CRITICALITY SAFETY WHEN USING GADOLINIUM AS A NEUTRONIC POISON WITH PLUTONIUM

R. H. Condit
J. S. Pettibone

September 1995

This is an informal report intended primarily for internal or limited external distribution. The opinions and conclusions stated are those of the author and may or may not be those of the Laboratory. Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.
DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial products, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

This report has been reproduced
directly from the best available copy.

Available to DOE and DOE contractors from the
Office of Scientific and Technical Information
P.O. Box 62, Oak Ridge, TN 37831
Prices available from (615) 576-8401, FTS 626-8401

Available to the public from the
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Rd.,
Springfield, VA 22161
DISCLAIMER

 Portions of this document may be illegible electronic image products. Images are produced from the best available original document.
Criticality safety when using gadolinium
as a neutronic poison with plutonium

Ralph. H. Condit and Joseph. S. Pettibone

Abstract

We have discovered that under special circumstances it is possible to have a criticality accident which can release tons of high explosive energy equivalent. We report a computer calculational experiment which illustrates this point so that the circumstances which might lead to such an accident can be avoided.

Our computer calculation gives the energy release in a criticality event involving a water solution of plutonium where (1) gadolinium has been used as a neutron poison and (2) the mass is then allowed to go slightly supercritical at room temperature. The reactivity has a strong positive temperature coefficient in this system. The result is that the self-heating from a small amount of fissioning of the plutonium in the delayed supercriticality regime raises the temperature to the prompt criticality regime and an explosive release of energy becomes possible. In this computer experiment the release is 19 tonnes (metric tons) of high explosive equivalent for a two-cubic meter sphere of water containing 50 grams of plutonium per liter.

Our concern is confined to gadolinium in moderating solutions and does not relate to any other known neutronic poison, because gadolinium is the only isotope having the special absorption characteristics that make possible this type of energy release.
This page intentionally left blank
1. Introduction

This note presents the results of a computer experimental calculation of energy release in a special criticality condition that might become possible when gadolinium is used as a neutronic poison. The results of the experiment are first presented and then discussed as they relate to possible criticality accidents. Our discussion is confined to gadolinium with its high temperature coefficient of capture cross-section and does not apply to other possible neutronic poisons which do not have this feature in the temperature ranges characteristic of fissile material processing or reactor operation.

In most criticality accidents and in the criticalities that are engineered into nuclear weapons the condition results from the movement of material. For example, two masses are brought together, a solution is poured into an oversized tank, or solids are stirred into suspension to give a geometry that supports a chain reaction. The energy release then tends to disassemble the configuration that supported the chain reaction. In this paper we direct attention to systems having different dynamics which are characterized by a positive temperature coefficient of reactivity. Such a system might be assembled at low temperature where it would be subcritical but at a higher temperature would become supercritical. Its self-heating would then drive it toward yet greater reactivity and a faster rate of neutron multiplication. Expressed mathematically,

\[ N = N_0 e^{\alpha t}, \]

where \( N \) is the number of neutrons at time, \( t \), after starting with \( N_0 \) neutrons and this number depends exponentially on a rate constant, \( \alpha \). In most criticality accidents, the value of \( \alpha \) is slightly positive but it remains fairly constant. In the situation which we discuss the \( \alpha \) depends on temperature and increases as the reaction progresses. Thus, the number of neutrons not only increases exponentially with time but the exponential factor of increase also increases with time.

