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Hypothetical helium migration in uranium dioxide fuel during neutron irradiation in terms of defect trap model.

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Motto

Since after more than 60 years of research fission gas behaviour is still a mater of controversial discussion, so the following Olander's statement in 1976 [a] in his book is still valid:

"Among the myriad phenomena that occur simultaneously in a nuclear fuel element under irradiation, none has so frustrated the designer, so challenged the experimentalist, or so intrigued the theorist as the behaviour of the fission products xenon and krypton".

D. R. Olander

I would like to the products xenon and krypton to add yet the noble gas helium.

[a] D.R. Olander, Fundamental aspect of nuclear fuel elements, *TID*-26711-P1 (1976).

Outline

- 1. Introduction.
- 2. Mathematics of helium release in terms of the Defect Trap Model .
- 3. Analytical analysis of the simplified differential equations of the proposed model of helium release from the $(U, Am)O_2$ specimen under irradiation in function of low and intermediate temperature.
- 3a. Prediction of helium atoms release from $(U, Am)O_2$ specimen under irradiation in function of low temperature.
- 3b. Prediction of helium atoms release from $(U, Am)O_2$ specimen under irradiation in function of low and intermediate temperature.
- 4. Conclusions

Outline

- 5. Appendix - **Anomalies of fission gas behaviour and their defect trap model predictions**

1. Introduction.

- The behavior of helium in uranium dioxide has been extensively investigated since the middle of the 1960s [1], due to its potential effects on the nuclear fuel thermal and mechanical properties as well as on the spent fuel behavior during long term storage and final disposal.
- Minor actinides (MA: Np, Am, Cm) generated in nuclear fuel during irradiation in reactor significantly contribute in the radio-toxicity heat generation of spent nuclear fuel and production of helium.
- In the minor-actinide-bearing blanket (MABB) concept based on (U, MA)O_{2-x} pellets incorporating a large amount of MA (up to 20 wt%) which are going to be irradiated for a long time on the periphery of the outer core [2], the large production of helium mainly resulting from the MA transmutation chains is expected.
- The impact of this helium on gas releases and blanket swelling must be identified

1. Introduction – cont.

- It is known also that large amount of noble gases are retained in the high burn-up structure (HBS) located in the fuel surface. Release of both the helium atoms and the fission gas of xenon atoms behave alike during annealing process (see Fig. 1) [3, 4, 5]. This let us to infer that transport (migration) and release of helium from the fuel under irradiation is also alike the fission gas products - the same mechanisms control these.

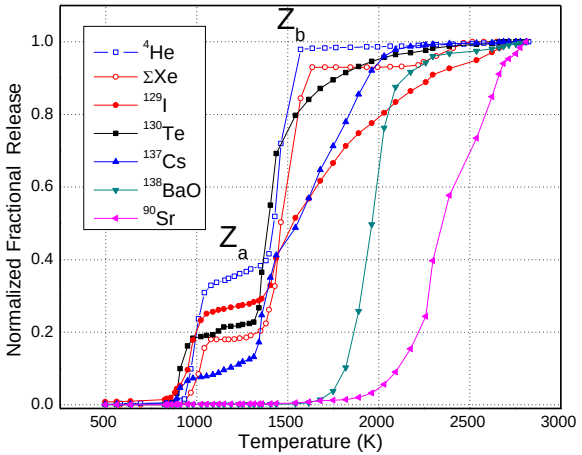

Fig. 1. Normalized fractional release of helium and fission products from highly irradiated UO_2 fuel [3, 4, 5].

1. Introduction – cont.

- It is believed that the helium atoms in the crystal structure of UO_2 occupy the most energetically favorable octahedral sites . If the potential barriers separating the given atom from the neighboring octahedral sites are sufficiently large, the atom can be effectively immobilized on millions of years.
- In our work [6] the helium atom immobilization in a deep potential well inside the crystallographic lattice was proposed. Applying the “Ab initio” calculations using the Wien2k program package [7] we estimated the static energy barrier between interstitial sites in perfect lattice UO_2+He. on about 4.15 eV.

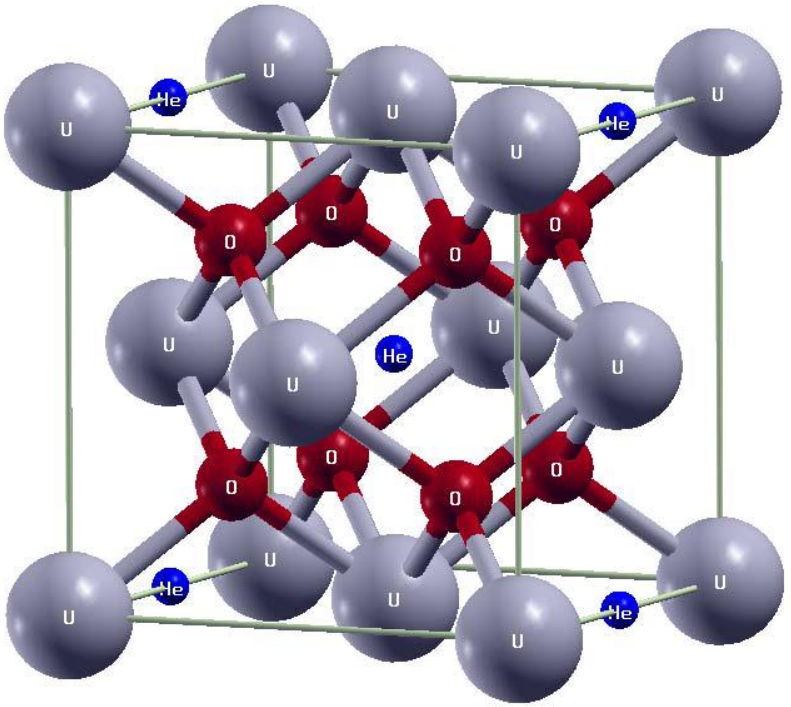

Fig. 2. The He atoms incorporation into the octahedral sites of the UO_2 lattice.

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1. Introduction – cont.

- This is clearly seen from the Fig.1 and Fig. 3 connecting the release of helium and xenon atoms during annealing at temperature about 1100 ^{o}C in Fig. 1 and the fuel structure at the annealing temperatures 920 K (647 ^{o}C) and 1500 K (1227 ^{o}C) in Fig. 3. The structure at the annealing temperatures 920 K (647 ^{o}C) has not changed in comparison with the received fuel structure, but the structure at the annealing temperature 1500 K (1227 ^{o}C) has changed showing that the grains increased what means that the re-crystallization process occurred (see Fig. 3) and the release occurred at the annealing temperature about 1373 K (1100 ^{o}C).

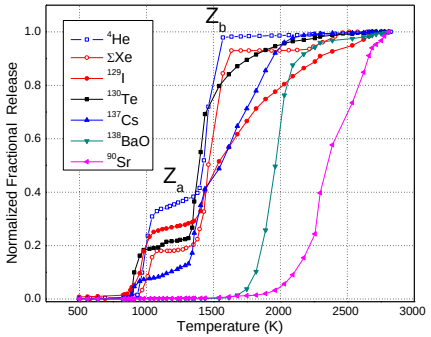

Fig. 1. Normalized fractional release of helium and fission products from highly irradiated UO_2 fuel.

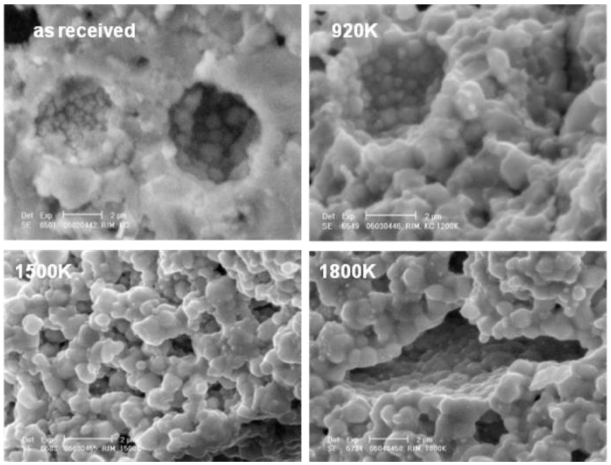

Fig. 3. High burn-up structure as received and after annealing at temperatures 920, 1500 and 1800 K.[3, 4, 5].

1. Introduction – cont.

- The critical temperature for the onset of the high temperature fission gas burst release is about 1100 ^{o}C for high burn-up fuel (above 50 MWd/kgU). This temperature (1100 ^{o}C) applies also to very high burn-up (over 100 MWd/kgU) since the stable state is reached already at the burn-up equal to 50 MWd/kgU (see Fig. 4 [8]). The curve of critical temperature for the onset of fission gas burst release in fact reflects the curve of grain re-crystallization temperature versus burn-up. The process of re-crystallization is the process of purging the contaminated lattice by the noble gases what means that the retained gas atoms in the fuel are released.

