TRANSIENT FISSION GAS RELEASE FROM UO_2 FUEL FOR HIGH TEMPERATURE AND HIGH BURNUP

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Abstract

In the present paper it is assumed that the fission gas release kinetics from an irradiated UO_2 fuel for high temperature is determined by the kinetics of grain growth. A well founded assumption that Vitanza curve describes the change of uranium dioxide re-crystallization temperature and the experimental results referring to the limiting grain size presented in the literature are used to modify the grain growth model. Algorithms of fission gas release due to re-crystallization of uranium dioxide grains are worked out. The defect trap model of fission gas behaviour described in the earlier papers is supplemented with the algorithms. Calculations of fission gas release in function of time, temperature, burn-up and initial grain sizes are obtained. Computation of transient fission gas release in the paper is limited to the case where steady state of irradiation to accumulate a desired burn-up is performed below the temperature of re-crystallization then the subsequent step temperature increase follows. There are considered two kinds of step temperature increase for different burn-up: the final temperature of the step increase is below and above the re-crystallization temperature. Calculations show that bursts of fission gas are predicted in both kinds. The release rate of gas liberated for the final temperature above the re-crystallization temperature is much higher than for final temperature below the re-crystallization temperature. The time required for the burst to subside is longer due to grain growth than due to diffusion of bubbles and knock-out release. The theoretical results explain qualitatively the experimental data but some of them need to be verified since this sort of experimental data are not found in the available literature.

1. INTRODUCTION

It is noticed that the radial distribution of concentration of retained fission gas in the fuel rod is in very close correlation with the radial grain size distribution. It is roughly the mirror reflection. It implies that the grain growth mechanism mainly determines the fission gas release for high temperature.

On the other hand it is noticed that the Vitanza curve is the threshold temperature change of uranium dioxide re-crystallization temperature with burn-up. This is well founded assumption.

Out-of-pile experiments show that during annealing the irradiated UO_2 samples bursts of fission gas release occur [1]. After a small burst release at relatively low temperature, a large burst release appears at high temperature. The critical temperature for high temperature burst release is about 1800°C for low burnup (about 7 MW•d/kg U) and decreases to about 1500°C for high burnup (30 MW•d/kg U).

The point defects induced by radiation begin to recover at 450–650°C and are completely almost recovered above 850°C, while defect clusters of dislocations and small intragrannular bubbles require 1150–1450°C [2].

Thermal recovery of radiation defects and microstructure change in irradiated UO_2 fuel studied by X-ray diffraction and transmission electron microscopy lead to the conclusion that the gas release kinetics from irradiated UO_2 is determined by the kinetics of thermal recovery of the radiation induced defects.

If the point defects, defect clusters of dislocation and small intragranular bubbles are thermally recovered at the temperatures below 1450°C, a natural question concerns the nature of forces which immobilise the noble gases. Hence an additional trapping process of inert gas atoms with the uranium dioxide material is suspected to occur.

The process of strong binding of the fission gas fragments with the irradiation defects is described in the literature as a process of chemical interaction with UO_2 [3]. It is assumed further that the vicinity of the fission fragment trajectory is the place of intensive irradiation induced chemical interaction of the fission gas products with UO_2 [3].

We can further assume that above a limiting value of fission fluency a more intensive process of irradiation induced chemical interaction occurs. Significant part of fission gas products is thus expected to be chemically bound in the matrix of UO_2.

Furthermore, it is expected that the gas can be released only in the process of re-crystallization. The higher burnup the higher amount of gas should be released and the lower re-crystallization temperature should be observed. Out-of-pile experiments [1] support this assumption since the critical temperature of fission gas burst during annealing decreases with burnup.

This that the critical temperature decreases with burn-up suggest that the re-crystallization temperature of UO_2 is changed by the process of chemical interaction. It is clear that during irradiation the grain growth should be observed above the re-crystallization temperature and grain subdivision below the temperature should be observed when saturation of fission damage is obtained. This means further that the re-crystallised region will be adjacent to the subdivided grains region and the appearance of interface between the two regions will be determined by the re-crystallization temperature. It can also be expected that in the re-crystallised grains the defects are swept out. This seems also to be natural that the chemically bound fission gas atoms replacing the uranium atoms in the crystallographic lattice will increase the fission gas product release. So the process of grain growth is the process of purging the contaminated lattice.

