HYPOTHETICAL CRITICALITY ACCIDENTS IN DILUTE PLUTONIUM-WATER SOLUTIONS: A POTENTIAL TRANSURANIC STORAGE HAZARD

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ABSTRACT

It has recently been noted that plutonium-water solutions may have positive temperature coefficients of reactivity at low fuel concentrations. A potential consequence could be the enhancement of a nuclear accident involving storage of certain types of liquid transuranic wastes. An example might be a large, shallow storage tank in which a plutonium-containing precipitate were accidentally disturbed and mixed with an overlying layer of water or organic liquid. Some computations of hypothetical accidents have been performed using a new methodology that was developed primarily for water solutions of uranyl nitrate and sulphate. It is concluded that significant enhancement of accident consequences is possible for certain combinations of parameters.

TEMPERATURE COEFFICIENTS OF REACTIVITY

The possibility of a positive temperature coefficient for a dilute plutonium-water solution was suggested by Suzaki, Miyoshi, and Hirose (1). Confirmation using multigroup transport theory was first reported by Mather et al (2). The most recent calculation was performed by Kornreich (3), using 69-energy-group transport theory with scattering kernels for bound hydrogen computed at intervals of 10 Celsius degrees. Spectral (neutron temperature) coefficients were calculated for poisoned and unpoisoned mixtures of plutonium (5% ^{240}Pu), gadolinium, and unit density water in infinite cylinders. All calculations were performed such that the reference state (293 K) was critical. Results are reproduced in Fig. 1, where the neutron temperature coefficients are expressed in dollars/degree and where the reactivity is $\Delta k/k$ divided by the delayed-neutron fraction for plutonium (0.0022).

The unpoisoned system exhibits a positive neutron temperature coefficient for concentrations below about 20 g/liter, while the poisoned system has a coefficient that changes sign between 50 and 60 g/liter and that is notably larger than that for the unpoisoned system at low concentrations. Therefore, the addition of this type of neutron absorber, while reducing neutron multiplication, may have an unexpected side effect.

Note that these are spectral (neutron temperature) reactivity coefficients, and not density effects (changing neutron leakage). Density decreases, produced by temperature rise as well as by the production of radiolytic gas, usually produce negative reactivity changes. The density effect, however, depends on the shape of the assembly, and it will be smaller for a short, wide cylinder than for a tall, thin one, suggesting the concern about a large, shallow storage tank containing a dilute solution.

DYNAMIC SIMULATIONS

One version of the dynamic simulation model, with applications to uranyl sulphate and nitrate pulsed reactors, was published in 1991 (4). The model uses one-region reactor dynamics with a two-term reactivity feedback

$$R = \alpha\Delta T + \varphi\Delta V$$

where R is reactivity in dollars, α is the neutron temperature coefficient of reactivity in dollars/degree, ΔT is the temperature increase, ϕ is the volume expansion coefficient of reactivity in dollars/unit volume, and ΔV is the volume change.

The differential equations for pressure and temperature are derived from an equation of state $P = P(T,V)$ and an energy balance. Acceleration is obtained from a momentum equation with dissipation

$$\ddot{y} = -\frac{1}{\rho}\nabla P - \theta\dot{y}$$

where y is the co-ordinate of the center of mass, ρ is the density, and θ is a parameter to be determined. In the one-region model, the pressure gradient is approximated by the gauge pressure at the bottom of the vessel divided by the solution height.

