NUREG/CR-2437 Vol. I

ANL-81-77 Vol. I

NUREG/CR-2437 Vol. I

ANL-81-77 Vol. I

MASTER

**LIGHT-WATER-REACTOR SAFETY
RESEARCH PROGRAM:
QUARTERLY PROGRESS REPORT**

January—March 1981

ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

**Prepared for the U. S. NUCLEAR REGULATORY COMMISSION
under Interagency Agreement DOE 40-550-75**

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NUREG/CR-2437 Vol. I

ANL-81-77 Vol. I

(Distribution
Code: R3)

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60439

NUREG/CR--2437 Vol. 1
DE82 007207

LIGHT-WATER-REACTOR SAFETY
RESEARCH PROGRAM:
QUARTERLY PROGRESS REPORT

January--March 1981

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Date Published: November 1981

Previous reports in this series

ANL-80-86	January--March 1980
ANL-80-107	April--June 1980
ANL-81-15	July--September 1980
ANL-81-42	October--December 1980

Prepared for the Division of Reactor Safety Research
Office of Nuclear Regulatory Research
U. S. Nuclear Regulatory Commission
Washington, D. C. 20555

Under Interagency Agreement DOE 40-550-75

NRC FIN No. A2016

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RESEARCH PROGRAM:
QUARTERLY PROGRESS REPORT

January--March 1981

ABSTRACT

This progress report summarizes the Argonne National Laboratory work performed during January, February, and March 1981 on water-reactor-safety problems. The research and development area covered is Transient Fuel Response and Fission-product Release.

NRC
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FIN Title

A2016 Transient Fuel Response and Fission-product Release

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EXECUTIVE SUMMARY

A mechanistic model for the prediction of microcracking (grain-boundary separation) during transient conditions has been generated within the context of the FASTGRASS computer code. A model based on the work of DiMelfi and Deitrich describing ductile/brittle behavior has been replaced by one based on the work of Beere and Speight, Chuang and Rice, and Chen and Argon. The theory underlying this new model is described and its proposed implementation in the prediction of DEH test results is outlined.

TRANSIENT FUEL RESPONSE AND FISSION-PRODUCT RELEASE

Principal Investigator:

J. Rest

A. Introduction and Summary

A physically realistic description of fuel swelling and fission-gas release is needed to aid in predicting the behavior of fuel rods and fission gases under certain hypothetical light-water-reactor (LWR) accident conditions. To satisfy this need, a comprehensive, computer-based model, the Steady-state and Transient Gas-release and Swelling Subroutine (GRASS-SST), its faster running version, FASTGRASS, and correlations (PARAGRASS) based on analyses performed with FASTGRASS are being developed at Argonne National Laboratory (ANL). This model is being incorporated into the Fuel-rod Analysis Program (FRAP) code under development by EG&G Idaho, Inc. at the Idaho National Engineering Laboratory.

The analytical effort is supported by a data base and correlations developed from characterization of irradiated LWR fuel and from out-of-reactor transient heating tests of irradiated commercial and experimental LWR fuel under a range of thermal conditions.

B. Modeling Fuel/Fission-product Behavior: FASTGRASS Model for Fuel Microcracking (S. R. Rajan, F. A. Nichols, and J. Rest)1. Introduction

A number of experiments have been performed to simulate accident transients in both thermal and fast reactors.¹⁻⁴ A wide spectrum of fuel response modes has been observed, ranging from very brittle to very ductile behavior.⁵ The ductile mode is associated with the development of substantial intergranular porosity, while the brittle mode is characterized by rapid grain-boundary separation (or microcracking). Extensive grain-boundary separation or microcracking has been observed in irradiated LWR fuel subjected to thermal transients by means of a direct electrical heating (DEH) technique.⁴ Such separation has also been observed in fuel tested in the PBF Reactor in Idaho and in commercial fuel that had undergone a power excursion. Assuming a failure to scram, the geometry of fuel that exhibited a "ductile" response could remain intact well into the transient, leading to large-scale fission-gas swelling as temperatures approach melting, and possibly frothing as melting proceeds (due to the availability of fission gas on the boundaries). The "microcracking" mode could lead to quicker transient release of fission gas from the grain faces to the edges and from the edges to the fuel exterior, and could result in particulate dispersal of the fuel.

