 23 kgs per canister, and with 25 meter spacing, the repository size would be ~1.4 km². The 0.23 km² area represents about 14% of the 400 acre Yucca Mountain repository. The 1.4 km² area would require about 86% of the same repository. Clearly, the costs of the \$9 billion Yucca Mountain program to be allocated to the Pu-glass program would be very large. Such a large allocation also assumes no opposition from the commercial fuel interests (an unlikely prospect).

The above calculations serve only to illustrate the effect of canister spacing on the economics. If the reader thinks that 25 meters would be too large a spacing, he is free to reduce it to whatever extent he can defend.

8. Consequences of the Bowman-Venneri Thesis on Pu-Glass Disposal in a Deep Borehole

The deep borehole concept is also being investigated for the:

- direct disposal of chopped Pu parts
- direct disposal of Pu oxide, Pu metal
- disposal of immobilized Pu (glass, ceramic, etc.)

Unfortunately, the Pu-glass canister disposal mode appears vulnerable to initiation of an autocatalytic nuclear excursion in a borehole.*

Assume that somewhere between 373 and 2,174 DHLW type canisters are placed in a single borehole. These numbers of canisters would be required if 50 MT are placed in canisters with loadings of 134 kgs down to 23 kgs in each. The vertical length of the column of canisters, assuming no spacing between canisters and ~3 meter canister lengths, would be somewhere between 1.1 km and 7 km to dispose of the postulated 50 MT of weapons plutonium in this fashion.

At 134 kgs Pu/canister, only one canister would be required after canister corrosion to form a potential initiator of a nuclear supercriticality (following loss of neutron poisons by leaching). At 23 kgs Pu/canister, three to four adjacent canisters could pool their plutonium in a single collection to initiate the nuclear eruption (again presuming loss of the nuclear poisons). The other lower and higher lying canisters would almost immediately become involved in the eruption, providing a very large total energy release.

Obviously, the canisters should not be placed in contact with each other. Bowman and Venneri calculate the maximum size of an underground fireball at about nine meters diameter. Clearly, we would separate the canisters vertically to prevent the domino effect. Assume that a spacing of ~25 meters was chosen. Add about three meters for the canister length. Thus, the vertical stack of spaced canisters would now be somewhere between 10 km and 61 km in total length, depending on the canister loading. If the boreholes were between 2 and 4 km deep and the canisters were confined to the bottom half of the boreholes, the number of boreholes to contain all the canisters would be somewhere between 5 and 61, depending on the number of canisters and the average depth of the boreholes. The drilling costs have been estimated in early projections at about \$60 million per hole. Thus, the borehole costs alone might be somewhere between \$300 million and \$3.7 billion.

