Letters to the Editor

Comment on "Reactivity Feedback Mechanisms in Aqueous Fissile Solutions"

Kornreich[1] reported results of theoretical studies made on reactivity feedback effects arising from the volumetric expansion of solutions (both uniform and distributed density) and from changes in neutronic spectral temperatures for a number of aqueous fissile solution systems. These studies are important for hazard evaluation of potential criticality accidents in homogeneous solutions of fissile material. The systems investigated were CRAC, KEWB-5, SILENE, SHEBA (all uranium-fueled assemblies), and plutonium solutions. Plutonium systems considered were infinite (poisoned and unpoisoned) and finite (with and without reflector) cylinders. Calculations were made by using the TWODANT transport theory code with an angular quadrature of order S_8. The 16-group Hansen-Roach cross-section set was used for calculating the volumetric feedback coefficients and a 69-group temperature-dependent cross-section set, generated from ENDF/B-VI data, for the neutron temperature feedback coefficients.

Reference 1 explains very well the physics of reactivity mechanisms in fissile solution systems. It does not, however, explicitly deal with the effect of radiolytic gas production on reactivity. The major conclusions—some of which confirm the results of previous studies—may be summarized as follows:

1. Volumetric reactivity feedback is primarily a geometry effect and decreases with an increase in system size, but the spectral temperature feedback coefficients are almost geometry independent and depend mainly on the solution's composition.

2. The magnitude of the spectral temperature coefficient for the 10.5 g Pu/ℓ solution infinite cylinder was significantly higher than the system's volumetric expansion feedback coefficient.

3. Infinite cylinders of low-concentration plutonium solution (<20 g Pu/ℓ) can produce a positive neutron temperature coefficient, and the addition of gadolinium poison to such systems makes the temperature feedback coefficient more positive (less negative).

4. Finite water-reflected cylinders of aqueous plutonium (98.5% ^{239}Pu) nitrate can have positive temperature coefficients, but the concentration at which reactivity changes sign from positive to negative is significantly higher (32 g Pu/ℓ) than that for the infinite cylinders. For unreflected systems, differences in the spectral negative temperature coefficients are mainly due to differences in their axial leakage. The greater the leakage, the greater is the negative temperature coefficient.

5. A significant discrepancy (nearly a factor of 2) between the experimental and the calculated reactivity feedback coefficients was noted but not explained.

6. The 69-group cross-section set can be used for accurate calculation of critical eigenvalues in fissile solution systems.

The purpose of writing this letter is to draw Kornreich's attention to my earlier work on the physics characteristics of power excursions in a homogeneous solution of fissile material since it has not been cited in his paper. The investigations[2] were carried out both by direct numerical integration of the point-kinetics equations and by the analysis of French criticality experiments.[3] For the former, a ^{233}U fueled uranyl nitrate experimental homogeneous solution reactor having a core volume of 4 to 7 ℓ was investigated. It had a 148-mm-i.d., 500-mm-high, and 3-mm-thick (radial) cylindrical Zircaloy core vessel surrounded by a 30-cm-thick BeO reflector. There were two safety mechanisms: a gravity drop of two cadmium absorber plates and a gravity drop of a large BeO reflector. A minimum of 10% of the core vessel volume was set apart for fuel solution expansion in the event of a power transient. The overall design of the reactor was based on the minimum critical mass concept.

The main reactivity feedback factors considered were those due to temperature and the radiolytic gas. Both these effects give rise to volumetric expansion of the fuel solution, which increases the height-to-diameter ratio of the core (geometrical effect) and decreases the density of the core (density effect) thereby removing the excess reactivity added by the external agency and limiting the nuclear excursion (power pulse size and fission energy yield). However, there is an essential difference between the two. The temperature reactivity effect is a permanent feature being proportional to the increase of temperature at any instant of time and acts promptly. But, the gas void reactivity effect is proportional to the amount of gas present in the fissile solution and is temporary since the gas bubbles migrate to the core solution surface and escape (for an open system). This gas void is formed from hydrogen and oxygen microbubbles that are produced by the radiolytic decomposition of water when fission fragments and other products of a fission reaction interact with the aqueous fissile solution directly. The volume of released gas has been measured to be between 5 to 6 ℓ/MJ ($H_2 + \frac{1}{2}O_2$) of fission energy. The time constant of the radiolytic gas void reactivity effect depends on the fuel solution volume, on the amount of gas produced per unit energy deposition (G_{H_2} factor), and on the velocity of gas bubbles escaping from the solution. For the reactor under study, this time constant varied from milliseconds to seconds depending on the time scale of reactivity addition.

