TRANSIENT FISSION GAS RELEASE
DURING FAST REACTOR TRANSIENTS

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TRANSPORT OF FISSION GAS RELEASE DURING FAST REACTOR TRANSIENTS

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This paper presents modelling work performed at BELGONUCLEAIRE in order to understand the processes by which fission products are released from oxide fuel during fast reactor transients, for heating rates up to 1000 K/s.

After a review of available experimental data, a model based on transient equiaxed grain growth is proposed which provides a simple interpretation of observed facts, including gas release, denuded zones and fuel swelling.

Application of this model shows that fission gas release is quite important in case of solid fuel, and stresses the impact of fuel burnup and stoichiometry on the fission gas release and swelling phenomena.
1. INTRODUCTION.

The successful operation of fast reactor oxide fuel up to high or very high burnups requires a quantitative understanding of fuel behaviour not only during steady-state irradiation, but also during normal and off-normal transients. The normal transients include power changes, load-follow, power ramps, while abnormal transients include mainly overpower or undercooling events. Important experimental programmes are underway in the frame of the European Fast Reactor Community, and they are described in other papers described at this conference [1,2,3].

This presentation focuses on the modelling work performed at BELGONUCLE-AIRE, in order to analyse and predict the processes by which gaseous fission products are released from oxide fuel during temperature transients, for heating rates up to about 1000 K/s. The latter limit is considered as boundary between operational and severe fast reactor transients, the latter ones having already been described previously [4].

2. THE TRANSIENT GAS RELEASE PROCESSES.

Temperature transients in oxide fuel are well known to lead to fission product release. There is, however, some paradox to be solved between steady-state and transient gas release. During steady-state, it is rather well established that gas release results from the diffusion of gas atoms towards the grain boundaries, the gas atoms are, in fact, the result of the resolution of small bubbles by fission spikes, so that in absence of fission, gas precipitation into the bubbles should lead to very low release fractions [5,6,7]. The paradox comes from the fact that, during temperature transients, extensive precipitation into bubbles is observed simultaneously with very high release fractions [4].

Four mechanisms have been proposed to solve this apparent contradiction between steady-state and transient gas release:

(i) during the transients, gas release occurs by diffusion of bubbles and not by the diffusion of gas atoms; the main driving force for bubble diffusion should be the large temperature gradient developed during, e.g. a power transient (see e.g. [8]);

(ii) during the important fission gas precipitation into bubbles, the latter become strongly overpressurized; this overpressure has been proposed as the driving force for thermally assisted gas atom resolution from the bubbles towards the surrounding matrix; the latter atoms diffuse then to the grain boundary and contribute so to the transient gas release [9];

(iii) more recently, it has been proposed that gas release could be due to the sweeping of the gas bubbles by moving dislocations, during annealing of the irradiated grains [10];

(iv) finally, transient fission gas release can be interpreted as the superposition of the diffusionless release of the grain boundary gas and the sweeping of the intragranular gas by grain boundaries during grain growth [4].

In this presentation, we focus on the latter mechanism as it allows a consistent interpretation of the observed experimental facts.
3. SHORT REVIEW OF EXPERIMENTAL DATA.

When submitting oxide fuel to temperature transients, striking microstructural changes are observed if the heating rate is of the order of 100 K/s or lower. These changes can be summarized as follows:

(i) extensive intragranular and intergranular bubble precipitation takes place;
(ii) when the transient is interrupted at temperatures lower than 2200°C, intergranular cracking is observed even in zones devoid of pretransient intergranular bubbles;
(iii) denuded zones are observed along the grain boundaries; in most cases, they are present only on one side of the grain boundary; HEDL measurements have concluded that the denuded zones are distributed uniformly in all directions [11];
(iv) the precipitating intragranular bubbles form a cloud, where the largest bubbles are found on the boundary [11];
(v) in many tests on samples irradiated at low temperature (burnup less than 3 at.%, linear power less than 200 W/cm), grain size variations are observed that must be attributed to the high temperature during the tests [12];
(vi) in instrumented tests, gas release is observed at temperatures as low as 1500°C in time scales of the order of the seconds [12].