The possibilities of positive temperature coefficients of reactivity and the resultant thermal instabilities are well known and have been much discussed as they relate to reactor design. (See, for example, ref. 1). Our contribution to the subject is to use computer codes developed for the design of nuclear weapon to calculate how much energy might be released. We select for our "experiment" a sphere having a volume of two cubic meters loaded with 50 grams per liter of plutonium. Without a neutronic poison this would have a
$K_{\text{eff}}$ (the ratio of change of fissions produced in the system with each succeeding neutron generation) of 1.5. Then, enough gadolinium is added to reduce the $K_{\text{eff}}$ to 1.002 at room temperature. We follow the energy release using the codes which employ (1) the best available neutron capture and fission cross-sections as a function of neutron energy (for example, ref. 2), (2) details of the energy distribution of neutrons as a function of time and position as calculated explicitly by the Monte Carlo method including thermal kinetics and doppler broadening of the resonances, (3) the equation of state of the water solution (pressure as a function of density and temperature), and (4) the kinetics of material movement as the materials are subjected to the forces resulting from the energy release.

We conclude from our computer experiment that the energy release can be larger than generally expected from criticality accidents, in the range of tons rather than pounds of high explosive equivalent. Criticality accidents in the past (there have been over thirty) have produced no physical damage outside the room in which they occurred (ref. 3). The contrast between this past experience and the possibility which we explore is suggested in Fig. 1. We want to point out that the probability of occurrence of any criticality accident today is very small because great efforts are taken specifically to avoid them. However, if one should occur under the special circumstances which we describe, the consequences could be greater than past experience might suggest. Risk is defined as the mathematical product of the probability of some occurrence times its consequence. The special type of accidental energy release which we describe can be avoided by not allowing the circumstances which might make it possible. Fortunately, there does not seem to be any current fissile processing technology which could run this risk.

It is well known that gadolinium has a neutron capture cross-section which decreases rapidly from room to higher temperatures (ref. 2). For this reason it would always be used with considerable caution in those cases where it is used at all. Furthermore, the use of gadolinium as a soluble additive to a plutonium or uranium solution would generally be recognized as bad practice (ref. 4). Nevertheless, these are just the features which we have selected for our calculation; the calculation must not be taken as reflecting actual practice. Even so, there may always be a temptation to use gadolinium in ways which could prove to be unwise, because gadolinium is attractive as a neutronic poison; its capture cross-section is very large, see Table 3, page 13 of this paper.
2. The basic physics of the process

The fission of a uranium or plutonium nucleus releases neutrons which have a mean energy of about 2 Mev. They travel at the high velocity of about $2 \times 10^9$ cm/sec, and have a mean free path in water of about 5 cm. Their velocities soon decrease in this moderator to thermal velocities, $2 \times 10^5$ cm/s (mean energies of 0.026 ev at room temperature), and the mean free path then becomes much shorter, 0.5 cm. They may experience something like 100 collisions before being absorbed by a fissile nucleus or a poison. The mean lifetime of a moderated neutron ranges around 10 - 30 μs, close to 10 μs in the system we have calculated using gadolinium. This is a short time compared with the time before disassembly, about 10 ms in moderated systems.

It is possible to capture neutrons before they initiate the fission of a U or Pu atom by use of neutronic poisons. Data for a number of neutronic poisons along with fission cross-sections for $^{235}$U and $^{239}$Pu are shown in Fig. 2. It appears that gadolinium isotopes 155 and 157 are unique in that their capture cross-sections drop rapidly with increasing temperature near room temperature and above. In Fig. 3 the ratio of the fission cross-section of plutonium to the capture cross-section of the two gadolinium isotopes is shown. It can be seen that at room temperature a small amount of $^{155}$Gd, 3×10^{-3} atom fraction of $^{239}$Pu would have the same capture-cross-sectional area as the fission cross-section of $^{239}$Pu, but greater amounts would be needed if the temperature were increased. As a result, the $K_{eff}$ would increase with temperature. In our experimental computation the temperature coefficient of $K_{eff}$ is $3.4 \times 10^{-4}$ per °C. Thus, a temperature increase of only 6°C could increase $K_{eff}$ from 1.000 to 1.002 which would be a prompt critical condition for a plutonium fuel. (Prompt criticality for $^{235}$U is achieved after $K_{eff}$ reaches 1.006.)