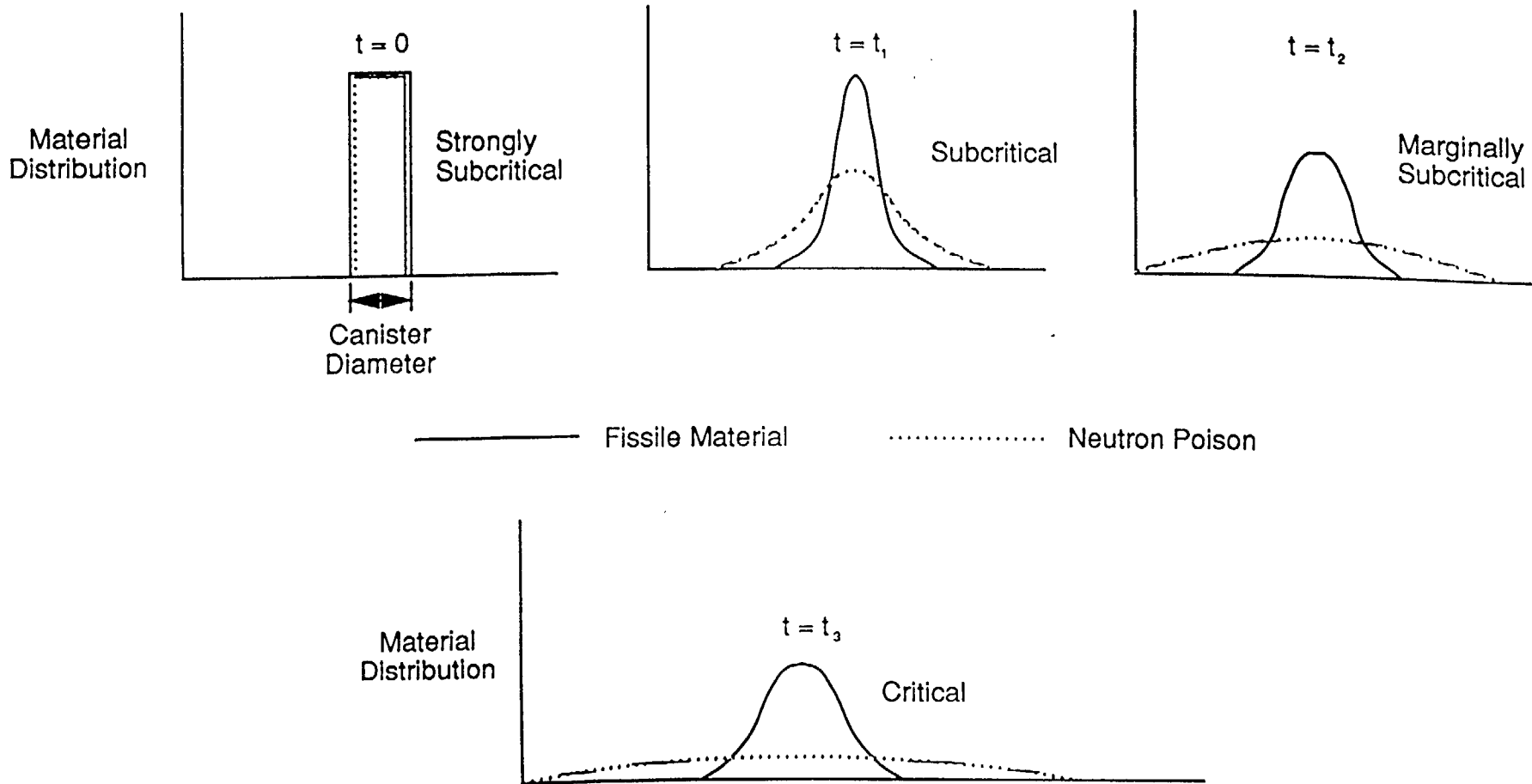
It is worth noting that in deep boreholes leaching of the Pu-glass may be much quicker than in near-surface emplacements. This is because at great depth and pressure, superheated water may be present, and it is a very aggressive leachant.

The deep borehole concept is, therefore, faced with the same unpalatable choices as the mined repository. Either prevent the domino effect of nuclear eruptions by separating the canisters vertically, and suffer the economic consequences, or accept the risk of the domino effect and live with the consequences of a very large blast. Pooling of the Pu/U from several small content canisters seems to have a larger likelihood of occurrence in a borehole than in the mined repository.

* This paper assumes that SiO₂ is the principal medium of the borehole. Bowman and Venneri have shown that the form of medium (tuff, granite, etc., except for salt domes) makes little difference.

Figure 1

Conceptual Spreading of Neutron Poisons and Fissile Material in an Underground Repository



11

DRAFT

9. Effects of the Bowman-Venneri Thesis of Nuclear Blasts on Non-Pu-Glass Waste Forms

This paper has pointed out the effects on the program for disposing of plutonium in borosilicate glass as a consequence of the nuclear eruptions that are possible with such forms in both mined geologic repositories and deep boreholes. The non-glass disposal forms have not been specifically treated in this paper. However, no obvious advantages of the non-glass forms are apparent to the authors. We would expect similar results from studies of the non-glass forms.