In this model it was assumed that radiolytic gas bubbles are nucleated at a rate proportional to the fission track density times the supersaturated concentration of dissolved gas, and that they grow quickly to a final radius r. Finally, the coefficients in the equation of state are the isothermal compressibility

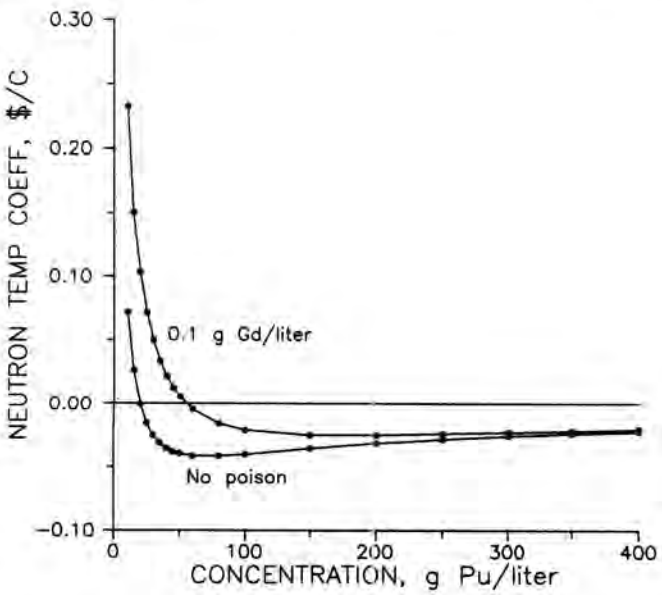

Fig. 1. Comparison of neutron temperature reactivity coefficients in poisoned and unpoisoned Pu-water solutions in critical infinite cylinders using 69-group cross-section sets (calculations by D. E. Kornreich, University of Arizona).

$$K = K_0 (1-f) + \frac{f}{P + 4\sigma/3r}$$

and the isobaric compressibility (thermal expansion coefficient)

$$\beta = \beta_0 (1-f) + \frac{f(P + 2\sigma/r)}{T(P + 4\sigma/3r)}$$

where f is the volume fraction of gas in the form of bubbles, σ is the surface tension, and the symbols with the subscript 0 refer to properties of the liquid. The liquid thermal expansion coefficient is represented by a cubic polynomial function of temperature.

As described in Ref. 4, two adjustable parameters were used to match peak power and peak pressure data from three pulsed reactor experiments. The resulting model was then adapted for use with dilute plutonium solutions (5). The model exhibited an interesting dynamic competition between prompt positive and delayed negative shutdown effects, and there was an extreme sensitivity to certain parameter values.

Meanwhile, a different model incorporating one-dimensional space-dependent hydrodynamics was being developed by Kimpland (6). This model has recently been improved by Kimpland, in research that will be reported in his forthcoming dissertation. Two-dimensional (r,z) hydrodynamics has been included, and a more realistic model for radiolytic gas has been devised.

The older model had two time regimes, one before saturation of dissolved radiolytic gas, in which the only volume effect was the liquid thermal expansion, followed by a second regime in which the formation of gas bubbles provided more rapid reactivity reduction and also a sudden increase in compressibility. This model failed to reproduce details of the pressure-time data, and it also was not able to reproduce the extremely rapid shutdown of the power pulses following the pressure peak.

The new model has three time regimes. In the first regime, the density effect of vapor in the thermal spikes produced by the fission tracks is included. These voids quickly collapse, but when the saturation concentration of dissolved gas is reached, bubbles of gas can grow rapidly. This marks the beginning of the second regime, in which gas voids are produced at a rate proportional to the fission track density, and in which the inertial pressure is suddenly augmented by the rapid production of gas bubbles. The third regime is marked by violent disassembly that occurs after the dynamic pressure has been relieved. This is roughly modelled by assuming that pressure relief induces extremely rapid cavitation throughout the solution. This model for gas production more nearly reproduces details of pressure-time data and nuclear power data.

Kimpland's model for gas production was incorporated into the one-region dynamic simulation, and one parameter (the production rate constant for gas bubbles after supersaturation) was adjusted in an attempt to match pulsed reactor data. One advantage of the new model for gas production is that the artificial dissipation term in the momentum equation is no longer needed in either the one-region or the many-region version. The one-region case with the new model for gas production was then adapted to create a new simulation model for plutonium solutions, and results are reported in the next section of this paper.

