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GEAP-12148
USAEC RESEARCH AND
DEVELOPMENT REPORT
NOVEMBER 1970

FISSION GAS RE-SOLUTION RATES:

LIMITATIONS OF FISSION GAS BUBBLE GROWTH BY THE RE-SOLUTION PROCESS

M. O. MARLOWE

U.S. ATOMIC ENERGY COMMISSION
CONTRACT AT(04-3)-189
PROJECT AGREEMENT 56

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VALLECITOS NUCLEAR CENTER, PLEASANTON, CALIFORNIA, 94566

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Prepared for the
United States Atomic Energy Commission
Contract No. AT(04–3)–189
Project Agreement 56

Printed in U.S.A. Available from the
Clearing House for Federal Scientific and Technical Information
National Bureau of Standards, U.S. Department of Commerce
Springfield, Virginia 22151

Price: $3.00 per copy; Microfiche $0.65

NUCLEONICS LABORATORY • GENERAL ELECTRIC COMPANY
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3356-NL-29
95-DAC-3/71

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Limitations of Fission Gas Bubble Growth by the Re–Solution Process

M. O. Marlowe

ABSTRACT

The experimental evidence and theoretical analyses of the process of re–solution of fission gas from bubble nuclei in irradiation environments is reviewed. Gas bubble nucleation kinetics are reviewed and it is shown that the model for gas–generation rate–controlled nucleation process is consistent with experimental evidence and the range of uncertainty in the reinjection probability per unit time. The growth of static fission gas bubbles from the nucleation stage in the presence of the re–solution process is considered. It is shown that the parameter (DF/b^2) where D is the gas diffusivity in the fuel, F is the fission rate, and b is the reinjection probability per unit time, must exceed approximately 10^9 cm^{-1} if bubble growth is to occur. That requirement makes intragranular bubble growth at temperatures below the columnar grain growth region and gas release temperature unlikely. It is shown that the present uncertainties in the reinjection probability give uncertainties in the temperature at which bubble growth can occur of the order of 600 to 800°C depending on fission rate.

I. INTRODUCTION

Irradiation of uranium dioxide generates gaseous fission products that can precipitate to form gas bubbles in the solid. These gases[1] (approximately 0.24 atoms of Xe and 0.04 atoms of Kr per atom of U–235 fissioned)* are essentially insoluble in the solid.[2-3] Thus, precipitation by formation of fission gas bubbles is expected whenever the atomic mobility of the gas[4] is sufficient to allow agglomeration of the gas atoms during their time of existence in the fuel material.**

In the high–temperature (above approximately 1700°C) regions of oxide nuclear fuels, which are characterized by columnar grain growth of the oxide, the solid has been shown[5] to be essentially free from fission product gases. This is consistent with continuous sweeping of the fission gases to the center of the fuel by migration of fabrication porosity[6] or fission gas bubbles[6-8] under the driving force of the temperature gradient in the fuel. Although some controversy exists over the kinctics of this redistribution process, the experimental evidence of very low fission gas content in this region of the fuel is undisputed.

At lower temperatures, some observers have found by using transmission electron microscopy techniques only a few observable fission gas bubbles.[5] The number density and size of the bubbles provides insufficient volume to account for more than a fraction of the generated fission gases believed to be contained in the fuel.[5-9] Indeed, replication techniques of electron microscopy[10] have failed to detect fission gas bubbles in the low temperature region of the fuel.

Other observers[11-13] using transmission electron microscopy, have reported a high density (approximately $10^{17}/cm^3$) of very small fission gas bubbles (20 to 40 Å diameter) in uranium dioxide irradiated at low temperatures (< 1150°C). It has been concluded[13] that only the best electron microscopes are capable of resolving the small bubbles. The limited astigmatism correction and additional degradation of the resolution of the microscopes due to charging of the non–conducting oxide fuel can make detection of the bubbles impossible.

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*Xe and Kr yields for a thermal neutron flux of 10^{13} neutrons/cm^2/sec and irradiation time of 3×10^7 sec (approximately 1 year).[1]

**An estimate of the minimum time for bubble formation as a function of temperature will be given later.

While it is reported[11,12] that "most" of the fission gas contained within the grains of the fuel at low temperatures is precipitated in these small bubbles, post–irradiation annealing results in readily observable (by electron microscopy) fission gas bubbles.[4,5,10-12,14-16] The growth of these bubbles during the initial stages of post–irradiation annealing is well described by a diffusional process.[4] Longer annealing times result in collisional coalescence of the bubbles as they undergo random Brownian motion.[14,17] Large (to at least 300 Å diameter) fission gas bubbles, grown by post-irradiation annealing are rapidly dissolved when reirradiated at low temperatures.[10,12,15-16] This, and the small size of the observed fission gas bubbles compared with that predicted for diffusional and collisional growth processes in the fuel below approximately 1500°C, have given credibility to a kinetic re–solution effect of the irradiation environment.[12,16,18-19]

Proposed mechanisms of the kinetic re–solution of fission gases from bubbles include a simple thermal spike model[10] in which all of the gas atoms in the volume encompassed by the thermal spike accompanying the fission event are assumed to be injected into solution. Thermal diffusion processes must then be sufficient to precipitate all of the gas atoms before the volume is again affected by a spike to allow observation of any bubbles. Another model[15] proposes that fission fragments passing close to the bubble surface blast (sputter) particles of the fuel from one bubble surface to the other side of the bubble, trapping some of the gas in the bubble beneath the deposited material. More recent detailed theoretical analyses of the re–solution mechanism[12,18-19] have been based on evaluation of the cross-section for transferring energy from the fission fragments or fast neutrons to the fission gas atoms and the minimum energy for reinjection of the fission gas atoms into the lattice. These studies have indicated that the most likely processes for reinjecting gas atoms from a bubble or a bubble nucleus a sufficient distance into the lattice to eliminate immediate thermal reprecipitation are direct collision processes with fast neutrons and fission fragments and fission fragment produced collision cascades. The most recent results,[16] however, indicate that the re–solution rate of gas from bubbles is higher than can be accounted for by those mechanisms.

The direct consequences of this kinetic re–solution effect of the irradiation environment is that during irradiation, there may be many more gas atoms in solution in the solid than thermodynamically stable. Thus, at a given temperature, neutron flux and fission rate, a "kinetic solubility limit" or supersaturation of the gases exists, which in some instances may be greatly in excess of the true thermodynamic solubility limit. This enhanced supersaturation can have profound effects on the predicted size and density of fission gas bubbles. It is the consensus[20] that the most important unknown in the area of understanding and prediction of oxide fuel behavior is the quantitative dynamics of the re-solution effect of the irradiation environment on the fission gases generated in the fuel.

