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AUTHOR(S): Dr. Charles D. Bowman

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Los Alamos Los Alamos National Laboratory
Los Alamos, New Mexico 87545

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Eliminating the Possibility at Chernobyl 4 of Recriticality with Positive Feedback

Charles D. Bowman
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The magazine *Science* carried an article in its latest issue entitled, "The Explosion That Shook the World" describing the status of the Chernobyl reactor core and the structure built for protection and containment. A substantial part of the article described efforts underway to monitor and avoid recriticality. We have recently published an article, "Underground Supercriticality from Plutonium and Other Fissile Material," in *Science and Global Security* Volume 5, p. 279 (1996) in which we discuss means by which plutonium and other fissile material stored underground could reach criticality with positive feedback and therefore explosive potential. This paper created a great deal of controversy because it introduced concepts which were not current practice into the little studied science of criticality for underground configurations. While there are still some reservations about our work after one year, the insights we have brought to this subject are generally accepted by those who have studied this work closely. The probability of the events described is said to be low but nobody seems willing to say whether they are acceptably low. In any case the study of remedial actions are recommended.

The Chernobyl rubble involving hundreds of tons of material is similar in some respects to the systems analyzed in the paper. I therefore read the science article with concern because the practices there to control criticality may well increase the probability of a second event at Chernobyl 4.

The only input I have on this is from the article. The key points of the story summarized briefly are as follows.

1. The explosion at Chernobyl 4 left a mass of fuel combined with graphite, concrete, water, and the materials dumped on the facility by helicopter. The configuration is uncertain as is the amount of fuel which remained inside the sarcophagus. There are a few means for access inside the sarcophagus and one of these was a room partly under the fuel mass. Neutron and gamma ray detectors were placed there.
2. The sarcophagus roof has been leaking for some time and it is estimated that 3000 m^3 of water has mixed with the fuel. There was therefore a concern about possible recriticality. The neutron monitors were in place to observe any increase in k_{eff} and criticality itself if it occurred.
3. After two weeks of heavy rainfall in 1990, the neutron detector rate increased by a factor of about 60 and stayed there for several days causing great concern to the scientific oversight team.
4. Finally a member of this team carried in a solution of gadolinium nitrite and poured it over the area where it appeared that the criticality might be reaching dangerous levels. Upon pouring on this solution, the neutron rate decreased as expected.
5. In order to keep the system subcritical, one kilogram of gadolinium in solution has been sprayed around inside the sarcophagus region every two weeks since that time. In the meantime the roof has continued to leak.

6. The science team at Chernobyl believes that the criticality problem would disappear if the water could be removed. Their intention therefore is to pump it out as soon as a place can be found to store it.

Criticality or near criticality

The increase of neutron rate by a factor of 60 is a strong indication that near-criticality or criticality was reached in 1990. The neutron multiplication is given by $1/(1-k_{\text{eff}})$ and the ratio of initial to final multiplication is $(1-k_f)/(1-k_i)$. If there were no multiplication at all initially ($k_i = 0$), to achieve an increase in neutron rate by a factor of 60 would require $k_f = 0.983$. It is unlikely that the true k_f could have risen from zero to 0.983 simply by the addition of the water. It is much more likely that the k_{eff} of the fuel in the rubble was near 0.9. In that case to reach a multiplication by 60, k_f must reach 0.9983. So the system was almost certainly very near criticality. I'm sure that the scientists on the site were aware of this and that this concern prompted the remedial action involving the gadolinium.

The *Science* article stated that the neutron rate remained stable at the high level for several days. If the system reaches criticality with negative feedback, additional heat would warm the fuel and moderator, the water would expand, the moderator effectiveness would decrease and the reactivity would start to decrease. But next the moderator would cool and the reactivity would increase. The net response to all of this is that the neutron output would remain fairly stable. These same principles operate in a reactor to keep the power level stable. The stable neutron rate strongly suggests that the system actually reached criticality and that k_{eff} was kept from rising higher by a negative temperature coefficient most likely associated with the expansion of the water as it heated. Such feedback would have required at least a few degrees rise in the temperature of the system which must involve at least several tons of material. The fission power therefore must have been in the range of 10 to 1000 KWt. If k_{eff} were not quite one and if k_{eff} was changing, the neutron rate would change also by a large amount in response to a very small change in k_{eff} . For example if k_{eff} had reached 0.99 and dropped back to 0.98, the neutron rate would have decreased by two. If k_{eff} had increased from 0.99 to 0.995, the neutron rate would have doubled. The fact that it increased by a very large factor (60) and then became stable strongly suggests that the system actually went critical with negative feedback in 1990.

