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HEATING-UP, NUCLEATION AND BOILING OF A CRITICAL SOLUTION OF FISSILE MATERIAL

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Abstract—Applied to a homogeneous solution of uranyl nitrate or plutonium nitrate, the physical mechanisms are discussed which control the mean nuclear power of a solution reactor. The short-time averaged power during the heating-up of a nonboiling solution is related to the mean flow rate of radiolytic gas which is necessary to yield a steady-state neutron flux. If the solution is boiling a similar relation to the flow rate of vapor is derived. This relation, however, can only be applied if the superheat of the solution permits vapor bubble growth. Otherwise, the minimum power for the required superheat will be obtained instead. These power relations are combined into a quasi-steady-state model which is discussed and verified by applying it to two experiments of the CRAC solution reactor.

1. INTRODUCTION

Relations for the nuclear power of critical solutions of fissile material have become of major importance in reprocessing. During the reprocessing of LWR and FBR spent fuel uranyl nitrate and plutonium nitrate solutions have to be stored in vessels which are designed small enough to prevent criticality of the fissile material in any achievable concentration. However, eight criticality accidents have been reported since 1958 which were mainly caused by an uncontrolled transfer of fuel solutions into unsuitable vessels. Therefore an accident analysis is required prior to the start-up of a reprocessing plant, in order to minimize the consequences of any conceivable excursion. This accident analysis should describe the whole excursion including the heating-up of the solution, the boiling phenomena and the self-controlled end after some minutes, caused by the evaporation of water. Previous models, however, were restricted to the first few power bursts as described below.

Experimental investigations of criticality excursions were first performed in the KEWB reactor, as reported by Dunenfeld & Stitt (1963). There, a short-time excursion of a highly-enriched uranyl nitrate solution was examined which was kept in a cylindrical or a spherical vessel. When criticality was initiated by pulling a control rod, only a single power burst of about 10–20 ms duration was observed. This single peak could be modeled using the space-independent kinetics equations for prompt and delayed neutrons. The reactivity feedback mechanism which caused the power to decrease again, was related to the temperature increase and to the sudden production of radiolytic gas bubbles which occurred as soon as a certain concentration of dissolved radiolytic gas was exceeded in the solution.

The first long-term excursions and boiling experiments were performed in the CRAC experimental facility, reported by Lécorché & Seale (1973). There a solution of highly-enriched uranyl nitrate was continuously fed into a vertical cylindrical vessel. After having reached the supercritical volume of the solution and even beyond the initial pulse further solution was added. This led to a series of power bursts which showed in general a decreasing power amplitude. If boiling of the solution was achieved, a constant power was obtained. The excursion could be terminated by draining the liquid, or it stopped by itself after a sufficient amount of water had evaporated from the solution.

In order to model this series of power bursts, the short-time model of Dunenfeld & Stitt (1963) was improved by Weber & Denk (1984) by including one-dimensional equations, and by adding an empirical relation for the succeeding escape of the radiolytic gas bubbles

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from the pool. This allowed them to model the renewed rise of the power after the first pulse. However, remarkable deviations from the experimental results occurred after a few periods, and application to a boiling liquid turned out to be impossible. Moreover, long computer times are expected if this model could be extended to include boiling.

In this paper it will be shown that, deviating from previous models, the mechanisms of nucleation and bubble growth are of major importance for the power of a boiling solution. Therefore, more care had to be taken to model the two-phase flow phenomena rather than the nuclear phenomena. Moreover, since only long-term power excursions were considered, the model was reduced to a quasi-steady-state approach which means that single power bursts were not included. This approach reduces computer times by several orders of magnitude.

2. THE QUASI-STEADY-STATE MODEL

The following model differs from previous models by the balance equations for neutrons and gas flow rates. Here, for a given temperature and a given concentration of fuel (uranyl nitrate or plutonium nitrate) a stationary neutron flux and a stationary flow rate of radiolytic gas or vapor are assumed. Thus the fast changes of the neutron flux and of the gas flow rate are neglected, compared with the slow changes of the temperature and the concentration of the fuel. This approach is called a quasi-steady-state model. It is the aim of this approach to calculate the history of the mean power rather than short-time power bursts.

The model is restricted to vertical, cylindrical vessels. A homogeneous distribution of fuel, temperature and void are assumed.

2.1. Neutron balance equation

Restricting to two energy groups, the stationary neutron diffusion equation requires the following condition for the geometrical buckling B^2:

$$B^2 = \frac{k_\infty - 1}{L^2 + \tau}, \qquad [1]$$

where k_∞ is the neutron multiplication factor of the infinite medium, L is the thermal diffusion length and τ is the Fermi age. For cylindrical vessels this geometrical buckling B^2 is related to the inner radius R of the vessel and to the liquid height H as

$$B^2 = \left(\frac{2.405}{R + \lambda}\right)^2 + \left(\frac{\pi}{H + 2\lambda}\right)^2, \qquad [2]$$

where λ is the extrapolation length. As no reflector around the vessel is considered here, a value of 2.5 cm can be taken to estimate the extrapolation length.

