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

July-September 1980

by

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RESEARCH PROGRAM:
QUARTERLY PROGRESS REPORT

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ABSTRACT

This progress report summarizes the Argonne National Laboratory work performed during July, August, and September 1980 on water-reactor-safety problems. The research and development area covered is Transient Fuel Response and Fission-product Release.

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TABLE OF CONTENTS

LIGHT-WATER-REACTOR SAFETY
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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. The interrelationship between fuel fracturing (microcracking), temperature scenario, and fission-gas-bubble response is investigated. The fission-gas-bubble behavior is described using the FASTGRASS computer code. A model based on the work of DiMelfi-Deitrich describing ductile/brittle fuel behavior has been implemented into the FASTGRASS Analysis. The predictions of fission-gas release, radial distribution of released gas, radial distribution of microcracking, and fuel temperatures are compared to the results of transient direct-electrical-heating experiments on irradiated LWR fuel.

TRANSIENT FUEL RESPONSE AND FISSION-PRODUCT RELEASE

Principal Investigators:
J. Rest and S. M. Gehl

A. Introduction and Summary

A physically realistic description of fuel swelling and fisison-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-base model, the Steady-state and Transient Gas-release and Swelling Subroutine (GRASS-SST), its faster-running version, FASTGRASS, and correlations based on analyses performed with GRASS-SST, PARAGRASS, are being developed at Argonne National Laboratory (ANL). This model is being incorporated into the Fuel-rod Analysis Program (FRAP) code being developed by EG&G Idaho, Inc., at the Idaho National Engineering Laboratory (INEL).

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 of Fuel/Fission-product Behavior (J. Rest and S. Zawadzki)

1. FASTGRASS Model for Fuel Microcracking

a. Introduction

Extensive grain-boundary separation (microcracking) has been observed in irradiated Light Water Reactor (LWR) fuel subjected to thermal transients by means of a direct electrical heating (DEH) technique.[1] Relatively strong correlations have been reported between the observed transient fission-gas release and the measured increase in pore-solid surface area due to the microcracking. Observations of the posttest microstructures have indicated that intergranular separations can be produced by the diffusion-controlled processes of growth and coalescence of fission-gas bubbles. However, crack-like separations have also been observed at radial positions where the predominant features on the grain surfaces are isolated fission-gas bubbles. That is, the gradual processes of bubble growth and coalescence can be interrupted by the more rapid process of crack propagation. Similar types of grain-boundary separation have also been observed in fuel tested in the PBF Reactor in Idaho and in commercial fuel that had undergone a power excursion.[1]

The high gas releases during DEH testing, the microstructures observed in transient-tested fuel, and the correlations between gas release and microstructural change indicate that the mechanisms responsible for the formation of the intergranular porosity can operate in series with intragranular

bubble diffusion to transport fission gases from the grain interiors to the di- fuel surface. In addition, the intergranular separations that form during e transient heating reduce the effective thermal conductivity of the fuel and thus affect the fuel-temperature scenario.

GRASS-SST[2,3] and FASTGRASS[4] are mechanistic computer codes for predicting fission-gas behavior in UO_2-base fuels during steady-state and transient conditions. FASTGRASS was developed in order to satisfy the need for a fast-running alternative to GRASS-SST. Although based on GRASS-SST, FAST-GRASS is approximately one to two orders of magnitude quicker in execution, depending on the complexity of the problem. The GRASS-SST and FASTGRASS transient analysis has evolved through comparisons of code predictions with the fission-gas release and physical phenomena that occur during reactor operation and DEH transient testing of irradiated light-water reactor fuel.

