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Comment on the Bowman and Venneri Analysis

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As one of the authors of two reports dealing with the urgent problem of disposition of excess weapon plutonium[1,2] I have followed the work of Bowman and Venneri [hereafter B&V], since our two primary approaches to WPu disposition end up with much or all of the WPu in the mined geologic repository. In the option for burning WPu as MOX in reactors of existing type, fuel bundles containing some 3 percent spent WPu[3] would eventually be entombed in heavy steel disposition casks after a decade or more of storage above ground. Since only about 2 tons of WPu would be loaded into reactors each year, in comparison with some 100 tons of U-235 into U.S. reactors annually, it is likely that only a single fuel bundle, of 5 in a typical steel disposition cask, would be spent mixed-oxide (MOX) fuel. The MOX bundle would contain some 9–18 kg of Pu, typically about 60 percent fissile isotopes.

The other mainline program would vitrify excess WPu in 2-ton borosilicate glass, stainless-steel-encased "logs" that would contain about 20 percent by weight highly radioactive fission products, with about 1 percent by weight WPu — likewise ending in the mined geologic repository after some years of cooling above ground. It remains to be determined what neutron absorber would be used to supplement the rather soluble boron of the glass; and the U.S. Department of Energy is not committed to implementing the 1 percent solution. In view of the wide public attention[4] given to the B&V work in its earlier form[5] which stated: "Yields around 300 tons of high explosive equivalent may be possible for spherical configurations containing about 100 kg of TFM (thermally fissile material)...," it is worthwhile explaining further the distinction between energy release and explosive effect.[6]

B&V assert that neutron poisons can be removed by groundwater in the course of time, leaving plutonium or other TFM dispersed in a very good neutron moderator, so that even a modest amount of TFM could become critical. Alternatively, they argue that Pu could be transported by groundwater and gradually concentrated in a similar configuration.

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Then they identify several situations that are "autocatalytic" so that once the system has exceeded prompt critical the fission heat could increase the degree of criticality by driving off excess water, by raising the temperature, or by dispersing TFM to reduce self-shielding.

B&V maintain that an underground repository is very different from similar accumulative accidents occurring on the surface, because the strength of the rock, or the lithostatic pressure prevents easy disassembly. They calculated the yield, initially, assuming that the reaction is quenched only when the surrounding rock suffers a major phase transition and density increase at a pressure of 30 GPa (0.3 megabars or 300,000 atmospheres). This then allows the expansion of the TFM-bearing region, increasing the neutron escape probability and thus bringing the system into a subcritical range.

But the authors note that even for k = 1.1 the yield increases by a factor *e* in one millisecond, while "the time for sound to move one meter in SiO_2 is about 200 microseconds so that a one meter radius system can adjust itself fairly well to the increasing energy deposition by fission without shock effects."

So these calculations are made in the quasi-static mechanical regime. This is very different from the case of underground nuclear weapons testing, in a strong-shock regime, where the strength (or weakness) of the rock is irrelevant. The slow energy release considered by B&V, on the other hand, is analogous to "hydrofracture", in which fluid under pressure is pumped down a well into a rock formation. For anisotropic stress, hydrofracturing can be used intentionally to introduce cracks in the rock, but even for isotropic lithostatic pressure, the injection of fluid so that fluid pressure at the base of the well exceeds the lithostatic pressure is likely to break the formation.

Let's see how much energy release would be required to make a cavity of about 200 cm radius at a depth in rock of about 300 m. With a rock density of 2.2 g/cm^3, the lithostatic pressure at 300 m depth is some 66 atm (or 66 bars or 6.6 MPa). So the order of magnitude of the work required to create this volume against lithostatic pressure alone is PV, where V is about 30 cubic meters. PV is thus about 200 MJ.

How much energy is required to produce this pressure depends on the equation of state (EOS) of the material. For Nevada tuff, the pore volume is about 20 percent so that if the pores are half filled with water, there is about 0.1 g/cc of water in the rock. A mole of water is thus contained in 180 cubic centimeters of rock; if this were all vaporized as a gas near the boiling point of water, it would correspond to a pressure of about 170 bar in 180 cubic centimeters of free volume. So to obtain the 66 bar that will produce the assumed

30 cubic meters cavity volume in the rock subject to that same lithostatic pressure, one needs to vaporize some 1.05 tons of water at a heat of vaporization of some 2.8 GJ/ton, for a total energy requirement of about 3 GJ.