10. Conclusions

Unless the Bowman-Venneri thesis can be discredited on physical grounds, DOE must approach the question of geologic disposal of plutonium very cautiously. Comparative cost analyses of the various alternatives should take into account prevention of the Bowman-Venneri type of criticality excursion. We suggest that the direct disposal of Pu-glass in a mined geologic repository may not appear economically attractive if that is done. Thus, the success of the vitrification program is threatened.

On balance, there is little doubt that we can, if we want, engineer a repository that can safely hold the plutonium without danger of criticality for ca. 10,000 years. The Egyptians did this sort of thing in their construction of tombs and pyramids 5,000 years ago, and even wood furniture and cloth from inside these have been recovered in good condition. Chinese structures thousands of years old have also survived. On the other hand, most of these were re-entered and looted within a few hundred years of construction, and many of the remainder have suffered the same fate in our own century. Except for the deep boreholes, this would likely be the fate of any repository we build. History shows that man's institutions typically last some hundreds of years at most; policies are even shorter lived. Someone within the next thousand years is likely to want this plutonium and unless cheaper alternatives are available, they will get it (and digging it up is not prohibitively expensive).

Even if the option of repository disposal of vitrified plutonium is abandoned, the other options for plutonium disposition will still remain valid. However, the deep underground disposal of spent MOX fuel can be questioned on the basis of the Bowman-Venneri thesis. One technical answer to that is chemical processing. Whether reprocessing will be required is an open question. The decay of plutonium to uranium, and the presence of the very low enriched uranium in the spent fuel probably means that the time of maximum vulnerability is just after canister destruction up to ~100,000 years. Beyond that time, only very low enriched uranium will remain, and that material must have a much lower vulnerability to a Bowman-Venneri type of supercriticality than the pure Pu-glass systems discussed above. Further study will be required to resolve this issue.

Beyond the MOX fuel disposal mode are the storage, Integral Fast Reactor, and Accelerator-Subcritical Reactor options. These are unaffected by the Bowman-Venneri thesis. Also unaffected is the possibility of using vitrification as a stabilization step for plutonium and storing the Pu-glass logs above ground while long-term disposition strategies are decided. Above ground storage eliminates the possibility of a Bowman-Venneri type of nuclear excursion.

11. Acknowledgments

The authors wish to express their appreciation to the following individuals for very helpful technical discussions and reviews of this paper: C. E. Apperson, R. W. Benjamin, C. D. Bowman, W. S. Durant, K. L. Ferguson, H. E. Hootman, W. R. McDonell, J. M. McKibben, and G. G. Wicks.

APPENDIX

Criticality Calculations for Plutonium Buried In DWPF Canisters

This is an evaluation of plutonium in 2 ft. by 10 ft. canisters in western soil. The proposition is that the boron and lithium poisons are leached from the canister and separate from the plutonium oxide which remains with the other glass components. The canister may slump and expand radially. Also, the plutonium and/or its uranium daughter products may leach from the glass and be forced a short distance into the surrounding soil. Is there a criticality concern?

The canister dimensions and plutonium loading are shown in Table 1 and the material characteristics are in Tables 2 and 3. In Table 2, three representative glasses are listed: glass with all components including PuO₂ at 171 g/l, glass with the boron and lithium compounds removed, and glass without boron and lithium but with water to an H/Pu ratio of 100. Two plutonium mixtures were considered, Pu-239 with 6% Pu-240 and all Pu-239. These mixtures maintain the ratios of SiO₂, MgO and Na₂O as used by Skiles and Mincey⁷, and maintain the plutonium concentration of 134 kg plutonium in a canister 2 feet diameter and 10 feet long. The glass density is theoretical density. Listed in this table are the compound weight fractions, the mixture density in g/cc and the H/Pu atom ratio for one wet glass mixture. Table 3 lists the soil properties used by Bowman and Venneri.¹

Table 1
Canister Characteristics

Diameter	60.96	cm.
Length	304.8	cm.
Vol.	890	Liters
Pu	134	kg
Pu	151	g/l
PuO ₂	171	g/l

