INTRAGRANULAR FISSION GAS BEHAVIOR DURING A SLOW THERMAL TRANSIENT

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ABSTRACT

A possible accident of interest to nuclear reactor designers is an accident where the core is successfully shut down but where the decay heat imposes a threat to the core integrity. One of the stages after the initiation of the accident, when the sodium has largely voided from the core region, may be characterized as one where the fuel undergoes nearly isothermal heating. The behavior of intragranular fission gas in this period of approximately 1500 seconds of slow isothermal heating is considered. The analysis, a spatially dependent multigroup formulation, considers bubble formation, bubble coalescence, and the migration of bubbles and/or gas atoms to grain boundaries; nonequilibrium bubbles are allowed. A finite difference technique is used for the spatially dependent solution. The analysis was used to predict intragranular fission gas behavior during 1) a thermal transient where the temperature increases linearly from 1160°K to 3000°K over a 1500 second period, and 2) a transient where the fuel is heated radially inward to a nearly uniform 2673°K in about 18 seconds and remains essentially isothermal at 2673°K for 12 more seconds (the FGR-34 studies at HEDL). The results for these transients admit the following conclusions: *For the 1500 second transient* - (1) The bubble behavior is dominated by coalescence, via random migration. The increased bubble size results in lower mobility and hence little of the intragranular fission gas reaches the grain boundary. The intragranular fission gas accounted for 14% increase in fuel volume and a gas release of 0.8% during the transient. (2) Allowing for nonequilibrium bubble condition results in transient swelling 15% below the results for the equilibrium treatment. (3) Grain size does not significantly affect intragranular swelling. Surface tension is an important factor. *For the 30 second transient* - (1) Highly overpressured bubbles are obtained in such transients. (2) The calculated bubble size distribution is one order smaller than the measured one because, among other factors, the attractive forces among overpressured bubbles that lead to enhanced coalescence is not included in the present analysis.

1. INTRODUCTION

A possible accident of interest to nuclear reactor designers is an accident sequence that involves a rupture of all piping connected to the reactor vessel with the core successfully shut down but where the decay heat imposes a threat to the core integrity [1]. The final analysis is to predict the sequence of physical processes leading to the disruption of the fuel element by melting or some dispersal mechanism caused by the fission product gas or volatile fission products. A major consideration in this analysis is the fuel swelling since recent DEH experiments [2,3] have shown the slower heating rates to lead to a larger net fuel swelling.

After the initiation of the accident, the sodium coolant will undergo a period of heat transfer via natural convection followed by a subcooled boiling and a bulk boiling time period involving around $2\mathrm{x}10^5$ seconds [1]. The fuel temperature during this time will be substantially reduced from the operating temperature and the intragranular fission gas will be essentially immobile. Once the sodium has largely voided from the core region, the heat transfer from the fuel element will be drastically reduced and the fuel will heat up nearly isothermally. It is this period of approximately 1500 seconds that is of major consideration herein with regard to fuel element behavior. In this paper we analyze the behavior of intragranular fission gas during this period of "isothermal" heating.

2. ANALYSIS

The isothermal intragranular fission gas model involves random gas motion resulting in bubble coalescence and migration to the grain boundaries. Resolution is not a factor for this transient. The formalism is a spatially dependent multigroup solution to the rate equation [4]. The main added features (beyond analyses available in the current literature) for this model are to allow for nonequilibrium states with bubble relaxation due to vacancy migration [5-7], and to allow for a linear distribution within each gas atom number group.

The time and spatial concentration C_i of bubbles that contain between n_i and n_{i+1} gas atoms are defined as

$$C_i(\rho,t) = \int_{n_i}^{n_{i+1}} C(n,\rho,t)dn \tag{1}$$

The multigroup form of the rate equation is

$$\frac{\partial C_i}{\partial t} = D_i\nabla^2 C_i - \sum_{j=1}^{i} A_{ij}(1-F_{ij})C_iC_j - \sum_{j=i}^{N} A_{ij}C_iC_j + \sum_{k=1}^{i-1}\sum_{j=1}^{k} A_{kj}F_{kji}C_kC_j \tag{2}$$

where

$A_{ij} = \alpha_{ij}4\pi(r_i+r_j)(D_i+D_j)$,

$\alpha_{ii} = 1/2$,

$\alpha_{i\neq j} = 1$,

ρ = bubble radius

D = bubble diffusion coefficient

t = time

is the generally used form of Chandrasekhar's result for the coalescence rate factor of bubbles with corresponding radii of r_i and r_j. F_{kji} is the fraction of bubbles appearing in group i due to coalescence between groups k and j. The coalescence terms are respectively j for $i \geq j$, the loss of bubbles from group i due to coalescence with group j for $i \leq j$, and the appearance of group i bubbles from the coalescence of smaller gas atom number bubbles.