It has been experimentally observed that a large fraction of transient fuel swelling is intergranular in nature. Hence, the behavior of fission gas on the grain boundaries is very critical in determining the accident

outcome. To understand the nature of intergranular gas behavior, one must first review the possible mechanisms in operation at the grain boundary. Recent literature on metallic alloys is very useful here.

Typically, metallic alloys, even if ductile at low temperatures, fail in an intergranular fashion⁶ at relatively low strains when held under tensile loading at temperatures of 0.3-0.9 of their melting points. This reduction in ductility and premature rupture under creep conditions (which can correspond to strains as low as 1%) has been attributed to the nucleation, growth and coalescence of cavities (or voids) on grain boundaries approximately normal to the applied stress. These two common types of cavities are often referred to as round (r-type) and wedge (w-type).⁷ The development of r-type voids leading to limited strains to failure is termed "creep cavitation." The formation of such voids on boundaries normal to the applied stress has been observed experimentally.⁷⁻⁹

In the present report, attention is directed towards the development and growth of r-type voids. At the high temperatures encountered in nuclear fuel, this form of cavitation may be more likely to occur than any brittle crack propagation effects. The reason for this assumption is given in the next section.

2. The DiMelfi-Deitrich Microcracking Model

The DiMelfi-Deitrich model^{5,10} was a pioneering effort in the understanding of grain-boundary separation due to overpressurization of grain-boundary gas bubbles during a thermal transient. The model provides a simple criterion for "ductile" or "brittle" behavior of the grain boundary by comparing the rate of departure from equilibrium due to internal overpressurization of the cavity with the rate of equilibration by grain-boundary vacancy diffusion into the bubble (as analyzed by Hull and Rimmer in Ref. 11).

A bubble arriving at a grain boundary during a transient is treated as a nucleate crack. As a consequence of this assumption, the gas bubble equilibrium equation turns out to be identical with the instability equation for the crack-like bubble. This implies that the gas bubble would grow as a brittle (elastic) crack in response to the slightest increase in the internal pressure during the transient. A gas cavity is in thermodynamic equilibrium (i.e., at a minimum in free energy) when

$$(p + \sigma)R = \gamma, \quad (1)$$

where

p = internal gas pressure

σ = the normal stress on the boundary

R = radius of curvature of the bubble surface

γ = the surface energy of UO_2 .

The RHS of Eq. 1 has γ instead of 2γ because of the simplified cavity geometry being considered (Fig. 1). However, there should be regions where the bubble is stable. Perturbing the equilibrium pressure slightly should have the effect of driving the system back towards equilibrium by a decrease in p (due to diffusional cavity growth or plastic relaxation of the matrix). The decrease of p can be achieved by brittle (elastic) crack growth only if two conditions are met:

- a. Sufficient energy must be available to create the new crack surfaces.
- b. There is a mechanism by which cracking can occur; i.e., the crack tip has to remain sharp in a Griffith sense, without crack blunting caused by plastic flow or diffusional processes.

Under normal or accident reactor conditions, the temperature would be high enough for diffusion as well as plastic relaxation to operate. Further work is needed, however, to determine whether these growth processes or brittle crack propagation predominates.

Fig. 1
Schematic of a Gas Bubble on a Grain Boundary Under Tension (Applied Normal Stress of σ)

3. Coupling of FASTGRASS/DEHTTD/DiMelfi-Deitrich Model

The DiMelfi-Deitrich model was coupled to FASTGRASS.¹² This version was interfaced with the DEH Transient Temperature Distribution (DEHTTD)⁴ code. The latter calculates the transient thermal histories for the DEH tests and is used to generate the input for FASTGRASS. The results of one of the DEH tests carried out at ANL were used to calibrate this coupled version. The results of this effort have been reported previously.¹² The agreement between observed and predicted pore-solid surface areas was found to be reasonable for the three tests to which the coupled code was applied. In view of the discussion in the previous section, however, it was felt that an alternate model for microcracking should be implemented.