This seems also to be natural that the chemically bound fission gas atoms substituting for example a uranium atom in the crystallographic lattice can form weak facets. At certain saturation conditions subdivision of the grains can occur and the increase in fission gas products release may be expected. So it can be stated either re-crystallization or subdivision have to occur in the saturation circumstances.

The fact that the process of grain subdivision for high burnup (70–80 MW•d/kg U) forms an extremely fine structure to a temperature as high as 1100 ° C and that the decrease in fission gas concentration in the fuel [4] supports this concept. Also the re-crystallised grain region is found to be adjacent to the subdivided grain region and in the re-crystallised grain region no defects or bubbles are observed [5]. This means that we can treat the re-crystallised volume of uranium dioxide as a fresh fuel where all the processes connected with irradiation start from the beginning. So the process of grain growth is the process of purging the contaminated lattice.

The decrease in critical temperature to about 1100°C for over 1% fractional fission gas release from the fuel and for high burn-up, reported by Vitanza et al. [6] well correlates with the experiments [1] and [4] mentioned above. This that the limiting grain size begins to increase practically at the temperature about 1000–1100°C for high burn-up (40 MW•d/kg U) [7]

supports the concept that the threshold temperature for UO_2 for high burn-up is about 1000°C where above the threshold temperature the re-crystallization takes place and below the threshold temperature the grain subdivision can occur under the condition of irradiation damage saturation.

This also gives evidence for the concept of chemical interaction of the fission gas atoms with the atoms of the fuel. It is assumed in this paper that all retained gas atoms in the lattice are released from the volume of re-crystallised grains.

2. UO_2 GRAIN GROWTH KINETICS

The above assumptions lead to the conclusion that the gas release kinetics from irradiated UO_2 fuel for high temperature is determined by the kinetics of grain growth. It is well known [8] that the irradiation damages introduced by fission events have two opposing effects on grain growth. The large concentration of fission gas atoms in the lattice introduced in fission spikes enhances the transfer of atoms across a boundary, increasing the rate of growth. It means that the higher is the burn-up the quicker is the process of grain growth–the stable state is quicker obtained. Conversely the impurities introduced by fission events inhibit grain growth by limiting the grain size.

Assuming that the Vitanza curve [6] describes the change of uranium dioxide re-crystallization temperature we can say that the grain growth rate depends on the burn-up in the way given by the best fit of the grain size change with the curve [9]. To obtain this we have modified the grain growth model of Ainscough et al. [10], which is generally considered to be the best available one in the open literature. These assumptions enable us to evaluate the change of grain growth in function of fuel burn-up. By help of this assumption we have been able to modify the coefficient $\underline{k}$ which is responsible for the grain growth rate.

According to the experimental data presented by Bagger et al. [7] much smaller values for the limiting grain size must be assumed at higher burn-up than in the Ainscough model. Fig. 1 presents the comparison of limiting grain size in function of temperature for the unirradiated fuel and the irradiated fuel [40 MW•d/kg U). The experimental results show that while decreasing the temperature the limiting grain size for burn-up of 40 MW•d/kg U decreases asymptotically to 5 μm and practically reaches the value at the temperature about 1000 °C.

Extrapolating the limiting grain size curve of irradiated fuel for low temperature we can see that the limiting grain size is not smaller than 5 μm, while for the unirradiated fuel the limiting grain size tends to zero. So, the limiting grain size in function of temperature for burn-up range 0–40 MW•d/kg U is to lie between these two curves. Taking into account these assumptions we finally obtained the modified differential equation of Ainscough of grain growth which describes the grain growth kinetics.

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

Where:

$$k = 5.27\times10^{7}\exp\left(-\frac{2.67\times10^{5}}{R\left(T+371\left(1-\exp\left(-\frac{B}{2700}\right)+0.041\times B\right)\right)}\right) \quad (2)$$

$$D_m = 2.23\times10^{3}\exp\left(-\frac{7620}{T-520(1-\exp(-\frac{B}{8400}))}\right) + 5\,(1-\exp(-\frac{B}{8400})) \quad (3)$$

where

D = grain size (μm),

D_m = limiting grain size (μm),

B = burn-up in MWd/tU

T = fuel temperature (K),
t = time (h).

