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AUTHOR(S): Dr. Charles D. Bowman

SUBMITTED TO:

Los Alamos

Los Alamos National Laboratory
Los Alamos, New Mexico 87545

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Current Status and Recommended Future Studies of Underground Supercriticality of Fissile Material

Charles D. Bowman

I. Introduction

More than a year has passed since we released our original report[1] pointing out the possibility of natural or induced rearrangement of fissile material underground into a critical mass, the possibility of positive feedback in underground configurations, the confinement of the rock to produce significant yield, and the possibility of venting or explosion. The nuclear weapons and repository storage groups at both Los Alamos and Livermore[2] have been critical of our work while others have defended our calculations on wet[3] and dry criticality[4]. The conditions we identified for positive and negative feedback are no longer contested. The role of confinement of the rock in enhancing the yield from the explosion is still unsettled, and that is addressed later in this paper. The likelihood of confinement, venting, or explosive dispersion also remains unsettled and that is addressed here as well. Some critics of our work have tried to show that the probability of reconfiguration by natural processes is very small[5]. They argue further that emplacement can be done in such a way as to make the probability even smaller. Of course these additional efforts will raise the cost of waste emplacement and the question arises as to how much is enough. The answer to this question seems to not be an easy one.

Nearly all criticism of our paper is based on evaluation of the prospects for reconfiguration of the fissile material by natural phenomena. The possibility of accidental or purposeful intrusion into the repository and consequent rearrangement of fissile material are not discussed as if reentry into the repository were not likely. However, a recent study from the University of California, Berkeley[6] asserts that recovery of plutonium from storage is over ten times faster and over ten times less expensive than making ^{239}Pu in a reactor or performing isotopic separation for ^{235}U. The International Atomic Energy Agency has concluded that geologic repositories must be guarded indefinitely[7]. Therefore, repositories would seem to be the natural choice for rogue nations or terrorists to obtain material for construction of nuclear weapons. The fact that all of the elements represented in the fission products have an isotopic distribution different from the natural isotope is one example of the uniqueness of materials which also could attract repository intrusion. Intrusion into the repository to purposely induce an explosion cannot be casually dismissed.

In addition to the supportive work at Savannah River referred to above, our work was extended by Choi and Pigford[8,9] who show that commercial spent fuel and MOX spent fuel both exhibit positive feedback underground even without separation of the ^{239}Pu from the uranium, the cladding, or the fission products. Furthermore, Choi and Pigford showed that the multipurpose canister (MPC) proposed for storage of the fuel, even when constructed as planned with boron neutron absorber in the steel, would become critical if the vacant space filled with water. If the MPC and its spent fuel were to disperse into the surrounding rock or backfill, this rearrangement would have positive feedback as well. It might not be an overstatement to say that many configurations for underground emplacement of fissile material planned before the appearance of our paper probably have the potential for reconfiguration by natural means into critical conditions with positive feedback.

II. The UC Berkeley Nuclear Engineering Department Study

Perhaps the most thorough study spawned to date by our work was that conducted by the University of California Berkeley (UCB) Nuclear Engineering Department[5]. This group intended to examine our criticality calculations including positive and negative feedback, the probability for forming the critical configurations, and the nuclear yield of supercritical configurations. Our calculations on wet underground criticality and positive and negative feedback were examined and confirmed by the study. The study also showed that plutonium in the form of metallic ingot-like configurations would exhibit the highest degree of positive feedback owing to the presence of self-protection in thick pieces. This was recognized early on in our Los Alamos work, but was not reported because plutonium was unlikely to be placed underground in this form and we could not see how such pure and thick configurations of plutonium would collect by natural or induced means from underground storage of plutonium. The issue, however, is of importance for interim storage of weapons plutonium and this subject is addressed later in this report.