An apparent anomaly is the reasonable FASTGRASS (and GRASS-SST) predictions[4] of DEH Transient-Gas Release made without including the phenomena of microcracking, in contrast to the strong experimental correlation[1] between Transient-Gas Release and Fuel Fracturing. R. J. DiMelfi and L. W. Deitrich have developed a model[5] for crack propagation in which the fuel response (ductile or brittle) is determined by comparing the rate of grain-boundary bubble growth as a sharp crack with the growth rate by mass transport. Preliminary qualitative comparison with the DEH test results indicated that the model has the capability to provide a good description of transient fuel behavior.[1] Based on these results, the DiMelfi-Deitrich model was coupled with FASTGRASS in conjunction with the existing treatment of long-range bubble interlinkage, to calculate gas release from the grain faces and grain edges.

These models have been interfaced with the DEH Transient Temperature Distribution (DEHTTD) code[1] used to calculate the transient temperature histories of the DEH tests. The transient heat-transfer equation is solved by DEHTTD, which accepts measured values of current, voltage, and surface temperature as input and uses expressions taken from the literature for the thermal and electrical conductivities of UO_2. A previous model[1,3] for the effect of intergranular separation on heat transfer has been modified to use the results of the DiMelfi/Deitrich crack propagation model.

The FASTGRASS/DiMelfi-Deitrich/DEHTTD code complex has been executed for the available DEH tests. Comparisons are made between the calculated and measured values for the transient gas release, the radial distribution of retained gas, and the radial distribution of pore-solid surface area. The interrelationships among the various physical mechanisms and related phenomena are examined, and the resolution of the above-mentioned anomaly is discussed.

b. Description of FASTGRASS

The most important differences between FASTGRASS and GRASS-SST are in the algorithms used for calculating the evolution of bubble density and size over time. In GRASS-SST, the bubble-size distribution is specified by calculating the densities of bubbles in each of a number of bubble-size classes. Each bubble-size class is characterized by an average number of atoms per bubble, the value of which differs from that of the preceding size class by a constant multiplier. (The number of size classes is a variable that is determined dynamically during a computer run.) Changes in the bubble-size distribution, caused by bubble coalescence and re-solution, for example, are determined by solving a large number of integral-differential equations for each time step. Solutions are carried out for bubbles on grain surfaces, along dislocations, and in the bulk matrix. The iterative solution of a large number of coupled equations is a major contributor to the computer run times of GRASS-SST.

In contrast to the multiclass description of the bubble-size distribution in GRASS-SST, FASTGRASS uses only one bubble-size class. Whereas the grain face and grain edge fission-gas-bubble behavior in FASTGRASS is based entirely on this single size-class description, the description of intragranular fission-gas behavior includes the kinetics of fission-gas-atom generation and migration and fission-gas-bubble/gas-atom interactions. Model improvements have been recently implemented for the description of bubbles on the grain edges. In GRASS-SST, the same delineation of bubble sizes in terms of the number of gas atoms per bubble is used for intergranular, intragranular, and dislocation bubbles. This is possible because enough size classes are available to fully characterize the distributions. In FASTGRASS, with only one bubble-size class available, separate descriptions of the size classes are necessary for the intragranular, grain face, and grain edge bubbles. The intragranular single gas atoms are characterized by number density. The grain face and grain edge bubbles are characterized by number density and the average number of atoms per bubble, S_i. The number densities are determined by solving the following set of equations:

$$\frac{df}{dt} = -a_0 f^2 - b_0 f + c_0, \tag{1}$$

$$\frac{dg_1}{dt} = -b_1 g_1 + c_1, \tag{2}$$

$$\frac{dg_2}{dt} = -b_2 g_2 + c_2, \tag{3}$$

and

$$\frac{dg_3}{dt} = -b_3 g_3 + c_3, \tag{4}$$

where f and g_1 are the single-atom and bubble densities, respectively, for the intragranular region, and g_2 and g_3 are the bubble densities, respectively, for the grain faces and edges. The other parameters are defined as follows:

a_0: Rate, in cm^3/s, at which intragranular gas atoms are lost due to nucleation into gas bubbles.