The equation (3) can be extended for higher burn-up than 40 MW•d/kg U since correlating the limiting grain size with the Vitanza curve we can see that for burn-up equal 40 MW•d/kg U the stable state is reached.

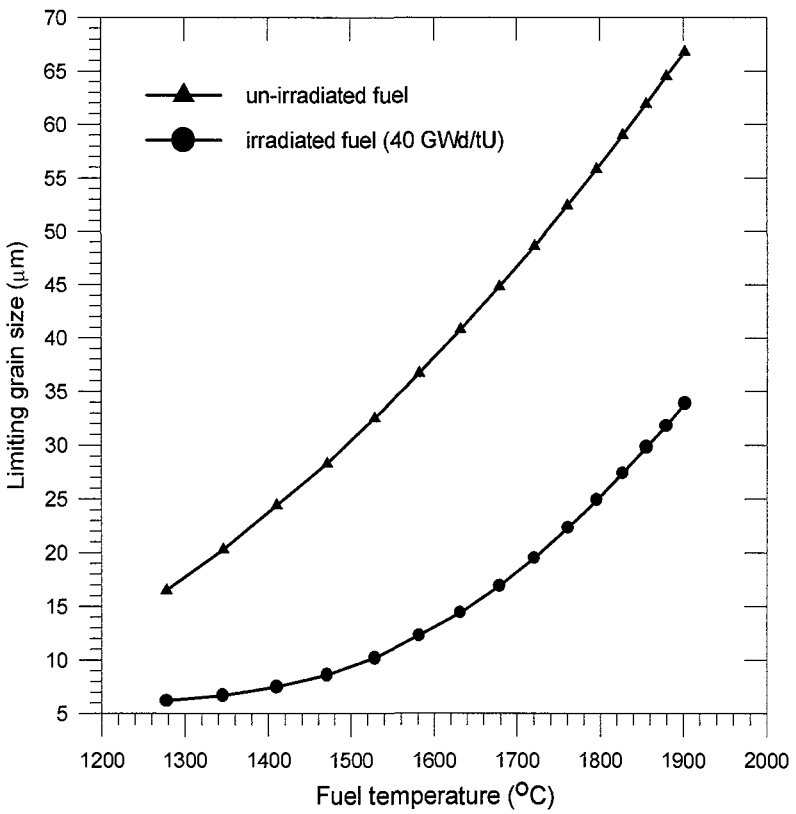

FIG. 1. Limiting grain size in function of fuel temperature for the un-irradiated and the irradiated fuel [7].

3. ALGORITHM OF FISSION GAS RELEASE DUE TO UO_2 GRAIN GROWTH

It is well known that the grain growth in polycrystalline materials is caused by a preferential shrinkage of smaller grains due to their relatively smaller radii of curvature. An average number of grains, (N_0), in a unit volume is of un-irradiated fuel:

$$N_o = \frac{1-\frac{p}{100}}{\frac{4}{3}\pi\left(\frac{D_o}{2}\right)^3} \tag{4}$$

where p –porosity in%, D_0–initial grain diameter.

At the elevated temperature the number of grains, N, in the unit volume is fixed by the limiting grain size, $D_{m.}$:.

$$N = \frac{1-\frac{p}{100}}{\frac{4}{3}\pi\left(\frac{D_m}{2}\right)^3} \tag{5}$$

The initial grain size, D_0, is easily measurable while the limiting grain size after the grain growth, $D_{m.}$, is determined by equation (3) at which grain growth ceases.

The release rate of re-soluted gas (chemically bound fission gas in the UO_2 lattice) and trapped in bubbles is to be determined. . Multiplying the rate of volume change of grain by the concentration of re-soluted gas atoms in the matrix, M_r, and trapped gas atoms in the bubbles, M_{tr}, we obtain the release from one grain, R_{go}:

$$R_{go} = \frac{1}{2}\pi(M_r + M_{tr})D^2\frac{dD}{dt} \tag{6}$$

where dD/dt is determined from the Ainscough's modified differential equation (1).

