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Dynamic Analysis of Nuclear Excursions in Underground Repositories Containing Plutonium

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A recent study performed at the Los Alamos National Laboratory postulates that plutonium-239 stored in underground repositories could lead to a nuclear explosion of up to a few hundred gigajoules. The study suggests that plutonium originally contained in glass logs could escape its containment and disperse into the surrounding native rock of the repository. This dispersion would then lead to an autocatalytic process that ultimately would lead to a catastrophic nuclear explosion. A computer model that simulates this autocatalytic process has been developed at the Los Alamos Critical Experiments Facility. The model has been used to determine the fission yield of such an event and the effects of that yield on the repository. The goal of this work is to quantify the consequences of the autocatalytic process, not to determine the probability of such an event occurring.

INTRODUCTION

One way to deal with excess weapons plutonium is to store this material in underground repositories, such as Yucca Mountain. The Pu-239 would be mixed with SiO_2 to form a glass log. This log would then be buried in the native rock of an underground repository. Recent reports[1,2] have postulated that this method of dealing with excess Pu-239 could lead to a catastrophic nuclear explosion of up to a few hundred tons of equivalent high-explosive yield. [*Editor's Note: The Bowman and Venneri paper in this issue suggests a yield up to a few hundred gigajoules.*] This event is triggered by an autocatalytic process, which causes a critical system to drive itself automatically to a supercritical system.

This report presents the results of a dynamic analysis of the autocatalytic process proposed in the reports referenced above. The goal is to simulate the autocatalytic process as accurately as possible and to determine its nuclear

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yield. No attempt was made to determine the probability of such an event occurring, or even if such an event is physically possible. The object of this study was to develop a dynamic model based on the scenario postulated in references 1 and 2 and quantify its consequences.

Autocatalytic Process

The fundamental premise underlying the autocatalytic process is the fact that the plutonium is originally placed in an underground repository in an extremely undermoderated condition. Thus, if the plutonium can escape its confining glass log and spread out into the surrounding rock of the repository, which for the most part is quartz, it will become more moderated. Several mechanisms by which the plutonium may be spread out have been proposed in reference 1, such as ground water finding its way into the repository and transporting plutonium into the surrounding rock, or earthquakes and volcanos disturbing the glass log and surrounding rock. Ground water seeping into the repository is assumed to carry away neutron poisons that were added to the glass log initially. Also, the ground water may cause steam explosions to occur, which would help to disperse the plutonium even more quickly. In addition to dispersing the plutonium, these events will crack and fracture the surrounding rock, allowing the plutonium to penetrate it. Given enough time and many such events, the plutonium could disperse into the surrounding rock until it reaches a critical configuration.

At this point, the power will begin to rise, causing the plutonium to heat up and eventually vaporize. It is assumed in reference 1 that all the energy of the fission fragments will be deposited into the plutonium only and not the surrounding rock. This assumption is made because it is believed that the plutonium will be gathered heterogeneously in the cracks of the surrounding rock and not be homogeneously distributed throughout the rock. Thus, the plutonium will heat up quickly and vaporize while the surrounding rock may only heat up negligibly due to the energy of fission neutrons that escape from the plutonium. It should also be noted that the system should be pushed sufficiently far above critical in order to overcome the negative temperature feedback of $-1 \times 10^{-5} \Delta k_{eff}/^{\circ}C$, which is given in reference 1. This negative temperature feedback will decrease reactivity \$5 for every 1000 °C increase in the plutonium. Assuming the plutonium vaporizes at 3000 K and starts out at 300 K, the system should start out at a reactivity greater than \$13.50.[b] This would allow the system to still be critical at the point of plutonium vaporization.

b Reactivity is measured as departure from critical in $\$ = \frac{k_{eff} - 1}{k_{eff}\beta}$, where β = delayed neutron fraction.

