

PLUTONIUM-WATER CRITICALITY HAZARD IN NUCLEAR WASTE DISPOSAL

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ABSTRACT

The possibility of autocatalytic nuclear excursions in plutonium processing and nuclear waste storage has recently received some attention. One source of concern is the positive neutron spectral temperature coefficient that has been predicted for solution concentrations below 20 g of plutonium per liter of water. This paper includes results of some new dynamic simulations for realistic dilute plutonium solution assemblies, showing comparisons of computed energy yields with and without the positive reactivity feedback. The primary application is to potential hazards in a large interim storage tank, where a sudden reconfiguration could have severe consequences.

INTRODUCTION

The possibility of a positive neutron spectral temperature coefficient in dilute water solutions of plutonium salts was suggested by Suzaki et al (1). Confirming computations were reported by Mather et al (2) and by Kornreich (3).

Previous computations for hypothetical plutonium-water assemblies (4,5) used an earlier version of our simulation models. The models were originally developed for uranium-water solutions, and benchmarked against solution reactor experiments and laboratory simulations of criticality accidents (6,7). Neutron temperature coefficients used in the new plutonium-water simulations were computed by Kornreich (3) and the critical configuration data and the volume expansion reactivity coefficients were computed by Lee (8,9). Some recent dynamic studies are reported by Newkirk (10) and by Hetrick and Newkirk (11). The dynamic computations were performed using the PC simulation language DESIRE (12).

SIMULATION MODELS

The dynamic simulation uses point reactor dynamics, with three types of reactivity feedback: neutron temperature, thermal expansion, and radiolytic gas voids. In uranium-water solutions and many plutonium-water solutions, all three feedbacks are negative, resulting in fairly stable systems. The dynamic model incorporates two types of inertial effects (pressures generated by bubble nucleation and mechanical lag in thermal expansion). Two types of hydrodynamic model have been used: a one-region lumped model, and a two-dimensional multi-region model. In both cases, volume-average densities and temperatures were used to calculate the reactivity feedbacks. It was observed that peak powers could be predicted well with the multi-region hydrodynamics using density feedback coefficients from transport theory calculations, but with the one-region model the transport theory feedback had to be multiplied by three to match peak powers. In order to match peak pressure data, it was necessary to invoke an adjustable parameter relating the rate of bubble production to the nuclear power (6,7).

Application of these models to plutonium solutions requires the assumption that the bubble dynamics are still valid in systems for which there is no experimental data. Also, excursions with positive feedback may be very large, stretching the credibility of simple dynamic models.

PLUTONIUM EXAMPLES

The cylindrical assemblies chosen for illustration are aqueous plutonium nitrate solutions having a concentration of 10 g Pu/liter and a neutron temperature coefficient $\alpha_T = +0.075 \text{ } \$/\text{K}$. Physical characteristics and corresponding values of the volume expansion reactivity coefficient α_v for the three cylinders are given in Table I.

Table I. 10 g/ Plutonium Nitrate Solutions ($\alpha_T = +0.075 \text{ } \$/\text{K}$)

H/D	R (m)	V (m ³)	α_v ($\$/\text{m}^3$)
1.30	0.35	0.35	-849
0.82	0.40	0.33	-687
0.70	0.43	0.35	-570

Values of α_v are transport theory results (8) multiplied by three for use in one-dimensional dynamic simulations. The prompt decay constant for all three assemblies is $\lambda = 22.8 \text{ sec}^{-1}$.

Energy yield vs initial reactivity for the first assembly (H/D = 1.30) is plotted in Fig. 1. One dollar of reactivity corresponds to a neutron reproduction factor that is above critical, where for plutonium is about 0.002 (i.e., $k = 1.002$). The role of positive feedback is illustrated by the comparison with computations for the same assembly without neutron temperature feedback. Note that 100 MJ is about 3×10^{18} fissions.

Systems such as these are statically unstable. If initially critical, a tiny perturbation will produce an increasing reactivity because $\alpha_T + \alpha_v \frac{V_0}{V} > 0$ where α_0 is the coefficient of thermal expansion at the initial temperature and V_0 is the initial volume. As the temperature rises due to nuclear heating, nonlinear thermal expansion of water introduces sufficient negative reactivity to reverse the reactivity increase. When the concentration of dissolved gas reaches saturation, the sudden appearance of gas bubbles produces a pressure pulse. At the same time there is a sudden increase in the solution compressibility, which provides pressure relief and a significant change in the hydrodynamic behavior. There may also be a second pressure pulse caused by inertial lag in thermal expansion.