b_0: Rate, in s^{-1}, at which intragranular gas atoms are lost due to diffusion to gas bubbles and migration to grain boundaries.

c_0: The rate, in atoms cm^{-3} s^{-1}, at which intragranular gas atoms are generated by fissioning and gas-bubble re-solution.

b_1: Rate, in s^{-1}, at which intragranular bubbles are lost by migration to grain boundaries and re-solution.

c_1: Rate, in atoms cm^{-3} s^{-1}, at which intragranular bubbles are produced by nucleation and diffusion of gas atoms to bubbles.

b_2: Rate, in s^{-1}, at which grain-face bubbles are lost by migration out of the annulus, migration to the grain edges, and re-solution.

c_2: Rate, in atoms cm^{-3} s^{-1}, at which grain-face bubbles are produced by diffusion of gas from the lattice, migration of gas into the annulus (up the temperature gradient) from the bordering annulus, and re-solution (i.e., bubble shrinkage).

b_3: Rate, in s^{-1}, at which grain-edge bubbles are lost by migration through the grain-edge interconnected porosity to the exterior of the fuel.

c_3: Rate, in atoms cm^{-3} s^{-1}, at which grain-edge gas bubbles are produced by the diffusion of grain-face bubbles to the grain edges.

The above parameters are, in general, functions of the average bubble size, S_i (i = 1, intergranular bubble; i = 2, grain-face bubble; and i = 3, grain-edge bubble), which is determined as follows. During each time step, the processes of bubble nucleation, gas-atom diffusion to bubbles, bubble coalescence, and bubble re-solution can lead to changes in the average bubble size. The rate, R_i at which the average-bubble size of the distribution grows due to the above processes is given by

$$R_i = \text{Growi-Srnki-Stabi}; \tag{5}$$

where Growi is the coalescence rate between i-size bubbles, Srnki is the re-solution rate of i-size bubbles, and Stabi is the rate at which i size bubbles are being produced.

If Srnki is greater than Growi, then the rate at which the average-bubble size of the distribution shrinks is given by

$$R_i = \text{Srnki} - \text{Growi} - \text{Stabi}. \qquad (6)$$

Note that a change in bubble size, S_i leads to a corresponding change in bubble density, g_i, since the total number of gas atoms in bubbles is conserved. Physically, the above formalism for determining dS_i/dt is based on an incremental approach to updating S_i as a function of time.

The coefficients (a_0, b_0, c_0, a_1, ... etc.) in Eqs. 1-4 are computed as a function of time by taking into simultaneous account the major mechanisms thought to influence fission-gas behavior.

Models are included for the effects of production of gas from fissioning uranium atoms, bubble nucleation and re-solution, bubble diffusion, bubble migration, bubble coalescence, gas-bubble/channel formation on grain faces, temperature and temperature gradients, interlinked porosity, non-equilibrium effects, microcracking and fission-gas interaction with structural defects on both the distribution of fission gas within the fuel and on the amount of fission gas released from the fuel. From these models, an equation of state for xenon, and bubble diffusivities based on experimental observations in the steady state and phenomenological modeling of bubble mobilities during transient non-equilibrium conditions, FASTGRASS calculates the fission-gas-induced swelling due to retained fission-gas bubbles in the lattice, on grain faces, and along the grain edges. It also calculates the fission-gas release as a function of time for steady state and transient thermal conditions. Fission gas released from the fuel is assumed to reach the fuel surface by successively diffusing from the grains to grain faces and then to the grain edges, where the gas is released through a network of interconnected tunnels of fission-gas-induced and fabricated porosity. The above models (with the exception of microcracking) have been described previously. *However, it is important to note that the GRASS model for gas bubble diffusion through solid UO_2 is unique in that it relates the bubble diffusivities to the fuel yield stress, heating rate, and vacancy mobility, as well as to fuel temperature and bubble radius.*

c. FASTGRASS Model for Determining Ductile/Brittle Fuel Response

The ability to determine whether microcracking will occur during a given thermal transient is an important element in the prediction of fuel temperatures and fission-gas release. In principle, a "classical" mechanical treatment, involving the high-temperature stress/strain relationships of UO_2, could be used to study microcracking. Besides being very complex, this approach would require knowledge of the mechanical properties of UO_2, including strain-rate effects, at high temperatures. Data in this area are sparse, and are almost nonexistent for temperatures in excess of 2400 K.

