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Might Underground Waste Repositories Blow Up?

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In "Underground Supercriticality from Plutonium and Other Fissile Material," C.D. Bowman and F. Venneri [hereafter B&V] consider possible scenarios in which a subcritical underground deposit of plutonium or other fissile material might be changed into a critical configuration. As they point out, underground criticalities occurred in Gabon some 1.7 billion years ago in deposits of natural uranium at a time when the percentage of chain-reacting U-235 in natural uranium was higher (3.7 percent) than today (0.711 percent).[1]

The Gabon deposits did not explode. When the fission heat drove off the neutron-moderating water in the deposit, they went subcritical until they cooled, the water percolated in again, and the chain-reaction started again with the cycle repeating itself over and over again until the reactors finally went permanently subcritical. B&V point out, however, that, if a deposit of the fissile material were "overmoderated," the boiling off of the water would increase rather than decrease the reactivity in a manner that they term "autocatalytic." As they point out, such an autocatalytic criticality did occur in the 1986 Chernobyl reactor accident.

The articles published here accompanying and commenting on the B&V paper do not contest that an autocatalytic criticality could occur if fissile materials were arranged in the configurations that B&V describe. However, the article by a group of Livermore scientists, "Comments on the Draft Paper, 'Underground Supercriticality from Plutonium and Other Fissile Material' by C.D. Bowman and F. Venneri" asserts that it is virtually impossible that such a configuration could develop in an underground depository. And the article by Robert Kimpland, "Dynamic Analysis of Nuclear Excursions in Underground Repositories Containing Plutonium," concludes that the energy release would be so small that it would merely heat up a small volume of the repository — not cause a nuclear explosion with a yield equivalent to hundreds of tons of

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high-explosive as estimated in the original draft of the B&V article,[2] or even the smaller but still potentially disruptive energy release estimated in the version of the B&V article published here.

The two central issues raised by the B&V article are therefore:

(i) Could a subcritical configuration of fissile material in a geological repository realistically be rearranged by natural processes to become autocatalytically supercritical?

(ii) Could such a supercritical configuration release enough energy to destroy the integrity of the repository?

COULD A SUBCRITICAL CONFIGUARTION OF FISSILE MATERIAL IN A REPOSITORY BE REARRANGED BY NATURAL PROCESSES TO BECOME AUTOCATALYTICALLY SUPERCRITICAL?

Leaching of the Waste Form

B&V take as their principal starting point a specific suggestion for underground disposition of excess weapons plutonium: mixing it into borosilicate glass "logs" about 3 meters long and 0.6 meters in diameter, such as those which are to be produced by the Defense Waste Processing Facility at the DoE's Savannah River site in South Carolina. They point out that the neutron absorbing boron in the glass is much more easily leached than the plutonium and consider possible criticalities if the boron were completely leached away and the resulting the plutonium-silica mixture began to disperse into the surrounding rock. According to their figure 1, on the order of 100 kg of Pu-239 mixed into a dry SiO_2 sphere imbedded in an infinite SiO_2 medium will go critical if the radius of the sphere is sufficiently large (more than about 0.5 meters).

This theoretical possibility is well known, however, and could probably be dealt with, if necessary, by adding other less leachable neutron absorbers to the glass.[3] Indeed, as the Livermore group points out, there is a considerable literature and associated licensing requirements on the possibilities of underground criticality in a radioactive waste depository. They argue that it is well within the capability of the current state of the art of waste-form and repository design to reduce the probability of any underground criticality to insignificant levels.

Migration or Dispersion of the Plutonium

Wasteforms such as borosilicate glass are designed to have a long life — on the order of the half-life of Pu-239 (24,000 years) under the low-water-flow conditions proposed for radioactive waste depositories. Therefore, by the time Pu-239 in a waste form was released and had traveled any significant distance, much of it would probably have decayed to U-235. But U-235 too can go critical.

The Livermore group points out that critical concentrations of Pu-239 or U-235 oxides could not build up as a result of migration in solution, because of their combinations of low solubility and not-very-high partitioning ratios between water and rock. (The very high concentrations of uranium found in some deposits were apparently precipitated there in a reducing environment. In most proposed repository locations, such as Yucca Mountain, the environment is oxidizing.) The Livermore group acknowledges that such concentration limits would not apply to plutonium being carried by a glass-derived colloid but points out that the mobility and distribution of such colloids could be controlled by the choice of burial medium. Immediately around the burial form, this would be determined by the backfill. Further away, it would be dependent upon the geological location.

In the version of their paper published here, B&V emphasize that the plutonium might be dispersed from the original deposit by repeated water-moderated criticalities until a radius is achieved where dry autocatalytic criticality can be sustained. This scenario appears more plausible than the migration and reconcentration scenario and, in the absence of high confidence that the neutron poisons will not be separated from the plutonium, might impose constraints on the plutonium loading of individual logs and on the spacing of the buried logs.