A four-dollar transient with this system (H/D = 1.30) has an energy yield of 115 MJ and a maximum kinetic energy of 0.13 kJ. The reactivity peaks at 4.029 and then declines rapidly. There are two pressure peaks (0.14 and 0.12 Mpa above atmospheric). For comparison, this transient is several times as energetic as the largest pulses in uranium-water solution reactors.

The second system in Table I ($H/D = 0.82$) is close to optimum (minimum critical mass). Energy vs initial reactivity is plotted in Fig. 2. The energy curve with positive feedback is no longer monotonic, and in part of the range it is much higher than the curve without neutron temperature feedback.

A five-dollar transient in this system is seen in Fig. 3, which shows part of the reactivity-time curve. This example was selected to illustrate the dynamic competition between positive and negative feedback that can yield two reactivity peaks before shutdown by disassembly. The reactivity peaks are at 5.19 and 5.65 dollars. The total energy is 290 MJ and the maximum kinetic energy is 34 kJ. There are two pressure peaks (0.17 and 0.79 Mpa).

The third case in Table I ($H/D = 0.70$) has the smallest value for the magnitude of ρ of the three systems listed. Energy vs initial reactivity is plotted in Fig. 4. A pulse initiated by a reactivity of 8 dollars (initial $k = 1.016$) has a total energy of 880 MJ and a maximum kinetic energy of 3.8 MJ. The two pressure peaks are 0.3 and 60 Mpa. Such a power pulse could be very destructive.

Note that the cases in Table I illustrate a trend of increasing accident severity as H/D is reduced. This is consistent with the expectation that $\rho \rightarrow 0$ in the limit of an infinite slab.

PRACTICAL APPLICATIONS

The above examples are illustrations of hypothetical accidents. Practical concerns would involve complicated scenarios that could take a plutonium solution storage tank from subcritical to supercritical in a very short time.

Possibilities include a sudden reconfiguration caused by an accident during handling, a turbulent introduction of water, an unintentional introduction of concentrated solution, or a natural disaster. Under certain conditions, recriticality and multiple power pulses could occur. Detailed modeling of realistic accidents would be very difficult.

CONCLUSIONS

Dynamic simulations of nuclear excursions in dilute plutonium-water solutions illustrate the possibility of dramatic increases in energy yield caused by positive reactivity feedback. More study is needed to improve the computational models and evaluate the sensitivity of the models. Improved models should include better computation of reactivity feedback from space-dependent densities, convection of gas voids and delayed-neutron precursors, and a model for explosive vaporization of a superheated solution.

