Mathematics in Defect Trap Model of Fission Gas Behaviour in Irradiated UO_2 Fuel.

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Abstract:- In this paper we focus on analysis of the defect trap model equations which one of them explains the peculiarities within the experimental results. Then, we give some computation results proving that solution of the coupled differential equations by help of the finite element method and the Runge-Kutta method enables to predict the fission gas behaviour outside the range of the available experimental database

Key-Words: - Fission Gas, Grain, Diffusion, Release, Knock-out, Finite Element Method.

1 Introduction

Fission gas behaviour is one of the central concerns in the fuel design, performance and hypothetical accident analysis. The recent increase in the range and quantity of published experimental information on stable and transient fission gas release, both in-pile and from laboratory heating experiments, has meant that existing models of fission gas behaviour have come under close scrutiny. Most models are able to explain some of the observations, but none are yet able to explain all of the data. This has led to the adoption of empirical expressions to describe transient fission gas release. A physically based description of fission gas behaviour is desirable for several reasons. It is necessary to extrapolate outside the range of the available experimental database in hypothetical accident analysis and this can only be done with confidence with a mechanistic understanding. Similarly the transients encountered in real situations or in the study of abnormal conditions are usually more complicated than the simplified transients employed in experiments.

This that the fission gas behaviour is still a subject of current concern is evidenced by the conclusions of recent international conferences referring to the subject [1 – 4].

2 Defect Trap Model

Mathematics of the defect trap model referring to the low and intermediate temperature presented previously [5 – 8] and supplemented recently with the description of fission gas behaviour due to grain growth process [9 – 10], what increased its application to high temperature, is as follows:

$$\frac{dM}{dt} = \beta_i f + \alpha_1 f M_r + g_3 f M_{tr} - \alpha_2 M - g N_{tr} M \quad (1)$$

$$\frac{dM_{tr}}{dt} = g N_{tr} M - g_2 f M_{tr} - g_3 f M_{tr} - \lambda M_{tr} - \frac{1}{2} M_{tr} D^2 \frac{dD}{dt} N \quad (2)$$

$$\frac{dM_r}{dt} = \alpha_2 M - \alpha_1 f M_r - \lambda M_r - \frac{1}{2} M_r D^2 \frac{dD}{dt} N \quad (3)$$

$$R = g_2 f M_{tr} (S \times r) + \frac{1}{2} \pi (M_{tr} + M_r) D^2 \frac{dD}{dt} N \quad (4)$$

$$\frac{dN_{tr}^{ko}}{dt} = g_1 f - (g_2 + g_3) f N_{tr}^{ko} \quad (5)$$

for $0 \le x \le r$

$$\frac{\partial N_{trI}^{D}}{\partial t} = D_b \nabla^2 N_{trI}^{D} - (g_2 + g_3) f N_{trI}^{D} \quad (6)$$

for $0 \le x \le r$,

$$\frac{\partial N_{trII}^{D}}{\partial t} = D_b \nabla^2 N_{trII}^{D} + g_1 f - g_3 f N_{trII}^{D} \qquad (7)$$

for $r \le x < \infty$,

$$N = \frac{1 - \frac{p}{100}}{\frac{4}{3}\pi\left(\frac{D_m}{2}\right)^3} \qquad (8)$$

$$\left\langle N_{trI}^{D} \right\rangle = \frac{1}{r}\int_0^r N_{trI}^{D} dx \qquad (9)$$

$$N_{tr} = N_{tr}^{ko} + \left\langle N_{trI}^{D} \right\rangle \qquad (10)$$

$$S = S_0 + S_1\left(1 - \exp\left(-\frac{B - B_0}{\tau}\right)\right) \qquad (11)$$

$$\frac{dD}{dt} = k\left(\frac{1}{D} - \frac{1}{D_m}\right) \qquad (12)$$

With

$$k = 5.27 \times 10^7 \exp\left(-\frac{2.67 \times 10^5}{R\left(T + 371\left(1 - \exp\left(-\frac{B}{2700}\right) + 0.041 \times B\right)\right)}\right) \qquad (13)$$

$$D_m = 2.23 \times 10^3 \exp\left(-\frac{7620}{T - 520(1 - \exp(-\frac{B}{8400}))}\right) + 5\,(1 - \exp(-\frac{B}{8400}))\,) \qquad (14)$$