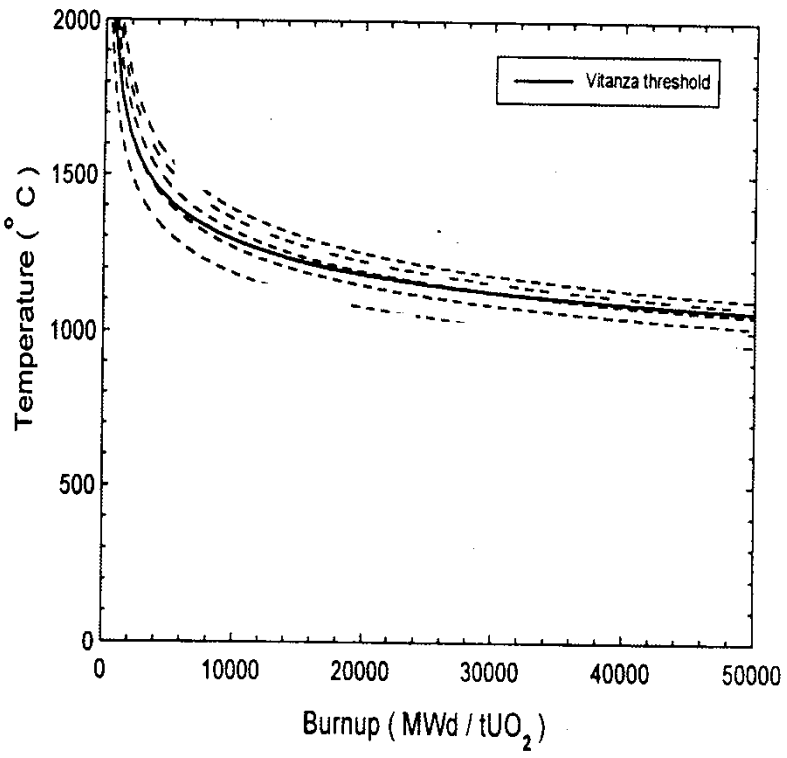

Fig. 4. Burn-up dependence of critical temperature for onset of burst release [8]. Incubation time (burn-up) as a function of the centre temperature [8].

1. Introduction – cont.

- Helium atoms resulting from the radioactive decay as α particles have the energy thousands of times greater than the ~ 4 electron volts barrier and can easily move in the crystal occupying free octahedral interstitial positions.
- The old mineral materials containing fissile isotopes hold fairly a lot of noble gases. These gases are not released during hundreds of millions of years from the old materials, e.g., the mineral thorianite at least 500 000 000 years old occurring in Ceylon contains as much as 10 cm^3 He/g, which is equivalent to the fission gas concentration at a burn-up of 300 000 MWd/t [9].

1. Introduction – cont.

- The grain subdivision can be observed in the Fig. 3 in the received sample picture. The sample shows the development of a sub-grain nanostructure with a typical grain size of 150 nm to 300 nm presented also in the earlier paper [10]. The sub-grain formation is often particularly clearly visible on the inner surfaces of pores or large fission gas bubbles [10].
- Associating the fact that xenon atoms are chemically bond with the fuel and the helium atoms can be immobilized in the uranium dioxide fuel because the octahedral interstitial positions in uranium dioxide are effective traps for helium atoms with the process of grain re-crystallization during which the xenon and helium atoms behave identically in terms of release we can infer that the hypothetical modeling of helium migration and release during irradiation is described by the defect trap model of fission gas behavior published earlier[11- 16].
- Given the experimental and calculation data above we propose the hypothetical modeling of helium migration and release during irradiation described by the defect trap model of fission gas behavior with certain modifications.

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2. Mathematics of helium release in terms of the Defect Trap Model .

- Given the experimental and calculation data above we propose the hypothetical modeling of helium migration and release during irradiation described by the defect trap model of fission gas behavior with certain modifications.

$$\frac{dM}{dt} = \lambda Am + \alpha_1 fM_r + g_3 fM_{tr} - \alpha_2 M - gN_{tr}M \qquad (1)$$

$$\frac{dM_{tr}}{dt} = gN_{tr}M - g_2 fM_{tr} - g_3 fM_{tr} - \frac{1}{2}M_{tr}D^2\frac{dD}{dt}N \qquad (2)$$

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

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

Mathematics of helium release in terms of the Defect Trap Model - cont.

$$\frac{dN_{tr}^{ko}}{dt} = g_1 f - (g_2 + g_3) f N_{tr}^{ko} \qquad \text{for} \qquad 0 \le x \le r \tag{5}$$

$$\frac{\partial N_{trI}^{D}}{\partial t} = D_b \nabla^2 N_{trI}^{D} - (g_2 + g_3) f N_{trI}^{D} \qquad \text{for} \qquad 0 \le x \le r, \tag{6}$$

$$\frac{\partial N_{trII}^{D}}{\partial t} = D_b \nabla^2 N_{trII}^{D} + g_1 f - g_3 f N_{trII}^{D} \qquad \text{for} \qquad r \le x < \infty, \tag{7}$$

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

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

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

Mathematics of helium release in terms of the Defect Trap Model - cont.

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

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

With

$$k = 5.27\times10^{7}\exp\left(-\frac{2.67\times10^{5}}{R\left(T + 371\left(1 - \exp\left(-\frac{B}{2700}\right) + 0.041\times B\right)\right)}\right) \quad (13)$$

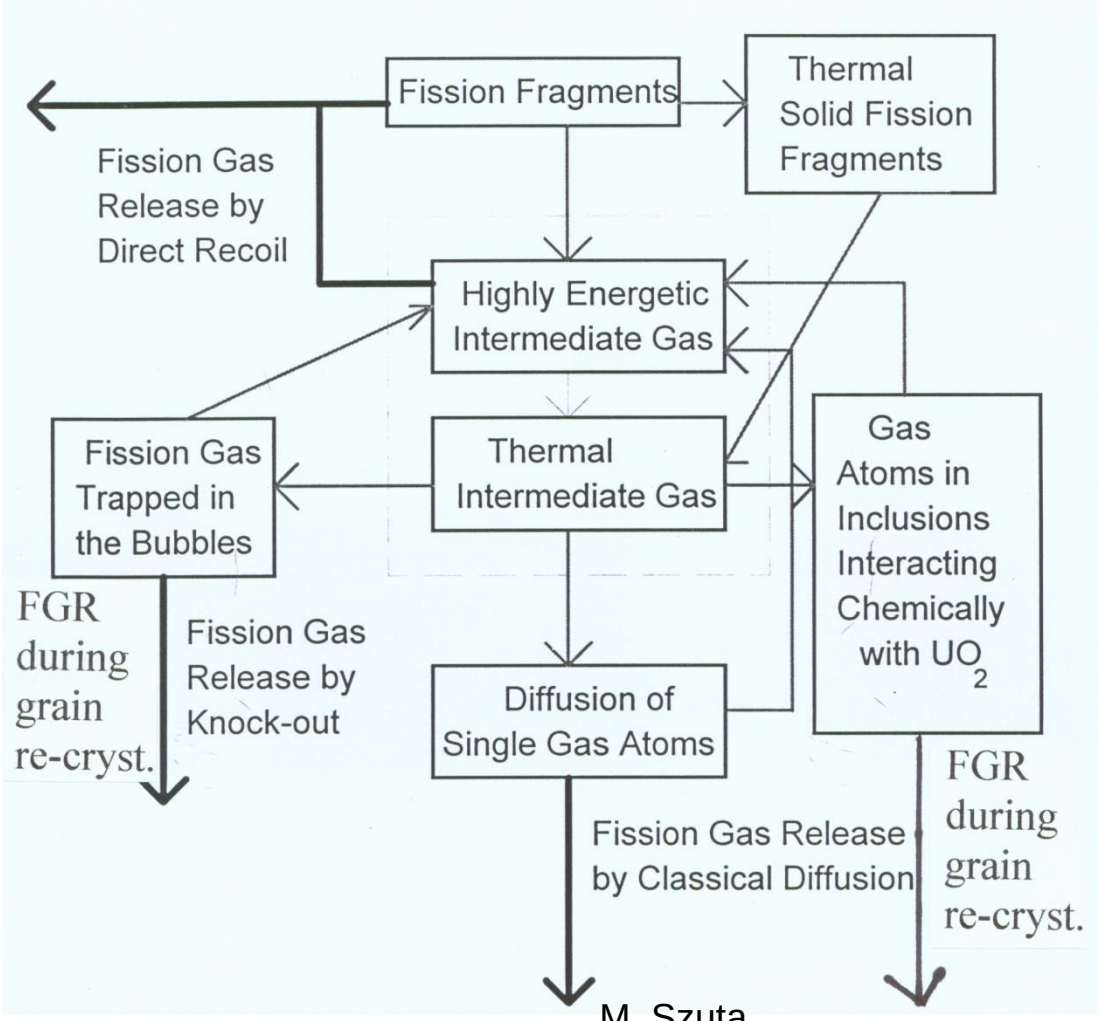
$$D_m = 2.23\times10^{3}\exp\left(-\frac{7620}{T - 520(1 - \exp(-\frac{B}{8400}))}\right) + 5\,(1 - \exp(-\frac{B}{8400}))\,) \quad (14)$$

Mathematics of helium release in terms of the Defect Trap Model - cont.