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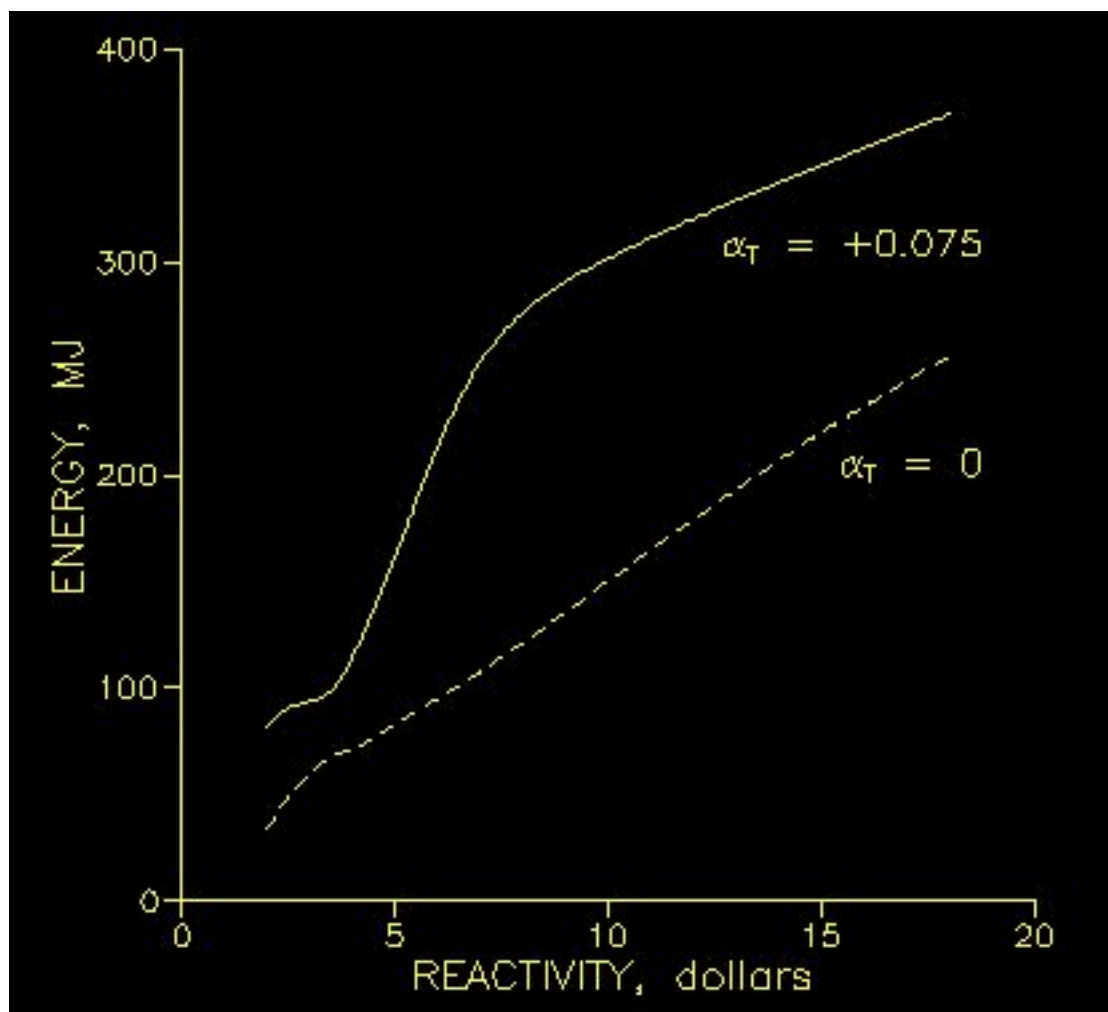


Fig. 1. Energy yield vs initial reactivity for Pu-water solutions (10 g Pu/L, $\alpha_T = +0.075$ \$/K, and $\alpha_v = -849$ \$/m³). Comparison with the curve for $\alpha_T = 0$ shows effect of positive feedback.