II. RE–SOLUTION DYNAMICS

A. GENERAL EQUATION (IDEAL GAS APPROXIMATION)

If redistribution of the fission gases has not occurred and other sinks for fission gases such as grain boundaries and fabrication pores are neglected, all of the fission gas generated is present either in solution in the lattice or precipitated in bubbles. If the gas is ideal and the vacancy flux is sufficient that the gas pressure is balanced by the surface energy restraint on the bubble, conservation of material requires that the following relationship be obeyed.

$$\int B\dot{F}\,dt = C + \frac{2\gamma R^2}{a^3 kT} \qquad (1)$$

where

- B is the number of gas atoms produced per fission event,
- $\dot{F}$ is the fission rate per unit volume,
- t is the irradiation time,
- C is the concentration of gas atoms in the lattice,
- γ is the surface energy of the fuel,
- R is the radius of the bubble,
- 2a is the bubble nucleation spacing,
- k is Boltzmann's constant, and
- T is the absolute temperature.

If the re-solution of gas atoms from bubbles into the lattice is possible, the instantaneous rate of change of the gas concentration in the lattice, dC/dt, is described as follows.[2]

rate of change of gas concentration in lattice	=	gas generation rate per unit volume	–	rate of precipitation in bubbles per unit volume	+	rate of re–solution of gas from bubbles per unit volume	(2)

The instantaneous gas generation rate per unit volume is the product of the number of gas atoms produced per fission event and the volumetric fission rate, *i.e.*, $B\dot{F}$. Ham[21] gives the precipitation rate per unit volume for spherical bubbles as $-3RDC/a^3$ where D is the diffusion coefficient of the gas atoms in the lattice. Snyder[22] has pointed out that the distribution of gas atoms around a bubble may be significantly different if a re–solution mechanism is operative than for normal precipitation process and this may alter the above formulation.

The re–solution rate per unit volume from the gas bubbles, is given by $3N/(4\pi a^3)$ if N is the number of gas atoms being reinjected into solution per unit time from a bubble.

Substituting the above forms in the rate Equation (2) gives:

$$\frac{dC}{dt} = B\dot{F} - \frac{3RDC}{a^3} + \frac{3N}{4\pi a^3} \quad . \tag{3}$$

This rate equation and the conservation of material relation [Equation (1)] should have beneral applicability. In fact, Cornell[4] treated a special case in his post–irradiation annealing study of the growth of bubbles. In that case, during the post-irradiation annealing, there was no additional generation of fission gas ($\dot{F} = 0$), but the gas total content was that which had been generated during irradiation, *i.e.*, $\int B\dot{F}t = C_o$; the concervation of material Equation (1) thus reduces to:

$$C_o = C + \frac{2\gamma R^2}{a^3 kT} \tag{4}$$

And, since during the post–irradiation anneal when there is no further fissioning, there is also no further reinjection of gas atoms into the lattice (N = O) the rate Equation (3) takes the simplified form:

$$\frac{dC}{dt_a} = \frac{-3RDC}{a^3} \tag{5}$$

where t_a is the annealing time. The solution to Equations (4) and (5) is:

$$\ln\left[\frac{R_f + R}{R_f - R}\right] = \frac{3R_f Dt_a}{a^3} \tag{6}$$

where R_f is the final bubble radius after complete precipitation. This final radius, R_f is obtained from the material balance Equation (4), when the concentration in solution C, has gone to zero.

An important consequence of the rate Equation (3) is the steady–state solution when the gas concentration in the lattice reaches a constant value, *i.e.*, the "kinetic solubility limit", C′. After the fission rate dependent "kinetic

solubility limit" for the particular irradiation conditions is achieved any additional gas generated does not contribute to changes in concentration in the lattice but only to changes in the bubble sizes. Nelson[18] and Clough, *et al.*,[12] treated this case but failed to recognize that in the steady state condition, the concentration in the lattice is constant, *i.e.*, $C = C'$ and $dC/dt = 0$. They thus obtained an erroneous equation and set of curves for the concentration of gas in solution as a function of irradiation time.* The correct solution requires evaluation of the transient part of the rate Equation (3) for determination of the duration of the transient, the "kinetic solubility limit" and the bubble size at the end of the transient.

B. RE–SOLUTION RATES FROM BUBBLES

The unknown in the quantitative application of these relationships (Equations 1 and 3) to the description of nuclear fuel behavior during irradiation is the quantity N, the number of gas atoms being reinjected into solution in the lattice per unit time from a given bubble. For any actual mechanism of re–solution of the gas atoms, including the fission fragment and/or fast neutron collision mechanism, it will be assumed that the re–solution rate from a bubble will be proportional to the density of the gas atoms in the bubble and the rate of the reinjection process, and hence the fission rate. These elementary considerations make the bubble size (R) and the equation of state of the fission gas important in determining the re–solution rate (N) from a particular bubble. As a result, the particular assumptions made regarding the bubble size dependence of the re–solution rate and the equation of state of the gas for a particular proposed mechanism of re–solution become important in both the evaluation of the theoretical models and in the prediction of fission gas behavior.

Nelson[18] and Manley[19] offer two extremes to the assumtpions made regarding the equation of state of the gas and the geometric assumptions made as to the efficacy of a particular event in reinjection of gas atoms into solution. Nelson[18] assumed that only very small ($\leqslant 50$ Å) bubbles need be considered in the re–solution process and as a result the volume occupied by each gas atom was assumed to be the minimum volume as given by a modified Van der Waals equation independent of the bubble size and temperature. Thus, one possible component to the size dependence of the re–solution rate (N) was eliminated by his formulation. Manley[19] on the other hand, assumed that the gas was ideal and as a result of the equilibrium surface energy restraint, the gas density was assumed to be inversely proportional to the bubble radius.

Nelson's[18] assumption of maximum gas atom density in the bubbles led him to assume that gas atoms undergoing recoil with neutrons or fission fragments will only be reinjected if the collision occurs within a critical distance, d, which he estimates to be 10–15 Å from the bubble surface. Otherwise, the energy gained by the gas atoms in the collisions will be lost through elastic scattering with the gas atoms. Manley[19] on the other hand, takes the approach that all collisions imparting sufficient energies to the gas atoms for reinjection into the lattice (he assumes 1 keV compared to Nelson's estimate of 0.3 keV) can result in reinjection.

For purposes of assessment of the importance of the re–solution process to fuel performance and behavior, the simplest approach is to use the probability of a particular atom within a bubble undergoing re–solution per unit time, b.[16] Following Turnbull and Cornell,[16] if N_b is the number of gas atoms in a bubble,

$$b = \left(\frac{1}{N_b}\right)\left(\frac{dN_b}{dt}\right) \quad \text{per unit time.} \tag{7}$$

Or, in terms of the re–solution rate N, previously described

$$N = b\, N_b. \tag{8}$$

*Note that Nelson[18] also misplaced the constant 3, in the precipitation rate term in the denominator instead, resulting in an under–estimate of the precipitation rate by a factor of 9.