4. An Alternate Microcracking Model

Baluffi and Seigle¹³ showed that vacancies could be produced at the grain boundary at a sufficient rate to allow cavity growth. Subsequently, in 1959, Hull and Rimmer¹¹ published their classic paper on cavity growth by grain-boundary diffusion driven by stress gradients normal to the boundary. A correction has been made to the Hull-Rimmer model by Weertman,¹⁴ who introduced the boundary condition of zero vacancy flux at the midpoint between two voids. The model has also been modified in various ways to account for boundary conditions^{14,15} and for non-equilibrium shapes of growing cavities.^{16,17} Other investigators have studied the importance of surface diffusion,¹⁶⁻¹⁸ as well as growth by lattice diffusion.^{19,20}

Differential thermal expansion produces large stresses in the fuel during a transient. Depending on whether they are compressive or tensile, these stresses will act to enhance or inhibit the gas bubble growth rate on grain boundaries normal to the stresses. Basically, the applied normal stresses create an excess of thermal vacancies at the grain boundary. These vacancies diffuse to voids which provide sinks of low chemical potential. The overall fuel creep rate is another factor that determines the rate of separation of boundaries due to intergranular bubble or crack growth.

Gas bubbles on a grain boundary acted upon by normal stresses can grow by a variety of mechanisms: diffusion control, plastic deformation (or creep) control, or brittle cracking along the boundaries.

Most theories of intergranular creep rupture of metallic alloys are complicated by the calculation of the nucleation rate of voids at the boundary (especially if second-phase particles are present). In nuclear reactor fuel analysis, this complication is avoided because fission gas bubbles already exist at the boundaries. Thus, the models developed for cavity growth should be directly applicable.

a. The Beere-Speight Diffusion-controlled Growth Model

Hull and Rimmer calculated the growth rate of voids on a grain boundary under the action of a tensile stress normal to the boundary, making the following assumptions (as enumerated by Beere):²¹

(1) Surface diffusion is very rapid, and maintains an equilibrium lenticular cavity shape as the cavity grows.

(2) The atom flux from the void surface "plates out" uniformly in the grain boundary, causing the grains to move apart as rigid bodies (i.e., no plastic deformation).

(3) During cavity growth, the diffusing material can be transported much more rapidly by grain-boundary diffusion than by lattice diffusion.

(4) The boundary behaves as a perfect source of vacancies, i.e., the local vacancy concentration is in equilibrium with the local normal stress.

(5) Vacancy fluxes in the boundary can be adequately described by classical diffusion theory.

(6) At the cavity tip, local equilibrium exists, so that the local applied stress is $2\gamma/R$, which is the capillarity compression stress on the boundary.

(7) When the grain boundaries move apart owing to atom plating, they can do so in an unconstrained fashion.

Consider an array of lenticular voids on a grain boundary (Fig. 2), with an internal pressure p , void radius a , and average inter-void spacing $2c$. Associated with each is a grain-boundary region of radius c . σ_∞ is the applied stress value far from the boundary. Let the normal traction in the grain boundary be $\sigma_n(r)$. The chemical potential of atoms in the boundary relative to the unstressed state is then²²

$$\Delta\mu(r) = -\sigma_n(r)\Omega, \quad (2)$$

where

$\Omega \equiv$ atomic volume.

Fig. 2. Schematic of Lenticular Gas Bubbles on a Grain Boundary, and the Variation of Normal Stress $\sigma_n(r)$ in the Radial Direction

The drift velocity V , the mobility B , and the driving force F are related by

$$V = B \cdot F \quad (3)$$

where

$B = D/kT$, the Nernst-Einstein relation between the mobility and the diffusivity.