Once the plutonium vaporizes, the autocatalytic process begins. This process is described in reference 2, which is a response prepared by the authors of reference 1 to an internal review of the draft of reference 1 performed by personnel at the Los Alamos National Laboratory. There it was assumed that the plutonium would vaporize at 3000 K, and the molecular velocity of the plutonium would be 45,000 cm/s. It was further assumed that the plutonium vapor would spread out through the surrounding rock at a velocity comparable to its molecular velocity. The worst-case scenario involves a glass log that initially contained 100 kg of Pu-239. In spherical geometry, 100 kg of Pu-239 homogeneously distributed in SiO_2 and surrounded by an infinite reflector of quartz would go critical at a radius of approximately 75 cm. It is assumed that this is the point at which vaporization would occur and the autocatalytic process begin. This assumes that the glass log and surrounding rock are quartz (pure SiO_2) with a density of 2.2 g/cm^3. The plutonium vapor spreads out rapidly and reaches its maximum k_{eff} of 1.18 at a radius of approximately 150 cm. Then the plutonium vapor continues to spread until it reaches a subcritical configuration at a radius of approximately 250 cm. See figure 4 of reference 1. [*Editor's Note: See figure 5 in the Bowman and Venneri paper, this issue.*] It is stated in reference 2 that the plutonium probably won't spread out through the surrounding rock unhindered, so it probably takes approximately 12 ms for the plutonium to spread from the critical radius, through supercritical, back finally to a subcritical radius. It is during this 12 ms period of the autocatalytic process that the potentially catastrophic nuclear yield will be produced. [*Editor's Note: The Bowman and Venneri paper in this issue does not estimate the period of the autocatalytic process.*]

THE MODEL

A computer model, which simulates the autocatalytic process described above, has been developed at the Los Alamos Critical Experiments Facility (LACEF). This model combines the neutron point kinetics equations with an equation of state for SiO_2 and energy, continuity, and momentum equations for SiO_2. It has been assumed that once the plutonium is vaporized, all the fission energy is deposited in the surrounding rock. Therefore, the model consists of a solid sphere of quartz with a radius of 150 cm. It is recognized that the actual scenario consists of a system whose radius increases from 75 cm all the way to 250 cm. For ease of computation a fixed geometry is assumed. This assumption is quite conservative, however, because testing of the model has shown that a negligible fraction of the total amount of energy generated is deposited into the system before it reaches maximum reactivity at 150 cm. Also, the

model dumps all the energy produced into the system of 150 cm, whereas in the actual system most of that energy should be deposited into a much larger system with a radius of 250 cm. This will result in over-estimating any mechanical effects. The model also restricts the outward expansion of the quartz sphere until the pressure of the core exceeds the compressive strength of the hypothetical surrounding rock. This constraint on the model is extremely conservative because it has already been assumed that the surrounding rock is cracked and fractured, otherwise the plutonium vapor would not be allowed to disperse. Also, it has been assumed that the original glass log and some of the surrounding rock have been broken to allow ground water to seep in and carry neutron poisons away. All the events that were necessary to help disperse the plutonium out to 75 cm, such as steam explosions or earthquakes, must have turned the original glass log and surrounding rock to rubble. The significance of this is that the Pu + SiO_2 system will be extremely porous, and no condensed state pressure can build up until the system expands to a point where the porosity has been eliminated. The model assumes a solid constrained sphere of SiO_2 because it best maintains the main theme of reference 1, which is that nuclear explosions can occur underground because of the confinement of the surrounding rock.

Neutron Point Kinetics Model

To calculate the power of the Pu + SiO_2 as a function of time, the point kinetics equations are used. These equations, in normalized form, are given by

$$\frac{dN}{dt} = \frac{\beta}{\Lambda}\left[(R\text{-}1)\ N + \sum_{i\text{-}1}^{6} \frac{\beta_i}{\beta} D_i\right] \tag{1}$$

and

$$\frac{dD_i}{dt} = \lambda_i (N\text{-}D_i) \tag{2}$$

where N is the fission power, R is the reactivity of the system in dollars, Λ is the mean generation time, and D_i is the relative precursor power. Because the reactivity of the autocatalytic process starts out near critical, moves to a maximum value, and then drops off to subcritical, a sine function is used to simulate the reactivity. An expression for the reactivity of this process is given by