Estimates of the probability of an atom being reinjected per unit time, b, can be obtained from Equation (7), if the number of atoms in a bubble can be determined at two times, *i.e.*,

$$b = \frac{-\dot{F}}{(\dot{F}t)} \ln (N_{b_1}/N_{b_2}).$$

Since, when a bubble is completely dispersed, N_{b_2} approaches unity, the value of b can be obtained from the minimum dose for complete dispersal, *i.e.*,

$$b = \frac{-\dot{F}}{(\dot{F}t_r)} \ln N_{b_1} \quad (9)$$

where t_r is the minimum reirradiation time at a fission rate $\dot{F}$ to completely redissolve a bubble initially contained N_{b_1} atoms.

Limiting values of the re–solution probability b, that have been obtained are listed in Table 1, with the experimental conditions on which the values are based.

In summary, it appears that for bubbles initially in the range 50 to 100 Å radius, a minimum value of 10^{-5}/sec for b is reasonable, as suggested by Turnbull and Cornell.[16] Maximum limiting values could be as low as 10^{-4}, but are less clearly defined on the basis of current data.

III. CONSEQUENCES OF THE RE–SOLUTION PROCESS

A. GAS VOLUME AT SMALL BUBBLE SIZES

It has repeatedly been shown[23,24] that for small bubble size, the ideal gas law greatly underpredicts the gas volume. In fact, the modified Van der Waals equation of state[2] for the gas and Harrison's[24] extrapolated equation of state both give essentially constant gas density (or volume per atom) for bubbles containing up to approximately 1000 gas atoms, independent of temperature or pressure. That minimum volume as given by the Van der Waals equation is approximately 8.5 X 10^{-23} cm^3/atom and, from Harrison's extrapolated equation of state it is approximately 6 X 10^{23} cm^3/atom. Assuming an intermediate value of 7 X 10^{-23} cm^3 per atom, the above considerations imply that the volume change due to gas generation is independent of whether or not the gas is in bubbles until the bubbles contain approximately 1000 atoms, and the corresponding bubble radius is approximately 26 Å. That is, the positive contribution to the volume due to fission gas generation $\Delta Vg/V$ is a linear function of burnup, *i.e.*,

$$\frac{\Delta Vg}{V} \sim (0.21 \text{ to } 0.27\%/10^{20} \text{ fissions/cm}^3) \text{ X } \dot{F}t \quad (10)$$

until the gas bubbles contain approximately 1000 atoms.

B. BUBBLE NUCLEATION SPACING

In order to evaluate the significance of this observation, it is necessary to consider the bubble spacing (2a) as set by the bubble nucleation process. Two postulates as to the rate controlling step in the nucleation process have been put forth. Greenwood, *et al.*,[25] developed a theory based on diffusion controlled nucleation in which gas bubble nuclei are assumed to form until the nuclei density is such that a newly generated gas atom, as it undergoes a diffusional random walk, is as likely to encounter a pre–existing nuclei as it is another single gas atom and thus to form a new bubble nucleus. When that condition is achieved nucleation is complete and the bubble half-spacing, a, is given by:

$$a = 6\sqrt{\frac{3D_g r_o^2 a_o}{2GZ}}$$

TABLE 1. Summary of Low Temperature Fission Gas Re–Solution Data

	Initial Bubble Radius (10^{-8} cm)	Initial Total Gas Content (cm^{-3})	Annealing Temperature (^{o}C)	Bubble Spacing (10^{-8}cm)	Gas Content in Bubble (N_{b_1}) (atoms)	Reirradiated Fission Rate ($f/cm^3/sec$) ($\dot{F}$)	Reirradiated Dose (f/cm^3) ($\dot{F}t$)	Probability of Reinjected per Unit Time (b) (sec^{-1})	Effective Volume of Fission Event in Reinjection Gas (b) (cm^3/fission)
Turnbull & Cornell[18]	50–75	$4.5–6 \times 10^{19}$	1500	–	~ 5500	10^{13}	7×10^{17}	$< 1.2 \times 10^{-4}$[a]	$< 1.2 \times 10^{-17}$
	50–75	$4.5–6 \times 10^{19}$	1500	–	~ 5500	10^{13}	3.3×10	$> 2.6 \times 10^{-5}$	$> 2.6 \times 10^{-18}$
Ross[10]	500	–	1500	–	–	?	1.4×10^{19}	–	–
Whapham[15]	50	6.6×10^{18}	1600	518[b]	~ 3900[b]	~ 4×10^{13}	2.2×10^{19}	$> 1.5 \times 10^{-5}$	$> 3.8 \times 10^{19}$
	150	6.6×10^{18}	1600	1225[b]	~ 5080[b]	4×10^{13}	2.2×10^{19}	$> 1.55 \times 10^{-5}$	$> 3.9 \times 10^{-19}$
Clough, et al.,[12]	13–50	6×10^{18} to 1.5×10^{20}	1500	360–437	~ 70–3000	?	10^{17}	–	–
						?	10^{16}	–	–

(a) Data that bubbles have not disappeared is inconclusive, therefore higher values of b are possible.

(b) Bubble spacing and gas content calculated from total gas content, bubble size and equation of state of gas with non–ideality considered, assuming all gas precipitated.

where

D_g is the diffusion coefficient of the gas in the fuel,

r_o is the radius of the stable bubble nuclei,

a_o is the lattice parameter of the fuel,

G is gas atom production rate per atom site, and

Z is the number of new lattice sites explored by the gas atom per jump.

The corresponding gas bubble density, n, is:

$$n = \left(\frac{3}{4\pi}\right)^{2/3} \left(\frac{2Z\,B\dot{F}M_F}{3D_g a_o N_A \rho_F}\right)^{2/3} \left(\frac{\rho_g}{m^*}\right)^{1/3} \tag{12}$$

where

M_F is the molecular weight of the fuel,

ρ_F is the theoretical density of the fuel,

N_A is Avogadro's number,

ρ_g is the gas density in the bubble, and

m^* is the number of gas atoms in a stable nucleus.

This diffusion controlled nucleation model gives a temperature dependent bubble spacing through the exponentially temperature dependent gas diffusivity, Dg. Note that any fission enhancement of the gas diffusivity serves to decrease the bubble density at a given temperature. The bubble density predicted by Equation (12) for UO_2 is plotted as a function of fission rate and gas diffusivity[4] (and temperature assuming no irradiation enhancement of gas diffusion) in Figure 1.* Also shown is the range of experimentally observed bubble densities, *i.e.*, 10^{16} to 10^{17} bubbles/cm[4,12,18] for irradiation temperatures to approximately 1130°C.[18] Note that the observed bubble densities are greater than predicted, at least above approximately 800°C, and that if a fission enhancement of gas diffusion as predicted by Nelson,[18] is included, the observed bubble densities are a minimum of approximately 25 times greater than the diffusion controlled nucleation model predicts.