The driving force for diffusion is given by the gradient in the chemical potential, i.e.,

$$F = -\nabla(\Delta\mu). \quad (4)$$

The atom flux in the grain boundary is thus given by

$$J = -\frac{D_g}{kT\Omega} \nabla(\Delta\mu), \quad (5)$$

where

$1/\Omega \equiv$ the concentration of the diffusing species.

To assure the conservation of mass in the boundary,

$$\nabla \cdot J = \beta; \quad (6)$$

i.e., the divergence of the flux should equal the "plating-out" rate of the atoms from the void deposition surface on the boundary. The rate β of atom deposition per unit area is assumed constant over the entire boundary.

This leads to the differential equation for diffusion,

$$\nabla^2(\Delta\mu) + \frac{\beta kT\Omega}{D_g} = 0. \quad (7)$$

The boundary conditions are as follows:

(1) At $r = a$, the chemical potential is $(\Delta\mu) = -2\gamma\Omega/R$, where R is the radius of curvature of the void surface.

(2) At $r = c$, $\partial/\partial r(\Delta\mu) = 0$; i.e., the stress gradient is zero at the midplane, a symmetry condition.

(3) A mechanical force balance yields

$$\int_a^c 2\pi r \sigma_n(r) dr = \sigma_\infty \pi c^2 + p \pi a^2 - 2\pi a \gamma \sin \theta. \quad (8)$$

Equation 8 can be solved to yield a value for β , the plating rate of atoms. The void growth rate is related to the amount of material deposited on the grain boundary. Since the grain boundaries (and hence the two void surfaces) are moving apart, there is an additional "jacking" contribution to the void volume growth that is not directly related to the atoms diffusing from the void surface, i.e.,

$$\frac{dV}{dt} = \beta \Omega \delta_g \Pi (c^2 - a^2) + \beta \delta_g \Pi a^2 \Omega = \beta \Omega \delta_g \Pi c^2 \quad (9)$$

where

$$\delta_g \equiv \text{the grain boundary width} \approx \Omega^{1/3}.$$

Substitution of β in Eq. 9 results in

$$\frac{dV}{dt} = \frac{2\pi D_g \delta_g \Omega}{kT} \frac{\left(\sigma_\infty + p - \frac{2\gamma}{R}\right)}{\ln\left(\frac{c}{a}\right) - \frac{3}{4} + \frac{a^2}{c^2} \left(1 - \frac{a^2}{c^2}\right)}, \quad (10)$$

which is equivalent to the Beere-Speight formula except that we have added the effect of the internal pressure p .

b. Non-equilibrium Cavity Shapes

The assumption that cavities retain their equilibrium shape may not always be valid. Very often, experimentally observed cavity shapes are flat and disklike or "penny-shaped." In a recent report on cavitation, Cane² and Greenwood²³ observed that cavity heights grew in the early part of the experiment, but stayed constant in the latter part. This suggests a transition from a quasi-equilibrium lenticular growth mode to a non-equilibrium crack-like mode where the void elongates in the plane of the grain boundary under conditions such that surface diffusion cannot keep up with grain-boundary diffusion in retaining an equilibrium lens-shape.

Chuang and Rice treated the problem of a long, crack-like cavity growing at a fixed rate in a grain boundary.¹⁶ Chuang et al. extended these results for non-equilibrium cavity shapes,¹⁷ explicitly including the effects of stresses normal to the boundary, and developed approximate "similarity" solutions for the full range between equilibrium and crack-like growth as treated by Chuang and Rice. For the latter, the surface flux at the crack tip is given by

$$(J_s)_{\text{tip}} = 2 \sin\left(\frac{\theta}{2}\right) \left(\frac{B}{\Omega}\right) \left(\frac{v}{B}\right)^{2/3}, \quad (11)$$

where

$$B = D_s \delta_s \Omega \gamma / kT$$

and

D_s = surface diffusivity

δ_s = surface diffusion layer thickness $\approx (\Omega)^{1/3}$

v = propagation velocity of the tip in the boundary plane.