SIMULATION RESULTS

For the one-region plutonium solution model, we selected a 40-cm radius cylinder having a fuel concentration of 10.5 g/liter. The parameters used in the equation of state, in the energy balance, and in the momentum equation were carried over from the uranium solution assemblies with adjustments for geometry and fuel concentration. Six-group delayed neutron parameters for thermal fission in plutonium were used, and an estimated value of 55 s^{-1} was selected for β/l.

From the 69-group transport theory, the neutron temperature coefficient of reactivity for this concentration is 0.072 \$/C (the lower curve in Fig. 1). Using two-dimensional transport theory (TWODANT) with 16-group Hansen-Roach cross-sections, the volume expansion reactivity coefficient was calculated as -0.289 \$/liter. A tall, thin cylindrical assembly would have a more negative volume coefficient, whereas a short, fat cylinder would have a smaller negative value. In the limit of large radius (an infinite slab) the volume reactivity coefficient would be zero.

Time-dependent power and reactivity for a reactivity input of four dollars are shown in Fig. 2. The computations were performed on a 386 PC, using DESIRE, a simulation software package (7). Note that the positive component of reactivity feedback appears first. After the reactivity peaks at 4.4 dollars, the power is ultimately limited by negative feedback from volume expansion. The maximum inertial pressure for this pulse is 0.58 MPa and the energy yield is 89 MJ. Radiation dose in the absence of shielding would be quite high, but the potential for mechanical damage is somewhat limited by the fact that only a very small fraction of the energy appears as kinetic energy (about 0.1 % in this example).

The magnitude of the pulse is larger if the positive neutron temperature coefficient of reactivity is increased. The largest coefficient in Fig. 1 is 0.233 \$/C. A second peak in reactivity appears when α exceeds about 0.224 \$/C, and a

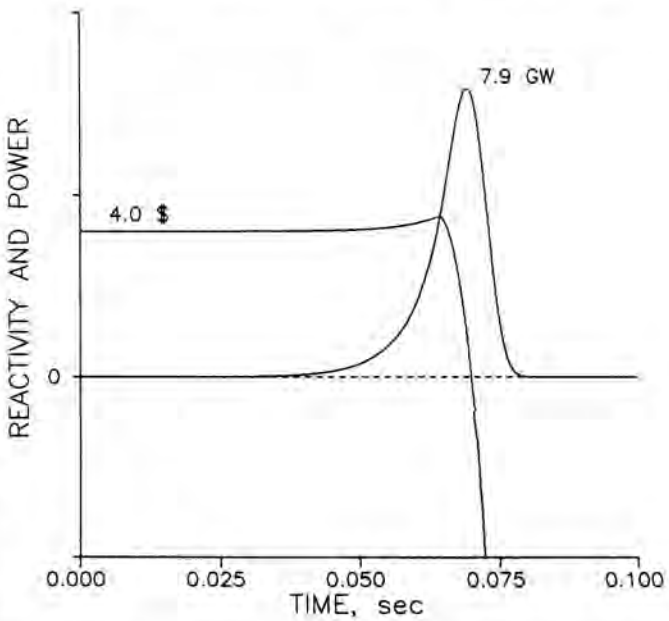

Fig. 2. Reactivity and power for a 4-dollar reactivity input in a Pu-water solution (finite cylinder, 10.5 g Pu/liter, neutron temperature coefficient 0.072 \$/C).