The main body of the UCB report is devoted to the analysis of migration and reconfiguration into a critical mass. Reconfiguration may be analyzed in terms of near-field and far-field migration where near-field means movement within a few meters of the original emplacement and far-field means migration over much longer distances. The UCB paper purports to show that far-field migration of plutonium requires a time much longer than the lifetime of plutonium ($T_{1/2}$ = 24,000 years) so that the material ends up migrating as ^{235}U. For the case of commercial spent fuel, the ^{238}U sufficiently dilutes the ^{235}U formed from ^{239}Pu so that critical configurations are unlikely. Weapons plutonium could be stored with a large amount of depleted uranium around it so that after the plutonium decayed to ^{235}U, it would again be mixed with ^{238}U. The present author (Bowman) believes that the plutonium can migrate faster than the Berkeley study assumes by being carried as a colloidal suspension of surface water making its way through fissures in the fractured rock which collect together to form streams which eventually reach the water table. The collected water from fissures may also be commonly seen as springs in wetter country, and springs also are found in the vicinity of Yucca Mountain. This phenomenon would collect the plutonium at the point where the plutonium reaches the water table where it would build in concentration perhaps reaching criticality. The build-up to criticality in the presence of high water concentration results in a strongly positively fedback condition.

Near-field reconfigurations to criticality are not considered carefully by the UCB study. The present author believes that the near-field situations are the most likely to exhibit reconfiguration to criticality. In summary, the UCB work is a useful effort but it is by no means exhaustive. While such studies of far-field migration might be extended, it is not clear that such studies will ever be able to usefully quantify the probability of the formation of positively fedback critical situations over periods of hundreds of thousands of years.

The UCB study also purports to present useful results on the nuclear yield from positively fedback configurations. Since there is no capability in the UCB for such studies, the UCB team tasked Lawrence Livermore National Laboratory investigators to produce some useful estimates using their weapons codes. The results of this work were nuclear yield estimates in the range of a few hundred tons of high explosive equivalent which is close to estimates in the report[1] which preceded publication. The present author questions the usefulness of these nuclear weapon codes which were constructed to calculate the generation of energy on a sub-microsecond scale of nuclear weapons rather than on the few millisecond scale of interest in underground supercriticality. The Livermore codes have been benchmarked against nuclear explosions but not against the 10,000 times slower systems of concern here.

The UCB report asserts that the probability of criticality with positive feedback is very small apparently intending to imply that it is too small to be of concern, but then it goes on to recommend the implementation of further mitigation means by the study of better engineered emplacement. This appears to be tacit recognition of the imprecise nature of such studies. The present author believes that the confirmation of our criticality calculations was the most useful result of their work. The work on material migration, nuclear yield, and yield confinement was interesting as a base for further work. The UCB effort, which was funded by the Los Alamos National Laboratory[10], might have been influenced unduly by the sponsor and by the advocates of geologic repository storage of nuclear waste.

III. Outstanding issues on supercriticality with repository storage
There are a number of issues on underground supercriticality deserving further study. The purpose here is to discuss them briefly and to suggest the most useful approach to these studies.

A. Near-field and far-field migration
The prediction of near- and far-field migration are perhaps the aspects of the underground supercriticality problem which are the most difficult to address with beneficial results as stated already above. However probably more could be done to address (1) the competition of water flow through rock pores compared with flow through fissures in the rock, (2) the disposition of surface water in summer and winter, and (3) the presence of decay heat and its role in drawing water up from the water table and driving an upwelling and down-return circulation of water through or around the waste. The present author expects that the value of such studies will be limited but worth doing.

B. Emplacement strategies
Modified emplacement strategies have been suggested[5,11] for reducing the possibility of formations of critical configurations such as using depleted uranium as the backfill or building an underground "roof" over the emplacement site to deflect the down-flow of surface water. These possible solutions, which might allow the waste to be emplaced with greater safety, will of course have to compete with the cost of destroying the fissile material by transmutation (fission) in a reactor or subcritical system.

C. Positive feedback analysis for ^{235}U
Our paper[12] includes a figure showing for ^{239}Pu k_{eff} as a function of temperature and water content in a mixture of water, rock, and plutonium. Figures also are included showing the bounds of positive and negative feedback for dry and wet systems containing plutonium. Since large amounts of highly enriched uranium spent fuel is presently destined for underground storage, it would be good to have similar analyses for ^{235}U. The 0.3 eV resonance plays a key role in ^{239}Pu, but such a resonance is not present in ^{235}U so situation for ^{235}U will be substantially different.

D. Explosion confinement mechanism
Key factors in the development of substantial nuclear yield from supercriticality underground are positive feedback and confinement by the rock. These obviously are not present for above-ground unconfined criticality. The rock confinement is a key issue since a supercritical system underground will continue to build energy until the system turns itself off by the increase in temperature or by expansion. If the rock prevents any expansion at all, only the temperature can operate and the energy developed can be enormous.