The width of the void in steady-state crack-like propagation is given by¹⁷

$$w = 4 \sin\left(\frac{\theta}{2}\right) \left(\frac{B}{v}\right)^{1/3}, \quad (12)$$

where the terms in the RHS are as described above.

c. Coupling of Diffusion, Plastic Deformation, and Non-equilibrium Effects

The assumption that cavities grow by grain-boundary diffusion alone leads to a linear dependence of the growth rate on applied stress, i.e., the rupture time is inversely proportional to applied stress. However, it has been known for some time that the rupture time in creep cavitation is often inversely proportional to the applied stress raised to a large power. Monkman and Grant²⁴ derived an empirical correlation of the form

$$\dot{\epsilon}_{ss} t_r = \text{constant}, \quad (13)$$

which has been verified for a wide range of metals and alloys for a wide range of strain rates and stresses.²⁵ $\dot{\epsilon}_{ss}$ is the steady-state creep rate and t_r is the rupture time. Deviation from this relationship can be expected for materials exhibiting large creep transients,²⁴ i.e., where rupture occurs while the material is still in the primary creep regime. The Monkman-Grant relation suggests that creep fracture is somehow linked to the "power-law" creep mechanisms, and that excluding the effects of plastic deformation on the cavitation process would lead to incorrect predictions of rupture times, especially at elevated temperatures.

The classical assumption for vacancy creation in the grain boundaries is one of uniform creation over the entire boundary. Conversely, the atoms leaving the cavity tip are assumed to plate out uniformly over the boundary, leading to a rigid-body translation of the grains. In many practical situations, however, the body undergoing creep cavitation deforms plastically, and the strain created by the plating out of atoms on the boundary near

the tip can be accommodated by dislocation creep of the matrix farther away from the tip. This implies that the vacancy creation rate (or atom plating rate) can be non-uniform over the boundary, i.e., matter flow can be accommodated locally near the cavity by the dislocation creep strain of the surrounding matrix. The effective diffusion distance is shortened, thereby speeding up the cavity growth rate. The combined effects of dislocation creep and diffusion can lead to cavity growth rates that are greater than those possible for either mechanism acting alone.

Beere and Speight²⁶ and Edwards and Ashby²⁷ made the first attempts to combine the effects of diffusion and creep on grain-boundary cavitation. Needleman and Rice introduced a finite-element solution to the coupled approach.²⁸ Chen and Argon have proposed an analytical closed-form model²⁹ which not only combines diffusional and plastic flow effects, but also accounts for the non-equilibrium crack-like, elongated shapes of cavities. Their model couples the Beere-Speight diffusion-controlled equilibrium model with the non-equilibrium cavity shape results of Chuang-Rice in a unique way. Their model is adopted as the basis for modeling the microstructural behavior in the present work.

Basically, they propose replacing the cavity half-spacing in the Beere-Speight model with an effective "diffusion distance." The Beere-Speight expression for the cavity growth rate can be rewritten as

$$\frac{dV}{dt} = \frac{2\pi D_g \delta g \Omega}{kT} \frac{(\sigma_\infty + p) \left(1 - \frac{r_{crit}}{a}\right)}{\ln\left(\frac{c}{a}\right) + \frac{a^2}{c^2} \left[1 - \frac{1}{4} \frac{a^2}{c^2}\right] - \frac{3}{4}}, \quad (14)$$

where

$$r_{crit} = (2\gamma \sin \theta) / (\sigma_\infty + p) \equiv \text{the critical radius below which the cavity sinters shut instead of growing.}$$