It also important to note some characteristics of experiments with adiabatic heating like SILENE [13] or VIPER [14]: when irradiated fuel samples are submitted to enthalpy increases ranging from 0.7 to about 1 kJ/g, they release about 20% of their gas content in time scales of the order of milliseconds. This observation suggests that some part of the release process is not controlled by diffusional processes, but rather by a diffusionless process. An example of this could be grain boundary cracking, with the concurrent instantaneous release of all the gases adsorbed on the grain surfaces.

The remaining part of the gas release process can be attributed to fuel grain growth during the transient, as immediately inferred from observation (v): this process is also consistent with the denuded zones and their isotropic distribution (cf. observation (iii)): when the grain grows, its moving boundary sweeps the intragranular bubbles which are in this way deposited on the grain boundary; however, as predicted by Speight and Greenwood [15], the boundary is only able to sweep the small bubbles, so that the largest ones are left behind, e.g. on the initial grain boundary position: this explains why the bubbles on the cloud boundary are larger (cf. observation (iv)).

The main objection to the interpretation of transient gas release in terms of grain growth is that the growth rates as predicted from out-of-pile tests on stoichiometric UO2 are far too low in comparison to what should be necessary to yield the observed transient gas release rates.
4. MODELLING OF TRANSIENT GAS RELEASE.

In order to solve the contradiction between grain growth rates as deduced from out-of-pile measurements on UO₂ and rates deduced from transients on irradiated (U,Pu)O₂, a theoretical analysis was performed of the boundary motion process, with the basic assumption that boundary mobility results from intergranular pore drag, and that the latter is controlled by gas phase transport across the pore. The resulting relation is a cubic grain growth law, with a rate given by:

\[
\frac{dG^3}{dt} = C \frac{p_{\text{vap}}}{T_{\text{p}}} T^{-3/2} \]

where \(G\) is the grain size, \(p_{\text{vap}}\) is the fuel vapour pressure, \(I_p\) is the inventory of gases inside the intergranular pores, \(T\) is the absolute temperature and \(C\) is a constant. This relation interprets the grain growth inhibition in irradiated fuel by the increase of the inventory \(I_p\). At high burnup, interlinkage of grain boundary bubbles can lead to a decrease of \(I_p\), while overheating of the irradiated fuel can increase \(p_{\text{vap}}\); the overall result is that very high grain growth rates can be obtained during transients with irradiated fuel.

The calculation of the fuel vapour pressure is quite delicate, because the detailed equilibria among fuel and fission products are not yet sufficiently understood. Considering for instance cesium, out-of-pile tests have shown \([16]\) that the uranate is in equilibrium with stoichiometric UO₂. However, when the fuel is overheated, one expects that the uranate will dissociate into gaseous cesium and hyperstoichiometric UO₂. These results which are valid for UO₂ can be qualitatively transposed to (U,Pu)O₂: one expects that the vapour pressure above irradiated mixed oxide fuel shall be high in comparison with that of unirradiated (U,Pu)O₂. Fig. 1 shows the variation of vapour pressure and stoichiometry with temperature, as one can expect when heating the outer fuel portion under the assumptions of chemical equilibrium in a closed system: these curves must be considered as tentative ones, but they show nevertheless the expected high vapour pressure, which is mainly due to UO₃.

The calculated grain growths during typical temperature transients are shown in Fig. 2. One has compared the transients with heating rates of 50 K/s and 5000 K/s, respectively. The graph shows that, while grain growth may be neglected in safety related transients (5000 K/s), significant grain growth may occur during operational transients.

Once the grain growth rate is known, it is easy to calculate the deposition rate of intergranular gas on the grain boundaries, i.e. in intergranular pores. This gas comes in addition to the gas adsorbed on the boundaries, which is liberated when a temperature of about 1100°C is exceeded. The total gas release into the intergranular pores is shown in Fig. 3. Again, one can neglect grain growth related effects for the high temperature ramp rate, but the opposite is true for the slow heating rate. In both cases, the release amounts to ca. 20 % of the retained gas when the limit temperature for release of the intergranular adsorbed gas is exceeded. At higher temperatures, the slow heating rate...
leads to practically total release at 2700°C, i.e. before melting is reached. This stresses the importance of grain growth on the gas release during operational transients.