A finite difference technique was used for the spatially dependent portion of the solution since the accuracy of applying a steady state gas release model to transient conditions has been questioned. Also, future model development will consider grain growth, as treated by Baldewicz [8], which requires a spatially dependent solution for the release of fission gas bubbles as obtained in the present analysis.

Temperature excursions and bubble coalescence result in a mismatch between the gas pressure and the surrounding matrix forces. Vacancy and interstitial motion allow the bubbles to volume adjust to an equilibrium condition. However, during transient conditions, highly overpressured bubble conditions can exist and the fuel swelling is directly affected. Presently, the volume adjustment is calculated by assuming the interstitial flux to be negligible and assuming the exponential factors to be small. Under these assumptions, the time rate of change of bubble radius r is given by

$$\frac{dr}{dt} = \frac{\Omega}{rkT} (\Delta P) D_v C_{ev} \qquad (4)$$

where γ is the surface tension and ΔP,the pressure mismatch, is given by

$$\left(\Delta P + \frac{2\gamma}{r} + \sigma\right)\left(\frac{4\pi r^3}{3} - nb\right) = nkT \qquad (5)$$

During a small time step, $t_1 - t_0$, the temperature, hydrostatic compression and the number of gas atoms are assumed to be constant. The resulting ΔP is then given by

$$\Delta P(t_1) = \Delta P(t_0) \exp - \int_{t_0}^{t_1} \left| \frac{4\pi nkTr^2}{(\frac{4}{3}\pi r^3 - nb)^2} - \frac{2\gamma}{r^2} \right| \frac{\Omega}{rkT} D_v C_{ev} dt \qquad (6)$$

Coalescing bubbles are relaxed in their initial state for half the time step and their final state for the second half. An option exists to allow instantaneous volume adjustment (equilibrium treatment) for comparative purposes.

3. RESULTS AND DISCUSSION

The resulting analysis is computer coded and used to predict intragranular fission gas behavior during a slow thermal transient, a linear temperature increase from 1160°K to 3000°K over a 25-minute period. The fuel was characterized by grains 7 μ in diameter (unless specified otherwise), a fission gas density of 1.4×10^{20} per cm^3, a surface tension of 1000 $ergs/cm^2$, and Gruber's surface diffusion model [9] as a fit to Gulden's data.

Figures 1 and 2 show the calculated intragranular fission gas swelling and release to the grain boundaries. The gas release, by random motion, is not large even for the 4 μ grain. The bubbles exist in an environment favoring volume increase and hence reduced mobility (mobility via surface diffusion). No driving forces, such as resolution, are occurring to reduce bubble size. Hence, coalescence is the controlling factor. Figure 3 shows the evolving bubble size distribution. The radial distribution peaks around 320 Å, but bubbles with radii around 600 Å are common. Some general observations for the present intragranular fission gas model computations are:

1. The bubble behavior is dominated by coalescence. The bubble population presents numerous traps for migrating bubbles which increase in size upon coalescence and hence their mobility is reduced. This results in little of the intragranular fission gas reaching the grain boundary by random diffusion.

2. The intragranular fission gas accounted for a 14% increase in fuel volume and a transient gas release of 0.8%.

3. Allowing for nonequilibrium bubble conditions resulted in transient swelling 15% below the results for the corresponding equilibrium treatment.

4. The initial fission gas density and the matrix surface tension were determined to be highly critical parameters. Halving the surface tension to 500 ergs/cm^2 or doubling the gas atom density to 2.8x10^{20} per cm^3 resulted in a transient volume increase of 31%; more than double the previous value.

5. Since the fission gas release to the grain boundary was small, larger grains (7.1 μ compared with 4 μ) resulted in a comparable net transient swelling. Coalescence dominates the smaller grains with a 4 μ grain having a transient swelling of 12% and a gas release of 2.2%.