The parameters L, τ and H are functions of the temperature T and of the mean void α generated by the rising bubbles. In first order this dependence is only caused by the variation of the density. This effect can be separated with a density factor D as

$$\tau = \tau_0 D^2 \qquad [3]$$

$$H = H_0 D, \qquad [4]$$

with

$$D = \frac{1 + \int_{T_0}^{T} \beta \, \mathrm{d}T}{1 - \alpha}.$$

Here β is the thermal expansion coefficient. The index 0 denotes the values of τ and H, respectively, at the reference temperature T_0 (20°C) and without any void.

Because the thermal spectrum of neutrons also depends on the temperature, a further correction should be added beyond the density factor D to calculate the diffusion length

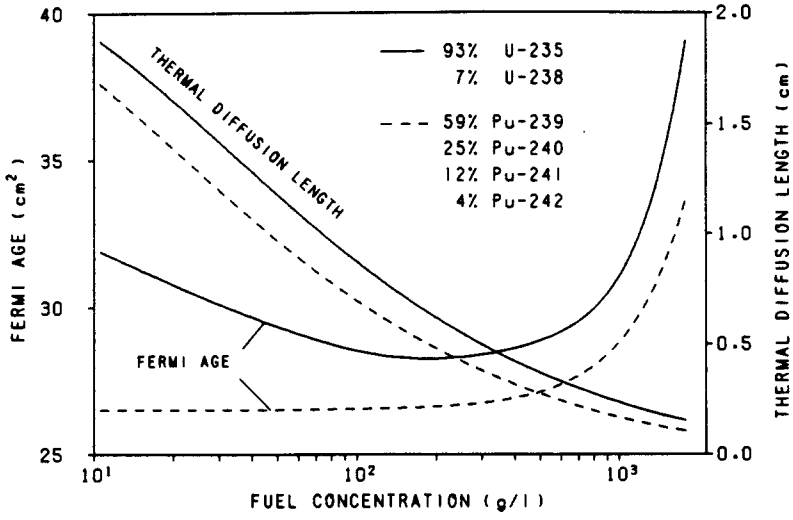

Figure 1. Thermal diffusion length and Fermi age at 20°C without any void.

L. We obtain

$$L = L_0\left(\frac{T}{T_0}\right)^{1/2} D. \qquad [5]$$

For a given enrichment the parameters L_0 and τ_0 depend only on the concentration of the fuel and, to a lesser extent, on the concentration of the nitric acid. In figure 1 these parameters are plotted vs the fuel concentration for a highly-enriched uranium nitrate solution and for a plutonium nitrate solution of a typical composition, such as occurs in LWR reprocessing plants. These data have been taken from the criticality handbooks by Carter *et al.* (1968) and by Heinicke *et al.* (1979), which were computed with the GAMTEC code, as described by Carter *et al.* (1965).

In heterogeneous reactors the multiplication factor k_∞ depends on the temperature of the fissile material, mainly because of the Doppler effect. In homogeneous fuel solutions this Doppler effect does not contribute significantly to the overall temperature dependence of the geometrical buckling, because the temperature is limited by the saturation temperature, and the density of the fuel is small compared with that of the moderator. Moreover, only the ratio of fuel density to moderator density determines k_∞, which is not altered by the void. Therefore, in first order, the dependence of k_∞ on the temperature and the void can be neglected.

In figure 2 the multiplication factor is plotted as a function of the fuel concentration for uranium nitrate and plutonium nitrate solutions as specified in figure 1.

When [1]–[5] are combined, the mean void α can be calculated as a function of the temperature and fuel concentration. If the extrapolation length λ is small compared to the liquid height, we obtain approximately

$$\alpha = 1 - \left(\frac{2.405}{R+\lambda}\right)\left[\frac{k_\infty - 1}{L_0^2\left(\frac{T}{T_0}\right) + \tau_0} - \left(\frac{\pi}{H_0 + 2\lambda}\right)^2\right]^{-\frac{1}{2}} \cdot \left(1 + \int_{T_0}^{T} \beta \, dT\right). \qquad [6]$$

This mean void is required to obtain a steady state for a critical system with a given temperature and a given fuel concentration. Next the mean power will be determined as a function of this mean void. Different relations will be obtained because different mechanisms exist to produce this void.

