Grain Boundary Processes and Transient Fission Gas Release During Fast Reactor Transients with a View to Determining the Loading of the Cladding

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ABSTRACT.

Due to their influence on clad loading, the fission gas phenomena due to the existence of fuel grain boundaries play an important role during fast reactor accidents. The recent transient gas release measurements are reviewed, and the fission gas inventory on grain boundaries is analysed for a few experiments. A simple transient gas release and swelling model is then proposed, which is based on diffusionless release (grain boundary cracking) and on equiaxed grain growth (grain boundary sweeping). Influence of fuel chemistry is emphasized.

1. INTRODUCTION

The determination of time and location of fuel pin failure plays an important role in the assessment of fast reactor accident scenarios. Pin failure results from the pressure exerted by pin inner gas and transient fuel swelling. This paper focuses on the transient gas release and swelling of solid oxide fuel, with emphasis on the surface processes connected to the existence of fuel grain boundaries.

Fission gases retained into the fuel correspond to an important pin disruption potential, which is not available as long as the fission gas remains either in solution in the oxide matrix or trapped in small bubbles. During normal fuel operation, a dynamic balance is established between fission gas precipitation into small intragranular bubbles, and fission-fragment induced resolution. High temperature excursions induce extensive gas precipitation into bubbles, resulting in swelling of the fuel. However, in solid fuel, the fast and important gas release observed at the same time appears in contradiction with the precipitation of the gas into the bubbles, and one has thus to explain how the gas appearing to be deeply trapped in intragranular bubbles can be rapidly transferred to grain boundaries or to open porosities. Different answers to that question have been proposed :

(i) the intragranular bubbles can be move either randomly (brownian motion), or under the influence of a temperature gradient (e.g. [1]) ;

(ii) thermal (thermodynamic) resolution of the gas contained in the bubbles is possible, if the bubbles are sufficiently overpressurized [2] ;

(iii) recrystallization of the oxide at high temperature can lead to the motion of dislocations or grain boundaries which are able to sweep the intragranular gas and bubbles [3].

2. REVIEW OF RECENT TRANSIENT FISSION GAS RELEASE MEASUREMENTS

Different experimental programmes have investigated the gas behaviour for a wide range of fuel heating rates, ranging practically from 1 K/s up to 10^6 K/s.

(i) For the fast heating rate tests [4,5,6], a rather clear picture is emerging : the fractional gas release is of the order of 15 to 50 % of the initially retained gas, as long as the fuel remains solid. To get a higher fractional release, one has to reach the fuel melting point. The gas release kinetics are unexpectedly fast, with characteristic times ranging from 2 - 4 milliseconds up to 50 milliseconds.

(ii) For the medium speed transients with time scales ranging from 0.2 to 2 seconds (e.g. [4,9]) post-test examination indicates extensive fuel morphology changes with, in some cases, intense gas bubble precipitation on grain boundaries, and in other cases, regions free of any bubbles adjacent to grain boundaries.

(iii) For still slower transients, numerous representative data can be found in the field of thermal reactor fuel ramp tests [10,11]. The gas release proceeds in two steps : a very fast release of 10 - 15 % which is instantaneous at the time scale of the experiments and a slower diffusional process, leading to a progressive and nearly complete gas release within a few hours.

The latter separation seems to correspond to two different physical phenomena : a diffusionless mechanism acting on very short time scales, and a diffusional process related to the slower microstructural changes which are sensitive to diffusion and surface properties.

3. THE GAS INVENTORY ON FUEL GRAIN BOUNDARIES.

The importance of the grain boundary fission gas inventory is due to the possibility of grain boundary fracture, and the corresponding instantaneous release of all the available gas. The distinction has to be made between equilibrium and non equilibrium grain boundary inventory : equilibration of the intergranular bubbles by surface and/or grain boundary diffusion or vapour transport requires some time, and during transients, significant deviations may be observed with respect to the equilibrium configuration.

Most observations pertaining the grain boundary bubbles are related to equilibrium grain boundaries. Extensive theoretical developments in that field have been achieved in the UK at Harwell and Berkeley (see e.g. [7] and [14]), and the morphology of equilibrium intergranular pores is now well understood. By taking into account pore shape, pore size and distribution, one obtains an equilibrium inventory which varies slowly with temperature, and that is approximately equal to $7\ 10^{14}$ atoms per square centimer of boundary.