$$R = 74.00 \sin\left(\frac{t}{0.006}\frac{\pi}{2}\right) \tag{3}$$

where t is the time in seconds and 74.00 represents the maximum reactivity

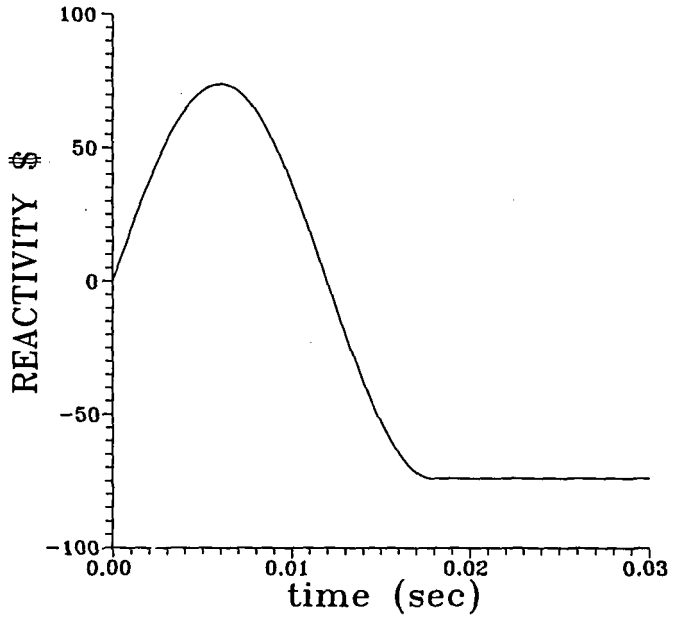

Figure 1: Reactivity vs time during the autocatalytic process.

the system reaches in dollars. Figure 1 shows a plot of the reactivity as a function of time. After its minimum value is reached, the reactivity is held constant. Only the effect of the plutonium vapor spreading out through the surrounding rock is accounted for in the reactivity equation. No other reactivity feedback mechanisms are included in the kinetics model. For example, the effects of thermal-neutron-spectrum hardening and the negative reactivity feedback due to fuel density redistribution in a constrained assembly are not included.

The most important parameter in the calculation of the power is the mean generation time. The system being modeled is unique in that the size of the core is continually changing. The core starts out as a sphere of radius 75 cm and ends up as a sphere with a radius of 250 cm. The volume of the core starts out at 1.767×10^6 cm^3 and ends up with a volume of 6.545×10^7 cm^3, which is a remarkable change. One would expect the mean generation time to change quite dramatically during this change. By using ONEDANT, a set of calculations was made to determine the mean generation time at various points during the autocatalytic process. At a radius of 75 cm (time = 0.0), the mean generation time is 7.3×10^{-5} s; at 150 cm (time = 0.006 s), the mean generation time is 1.3×10^{-4} s; and at 250 cm (time = 0.012 s), the mean generation time is

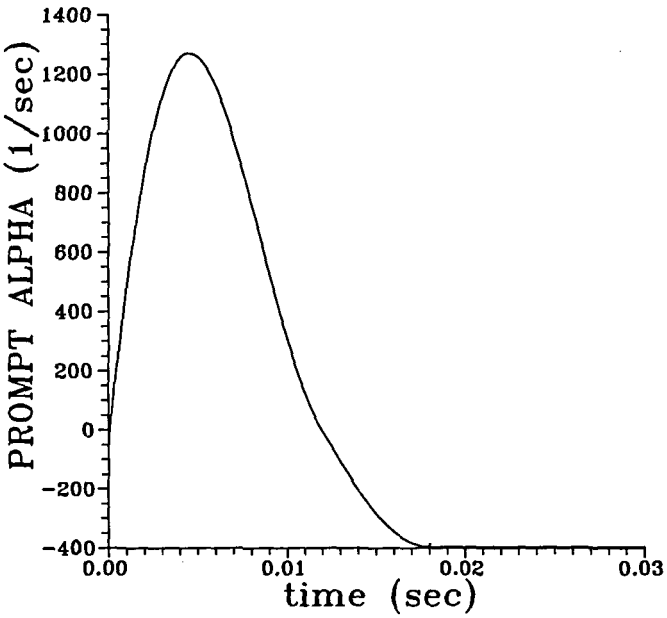

Figure 2: Prompt Alpha vs time during the autocatalytic process.

4.0×10^{-4} s. An expression for β/Λ as a function of time is given by

$$\frac{\beta}{\Lambda} = 28.48 - 2268t + 28056t^2 \qquad (4)$$

This expression is simply an empirical fit that matches the known mean generation time with its corresponding reactivity in time. After the system becomes subcritical (time = 0.012 s) the mean generation time stays constant at 4.0×10^{-4} s. The prompt alpha for the pulse, which is strongly dependent upon the mean generation time, is shown in figure 2 as a function of time. The mean generation times stated above are conservatively short because the model used to calculate them assumed that the plutonium was homogeneously distributed throughout the quartz. Again, it should be noted that the plutonium spreads out through cracks in the surrounding rock, which means that the plutonium is distributed more heterogeneously than homogeneously. This will cause the actual critical system to have a larger dimension than a system with the plutonium homogeneously distributed. Thus, a larger system will mean a longer mean generation time.