- where
- *Ntr* - concentration of bubbles in the surface layer, - bubbles created in the surface layer, - bubbles diffused into the surface layer from the bulk, - bubbles in the bulk, r - fission product range, λ – decay constant of americium isotope , Am –americium concentration, *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), D_m - limiting 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.
- Expression λAm in the equation (1) describes helium atoms production rate.

2. Mathematics of helium release in terms of the Defect Trap Model - cont.

Fig. 5. A schematic diagram of fission gas behavior in the UO_2 fuel during irradiation



In general, in the irradiated UO_2 fuel to the burn-up higher than 10^{18} fissions/cm^3 (~ 0.04 GWd/tU) the fission gas products occur in two extreme states: gas trapped in the bubbles and gas immobilized in the crystal. Each grain is assumed to be single crystal of UO_2.

Mathematics of helium release in terms of the Defect Trap Model - cont.

- We expect many anomalies to be predicted for helium behavior under irradiation by solving the proposed mathematical construct as it is observed for fission gas Kr and Xe pertaining to the burn-up range of 0.4 – 40 GWd/tU as it was collected in the last presentation [17].

.. Mathematics of helium release in terms of the Defect Trap Model - cont.

- In the above mathematical model we have introduced the following innovations and new processes:
- a) Single gas atom diffusion in the $(U, Am)O_2$ in excess of fission fluency 10^{17}fissions/cm^3 is considered negligible,
- b)- The only way of noble gas release is knock-out - The rate in which the fission track does cross the surface and the kinetically excited gas atoms of the bubble follow the fission fragment out of the bubble – this process is termed here as a knock-out release process. . The fission gas escape from the fuel through channels which are formed instantaneously, between the bubble traps and the fuel surface, by the fission products.
- c) . The knock-out process affects the UO_2 fuel surface layer to a depth of not more than 10 μm (the maximum fission product range in UO_2 fuel

Mathematics of helium release in terms of the Defect Trap Model - cont.

- d) The quantity of the fission gas released during irradiation depends on the total surface of the specimen.
- e) - The direct recoil pertaining to UO_2 molecules and, together with them, the noble gas atoms, which mostly have sufficient energy to re-enter the specimen, is also disregarded. It is distinguished from the knock-out release.
- f) While Kr and Xe atoms are chemically bond in the uranium dioxide fuel, the helium atoms are trapped in the octahedral interstitial sites.
- g) The concentration of bubbles in the surface layer consists of the average concentration of the created in the surface layer what is independent on temperature and the concentration of the diffused bubbles into the surface layer what is dependent on temperature .

.. Mathematics of helium release in terms of the Defect Trap Model - cont.

- h) A new notion of bubble diffusion during irradiation. Gas atoms caught up in the thermal spikes (kinetically excited) are free to migrate as random walkers. Consequently, the bubble migration is an entangled process of the following sequence: kinetic excitation of gas atoms in the bubbles (radiation induced evaporation-condensation process) and gas atoms immobilized in the bulk (bound chemically in the fuel and trapped in the octahedral interstitial sites), intermediate gas formation and bubble formation at their new location – this is radiation induced bubble diffusion.
- i) Grain recrystallization leads to noble gas release.
- k) The fission gas atoms released into the grain boundary channels connecting the total surface area of the pellet with the open volume (free volume) of the fuel rod do not diffuse there but are pressed by another released atoms as in a pump.

3. Analytical analysis of the simplified differential equations of the proposed model of helium release from the $(U, Am)O_2$ specimen under irradiation in function of low and intermediate temperature.

- In this section we focus on calculation of hypothetical release rate of helium atoms in function of temperature from the specimen under neutron irradiation predicting the release consisting of a temperature-dependent component and a constant temperature-independent component as an expected anomaly - peculiarity.
- This peculiarity can be explained by assumption that the same carriers brings the helium gas to the specimen surface, even though the release is by two different processes: the temperature independent process and the temperature dependent process. The carriers of helium gas and the fission gas products accomplishing the above assumption can only be the bubbles.

3a. Prediction of helium atoms release from (U, Am)O_2 specimen under irradiation in function of low temperature.

- Neglecting the terms $\alpha_1 f M_r$, $g_3 f M_{tr}$ and the terms describing the process of grain recrystallization in equations (1) – (4) we limit the behavior of noble gases to the thin surface layer of the specimen. So we get the following simplified equations of the defect trap model:

$$\frac{dM}{dt} = \lambda Am - \alpha_2 M - g N_{tr} M \qquad \textbf{(1a)}$$

$$\frac{dM_{tr}}{dt} = g N_{tr} M - g_2 f M_{tr} \qquad \textbf{(2a)}$$

$$\frac{dM_r}{dt} = \alpha_2 M \qquad \textbf{(3a)}$$

$$R = g_2 f M_{tr}(S \times r) \qquad \textbf{(4a)}$$

$$\frac{dN_{tr}^{ko}}{dt} = g_1 f - (g_2 + g_3) f N_{tr}^{ko} \quad \text{for} \quad 0 \le x \le r \qquad \textbf{(5a)}$$

3a. Prediction of helium atoms release from (U, Am)O_2 specimen under irradiation in function of low temperature.

- So the solution of the simplified differential equations for the stable state and step function of fission rate allows us to determine M_{tr}.

$$M_{tr} = \frac{\lambda Am\, gN_{tr}}{g_2 f\, (\alpha_2 + gN_{tr})} \qquad (15)$$

- When the density of bubbles is assumed equal to the density of bubbles formed in the thin surface layer then we get amount of gas released only from the thin surface layer which is not dependent on temperature using the equation (4a).

3b. Prediction of helium atoms release from (U, Am)O_2 specimen under irradiation in function of intermediate temperature.

- However the concentration of bubbles in the surface layer consists of the average concentration of the created in the surface layer (N_{tr}^{ko}) and the concentration of the diffused bubbles ($N_{trI}^{D}(x)$) into the surface layer. In order to solve analytically the problem of bubble distribution in the UO_2 single crystal, it is assumed to write separate equations of simplified equation (6) for bubbles which diffused from the bulk of the UO_2 crystal:

$$D_b \nabla^2 N_{trI}^{D} - (g_2 + g_3) f N_{trI}^{D} = 0 \qquad for \quad 0 \le x \le r, \quad (6a)$$

- The solution of eq. (6a) worked out below applies to a semi-plane of UO_2 single crystal with spatially uniform temperature and fission rate density distribution and steady state.

3b. Prediction of helium atoms release from (U, Am)O_2 specimen under irradiation in function of intermediate temperature – cont.

- The boundary conditions associated with the problem are obtained by assuming that the bubble concentration in the bulk of the fuel up to a depth of 10^{-3} cm (the maximum fission product range in the UO_2 fuel) is equal to $N^D_{trI} = g_1/g_3$ . . Using the pertinent boundary conditions and the eq. (9), one obtains the solution for the averaged diffused bubble concentration in the surface layer. Finally, after some simplification, we find:
-

$$< N^D_{trI} > \approx 10^3 \frac{g_1}{g_3} \sqrt{\frac{D_b}{f(g_2 + g_3)}} \qquad (16)$$

3b. Prediction of helium atoms release from (U, Am)O_2 specimen under irradiation in function of intermediate temperature – cont.

- Analyzing the experimental results of ^{88}Kr steady state release rate (Fig. 6) from UO_2 single crystal for ϕ = 1.1x10^{13} neutrons/cm^2·s(f ~ 10^{12} cm^{-3}s^{-1}) at 923 K (650 ^{o}C) we see that the release rate is twice as big as in the temperature independent region.
- This means that the value of diffused bubble concentration (N_{tr1}^{D}) is equal to that (N_{tr}^{ko}) formed in the layer where knock-out occurs.