2.2. Nonboiling liquids

If the temperature of the solution stays below the saturation temperature, the required mean void can only be produced by radiolytic gas bubbles. This gas will mainly consist

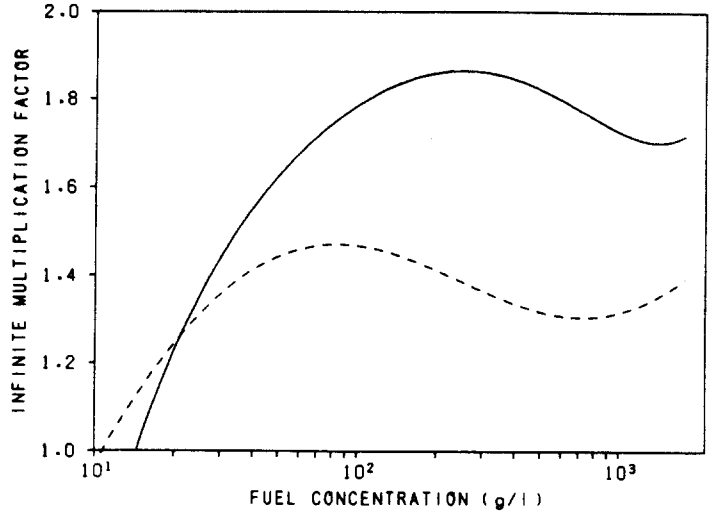

Figure 2. Neutron multiplication factor of an infinite medium for the fuel solutions specified in figure 1.

of H_2 and O_2. If a quasi-steady-state is assumed, the total volumetric gas flow rate $\dot{V}_G$ from the surface of the solution is proportional to the total power Q in the solution.

Experimental data for fission-product radiolysis and α-radiolysis of H_2 and O_2 have been provided by Bibler (1974), Savel'ev *et al.* (1967) and Spiegler *et al.* (1962). Combining these data we obtain the fit

$$\dot{V}_G = [(1 - 0.36\, C_N^{0.45})(2.25 - 0.2\, C^{0.33}) + 0.15 + 0.17\, C_N^{0.7}] \cdot 2.82\, Q, \qquad [7]$$

for $\dot{V}_G$ [l/s] and Q [MW]. Here C_N is the molarity of the nitric acid [mol/l] and C is the fuel concentration [g/l]. Equation [7] was compared with the measured gas flow rates of the two solution reactors KEWB, as reported by Spiegler *et al.* (1962), and SILENE, as reported by Barbry & Manaranche (1982). These data are generally underpredicted by 10–20%, but the dependence on the fuel concentration is well-reproduced.

Assuming that the local gas flow rate increases linearly with the height in the liquid, the mean superficial gas velocity u_{SG} within the liquid is approximately half of the superficial gas velocity above the liquid. Then we obtain

$$u_{SG} = \frac{\dot{V}_G}{2\pi R^2}. \qquad [8]$$

A relation between the superficial gas velocity and the mean void can be derived from the drift flux model of Wallis (1969). If $\alpha \leqslant 0.4$, we obtain

$$u_{SG} = v_\infty \alpha (1 - \alpha), \qquad [9]$$

where v_∞ is the rise velocity of single bubbles. An estimate of this velocity can be derived from the period of oscillations observed in the CRAC experiments. We assume that the time difference between two successive power bursts is essentially determined by the time which a swarm of bubbles takes to escape from the liquid. As the rise velocity will be constant if the bubbles are sufficiently small, we estimate from the results of Lécorché & Seale (1973) that

$$v_\infty \approx 5\ \text{cm/s}. \qquad [10]$$

Now [6]–[10] can be combined to yield an explicit equation for the mean power as a function of a given temperature, fuel concentration and liquid mass. This power will be denoted as Q_N for further distinction, where the index N is chosen to indicate nonboiling liquids.

2.3. Fully developed nucleate boiling

If the saturation temperature can be achieved, nearly all the heat produced is needed for evaporation. Then the relation between the gas flow rate $\dot{V}_G$ and the mean power Q is given as

$$\dot{V}_G = \frac{Q}{\rho_G \Delta h}, \qquad [11]$$

where ρ_G is the density of the vapor, and Δh is the enthalpy of evaporation. The flow rate of the radiolytic gas can be neglected compared to this flow rate.

We expect that vapor bubbles will be larger than radiolytic gas bubbles, so we propose the relation of Zuber & Findlay, as reported by Wallis (1969), instead of [9]. This relation reads

$$u_{SG} = 1.53 \frac{\alpha}{1 - 1.2\alpha} \left[\frac{\sigma (\rho_L - \rho_G) g}{\rho_L^2} \right]^{\frac{1}{4}}, \qquad [12]$$

where σ is the surface tension, g is the acceleration due to gravity and ρ_L is the density of the liquid.

Combining [6], [8], [11] and [12] we obtain again an explicit equation for the mean power, which is denoted as Q_B, where the index B is chosen to indicate boiling. This achievable power Q_B will always be much smaller than the power Q_N, which is achieved if only radiolytic gas bubbles are generated. Since only a small part of the nuclear power produces radiolytic gas, a greater power is needed for nonboiling liquids to produce the same required void. However, if the equation for the power Q_B were applied to verify the CRAC experiments, the mean power would be underpredicted by about an order of magnitude. From this discrepancy we conclude that a further physical mechanism exists which controls the power when the liquid is boiling. This mechanism will be discussed next.