where

N_{tr} - concentration of bubbles in the surface layer, N_{tr}^{ko} - bubbles created in the surface layer, N_{trI}^{D} - bubbles diffused into the surface layer from the bulk, N_{trII}^{D} - bubbles in the bulk, r - fission product range λ - decay constant of isotope i, β - formation yield of the intermediate gas of isotope i, f - fission rate, t - time, x - distance into the fuel from the sample surface, r - fission product range, D_b - diffusion coefficient of bubbles, B - burn-up in MWd/tU, M - concentration of intermediate gas atoms, M_{tr} - concentration of gas atoms in the bubbles, M_r - concentration of gas atoms in the matrix, S - total surface area, D - grain size (μm), T - fuel temperature (K),
g, g_1, g_2, g_3, α_1, α_2, S_0, S_1, B_0, τ - constants.

The coupled equations of the defect trap model describe the behaviour of fission gas behaviour in full range of temperature without any artificial assumptions. Simply, the different processes in different ranges of temperature are significant or are negligible.

3 Anomalies Within the Experimental Results

Many anomalies exist within the experimental results. The main and most important assumption to explain the anomalies is that the single gas atom diffusion does not occur in the UO_2 fuel for the fission fluency higher than 10^{18} fissions/cm^3 (burn-up > 0.04 MWd/kgU).

The peculiarity that the fission gas release is caused by a combination of two basic processes: a temperature independent process and a temperature dependent process [11], is explained by the assumption, that the main contribution to the fission gas release from the UO_2 single crystal, is from the bubble traps created in the thin surface layer and from these bubbles diffused into the layer (eqs 6 and 7) from the bulk, by knock-out process (eqs. 4 and 10), during low and intermediate temperature irradiation.

This that the fission gas release is dependent on the decay constant, and the fission gas have the same proportions of isotopes for all temperatures both in the temperature dependent region and the non-temperature dependent region [11] is explained by introducing the decay constant λ into the eqs (2 –3).

The above assumptions explain also the following anomalies occurring in the low and intermediate temperature range:

- The release of fission gases in the non-temperature dependent region is oscillating function for the oscillating fission rate but the gas release oscillations are not instantaneous with fission rate oscillations

while holding the specimen at constant temperature [12],
- Fission gas release in non-temperature region during the neutron flux oscillation is greater when the flux is decreasing than when the flux is increasing (hysteresis loop) [12],
- Fission gas release rate in non-temperature dependent region is higher than the steady-state release for the same flux [12],
- Fractional fission gas release in the temperature dependent region is inversely proportional to fission rate 10^{12} – 10^{13} fissions/ $cm^3 \cdot s$, is nearly constant in the range 10^{13} – 10^{14} fissions/ $cm^3 \cdot s$. Fission rates above 10^{14} fissions/ $cm^3 \cdot s$ cause accelerated fission gas release [13],
- Fission gas release in the temperature dependent region is a periodic function for a sinusoidally changing fission rate with a more complex form [12].
- Fission gas release in the temperature dependent region is a periodic function for a sinusoidally changing temperature while specimen was maintained at a constant fission rate but the maximum release occurred earlier before the specimen reached its maximum values at the same time [12].
- Diffusion coefficients are not unique functions of temperature but also depend on irradiation parameters e.g. burnup and ratings:
- Increasing the prior irradiation exposure the diffusion coefficient is extensively reduced. The value of diffusion coefficient strongly depends on the decay constant of the isotope considered [14, 15],
- Diffusion coefficient proportional to the square of the rating within the range 2 – 9 10^{12} fissions/ $cm^3 \cdot s$. However for high fission rate, greater than 9 10^{12} fissions/ $cm^3 \cdot s$ the diffusion coefficient has been found to be proportional to the square root of fission rate between 800 1400 ^0C, directly proportional to fission rate at low temperatures and independent of fission rate at higher temperatures [16, 17, 18],
- The diffusion coefficient exhibits athermal characteristics below 800 ^0C [16, 17, 18],
- Contrary to expectations of the classical diffusion the tendency for xenon to diffuse as fast as krypton is observed [18].
- An abrupt burst of fission gas is emitted from the single crystal UO_2 when the temperature is increased [11] and from the leaking rods when power was increased [19].

