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COMMENTS ON "NUCLEAR EXCURSIONS" AND "CRITICALITY ISSUES"

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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.

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