Since BeO has a very large absorption mean free path for thermal neutrons (λ_a for beryllium ~ 1500 cm), it moderates and reflects a considerable fraction of neutrons back into the core. This introduces a finite delay time that may be long compared with the core neutron lifetime. The effect of these reflector-returned neutrons on reactor dynamics was accounted for by defining an overall neutron lifetime (l_{eff}) for the entire reactor. Calculations using the one-dimensional static (without delayed neutrons) transport theory code DTF-IV in an alpha mode showed that the effect of a BeO reflector for the well-reflected thermal core was to stretch the mean prompt neutron lifetime by a factor of ~2 (l_{eff} = 33 to 62 μs) over the core neutron lifetime (l_c = 16 to 30 μs). This higher value of l_p had a beneficial effect on the safety of the solution reactor by making the power transient milder and by reducing the peak pressure by a factor of ~1.5. Neutron effectiveness calculations for the system performed by using the multigroup flux and adjoint flux values obtained from the DTF-IV code showed that delayed neutrons were ~22% more effective (β_{eff} = 1.22 β; β = 0.00266) than the prompt neutrons in causing fission, but for the bare equivalent system, β_{eff} was ~28% higher. This lower value of β_{eff} for the reflected system happened because in the presence of a strongly moderating reflector such as BeO surrounding the core, both prompt and delayed neutrons were rapidly thermalized and were either returned to the core or leaked out of the reactor assembly. This partly smeared out the difference in energy between the prompt and the delayed neutrons resulting in a lower value for β_{eff}. The higher reactivity effect (greater than unity) of delayed neutrons was expected since the core was highly thermalized with the majority of the fissions occurring in the thermal region and since the lower energy of delayed neutrons ($\bar{E}_{dn}$ ~ 400 keV) compared with prompt neutrons ($\bar{E}_{pn}$ ~ 2 MeV) means a higher fission probability and a higher nonleakage probability from the system while slowing down.[4] Variation of neutron effectiveness in the fuel solution concentration range of interest (140 to 80 g/ℓ) was found to be negligible. Since beryllium has a photoneutron threshold of only 1.67 MeV, the contribution of delayed photoneutrons (produced entirely in the reflector by fission product gammas) to the reactor period was investigated by using the Inhour equation. A nine-group mathematical model for the beryllium delayed photoneutrons was used, and the total photoneutron worth was conservatively assumed to be 0.08 mk. It was found that the photoneutron reactivity effect was negligible for periods less than ~10 s.

The dynamic calculations were performed for the case of a 5.9-ℓ-vol-solution core (fuel concentration = 90 g/ℓ) with a reciprocal core heat capacity equal to 43.2°C/MJ, a temperature reactivity feedback coefficient equal to −0.05 mk/°C, a void reactivity feedback coefficient equal to −270 mk per unit fractional void change, a mean prompt neutron lifetime equal to 40 μs, and β_{eff}(1 \$) equal to 0.00325. The power excursion was initiated by adding reactivity to an initially just-critical reactor at rest by the sudden insertion of a "step," the step occurring in a time interval short compared with the mean prompt neutron lifetime. Step reactivities inserted were in the range of ~13 to 1 \$ corresponding to stable reactor periods T of ~1 to 20 ms. The radiolytic void volume $V(t)$ at time t was calculated by using a pressure model developed by Spiegler et al.[5] A dynamic pressure wave was produced during short period (milliseconds) transients by the build-up and subsequent relaxation of the inertial pressure field generated by the accumulation and growth of H_2 gas bubbles (nucleation time of gas bubbles ~200 μs) produced during radiolysis. The threshold energy for the appearance of this shock wave was calculated to be 0.69 MJ with the peak-to-average flux ratio equal to 1.15. As the pressure relaxed by the actual physical expansion of the solution, a large negative reactivity was produced, which shut down the system. The pressure relaxation time constant RC was calculated to be 2.7 ms. Peak pressure varied from 726 to 61 psi for reactivity steps ranging from 13.3 \$ (T = 1 ms) to 2.2 \$ (T = 10 ms). The pulse width (full-width at half-maximum) θ was found to be given by

$$\theta = T + 0.7RC \ .$$

Since the shock wave was felt by the core vessel, which had to withstand its physical impact, from a reactivity safety point of view, the peak pressure was restricted to a nominal value of 100 psi. Consequently, the maximum reactivity step (or equivalent ramp) was limited to 3 \$ and the reactor period to ≥6 ms. The corresponding peak power varied from ~6000 to 70 MW. Figure 1 shows the typical behavior of the calculated power in pulse operation when the core was submitted to a 2.2 \$ reactivity step. Alongside the figure is shown a similar power trace from the SILENE experiments.[6] Figure 2 shows that the integrated prompt burst fission yield could vary from ~3 × 10^{16} to 6 × 10^{17} fissions for step reactivity injections ranging from ~5 to 42 mk (~1.5 to 13 \$). Also plotted in the figure are data obtained from the experimental solution pulse reactor SILENE