In a supercritical assembly produced by bringing two parts together the $K_{eff}$ will have become greater than 1.00 but does not change much thereafter until disassembly occurs. In accidents the disassembly normally terminates the chain after about $10^{17}$ to $10^{18}$ fissions have occurred, perhaps a few pounds of high explosive equivalent. There may be about the same number of neutron lifetimes before disassembly in the unpoisoned as in the Gd-poisoned systems, but the amount of material which fissions in the Gd system will be greater. In our computer experiment 3×10^{21} fissions occurred.
3. Assumptions for the calculation

For the purposes of this calculation we have assumed a sphere of a water solution of plutonium. The volume of the solution is taken as approximately 2.0 cubic meters with the plutonium concentration being 50 grams per liter. The total amount of plutonium is 100 kilograms. The plutonium is taken to be weapons grade, 94% $^{239}$Pu and 6% $^{240}$Pu. This actually contains 93% $^{239}$Pu and about 1% $^{241}$Pu, but the neutronic behavior of these two is quite similar. The nuclear data include the cross-sections for fission and capture of the two principal Pu isotopes as a function of neutron energy.

The equation of state (EOS) for pure water has been assumed. Actually, the equations for water which we have used in the calculations of EOS are not well behaved at room temperature and pressure; these parameters are too low for normal weapons calculations. Therefore we had to constrain the density to a value of 0.89 grams/cm$^3$ during the early parts of the computer run and the pressure was computationally defined as zero in order to suppress irrelevant effects which would otherwise occur during the initial slow heating stage, i.e. during the first 58 milliseconds. This still allows the neutron multiplication to progress exactly as it would in a real system. The total amount of water was 2.0 tonnes, i.e., 2.0 m$^3$ at density 1.00. The reduction in density means an expansion of the sphere as is reflected in the ordinate of fig. 11.

An iron shell having a thickness of 2 cm was placed around the sphere as a container. The neutron reflectivity and neutronic capture properties of the iron and the water were those appropriate for real materials. The masses of materials are 2,000 kg of water, 100 kg of plutonium, 1.8 kg of gadolinium, and 1238 kg of iron. The EOS and density of the iron were set to that of water for computational simplicity. The iron will act as a tamper during the expansion of the sphere. By slowing down the disassembly a little it will allow the energy generation to carry for a little longer than if it were absent. The maximum energy produced and pressures generated are sufficiently great that this extra mass of the iron has little effect on the disassembly rate and has only a small effect on the total energy release. This containment is a reasonable approximation to the confinement which might be expected in any actual system.
4. The computer program

We have used a computer program developed at LLNL. The Cray YMP computer was used. Over 30 hours of computer time were required for the calculation and the process was followed for about 1000 neutron lifetimes.

We have undertaken a 1-dimensional calculation as being appropriate for spherical symmetry. The program could be run in a 2-dimensional version, but that was not deemed necessary. The material encompassed in the calculation is 1/100th of the total sphere volume. The zones as one progresses along this sector from the center to the surface are all weighted in accordance with the volumes which they would actually include. The data presented in figs. 4-13 are all for this 1/100th of the sphere. Thus, the total energy release is 100 times that given in figs. 5-7.

Before calculating the energy release we calculated the amount of gadolinium poison required to bring the $K_{eff}$ down from ~ 1.5, what it would be without any poison, to 1.00. It turns out that a Gd/Pu atom ratio of 0.028 is required. A little over 1/2 of the excess $K_{eff}$ is recovered at the point of maximum heating.

5. Results of calculations

The results of the calculation are shown in figs. 4 - 14. In all of these figures the abscissa is time in units of shakes, $10^{-8}$ seconds. See Table 2 for explanations of the units used. The times displayed extend to $6 \times 10^6$ shakes, 60 milliseconds (ms). Captions for the figures explain the results in detail. In fig. 4 the neutron density is shown as a function of time and it can be seen that for most of the experiment it was increasing with a constant exponential growth rate, but as the temperature began to rise significantly in the time between 58 and 60 ms, fig. 7, the rate of neutron multiplication also began to rise.