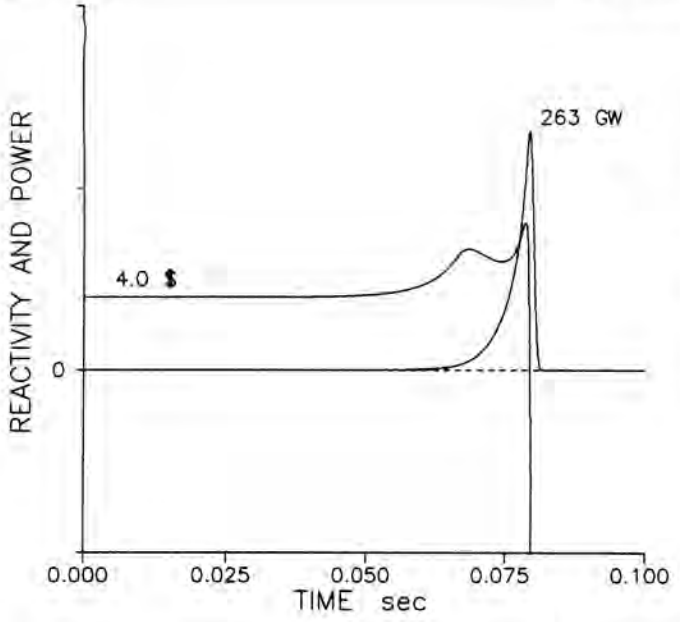

Fig. 3. Reactivity and power for a 4-dollar reactivity input in a Pu-water solution (finite cylinder, 10.5 g Pu/liter, neutron temperature coefficient 0.2247 $/C).

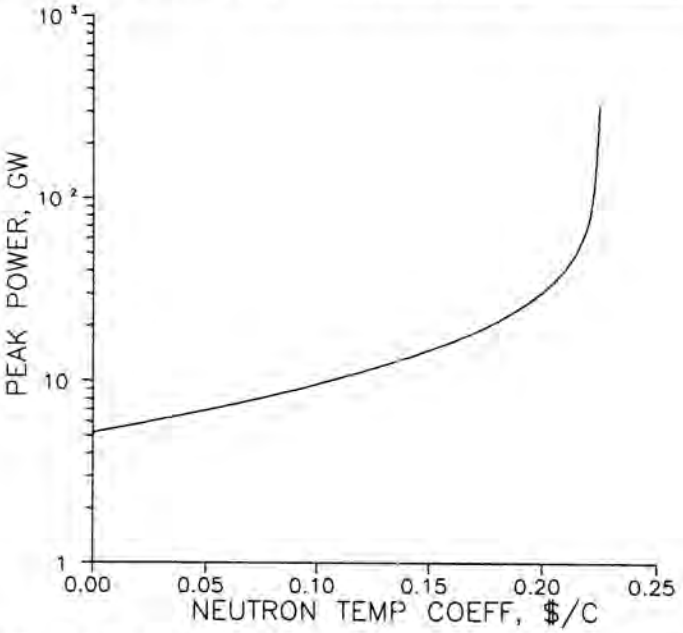

Fig. 4. Peak power vs neutron temperature coefficient for a 4-dollar reactivity input.

computer example was selected at $\alpha = 0.2247$ and is shown in Fig. 3.

The dynamic competition between the positive and negative reactivity feedbacks is evident in Fig. 3. The reactivity first increases in response to the positive feedback, but the negative effect of expansion overtakes this and causes the reactivity to peak at 6.6 dollars. The reactivity falls to about 6.0 dollars before the positive feedback catches up. The result is a maximum reactivity of 8.1 dollars followed by a large peak power and a violent disassembly. The peak pressure is 49 MPa, the energy yield is 1073 MJ, and the maximum kinetic energy is about 5 %.

Effects of varying the temperature coefficient are shown in Fig. 4 (peak power), Fig. 5 (maximum reactivity) and Fig. 6 (energy yield). As expected, increasing α while keeping ϕ constant results in larger pulses that become very sensitive to the value of α. The sharp discontinuity in maximum reactivity in Fig. 5 denotes the appearance of a second peak in reactivity that exceeds the height of the first peak after an extremely small increase in α.

Reducing the magnitude of the volume reactivity coefficient ϕ, while keeping α constant, will also produce larger pulses. This corresponds to moving toward larger, squat cylinders having the same fuel concentration. With $\alpha = 0.072$ $/C, the magnitude of the pulses becomes very sensitive to the value of ϕ for $|\phi|$ less than 0.08 $/liter. The calculated peak power at $\phi = -0.075$ $/liter is 643 GW. However, the tendency toward a second peak in reactivity was not observed.