Table 2
Glass Characteristics

	Wt. Frac.	Wt. Frac.	Wt. Frac.
SiO ₂	0.7206	0.8490	0.5472
B ₂ O ₃	0.0749	-	-
Li ₂ O	0.0655	-	-
MgO	0.0187	0.0221	0.0142
Na ₂ O	0.0562	0.0662	0.0426
PuO ₂	0.0642	0.0627	0.0933
H ₂ O	-	-	0.3091
Density	2.66	2.72	1.83
H/Pu	-	-	100

Table 3
Soil Characteristics

	Wt. Frac
SiO ₂	0.716
Al ₂ O ₃	0.121
H ₂ O	0.040
K ₂ O	0.035
CaO	0.024
Density	2.69

The Joshua J70 Modules HRXN-KENO were used in this analysis. HRXN computes atom densities and prepares mixture cross sections in the Hansen-Roach 16-group energy structure. KENO is a Monte Carlo module which computes the effective multiplication factor. Results for several cases are in Table 4.

Table 4
Monte Carlo Results

Single Canister	keff 1 sig.		keff 1 sig.	
	0% Pu-240		6% Pu-240	
Full Glass Mixture	0.292	0.002	0.280	0.001
Mixture w/o B or Li	0.906	0.004	0.854	0.006
Mixture with water H/Pu=10	1.174	0.008	0.979	0.008
Mixture with water H/Pu=20			1.086	0.008
Mixture with water H/Pu=40			1.230	0.007
Mixture with water H/Pu=60			1.325	0.008
Mixture with water H/Pu=100	1.582	0.004	1.455	0.006

Canisters in linear array	keff 1 sig.		keff 1 sig.	
	0% Pu-240		6% Pu-240	
One can	0.906	0.004	0.854	0.006
Three cans	0.930	0.004	0.850	0.007
Five cans	0.933	0.004	0.847	0.007
Seven cans	0.940	0.006	-	-

Single Canister	keff 1 sig.		keff 1 sig.	
	0% Pu-240		6% Pu-240	
Expanded radius Fixed Volume				
R = 30.48 : H/D = 5.0	0.906	0.004	0.854	0.006
R = 35 : H/D = 3.3	0.968	0.004	0.888	0.007
R = 40 : H/D = 2.2	1.014	0.004	0.939	0.007
R = 50 : H/D = 1.1	1.074	0.004	0.983	0.008
R = 60 : H/D = 0.7	1.076	0.009	0.973	0.008
R = 70 : H/D = 0.4	1.040	0.009	0.952	0.009

Single Canister Expanded radius Reduced Pu concentration	keff 1 sig.		keff 1 sig.	
	0% Pu-240		6% Pu-240	
R = 30.48 : 151 g Pu/l	0.906	0.004	0.854	0.006
R = 45 : 69 g Pu/l			0.870	0.008
R = 60 : 39 g Pu/l			0.895	0.006
R = 90 : 17 g Pu/l			0.940	0.008
R = 120 : 9.7 g Pu/l	1.174	0.008	0.976	0.008
R = 150 : 6.2 g Pu/l	1.202	0.008	1.020	0.006
R = 180 : 4.3 g Pu/l	1.167	0.007	1.008	0.006

These data show that the single canister is safe with the boron and lithium leached away, however, is not safe if water intrudes. The addition of water to make the H/Pu atom ratio as low as 10, which corresponds to about 2% by weight water, significantly increases the multiplication factor.

The canisters stacked one above the other to make a long cylinder with no boron or lithium or water maintain a multiplication less than unity.

If a single canister without boron, lithium or water slumps to a cylindrical shape with fixed volume approaching an H/D ratio of unity, it becomes unsafe with Pu-239 only but maintains k_{eff} less than unity with Pu-240.