The FASTGRASS approach to modeling ductile/brittle behavior of oxide fuels is based on a model developed by DiMelfi and Deitrich.[5] In their model, the growth of a grain-boundary bubble under the driving force of internal pressurization is estimated. The volume growth rates due to crack propagation and to diffusional processes are compared to determine the dominant mode of volume swelling. Knowledge of the mechanical properties of UO_2 is not required.

The underlying structure of the model can be summarized as follows: A fission-gas bubble on a grain boundary can be viewed as a crack nucleus. It can be shown that such a crack will propagate unstably if the internal bubble pressure exceeds that required for bubble equilibrium, i.e., if

$$p > \frac{\gamma_s}{\rho} - \sigma \tag{7}$$

where p is the internal bubble pressure, γ_s is the fuel-gas surface energy, ρ is the bubble radius of curvature, and σ is the tensile stress normal to the boundary.

Further, if a bubble, initially at equilibrium, is subjected to transient heating, the internal pressure will increase above the equilibrium value. Under these conditions, crack propagation will occur unless diffusional growth of the bubble occurs rapidly enough to maintain equilibrium conditions.

During most thermal transients, the initial mode of bubble growth will be crack propagation. The cracked bubble may be able to reattain its equilibrium shape by diffusional transport of material along the grain boundary. However, if the heating rate is sufficiently high, repressurization can take place. Thus, the competition between diffusional growth and crack growth determines whether bubbles tend to remain isolated or rapidly become part of an interconnected network of microcracks.

In the DiMelfi-Deitrich analysis, the dominant mode of bubble growth is determined by comparing the rates of volume swelling due to crack propagation and diffusional growth. In practice, this is done by comparing the instantaneous value of the grain-boundary diffusion coefficient, D_g, with the minimum value needed to maintain bubble equilibrium, D_g^{min}. (The calculation of D_g^{min} is discussed in detail in Ref. 5.) If $D_g < D_g^{min}$, cracking dominates; this behavior is termed "brittle." If $D_g > D_g^{min}$, diffusional growth or "ductile" behavior dominates.

The minimum diffusion coefficient D_g^{min} is given by

$$w \cdot D_g^{min} = \left(\frac{mk}{\gamma_s}\right)^2 \frac{k\lambda AT}{HL\Omega(\Delta p)}, \tag{8}$$

where w is the grain-boundary width, γ_s is the surface energy of UO_2, L is the bubble length, Ω is the molecular volume of UO_2, H is a geometric factor, k is Boltzmann's constant, T is the temperature, A is the instantaneous heating rate, λ is the average bubble spacing in the grain boundary, and Δp is the pressure in excess of that for an equilibrium grain-boundary bubble. In deriving Eq. 8, the ideal gas law and zero normal stresses on the grain boundary were assumed. FASTGRASS provides the gas-bubble input to Eq. 8 as a function of time.

The calculation of D_g^{min} must evolve from a consistent scheme to determine Δp, the driving force for grain-boundary diffusion. DiMelfi has proposed such a scheme[6] by considering the force balance that results in crack arrest.