COULD A SUPERCRITICAL CONFIGURATION RELEASE ENOUGH ENERGY TO DESTROY THE INTEGRITY OF THE REPOSITORY?

This is the central point of the B&V paper. Specifically, B&V examine the case of a critical configuration of about 100 kg of plutonium uniformly dispersed in a sphere of SiO_2 with a radius of about 1 meter. According to their scenario, the plutonium will heat up and vaporize the associated rock and the resulting gas bubble will expand. As the vapor expands, the reactivity will first increase and then decrease (see their figure 5[4]), becoming subcritical when a radius of about 2 meters is achieved. Creating a cavity with a radius of 2 meters in solid rock would require large pressures which would require large energy releases

estimated at about 80 tons high-explosive equivalent by B&V — although the energy release would be slow enough so that only a relatively small fraction would be converted into a shock wave. B&V suggest that the greatest hazard would be that the high-pressure gas containing most of the fission energy might vent, carrying plutonium to the surface.

Robert Kimpland in his article, "Dynamic Analysis of Nuclear Excursions in Underground Repositories Containing Plutonium," estimates, making assumptions that he considers conservative, an energy release about an order of magnitude lower than B&V. The principal physical reason for the difference appears to be that Kimpland does not assume that the rock confines the plutonium-containing gas but rather that the gas expands relatively freely through cracks and pores in the rock at an average radial speed of about 150 meters/second — about one third of the molecular velocity of plutonium at 3000 K. Kimpland then calculates the fission energy release from the expanding gas during the period that it is supercritical. Assuming that the heat released is absorbed in a sphere of rock 1.5 meters in radius, he finds an average temperature increase for the rock of only 1250 °C, about half the vaporization temperature of SiO_2. Thus, virtually all of the fission energy would be left as heat which would slowly diffuse into the surrounding rock.

Kimpland argues that his assumption that the plutonium-containing vapor can expand into the rock is plausible since the events which would have dispersed the plutonium into its supercritical configuration in the first place would have "turned the original glass log and the surrounding rock to rubble." It would be desirable to have a model for the expansion of the plutonium-oxide vapor which would calculate the radial expansion velocity as a function of crack density, size and curvature, taking into account condensation of plutonium-oxide on the crack surfaces. Comments in the Livermore review suggest that an expansion as rapid as assumed by Kimpland may be implausible. However, the perfect confinement assumed by B&V is also implausible.

CONCLUSIONS

The set of papers published here represent just the beginning of the more serious analysis of the possibilities of underground criticalities as a result of the burial of fissile materials. B&V have not proved that the underground disposal of fissile material is unacceptably hazardous. There are many measures that can be taken in the choice of geology and design of the disposal matrix, packaging and backfill to reduce the likelihood of conditions that could lead to a criticality. And, even if a supercriticality were to occur, the resulting hazard

might, as Kimpland argues, be negligible. However, the critics have thus far not produced a treatment of all the phenomena involved thorough enough to lay the issue to rest. At this point a treatment that can legitimately claim to be definitive remains to be done.

In the meantime, the scientific and political debates over the burial of radioactive waste will continue to interact strongly. The publicity about the early B&V conclusions before peer review may have unnecessarily alarmed the public.[5] However, the result has been to decisively put the issue of possible criticalities in underground fissile-material repositories on the policy "map." Some analysis of these possibilities may already be a licensing requirement for geological repositories. However, as this set of papers illustrates, a great many uncertainties remain in such calculations. The political flap over the B&V paper will therefore hopefully have a positive effect in forcing the development of more sophisticated analyses.

NOTES AND REFERENCES

1. The percentage of U-235 in the earth's natural uranium has been declining since its original creation in a supernova because its half-life is shorter (0.71 billion years) than that of non-chain-reacting U-238, the other major isotope in natural uranium (4.5 billion year half-life).

2. C. Bowman and F. Venneri, "Underground Autocatalytic Criticality from Plutonium and Other Fissile Material," (Los Alamos National Laboratory, LA-UR-94-4022 (draft), March 1995.

3. See e.g. Management and Disposition of Excess Weapons Plutonium (Washington, D.C.: National Academy Press, 1994), pp. 191-192.

4. Note, however, that the neutron multiplication factors (k_{eff}) shown in figure 5 are for a constant temperature of 25°C. As is shown along the zero-water side of the 3-dimensional plot in figure 2, k_{eff} decreases with increasing temperature.

5. Gary Taubes, "Blowup at Yucca Mountain," Science 268 (June 30, 1995), pp. 1836-9.