Nelson[18] assumed that the diffusional process was not rate controlling, but that in the radiation environment diffusion is sufficiently rapid that the nucleation rate is limited by the gas atom production rate. Under those conditions, nucleation is complete when the gas atom production rate per unit volume is equal to the re–solution rate from the nuclei, which gives:

$$a = 3\sqrt{\frac{3bm^*}{4\pi B\dot{F}}} \quad \text{or} \quad n = \frac{B\dot{F}}{bm^*} \tag{13}$$

The bubble density as given by Equation (13) is plotted in Figure 2 as a function of fission rate and the gas atom probability of reinjection per unit time, b.*

*It was also assumed that two gas atoms are sufficient to form a stable bubble nucleus, *i.e.*, $m^* = 2$.

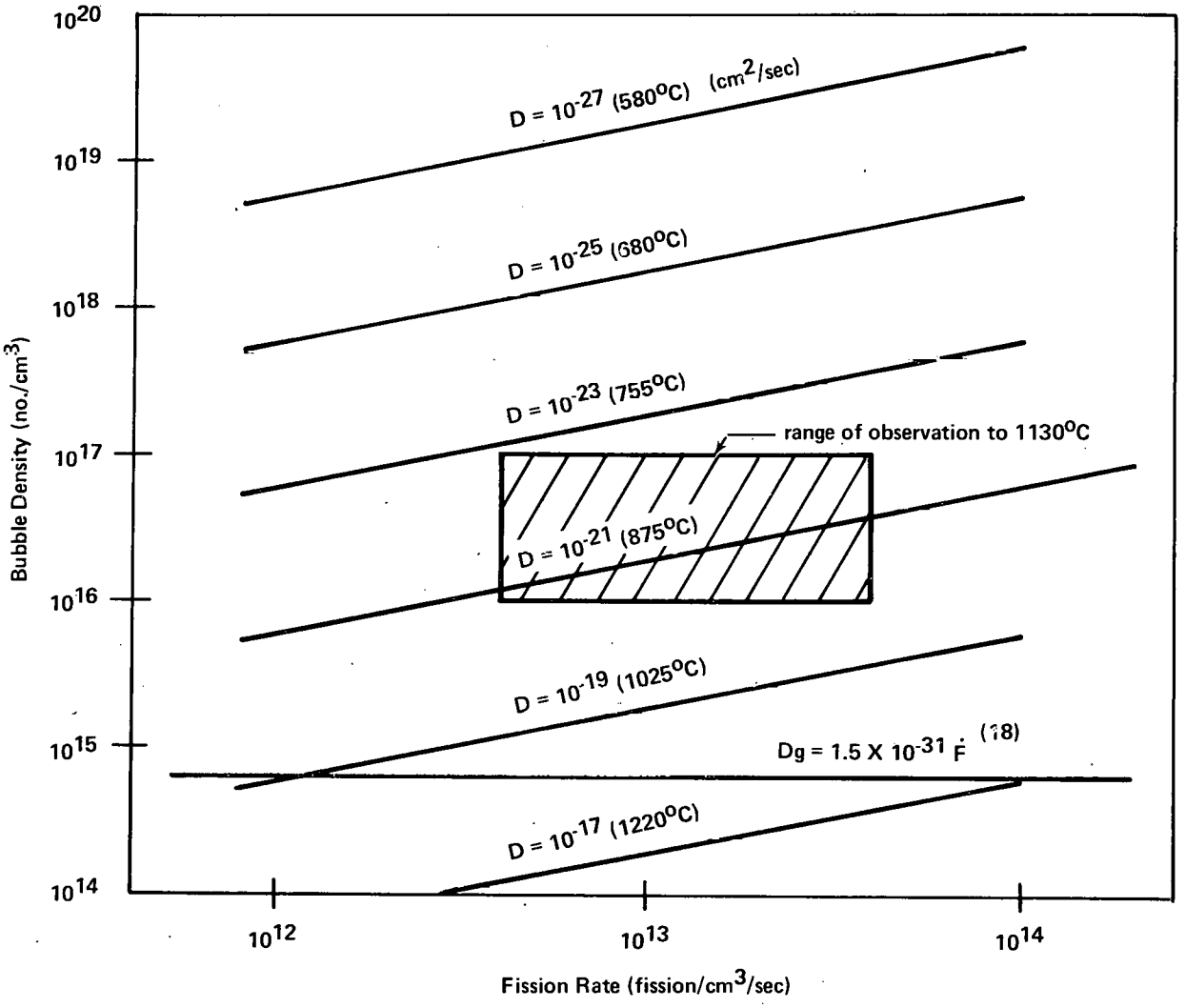

FIGURE 1. BUBBLE NUCLEUS DENSITY AS GIVEN BY A DIFFUSION–CONTROLLED NUCLEATION PROCESS AS A FUNCTION OF FISSION RATE AND GAS DIFFUSIVITY (TEMPERATURE).