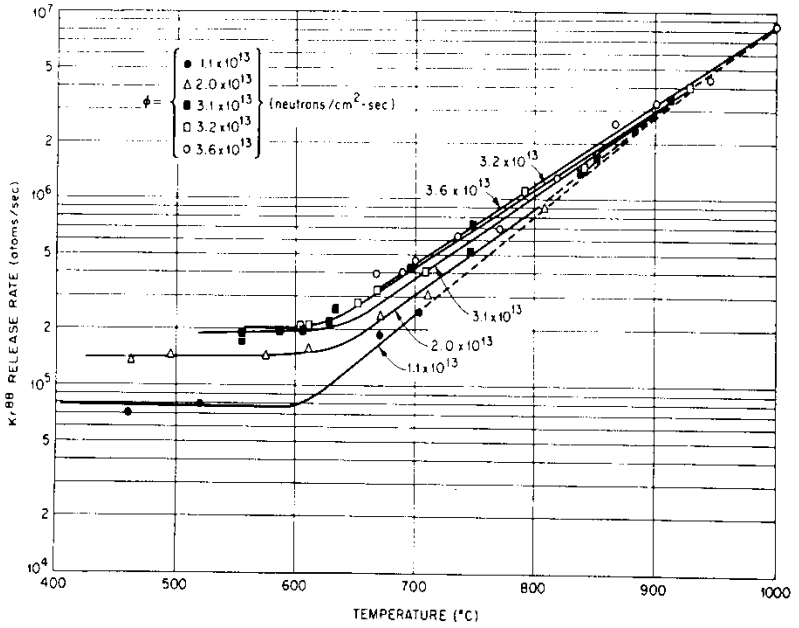


Fig. 6. Steady-state release rate of Kr^{88} from UO_2 single crystal [18, 19].

3b. Prediction of helium atoms release from (U, Am)O_2 specimen under irradiation in function of intermediate temperature – cont.

- Considering the experimental conditions and that $N_{tr}^{ko} = g_1/(g_2+g_3)$ for steady state we obtain:

$$10^3 \frac{g_1}{g_3}\sqrt{\frac{D_b(923K)}{f(g_2+g_3)}} = \frac{g_1}{g_2+g_3} \qquad (17)$$

- Using g_1=10, $g_2 = 10^{-15}$ cm^3, g_3=10^{-17} cm^3 and f = 10^{12} cm^{-3}s^{-1} we find that:

$$D_b(923\,K) = 10^{-6}\frac{g_3^2}{g_2+g_3} f \approx 10^{-11}\,cm^2/s \qquad (18)$$

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3b. Prediction of helium atoms release from (U, Am)O_2 specimen under irradiation in function of intermediate temperature – cont.

- We will use the activation energy equal to 2.3864x10^5 J/mole (57 kcal/mole) deduced from the experimental results in ref. [18, 19] which is for ^{88}Kr, and the value of the "pre-exponential" constant is estimated from eq. (18).
- The diffusion coefficient for bubble migration during irradiation thus derived may be expressed by the following equation:

$$D_b \approx 3.105\, e^{-\frac{2.3864x10^5}{RT}}\ cm^2/s \qquad (19)$$

- where R is the universal gas constant (J/K·mole) and T is the absolute temperature.

3b. Prediction of helium atoms release from (U, Am)O_2 specimen under irradiation in function of intermediate temperature – cont.

- Applying the equations (15), (16), (10) and (4a) we can to evaluate release rate of helium atoms from the specimen identical as in the Carroll's and Sisman's [19] experiment with the difference that 20% of americium dioxide (~ 2.21 g) and 80% of uranium dioxide is in the sample instead 100% of UO_2.

$$M_{tr} = \frac{\lambda Am\, gN_{tr}}{g_2 f\, (\alpha_2 + gN_{tr})} \qquad (15)$$

$$< N_{trI}^{D} > \approx 10^3 \frac{g_1}{g_3} \sqrt{\frac{D_b}{f(g_2 + g_3)}} \qquad (16)$$

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

$$R = g_2 f M_{tr} (S \times r) \qquad (4a)$$

3b. Prediction of helium atoms release from $(U, Am)O_2$ specimen under irradiation in function of intermediate temperature – cont.

- Data needed to estimate the release of helium from the specimen:
- To solve the release rate of helium atoms from the specimen the following appropriate values for the constants were used: $g=10^{-19}$ $s^{-1}\cdot cm^3$, $g_1=10$, $g_2=10^{-15}$ cm^3, $g_3=10^{-17}$ cm^3, $\alpha_1=10^{-16}$ cm^3 and $\alpha_2=0.999$ s^{-1}, fission rate f ~ 1.0 10^{12} fissions/$cm^3\cdot s$. Decay constant of ^{241}Am is equal to = 5.085 10^{-11} s^{-1}; Fission accumulated yield of ^{88}Kr is β_i = 3.53%.
- Specimen sizes and required data for performing the calculation:
- The specimen [18, 19] was in the form of two thin plates, about 0.040-in. thick and 0.5-in. wide, geometrical surface S= 5.874 cm^2, volume V = 0.514 cm^3, weight W=5.63 g in the experiment [18, 19]

3b. Prediction of helium atoms release from (U, Am)O_2 specimen under irradiation in function of intermediate temperature – cont.

- The average density of the (U, Am)O_2 specimen fuel d = 10.986 g/cm^3 where 20% of AmO_2 (0.199 cm^3; 2.21 g) and 80% of UO_2 (0.8 cm^3; 8.776 g). For the same volume (V = 0.514 cm^3) as in the experiment [18, 19], the considered (U, Am)O_2 specimen weighted 5.65 g.
- 2.21 g of AmO_2 molecules (~0.2 cm^3) is equivalent to 8.06 10^{20} Am-241 atoms Production rate of helium by 2.21 g of AmO_2 is equal to Am = 4.1036 10^{10} cm^{-3} s^{-1} while production rate of ^{88}Kr is equal to $f\beta_i$ = 3.53 10^{10} cm^{-3} s^{-1} for fission rate f ~ 1.0 10^{12} fissions/cm^3·s. The production rates are comparable.

5b. Prediction of helium atoms release from $(U, Am)O_2$ specimen under irradiation in function of intermediate temperature – cont.

- Calculation results of hypothetical steady-state release rate of helium from the $(U, Am)O_2$ specimen crystal and bubble density in thin surface layer of the crystal in function of temperature during neutron irradiation are presented in Table 1 and in Fig. 7.

Table 1. *Hypothetical bubble density in thin surface layer of UO_2 and AmO_2 mixture single crystal and steady-state release rate of helium release from the crystal in function of temperature during neutron irradiation.*

T [^oC]	350	450	550	650	750	850	1000
$< N_{tr}^{D} >$ [cm^{-3}] 57 kcal/mole	$5.51\ 10^{12}$	$3.28\ 10^{14}$	$3.24\ 10^{15}$	$1.0\ 10^{16}$	$4.66\ 10^{16}$	$1.55\ 10^{17}$	$7.11\ 10^{17}$
$N_{tr}= N_{tr}^{ko} + < N_{tr}^{D} >$ [cm^{-3}]	$1.00\ 10^{16}$	$1.03\ 10^{16}$	$1.32\ 10^{16}$	$2.0\ 10^{16}$	$5.66\ 10^{16}$	$1.65\ 10^{17}$	$7.21\ 10^{17}$
R [He atoms/s]	$2.39\ 10^{5}$	$2.46\ 10^{5}$	$3.16\ 10^{5}$	$4.77\ 10^{5}$	$1.35\ 10^{6}$	$3.95\ 10^{6}$	$1.72\ 10^{7}$

3b. Prediction of helium atoms release from $(U, Am)O_2$ specimen under irradiation in function of intermediate temperature – cont.