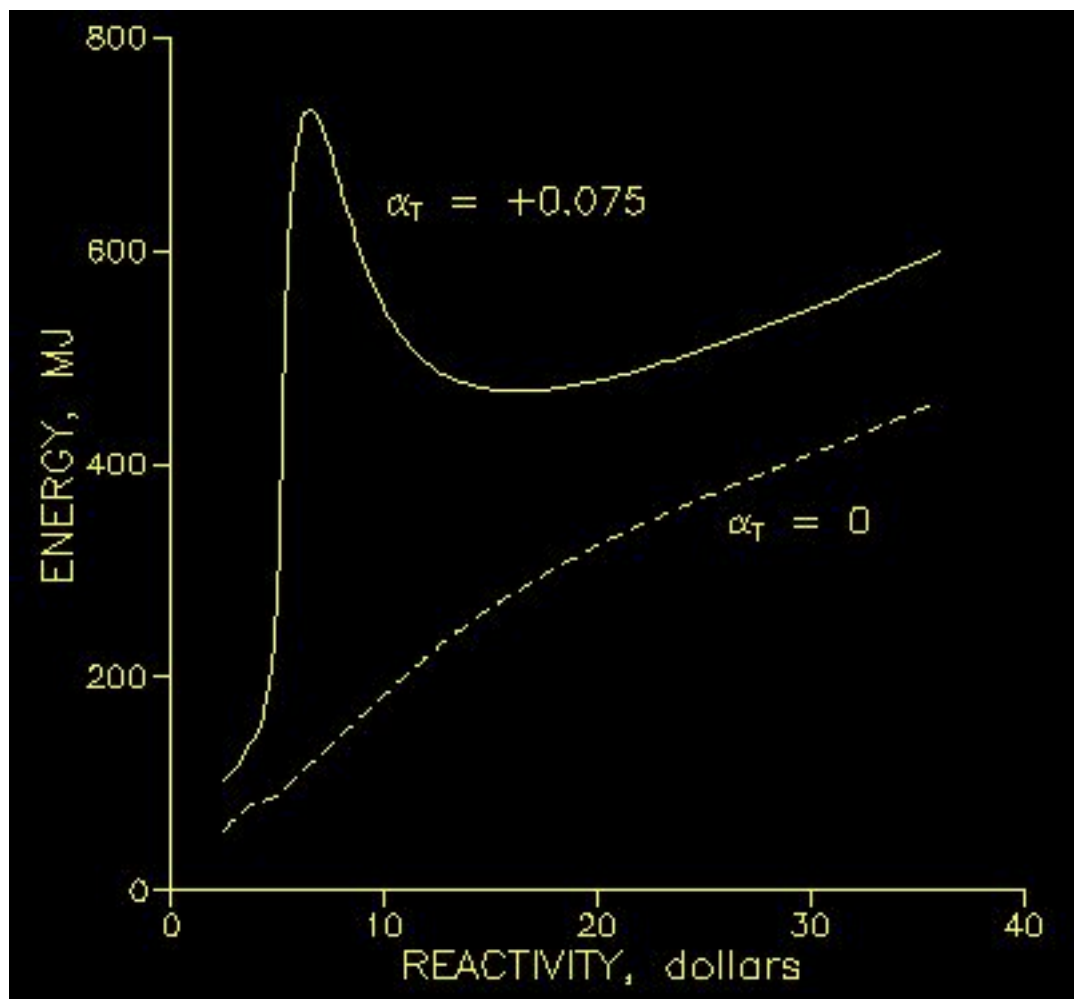


Fig. 2. Energy yield vs initial reactivity for $\nu = -687 \text{ \$}/\text{m}^3$. Reprinted from Ref. 11 by permission (copyright 1997 by the American Nuclear Society, La Grange Park, Illinois).

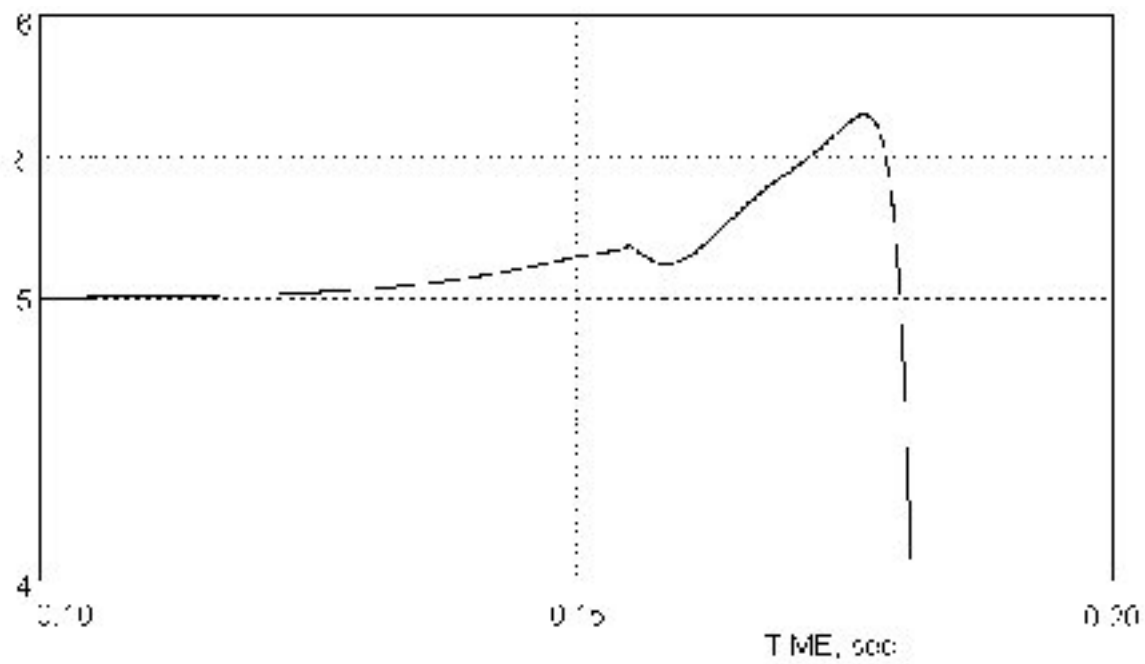


Fig. 3. Reactivity in dollars vs time for a five dollar pulse (initial $k = 1.01$) in the system of Fig. 2, illustrating two peaks in transient reactivity.

