ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60439

Transient Swelling Studies
with the GRASS Code

by

J. W. Dias* and R. B. Poeppel

Materials Science Division

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*Present address: Dept. of Energy and Kinetics,
School of Engineering and Applied Science,
University of California, Los Angeles

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ABSTRACT

The gas-release and -swelling subroutine computer code was used to calculate fission-gas swelling of an oxide fuel during a hypothetical reactor power transient. The effects of bubble-diffusion coefficient, temperature, hydrostatic pressure, fission-induced gas-atom re-solution, temperature gradient, grain boundaries and dislocations, duration of the transient, pretransient bubble size, and pretransient burnup on the resultant swelling were investigated. The results indicate that the diffusion coefficient, temperature, pressure, duration, and burnup have significant effects, whereas the other parameters do not. The investigation must be considered preliminary in that the calculations within the gas-release and -swelling subroutine are made at constant temperature. The transient temperature was approximated by taking small, constant temperature-time steps. The effect of step size on the results was not investigated.

I. INTRODUCTION

An attempt has been made to verify the applicability of the Gas Release and Swelling Subroutine (GRASS) code¹ to calculate swelling of oxide fuel under power-transient conditions. The power and temperature history obtained from Bohl and Smith² for earlier transient studies with GRASS was used.³ The previous and present results were compared and their differences analyzed. Furthermore, some of the most pertinent parameters were analyzed to investigate their effects on swelling. It is hoped that the findings of these studies will be of value for a better understanding of bubble behavior during reactor transients.

A. Gas Swelling

The observation of helium-filled bubbles moving through solid copper foils by Barnes and Mazey⁴ in 1963 has led to the knowledge that bubbles can collide and coalesce into one bubble with the net result that the total bubble volume increases. This net increase, often referred to as

"gas swelling," has been the subject of many studies, both theoretical and experimental, aimed at improving fuel design.

More recently, gas swelling has also become of interest in reactor-safety analysis due to the possibility that it may be a major mechanism of fuel failure during a reactor transient. Reports from HEDL⁵ on TREAT transient tests with mixed-oxide fuel pins preirradiated in EBR-II have indicated an increase of as much as 3.6% in fuel volume due to the formation of bubbles.

B. The GRASS Code

A model⁶ to calculate in detail the in-reactor behavior of fission gas in a mixed-oxide fuel has been incorporated in GRASS, which has been written for inclusion in the LIFE⁷ code. LIFE supplies the following operating conditions to GRASS: temperature, temperature gradient, fission rate, and hydrostatic stress state. Grain size and dislocation density may also be specified by LIFE, but at present they are determined within the GRASS code. GRASS may operate independently of LIFE, if the operating conditions are given, and may also be used in parametric studies.

The basic model assumes gas atoms nucleate to form bubbles that move both by random motion and under the influence of a temperature gradient. The bubbles may coalesce to form larger bubbles. The gas pressure in the bubbles is balanced by capillarity and hydrostatic stress in the fuel material. The gas in the larger bubbles is less dense than in the smaller bubbles. The bubbles are attracted to and pinned by structural defects, especially dislocations and grain boundaries. Bubbles can move along the defects, where they continue to grow by coalescence, until they are pulled off by the temperature-gradient force and then migrate to the central void. Included in the calculation of growth is a negative term that describes the rate of radiation-induced re-solution.

GRASS computes the evolution of the bubble-size distribution as a function of position in the fuel for three types of bubbles: (1) bubbles not pinned to structural defects, (2) bubbles pinned to dislocations, and (3) bubbles pinned to grain boundaries. The criteria derived by Weeks *et al.*⁸ are used to determine when a bubble is sufficiently large to be pulled free from the defect. Bubble mobility and velocity are computed on the basis of three separate mechanisms for bubble motion: (1) surface diffusion, (2) volume diffusion, and (3) evaporation condensation. In each fuel region and for each bubble size, the mechanism that gives the largest mobility and/or the highest velocity is used. Bubble-coalescence probability is computed following Gruber,⁹ but in each case the mechanism that gave the greatest probability was used. Where two or more mechanisms dominate, the calculation of the coalescence probability is based on the sum of the probabilities calculated separately from the various mechanisms.