2.4. Partly suppressed nucleate boiling

If a fuel solution is boiling, nucleation can occur heterogeneously at the walls of the vessel and homogeneously within the liquid. In the following considerations we restrict ourselves to the homogeneous nucleation, which we expect to be dominant in large vessels. Spiegler *et al.* (1962) discuss the microscopic details of this homogeneous nucleation: if the fission products are slowed down, along a track of about 4 μm, they produce microscopic bubbles which consist of vapor and a small amount of radiolytic gas. Ghormley (1958) has shown that the diameter of these bubbles is approx. 1.4 μm. If the liquid is not sufficiently superheated these bubbles collapse within about 10^{-8} s, according to the model of Zwick & Plesset (1954). Afterwards smaller radiolytic gas bubbles remain which have a diameter of about 0.1 μm, and which collapse after about 10^{-4} s if the solubility of the radiolytic gas has not been exceeded.

These temporary vapor bubbles do not contribute significantly to the mean void but, neglecting the walls, they constitute the only nucleation sites. As a pressure difference of 0.16 MPa within these bubbles can be calculated from the bubble diameter, a local superheat of about 29°C is required for a growth of the microscopic vapor bubbles. This superheat, however, also leads to convective heat transport to the upper surface where it causes further evaporation, so that only part of the heat is available for bubble growth inside the liquid. Therefore, only a certain fraction of the total produced vapor will contribute to the mean void inside the liquid, the other part is produced at the upper surface in our restricted model. This means that [8] is invalid in this case.

The heat transfer from these local superheats cannot be determined in detail with our space-independent model. Instead, we can calculate the integral fraction of vapor which is produced as bubbles and the overall mean power which will be achieved. This power cannot be smaller than the power we need for the first nucleation in the absence of any bubbles. Therefore, we first consider the transient state of the onset of boiling, and by this the initial power which leads to a global superheat of 29°C. If we apply the results of an experiment of Fieg (1978) to determine the heat transfer from a volumetrically heated liquid in a vertical, cylindrical vessel the following relations hold:

$$\left.\begin{aligned} &\mathrm{Nu} = 1.67\,\mathrm{Ra}^{0.206} \quad \text{if} \quad \frac{H}{2R} \approx 1 \\ &\text{and} \\ &\mathrm{Nu} = 0.985\,\mathrm{Ra}^{0.207} \quad \text{if} \quad \frac{H}{2R} \approx 0.25. \end{aligned}\right\} \qquad [13]$$

Here Nu is the Nusselt number and Ra is the internal Rayleigh number, which are defined as

$$\mathrm{Nu} = \frac{q\,H^2}{k\,\Delta T} \quad \text{and} \quad \mathrm{Ra} = \frac{\beta\,g\,q H^5}{k\,\kappa\,\nu}. \qquad [14]$$

In these definitions q is the power density, k is the thermal conductivity, ΔT is the superheat, κ is the thermal diffusivity and ν is the kinematic viscosity. Equations [13] include some heat losses to the side walls and to the bottom of the vessel which should account approximately for the nonuniform power profile in this case of pure homogeneous boiling.

Equations [13] determine the internal heat which is required for a given temperature difference between the liquid and its upper surface. Using the properties of a uranyl nitrate solution at saturation temperature, [13] can be simplified to yield the initial power for the required superheat, which is also the minimum power for vapor bubble growth. We obtain

$$Q_{\min} = \mathrm{Fr}\,\frac{\pi R^2}{H^{0.2}}, \qquad [15]$$

with

$$\mathrm{Fr} \approx 70 \quad \text{if} \quad \frac{H}{2R} = 1 \quad \text{and} \quad \mathrm{Fr} \approx 35 \quad \text{if} \quad \frac{H}{2R} = 0.25.$$

Here $Q_{\min}$ is in W and R and H in cm. Of course, this relation is independent of the fuel concentration. It describes the third physical mechanism which controls the mean power.

Until now we have only considered a transient state, the onset of boiling. In the following considerations, however, we will show that this mechanism can also explain the steady power which was observed during boiling in most of the experiments. In order to decide which mechanism will be obtained if the liquid is boiling, we distinguish the following three cases:

(a) Fully developed nucleate boiling can only occur if the power Q_B, calculated in section 2.3, exceeds the minimum power, i.e. if $Q_B \geqslant Q_{\min}$. Then, even the low power Q_B will be sufficiently high to produce a global superheat $>20°C$. In this case the power Q_B will be obtained as the mean power, and the total amount of vapor produced will contribute to the void in the liquid. However, this state could never be achieved in any of the CRAC experiments.

(b) On the other hand, growth of the bubbles will be completely suppressed if the power Q_N, which is the power without any vapor as calculated in section 2.2, is less than the minimum power ($Q_N \leqslant Q_{\min}$). Then, even the highest achievable power, Q_N, is never sufficient to produce a global superheat of 29°C. Although the saturation temperature has been reached, we expect a similar situation to that in nonboiling liquids. In this case the power Q_N will be obtained and all vapor should be produced at the upper surface. However, bubble growth can still occur periodically from macroscopic radiolytic gas bubbles. This latter effect will be discussed in more detail in section 3.