In our first work[1], we took the phase change to stiskovite as the relevant pressure. This happens at a very high pressure of 30 GPa and confinement by such a pressure would result in a substantial yield. Early evaluators of our paper argued instead that the lithostatic pressure of the rock above the waste was the relevant pressure[11]. This pressure is of course orders of magnitude lower than the phase-change pressure and would allow only a quite modest nuclear yield. However, even though the energy generation e-folding time is only of the order of one millisecond, this time is much too short for a sound wave to travel to the surface and back to tell the system where the surface is. The actual confinement strength appears therefore to be the compressive strength of the rock. This is the pressure at which a specimen of the rock in the shape of a right circular cylinder gives way when pressed on its flat surfaces placed between two flat plates without confinement on the circular surface. Of course the rock underground is confined in all directions. When the circular cylinder is confined on the radius by the compressive strength as determined above, the compressive strength is increased typically by a factor of about five. For tuff this confined pressure is about 250 MPa; the pressure may be as much as a factor of ten higher for stronger rock such as basalt[14]. It seems probable therefore that the minimum yield would come from an explosion in sand or gravel.

It is interesting to note that the compressive strength underground of rock giving way in the presence of nuclear explosions has been found not to differ much from one rock type to another in spite of this factor of ten difference in static compressive strength[13]. The present author suggests that the reason for this is that for an ordinary nuclear explosion the expansion of the rock is preceded by a shock wave which crushes the rock and destroys its integrity before the slower developing pressure from the gas is felt by the rock. The compressive strength of crushed rock (sand) is not expected to vary much from one species to another. The underground supercriticality events we discuss proceeds too slowly to produce a shock wave. Therefore the measured compressive strength with confinement on the circular surface appears to be the relevant confinement pressure which defines the yield.

E. Yield Vs fissile mass and volume

Our thesis[1] is that systems underground which contain fissile material can become critical with positive feedback which will lead to an explosion. When the pressure grows to the point that it begins to exceed the compressive strength of the surrounding rock, the rock will give way and allow the gas ball to expand until the pressure decreases to the compressive strength of the rock. The equations of state of the gaseous rock gives the temperature at which the compressive strength is reached for a given density of the gaseous rock. For granite with a compressive strength of 1.25 GPa, the temperature at this pressure for a density of unity is[15] about 0.35 eV. The reactivity will decrease as the temperature increases and as the volume increases. However since the confinement pressure is fixed, the temperature and the volume for a system are not independent. Therefore the yield will depend on the amount of fissile material, the volume of rock through which the fissile material is distributed, the maximum degree of excess reactivity which is generated by the positive feedback and the final *volume or temperature* but not both. In considering the safety of storage, the spread in yields, influenced by things such as heat-driven water transport, is of substantially less interest than the maximum yield possible. It would be useful to generate this information for both ^{239}Pu and ^{235}U using the maximum value which k_{eff} could reach under the most favorable conditions.

F. Confinement, venting, or explosion

After the generation of energy ceases, the system will be left as a large confined gas ball at the pressure of the compressive strength of the rock. This static condition might be maintained by the rock until the gas slowly finds its way out of the spherical ball. The pressure might also be relieved by the gas finding its way to the surface and venting. Finally the pressure might be relieved by explosively ejecting the rock above the gas ball. One first guess about how such ejection would take place might be the ejection of the column of rock directly above and of the same diameter as the gas ball. However, since the rock is not cracked in such a cylindrical fashion, it would be necessary to cause the rock to shear and then to slide against the fixed rock as it is ejected. The actual pressure required to shear the rock in this way is much greater than the pressure associated with the weight of the rock (the lithostatic pressure). Therefore an explosion would probably occur by the ejection of rock which is already cracked. The shape of the rock volume explosively ejected would be in the form of an inverted cone with an appropriate half-angle and the gas ball at the apex. Using a suitable model for estimating the half-angle, it should be possible to determine the depth of burial required to avoid an explosion but not sufficient to avoid venting of the hot gas. A different model for the depth of burial to avoid venting might be developed using an approach similar to that for preventing explosion ejection. These criteria could then be applied to the yield estimated Vs fissile mass and volume to develop criteria for a burial depth to avoid venting, or to avoid an explosion. The result of such a study might indicate a given depth of burial to avoid venting for emplacements containing a given amount of material distributed in a given volume. The burial depth would vary for the same amount of fissile material but with a lesser or greater volume. Trade-offs therefore could be examined such as whether to bury many smaller batches of plutonium at a shallow depth or larger and fewer batches at a greater depth.