6. It was assumed that a reduction in the initial gas atom density might allow more gas atoms to reach the grain boundary because of the reduced probability for coalescence. While the fractional release did increase noticeably with the decrease in initial density, the actual number of gas atoms reaching the grain boundary was reduced. Hence, the higher initial gas atom densities are expected to result in a larger intergranular swelling. An intergranular fission gas model is under development and will be coupled, upon completion, to the present intragranular model.

7. If the bubble diffusion coefficient is arbitrarily increased by a factor of 1000, intragranular swelling increases from 14% to 41% and gas release increases from 0.8% to 4.2%.

Experiments for slow thermal transients (∿1°C/sec) do not exist. Thermal transient experiments have been performed at Argonne (DEH tests) and at HEDL (the FGR series). The DEH experiments tend to have large thermal gradients if the centerline temperature approaches the melting temperature; hence, biased bubble motion is important and the present model is not applicable. The work at HEDL does however, offer thermal histories that have desirable features for comparative studies.

The FGR-34 study was performed on fuel section PNL-2-4-D which had a burn-up of 3.9 atom % heavy metal. This fuel was heated radially inward to a nearly uniform 2400°C in about 18 seconds and remained essentially isothermal at 2400°C for 12 seconds longer. While very obvious differences exist between the present analysis and this experiment (for example two orders of magnitude difference in transient time and the early phases of the experimental heating resulted in moderate thermal gradients), the isothermal feature is interesting. The present analysis was used to predict the intragranular fission gas behavior during this FGR-34 isothermal transient. The calculations predict highly overpressured bubble conditions to exist on this time scale. This occurrence corresponds with the reported observations. However, the calculated bubble size distribution was much smaller than the experimental values, Fig. 4.

An important observation reported by HEDL was an intragranular zone within a few microns of the grain boundaries denude of observable bubbles [10]. These authors claimed that this occurrence could not be correlated with grain growth or biased bubble motion from the thermal gradient. Rather, they postulated that the nonequilibrium bubbles were attracted to the boundary free surface. A similar attractive force exists between overpressured bubbles which could lead to enhanced coalescence [11]. The discrepancy between the calculations and observations could be due to this nonequilibrium effect or due to errors in extrapolating Gulden's diffusion data to higher temperatures. Work is continuing on including a grain growth model, and intergranular model, a spatial solution for the vacancy concentration and examining the nonequilibrium forces.

It is suggested that serious consideration be given to appropriate "isothermal" transient experiments on well-characterized pre-irradiated fuel in order to provide a better empirical foundation for further theoretical development on intra- and intergranular fission gas behavior in the regime.

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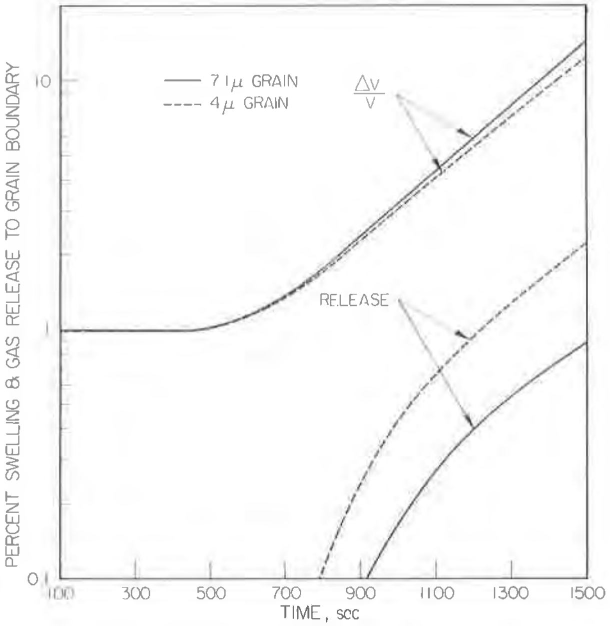

Figure 1. Intragranular swelling and gas release to the grain boundary in an isothermal transient. Nonequilibrium treatment.