To evaluate the effects of crack growth versus equilibrium bubble growth on such properties as intergranular swelling, grain-boundary areal coverage, interconnected porosity, and gas release, some of the geometric aspects of the DiMelfi-Deitrich model must be transmitted back to the FASTGRASS and DEHTTD codes at each instant of time. For example, the thermal conductivity, F_c, of UO_2 is given by

$$F_c = F_c^\circ \left[1.0 - C_2 \sum_i C_1 \left(\frac{A_c}{A_e} \right) i x_i h_i \right] = F_c^\circ \left(1.0 - C_2 S_V \right) \qquad (9)$$

where F_c° is the thermal conductivity of uncracked, stoichiometric UO_2, C_1 and C_2 are constants, $(A_c/A_e)_i$ is the ratio of the areal coverage of a crack to that of an equilibrium bubble,[6] x_i is the projected grain-face areal coverage per unit volume of bubbles, h_i is a calculational time step, and S_v is the pore-solid surface area. The constant C_1 is nonzero whenever $D_g < D_g^{min}$ (see Eq. 8). The effect of microcracking on the thermal conductivity of UO_2, as described in Eq. 9, can result in values for $F_c \sim 50\%$ of the value of dense fuel. A change of this magnitude will have a strong effect on calculated temperature profiles. As an example, DEHTTD calculations that ignored the effect of microcracking gave errors >600 K in the center temperature. The need to include the effect of microcracking on thermal conductivity is not limited to electrical heating. A correction to thermal conductivity is also required for nuclear transients in which microcracking impedes radial heat flux.

d. Results

FASTGRASS was executed with the LIFE-LWR fuel behavior code[7] for the steady-state irradiation of a fuel rod in the H. B. Robinson reactor in order to generate the required initial conditions for a transient analysis. Subsequently, FASTGRASS was executed with the DEHTTD code for a series of DEH tests. The calculational scenario is as follows: After receiving the DEH test operating conditions, DEHTTD calculates the radial temperature profile and passes this information to FASTGRASS. FASTGRASS then calculates the fission-gas response and utilizes these results for the calculation of fuel microcracking. In turn, if microcracking occurs, the fission-gas release, retention, and

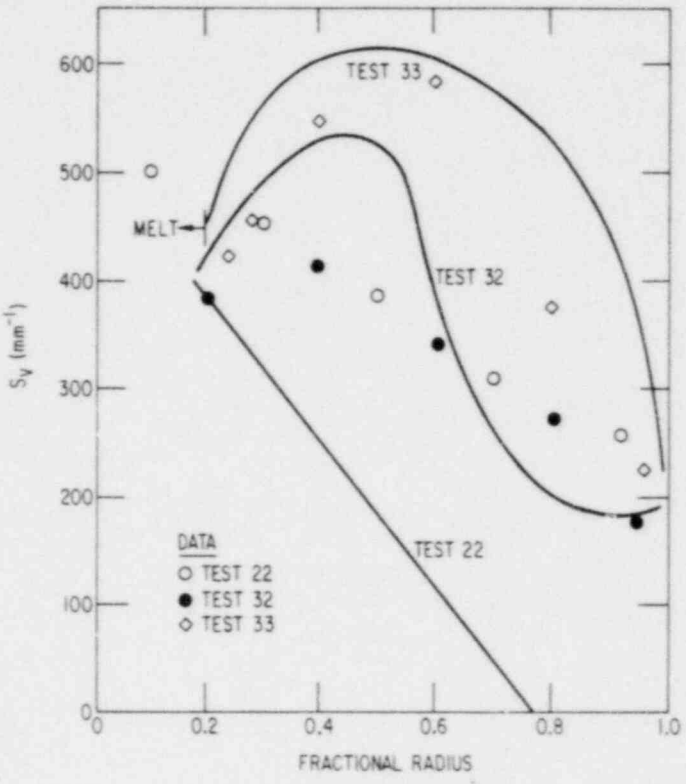

Fig. 1. FASTGRASS Calculations of Pore-solid Surface Area (S_V) as a Function of Pellet Radius for Tests 22, 32, and 33, Compared with Experimental Data

swelling results are updated accordingly. Finally, the microcracking results are passed back to DEHTTD where the thermal conductivity expression is modified according to Eq. 9, and the calculation proceeds to the next time step.