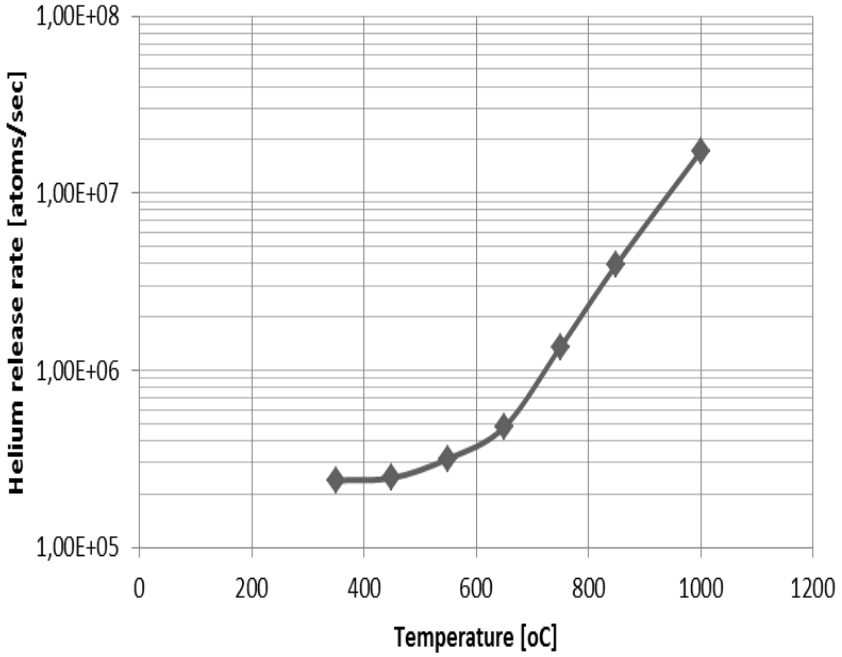

Fig.7. Hypothetical steady-state release rate of helium from the $(U, Am)O_2$ specimen

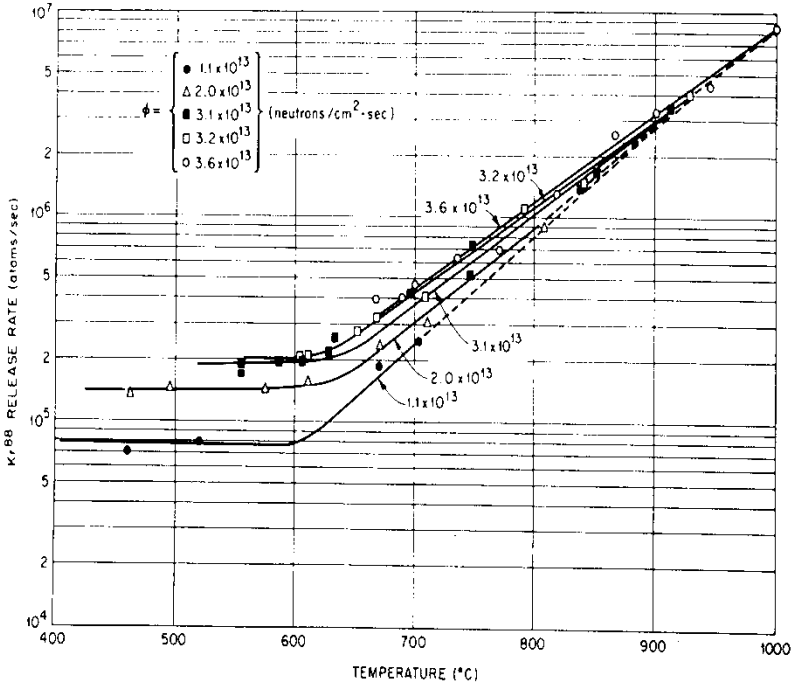

Fig. 6. Steady-state release rate of Kr^{88} from UO_2 single crystal [18, 19].

- **4. Conclusions**
- Both the shape of the predicted helium release rate and the experimental krypton release rate from the $(U, Am)O_2$ single crystal specimen (see Fig. 7) and UO_2 single crystal (see Fig. 6) respectively in function of temperature under neutron irradiation are alike.
- This let us to infer that we should not to bother of increased pressure under the clad of the fuel rod due to helium release under condition that the ratio of Am/U in the fuel pellets does not cross 0.2 – proper ratio of actinides to uranium in the fuel called “blanket” is required.
- Also specific values for the rate of helium and krypton release are similar as the rate of helium production ($4.1036\ 10^{10}$) and krypton - 88 production ($3.53\ 10^{10}$) are similar.

4. Conclusions

- This calculations and the comparison of release rate of helium with the Kr-88 release rate confirm us that the anomaly of the release consisting of a temperature-dependent component and a constant temperature-independent component can be expected.
- This in turn supports the new notion that the bubble migration is an entangled process of the following sequence: „kinetic excitation of gas atoms in the bubbles and gas atoms bound chemically and immobilized helium in the octahedral positions in the fuel, intermediate gas formation and bubble formation at their new location" – this is radiation induced bubble diffusion. Since the atoms in the bubbles are not distinguishable we can assume simplification by writing the algorithms for the radiation induced bubble diffusion.

5. References

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- Thank you for the attention.

Experimental data.
Bubble behavior in irradiated UO_2 fuel during irradiation

Table X. Results of four uranium dioxide pellets irradiated over a wide temperature range and a different burn-up (R.M. Cornell – 1970).

	Pellet A	Pellet B	Pellet C	Pellet D
Doses (fissions /cm^3)	$3.5x10^{19}$	$2.3x10^{20}$	$4.6x10^{20}$	$2.6x10^{20}$
Pellet center temperature (^oC)	1640	920	910	1275
Pellet surface temperature (^oC)	700	650	640	560
Bubble concentration (cm^{-3})	$1.2\text{-}3.8x10^{17}$	$2.0\text{-}3.3x10^{17}$	$2.6\text{-}3.4x10^{17}$	$1.0\text{-}2.4x10^{17}$
Bubble diameter (nm)	1.7-2.8	1.6-2.1	1.7-2.1	2.1-3.1
Gas content in bubbles (cm^{-3})	$0.95\text{-}1.3x10^{19}$	$0.8\text{-}1.45x^{19}$	$1.0\text{-}1.3x10^{19}$	$1.15\text{-}1.5x10^{19}$
Total gas content (cm^{-3})	$1.7x10^{19}$	$5.0x10^{19}$	$10.0x10^{19}$	$5.6x10^{19}$

5. Anomalies of fission gas behaviour and their defect trap model predictions.

Appendix (Supplement).

Marcin Szuta

Fission gas release is caused by a combination of two basic processes: a temperature independent process and a temperature dependent process [1].

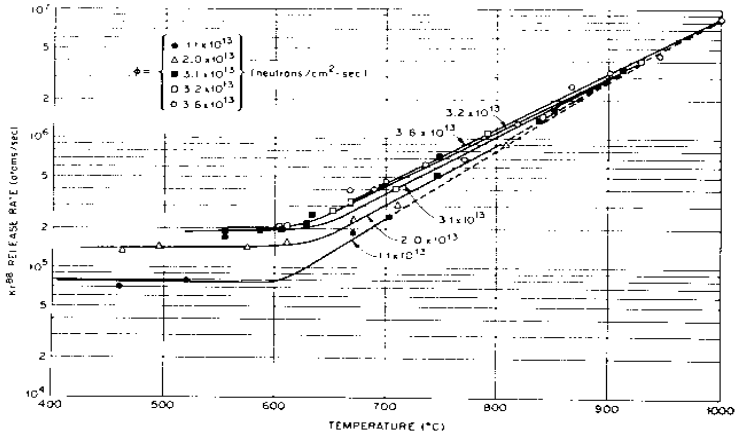

Fig. 1;3. Steady-state release rate of Kr^{88} from UO_2 single crystal [1].

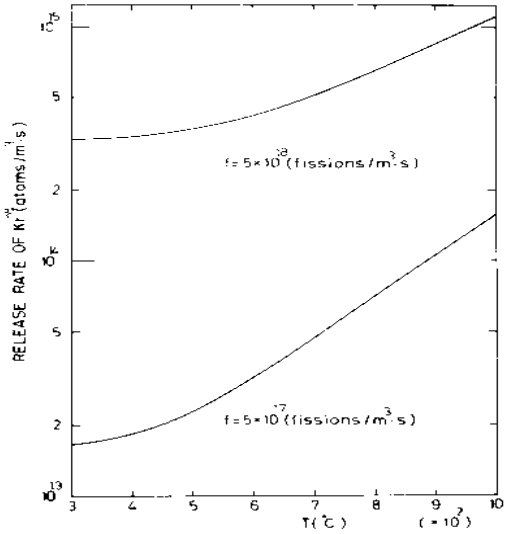

Fig. 1a. Theoretical fission gas release rate of ^{88}Kr as a function of temperature [2].

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 [1].