(c) If the power Q_N is in excess of the minimum power $Q_{\min}$, but the power Q_B of fully developed nucleate boiling is below it, i.e. if $Q_B \leqslant Q_{\min} \leqslant Q_N$, we expect partly suppressed nucleate boiling. Then only as much power will

be achieved as is required for the global superheat of 29°C. Thus the power Q_{min} will be obtained, independent of Q_B and Q_N.

The following argument supports this conclusion: any power below Q_{min} will never lead to evaporation within the liquid, which would result in an increased power Q_N. On the other hand, any power in excess of Q_{min} leads to fully developed nucleate boiling which, in turn, would result in a power $Q_B < Q_{min}$. Therefore, we can conclude the occurrence of Q_{min} from a contradiction. This implies that we require the simultaneous distribution of bubbles and superheats.

The fraction f of the vapor which is produced in bubbles can now be determined from the overall mean power Q_{min}. The volumetric flow rate of the vapor bubbles is still proportional to the superficial velocity, as described by [6] and [12]. Therefore, this flow rate is given by [11], if Q is chosen to be Q_B. On the other hand, the total flow rate above the liquid is proportional to Q_{min}. These arguments yield the following simple equation:

$$f = \frac{Q_B}{Q_{min}}. \qquad [16]$$

Under real conditions there will also be some heterogeneous nucleation at the side wall and at the bottom of the vessel. Usually the nucleation sites of the walls initiate vapor bubbles with a superheat which is far less than the superheat for a homogeneous nucleation, as described above. However, this contribution is small if the vessel is large and if the walls are cold. For the extreme case that this heterogeneous nucleation dominates (e.g. if the vessel were filled with grids or rods), we conclude from the considerations above that the power Q_B of fully developed nucleate boiling will always occur. Therefore any general case with a finite contribution of the walls will lead to a certain power between Q_B and Q_{min}. This power, however, cannot be determined with the tools of our model, since an accumulation of bubbles near the walls, and so at least a one-dimensional distribution of the void, must be considered.

2.5. Computation of a total excursion

Now the relation for the mean power can be inserted into the balance equations for heat and mass. For a nonboiling liquid, the space-independent heat equation, including heat losses to the surrounding atmosphere, reads

$$m_L c_p \frac{dT}{dt} + \dot{m}_{in} c_p (T - T_{in}) = Q_N - h_w A (T - T_\infty). \qquad [17]$$

Here m_L is the actual liquid mass, $\dot{m}_{in}$ and T_{in} are the mass flow rate and the temperature of additional liquid, respectively, c_p is the heat capacity, Q_N is the power as determined in section 2.2, h_w is the heat transport coefficient, A is the wetted area of the side wall and of the bottom of the vessel and T_∞ is the temperature of the surrounding atmosphere.

The liquid mass m_L and the height of the cold liquid, H_0, are related by

$$m_L = \rho_{L0} \pi R^2 H_0, \qquad [18]$$

where ρ_{L0} is the density of the solution at the reference temperature T_0.

If the fuel concentration of the added liquid differs from that of the accumulated liquid, the fuel concentration varies with time according to

$$m_L \frac{dC}{dt} = (C_{in} - C)\dot{m}_{in}. \qquad [19]$$

If the solution is boiling the mass balance of the solution reads

$$\frac{dm_L}{dt} = \dot{m}_{in} - \frac{1}{\Delta h}[Q - h_w A (T - T_\infty) - \dot{m}_{in} c_p (T - T_{in})]. \qquad [20]$$

There Q is one of the three powers Q_{min}, Q_B and Q_N, according to the considerations in section 2.4. In this case the heat equation determines the increase in the fuel concentration as

$$m_L \frac{dC}{dt} = (C_{in} - C)\dot{m}_{in} + \frac{C}{\Delta h}[Q - h_w A(T - T_\infty) - \dot{m}_{in} c_p (T - T_{in})]. \quad [21]$$

Equations [17]–[21] were solved numerically using an explicit finite-difference method of first order.

3. APPLICATION TO THE CRAC EXPERIMENTS

In order to verify the model it was applied to some experiments of the CRAC test facility. This solution reactor is described by Lécorché & Seale (1973), together with a review of the various experiments. Two boiling-type experiments have been selected for discussion here.

The experiment CRAC 16, which is described in more detail by Barbry *et al.* (1970), was conducted in a vertical, cylindrical vessel of 300 mm dia. An aqueous solution of uranyl nitrate with an enrichment of 93%, and a concentration of 76.6 g/l U-235, was continuously fed to the vessel with a steady mass flow rate of 443 l/h, until 54 l. had been inserted. At a height of 42 cm the solution became critical, causing the temperature to rise within about 5 min to the saturation temperature. The addition of liquid was terminated 205 s after the solution became critical.