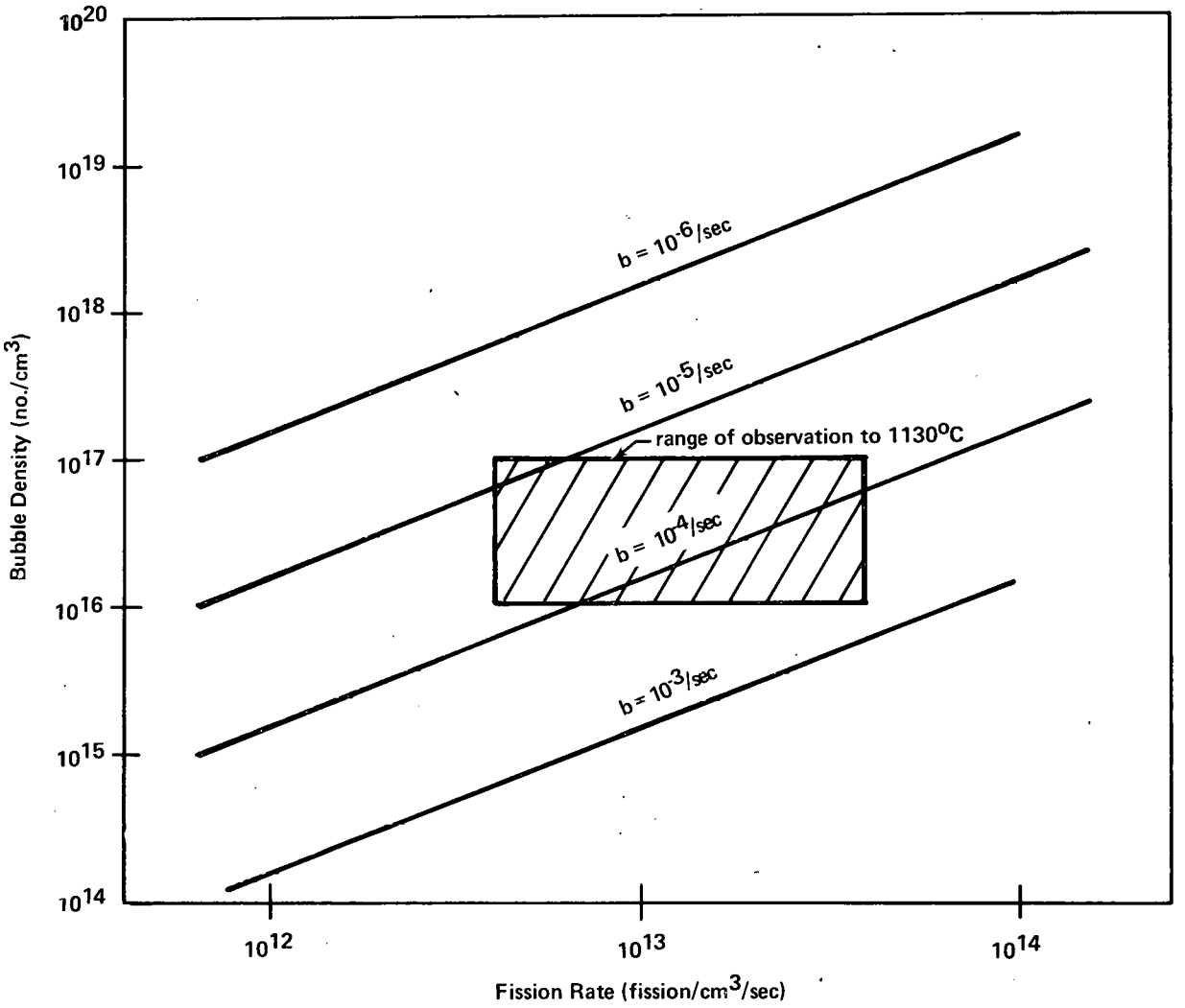

FIGURE 2. BUBBLE NUCLEUS DENSITY AS GIVEN BY A GAS GENERATION – RATE CONTROLLED NUCLEATION PROCESS AS A FUNCTION OF FISSION RATE AND GAS ATOM REINJECTION PROBABILITY PER UNIT TIME.

The observed gas bubble density as calculated by this model which assumes that gas generation is the rate controlling step in the bubble nucleation process is well described if the reinjection probability, b, is in the range 10^{-4} to 10^{-5}, as previously estimated from the bubble disappearance experiments. It is clear that an experimentally self-consistent bubble nucleation and re–solution model is obtained using Nelson's model for the nucleation spacing. The diffusion rate controlled nucleation model does not give as good agreement with experimental observations.

C. GAS BUBBLE GROWTH (LIMITING CASES)

The rate of bubble growth can be determined for the case of small bubbles (< 1000 atoms) by assuming a constant gas density as follows. The kinetic equation (Equation 3) expressed in terms of the reinjection probability b (Equation 8), is:

$$\frac{dc}{dt} = B\dot{F} - \frac{3RDC}{a^3} + \frac{3bN_B}{4\pi a^3} \tag{14}$$

The constant gas density restriction gives the following relation between the number of atoms per bubble N_B and the bubble radius, R.

$$\frac{3N_B}{4\pi R^3} = \rho_g \tag{15}$$

Substitution of that relationship into a modified mass balance equation, *i.e.,*

$$B\dot{F}t = \frac{C + 3N_B}{4\pi a^3} \tag{16}$$

followed by differentiation, gives:

$$\frac{dc}{dt} = B\dot{F} - \frac{3\rho_g R^2}{a^3}\left(\frac{dR}{dt}\right). \tag{17}$$

Combining Equations (15) and (18), then gives the desired result:

$$\frac{dR}{dt} = \frac{-bR}{3} - \frac{DR^2}{a^3} + \frac{DB\dot{F}t}{\rho_g R} \tag{18}$$

The three terms in Equation (18) contributing to the growth rate of the bubbles (dR/dt), arise from the following physical phenomena:

$\frac{DB\dot{F}t}{\rho_g R}$, the only positive term is the result of precipitation of the gas in the bubbles.

$-\frac{bR}{3}$, is the decrease in bubble growth rate as a result of the re–solution process which arises from fewer gas atoms per bubble at any given time.

$\frac{-DR^2}{a^3}$, is the decrease in bubble growth rate arising from noninstantaneous precipitation of the gas which produces a nonequilibrium gas concentration in the lattice even in the absence of the re–solution process.

Using Nelson's[18] relationship for the nucleation spacing, *i.e.*, Equation (13), gives:

$$\frac{dR}{dt} = -\left(\frac{b}{3}\right)R - \left(\frac{4\pi B}{3m^*}\frac{D\dot{F}}{b}\right)R^2 + \left(\frac{BD\dot{F}}{\rho_g}\right)\frac{t}{R}. \tag{19}$$

This relationship should only apply after the nucleation stage is complete. If gas generation is indeed the rate controlling step in the bubble nucleation process, it is clear that the time to formation of the nuclei, t_o, is simply the time to generate the amount of gas per unit volume required to form the nuclei. Mathematically that condition is expressed as follows:

$$t_o = \frac{3m^*}{4\pi a^3 B\dot{F}} = \frac{1}{b} \tag{20}$$

and the radius of the stable nucleus, R_o, is

$$R_o = \left(\frac{3m^*}{4\pi\rho_g}\right)^{1/3} \tag{21}$$

which is, approximately 3.2×10^{-8} if two gas atoms form a stable nucleus ($m^* = 2$) and $\rho_g \cong 1/(7 \times 10^{-23})$ $(cm^3)^{-1}$. Thus, the initial boundary conditions on Equation (19) are $t = t_o$, $R = R_o$.

Replacing the radius and time with dimensionless quantities: $R^* = R/R_o$ and $t^* = t/t_o$, and substituting from Equation (20) for t_o gives

$$\frac{dR^*}{dt^*} = -\frac{R^*}{3} - \left[\frac{4\pi BR_o}{3m^*}\left(\frac{D\dot{F}}{b^2}\right)\right]R^{*2} + \left[\frac{B}{\rho_g R_o^2}\left(\frac{D\dot{F}}{b^2}\right)\right]\frac{t^*}{R^*} \tag{22}$$

where the initial boundary conditions now become:

$$R^* = 1.0, t^* = 1.0.$$

If bubble growth is to occur, clearly from Equation (22), dR^*/dt^* must be positive and hence,

$$\left(\frac{B}{\rho_g R_o^2}\frac{D\dot{F}}{b^2}\right)\frac{t^*}{R^*} > \frac{R^*}{3} + \left[\frac{4\pi BR_o}{3m^*}\left(\frac{D\dot{F}}{b^2}\right)\right]R^{*2} \tag{23}$$

Evaluating this inequality at the initial condition, *i.e.*, at the cessation of nucleation when $t^* = 1$ and $R^* = 1$, gives:

$$\left(\frac{D\dot{F}}{b^2}\right) > \frac{m^*\rho_g R_o^2}{(3m^*B - 4\pi B\rho_g R_o^3)} \tag{24}$$

Assuming two gas atoms per nuclei ($m^* = 2$), $\rho_g = 1/(7 \times 10^{23})$ $(cm^3)^{-1}$ and $B = 0.3$ as before and obtaining R_o from Equation (21), gives:

$$\left(\frac{D\dot{F}}{b^2}\right)_{min} > 8.3 \times 10^8\ cm^{-1}. \tag{25}$$

At lower values of the quantity $(D\dot{F}/b^2)$ *no bubble growth* is predicted beyond the nucleation stage.