Finally, the evolution of fuel swelling can be calculated as soon as the inventory of gases on the grain boundaries and the corresponding pressure are known. The inventory of the gases results from the balance between gas deposition rate and gas percolation rate towards the fuel pin free volumes. The latter percolation rate is a complex function of the intergranular bubble network and size, of the pressure gradients and of the pressure itself. Regarding the intergranular pore size, a useful correlation has been found between the latter radius \( R \), the denuded zone width \( w \), and the current and initial grain sizes \( G \) and \( G_0 \) respectively, i.e.

\[
R = w = \frac{1}{2} (G - G_0)
\]

The resulting calculated transient swelling evolutions are shown in Fig. 4 for unrestrained conditions. Again, there is a large sensitivity to the heating rate: for the high heating rate, the maximum swelling is of the order of 12 \% only, while values close to 30 \% are obtained for the slow heating rate. The final drop in swelling results from the fact that, once most of the gas has been released, there is no more driving force for swelling, so that fuel sintering does take place and the fuel density increases.

5. **COMPARISON WITH EXPERIMENTAL DATA**

In order to compare model predictions with experimental data, the two HRAl experiments FGR-9 and FGR-10 have been selected [12]. The main interest of these two experiments is that the test FGR-10 is a repetition of the test FGR-9, but with interruption of the indirect electrical heating after about 7.5 seconds, while heating continued in FGR-9 up to about 15 seconds.

The transient gas release during these two experiments can be indirectly deduced from pressure measurements, and from the gas inventory in the test capsule after its cooldown.

The comparison between measured and calculated gas release fractions is shown on Fig. 5. The final calculated release fractions are in excellent agreement. The reason for the difference between the calculated and measured gas release kinetics is to be attributed to the underestimation of the fuel temperatures. Indeed, the temperatures were calculated assuming axisymmetric position of the sample in the test capsule, while post-irradiation examination data showed contact between the sample and the heating capsule wall.

Nevertheless, the model predicts release rates that are of the right order of magnitude, therefore providing support for the proposed gas release mechanism.
6. CONCLUSIONS.

The fuel behaviour during operational transients appears to be far more complex than during safety-related transients. Indeed, the time scales are significantly longer for operational transients, and this leaves more time for the fuel restructuring processes, mainly for grain growth.

The latter process appears to provide a satisfactory interpretation of the observed experimental facts, as far as fuel chemistry is taken into account: indeed, at high temperature, the fuel-fission product compounds dissociate, and their oxygen content is transferred to the fuel phase, which enhances the vapour transport controlled processes like grain growth.

The gas release due to grain boundary sweeping is relatively slow in comparison with the diffusionless release of the gas adsorbed on the grain boundaries which takes place as soon as a limit temperature of the order of 1100°C is exceeded.
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FIG. 1
EVOLUTION OF GRAIN SIZE DURING TEMPERATURE RAMPS ON PREIRRADIATED FUEL

△ 5000 K/S
○ 50 K/S

FIG. 2
EVOLUTION OF GAS RELEASE DURING TEMPERATURE RAMPS ON PREIRRADIATED FUEL

![Graph showing the evolution of gas release during temperature ramps. The graph plots fractional release against temperature in °C. Two curves are shown: one for 5000 K/s and another for 50 K/s.](image)

Fig. 3

Δ 5000 K/s
○ 50 K/s
EVOLUTION OF FUEL SWELLING DURING TEMPERATURE RAMPS ON PREIRRAPIDATED FUEL

FIG. 4

III/4-12
COMPARISON BETWEEN CALCULATED AND MEASURED GAS RELEASE FOR THE HEDL TESTS FGR-9 AND FGR-10

FIG. 5

III/4-13