The power was unsteady during the heating up. A mean power of about $3 \cdot 10^{15}$ fissions/s (90 kW) was obtained. When the solution started boiling the oscillations damped out, and the power was reduced to $4 \cdot 10^{14}$ fissions/s (12 kW). After 12 min the excursion was terminated by draining the solution. Up until then nearly 10^{18} fissions had occurred.

This experiment was simulated numerically with the quasi-steady-state model, using a time step of 2 s. Results are shown in figures 3–5. Due to the approximations of this model the oscillations could not be reproduced, but the integrated mean power agreed well with the observed increase of the total energy.

When the saturation temperature has been reached the model predicts partly suppressed nucleate boiling. The predicted power is higher than the observed one by about a factor of 2. The discrepancy is believed to be due to the heterogeneous nucleation at the side wall. Only 4–6% of the vapor contribute to the mean void within the liquid. A superheat of

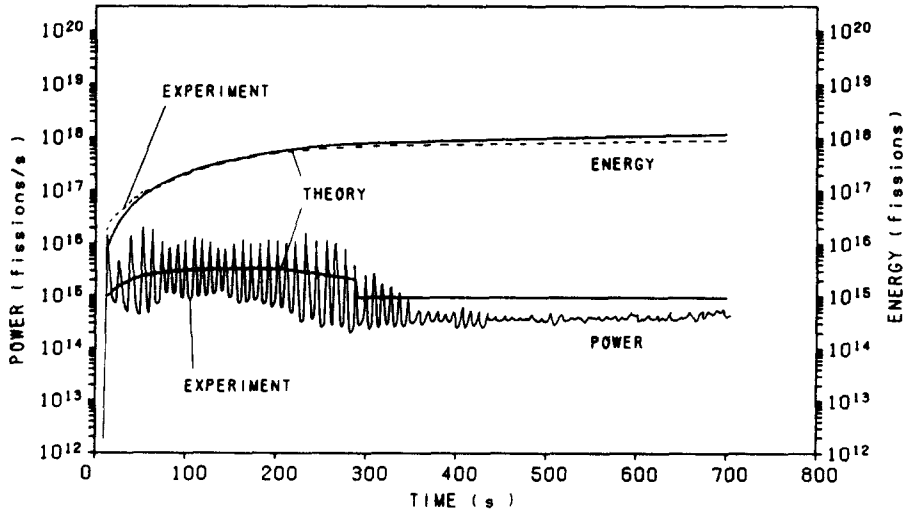

Figure 3. History of power and total energy released during the experiment CRAC 16 (Barbry *et al.* 1970) and comparison with the model.

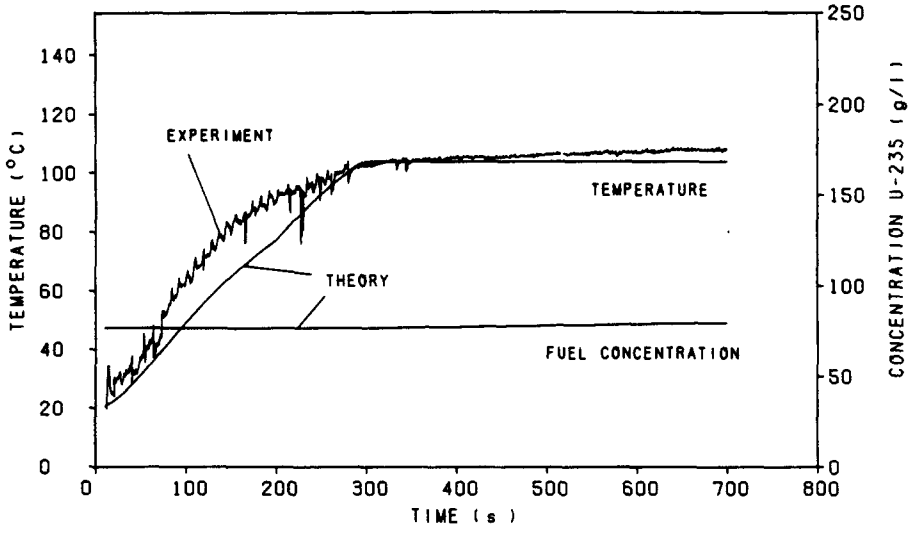

Figure 4. Increase of temperature and fuel concentration simulated for the experiment CRAC 16.

29°C was not recovered by the thermocouple during the experiment, probably because its surface provided further suitable nucleation sites.

The experiment CRAC 43, described by Barbry *et al.* (1971), was performed in a larger vessel of 800 mm dia. A higher fuel concentration of 188 g/l was used; 76.4 l. of uranyl nitrate solution were inserted with a mass flow rate of 1407 l/h. In this experiment the mean power was higher by about a factor of 5. The excursion stopped by itself. Liquid addition was terminated 19 s after the solution became critical.