The rate of energy production, fig. 5, parallels the density of neutrons. The total energy produced is shown on a log scale, fig. 6, and a linear scale, fig. 7. Note that these data are for a 1/100th segment of the sphere. The total energy produced is 0.8 jerks for this segment or 80 jerks for the whole sphere or $8 \times 10^{10}$ joules. This is equivalent to 19 tonnes (metric tons) of high explosive equivalent. This corresponds to $3 \times 10^{21}$ fissions in 100 kg of Pu, $2.53 \times 10^{27}$ atoms. About $10^{-6}$ of the plutonium atoms undergo fission.
From fig. 6 it can be seen that the principal release of energy occurred over a period of about 0.1 ms, from 58.8 to 58.9 ms. Inspection of fig. 10 reveals the way in which the pressure pulse from the interior propagated to the surface of the sphere and blew it away with the rest of the material following. The surface velocity, fig 11, reached a value of $5.5 \times 10^{-3}$ cm/shake or 5500 meters/second.

The neutron multiplication rate, $\alpha$, is shown in fig. 12. This grew to a strongly positive value at 58.7 to 58.8 ms and then became negative after 58.9 ms. The chain reaction then began to shut down. This is reflected in the $K_{\text{eff}}$, see fig. 13, which increases from the 1.002 at the start of the calculation to about 1.3 and then drops below 1.00 at the conclusion of the process.

6. An approximation formula and limitations on its applicability

It is possible to roughly calculate yields from other systems having the same geometry and homogeneous distribution of fissile material as in our computer experiment by using an approximate scaling formula. To derive this we start with the growth rate of the number of neutrons,

$$\alpha = \frac{(K_{\text{eff}} - 1)}{\tau}, \quad (2)$$

where $K_{\text{eff}}$ is the increase in fissions in each generation over that in the previous generation and $\tau$ is the neutron mean life time, in effect, the time between generations. The $K_{\text{eff}}$ in our experiment is shown in fig. 13 as a function of time. The lifetime does not change greatly with temperature, because it is the product of two terms, velocity, $v$, and fission cross-section which decreases roughly as $1/v$. However, it does depend inversely on the concentration of fissile material in the solution. Thus, the scaling relationship will include the growth rate of neutrons as being in proportion to the concentration,

$$\alpha \sim C. \quad (3)$$

The quantity, $K_{\text{eff}} - 1$, is just barely greater than zero at the beginning of the present calculation. In the system we are considering the $K_{\text{eff}}$ would be considerably greater than 1.00 if there were no neutronic poison because
the concentrations and volumes of plutonium solution will be assumed relatively large. This excess plutonium is just balanced by the gadolinium.

\[ K_{\text{eff}} \text{ (no Gd) } - 1 = \Delta K_{\text{Gd}}. \] (4)

Any slight supercriticality would lead to the generation of heat, a decrease in \( \Delta K_{\text{Gd}} \) and an increase in \( K_{\text{eff}} \) to the prompt critical regime. As noted in the discussion above, the computation shows that as the system heats up the \( K_{\text{eff}} \) increases to a value which recovers about 1/2 of the \( \Delta K_{\text{Gd}} \).

The yield, \( Y \), is going to be proportional to a term measuring the kinetic energy imparted to the mass, \( M \), (water, dissolved plutonium, and steel shell) as it disassembles with velocity, \( V \).

\[ Y \sim \frac{1}{2} M V^2. \] (5)

The velocity will be the rate of disassembly during the last generation of neutrons and should be proportional to the radius which will be expanding at that time. In other words, at the cut-off in energy production one can think of the radius as roughly doubling during a time which equals one neutron generation.

\[ V \sim R_0 \alpha. \] (6)

Therefore, combining (5) and (6) gives

\[ Y \sim M R_0^2 \alpha^2. \] (7)