The upper limit of applicability of the bubble growth Equation (22) is reached when the gas density is no longer constant as more gas is precipitated into the bubbles, *i.e.,* when the number of gas atoms per bubble exceeds approximately 1000. The corresponding bubble radius R_{1000} is:

$$R_{1000} = \left(\frac{3 \times 1000}{4\pi\rho_g}\right)^{1/3} \tag{26}$$

Comparison of Equations (21) and (26) gives the following upper limit for the value of R^* if the number of atoms in the stable nucleus is two (*i.e.,* $m^* = 2$).

$$\frac{R_{1000}}{R_o} = R^*_{1000} = \left(\frac{3 \times 1000}{4\pi\rho_g}\right)^{1/3} \left(\frac{4\pi\rho_g}{3m^*}\right)^{1/3} \cong 8 \tag{27}$$

Unfortunately no exact solution was obtained for the bubble growth Equation (22) and exact times to achieve 1000 atoms per bubble could not be calculated.

Very tight upper and lower limits on the time to achieve 1000 atoms per bubble can be calculated as follows; however, if it is assumed that after nucleation has ceased, the bubble growth rate is always positive, *i.e.,* $dR^*/dt^* \geqslant 0$, then a minimum value for the dimensionless time t^*_{min} to achieve a particular value of $R^* = R^*_i$ is given as follows from Equation (22):

$$\frac{dR^*}{dt^*} = 0 = \frac{-R^*_i}{3} - \left[\frac{4\pi BR_o}{3m^*}\left(\frac{D\dot{F}}{b^2}\right)\right] R^{*2}_i + \left[\frac{B}{\rho_g R_o{}^2}\left(\frac{D\dot{F}}{b^2}\right)\right] \frac{t^*_{min}}{R^*_i} \tag{28}$$

which, when solved for t^*_{min} gives:

$$t^*_{min} = \frac{\frac{R^{*2}_i}{3} + \left[\frac{4\pi RR_o}{3m^*}\left(\frac{D\dot{F}}{b^2}\right)\right] R^{*3}_i}{\left[\frac{B}{\rho_g R_o{}^2}\left(\frac{D\dot{F}}{b^2}\right)\right]} \tag{29}$$

Similarly, recognizing that the *maximum* growth rate dR^*/dt^*_{max} is the initial value at $R^* = 1, t^* = 1$, then an upper limit on the "time" to achieve a given value of $R^* = R^*_i$ can also be obtained as follows:

$$\frac{dR^*}{dt^*_{max}} = \frac{-R^*_i}{3} - \left[\frac{4\pi BR_o}{3m^*}\left(\frac{D\dot{F}}{b^2}\right)\right] R^{*2}_i + \left[\frac{B}{\rho_g R_o{}^2}\left(\frac{D\dot{F}}{b^2}\right)\right] \frac{t^*_{max}}{R^*_i} \tag{30}$$

or solving for t^*_{max} and substituting from Equation (3) gives:

$$t^*_{max} = t^*_{min} + \left(\frac{R^*_i}{\left[\frac{B}{\rho_g R_o^2} \left(\frac{\dot{DF}}{b^2} \right) \right]} \right) \frac{dR^*}{dt^*_{max}} \tag{31}$$

The maximum growth rate dR^*/dt^*_{max} is obtained by substitution in Equation (22) for $R_i = 1$, $t^* = 1$, *i.e.*,

$$\frac{dR^*}{dt^*_{max}} = -\frac{1}{3} + \left[\left(\frac{B}{\rho_g R_o^2} - \frac{4\pi B}{3m^*} \right) \left(\frac{\dot{DF}}{b^2} \right) \right] \tag{32}$$

These limiting quantities are listed as a function of the parameter (DF/b^2) (cm^{-1}) in Table 2.

TABLE 2. Limiting Values of the Dimensionless Time, t^*, to Achieve 1000 Gas Atoms per Bundle

$\frac{\dot{DF}}{b^2}$ (cm^{-1})	$\frac{dR^*}{dt^*_{max}}$	t^*_{min} to achieve 1000 atoms/bubble	t^*_{max} to achieve 1000 atoms/bubble
10^8	(–)	(a)	(a)
8.3×10^8	(0)	(a)	(a)
9×10^{18}	0.0281	503.13	503.14
10^9	0.0683	503.01	503.04
5×10^9	1.6748	502.18	502.31
10^{10}	3.6830	502.08	502.22
10^{11}	39.830	501.98	502.14
10^{12}	401.3	501.97	502.14

(a) *No* growth expected for $\dot{DF}/b^2 \leq 8.3 \times 10^8\ cm^{-1}$, since initial, *maximum* growth rate is *negative.*

Note that the dimensionless "time" to achieve 1000 atoms per bubble is essentially independent of $(\dot{DF}/b^2)$ and is equal to approximately 500 at any value of $(\dot{DF}/b^2)$ large enough to allow *any* bubble growth $(dR^*/dt^*_{max} = +)$.

D. GAS ATOM REINJECTION ON PROBABILITY: DEPENDENCE ON FISSION RATE

Having determined a minimum limiting value for the parameter ($D\dot{F}/b^2$) for bubble growth to occur, and having shown that the dimensionless time $t^* = t/t_o = t_b$ to exceed approximately 1000 gas atoms per bubble is essentially constant and approximately 500 if any bubble growth can occur, it is important to re–examine the reinjection probability per unit time, b. Recall that (Section II–B) the re–solution rate or reinjection rate of gas atoms into the lattice is intuitively expected to be dependent on the fission rate (*i.e.,* the *rate* at which the reinjection process is occurring) and the gas density in the bubbles. Since for the cases considered here, *i.e.,* up to approximately 1000 gas atoms per bubble, the gas density is constant, that contribution to possible variation in b, the reinjection probability per unit time, need not be considered. Further, since the maximum radius being considered $R_{1000} \cong 26$ Å is not far removed from Nelson's[18] estimate of the maximum injection distance through the dense gas (10–15 Å), it is expected that variation in b and hence in bubble growth rates from that source would be secondary in nature. Conversely, the fission rate is expected to be the primary variable in determining the reinjection–probability per unit time, whereas in the analysis presented to this point, that dependence or fission rate has been neglected. To rectify that condition we can define a new variable, b′ by the relation:

$$b = b'\dot{F} \tag{33}$$

which requires that b′ have the units of volume per fission. Thus, we can interpret it as an effective *volume* in which reinjection occurs because of a fission event.