Remedial steps with gadolinium

The gadolinium was added "in the heat of the moment" to reduce the reactivity and the action succeeded. At that point the system was held subcritical by the gadolinium, but it was an unfortunate choice. The absorption cross section for Gd decreases as the neutron velocity increases but it decreases much more rapidly than inversely with the velocity of the neutron. The reason for this is that gadolinium owes its large thermal absorption cross section to two near-thermal resonances. The isotope ^{155}Gd has a resonance at 0.0268 eV (0.0254 eV is 20 °C thermal energy) and the more important isotope ^{157}Gd has a resonance at 0.0314 eV. Both resonances lie below the boiling point of water. Since fission cross sections decrease roughly inversely with neutron velocity, a rise in temperature of the moderator as a result of fission heat generation will result in an increase in reactivity with increase in fission power. This positive feedback is unacceptable because it could lead to uncontrolled energy release. Whether the feedback is positive or negative is crucial in that criticality with negative feedback in such a system is bothersome but benign in some respects whereas one must expect serious consequences from positive feedback.

Since the addition of the gadolinium reduced the neutron rate, it is clear that the reactivity of the system was brought under control by the gadolinium. Gadolinium has been added every two weeks since. Of course some of the gadolinium dissolved in water might have

been carried away by water leaking away from the region of the fuel. Since additional water has leaked into the system over the six years since 1990 and gadolinium has been added, it is no longer clear that the reactivity is under the control of the gadolinium. It might be under the control of the water in the sense of overmoderation.

Undermoderated and overmoderated systems containing water

The fact that the k_{eff} increased and that the system apparently went critical with negative feedback under the addition of water implies that the probable criticality event in 1990 was of an undermoderated nature. In an undermoderated system the addition of more water increases the neutron moderation enhancing the fission cross sections seen by the neutrons. In an overmoderated system the addition of water decreases the reactivity because the neutrons are already sufficiently well moderated by the water and other material already present so that the additional water does not enhance the moderation. On the contrary, the additional hydrogen in the water increases the neutron absorptive properties of the system. While the system has been held subcritical first by the gadolinium over the past six years, rains might have transformed the system since from an undermoderated one to an overmoderated one.

The overmoderated condition may be one of strong positive reactivity feedback, since fission heat can decrease the water density when the temperature is below the boiling point and decrease it further when the boiling point is reached or exceeded. Reduction in the water content reduces the neutron absorption of the water which increases the reactivity. If the system contained gadolinium and was in an overmoderated condition as well, the positive feedback from both effects would be doubly strong.

Remedial steps by water removal

Scientists at Chernobyl and their advisors blame the water for the reactivity problems, and would like to remove it. However a suitable storage place for this radioactive water with volume estimated at 3000 m^3 is not available. If the system is undermoderated, there is little danger in pumping away the water. If it is in an overmoderated condition, there is a significant possibility of reaching criticality with strong positive feedback if the water is removed. The water might be pumped away very slowly while the reactivity is monitored by measuring the neutron rate. If the reactivity goes down as the water is removed, this would show that the system was in an undermoderated condition. If the neutron rate increases as the water is removed, it might signify that the system is overmoderated and that the removal of the water is not a practical solution. Another danger in removal of the water is a sudden slumping of the fuel resulting from some of the fuel weight no longer being carried by the buoyancy of the water or from other effects. A large introduction of reactivity by such an event even with negative feedback could have serious consequences.

Suggestions for risk reduction

The proclivity toward positive feedback resulting from the addition of the gadolinium should be corrected if possible. Perhaps the best way to accomplish this would be to replace or overwhelm gadolinium with cadmium, perhaps Cd Cl_2 . It appears that if one added as much absorptive power of Cd as that from the Gd already present, the net effect would be a negative temperature coefficient for the system with respect to the neutron absorbers. Since the thermal cross section for Gd is 20 times larger than that for Cd, 20 times as much Cd must be added to the system as Gd. Of course one must be concerned that this poison replacement or mixing approach might fail to some degree since the Cd added might not reach the same part of the rubble as was reached by the Gd. Whether to remove the water is a difficult issue complicated by the uncertainty of whether the system is overmoderated or undermoderated and by the possibility of sudden slumping caused by the physical changes associated with the reduction in the water content.

Closing comments

It is of the greatest importance to learn whether the feedback, if criticality were reached, would be positive or negative and to take appropriate steps to assure negative feedback. The addition of the gadolinium certainly provided a source of positive feedback and compensating by adding cadmium should be considered. The continued leakage of the sarcophagus over the past six years since the first near critical or critical condition occurred might have increased the water content to the point of overmoderation with positive feedback potential from that as well. The avoidance of positive feedback is of course a requirement in all modern deployable nuclear reactor designs, and the same conditions should be established if at all possible for the Chernobyl 4 rubble.

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