The mass will be proportional to the volume, since the density of the solution will not depend strongly on the concentration of plutonium,

\[ M \sim R_0^3. \] (8)

Thus, it appears that the yield of systems, combining (7) and (8), can be related to a computed value using the equation,

\[ Y \sim \alpha^2 R_0^5. \] (9)
Combining (3) and (9) gives the yield as a function of fissile material concentration, $C$, and sphere radius, $R_o$.

$$Y \sim C^2 R_o^5. \quad (10)$$

In the following tables some possible yields are calculated using this formula together with a standardization point, our experimental result that the energy release for 2 cubic meters solution volume of 50 g/l of plutonium gives 19 tonnes explosive equivalent.

We need to point out that the formula is limited in applicability by a phenomenon which has not been discussed thus far, the absorption of neutrons by $^{240}\text{Pu}$ at 1 ev neutron energies. It turns out that this strong resonance absorption cuts off the reaction at temperatures near 5000°C and thereby limits the yield to about 10 tons explosive equivalent per cubic meter of solution. This phenomenon does in fact play some role in the final cut-off in energy production in our experiment. Very great (many kilotons) yields would not be possible. This limitation would not apply when using pure $^{239}\text{Pu}$ or pure $^{235}\text{U}$.

**Table 1: Masses of plutonium (kg)**

<table>
<thead>
<tr>
<th>Volume (meters$^3$)</th>
<th>Radius (meters)</th>
<th>Concentration (grams per liter):</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>5</td>
</tr>
<tr>
<td>1.00</td>
<td>0.62</td>
<td>*</td>
</tr>
<tr>
<td>2.00</td>
<td>0.78</td>
<td>10</td>
</tr>
<tr>
<td>4.00</td>
<td>0.98</td>
<td>20</td>
</tr>
</tbody>
</table>

* This is a subcritical configuration.

** This is the example selected for the computer calculation.
Table 2: Energy yields given by the formula, tonnes of HE equivalent

<table>
<thead>
<tr>
<th>Volume (meters$^3$)</th>
<th>Radius (meters)</th>
<th>Concentration (grams per liter):</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>5</td>
</tr>
<tr>
<td>1.00</td>
<td>0.62</td>
<td>0</td>
</tr>
<tr>
<td>2.00</td>
<td>0.78</td>
<td>0.04</td>
</tr>
<tr>
<td>4.00</td>
<td>0.98</td>
<td>1.2</td>
</tr>
</tbody>
</table>

** This is the value obtained by computer calculation

( ) Values in parentheses assume the absence of all $^{240}$Pu. When present, it limits yields to about 10 tonnes per cubic meter of solution.

7. Discussion

This computer experiment has been, in effect, the design of a nuclear explosive. It would be a very inefficient explosive, because 100 kg of plutonium would be employed to give a yield of about 19 tonnes explosive equivalent or 0.2 tonnes per kg of Pu. About one plutonium atom in a million would undergo fission. It should be clear that our "device" would only be built by accident. However, it should also be noted that the phenomena we describe might occur in any moderated system in which gadolinium was used for the marginal control of criticality, regardless of whether it was in solution or in solid materials distributed around the fissile material. Therefore, our calculation might have implications for a broader range of fissile material handling than our specific calculation might at first seem to imply, and we believe that further study of the possible problems with using gadolinium would be appropriate.

We do not plan further studies ourselves, but the computational technology is available for such a task. We look to others to judge the relevance of this experiment to situations of practical importance.

Under current practice the type of accident we suggest is highly improbable. It is generally recognized that the use of soluble poisons as either a primary or backup means for assuring criticality safety, whether gadolinium or any other poison, is questionable practice because it would always be difficult to assure that the design amount has in fact been added in
every case. It should also be pointed out that this discussion cannot apply to nuclear reactors, especially commercial reactors. They contain a large amount of $^{238}\text{U}$ which is itself a good absorber of neutrons. Even though gadolinium may be used as a neutronic shim or as a burnable poison in reactors, its function is to decrease the reactivity at a time when a fresh load of fuel would otherwise have too much reactivity. Such systems are designed to have negative temperature coefficients and our scenario would not apply.