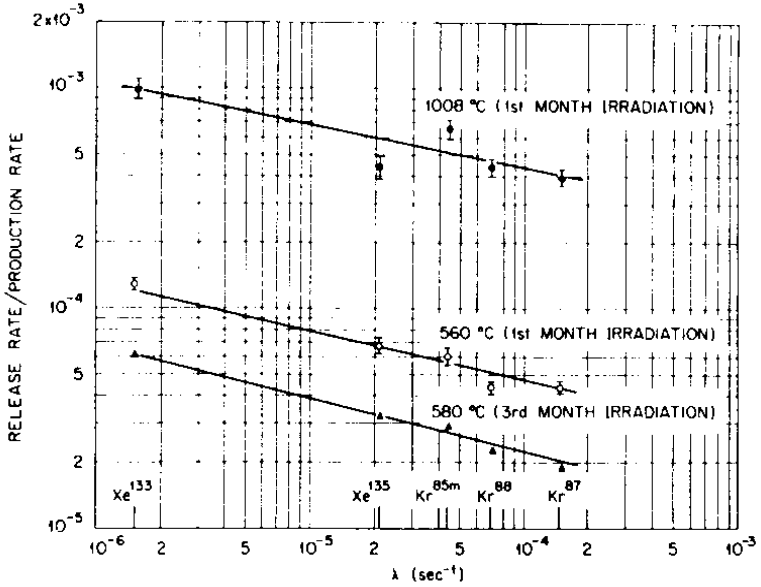

Fig. 3. Dependence of fractional release upon decay constant, UO_2 single-crystal (Kr^{85m} plotted for fission yield of 2 %) [1]

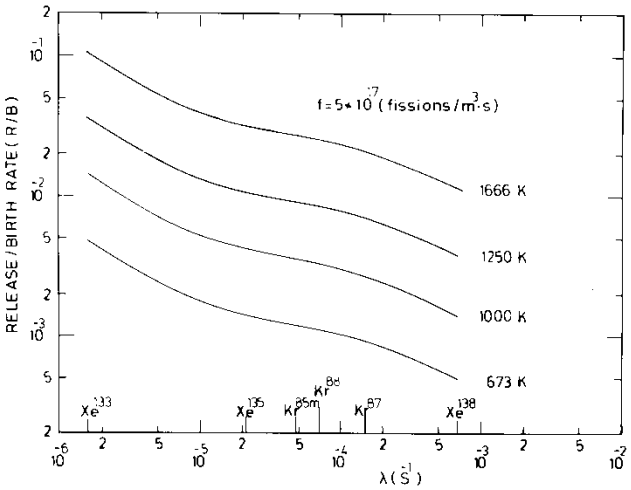

Fig. 3a. Theoretical fractional fission gas release rate as a function of the decay constant [2].

The fission gas release is a function of fission rate:

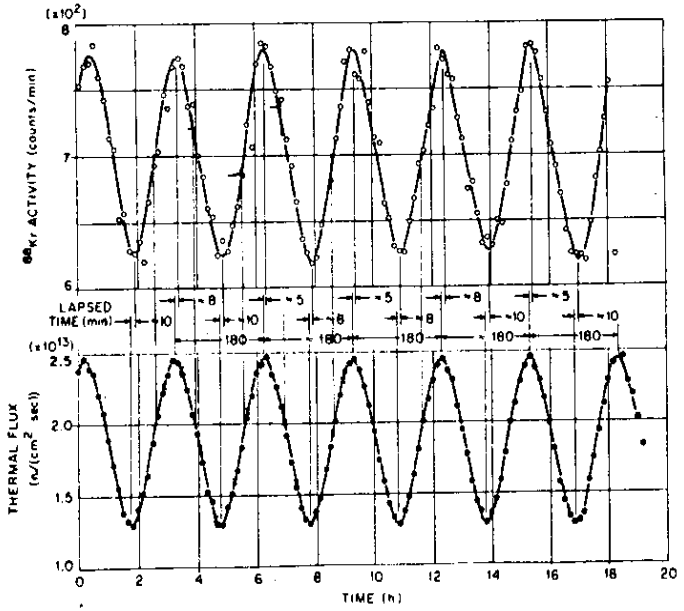

Fig. 4. UO_2 single crystal, 3 h flux cycle, 575° C [4].

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,

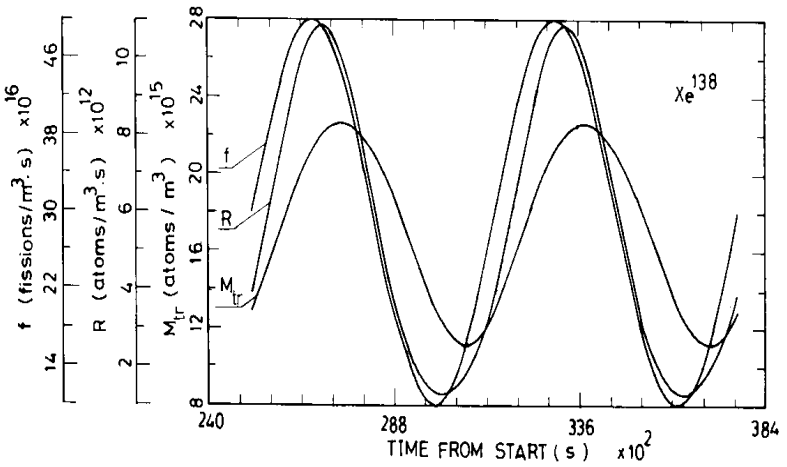

Fig. 4a. Theoretical release rate R and concentration of gas trapped in the bubbles M_{tr} as a function of time for oscillating fission rate f [5].

c2. Fission gas release in non-temperature dependent region during the neutron flux oscillation is greater when the flux is decreasing than when the flux is increasing (hysteresis loop) [4],

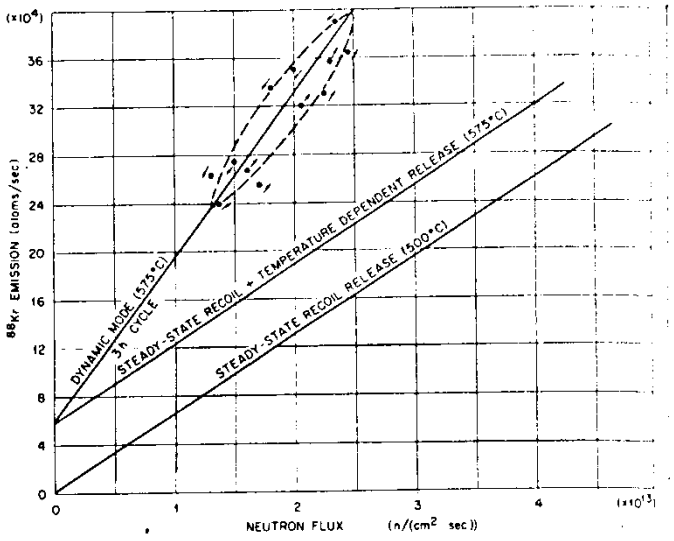

Fig. 5. UO_2 single crystal [4].

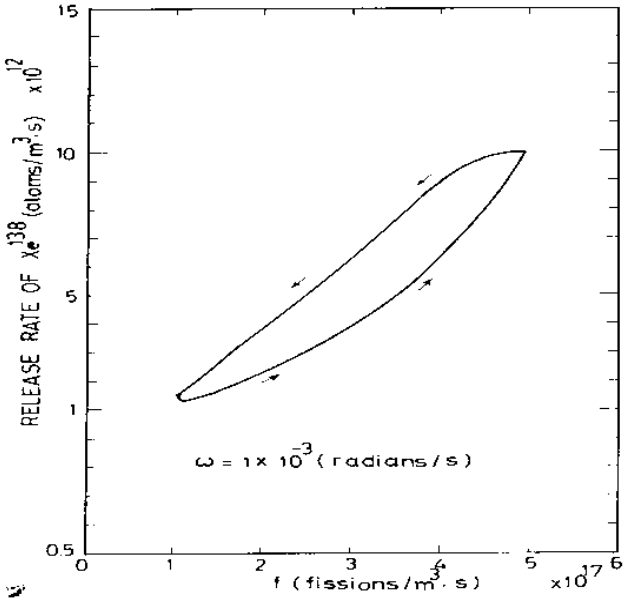

Fig. 5a. Theoretical fission gas release rate of ^{138}Xe as a function of the oscillating fission rate [5].

c3. 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 [6],

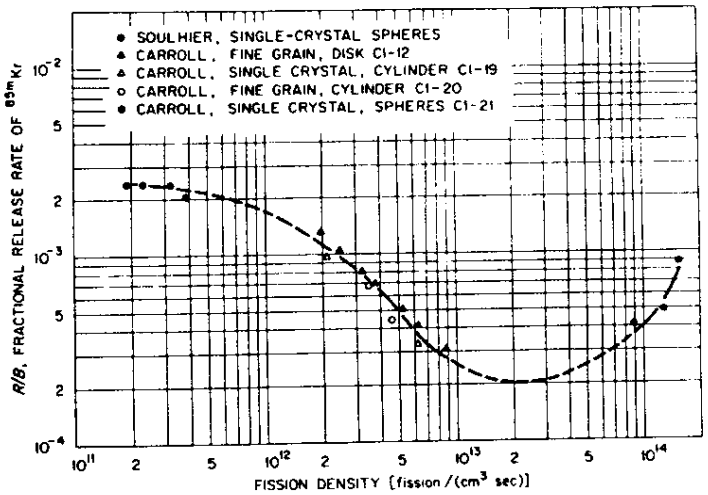

Fig. 6. Fractional release of Kr^{85m} from high density stoichiometric UO_2 during irradiation at 1400° C [6]