Increasing the input reactivity produces larger pulses, and, for sufficiently large α, a second peak in reactivity can also be observed. However, this model for radiolytic gas did not show any discontinuities in curves of peak power vs input reactivity of the type reported for the older gas production model used in Ref. 5.

We have not attempted to model the complicated mechanisms by which the reactivity could suddenly become sufficiently positive to produce large power pulses. An example

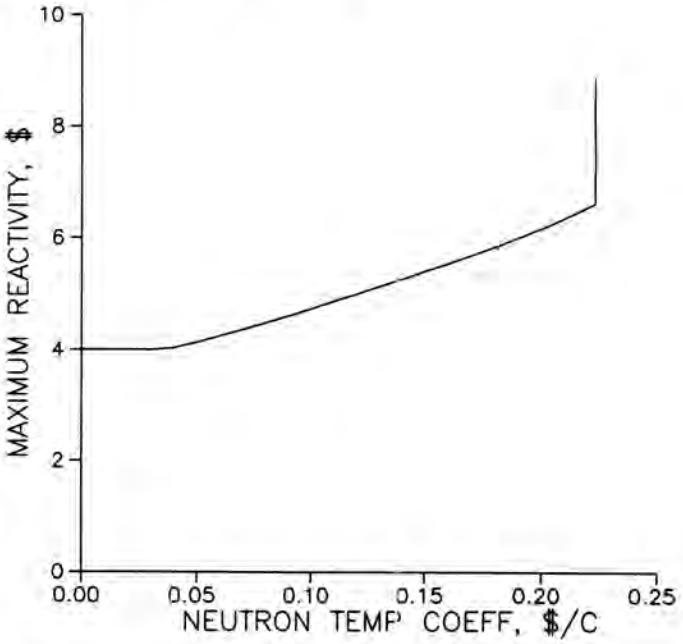

Fig. 5. Maximum reactivity vs neutron temperature coefficient for a 4-dollar reactivity input.

might be a large, shallow tank in which a plutonium-containing precipitate were accidentally disturbed and mixed with an overlying layer of water or organic liquid. Other examples can be imagined for fuel processing plants.

CONCLUSION

We have described some interesting new features that might appear in hypothetical criticality accidents involving positive components in the reactivity feedback, and we have suggested that there may be reason for concern about storage of certain types of liquid transuranic wastes. We are

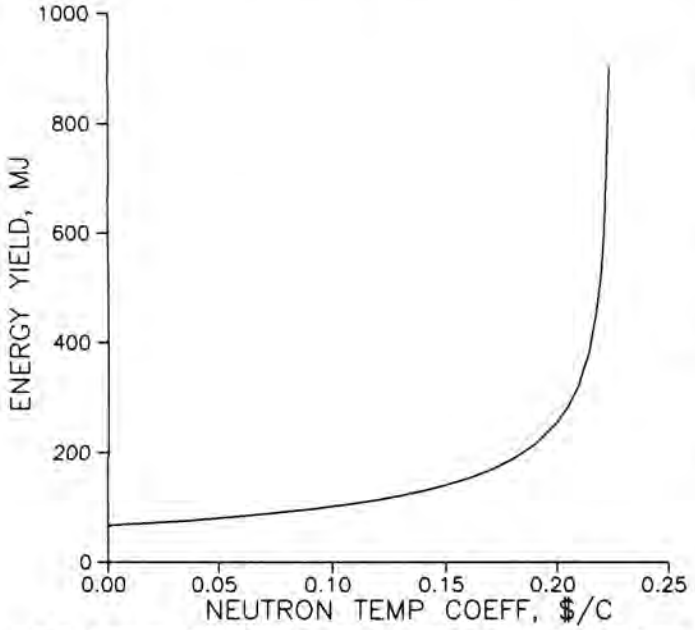

Fig. 6. Energy yield vs neutron temperature coefficient for a 4-dollar reactivity input.

continuing to refine our models and to study the sensitivity to parameter variations.

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