When fuel is irradiated at low temperatures, the radiation induced defect product rate exceeds the thermal defect recovery rate, and the state of the grain boundary is far from equilibrium. One has thus to rely on an empirical approach to estimate the corresponding fission gas inventory. A simple way to measure the intergranular gas inventory is to heat the oxide up to a temperature where the grain boundary can be broken, fast enough to prevent other release mechanisms. Table I collects some results obtained by transient nuclear heating. One sees that the maximum boundary inventory obtained by assuming that the bubbles are in equilibrium is 10 to 30 times less than what can be measured during transient heating.

4. A SIMPLE MODEL FOR TRANSIENT SWELLING AND RELEASE

The experimental observations suggest that transient gas release and swelling could be modelled on basis of two phenomena. The first one is the diffusionless gas release due to boundary cracking ; the second phenomenon is grain boundary motion. The latter process is indeed consistent with the observed denuded grain boundaries [15] ; Speight and Greenwood's bubble sweeping theory [16] explains the presence of larger bubbles near the outer regions of the intragranular bubble cloud [15] ; the approximately isotropic distribution of the width of the denuded zones [15] indicates that it is surface energy which drives boundary motion, rather than temperature gradient. We identify thus the second phenomenon with transient equiaxed grain growth.

In steady state, we assume complete gas retention when the temperature is lower than a burn-up dependent limit T_L. Complete release is assumed above T_L. For $T < T_L$, one considers that 20 % of the produced gas is stored on the grain boundaries. The boundary gas inventory I_b is thus equal to $I_{b_o} = 0.2\ Y\ B$, where Y is the gas yield and B the burn-up. The intragranular inventory I_g is equal to $I_{g_o} = 0.8\ Y\ B$.

During the transient, one considers that the grain boundaries are fractured when T exceeds T_L ; the grain boundary gas can thus escape rapidly with a time constant τ_b equal to 2 milliseconds. The boundary inventory is described by :

$$\frac{dI_b}{dt} = -\frac{I_b}{\tau_b} \quad ; \quad I_b(t=0) = I_{b_o} \tag{1}$$

During boundary motion, intragranular gas is collected into intergranular pores ; the corresponding inventory I_p is described by :

$$\frac{dI_p}{dt} = -\frac{I_p}{\tau_p} - I_{g_o}\frac{d}{dt}\left(\frac{G_o}{G}\right)^3 \quad ; \quad I_p\ (t=0) = 0 \tag{2}$$

where G_o and G are respectively the initial and current grain sizes, and τ_p, a percolation time constant of the order of 200 milliseconds. The grain growth is considered to be controlled by vapour transport across equilibrated boundary pores ; this leads to

$$\frac{dG^3}{dt} = C\,\frac{P_{vap}}{I_p}\,T^{-3/2} \tag{3}$$

where C is a constant, and p_{vap} is the fuel vapour pressure. Eq. (3) is in excellent agreement with Mc Ewan and Hayashi results for fresh UO_2 and UO_{2+x}, as well as irradiated UO_2 [13]. The radius R_p of the intergranular pores is assumed to be equal to R_{po} + $1/2(G - G_o)$, and the transient swelling ϵ is calculated according to

$$\epsilon = kT \left[\frac{R_p I_p}{2\gamma_s + p R_p} + \frac{I_b}{p} \right] \tag{4}$$

where p is an external pressure, and γ_s the surface energy.

A key variable in the model is the fuel vapour pressure which depends on fuel chemistry : at high temperature, oxygen rich fission product compounds dissociate and increase the fuel stoichiometry and vapour pressure. This is shown by Fig. 1, where one has assumed chemical equilibrium.

Typical model results are provided in Fig. 2, where one has considered a fuel irradiated at 800°C up to 5 at.% burn-up, and heated up to 2800°C with ramp rates ranging from 10 to 10^7 K/s. Transient grain growth appears to be significant for ramp rate up to about 1000 K/s. In that regime, denuded grain boundaries are predicted, as well as important gas release. Above 1000 K/s, only boundary gas can escape before melting ; above 10^6 K/s, it cannot even escape anymore. The swelling calculated under 1000 K/s is found to be in the range 10 to 50 %, and it is quite sensitive to ramp rate and external pressure. The large swelling calculated above 1000 K/s results from the expansion of the boundary gas and corresponds to fuel fragmentation. The model predicts thus fairly different transient fuel behaviours below and above 1000 K/s ; the latter ramp rate corresponds to the limit between ductile and brittle fuel behaviour.