It is possible to envision that at some time there might be economic incentives for processing fissile materials in larger batches than those presently permitted. The use of neutronic poisons might then appear to be one source of help in this regard. Where work with radioactive materials already requires special containment and removes personnel from the process area there could be arguments for allowing neutronic poisons to backup other safeguards (ref. 4). It might be assumed that a criticality, were it to occur, would not subject personnel to serious additional radiation exposures in such facilities. We are saying that in the special case when using gadolinium, the energy release might be much greater than presently contemplated and therefore its use should be questioned.

The poisons would not have to be dissolved in solutions but could be incorporated in container walls or Raschig rings. If gadolinium were used but not present in great excess the phenomena which we describe might become possible.

In another scenario, the cleanup operations following a reactor or other accident involving fissile materials might call for the use of neutronic poisons to preclude a criticality excursion in spilled materials. In the urgency of the situation there might be a temptation to use gadolinium. In such systems an initial rise in temperature might occur during some sort of chemical reaction and this could set the stage for the phenomena we describe.

8. Conclusions

We have carried out a computer experiment to suggest a possible consequence of the use of gadolinium as a neutronic poison. It might set the conditions for a criticality accident having an energy release of many tonnes of explosive equivalent rather than the few pounds experienced in such accidents to date.
This finding is restricted to gadolinium when used in a moderating system with $^{239}$Pu or pure $^{235}$U. The probability of an accident of the type which we examine is very small. Nevertheless, as a possibility, it deserved serious study.

The computer program used is one which has been developed for designing nuclear weapons and could be adapted to the analysis of many cases of more direct interest than our simple geometry. It might be worth while to extend this research, but we do not have specific plans to do so at this time.

Research on criticality phenomena is not as active now as it has been in the past, because it is generally felt that the important issues have been settled. This means that technicians entering the field at this time may become complacent. We are concerned that at some time in the future the attractive features of gadolinium may lead to its use in a situation where the then-current wisdom may not appreciate its possible dangers.

References:
Table 3: Isotope abundances in natural gadolinium

<table>
<thead>
<tr>
<th>Mass (atomic mass units)</th>
<th>% concentration in natural material</th>
<th>Capture cross-section thermal neutrons (barns)</th>
</tr>
</thead>
<tbody>
<tr>
<td>152</td>
<td>0.2</td>
<td></td>
</tr>
<tr>
<td>154</td>
<td>2.1</td>
<td></td>
</tr>
<tr>
<td>155</td>
<td>14.8</td>
<td>70,000</td>
</tr>
<tr>
<td>156</td>
<td>20.6</td>
<td></td>
</tr>
<tr>
<td>157</td>
<td>15.7</td>
<td>160,000</td>
</tr>
<tr>
<td>158</td>
<td>24.8</td>
<td></td>
</tr>
<tr>
<td>160</td>
<td>21.8</td>
<td>100.0</td>
</tr>
<tr>
<td>100.0</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Table 4: Units and conversion factors

Many of the units which we use come from nuclear weapons technology.

**Time**

1 shake = 10^{-8} seconds

**Energy**

1 jerk = 10^9 joules, = 2.39x10^8 calories

1 tonne (1000 kg) of high explosive equivalent = 4.2x10^9 joules

Energy release from fission (170 Mev/fission, prompt available energy release) = 2.7x10^{-11} joules/nucleus

**Rate of energy production**

1 jerk/shake = 10^{17} joules/second

**Temperature**

1 ev = 1.16x10^4 K, 10^3 K = 0.0862 ev, and 25°C = 0.026 ev.