The peculiarity that the burst fission gas release occurs, when the critical temperature (higher than the intermediate temperature) is crossed, is explained by grain growth process. The fission gas release kinetics from the irradiated UO_2 fuel for high temperature is determined by the kinetics of grain growth (eq. 12). The grain growth process lets also to explain the experimental anomaly that the fission gas retention in the central part of the fuel rod, where the temperature is the highest one, is higher than the retention in the intermediate radial position. [20 – 21].

The coupled equations of the defect trap model supplemented with the algorithms of fission gas behaviour due to re-crystallization of uranium dioxide grains are solved numerically using the modified Runge-Kutta method for the ordinary differential equations (1 – 4, 12) and the explicit finite-difference technique - Crank-Nicholson scheme for the partial differential equations (6 – 7).

4 Examples of Computation Results

Experimental observations show [11, 19, 20 and 21] that during transient tests, bursts release occur of two types. The main difference between this two types of fission gas release is that they refer to the range of about 0.1 – 1 % fractional release for the first type, and to the range of about 1 – 95 % fractional release for the second type.

Computation, as an example, of transient fission gas release is limited to the case when the steady state of irradiation to accumulate a desired burn-up is performed below the temperature of re-crystallization and then the subsequent step temperature increase follows.

We have considered two kinds of step temperature increase for different burn-ups: the final temperature of the step increase is still below the re-crystallization temperature, the final temperature after the step increase is above the re-crystallization temperature.

Calculations show that bursts of fission gas are predicted when the temperature is increased in both kinds. The amount of gas liberated for the final temperature above the re-crystallization temperature is much higher than for the final temperature below the re-

crystallization temperature. This is clearly seen on FIG. 1. and FIG. 2. These two figures show the theoretical krypton 87 release rate in function of time when fuel temperature is increased from 865 ^{o} C to 1240 ^{o} C at constant fission rate of 3.3 10^{12} fission/cm^3·s and fuel burn-up of 40 MWd/kgU but for two different initial grain size of 5 μm (FIG. 1.) and of 9 μm (FIG. 2.).

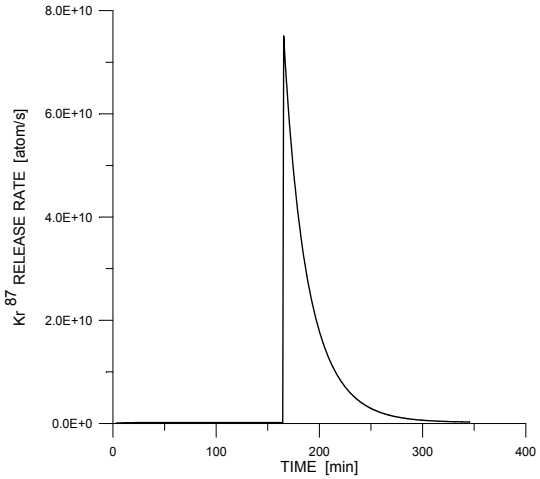

FIG. 1. Theoretical krypton release when fuel temperature is increased from 865 ^{o} C to 1240 ^{o} C at constant fission rate of 3.3 10^{12} fission/cm^3·s, initial grain size of 5 μm and burn-up of 40 MWd/kgU.

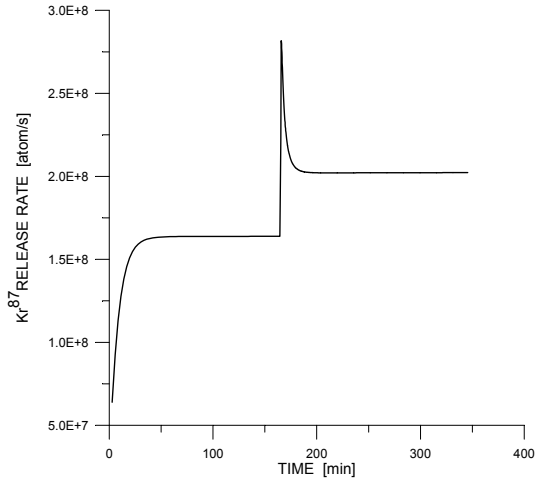

FIG. 2. Theoretical krypton release when fuel temperature is increased from 865 ^{o} C to 1240 ^{o} C at constant fission rate of 3.3 10^{12} fission/cm^3·s, initial grain size of 9 μm and burn-up of 40 MWd/kgU.

For the initial grain size of 5 μm (FIG. 1.) the re-crystallization temperature is crossed and that is why the grain growth begins and in consequence the release rate is much higher than for the initial grain size of 9 μm (FIG. 2.) where grain growth does not occur.