The procedure of calculating the bubble-size distribution consists of dividing the bubbles into equal size ranges on a logarithmic scale and averaging properties over the ranges.⁶ The entire distribution can then be described by a relatively small number of equations. The bubble sizes are defined according to the number of gas atoms. The gas is assumed to obey the extrapolated equation of state of Harrison.¹⁰ This is essentially a van der Waals equation, in which the vander Waals constant is temperature- and pressure-dependent. When a hydrostatic stress is present, the bubble radius is a root of a quartic equation.

Based on these models for the behavior of individual gas bubbles, differential equations can be written that describe the rate of change of the number of bubbles in every size range. The equations are written in the form

$$\dot{f}_i = -a_i f_i^2 - b_i f_i + c_i, \quad (1)$$

where

f_i = number density of bubbles in the i th size range;

$\dot{f}_i$ = time derivative of f_i ;

a_i = rate at which bubbles are lost from the i th range through coalescence with bubbles of the same size;

b_i = rate at which bubbles are lost from the i th range through coalescence with bubbles of different sizes and by re-solution, migration, and diffusion;

c_i = rate at which bubbles are formed in the i th range;

and

$$f_i, a_i, b_i, c_i \geq 0.$$

The index i takes values from 1 to $i_{\max}$. The parameters b_i and c_i are functions of f_j , $j \neq i$; i.e.,

$$b_i = b_i(f_1, \dots, f_{i-1}, f_{i+1}, \dots, f_{i_{\max}})$$

and

$$c_i = c_i(f_1, \dots, f_{i-1}, f_{i+1}, \dots, f_{i_{\max}}).$$

The evolution of the bubble-size distribution is found as the solution to Eq. 1. In general, the solution must be found numerically, but in GRASS a more efficient approximate analytical solution is used in the numerical analysis.

Fuel swelling is calculated from the distribution in a straightforward manner using

$$\Delta V = \sum_{\substack{\text{bubble} \\ \text{types}}} \sum_{i=1}^{\text{imax}} f_i \times \frac{4}{3} \pi r_i^3, \quad (2)$$

where the double summation is over all bubble types and sizes, ΔV is the total bubble volume, and r_i is the average radius of bubbles in the i th size range. The swelling is calculated separately for each of the three fuel zones: columnar, equiaxed, and unrestructured.

Bubbles migrate through the fuel, and the rate of leaving any zone is included in the term b_i in Eq. 1. The rate at which gas leaves the columnar zone is of particular interest. This gas-release rate is given by

$$\text{GR} = A \times \sum_{\substack{\text{bubble} \\ \text{type}}} \sum_{i=1}^{\text{imax}} f_i \times v_i \times S_i, \quad (3)$$

where the bubbles pinned to dislocations are not included in the sum, GR is the rate at which gas atoms are released, A is the area of the inner boundary of the columnar-grain region, v_i is the average velocity of bubbles in the i th size range, and S_i is the average number of atoms per bubble for bubbles in the i th range.

C. FFTF Data

Values for temperature, linear power, and hydrostatic pressure as a function of time for the midplane portion of an FFTF fuel pin during a hypothetical reactor power ramp that results from an ~ 5 \$/sec reactivity insertion were obtained from calculations with SAS2A.² The transient was assumed to have started after the fuel had undergone a burnup of 6 at. %, operating at a steady-state linear power rating of 6.36 kW/ft. Plots that show the histories mentioned above for the columnar region--our studies were conducted for this region only--are presented in Figs. 1 and 2. In Fig. 1, R_1 and R_2 are the inner and outer radii of the columnar zone.