By introducing a new variable

$$\lambda \equiv \left(\frac{D_g \delta g \Omega \sigma_\infty}{kT \dot{\epsilon}_\infty} \right)^{1/3},$$

where $\dot{\epsilon}_\infty$ is the far-field creep rate of the matrix, and replacing the half-spacing by $(a + \lambda)$, one obtains

$$\frac{1}{a^3 \dot{\epsilon}_\infty} \frac{dV}{dt} = \frac{2\pi \left(\frac{\lambda}{a}\right)^3 (\sigma_\infty + p) \left(1 - \frac{r_{crit}}{a}\right)}{\ln\left(\frac{a + \lambda}{a}\right) + \left(\frac{a}{a + \lambda}\right)^2 \left[1 - \frac{1}{4} \left(\frac{a}{a + \lambda}\right)^2\right] - \frac{3}{4}}, \quad (15)$$

Clearly, the substitution of $(a + \lambda)$ for the half-spacing is only valid when $(a + \lambda) < c$. Otherwise, c is retained.

The cavity growth rate can be related to the cavity shape by the conservation equation

$$\frac{dV}{dt} = 4\pi a \Omega (J_s)_{\text{tip}} \quad (16)$$

The tip flux $(J_s)_{\text{tip}}$ is obtained from the solution of Chuang et al.¹⁷ for the two limiting cases (quasi-equilibrium and crack-like growth). Noting that the cavity volume can be related to the cavity radius by

$$V = \frac{4}{3} \pi a^3 h(\theta), \quad (17)$$

where

$$h(\theta) = (1 - 3/2 \cos \theta + 1/2 \cos^3 \theta) / \sin^3 \theta,$$

and substituting the expression for $(J_s)_{\text{tip}}$ from Chuang et al.,¹⁷ we obtain

$$\left(\frac{4\pi h(\theta)}{\epsilon_\infty a} \frac{da}{dt} \right)_{\text{quasi-equi}} = 2\pi \left(\frac{\lambda}{a} \right)^3 \left\{ \ln \left(\frac{a + \lambda}{a} \right) + \left(\frac{a}{a + \lambda} \right)^2 \left[1 - \frac{1}{4} \left(\frac{a}{a + \lambda} \right)^2 \right] - \frac{3}{4} \right\}^{-1} \quad (18a)$$

$$\left(\frac{4\pi h(\theta)}{\epsilon_\infty a} \frac{da}{dt} \right)_{\text{crack-like}} = \alpha \left(\frac{\lambda}{a} \right)^{5/2} \left\{ \ln \left(\frac{a + \lambda}{a} \right) + \left(\frac{a}{a + \lambda} \right)^2 \left[1 - \frac{1}{4} \left(\frac{a}{a + \lambda} \right)^2 \right] - \frac{3}{4} \right\}^{-1} \quad (18b)$$

where

$$\alpha \left(\theta, \frac{D_g \delta_g}{D_s \delta_s}, \frac{\sigma_\infty \lambda}{\gamma} \right) = \frac{4\pi h(\theta)}{\left[4 \sin \left(\frac{\theta}{2} \right) \right]^{3/2}} \left[\left(\frac{D_g \delta_g}{D_s \delta_s} \right) \left(\frac{\sigma_\infty \lambda}{\gamma} \right) \right]^{1/2}.$$

α is a coupling parameter, and a measure of the effectiveness of boundary diffusion relative to surface diffusion. The mode yielding a faster growth rate is assumed to be favored. The transition from quasi-equilibrium to crack-like growth occurs when

$$\alpha = 2 \left\{ \left(\frac{\lambda}{a} \right) \left[\ln \left(\frac{a + \lambda}{a} \right) + \left(\frac{a}{a + \lambda} \right)^2 \left[1 - \frac{1}{4} \left(\frac{a}{a + \lambda} \right)^2 \right] - \frac{3}{4} \right] \right\}^{1/2}. \quad (19)$$

5. Research in Progress

The new microcracking model is being coupled with FASTGRASS, and the coupled version with the DEHTTD code. DEH test conditions will be fed into this code complex, and the prediction of gas release and pore-solid surface area (separated boundaries) will be compared with the observed values. Preliminary results indicate that the new model will have a wide degree of applicability, and will be better able to predict the nature of overall transient fuel response. We are also considering an analogous model developed by Beere.²¹

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