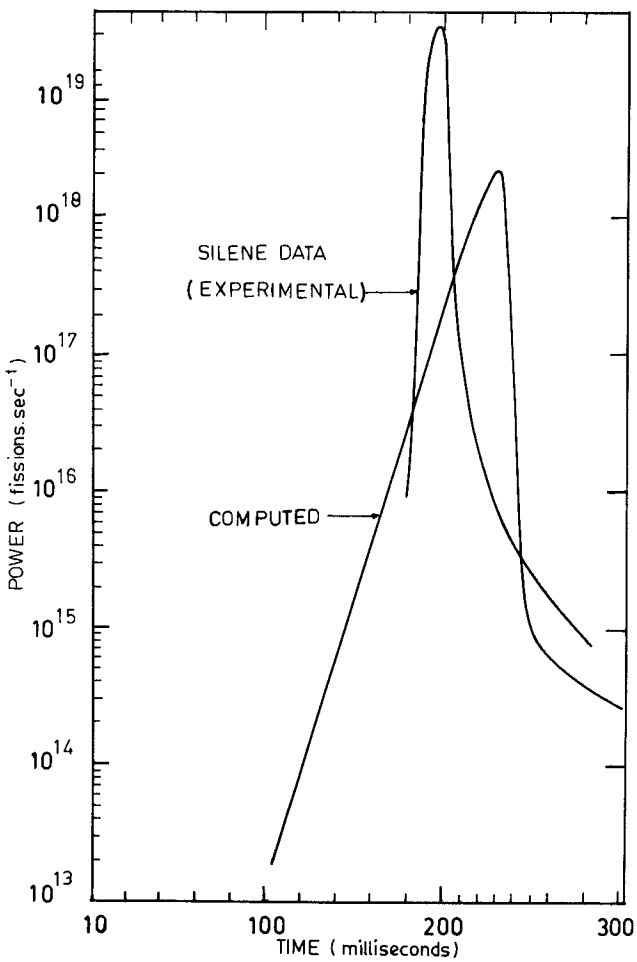

Fig. 1. Power behavior in pulse operation of a homogeneous solution reactor.

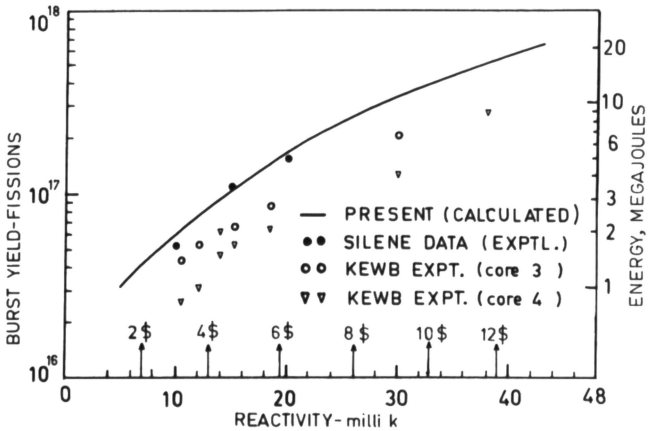

Fig. 2. Variation of total energy in burst as a function of injected reactivity in pulse operation of three homogeneous solution reactors.

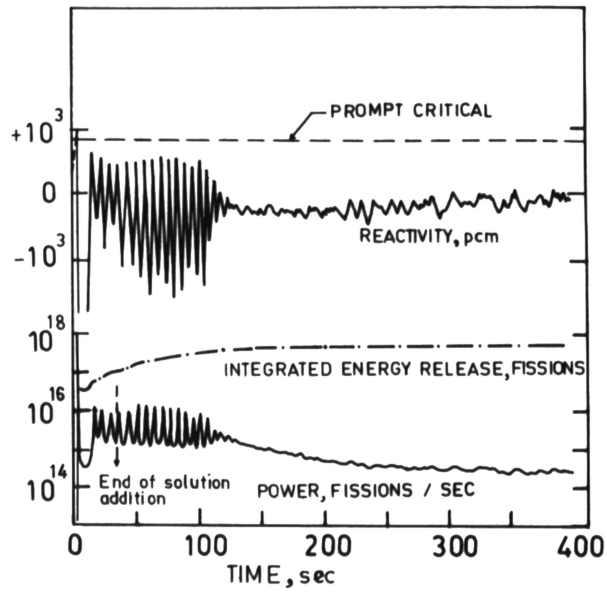

Fig. 3. Time variation of instantaneous power, integrated energy release, and reactivity during a typical free evolution power excursion in a homogeneous solution reactor (experimental).