It is worth mentioning that the hydrostatic-stress evolution in Fig. 2 was obtained from a calculation with SAS2A in which the effect of fission-gas swelling was not included. For the sake of accuracy, GRASS³ and SAS2A should be coupled because, in reality, the swelling interacts with the fuel stress. Furthermore, the stress calculations were performed with a version of SAS2A that did not include fuel plasticity or fuel cracking. The stresses shown in Fig. 2 are believed, therefore, to be an overestimate of the actual stresses that would occur in the fuel during the power transient.

Fig. 1. Linear Power and Temperatures in a Prototypical FFTF Fuel Element during a Hypothetical Power Ramp. ANL Neg. No. 306-1971 Rev. 1.

Fig. 2. Hydrostatic Pressure, as Calculated by SAS2A, Developed in a Prototypical FFTF Fuel Element during Hypothetical Power Ramp Described in Fig. 1. ANL Neg. No. 306-1977.

Data on the amount of fission gas and bubble distribution in the columnar region at the onset of the accident for the present studies were obtained from a recent run with GRASS at steady-state conditions up to a 6 at. % burnup. The results are plotted in Fig. 3. The amount of fission

Fig. 3

Pretransient Fission-gas Bubble-size Distribution after 6 at. % Burnup, as Calculated with GRASS. ANL Neg. No. 306-1980 Rev. 2.

gas in the fuel region for that burnup is 9.67×10^{-5} mole. Most of the gas remains in solution within the fuel, but a small peak occurs in the distribution at a bubble size of $17 \text{ \AA}$ (radius).

Fig. 4. Fission-gas Swelling, Calculated with GRASS, during Transient Described in Figs. 1 and 2. ANL Neg. No. 306-1973 Rev. 1.

D. Swelling Results

Since GRASS is not coupled to any transient code (e.g., SAS2A) at present, a short program to describe the temperature, power, and stress histories was written. The entire transient history was divided into small time steps, over each of which GRASS was called and operated in a quasi-static manner.

Figure 4 shows the time-swelling history for the hypothetical FFTF power transient described in Sec. I.C. Swelling, as defined in the present report, is the ratio of the volume of fission gases contained in the fuel region to the total volume of the fuel region. At the onset of the excursion, the swelling is 3.363%, which is how much fuel has swelled because of fission-gas bubble growth at a steady-state burnup of 6 at. %.

From then until 0.35 sec has passed, gas swelling actually decreases because of the increase in hydrostatic stresses within the fuel that act on the bubbles. At this point, however, the rate of coalescence of bubbles begins to increase considerably because of the rapid increase in temperature, and, as a result, a dramatic increase in bubble volume occurs. At 0.475 sec, the trend reverses; slower rates of coalescence, which are inversely proportional to bubble size, in addition to tremendously high pressures (on the order of 4000 atm), reduce the bubble-gas volume again. When the first portion of fuel starts melting (at 0.53725 sec), gas swelling is only 3.221%, which is even lower than the value at the beginning of the accident. If high stresses did not occur within the fuel, swelling would be rather high, as indicated in Fig. 5, for a calculation at a constant hydrostatic pressure of 10 atm throughout the transient. Transient swelling (the difference between swelling at the beginning and end of the transient) reached a maximum value of 3.17%.

Figure 6 shows the evolution of the bubble-size distribution during the transient. Until after ~ 0.4 sec, the distribution does not vary; afterward, it shifts to the right, which indicates precipitation of bubbles. Throughout the transient, the bubble-number density peaks at a bubble size of 256 atoms/bubble. Note in the table in Fig. 6 the decrease in the radius of a 256-atom bubble with time, which, as has been explained, is due to the effect of the increasing hydrostatic pressure on the bubbles.

Fig. 5. Fission-gas Swelling, Calculated with GRASS, during Transient Described in Fig. 1 with Hydrostatic Stress Held Constant at 10 atm. ANL Neg. No. 306-1972.

Fig. 6. Evolution of Fission-gas Bubble-size Distribution during Transient. ANL Neg. No. 306-1984.