This experiment was simulated numerically with a time step of 1 s. Results are shown in figures 6–8. During the heating up the mean power is overpredicted by nearly a factor of 2. In this experiment the pool diameter exceeded the liquid height by more than a factor of 5. This geometry is rather unsuitable for a space-independent model because, due to the nonuniform power profile, high differences in temperature and void are expected between the center of the pool and the outer parts of the liquid. In spite of this restriction an agreement was achieved which should be good enough for an accident analysis.

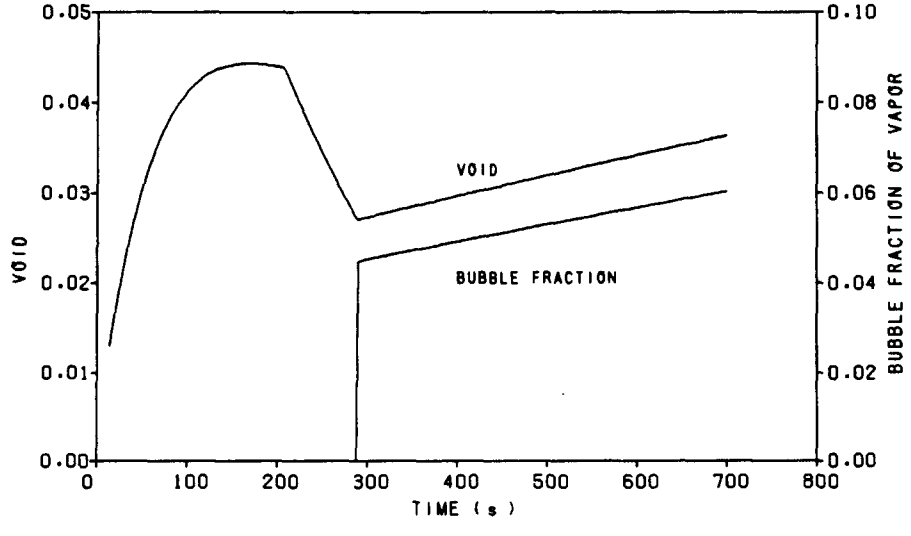

Figure 5. Model predictions of the void and the fraction of vapor released as bubbles during the experiment CRAC 16.

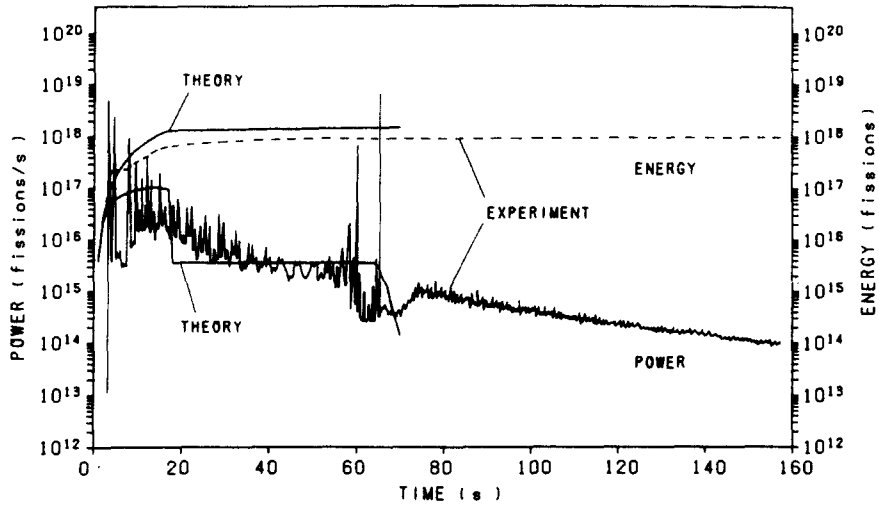

Figure 6. History of power and total energy released during the experiment CRAC 43 (Barbry *et al.* 1971) and comparison with the model.

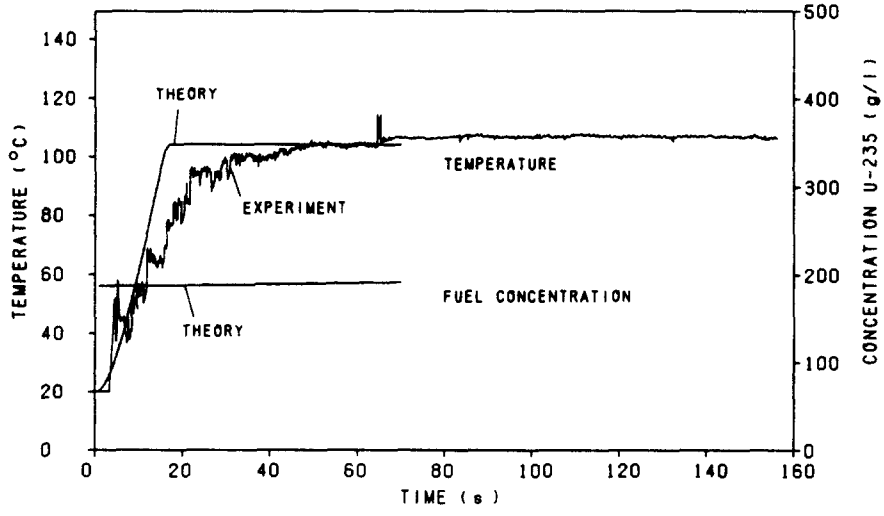

Figure 7. Increase of temperature and fuel concentration simulated for the experiment CRAC 43.