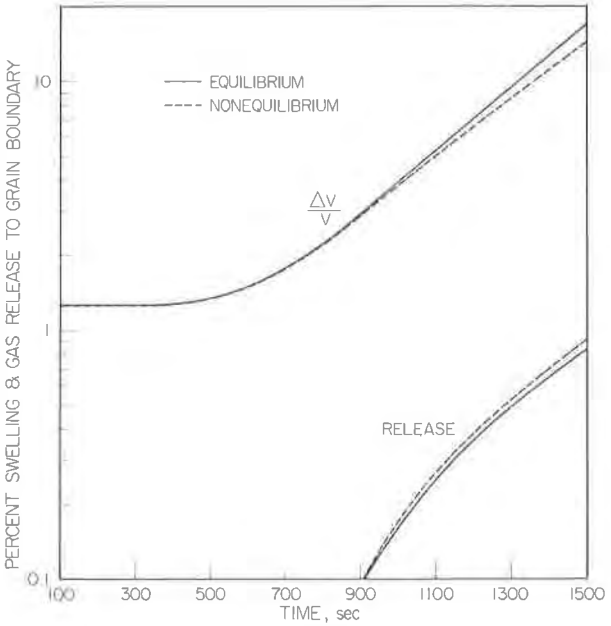

Figure 2. Intragranular swelling and gas release to the grain boundary in an isothermal transient. Equilibrium and nonequilibrium treatments for 7.1 μ grain.

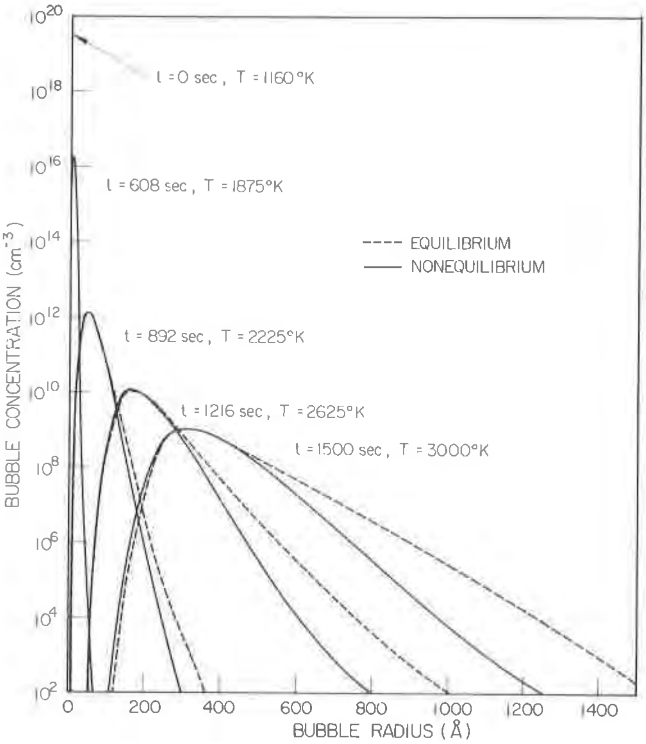

Figure 3. Bubble size distribution during an isothermal transient.

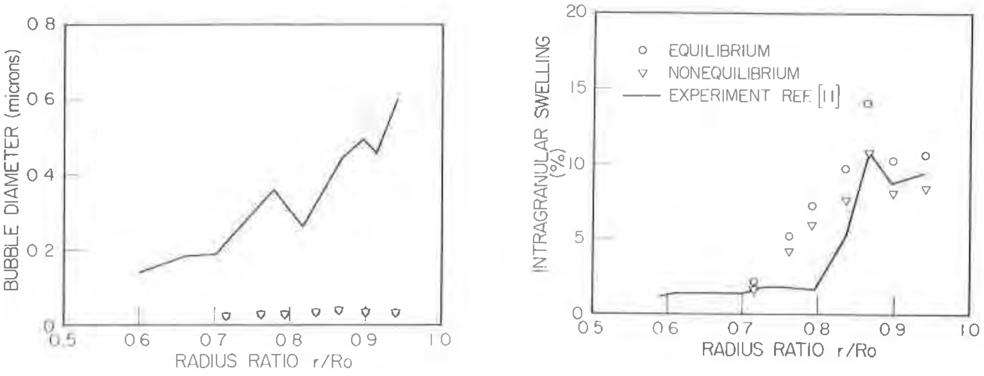

Figure 4. Bubble diameter and intragranular swelling as a function of location in the pellet. R_0 is the pellet radius.