II. SWELLING-PARAMETER STUDIES

The present studies and those of Makenas and Poeppel³ for the same transient conditions are compared here. Their results for unrestrained and restrained swellings are plotted in Figs. 7 and 8, respectively. Much larger values of swelling were obtained, compared with the present calculations. Their unrestrained fuel swelling, for instance, is nine times as large. In light of these differences, parametric investigations of various quantities that influence bubble precipitation were undertaken and are described below. Some recent improvements had been made in the code, and it was thought these might be a cause of the difference. It appears now, however, that the major contribution to the higher swelling rates was the bubble-diffusion coefficient. All the parametric studies are described below.

A. Diffusion Coefficient of Bubbles

The bubble-diffusion coefficient D used in GRASS is derived from several sets of diffusion data and theoretical considerations. The value of D used at 1500°C, shown in Fig. 9 as a function of the bubble radius, has several attributes:

Fig. 7

Unrestrained Fission-gas Swelling,
Calculated by Makenas and Poeppel,
during Transient Described in Fig. 1.
ANL Neg. No. 306-1979.

Fig. 8

Restrained Fission-gas Swelling, Calcu-
lated by Makenas and Poeppel, during
Transient Described in Figs. 1 and 2.
ANL Neg. No. 306-1978.

Fig. 9

The Bubble-diffusion Coefficient at
1500°C as a Function of Bubble Radius.
ANL Neg. No. 306-1976 Rev. 1.

1. In the limit of decreasing bubble radii, the value of D is equal to the value for the diffusion coefficient of atomic species of xenon in UO_2 as quoted by Cornell¹¹ and Nelson.¹²

2. For gas bubbles with radii $< 0.1 \mu\text{m}$, the value of D is equal to the value of the bubble-diffusion coefficient reported by Gulden¹³ for small bubbles undergoing random motion.

3. For bubbles with radii $< 0.5 \mu\text{m}$, the mathematical expression for D is

$$D(r) = 2.1 \times 10^{-4} \exp\left(-\frac{91,000}{RT}\right) \left(\frac{r}{r_1}\right)^{-1.6234}, \quad (4)$$

where R is the gas constant, T is the absolute temperature, r is the bubble radius, and r_1 is the radius associated with a bubble that contains a single gas atom. For $r = r_1$ at 1500°C , $D = 1.3 \times 10^{-15} \text{ cm}^2/\text{sec}$, the value obtained by Cornell for a fission-gas atom.

4. For gas bubbles with radii between 0.5 and $10 \mu\text{m}$, the value of D is based on a calculation assuming the bubbles diffuse by the surface-diffusion mechanism¹⁴

$$D = \frac{3}{2\pi} \cdot \frac{D_s \rho \Omega}{r^4}, \quad (5)$$

where D_s is the surface-diffusion coefficient, ρ is the thickness of the high-diffusivity layer, Ω is the molecular volume of fuel, and r is the radius of the bubble.

5. For gas bubbles with radii greater than $10 \mu\text{m}$, the value of D is based on a calculation assuming the bubbles diffuse by the vapor-phase diffusion mechanism¹⁵

$$D = \frac{9 P_v \Omega^2}{32 \pi \sigma^2 P r^3} \left[\frac{(M_1 + M_2) R T}{2 \pi M_1 M_2} \right]^{1/2}, \quad (6)$$

where P_v is the partial pressure of UO_2 , $\pi \sigma^2$ is the cross-sectional area for collisions between fuel vapor and rare-gas molecules, P is the gas pressure in the bubble, M_1 is the molecular mass of the fission gas, and M_2 is the molecular mass of UO_2 .

The value of D used for temperatures other than 1500°C was obtained by extrapolating the values shown in Fig. 9. Cornell¹¹ reported an activation energy of $91,000 \text{ cal/mole}$ for the diffusion of atomic xenon; therefore this value was used in an Arrhenius expression for the diffusion

coefficient of a bubble that contains a single gas atom. The dependence of D on bubble radius was assumed to remain in the form given by Eq. 4; i.e.,

$$D(r) = 2.1 \times 10^{-4} \exp\left(-\frac{91,000}{RT}\right)\left(\frac{r}{r_1}\right)^x, \quad (7)$$