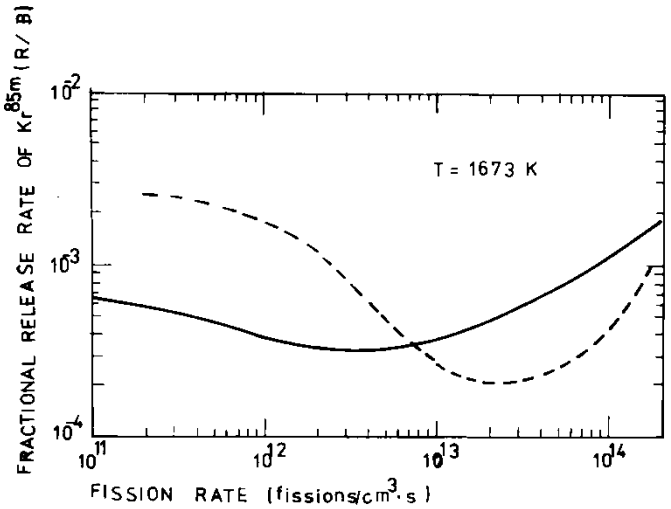

Fig. 6a. Comparison of theoretical fractional release of ^{85m}Kr as a function of fission rate for data of the fuel specimens from ref. [6] (continuous plot [5]) with experimental data (dashed plot [6]) copied from fig. 1 of ref. [6] (see fig. 6).

c4. Fission gas release in the temperature dependent region is a periodic function for a sinusoidally changing fission rate with a more complex form [4].

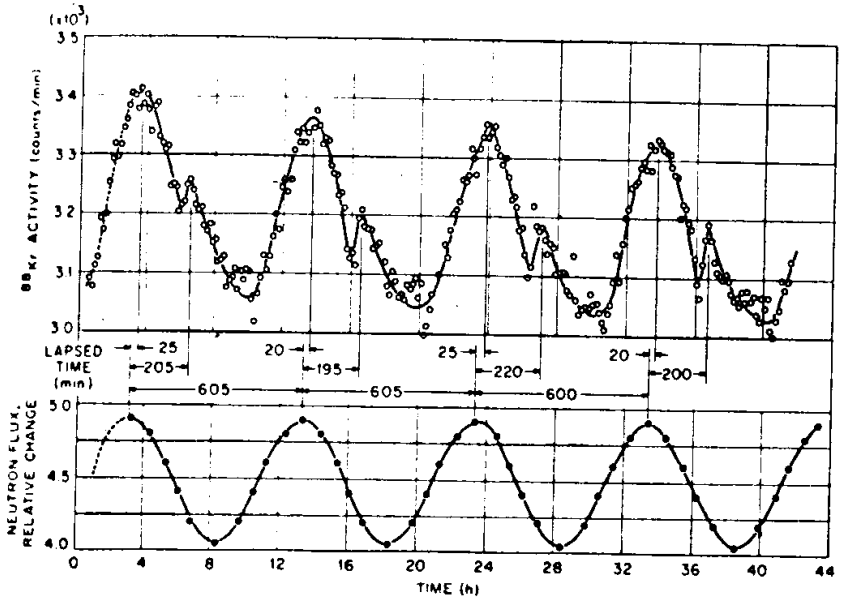

Fig. 7. UO_2 single crystal, 10 h cycle, 840° C

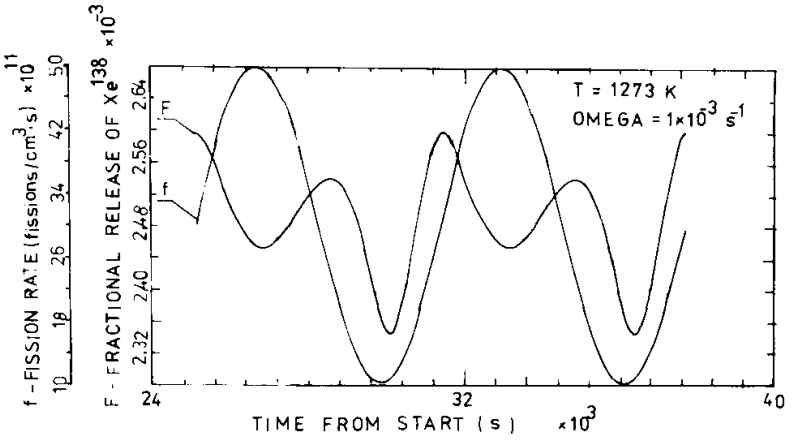

Fig. 7a. Theoretical fission gas behaviour of ^{138}Xe as a function of time for oscillating fission rate [2].

e) **Diffusion coefficients are not unique functions of temperature but also depend on irradiation parameters e.g. burn-up and ratings:**

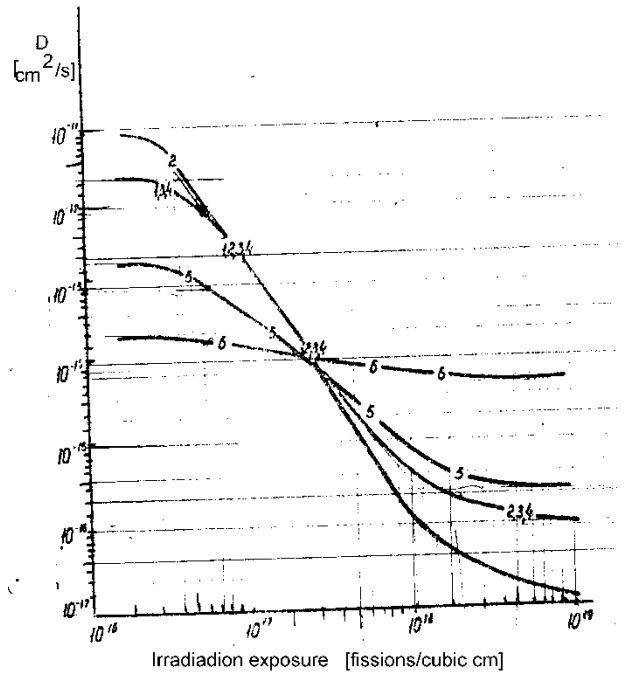

Fig. 10. Dependence of apparent diffusion coefficient irradiation exposure. 1-Xe133 UO_2, 2-Xe133 UC, 3-Xe133 UC_2, 4 -Xe133.UN, 5 – Xe135, UO_2, UC, UC_2, UN, 6 – Xe – 138, UO_2, UC, UC_2, UN [9].

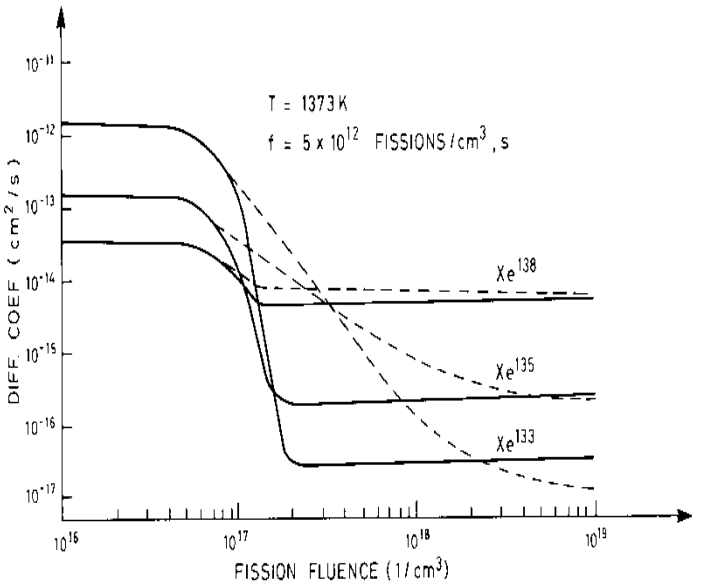

Fig. 10a. Comparison of the calculated apparent diffusion coefficient as a function of burn-up (continuous plots [10]) with the experimental data (dashed plots [9]) copied from fig 10 [10].

d). Equilibrium state obtainment of non-temperature-dependent fission gas release rate is dependent on the decay constants of the radioactive gas isotopes.