The constant C_2 in Eq. 9 was determined by matching the calculated and observed melt radii in Test 33. The resulting value was used for all other DEH tests.

Figure 1 shows the results of FASTGRASS calculations of pore-solid surface area (S_V) as a function of pellet radius for DEH Tests 22, 32, and 33, and measured values of S_V for the same tests (this pore-solid surface is presumably produced mainly by fuel microcracking.[1] The predictions for Test 33, which were normalized to the data by adjustment of the constant C_2, agree reasonably well with the measured values. The predicted values of the pore-solid surface area for Tests 22 and 32 are in reasonable agreement with the data. *It should be noted that the above model for fuel microcracking does not include the effect of externally applied stresses (e.g., those due to differential thermal expansion). Experimentally, external stress has been observed to have a pronounced effect on the amount and distribution of microcracks.*[1]

The predicted total fractional gas release during the transient is 13.6, 28, and 43.6%, respectively, for Tests 22, 32, and 33; the corresponding measured values are 13.1, 16.1, and 40%. Figure 2 shows the predicted radial profile of fission-gas release during DEH Test 33 compared with the data. The predictions of total fractional gas release and radial distribution of released gas for Tests 22 and 33 are in reasonable agreement with the data, while the gas release for Test 32 is overpredicted. This high gas release prediction for Test 32 is due, at least in part, to the overprediction of fuel microcracking [e.g., see Fig. 1 and Eq. 3; central melting for Test 32 was predicted to occur, while none was observed].

The question remains as to why FASTGRASS gives reasonable predictions[4] for transient gas release for DEH Test conditions even without

considering the effects of microcracking (and, for that matter, why the FASTGRASS prediction of transient gas release for Test 22 is in good agreement with the data while at the same time the radial distribution of pore-solid surface area is under-predicted). The answer to this apparent anomaly is as follows: In the absence of any model for microcracking (or in the event of an underprediction of microcracking), the fission gas which would have been vented via the cracks is predicted to remain on the grain boundaries. This fission gas causes the deformation of the grain edges (i.e., grain-edge fission-gas-bubble swelling) and the subsequent increased long-range interconnection of grain-edged tunnels. This connection of grain-edge tunnels provides the pathways for the enhanced fission-gas release.

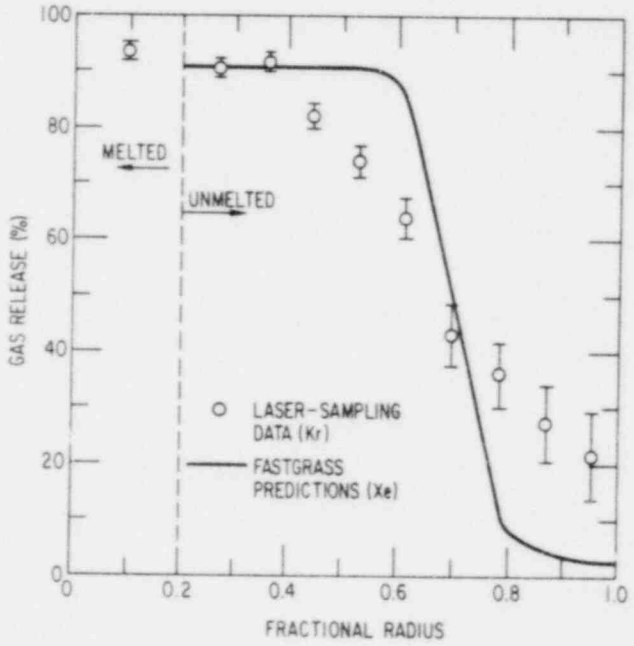

Fig. 2

FASTGRASS Calculations of the Radial Profile of Fission-gas Release during DEH Test 33, Compared with Experimental Data

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