Fig. 10

Swelling during Transient, Calculated with GRASS, Using Various Values of Q , the Activation Energy for Bubble Diffusion. ANL Neg. No. 306-1969 Rev. 2.

where the exponent x is temperature-dependent. With T equal to 1773°K (corresponds to 1500°C), x is -1.6234 , and Eq. 7 is identical to Eq. 4. The value of x for other temperatures is obtained by assuming that the diffusion coefficient of a $50\text{-}\text{\AA}$ bubble D_{50} obeys an Arrhenius relationship with an activation energy Q ,

$$D_{50} = D'_{50} \exp\left[-\frac{Q}{R}\left(\frac{1}{T} - \frac{1}{1773}\right)\right], \quad (8)$$

where D'_{50} is the value of D_{50} at 1773°K . Equations 7 and 8 combine to yield

$$x = \frac{\ln D_{50} - \ln (2.1 \times 10^{-4}) + \frac{91,000}{RT}}{\ln\left(\frac{50}{r_1}\right)}. \quad (9)$$

Swelling calculations were made with GRASS for $Q = 45.5, 91, 113.75$, and 136.5 kcal/mole; the results are shown in Fig. 10. These calculations were made for the transient conditions described in

Figs. 1 and 2 and for the initial bubble distribution shown in Fig. 3. The results in Fig. 10 for $Q = 136.5$ kcal/mole are approaching the results of Makenas and Poeppel shown in Fig. 8, in which the value of Q used to extrapolate bubble-diffusion coefficients was 144 kcal/mole. The difference in the swelling in the two cases at the initiation of the transient is the result of different initial bubble-size distributions. Makenas and Poeppel assumed all gas was in solution before the transient.

B. Thermodynamic Effects versus Bubble Precipitation

Bubble-gas behavior under thermodynamic effects (dictated by the equation of state⁹) compared with bubble growth by coalescence during a fuel power excursion is discussed. These two features were separated by calling GRASS for a small time step, thus not allowing sufficient time for

coalescence, at the beginning of every time step. The results are shown in Fig. 11. The vertical jump at the beginning of every time step in the

Fig. 11

Stepwise Swelling during Transient that Exhibits Separate Effects of Bubble Coalescence and Thermodynamic Behavior of Fission Gas. ANL Neg. No. 306-1975 Rev. 1.

(bubble distribution is peaked at 64 atoms/bubble with a corresponding radius of $10 \text{ \AA}$) becomes more significant.

C. Re-resolution

The lower swelling rates obtained with the current version of GRASS were initially thought to be the result of the inclusion of re-resolution. In GRASS, the rate $-dN_b/dt$ at which a bubble containing N_b gas atoms loses atoms by re-resolution is assumed to be proportional to fission rate $\dot{\phi}$ and the re-resolution constant b' (per unit fission rate), as given by ¹⁶

$$-\frac{dN_b}{dt} = b' \dot{\phi} N_b. \quad (10)$$

Based on the work by Turnbull and Cornell,¹⁷ the value of b' is expected to be between 10^{-18} and 10^{-17} . The product $b'\dot{\phi}$ is also known as re-resolution parameter b , which specifies the probability per second of a particular gas atom within a bubble undergoing re-resolution.

Runs were performed with GRASS to analyze the effects of re-resolution on transient swelling. For no re-resolution, b' was set equal to 10^{-22} . The

curve indicates either an increase or a decrease in gas volume due to thermodynamics effects. Throughout the entire transient, except for one step at ~ 0.37 sec, these effects are negative because of the rapidly rising stresses within the fuel. One should also note the slopes of the straight lines that connect the time nodes. They indicate the rates at which bubble coalescence takes place. At 0.475 sec, the rates begin to decrease as a result of the increase in bubble size.