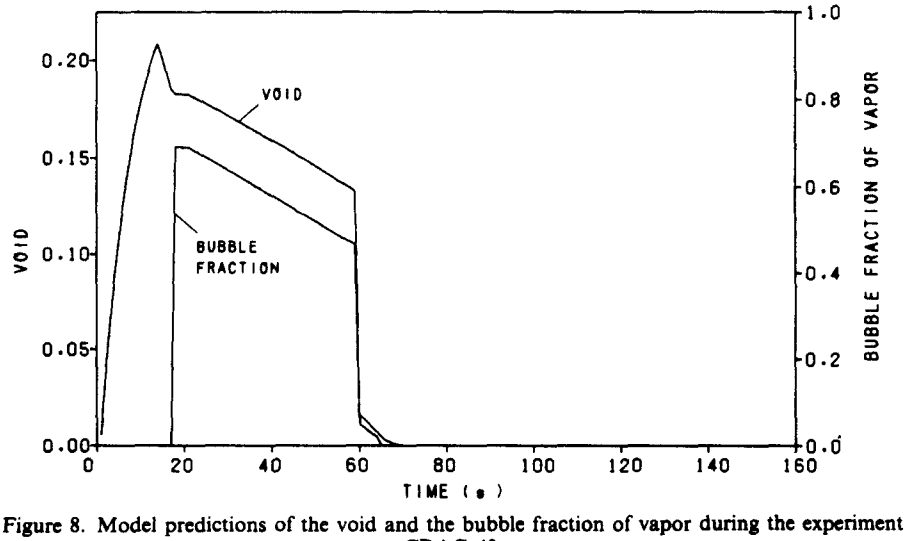

Figure 8. Model predictions of the void and the bubble fraction of vapor during the experiment CRAC 43.

Boiling was obtained in the experiment during the time interval from 40 to 60 s. Here again, partly suppressed nucleate boiling is predicted by the model, and the mean power is well-reproduced. The bubble fraction of vapor exceeded 70%. In contrast to the experiment CRAC 16, this fraction decreased quickly—probably faster in the experiment than predicted by the model.

After about 60 s some unexpected power bursts occurred, followed by a sudden superheat which was indicated by a thermocouple. Barbry *et al.* (1971) reported that an unknown amount of liquid was ejected from the vessel at that time. To simulate this loss of liquid it was assumed in the model that 2 l. of liquid were taken from the pool after 60 s. A sharp decrease of the bubble fraction of vapor resulted due to this correction, whereas the power remained on a high level. After 64 s the growth of the vapor bubbles was predicted to be completely suppressed. The power was then equal to Q_N, which decreased rapidly with time.

The considerations about partly suppressed nucleate boiling explain the observed power bursts as vapor explosions. Beyond our model predictions we assume that bubble growth was already completely suppressed after 60 s. However, each swarm of radiolytic gas bubbles which occurred periodically, like in a subcooled liquid, could actuate vaporization again. As the power was rather high these sudden evaporations could even occur explosively, ejecting liquid from the pool.

After 70 s the solution became subcritical and the excursion stopped by itself. The low power decrease which then followed is only due to delayed neutrons which are not included separately in our model. Therefore, the model predictions end at 70 s. Only 3% of the total energy released was produced by these delayed neutrons after 70 s, so we can justify this neglect.

4. CONCLUSIONS

The simple quasi-steady-state model provides short computer times and allows us to study a great number of previous experiments, and even real accidents, for the first time. As most of the model is algebraic, an advantageous insight into the physics of the excursions can be obtained.

Only one empirical parameter, the rise velocity of single radiolytic gas bubbles, was taken from the CRAC experiments, all other parameters were derived from independent experiments. Therefore the good agreement between the model and the CRAC experiments encourages application of the model to the real conditions of reprocessing plants. However, deviations of about a factor of 2 are expected for these applications.

It was the aim of this model to demonstrate the great importance of two-phase flow phenomena, especially if boiling solutions are concerned. Evidently, further improvements could be obtained in future models if a more detailed neutron balance were applied. Limits of the model will be attained if it is applied to shallow pools with a high power density. Then the distributions of temperature and void become significantly two-dimensional, whereas the model is space independent. For the same reason a nonuniform distribution of the fuel in the solution can lead to remarkable errors. Moreover, if heterogeneous nucleation sites dominate the power during boiling will be overpredicted, which can also only be modeled if at least one-dimensional equations are applied.

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