Another key parameter in the kinetics equations is the initial power of the system. For the autocatalytic process to occur, the plutonium must be completely vaporized. It is assumed that this vaporization occurs, because of some

event that causes the system to go supercritical initially. Because the history of the system is not known, in any detail, prior to the autocatalytic event, the model assumes that the initial power is 500 MW. This power should be conservatively high enough to ensure that the 100 kg of plutonium are completely vaporized in the period of time leading up to the autocatalytic event. The total amount of fission energy produced during the autocatalytic event is directly proportional to the initial power. It is believed that 500 MW is a very conservative upper limit for the initial power of any actual event.

Equation of State

During the autocatalytic process, the fission energy generated will be deposited into the remains of the glass log and, later, in the surrounding rock. To determine the mechanical stresses produced by this energy deposition, an equation of state for SiO_2 has been developed. This equation is given by

$$\frac{dP}{dt} = \frac{\alpha}{\kappa}\frac{dT}{dt} + \frac{1}{\rho\kappa}\frac{d\rho}{dt} \tag{5}$$

where P is the pressure of the SiO_2, α is the isobaric compressibility of SiO_2 (5.5×10^{-7} $^{\circ}C^{-1}$), κ is the isothermal compressibility of SiO_2 (2.74×10^{-5} MPa^{-1}), ρ is the density of the SiO_2, and T is the temperature of SiO_2.[3,4] An expression for the temperature of the SiO_2 as a function of time and position is given by

$$\frac{dT}{dt} = \frac{N}{MC_P}\frac{R}{r\pi}\sin\left(\frac{r\pi}{R}\right) \tag{6}$$

where M is the mass of the SiO_2, C_P is the specific heat of SiO_2 (7.5×10^{-4} MJ/kg/°C), r is the radial position, and R the radius of the SiO_2 sphere.

To determine the acceleration of the SiO_2 and its change in density, simple momentum and continuity equations are used. In spherical coordinates these equations are given by

$$\frac{\partial U}{\partial t} = -\frac{1}{\rho}\frac{\partial P}{\partial r} \tag{7}$$

and

$$\frac{\partial \rho}{\partial t} = -\frac{1}{r^2}\frac{\partial r^2 \rho U}{\partial r} \tag{8}$$

where U is the velocity of the SiO_2.

A multi-region model was used to simulate the SiO_2. The quartz sphere was split up into ten concentric shells of equal thickness. Each shell or region contains its own equation of state and its own energy, momentum, and continuity equations. These equations were made discrete and solved numerically on a dynamic system-simulation software package.[5]

RESULTS

Figure 3 shows the model's predicted power pulse for the autocatalytic process. The spatially averaged peak power of the burst was 3.83×10^6 MW and the total fission energy yield for the prompt burst was 2.85×10^4 MJ or about 9.48×10^{20} total fissions. Figure 4 shows the core-averaged temperature of the quartz sphere and the pressure that was built up during the autocatalytic process. The fission energy generated caused the quartz to heat up an average of 1250 °C and it caused a pressure buildup of 24.6 MPa throughout the quartz.

CONCLUSION

The computer model presented above attempts to simulate supercritical excursions of plutonium and SiO_2 in underground repositories. In particular, the model tries to simulate the "dry" autocatalytic process postulated in references 1 and 2 as accurately as possible. The model's predicted total energy yield is at least two orders of magnitude less than the kinetic energy yield estimated in reference 1. The fact that the mean generation time continually changes as the plutonium vapor spreads out through the surrounding rock has a dramatic effect on the e-folding period during the autocatalytic process. The mean generation time increases by almost a factor of 6 during the course of the pulse. The effect of the mean generation time on the pulse can be seen from figure 2. As the mean generation time increases, the prompt alpha increases less with increasing reactivity.