For a bubble in equilibrium with its surroundings, the bubble-gas pressure is equal to the sum of the local stress (hydrostatic pressure) and the surface tension pressure. One can then see that high stresses within the fuel, which might occur in severe power-ramp accidents, can cause excessive compression of bubbles. These features are illustrated clearly in Fig. 11. At about 0.4 sec, for example, the effect of the stress ($\sim 2500 \text{ atm}$) on the bubbles

resultant time-swelling evolution was identical to that for the standard case (Fig. 4), indicating that the role of re-solution is not important, at least in short-time transients. These results were rather surprising, since re-solution has been shown to affect bubble-size distribution substantially during long-term fuel irradiation.^{16,17}

The absence of an effect of re-solution on transient swelling results in a transient swelling that depends only on the time-temperature, stress history of the transient and not explicitly on the pin power, the fission rate, or the neutron flux. In Secs. II.D and II.G, the transient swelling will also be shown (within reasonable limits) to be independent of thermal gradient and initial bubble size.

D. Thermal Gradient

Bubbles in GRASS are allowed to diffuse at random and to migrate in the thermal gradient. In either case, they move through the fuel by surface diffusion, volume diffusion, or vaporization-condensation of fuel material within the bubble. As a result of the driving thermal force, a bubble moves with a velocity $V = (kT)^{-1}DF$, where k is the Boltzmann constant, and F is the thermal-gradient force. This force is described by¹⁴

$$F = \frac{4}{3} \frac{\pi r^3}{\Omega} \frac{Q^*}{T} \beta \frac{dT}{dx}, \quad (11)$$

where Q^* is the heat of transport per molecule, and dT/dx is the local temperature gradient in the fuel. The term β is a numerical factor that depends on the relative conductivities of the bubble and matrix material.

Gruber⁹ has computed the rate at which coalescence follows bubble collisions that result from either random motion or biased migration. For random motion, he has used the expression suggested by Chandrasekhar¹⁸ to describe the rate of colloid coagulation,

$$J_{ik} = 4\pi D_{ik} r_{ik} f_i f_k, \quad (12)$$

where J_{ik} is the rate of coalescence between i and k bubbles; D_{ik} is the appropriate diffusion coefficient, given by $D_i + D_k$; and r_{ik} is the sum of the bubble radii $r_i + r_k$. For biased migration, the expression J_{ik} is given by

$$J_{ik} = \pi(v_i - v_k)(r_i + r_k)^2 f_i f_k, \quad (13)$$

where v_i and v_k are the velocities of i and k bubbles, respectively.

To assess the relative importance of the modes of migration in the analysis of transient swelling, a run was performed with J_{ik} for biased

migration set equal to zero. The results were identical to those in Fig. 4 for the standard case, suggesting that bubble coalescence in a short transient occurs by random motion only. Although this seems rather striking, it has been observed in some fuel irradiated in TREAT¹⁹ that the spatial distribution of the porosity remains relatively uniform after the transient. This could be an indication that biased migration is negligible during a transient.

E. Grain Boundaries and Dislocations

In GRASS, bubbles are susceptible to pinning on structural defects. The code assumes a random nature for the dislocations and grain boundaries within the fuel. When bubbles impinge on structural defects, all bubbles smaller than certain critical sizes are assumed to be pinned. As the bubbles grow above the critical sizes, they are assumed to be pulled through the bulk by the temperature gradient.

The effects of grain boundaries and dislocations were investigated by calculating transient swelling with and without those defects and by comparing the results (Fig. 4). The time-swelling evolution obtained was exactly the same as the standard, suggesting that structural defects do not affect the outcome of power-excursion accidents. These results are due to the low concentration of gas on grain boundaries at the onset of the transient (Fig. 3). In reality, however, this concentration--depending on temperature, power, and burnup conditions--is usually much higher than intragranular gas concentration. For mixed-oxide fuel, for instance, operating at 1400°C and 3.3×10^{13} fissions/sec-cm³, as much as 80% of the gas has been observed on the grain boundaries after 0.84 at. % burnup.²⁰ If this were the case,

one would probably expect that most of the transient swelling would be due to thermal expansion of the gases on the grain boundaries.

Fig. 12. Swelling during a Transient with Transient Time Increased by a Factor of 60. ANL Neg. No. 306-1983 Rev. 1.