If the plutonium material expands radially into the surrounding soil (but does not shrink axially) so the fissile density decreases, the system can reach conditions with k_{eff} greater than unity with and without the Pu-240

Both Pu-239 and Pu-240 decay by alpha emission to U-235 and U-236, respectively, with half lives of thousands of years. An estimation of the reactivity effect was made by computing the infinite multiplication factor, K_{inf} , for the dry mixture with no boron or lithium. For this computation, a direct substitution of a uranium atom was made for each plutonium decay. Half lives of 24,100 years and 6,570 years were used for Pu-239 and Pu-240, respectively. Results are in Table 5

Table 5
Change in Infinite Multiplication Factor

Decay Time Years	K_{inf}
0	1.728
10,000	1.699
50,000	1.654
100,000	1.628

This indicates that the substitution of U-235 for Pu-239 and U-236 for Pu-240 does not markedly change the multiplication factor.

DRAFT

WSRC-TR-95-0036

References:

1. C. D. Bowman and F. Venneri, "Underground Autocatalytic Criticality From Plutonium and Other Fissile Material Stored Underground", DRAFT Unpublished LANL Report.
2. U.S. Department of Energy, Office of Fissile Materials Disposition, "FY 95 Program Plan", October, 1994.
3. G. G. Wicks, "Nuclear Waste Glasses; Corrosion Behavior and Field Tests", Corrosion of Glass, Ceramics and Ceramic Superconductors", D. E. Clark and B. K. Zoitos, eds., pp. 218-268, Noyes Publications, Park Ridge, N.J. (1992).
4. G. G. Wicks, J. M. McKibben and M. J. Plodinec, "Vitrification for Disposition of Excess Plutonium", presented at Waste Management '95, Tucson, AZ, February 26-March 2 (1995).
5. J. M. McKibben, et al., "Vitrification of Excess Plutonium", WSRC-RP-93-755, May, 1993.
6. M. J. Plodinec, "Development of Glass Compositions for Immobilization of SRP Waste", DP-1517, February, 1979.
7. S. K. Skiles and J. F. Mincey, "K-Infinity Analysis of Weapons-Grade Plutonium Dioxide in Defense Waste Processing Facility Borosilicate Glass (U)", WSRC Report, SRT-CMA-93021, May, 1993.

Title:

**COMMENTS ON "NUCLEAR
EXCURSIONS" AND "CRITICALITY
ISSUES"**

Author(s):

**Gregory H. Canavan, DDP
Stirling A. Colgate, T-6
O'Dean P. Judd
Albert G. Petschek, DDP
Thomas F. Stratton**

Submitted to:

For Discussions outside the Laboratory

Date: 7 March 1995

**Los Alamos
NATIONAL LABORATORY**



Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the University of California for the U.S. Department of Energy under contract W-7406-ENG-84. By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. The Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy.

Form No. 12870
ST 2088 10-91

9503170013 7 pp.

COMMENTS ON "NUCLEAR EXCURSIONS" AND "CRITICALITY ISSUES"

Gregory H. Canavan, Stirling A. Colgate, O'Dean P. Judd

Albert G. Petschek, Thomas F. Stratton

Los Alamos National Laboratory

Technical reviews of papers on criticality and energy release from underground storage of fissile material concluded the probability of each of the steps required is vanishingly small and the probability of occurrence of all of them is essentially zero. Even if they could occur, any release would be too small and slow to produce significant consequences in the repository or on the surface

The Laboratory provided technical reviews of papers by Drs. Bowman and Venneri. The first, entitled "Nuclear Excursions and Eruptions from Plutonium and Other Fissile Material Stored Underground"¹ ("Nuclear Excursions") was reviewed in December, 1994, and a written response was submitted to the authors through Laboratory management. The second, entitled "Criticality Issues for Thermally Fissile Material in Geologic Storage"² ("Criticality Issues"), which was a response to the issues raised in the December review, was reviewed in February, 1995. This review summarizes the assessment of both. Very recently, the authors released a third paper, entitled "Underground Autocatalytic Criticality from Plutonium and Other Fissile Material"³ ("Underground Autocatalytic Criticality"). However, it is largely a compilation, without correction, of materials from the first two; thus, our comments apply to it as well.