Duration of the two bursts are different since grain growth kinetics is responsible for the fission gas release rate (FIG. 1.) for the first one and diffusion of bubbles from the bulk to the total surface layer of the fuel is responsible for the second one (FIG. 2.)

Release rate both before and after the bursts for the stable state are equal because in this time the knock-out release process only exists. Both stable state values of the release bursts are equal. The stabilised release rate after the burst is a little bit higher than before the burst due to the step increase of temperature for both cases.

Duration of these bursts are different. It is far longer when the final temperature crosses the re-crystallization temperature. The duration of this burst is dependent on burn-up. The higher is the burn-up the shorter is the release burst.

6 Analytical Analysis

One of the mentioned above anomalies existing within the experimental results, namely the fission gas release rate in non-temperature dependent region is higher than the steady-state release for the same flux, was undertaken to explain by analytical solution of simplified equations for the non-temperature dependent region.

For low temperature (< 600 ^{o}C) and small fission rates, the terms $\alpha_1 f M_r$, $g_3 f M_{tr}$ $1/2 M_{tr}$ $D^2 dD/dt$ N and $1/2 M_r$ $D^2 dD/dt$ N can be neglected in eqs (1) – (2) since their values are small. This means that the gas already chemically immobilised and trapped in the bubbles is assumed not to appear in the intermediate state again and the process of re-crystallisation does not occur.

The analytical solution of the simplified equation for the fission rate changing sinusoidally with time, $f(t)=f_o+f_1 \sin \omega t$, supports the peculiar experimental observation. To start up the problem the following reduced set of equation, are to be solved:

$$\frac{dM}{dt} = \beta_i f - \alpha_2 M - g N_r M \qquad (15)$$

$$\frac{dM_{tr}}{dt} = gN_{tr}M - g_2 fM_{tr} - \lambda M_{tr} \qquad (16)$$

The equation set for the dynamic case becomes a set of time dependent non linear equations. Solution of this problem is based on the perturbation theory.
The eq. (15) for the case of oscillating fission rate is still linear and can be solved straightforwardly.

Inserting the solution of differential equation (15) into eq. (16) and linearizing it, one can obtain a first approximation to M_{tr}^{o} by solving the following equation:

$$\frac{dM_{tr}^{o}}{dt} + (g_2 f_o + \lambda)M_{tr}^{o} = gN_{tr}M \qquad (17)$$

The second approximation M_{tr}^{1} is given by a solution of the following linear differential equation:

$$\frac{dM_{tr}^{1}}{dt} + (g_2 f_0 + \lambda)M_{tr}^{1} = gN_{tr}M - g_2 f_1 \sin \omega t\, M_{tr}^{1} \qquad (18)$$

The term $f_1 \sin \omega t$ is small in comparison with f_o and is the perturbation. For the asymptotic state the solution of eq. (19) becomes:

$$M_{tr}^{1} = \frac{\beta_i f_o g N_{tr}}{(g_2 f_o + \lambda)(\alpha_2 + gN_{tr})} + \frac{g_2 f_1 C}{2(g_2 f_o + \lambda)} + K \sin \omega t + L \cos \omega t + P \sin 2\omega t + W \cos 2\omega t \qquad (19)$$

where C,K,L,P,W are coefficients depending on f_o, f_1 and ω.

The solutions of the linearized eqs. (17) and (18) were convened over by using the variation of constants method.

Eq. (19) shows the concentration of gas atoms trapped in the bubbles is a periodic function for sinusoidally changing fission rate.

The mean value of the asymptotic eq. (19) is:

$$M_{tr}^{1} = \frac{\beta_i f_o g N_{tr}}{(g_2 f_o + \lambda)(\alpha_2 + gN_{tr})} + \frac{g_2 f_1 C}{2(g_2 f_o + \lambda)} \qquad (20)$$

The first term on the right hand side of eq. (20) is the solution of eqs (15) and (16) for steady state for the step function of fission rate $f=f_o$. Hence, it may be concluded, the mean value of the fission gas release rate for the asymptotic state during oscillation of the fission rate will be greater than the steady state release rate for the step function of fission rate equal to the mean value of the oscillating fission rate

7 Conclusion

The algorithms of fission gas behaviour in UO_2 fuel described above explain the peculiar experimental results and let predict the transient fission gas behaviour outside the range of the available experimental database.

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