F. Transient Time

To assess the effects of transient time on swelling, a GRASS calculation was performed with only the transient time changed. The transient time was scaled by a factor of 60 to about 30 sec. The results, plotted in Fig. 12, indicate a higher peak swelling (3.913%) compared with that for the 0.525-sec-transient-time case (3.415% in Fig. 4). Swelling was also calculated in hypothetical incidents for which the fuel temperature was assumed to vary linearly with time from 2552 to 4982°F, with the hydrostatic pressure constant at 100 atm and

an initial gas concentration of 3.234×10^{20} atoms/cc. No bubbles were assumed to have precipitated before the transient. The swelling is shown as a function of temperature in Fig. 13a for transient times of 5, 10, 15, 25, 35, and 50 sec. Note that swelling varies dramatically with transient time, especially after the local average temperature has attained $\sim 4700^\circ\text{F}$. This is due to the greater duration of time allowed for bubble coalescence. The fact that, at $\sim 4700^\circ\text{F}$, bubble precipitation increases more dramatically suggests the existence of a "temperature threshold" for bubble precipitation.

(a)

(b)

(c)

Fig. 13

Swelling during a Linear Temperature Rise as a Function of Temperature for Various Transient Times and an Initial Bubble Density. (a) $3.234 \times 10^{20}/\text{cc}$, containing one atom each; (b) $2.0575 \times 10^{19}/\text{cc}$, containing 16 atoms each; and (c) $1.2734 \times 10^{18}/\text{cc}$, containing 256 atoms each. ANL Neg. Nos. 306-1985, -1981, and -1982.

G. Initial Bubble Size

Analyses were also made to investigate the effects of initial bubble-size distribution on swelling. Again, instead of the SAS2A output, the linear

Fig. 14. Final Swelling after a Linear Temperature Rise as a Function of Transient Time for Three Pretransient Bubble-size Distributions. ANL Neg. No. 306-1974.

temperature ramps described in Sec. II.F were used. The results plotted in Fig. 13a for 3.234×10^{20} bubbles/cc that contain one atom each at the onset of the transient, as well as those in Figs. 13b and 13c for 2.0575×10^{19} bubbles/cc that contain 16 atoms each and 1.2734×10^{18} bubbles/cc that contain 256 atoms each, respectively, were used in the analysis. The results from Fig. 13 were then used to generate the curves in Fig. 14, in which the final swelling at the end of the transient is plotted as a function of the transient time. The coincidence of the three curves for the three initial bubble-size distributions suggests that, within the range of initial sizes and transient times investigated, these initial distributions have little effect on the final swelling.

H. Steady-state Fuel Burnup

Perhaps one of the most important factors in the studies of transient swelling is the amount of burnup attained by the fuel

before the transient, since it indicates the amount of fission gas and the gas-bubble distribution within the fuel. It has been shown that the role of initial gas-bubble distribution is not significant in transient swelling studies. Therefore, the effects of steady-state burnup are essentially those of the amount of fission gas still retained by the fuel after the steady-state irradiation.

Transient GRASS runs were performed with initial bubble distributions (obtained with GRASS) for 1-6 at. % steady-state burnup. The same transient mode (obtained with SAS2A for the FFTF fuel pin) was used in all runs. The results are plotted in Fig. 15. Steady-state swelling (at the onset of the transient) can be seen to be directly proportional to the amount of fuel burnup. Unfortunately, a meaningful analysis of transient swelling is hindered by the large values for hydrostatic pressure used in these calculations. Figure 15 indicates that, due to the presence of larger gas bubbles, which are more easily compressed, transient swelling may decrease with steady-state burnup. The rate of bubble precipitation increases with steady-state burnup, mainly as a result of the higher content of fission gas in the fuel.

Fig. 15

Fission-gas Swelling, Calculated with GRASS, during Transient Described in Figs. 1 and 2 for Fuel Irradiated to Various Burnups (in atomic percent). ANL Neg. No. 306-1970.