The papers primarily discuss the underground emplacement of glassy logs containing weapons plutonium, and purport to demonstrate that after on the order of 10,000 years, geologic action will increase their reactivity to the point where criticality, auto-catalytic action, and explosive energy release are probable. The significant difference between the papers is that the first ascribes the increase in reactivity to the dilution of plutonium in a dry silicon dioxide medium, while the second two ascribe the increase of reactivity to the concentration of plutonium in a wet silicon dioxide medium.

The review concluded that the discussion in the papers does not describe a credible sequence of geologic events leading to super criticality and explosive energy release. The probability of each of the necessary steps—increase in reactivity to criticality, auto-catalysis, and explosive energy release—is vanishingly small, and the probability of occurrence of all three is essentially zero. Moreover, even if these steps could occur, any energy release would be too small and slow to produce any significant consequences either in the repository or on the surface. Indeed, any surface effects would occur on times of tens of thousands of years, which are so long as to be outside the time scale of any credible scientific prediction.

Emplacement, dispersal, and criticality. The geological situations discussed in "Nuclear Excursions" were too unrealistic to provide a useful framework for analysis or to validate the proposed scenario. That was pointed out in the review, but those situations were still used in "Criticality Issues." "Nuclear Excursions" postulates the emplacement of fissile materials in geologic formations of pure silicon dioxide, which is a weak neutron absorber, is not a common geologic material, and has not been proposed as a repository material. Other elements present in all geologic formations absorb neutrons much more strongly than pure silicon dioxide, which reduces the reactivity of the mixture. Although the papers mention minor soil constituents with very large absorption cross sections, their calculations ignore them. The papers offer unsupported estimates that including them would increase the critical mass by 50%. When they are properly included, it may not be possible to achieve criticality for the assumed conditions even with pure Pu-239. It is not possible to be more quantitative in our response without further analysis of weapons Pu and spent fuel in realistic media, which is not performed in these reports. That must be done in a more careful subsequent project.

The papers perform most of their calculations for pure Pu-239. The weapons plutonium of interest has a significant fraction of Pu-240, a strong absorber that further reduces reactivity. Even for the maximum loadings postulated in "Nuclear Excursions," weapons plutonium could never disperse to a condition of criticality in real, dry repository materials. It is argued that the Pu-240 would decay, leaving the more reactive Pu-239, but that would happen over several times the 6,500 year half life of Pu-240. Even then the Pu-240 would be replaced by its daughter U-236, which is a weaker but still noticeable absorber, degrading the thermally fissile mixture.

The assumption of significant dispersion of plutonium into the surrounding geologic medium is without justification. Geologic processes would take millions of years, by which time plutonium would have decayed to uranium-235, which is less reactive than Pu-239. We have not discovered a credible process that would produce more rapid dispersal. Anthropogenic measures are unlikely and are routinely accounted for in repository analyses. "Criticality Issues" argues that water flowing down through the repository would dissolve the glass log in 1,000 years and leave a fragile powder, but its calculation overestimates the amount of rainfall on—and water within—the repository by factors of 1,000, so the correct time scale for dispersal is about a million years.⁴ Moreover, the temperature gradients driving the process are overestimated by an order of magnitude, and the leaching process could leave a residue as strong as the original log.

Autocatalysis. The papers' assumptions about the behavior of the fissile mixture near criticality are not credible. Based on their improper interpretation of published equations of state, "Nuclear Excursions" and "Underground Autocatalytic Criticality" assumed the rock in which the fissile material is placed is rigid and would prevent the expansion of the material. Rock is compressible, and even at depths of several kilometers, lithostatic stresses are small and

anisotropic, so that confining stresses are small. Even if the mixed material became critical, it would slowly heat and expand, which would decrease its reactivity below critical. Then its neutron flux would drop, and it would cool.⁵ Thus, these dry mixtures have the negative temperature coefficients characteristic of most fissile assemblies, as discussed in detail in the open meetings of the review, and would not be autocatalytic for material motion over geologic time scales.