Evaluating the effective reinjection volume per fission event, b′, from the previously mentioned experimental results (see Table 1), gives the following limiting values for b′:

$$2.6 \times 10^{-18} < b' < 1.2 \times 10^{-17}\ \text{cm}^3/\text{fission}.$$

The total number of atoms displaced from their lattice position by the fission fragments N_d, is estimated to be in the range 10^4 to 10^5 per fission event.[26] Assume for the present that some fraction E of such atomic displacements could be equivalent to a reinjection event. If the size of the bubble nuclei is not large (a few atoms) such an assumption may be valid. The probability of a gas atom in a bubble nucleus being reinjected per unit time by such a displacement is given by the product of the number of atoms displaced per fission event, N_d, the number of fissions occurring per unit time in the volume of fuel associated with the bubble, $(4\pi a^3\dot{F}/3)$, and the probability that the volume of the fission fragment collision path encompasses the bubble nucleus, *i.e.,* (R_o^3/a^3). Therefore, the predicted reinjection probability per unit time, b, is given by the relation:

$$b = N_d \times \frac{4\pi a^3 \dot{F}}{3} \times \frac{R_o^3}{a^3} \times E = \left(\frac{4}{3}\pi N_d R_o^3 E\right)\dot{F} \tag{34}$$

Evaluating the quantities as before and assuming 100% efficiency (E = 1) in the displacement of gas atoms as reinjection processes gives:

$$1.4 \times 10^{-18} \times \dot{F} < b < 1.4 \times 10^{17} \times \dot{F}\ \text{sec}^{-1}$$

or

$$1.4 \times 10^{-18} < b' < 1.4 \times 10^{-17}\ \text{cm}^3$$

based on the uncertainty in N_d (*i.e.,* $N_d = 10^4$ to 10^5 atoms displaced per fission event). This simple model, which assumes that displacement of a lattice atom is essentially equivalent to reinjecting of a gas atom from a bubble nucleus in which the gas is so restrained as to be incompressible, is within the range of uncertainties in experimental results and uncertainties in the number of atoms displaced per fission event.

E. COMPOSITE DESCRIPTION OF GAS BEHAVIOR

From the preceding sections a composite picture of fission gas behavior emerges. This behavior can be discussed in terms of three regions of behavior, depending on the values of the dimensionless "time," t,* and the dimensionless bubble "radius," R,* which are just the ratios of the quantities to their values at the cessation of nucleation, and also depending on the parameter $(D/b'^2\dot{F})$.

1. Nucleation Stage

For $t^* < 1$, the nucleation stage is incomplete, and all of the gas can be considered to be solution. The positive volume contribution of the gas is approximately $0.27\%/10^{20}$ fissions/cm^3.

2. Initial Bubble Growth

a. If the parameter $(D/b'^2\dot{F})$ does *not* exceed approximately 10^9 cm^{-1}, there is *no* bubble growth and all of the gas can be treated as in solution and, as above, the positive volume contribution of the gas is approximately $0.27\%/10^{20}$ fissions/cm^3.

b. If the parameter $(D/b'^2\dot{F})$ is equal to or exceeds approximately 10^9 cm^{-1}, bubble growth is expected. However, the bubbles will not contain more than 1000 gas atoms until $t^* \cong 500$. Until that time, the positive volume contribution of the gas is independent of whether bubbles have formed or not and remains $\cong 0.27\%/10^{20}$ fissions/cm^3.

c. If the parameter $(D/b'^2\dot{F})$ is equal to or exceeds approximately 10^9 cm^{-1} and $t^* \geqslant 500$, bubble growth must be considered with a model using a nonconstant gas density (approaching ideal gas behavior at large bubble sizes, *e.g.,* Reference 27).

The restriction that the parameter $(D/b'^2\dot{F})$ must exceed approximately 10^9 cm^{-1} before the gaseous contribution can become nonlinearly time dependent has another important implication. That condition $(D/b'^2F = 10^9$ cm$^{-1})$ is graphically depicted in Figure 3 for the experimental range of uncertainty in the effective reinjection volume per fission, b′, and for the model based on reinjection of displaced atoms. The temperatures corresponding to the out–of–pile gas diffusivities are from Cornell[4] and Belle, *et al.*[28] Also shown is the predicted fission induced gas diffusivity[18] (which is of little importance if the prediction is correct) and the temperatures corresponding to the gas diffusivity if there is *no* enhancement of the gas diffusivity due to the radiation environment. The range of fission rates over which some experimental evidence exists for calcualtion of b or b′ is shown as a double cross–hatched area in Figure 3.

The range of b′ values based on experimental results (meager though they are) shows that there is no predicted gas bubble growth at temperatures below which grain growth, columnar grain growth and gas release are predicted to occur. The exact temperature at which bubble growth can occur at a given fission rate is uncertain within approximately 600 to 800°C because of uncertainties in gas diffusivity and the reinjection parameters b and b′. Thus, it appears that if the parameter $(D/b'^2\dot{F})$ has a sufficiently high value for bubble growth to occur, the gas is also subject to migration and release. The consequence is that very little, if any, of the fuel is expected to have a significant intragranular fission gas bubble population except in transit to the fuel center during operation.

This situation may not obtain at the grain boundaries in the fuel, since in that case $(D/b'^2\dot{F})$ may be higher than in the bulk of the fuel as a result of greater gas diffusivities on and near the grain boundaries.

F. TOTAL FUEL SWELLING

From the previous section it is apparent that the positive contribution of the fission gases to fuel swelling throughout much of the fuel (*i.e.,* if $D/d'^2\dot{F} < 10^9$ cm^{-1}) and for much of the life of the fuel (to $t^* = 500$, even if $(D/b'^2F > 10^9$ cm^{-1}), is simply 0.21 to $0.27\%/10^{20}$ fissions/cm^3.