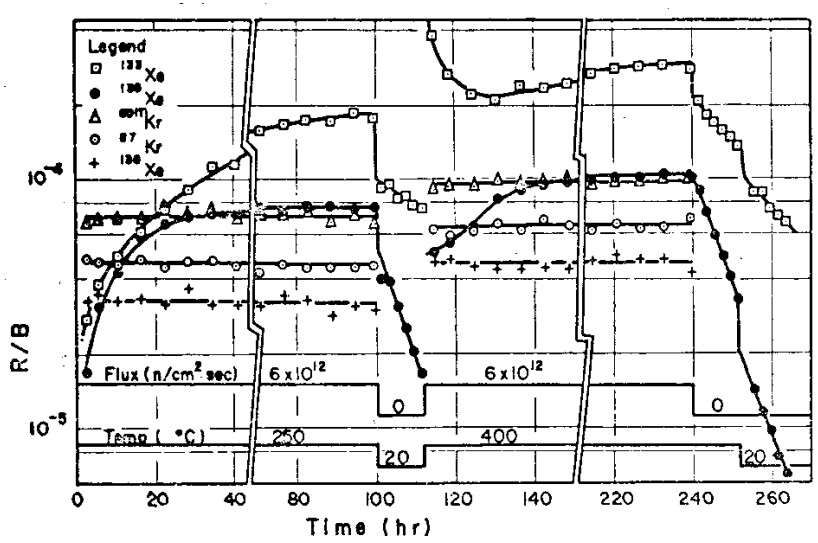

Fig. 8. Fractional fission gas release from UO_2 against irradiation time [7].

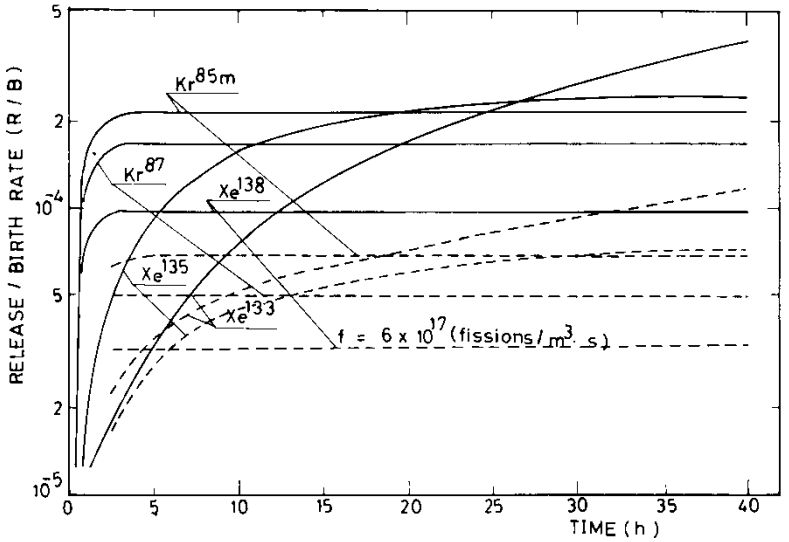

Fig. 8a.. Comparison of theoretical fractional fission gas release as a function of time for the step function of fission rate for data of fuel specimen from ref. [7] (continuous plots [5]) with experimental data (dashed plots [7]) copied from fig. 1 of ref. [7].

e2) The diffusion coefficient exhibits athermal characteristics below 800 ^{0}C [11],

e3. Contrary to expectations of the classical diffusion the tendency for xenon to diffuse as fast as krypton is observed [12].

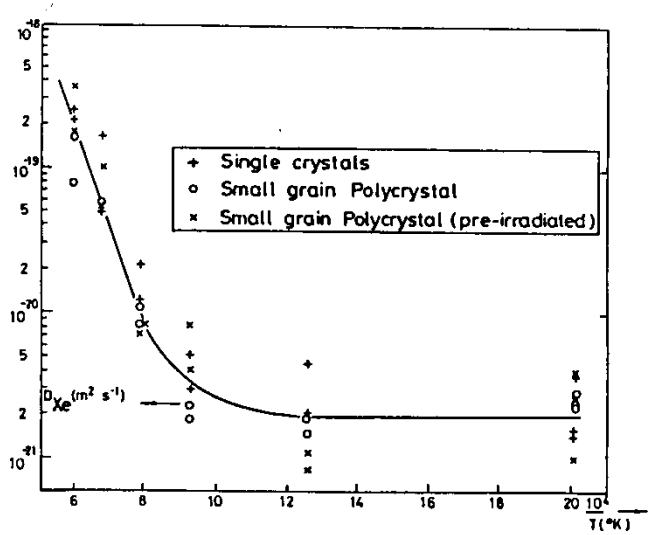

Fig. 11 Data for xenon diffusion coefficient plotted against reciprocal temperature [11].

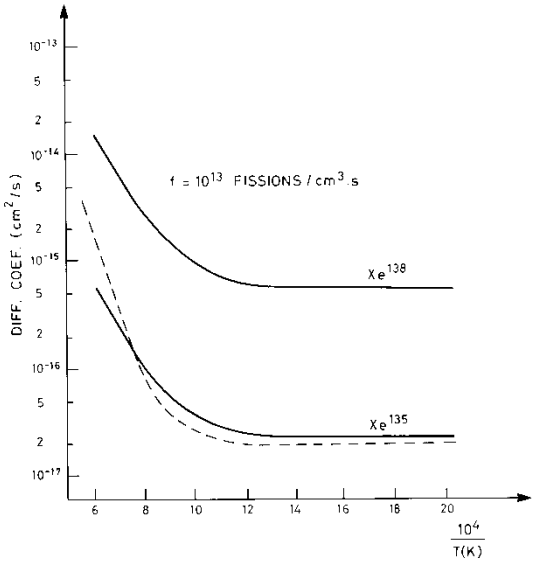

Fig. 11a. Comparison of the calculated apparent diffusion coefficient versus reciprocal temperature for xenon isotopes (continuous plots) with the experimental data (dashed plots) [10].

f. An abrupt burst of fission gas is emitted from the single crystal UO_2 when the temperature is increased (rather than a smooth transition to the higher release rate) [13].

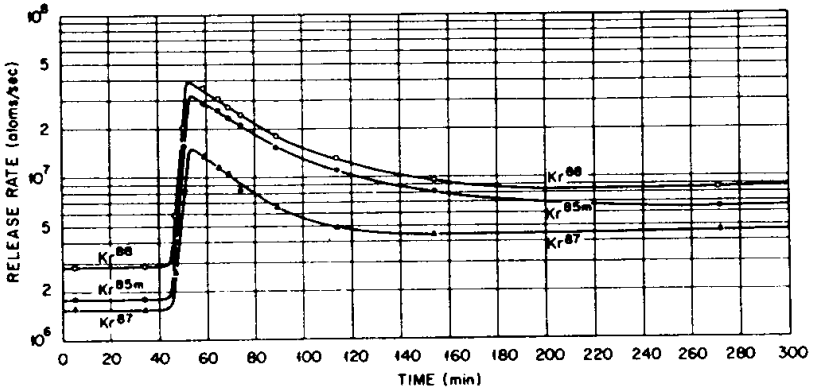

Fig. 12. Krypton release when single crystal UO_2 specimen A temperature was increased from 865^o C to 1040 ^{o}C at constant flux. of 3.3×10^{13} neutrons/cm^2 sec [1]

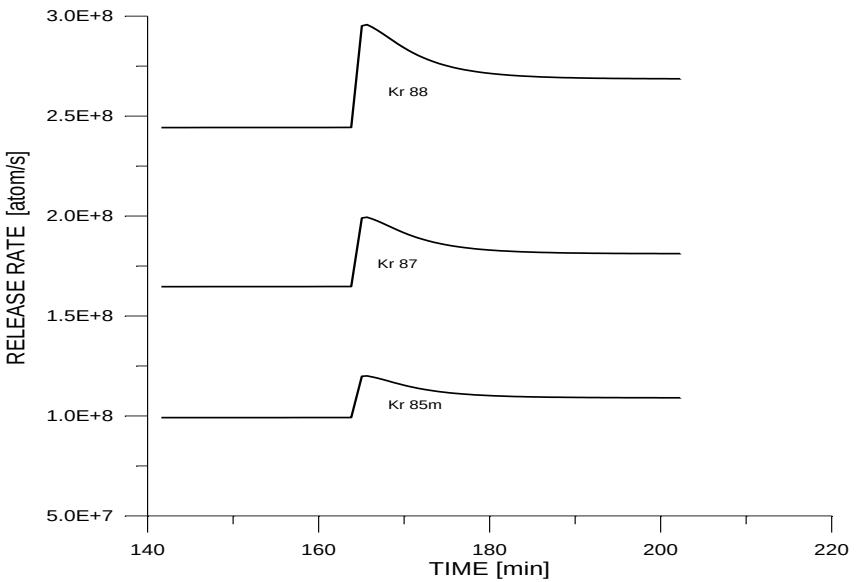

Fig. 12a. Theoretical krypton release when fuel temperature is increased from 865 ^{o} C to 1040 ^{o} C at constant fission rate of $3.3\ 10^{12}$ fission/cm^3·s, initial grain size of 9 μm and burn-up of 35 MWd/kgU [14].

Anomalies of fission gas behaviour and their defect trap model predictions.

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