(Ref. 6) and the KEWB experiments.[7] The data from SILENE (β_{eff} = 0.00794, l_p = 33 μs, and temperature reactivity coefficient = −0.52 mk/°C), which had a six to seven times larger core solution mass than the present one, are in better agreement with our calculations. The lower yield in KEWB compared with the present results could be due to its higher β_{eff} (0.0308) and higher temperature coefficient of reactivity (−0.30 mk/°C), though l_p for KEWB was between 20 to 26 μs compared with the present value of 40 μs.

Analysis of the French criticality experiments[3] conducted with aqueous uranyl (uranium enriched to 93%) nitrate solution showed that the major part (mostly 80 to 100%) of the energy release occurred during the pulsing/oscillating portion. In the majority of the experiments, energy in the first pulse contributed to ~10% of the total yield; in 12 experiments, this fraction exceeded 12% becoming as high as 60% of the total in a couple of them. These criticality excursions were performed in stainless steel cylinders of 30-cm diameter (3-mm wall thickness) and 80-cm diameter (4-mm wall thickness). Concentration of ^{235}U ranged from 20 to 400 g/ℓ, and critical solution volume from 19 to 231 ℓ. Both step and ramp reactivity excursions were made with the maximum step limited to <55 ¢, and the ramp reactivity insertion rate range from ~1 to <96 ¢/s. The minimum instantaneous period obtained in the experiments extended from a few seconds to less than a millisecond. The observed peak powers varied from $\sim 10^{15}$ fission/s (~30 kW) to $\sim 3 \times 10^{19}$ fission/s (~1000 MW). Total yield (integrated fissions) in the experiments varied from 2×10^{16} to 5×10^{18}.

The total yield Y_f in number of fissions when plotted against the duration of the experiment was found to be approximately five times greater in experiments with an 800-mm-diam tank then with the 300-mm-diam tank. This was due to the larger quantity of fissile solution in the former. It was found empirically that the total yield in an experiment was directly proportional to the product of the total solution volume in cubic centimetres and the total potential reactivity introduced, the proportionality constant (in fissions per cubic centimetres) increasing approximately in the ratio of tank diameters. It was $\sim 2 \times 10^{14}$ for the 30-cm-diam tank and $\sim 5.5 \times 10^{14}$ for the 80-cm-diam tank. To gain greater insight into the physics of criticality excursions in fissile solution systems when reactivity is added linearly in time, data from the CRAC-23 experiment (Fig. 3) were analyzed. In this experiment, reactivity was added at the rate of ~0.31 \$/s (1 \$ = 8.15 mk = 815 pcm in this experiment) for 38.6 s (time measured from delayed criticality). It is seen that after reaching prompt criticality, power rises rapidly reaching a peak value of $\sim 1.1 \times 10^{18}$ fission/s at $t \sim 4.6$ s due to the large negative reactivity feedback produced by ~5 ℓ of radiolytic gas during the prompt critical spike (increase in solution temperature during this time was ~10°C). Following this, the system gains reactivity because of the continuing addition of fissile solution and because of the release of radiolytic gas, and the power rises for the second time after reaching a minimum at $t \sim 9$ s. As the power level increases, more fissions take place producing more bubbles, which results in the negative reactivity feedback, and power reaches its second maximum. This process continues. Calculations of the delayed neutron effective source strength[8] in the sub-prompt-critical phase showed that its variation with time was between 30 to 40%, but the oscillations in power are seen to change by a factor of ~10. This means that it is the reactivity swing due to bubble production and escape that gives rise to the series of 19 oscillations in reactivity and power that occur in phase with their maxima and minima at the same points on the time axis. These oscillations last until ~130 s when the reactivity loss due to thermal expansion, radiolytic gas formation, and steam bubble production exceed the reactivity gain of ~97 mk due to the solution addition. Following this, the system becomes sub-delayed critical ($\Delta k \approx -300$ pcm), and the reactor power falls because of the decay of delayed neutron precursors (there was no extraneous source in this experiment) eventually tending toward a condition of equilibrium at a relatively low value of ~9.7 kW or 3×10^{14} fission/s when reactivity reaches its constant value at ~300 s. Long-term oscillations in power appearing in the experiments after this phase were due to heat exchanges. Total yield in this experiment was measured to be $\sim 5.3 \times 10^{17}$ fissions of which ~81% (4.3×10^{17} fissions) was produced in the oscillating portion. Assuming that all the gas produced in the core solution volume by radiolysis results in a uniform production of bubbles all of which escape from the solution, the void fraction V_f −