The product of release rate from one grain, R_{go}, and the number of grains, N, at elevated temperature, defined by equation (5) determines the release rate from a unit volume.

The defect trap model presented previously [9, 11, 12, 13 and 14] can be supplemented with the description due to grain growth process, according to the aforementioned assumptions:

$$\frac{dM}{dt} = \beta_i f + \alpha_1 f M_r + g_3 f M_{tr} - \alpha_2 M - g N_{tr} M \tag{7}$$

$$\frac{dM_{tr}}{dt} = g N_{tr} M - g_2 f M_{tr} - g_3 f M_{tr} - \lambda M_{tr} - \frac{1}{2} M_{tr} D^2 \frac{dD}{dt} N \tag{8}$$

$$\frac{dM_r}{dt} = \alpha_2 M - \alpha_1 f M_r - \lambda M_r - \frac{1}{2} M_r D^2 \frac{dD}{dt} N \tag{9}$$

$$R = g_2 f M_{tr}(S \times r) + \frac{1}{2}\pi(M_{tr} + M_r)D^2 \frac{dD}{dt} N \tag{10}$$

Where

$$\left\langle N_{trI}^{D} \right\rangle = \frac{1}{r}\int_0^r N_{trI}^{D} dx \tag{11}$$

$$N_{tr} = N_{tr}^{ko} + \left\langle N_{trI}^{D} \right\rangle \tag{12}$$

$$S = S_0 + S_1\left(1 - \exp\left(-\frac{Bu - B_0}{\tau}\right)\right) \tag{13}$$

$$\frac{dN_{tr}^{ko}}{dt} = g_1 f - (g_2 + g_3) f N_{tr}^{ko} \quad \text{for} \quad 0 \le x \le r \tag{14}$$

$$\frac{\partial N_{trI}^{D}}{\partial t} = D_b \nabla^2 N_{trI}^{D} - (g_2 + g_3) f N_{trI}^{D} \quad \text{for} \quad 0 \le x \le r, \tag{15}$$

$$\frac{\partial N_{trII}^{D}}{\partial t} = D_b \nabla^2 N_{trII}^{D} + g_1 f - g_3 f N_{trII}^{D} \quad \text{for} \quad r \le x < \infty, \tag{16}$$

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_i – 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,

Bu – burnup,

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,

g, g_1, g_2, g_3, α_1, α_2, S_0, S_1, B_0, τ –constants.

It is assumed that the total surface area versus burn-up described by Eq. 13 does not change during the process of grain growth. The coupled Eqs. (7- 16) are solved numerically using the modified Runge-Kutta method and the explicit finite-difference technique; Crank-Nicholson scheme.

4. BURSTS OF FISSION GAS RELEASE

Knowledge of fission gas release mechanisms during transients at high burn-up is very important from the exploitation point of view. The experimental programmes on the subject are mainly concerned with the hot cell examination after irradiation, without information on the detailed kinetics of the phenomena.

In order to contribute to filling this gap, the analysis of fission gas release for temperature transient (which occurs during power transient) is performed and the theoretical results are compared with the few experimental results available in the literature.

Experimental observations show [7, 15, 16 and 17] 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.

It is considered that the first type of fission gas release is proportional to the total fuel surface area, but the second type depends on the volume of the sample. Below the re-crystallization temperature, the main contribution to the fission gas release from the UO_2 fuel is from the bubble traps by knock-out process and diffusion of bubbles [11–14]. The knock-out process affects the UO_2 total fuel surface layer to a depth not more than 10 μm–the fission fragment range. The bubble migration is due to the following sequence "kinetic excitation of gas atoms, intermediate gas formation and bubble formation at their new location". Since the same knock-out release mechanism applies to the bubbles created in the thin surface layer and to the bubbles diffused into the layer from the bulk, the combination of temperature independent release process and temperature dependent release process is explained. So, it is sometimes doubtful whether gas release measurements reflect the volume characteristics of the solid or the surface characteristics.

Above the re-crystallization temperature, the main contribution to the fission gas release is due to purging the contaminated lattice from fission gas atoms by the re-crystallization. At the beginning of the re-crystallization process, the surface of the grains is also affected which takes part in forming the total surface area.