Clearly, no significant kinetic energy was produced during the pulse. The only motion produced was a redistribution of core material from the center of the core outward. The fission energy generated only raised the temperature of the quartz 1250 °C, which produced a negligible pressure buildup of 24.6 MPa. Given a compressive strength for quartz of 1067 MPa, the surrounding rock can contain the quartz sphere without any difficulty.[4] Even if the quartz sphere was allowed to expand freely after the prompt burst, it would only generate 0.22 MJ of kinetic energy or about 0.11 lb of equivalent high-explosive

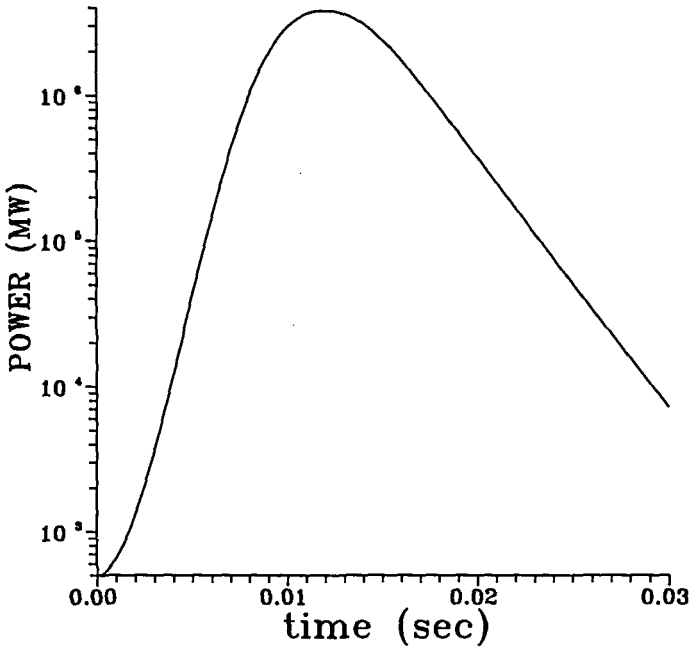

Figure 3: Model's predicted power pulse for the autocatalytic process.

yield. This calculation was made by giving the quartz sphere an initial pressure of 24.6 MPa and letting the outer boundary of the sphere expand against a vacuum. The model predicted a maximum dynamic pressure of 0.016 MPa. By multiplying the dynamic pressure by the volume of the quartz, the kinetic energy can be crudely approximated.

At a radius of 150 cm the system contained over 31,000 kg of quartz, which provided a huge energy sink. It should be noted that at the 12 ms point during the burst, only 44 percent of the total fission yield had been generated. This means that in an actual system, over 50 percent of the 1.03×10^4 MJ yield should be deposited into a sphere at least 250 cm in radius, which would contain over 1.4×10^5 kg of quartz. Due to the large thermal inertia of the quartz, no significant mechanical stresses were produced even with the extremely conservative constraints placed on the model. It is the opinion of the author that no kinetic energy could be produced by the autocatalytic process described in this report. The sheer size of the system considered, combined with the long mean generation time, makes it impossible for any nuclear explosion to occur. The integrity of an underground repository will not be compromised by the autocatalytic process postulated in references 1 and 2.

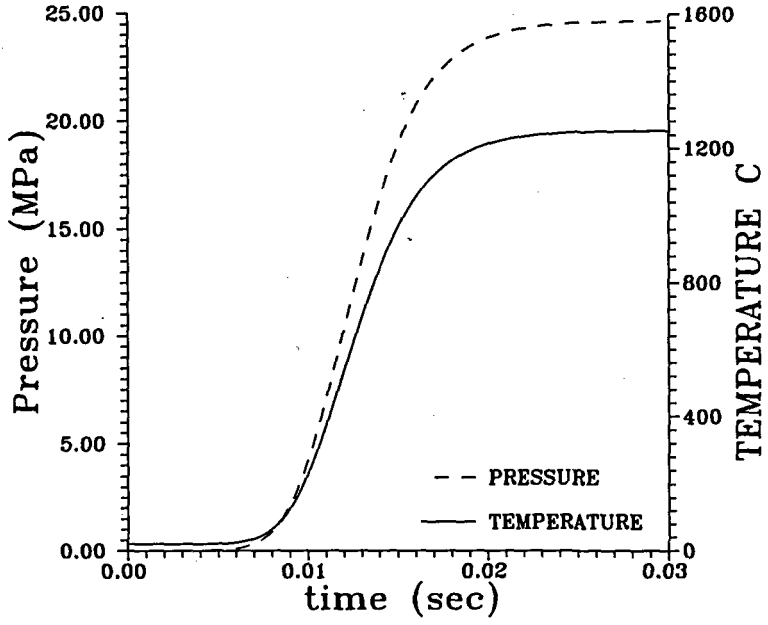

Figure 4: Model's predicted pressure and temperature behavior during the autocatalytic process.

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