**Pressure**

Jerks/cm^3 = 10^{10} bars, approx = 10^{10} atm

**Cross-sections for fission and neutron capture**

1 barn = 10^{-24} cm^2, used here as units of neutron capture or fission cross-section of atomic nuclei.

**Mass**

1 kg of Pu = 4.19 gram atoms of Pu.

1 kg Pu = 2.53x10^{24} atoms (counting 6x10^{23} atoms/gram atom).
Table 5: **Radial positions in the sphere and shell**

Radial positions in the sphere are tied to cell positions in the computer calculation. In some figures the cells are indicated by capital letters and the captions then give the numerical designator of these cell. In this table these cell positions are converted to initial radial distances from the center of the sphere. As the sphere expands and disassembles the cells move with the material in the sphere; their distances from the center increase.

Note that in all of the graphs of computer output the calculations are for 1/100\textsuperscript{th} of a complete sphere of material. This was done for calculational convenience. This means that to get the total numbers of neutrons and energy releases we need to multiply the ordinates by a factor of 100.

<table>
<thead>
<tr>
<th>Cell nr in orig cmptr calcn</th>
<th>Position of cell</th>
<th>Notes</th>
<th>Letter label given to curve in Fig 11.</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>4.55</td>
<td>a</td>
<td>A</td>
</tr>
<tr>
<td>3</td>
<td>6.06</td>
<td></td>
<td>B</td>
</tr>
<tr>
<td>12</td>
<td>19.71</td>
<td></td>
<td>C</td>
</tr>
<tr>
<td>22</td>
<td>34.87</td>
<td></td>
<td>D</td>
</tr>
<tr>
<td>32</td>
<td>50.03</td>
<td></td>
<td>E</td>
</tr>
<tr>
<td>42</td>
<td>65.18</td>
<td></td>
<td>F</td>
</tr>
<tr>
<td>52</td>
<td>80.34</td>
<td></td>
<td>G</td>
</tr>
<tr>
<td>55</td>
<td>84.89</td>
<td>b</td>
<td>H</td>
</tr>
<tr>
<td>57</td>
<td>95.17</td>
<td>c</td>
<td>I</td>
</tr>
</tbody>
</table>

Notes:

- **a**: outer surface of "cell 2" sphere
- **b**: outer surface of water sphere and inner surface of iron shell
- **c**: outer surface of iron shell
The probability of occurrence of any criticality accident is small. It has generally been assumed that the magnitude of energy release would be no more than a few pounds of high explosive equivalent. The thesis of this paper is that if gadolinium were used as a neutronic poison there could be special circumstances where the energy release would be over a tonne of high explosive equivalent. Risk is the probability times the consequence of some event. Therefore, we believe that the risk with using gadolinium in certain ways may be higher than generally recognized.
Fig. 2: Neutron cross-sections for capture and for fission.

The neutron capture cross-sections for several isotopes and the fission cross-sections for $^{235}$U and $^{239}$Pu are shown as a function of neutron energy (Data from ref. 2).
Fig. 3: The ratio of fission of $^{239}$Pu to the capture cross-section of $^{155}$Gd and $^{157}$Gd.

The abscissa is in energy and also temperature. The ordinate is cross-section in barns.
Fig. 4: The number of neutrons in the system as a function of time.

The time scale, abscissa, is in shakes. In this figure it spans from 0 to 60 milliseconds. The ordinate is the number of neutrons in 1/100th of the sphere. The data is not a smooth curve, because the concentration will fluctuate with time and the Monte Carlo calculation reflects this.
Fig. 5: The rate of energy production as a function of time.

The time scale, abscissa, is in shakes. In this figure it spans from 0 to 60 milliseconds. The ordinate is jerks per shake. Data is for 1/100\textsuperscript{th} of the sphere.
Figure 6: The total energy produced as a function of time, log scale.