III. CONCLUSIONS

1. It has been shown that calculated swelling is sensitive to the value of the heat of activation used to extrapolate the bubble-diffusion coefficient to high temperatures.
2. Bubble-size behavior during a power transient results from the competitive interplay between temperature and fuel-stress effects upon the bubbles. Temperature effects by means of bubble mobility and thermal-gas expansion cause bubble growth, whereas fuel compressive stresses have the opposite effect on the bubbles. The studies have indicated that swelling during the transient may reach values lower than those at the onset of the transient. This may not be true, since it is believed that the hydrostatic pressure computations were overestimated. More reliable stress computations could be accomplished by coupling GRASS with SAS2A.
3. Fission-rate-induced re-resolution of gas atoms from bubbles into the fuel matrix does not seem to be an important factor in transient swelling calculations.
4. Biased migration of bubbles due to thermal gradients has no effect on transient bubble behavior. Therefore, one concludes that bubbles migrate by random motion only, and this may be the reason for the constant bubble spacing that occurs during a transient.
5. Although, in our runs, bubbles on grain boundaries and dislocations did not affect transient swelling, it is believed that in many real situations most of the fission gas is contained in large intergranular bubbles. Should this prove to be the case, it is likely that gas swelling would take place mainly by thermal expansion of those large bubbles.

6. Transient time has been shown to affect transient swelling considerably because of the longer duration of time allowed for bubble coalescence.

7. It has been observed in every calculation that a "temperature threshold" exists for bubble precipitation.

8. It has been shown that initial bubble-size distribution is not a significant factor in transient swelling.

9. The rate of bubble precipitation increases with the amount of fission gas retained in the fuel at the beginning of the transient.

One can conclude that, at least within the framework of the GRASS code, linear power rating (to which re-resolution is directly proportional), temperature gradient (under which biased migration can occur), and initial bubble-size distribution are not significant in transient-swelling studies. On the other hand, one must consider the effects of the amount of fission gas in the fuel at the onset of the transient, activation energy for bubble diffusion, local temperature, local fuel stresses, amount of gas on grain boundaries relative to that within the fuel matrix, and transient time.

It should be emphasized that the present results can only be considered reliable in a qualitative manner, since the FFTF data selected are hypothetical, and the results are not subject to experimental verification. Therefore, it is suggested that analyses be performed against real data from TREAT transient tests. It should also be pointed out that, in the present studies, no attempt was made to assess the ability of the GRASS code to handle short-time bubble-coalescence phenomena. For instance, the rate of bubble formation as given by Eq. 12 is only an approximation to the more complete expression for coagulation derived by Chandrasekhar,¹⁸

$$J_{ik} = 4\pi D_{ik} r_{ik} f_i f_k \left[1 + \frac{r_{ik}}{(\pi D_{ik} t)^{1/2}} \right], \quad (14)$$

where t is the time measured from the initiation of coagulation. In the present report, the second term inside the brackets was dropped, assuming it would be negligible for long-term bubble-coalescence rate. In short-time transients, however, it is not obvious that the second term is negligible. The inclusion of this second term in the rate equations for bubble coalescence will require a modification to the numerical analysis used in GRASS.

The fact that, when two bubbles coalesce, some time is required for the resultant bubble to reach equilibrium volume¹⁴ makes the assumption of instantaneous equilibrium of volume implied in GRASS doubtful.

The rate at which the volume adjusts depends on how rapidly vacancies flow from the fuel matrix into the bubble.²¹ Therefore, attempts should be made to assess the relaxation time for effecting the volume adjustment and to compare it with the times invoked by GRASS to solve Eq. 1 during short-term transients.

The numerical integration used in GRASS assumes constant temperature. To circumvent this deficiency, small time steps during the temperature ramp were chosen for the coalescence calculations. During each of these steps, the temperature was assumed constant and was incremented from step to step. No analysis was performed to investigate the effect of step size. A more rigorous procedure would be to include the thermal variation in the integration of the coalescence equations. This, and the coupling of GRASS with a code such as DEFORM that performs the mechanical analysis, will vastly improve the validity of the results.

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