G. Coupled explosions domino fashion.
Depending on the amount of fissile material buried in each batch, the spacing of the batches, the yield of the explosion, and the final gas ball diameter, it is possible that one explosion could trigger a similar explosion of its neighbors and therefore for the explosions to multiply domino fashion throughout the repository into a colossal event. The conditions which might lead to such an event should be examined carefully. The result might be a prescription for spacing between batches or adjustment of batch size sufficient to eliminate the coupled explosion possibility.

H. Criticality effects for commercial spent fuel and MOX spent fuel.
Commercial spent fuel and MOX spent fuel are of particular interest because they both represent uniform waste forms and they would account for the bulk of the stored high-level waste over the long term if present U. S. planning is not changed. Pigford and Choi[8,9] have shown that these waste forms will exhibit positive feedback, but the yield from such feedback has not been estimated. Also mitigation means have not been examined for positive-fedback criticality for this material. An extension of the Choi-Pigford work is needed.

IV. Other criticality issues from fissile material underground
The purpose of our original work on underground supercriticality was to point out the major difference between accidental or spontaneous criticality above ground and below ground. Below ground, the feedback following criticality may be positive or negative whereas above ground where the fissile material is unconfined the feedback is almost always negative. In addition the strength or mass of the rock surrounding a critical system below ground confines the system allowing the energy to build to much higher levels than is possible above ground. The concern for these positively fedback supercriticality events exists for any situation where fissile material has been placed underground. Some other situations besides Yucca Mountain deserving attention are described next.

A. Other nations' geologic repositories

Our paper evaluated the possibility of underground supercriticality using rock type and composition characteristic of that at Yucca Mountain since that is the site under investigation for holding all types of high level nuclear waste that have been generated in the U. S. Much of the response to our paper has been denial that our analysis could have any relevance to the Yucca Mountain site. It is the present author's impression that the strategy for resolution of the issue is continuing denying that such events deserve attention but to modify the emplacement strategy to significantly reduce the risk of these previously unforeseen possibilities. However this response avoids the perhaps more important issue that the U. S. policy is against waste burn-up and for implementation of permanent underground storage *the world over*. Since there are approximately 30 countries with nuclear power, each of them would have to develop its own geologic storage site if they wished to accommodate U. S. policy.

Those sites would have characteristics much different from Yucca Mountain. Most would be below the water table and they might be in clay or granite. Even if it can be shown eventually that fissile material can be emplaced so as to eliminate concerns for underground supercriticality at Yucca Mountain, this success says little about the possibilities for eliminating such criticality situations for other countries' repositories. As things now stand, the U. S. risks pushing other countries into a waste disposition system which might be made safe for us but not assuredly safe for them. For this U. S. policy to continue to make sense, it is necessary that the U. S. not only devise effective means for avoiding underground supercriticality here but for conditions in every other country as well.

B. Chernobyl recriticality

It was recently reported that detectors inside of the leaking sarcophagus at Chernobyl recorded a 60-fold increase in the neutron counting rate in 1990 following a two week period of heavy rainfall[16]. The rate stayed high for several days. The large increase in rate implies that the system was either very near criticality or it had in fact reached criticality. It was assumed that this event was caused by water leaking into the system. We showed[1, 12] that criticality reached initially this way has negative feedback so that the system is self-controlling. The monitoring scientists were greatly concerned about this problem and so finally a brave scientist[17] raced inside the sarcophagus and poured a solution of gadolinium inside and the neutron rate subsided as a result of this action. Periodically gadolinium solution has been sprayed around inside of the sarcophagus since.

The Chernobyl rubble criticality thereby came under the influence of gadolinium. Because this element has a cross section which falls more rapidly than inversely with the velocity, the rubble now would exhibit positive feedback (positive temperature coefficient) if the system went critical again. Water apparently has continued to leak into the system for the past six years and perhaps enough has leaked in (it's estimated now to be 3000 tons) that the system might be in an overmoderated condition. In that case if the water is removed, the criticality could increase and if the system became critical the system would exhibit positive feedback for a different reason than that provided by the gadolinium. The authorities at Chernobyl according to *Science* blame their criticality problems on water and would remove it immediately if they had a place for storage. It is possible that both overmoderation and the gadolinium could jointly contribute to positive feedback. It is difficult to imagine the consequences for worldwide nuclear technology of recriticality with positive feedback from the Chernobyl rubble. It would be highly desirable to find out as soon as possible whether the Chernobyl rubble is in a positive or negative feedback condition before any further remediation actions are undertaken.