The time scale, abscissa, is in shakes. In this figure it spans from 0 to 60 ms (milliseconds). The ordinate is jerks. Data is for 1/100th of the sphere. Note that the total energy produced up to 58 ms is .003 jerks for 1/100th of the sphere or $3 \times 10^8$ joules for the whole $2 \, m^3$ or an average of $1.5 \times 10^2$ joules/cm$^3$, roughly 36 cal/gram of solution.
Fig. 7: The total energy produced as a function of time, linear scale.

The time scale, abscissa, is in shakes. In this figure it spans from 58 to 60 milliseconds. The ordinate is jerks. Data is for 1/100th of the sphere. The total energy produced is 0.8 jerks for this segment or 80 jerks for the whole sphere which equals $8 \times 10^{10}$ joules. Note that the principal release of energy occurs between 58.8 and 59.0 milliseconds, a span of about 0.2 millisecond.
Fig. 8: Temperature as a function of time.

The temperatures at six different points in the sphere are given as a function of time. The units along the abscissa are shakes and span from 58 to 60 milliseconds. The temperature increase up to 58 milliseconds is small. We only present the data from that time onward when the center had reached about 150°C. The ordinate is measured in units of kiloelectron volts, kev. The maximum temperature near the center of the sphere reaches about 1.00×10^{-3} kev or 1.00 ev which is 11,600 K. The positions are given for successive distances from the center, A ... F, see Table 5. A is cell 12, B is 22, C is 32, D is 42, E is 52, and F is 55 (the outer surface of the water sphere).
Fig. 9: Densities as a function of time.

Densities of the water at six different points in the sphere are given as a function of time. The units along the abscissa are shakes and span from 58 to 60 milliseconds. The ordinate is a linear scale. Note that the initial density has been set at 0.89 g/cm$^3$ as discussed in the text. This does not affect the dynamics of the neutron chain reaction. The positions are given for successive distances from the center, A ... F, see Table 5. A is cell 3, B is 12, C is 22, D is 32, E is 42, F is 52, and G is 55 (the outer surface of the water sphere).
Fig. 10: Pressures as a function of time.

Pressures at six different points in the sphere are given as a function of time. The units along the abscissa are shakes and span from 58 to 60 milliseconds. The ordinate is in units of jerk/cm$^3$ where 1 jerk/cm$^3 = 10^{10}$ bars. The maximum pressure at the center rises above 150 kilobars. The positions are given for successive distances from the center, A ... F, see Table 5. A is cell 3, B is 12, C is 22, D is 32, E is 42, F is 52, and G is 55 (the outer surface of the water sphere).
Fig. 11: Radial positions of material points as a function of time.

The units along the abscissa are shakes and span from 58 to 60 milliseconds. The ordinate is in centimeters for the distances in the calculational model. That is, they reflect the fact that the initial density of the water was taken at 0.89 g/cm$^3$. The mass of the water was 2.0 kg. The positions are given for successive distances from the center, A...F, see Table 5. A is cell 2, B is 3, C is 12, D is 22, E is 32, F is 42, G is 52, H is 55 (the outer surface of the water sphere), and I is 57.
Fig. 12: Velocity as a function of time.

The velocity at the surface of the sphere is given as a function of time. The units along the abscissa are shakes and span from 58 to 60 milliseconds. The velocity at 60 milliseconds is about \(5.5 \times 10^{-3}\) cm/shake or 5500 meters/second.
Fig. 13: Neutron generation rate, $\alpha$, as a function of time.

The neutron generation rate is given as a function of time with the ordinate measured in reciprocal shakes. The peak alpha is $2.5 \times 10^{-4}$/shake or $2.5 \times 10^4$/second and then becomes negative after the energy pulse drops off.
The number of neutrons produced for each neutron consumed, the $K_{\text{eff}}$, is given as a function of time. As the temperature rises the capture cross-section for the gadolinium decreases and so fewer neutrons are lost from the chain reaction. After disassembly of the sphere, the $K_{\text{eff}}$ becomes smaller than 1.00 and the chain reaction dies out.