CONCLUSIONS

This presentation has focused of the fission gas processes which are responsible for the swelling of solid oxide fuel during fast reactor transients. Two main points have to be stressed :

(i) The grain boundary fission gas inventory can lead to diffusionless gas release, through fracture of the grain boundaries ; this means oxide fragmentation, in case of unrestrained fuel, and gas readily available for pin pressurization, if the cladding is still intact.

(ii) For longer time scales, grain boundary motion appears to transfer intragranular gas towards the grain boundaries. Of special importance is the sensitivity of this process to fuel chemical changes : it is indeed possible, by heating previously cold oxide, to obtain vapour pressures quite sufficient to promote fast grain growth.

ACKNOWLEDGMENTS.

This work was supported by the Commission of the European Communities, under study contract ECI-1076-B-7221-83-B. The author would like to thank the chairman and the members of the Whole Core Accidents Code Group, of the Fast Reactor Safety Working Group of the CEC, for their helpful suggestions and for their continuous interest in the present study.

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TABLE I

Observed intergranular fission gas inventories

Experiment	Burn-up at.%	Grain size micron	Local retained fission gas cc.N/g.ox	Local released fission gas cc.N/g.ox	Boundary gas inventory at/cm^2
TREAT-H3	2.3	4	0.52	0.39	1.5 10^{16}
SILENE-TP9	5	20	0.40	0.09	1.8 10^{16}
VIPER-MINIPINS	1 - 2	≅ 10	0.28-0.41	.04-.06	0.4-1.2 10^{16}
HFR-KWU/CE	4.5	6	1.20	0.36	2.1 10^{16}
Equilibrium grain boundary gas inventory					7 10^{14}

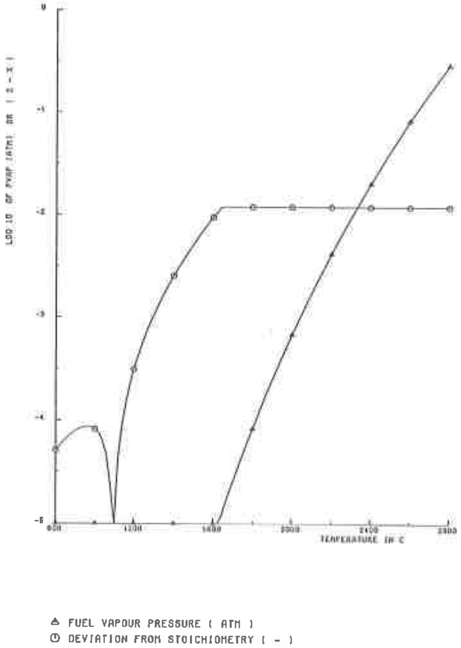

Figure 1 Variation of solid fuel transient swelling and fission gas release with temperature ramp rate (free fuel expansion with p = 0.1 MPa).

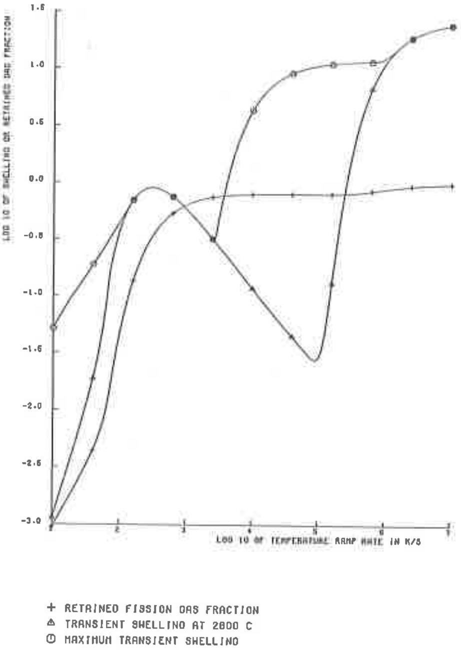

Figure 2 Evolution of oxide fuel stoichiometry and vapour pressure at chemical equilibrium during a 100 K/s temperature transient.