5. COMPUTATION RESULTS

Computation 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. Two kinds of step temperature increase for different burn-ups are considered:

— 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. 2. and FIG. 3. These two figures show the theoretical krypton 87 release rate in function of time when fuel temperature is increased from 865°C to 1240°C at constant fission rate of 3.3 10^{12} fission/cm^3·s and fuel burn-up of 40 MW•d/kg U but for two different initial grain size of 5 μm (FIG. 2.) and of 9 μm (FIG. 3.). For the initial grain size of 5 μm (FIG. 2.) 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. 3.) 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. 2.) 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. 3.).

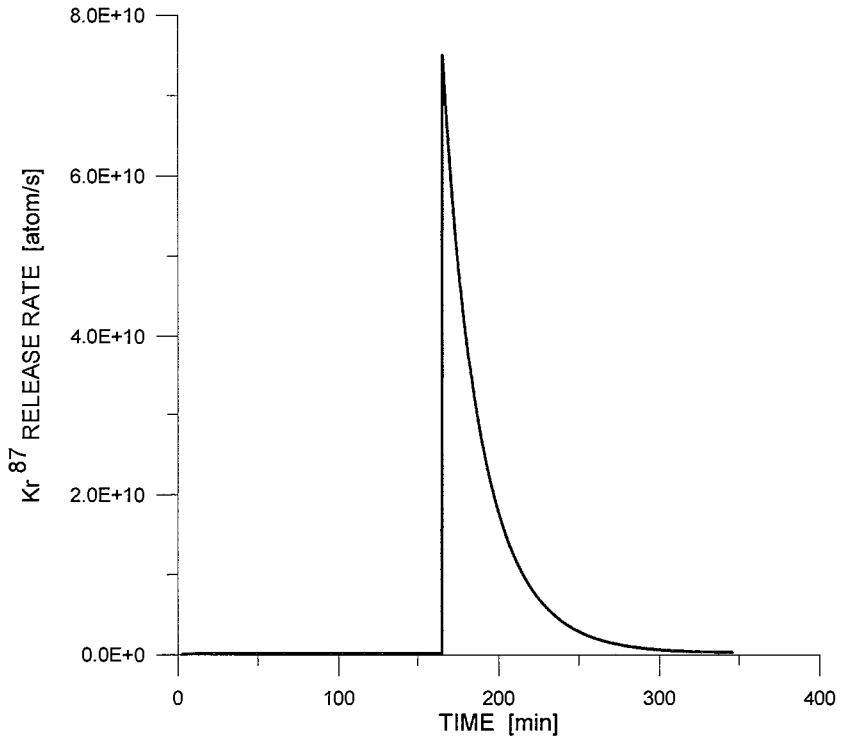

FIG. 2. Theoretical krypton release when fuel temperature is increased from 865 ° C to 1240 ° C at constant fission rate of 3.3 10^{12} fission/cm^3·s, initial grain size of 5 μm and burn-up of 40 MW•d/kg U.

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. This is seen in FIG. 4 which presents theoretical krypton burst half decrease duration in function of burn-up at constant fission rate of 3.3 10^{12} fission/cm^3·s, initial grain size of 5 μm and 6 μm when fuel temperature is increased from 865°C to 1240°C. The smaller is the initial grain size the shorter is the duration of release burst.