C. Nuclear waste storage lakes in Russia

Much of the radioactive waste from the reprocessing of fuel to produce weapons plutonium in Russia was stored in open lakes. Russian authorities have said that for several years in the early period of plutonium production, about 15 % of the plutonium was lost to the waste stream. While the recovery fraction from the spent fuel was improved later, plutonium measured in tons might exist in these lakes. Presumably it has precipitated and mixed with the mud. The remediation planned is to remove the water and then to dig up the plutonium-contaminated mud for proper handling. We show in our paper[1,12] that if the plutonium concentration in the lake bottom mud is high enough (for example a mole fraction of 0.0004 in mud containing 70 % water), criticality with positive feedback might be reached when the water is removed and the mud begins to dry. The nuclear yield per kilogram of plutonium from such an event is unlikely to be large since the amount of overburden is unlikely to be large. However nuclear technology would suffer a serious blow if a presumably well planned clean-up process went awry.

D. Bunker storage of excess weapons plutonium

The UCB analysis of our paper confirmed the existence and properties of positive feedback we described. The UCB work also called attention to positive feedback associated with highly concentrated plutonium such as plutonium distributed as thick chunks, slabs, spheres or shells. This feedback is associated with the self-shielding which exists for plutonium in the form of thick pieces and the removal of the self-shielding if the system reaches criticality and the plutonium vaporizes. The earliest draft of our underground supercriticality paper also recognized this source of positive feedback, but it was omitted from our final paper for reasons other than technical. We were pleased to have the opportunity to review the UCB report before it was finalized and made several suggestions. One of those was that this particular source of positive feedback would have relevance to the present means of storage of weapons pits. The UCB response was that it took no position on the relevance of the strong positive feedback they discuss to plutonium storage as heavily self-shielded fissile material.

The induced collapse of the storage bunkers which might contain 1000 pounds each or so of plutonium[18] would cause the concrete and earth overburden to mix to some degree with the plutonium with criticality being a possible or even likely result. Such a supercritical system would probably exhibit stronger positive feedback than any system considered in our paper. While the 4 ft. of earth and concrete overburden and side-berm might be only a few hundred tons altogether and therefore modest compared to that for storage of waste deep underground, it might be sufficient with the large positive feedback to cause nuclear energy release in the range perhaps of several tons of high explosive. Many such bunkers may be spaced close to one another. If the yield is large enough, it might be sufficient to induce the collapse of a neighboring bunker. Neither the possibility of a strongly positively fedback criticality nor the destruction of all of the bunkers domino fashion from the induced collapse of one of these is included in the Programmatic Environmental Impact Statement for excess plutonium storage which is now under review. A more accurate estimate could and should be made of the yield from such criticality.

VI. Closing comments

The present author views the UCB and the Choi-Pigford studies as a useful beginning. Much remains to be done as stated above on issues such as near-Vs far-field migration, positive feedback effects for ^{235}U, the role of compressive strength in enhancing the nuclear, the yield dependence on fissile mass and the fissile material dispersal volume, and the matters of confinement, venting, or explosion. Much of the above should be carried forward in the context of Yucca Mountain. However repository conditions in other countries will be substantially different from Yucca Mountain probably implying independent studies for each foreign site. It would be desirable therefore, if possible, to develop criteria for dealing with underground criticality which are relevant to all repositories rather than just one. This effort would be simplified by the fact that most will hold only spent commercial nuclear fuel and spent MOX fuel.

The science of underground criticality is not just relevant to waste placed in repository storage, but also to environmental remediation efforts in several contexts such as at Chernobyl, The Chelyabinsk area, and at sites for excess weapons plutonium interim storage. Perhaps the Nation will take the risk of underground supercriticality seriously enough to avoid dangers in contemplated actions involving fissile material and to avoid the necessity for future corrective measures which may be risky and expensive. After Chernobyl, nuclear technology cannot endure another unexpected nuclear explosion of even modest magnitude. We hope that our underground supercriticality paper will be viewed as a useful contribution to preventing such unexpected events.

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