The fission heat would thus be about 3/4 ton HE but the mechanical energy (communicated to the rock support) would be only 200 MJ/(4 GJ/ton) = 0.05 ton HE. So in wet rock a total heat release of about one ton HE-equivalent would be enough to produce steam pressure equal to the lithostatic pressure to create a cavity large enough to render 100 kg of Pu subcritical.

It is possible in principle that the rock is so very porous that the steam produced at this rate (within a millisecond or so) can simply whistle through the rock without producing significant body forces, in which case there will be little mass disassembly due to the energy release. Experiment and additional calculation can resolve this question.

It might be imagined that the plutonium is rather concentrated in veins within the "sphere" so that the rock is not significantly heated and the Pu itself is vaporized. Indeed, TFM in veins constitutes still another autocatalytic mechanism, since the self-shielding of the TFM diminishes with increase in neutron temperature, and a system of TFM and dispersed thermal absorbers would become more reactive with increase of temperature. The transport of the vaporized Pu would need to be considered, as well.

Creating volume for expansion by exceeding the lithostatic pressure may be too slow a process, but crushing the rock by exceeding its compressive strength is much faster, although one would not gain the entire pore volume[7]. The mechanical energy required to gain volume V at a crushing pressure P is PV. To gain 10 cubic meters of volume at crushing pressure of 50 MPa would involve 500 MJ of mechanical energy; if yielding takes place at 250 MPa, the mechanical energy would be 2.5 GJ, or about 0.6 tons of high explosive equivalent. The 60 grams of fission products produced from even a 100 ton energy release is very little radioactivity compared with the 400–100 kg of fission products emplaced with every 100 kg of Pu in spent reactor fuel.

My own judgment is that the consequences of supercriticality in any Yucca Mountain emplacement of 1 percent WPu in borosilicate glass, or 4 percent RPu in spent MOX fuel[8] will turn out to be negligible, in that the kinetic energy or true "explosion" aspects will be found to be small, and gas will dissipate within the mountain rather than coming to the surface. These, however, are only conjectures on my part, and more work should be done.

No matter how real or unreal the prospects for explosive criticality in the mined repository, they will not occur without differential leaching and transport by water. And Pu-239 will no longer be a concern after about 50,000 years, while the U-235 in non-MOX LWR spent fuel is only about 1 percent in U-238.

Moreover, as noted in a 1993 paper[9] there is the possibility of engineered diversion systems; and the authors cite a USGS proposal to use ceramic umbrellas to divert water from waste packages, and also waste-package covers consisting of conical layers of gravel and sand. Indeed, an underground analog of a sloping, layered tile roof, with the tiles made of ceramic or granite on crushed-rock support, combined with sand and gravel differential-capillarity barriers seems to me a useful approach to prevent underground water access to the individual waste packages.

Evidently, DOE should task at least one of its laboratories or contractors to create a flexible computer simulation program that could model the autocatalytic process, to replace these conjectures by numerical results, for various assumed environments.

NOTES AND REFERENCES

1. W.K.H. Panofsky (Study Chair), "Management and Disposition of Excess Weapons Plutonium," Report of the National Academy of Sciences, Committee on International Security and Arms Control, (National Academy Press, Washington, D.C. 1994).

2. J.P. Holdren (Chair), "Management and Disposition of Excess Weapons Plutonium:" Reactor-Related Options, Report of the National Academy of Sciences, Committee on International Security and Arms Control, Panel on Reactor-Related Options for Disposition of Excess Weapons Plutonium, (National Academy Press, Washington, D.C. 1995).

3. The pencil-thin fuel "rods" are zirconium alloy tubes containing PuO_2/UO_2 ceramic cylinders.

4. W.J. Broad, *The New York Times*, March 5, 1995.

5. Los Alamos report LA-UR-94-4022A, March 1995.

6. The energy release of high explosive is taken as 4.2 MJ/kg.

7. Because grains do not pack without voids.

8. Which after 50,000 years would contain about 3 percent U-235 formed from the decay of Pu-239.

9. E.C. Taylor, L.D. Ramspott, and W.M. Sprecher, "Demonstration of Safety for Geologic Disposal" in *Scientific Basis for Nuclear Waste Management XVII*, edited by A.Barkatt and R.A. Van Konynenburg (Materials Research Society, Pittsburgh, PA) pp. 199-207.