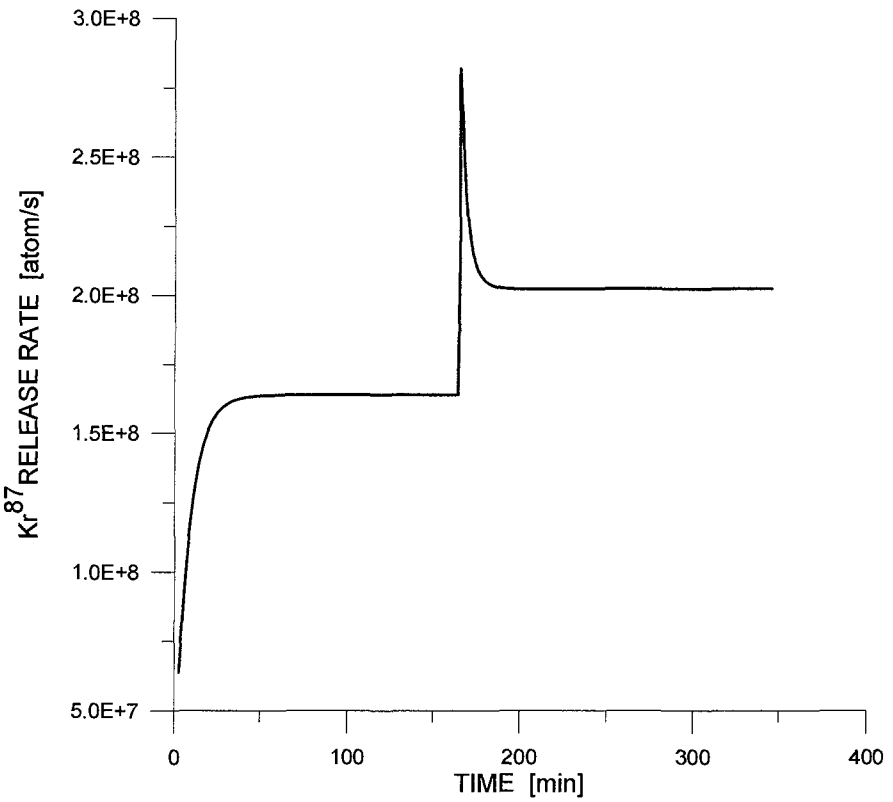

FIG. 3. Theoretical krypton release when fuel temperature is increased from 865 ° C to 1240 ° C at constant fission rate of 3.3 10^{12} fission/cm^3 s, initial grain size of 9 μm and burn-up of 40 MW•d/kg U.

Maximal release rate of the burst generally increases with burn-up except the initial grain size is very close the limiting grain size. In FIG. 5. The curve of maximal release rate for the initial grain of 6 μm starts to decrease when the burn-up reaches the value of 30 MW•d/kg U. For this burn-up the limiting grain size is equal 6.59 μm at the temperature of 1240 ° C. For burn-up of 35.3 MW•d/kg U the limiting grain size becomes about 6.01 μm what is very close the initial grain size and crossing the burn-up of 36 MW•d/kg U the limiting grain size becomes lower than the initial grain size. This means that the grain growth vanishes and the gas release vanishes as well.

The experimental results presented in ref. [15] indicate that an abrupt burst of fission gas was emitted when the single crystal UO_2-specimen temperature was increased. FIG. 6. presents the theoretical krypton release for the same temperature conditions and fission rate as in the experiment carried by R.M. Carroll et al. (FIG. 8. of Ref. [15]). The defect trap model of fission gas release permits a qualitative interpretation of the results. The times required for the burst to increase and subside are of the same about value as in the experiment. The values of release rate before the burst and after the burst are of the same order except the value during the burst. The burst in the experiment is bigger than the burst obtained theoretically. The total surface area in the theoretical calculations for the unit volume of 1 cm^3 is about 135 times bigger than in the experiment of R.M. Carroll et al. [15] than in the experiment. Taking this into consideration we can state that the theoretical krypton isotopes release rate is of the same order as in the experiment for the stable state.