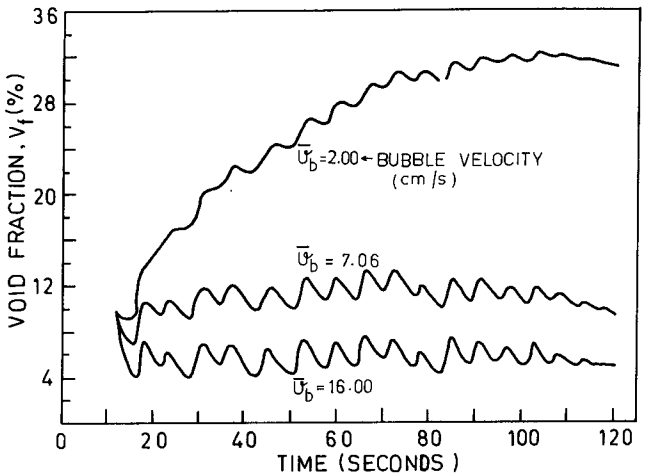

Fig. 4. Computed time variation of void fraction obtained from analysis of experimental data.

defined as the ratio of void volume (bubble volume plus increase in volume due to thermal expansion) to the total volume (void volume plus the solution volume)—at any instant of time was computed from the following simple equation taking the radiolytic gas yield G_{H_2} as 0.005 cm^3/J and the time interval $\Delta t = 10^{-3}$ s.

Note that

$$\Delta V_b = G_{H_2} \cdot \Delta E - A \cdot \bar{V}_b \cdot V_{bf}(0) \cdot \Delta t \ ,$$

where

$\Delta V_b, \Delta E$ = incremental changes in the volume of bubbles and energy, respectively, during Δt

A = cross-sectional area of the core tank

$\bar{V}_b$ = average bubble velocity

$V_{bf}(0)$ = bubble fraction at the beginning of Δt

$\Delta E = \int P.$

The term Δt was computed from the experimental power data of Fig. 3. Figure 4 gives plots of V_f against time for three different values of $\bar{V}_b$. It is seen that for $\bar{V}_b \sim 7.1$ cm/s, the void fraction oscillates with an amplitude of ~2 to 3% around an average value of ~12%. The number of oscillations in power equals the number of oscillations in void fraction, and their points of maxima and minima occur at the same time points. During the oscillation period (~15 to 120 s), the system crosses delayed criticality 35 times, and the void fraction at $k = 1$ increases from ~6 to 13%. After the termination of the solution addition at $t \approx 39$ s, at points where $k(t) = 1$, the excess reactivity (~97 mk) in the system exactly balances the 13% void fraction. Therefore, the observed swing of ~20 mk in reactivity after solution addition has stopped should correspond to the 2 to 3% swing in V_f. Thus, the oscillations in reactivity and power are due to the appearance and disappearance of the gas bubbles. The figure also shows that the total number of oscillations in void fraction (and hence reactivity and power) is relatively insensitive to the actual magnitude of the bubble velocity. The magnitude of $\bar{V}_b$ obtained from the foregoing analysis of the experimental power trace was found to be much more sensitive to the value of V_{f0} immediately after the first prompt-critical power burst than to the value of G_{H_2}.

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Response to "Comment on 'Reactivity Feedback Mechanisms in Aqueous Fissile Solutions'"

Thank you for bringing to my attention the work you have performed regarding excursions in fissile solution assemblies. Reference 1 discusses the reactivity effects that arise from temperature changes in the assembly. It is correctly noted that temperature-related reactivity effects are permanently present in the dynamics of an excursion and that void reactivity effects are more difficult to model as bubbles move vertically through regions of varying worth and eventually out the solution's surface. Void reactivity effects are not discussed in Ref. 1, as it focuses strictly on the effects of temperature on reactivity feedbacks, where the temperature feedback is divided into two parts: density effects and flux spectral shifting effects. A goal of the temperature feedback analysis was to determine if low-concentration plutonium solution assemblies could have a net positive temperature feedback coefficient. In an excursion in such an assembly, the shutdown mechanism is void production. It is certainly true that void reactivity effects play a crucial role in the dynamics of solution assemblies.