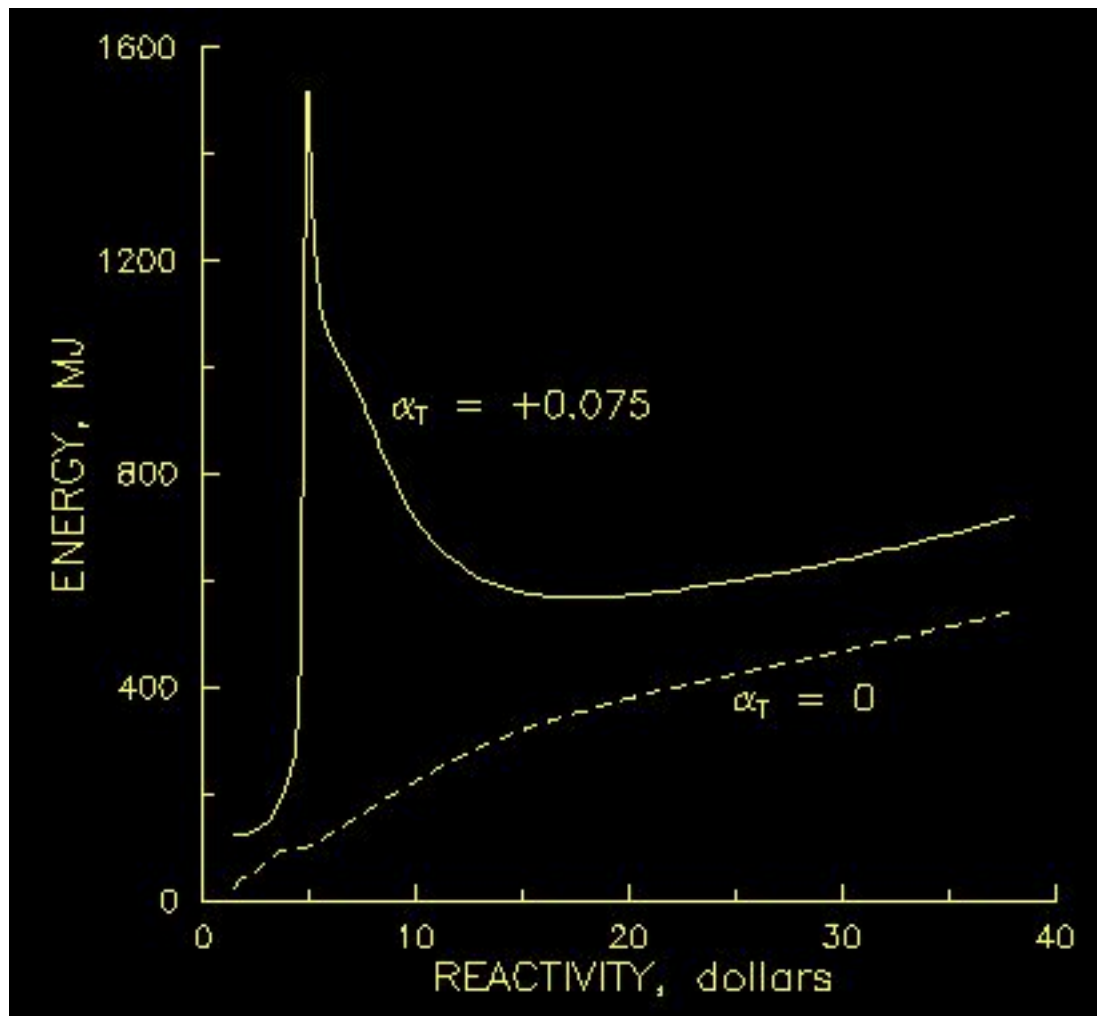


Fig. 4. Energy yield vs initial reactivity for $\nu = -570 \text{ } \$/\text{m}^3$.