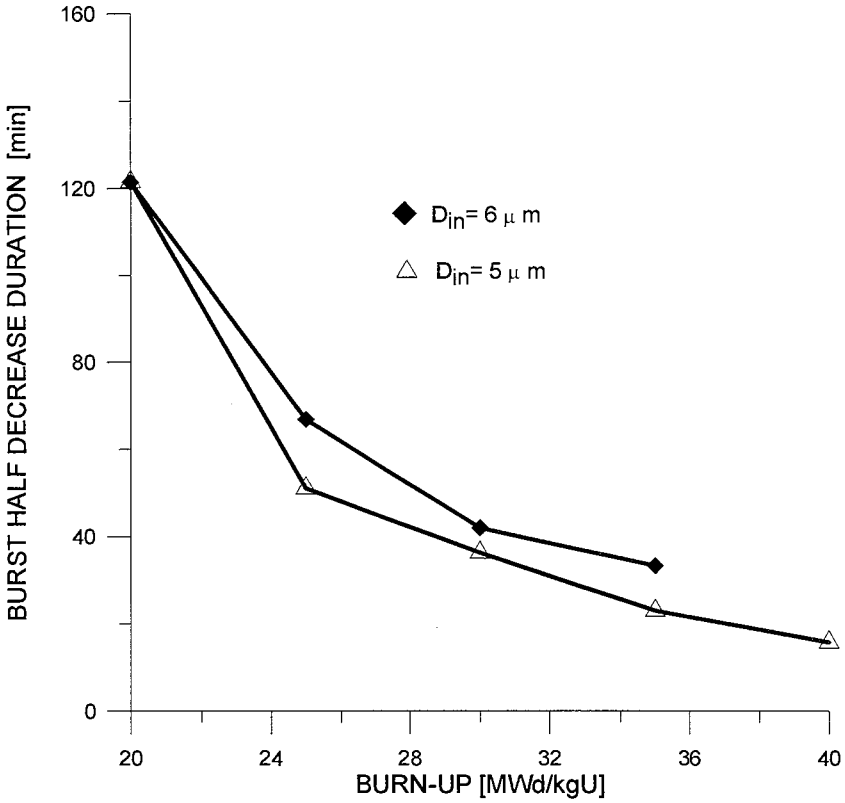

FIG. 4. Theoretical Kr^{87} burst half decrease duration in function of burn-up at constant fission rate of 3.3 10^{12} fission/cm^3·s, initial grain size of 5 μm and 6 μm when fuel temperature is increased from 865 ° C to 1240 ° C

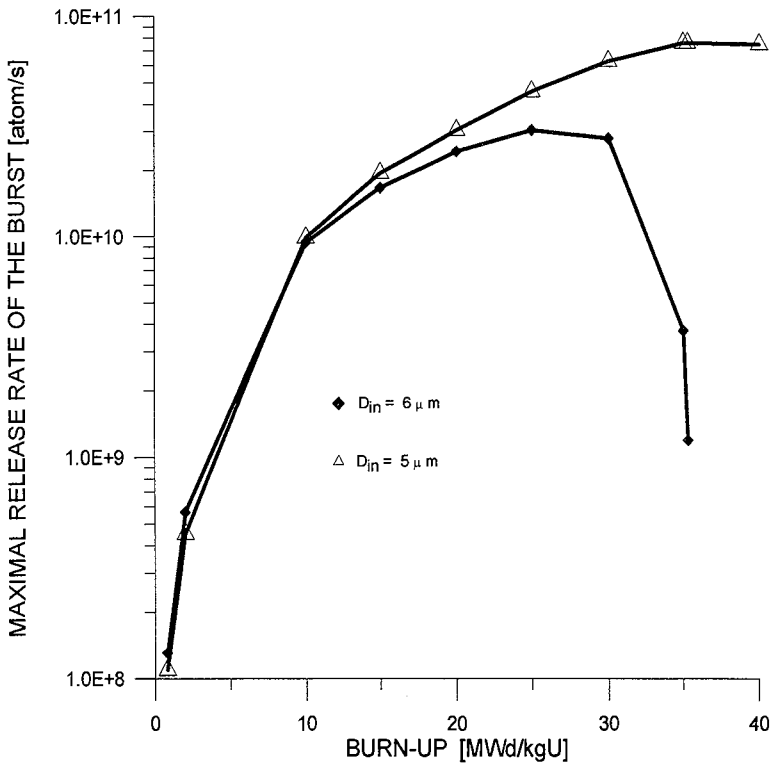

FIG. 5. Theoretical maximal Kr^{87} release rate of the burst in function of burn-up at constant fission rate of 3.3 10^{12} fission/cm^3·s, initial grain size of 5 μm and 6 μm when fuel temperature is increased from 865 ° C to 1240 ° C