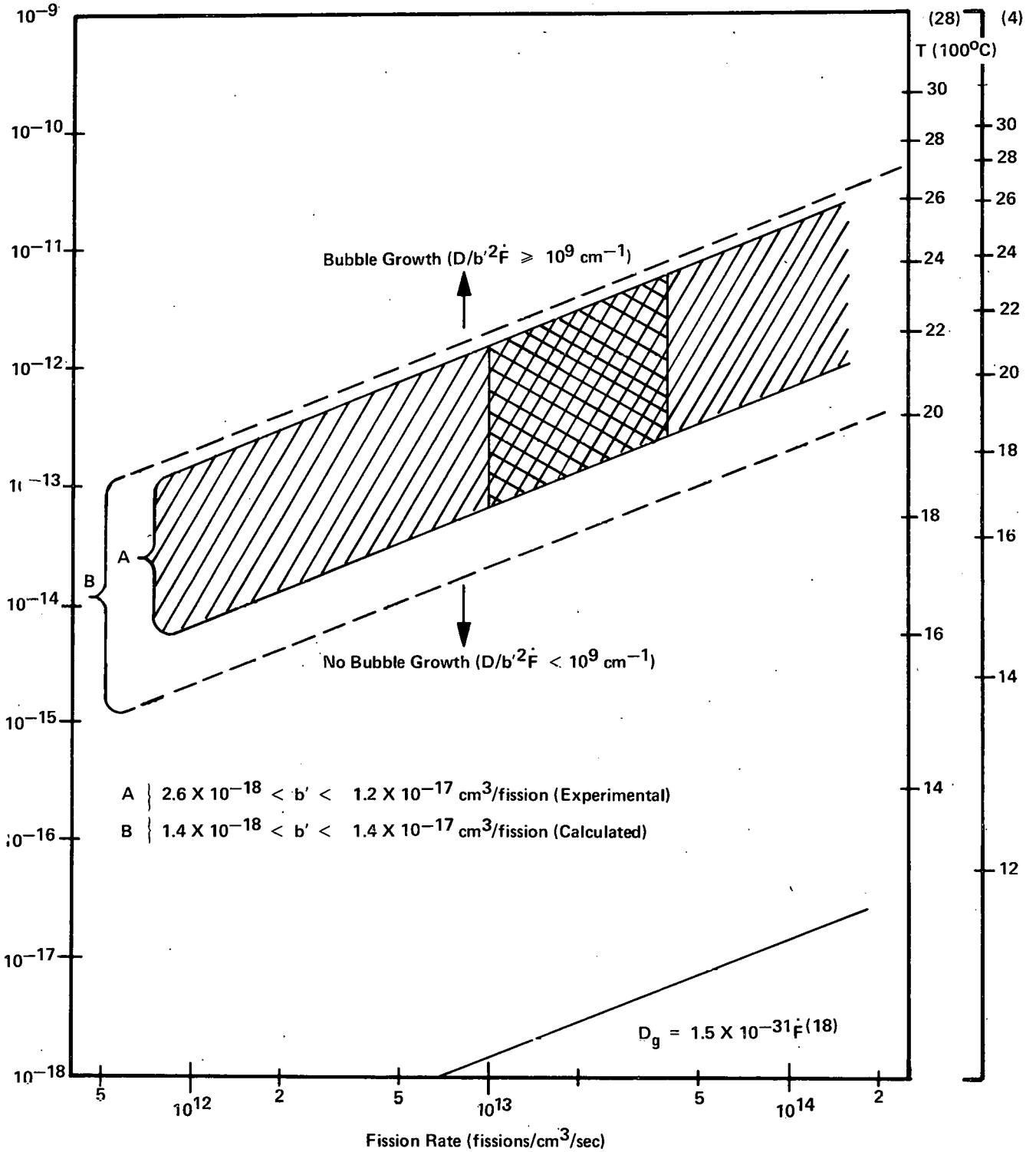

FIGURE 3. LIMITING CONDITIONS FOR BUBBLE GROWTH AT CONSTANT FISSION RATE

Anselin[29] has considered the positive volume change resulting from the generation of solid fission products and has obtained a maximum value of $0.54\%/10^{20}$ fissions/cm^3 for UO_2 fuel in a thermal reactor.

The negative volume change accompanying the fission process as a result of destruction of the matrix uranium atoms was also considered by Anselin[29] for which he obtained $-0.41\%/10^{20}$ fissions/cm^3.

Combining all three terms, *i.e.*, the negative volume change due to matrix destruction, and the positive contributions of the solid and gaseous fission products, in the time and ($D/b'^2\dot{F}$) ranges described above is:

$$-0.41\% + 0.54\% + 0.27\% = 0.40\%/10^{20} \text{ fissions/cm}^3$$

This is very close to the average figure of $0.35\%/10^{20}$ fissions/cm^3 derived from some irradiations performed under varied conditions.[28]

G. QUALIFICATION

It must be noted that the above treatment strictly speaking treats static gas bubbles and bubble nuclei. At high temperatures it is expected that collisional coalescence of migrating (random and biased) bubble nuclei may rapidly make a dominant contribution to bubble growth kinetics invalidating the treatment presented here. It should also be noted that because of the high values of gas diffusivity (and temperature) to which the gases may be essentially all in solution, the mobility may be sufficient to allow rapid bubble growth and swelling once the neutron flux is removed, or decreased, *e.g.*, on cooling during reactor shutdown, or during a controlled slow restart to power. Such behavior could lead to wide variations in experimentally observed fission gas swelling behavior, and could contribute to the observed gas release bursts on reactor shutdown and restart.

IV. SUMMARY AND CONCLUSIONS

The process of fission gas bubble nucleation and growth of static bubbles in nuclear fuels was analyzed. The effect of re–solution of the fission gases in the irradiation environment was treated. It was shown that all of the available data on the disappearance of fission gas bubbles grown large by post–irradiation annealing on re–irradiation, support a value of 10^{-4} to 10^{-5} sec^{-1} for the reinjection probability per unit time b, of a gas atom in a small gas bubble. It was concluded that the volume contribution of the gas atoms is essentially independent of whether the gases are in solution in the lattice or precipitated in bubbles up to a gas content of approximately 1000 atoms per bubble.

Using the above range of values for the reinjection probability per unit time it was shown that the available experimental evidence of gas bubble densities support a model for the nucleation process in which gas generation is the rate controlling step rather than a diffusion controlled nucleation process.

The gas bubble growth model which includes the re–solution mechanism and the gas–generation rate as the rate controlling step in the bubble nucleation process was used to show that no fission gas bubble growth is expected for values of the parameter ($D\dot{F}/b^2$) less than 10^9 cm^{-1}.

The model was also used to show that the growing bubbles do not attain a gas content of 1000 atoms until an irradiation time of approximately 500/b, regardless of the value of ($D/b'^2\dot{F}$). The model indicates that with the re–solution process there should be essentially no observable fission gas bubbles in UO_2 fuel under steady–state neutron flux below the grain growth temperature. Since above that temperature rapid gas release is a predominant characteristic, fission gas bubbles make a very small contribution to reactor fuel behavior at a steady–state fission rate. However, under conditions of elevated temperature but reduced or removed neutron flux, rapid bubble growth and swelling is expected.

The total positive gaseous fission product contribution to fuel swelling for $D/b'^2\dot{F} < 10^9$, and for $t < (500/b)$ was estimated to be 0.21 to $0.27\%/10^{20}$ fission/cm^3. With the estimated solid fission product positive volume contribution

and the negative volume change resulting from destruction of the matrix, the estimated total swelling rate in the above range of conditions is estimated to be less than 0.34 to 0.40%/10^{20} fission/cm^3. This is in very good agreement with the average observed values of 0.35%/10^{20} fission/cm^3.

It is concluded that further analysis of the gaseous swelling is required for values of $(D/b'^2\dot{F}) > 10^9$ at times greater than 500/b. Also, further experiments are necessary to narrow the range of uncertainty in the reinjection probability per unit time, b, and to determine its bubble size dependence, if any. The current uncertainty in b gives uncertainties in the temperature at which the gas is no longer in kinetic solution of the order of 600 to 800°C, depending on fission rate (see Figure 3).

The results of this model indicate that whereas total fuel swelling may be of the order of 0.35%/10^{20} fissions/cm^3 under steady–state operating conditions, power cycling and reactor shutdown and restart, *i.e.*, transient conditions of high temperature and low or reduced fission rate could cause large, rapid change in the volume of the fuel because of bubble growth.

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