"Criticality Issues" again argued that fissile material could diffuse to criticality, although it shifted its argument to SiO_2 with high amounts of water, which have higher reactivity.⁶ However, the physics for such media is essentially the same as that for dry rock.⁷ There are two parts to the argument, depending on whether the mixture approaches criticality from the under moderated or over moderated side. From the under moderated side, as the mixture reached criticality, it would heat slightly. That would expel some water, which would reduce its reactivity, after which it would cool.⁸ This is closely related to the stabilization of dry media by a negative temperature coefficient.

From the over moderated side, as the mixture gradually passed through criticality, it would heat slightly—though not enough to expel significant water—which would cause it to expand. That would reduce its reactivity, after which it would cool.⁹ Thus, over moderated, heavily hydrated mixtures generally also have negative temperature coefficients.¹⁰ Thus, there is nothing new in the papers on wet media, which just repeat the stability errors made in "Nuclear Excursions" in a different context.

A key feature not addressed in the papers reviewed is importance of the evolution in time of the criticality and temperature of the mixtures. For those of interest, the time scale for the increase of reactivity is very long—tens to hundreds of thousands of years. Thus, the excess levels of criticality and hence the time scales for the release of energy are correspondingly long—thousands to tens or hundreds of thousands of seconds. And the temperature increases are fractions of a degree. The slowness of those processes dominate the faster time-dependent processes postulated but not analyzed in the reports.

There are some scientifically interesting interactions between the negative temperature coefficient of such mixtures from expansion and the potentially small positive coefficient from absorption and Pu-239 resonance broadening, but those effects are delicate and comparable even at very high levels of hydration. Unfortunately, they cannot be evaluated from the calculations in "Criticality Issues," which were apparently all performed for cold soil, pure SiO_2 , and pure Pu-239. All three of those restrictions would have to be removed to provide an assessment beyond that in "The Myth of Nuclear Explosions at Waste Disposal Sites," which predicts overall stability.¹¹

Energy release. Even if dispersion and criticality are assumed, the conclusion that an explosion would occur is incorrect. "Nuclear Excursions" postulates "auto-catalytic" behavior in which the release of energy leads to greater criticality, but the discussion above shows that in dry repository material, the release of energy instead reduces criticality and shuts the reaction off.

"Criticality Issues" postulates auto-catalytic behavior in hydrated mixtures, but the discussion of the previous section shows that to the extent that the phenomenon has been quantified by earlier work, the release of energy reduces criticality there, too. Temperature increases appear to be limited to at most fractions of a degree for plausible dispersal times.

The postulated mechanisms for explosion are not credible. The essential feature of explosive process is the rate at which energy is released. The papers do not calculate it; they do not even estimate it. They simply assume it. For the largest realistic rates the most that appears possible is heating and evaporation of some water before a smooth shut down. There is no credible mechanism for releasing energy on a time scale short enough for even a steam explosion. A nuclear explosion must make the transition from critical to highly supercritical in a fraction of a second. A credible means to force such a transition in a repository has not been found.¹² Thus, the assertion that an explosion would occur is incorrect.

Even if dispersion, criticality, and energy release are *assumed*, which appear virtually impossible on the basis of the arguments above, there would be no serious consequences elsewhere in the repository or on the surface. Even if an explosion could occur, careful calculations indicate that the energy released would be on the order of a few percent of that from the natural decay of the Pu over the same time scale. Detailed hydrodynamic calculations indicate that the containment volumes from such explosions would be very small compared to the nominal spacing between storage elements; thus, there could not be any coupling between storage elements or any possibility of greater energy releases through synergisms.¹³

Relation with other work. That the critical mass may be reduced by dilution by moderating material, as discussed in the paper, is well understood by the nuclear community. Fermi used it to full advantage when he assembled the first pile under the grandstand at Stagg Stadium.¹⁴ Fermi also used the advantages of heterogeneity in minimizing resonance losses in natural uranium, although that is irrelevant to the discussions of Pu reactivity here.

The National Academy of Science report does not suggest emplacement of weapons plutonium in the manner discussed by "Nuclear Excursions," although it did comment on the advantages of higher fissile loadings. The Academy was alert to the potential for criticality and qualified its recommendations by stating that further analysis and discussion were needed before deciding on the best and safest geologic disposition of weapons and reactor spent fuel.