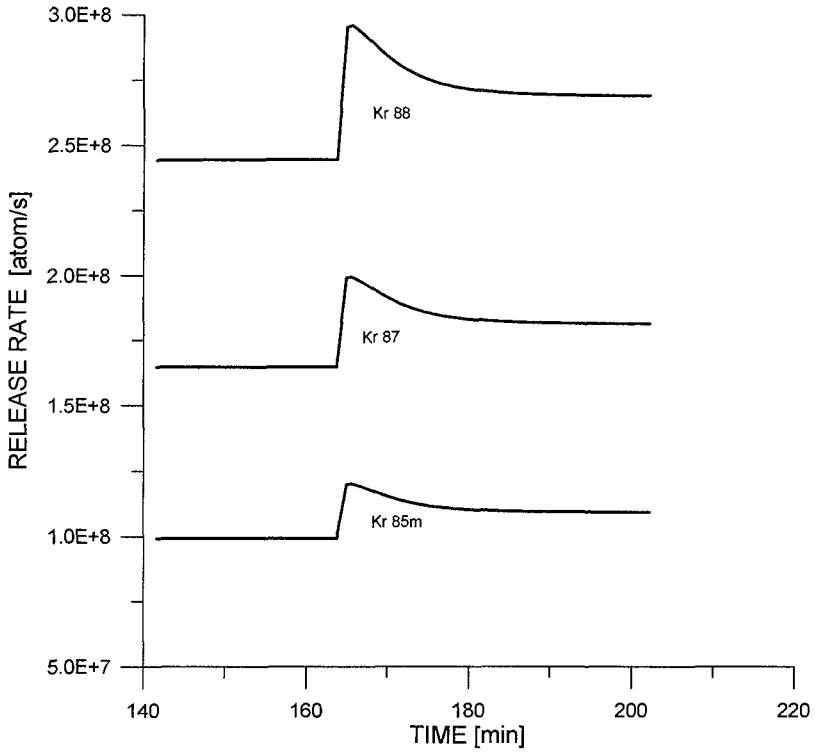

FIG. 6. Theoretical krypton release when fuel temperature is increased from 865 ° C to 1040 ° C at constant fission rate of 3.3 10^{12} fission/cm^3·s, initial grain size of 9 μm and burn-up of 35 MW•d/kg U.

Also the experimental results presented in ref. [16] show that an abrupt burst of fission gas was emitted when the UO_2 fuel temperature was increased. So both from single crystal and UO_2 fuel an abrupt burst of fission gas is emitted what means that the same process is responsible for this. Some of the theoretical results explain qualitatively the experimental data but some of them need to be verified since this sort of experimental data are not found in the available literature.

6. CONCLUSIONS

Two opposing effects of enhancement and inhibition of irradiation damage introduced by fission effect on grain growth have a significant impact on fission gas release for high temperature. The large concentration of point defects in the lattice introduced in fission spikes enhances the transfer of atoms across a boundary, increasing the rate of growth. Conversely the impurities introduced by fission events inhibit grain growth by limiting the grain size.

There is no doubt that during the process of grain growth the fission gas products retained in the matrix of uranium dioxide being there immobilised are released. It is natural that the process of grain growth is the process of purging the contaminated lattice. So we can treat the re-crystallised volume of uranium dioxide as a fresh fuel where the processes connected with the radiation start from the beginning.

Supplementation of the defect trap model of fission gas release by the grain growth process let to form complementary model for low, intermediate and high temperatures.

It can be concluded that the additional assumptions made in this model make it possible to give the experimental results described in the literature, regarding fission gas release from UO_2 fuel during high temperature irradiation a satisfactory qualitative interpretation.

The higher is the initial grain size, the higher is the threshold temperature of re-crystallization of the grain. It means, if the limiting grain size for the appointed temperature is equal to the initial grain size, then the re-crystallization does not occur. In consequence, the fission gas release due to re-crystallization also does not occur and the retained gas concentration remains on the level of accumulation.

Calculations show that bursts of fission gas are predicted for the same step function of temperature both when the initial grain size is below and above the limiting grain size. The release rate of gas liberated for the final temperature above the re-crystallization temperature is much higher than for final temperature below the re-crystallization temperature. The time required for the burst to subside is longer due to grain growth than due to diffusion of bubbles and knock-out release.

The model gives theoretical results which need to be verified since this sort of experimental data are not found in the available literature.

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