Summary. We should always be alert to unintended consequences and open to discussions that illuminate potential dangers in nuclear waste storage. "Nuclear Excursions" argued that there were serious dangers in proposed repository concepts, but review found the paper's major assumptions flawed and its major conclusions incorrect for fundamental, technical reasons, which were stated in detail and in writing. "Criticality Issues" did not respond to those criticisms; instead, it introduced a new scenario, in which it made the same technical errors in a new context.

Those errors were combined for publication in "Underground Autocatalytic Criticality." We find no technical merit in these papers. However, they treat technical matters and apparently contain no classified material; thus, in accord with the Laboratory's policy of open and unrestricted research and discussion on unclassified matters, the authors should be free to submit their paper for publication in a peer reviewed journal.

We do not find any value in these two papers that would justify their publication, and do not see how to produce such a paper from them. They contain fundamental errors in concept and execution. They show no grasp of such elementary concepts as the time scale for the approach to criticality, the rate of energy release, and the crucial role of the negative temperature coefficient of the systems treated. Moreover, they show no appreciation of these points even after they were pointed out clearly in the review by those who do did understand them. That is compounded by the shifting scenarios on which the papers are based and the alarmist estimates of potential effects, which have become less credible and more shrill throughout the review process.

The authors have shown little interest in technical suggestions or inclination to respond to them; thus, it would not appear to be useful to continue this one-sided discussion. However, it would be irresponsible for the Laboratory to disseminate untested opinions in this visible and controversial area. Thus, if this program is continued, and these individuals remain associated with it, the laboratory would be well served by establishing a permanent red team, funded by this program and composed of members from the cognizant technical divisions, with the responsibility of independently checking the calculations done by those in the program.

HR-10-1990 23-14

References

1. C. Bowman and F. Venneri, "Nuclear Excursions and Eruptions from Plutonium and Other Fissile Material Stored Underground," draft, 22 November 1994.
2. C. Bowman and F. Venneri, "Criticality Issues for Thermally Fissile Material in Geologic Storage," draft, February 1995.
3. C. Bowman and F. Venneri, "Underground Autocatalytic Criticality from Plutonium and Other Fissile Material," Los Alamos National Laboratory document LA-UR-94-4022 (draft), March 1995.
4. T. Kunkle, "Bowman and Venneri, Part Deux," Los Alamos memo EES-5, DDEES/CEP-95-006, 15 February 1995.
5. G. Canavan, "Time Dependence of Neutron Density and Material Temperature in Thermally Fissile Mixtures," Los Alamos National Laboratory report, March 1995.
6. W. Stratton, "Neutron Transport Reactivity Calculations of Plutonium Mixed with and Reflected by Silicon Dioxide," Los Alamos memo, 14 December 1994, Fig. 7 and Table V.
7. W. Stratton, "Pu/SiO₂ Overmodulation and Autocatalysis," Los Alamos memo, 10 January 1994.
8. K. Despain, "k_{eff} vs. Assembly Radius from MCNP Calculations—Weapons Grade Pu in Tonopah Springs Rhyolite," Los Alamos memo for blue team, December 1994.
9. G. Canavan, "Time Dependence of Neutron Density and Material Temperature in Thermally Fissile Mixtures," *op. cit.*
10. W. Stratton, "The Myth of Nuclear Explosions at Waste Disposal Sites," LA-9360, October 1983.
11. W. Stratton, "The Myth of Nuclear Explosions at Waste Disposal Sites," *op. cit.*
12. J. Karmerdiener, "'Bowman-Venneri paper on 'Nuclear Excursions'," Los Alamos X-2 memo to Jas Mercer Smith, Blue Team Leader, 23 December 1994.
13. J. Mercer-Smith, "Super criticality Blue Team Preliminary Report," Los Alamos memo, December 1994.
14. E. Fermi, "Elementary Theory of the Chain-reacting Pile," *Science*, 10 January 1947.