

Analysis of noble gas accumulated in the irradiated UO₂ fuel and in mineral materials containing fissile atoms

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Abstract— Both the old mineral materials containing fissile isotopes and the neutron irradiated uranium dioxide fuel hold fairly a lot of noble gases.

Appreciable fraction of the inert gases is immobilized in the uranium dioxide fuel after irradiation exposure in excess of 10^{15} fissions/cm³.

Noble gas atoms can be trapped by the point defects, clusters of the point defects, clathrates and small bubbles in the irradiated material.

It is suggested that above a fission fluency threshold about 10^{15} fissions/cm³ occurs an additional trapping process based on the irradiation induced chemical bonding process.

It is inferred further that as well in the minerals and in the irradiated UO₂ fuel the gas release kinetics is determined by the kinetics of thermal recovery of the radiation induced defects in the low temperature range.

It is surmised that the same amount of damage caused by alpha decay of uranium is much less effective than caused by fission of uranium for the immobilization of noble gases since only during fissioning there are formed conditions for strong chemical binding between the noble gas atoms and the mineral materials.

Finally it is concluded identical nature of noble gas accumulation mechanisms in the irradiated UO₂ fuel and in the old mineral materials containing fissile atoms.

Spontaneous fission fluency for the minerals containing fissile atoms of about 2 billions years old is about 10^{15} fissions/cm³.

Alike the old minerals containing fissile isotopes and the irradiated UO₂ fuel reveal during annealing three peaks of gas release in the low temperature range. In the range only several percent of the total amount of gas as well in the mineral materials and the uranium dioxide fuel is released. It still holds significant part of noble gases after annealing at a temperature about 1400 °C.

Keywords—mineral materials, noble gases, uranium dioxide fuel, single crystal, irradiation defects.

I. INTRODUCTION

BOTH the old mineral materials containing fissile isotopes and the neutron irradiated dioxide material 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 cm³ He/g, which is equivalent to the fission gas concentration at a burn-up of 300000 MWd/t [1].

Also appreciable fraction of the inert gases is immobilized in the uranium dioxide material after irradiation exposure in excess of 10^{15} fissions/cm³ [2].

It is proved also that irradiating the UO₂ pellets in the presence of natural xenon, part of the gas atoms is imbedded into the pellet. The xenon is found to be firmly attached to the UO₂ surface such that only 1 % of the attached gas can be removed after annealing samples for over 12 hours at 1400 °C [3], [4].

On the other hand, some minerals containing fissile isotopes still hold significant part of xenon after annealing at 1325 °C [5].

II. EXPERIMENTAL DATA

The uraninite and nasturan minerals are the most suitable uranium minerals to be compared with the UO₂ fuel since they have high percentage of uranium which occur in the form of uranium oxides. Both the minerals and the UO₂ fuel have a cubic crystal structure of the fluorite type (CaF₂) – ionic crystal.

The particular feature of the gas release (Xe, Kr, Ar, He) from the uranium minerals during annealing is that it has three main peaks for all the gases principally at the same temperatures. For instance, for the uraninite Big Lopot the three peaks lie in the temperature ranges 200 – 300 °C, 700 – 800 °C, 1100 – 1200 °C, for the uraninite MF – 200 -300 °C,, 900 – 1000 °C, 1200 – 1300 °C and for the nasturan G1 – 200 – 300 °C, 600 700 °C, 1000 – 1100 °C. The most common temperature range for all the minerals is the first peak, the second peak and especially the third peaks are a little bit different for the different minerals (see Fig. 1).

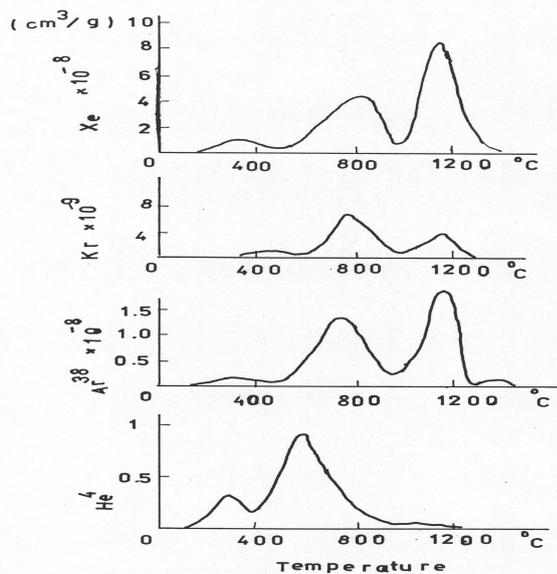


Fig. 1. Differential curves of radiogenic gas release from the uraninite Big Lopot during annealing in cm³/g (copied from Fig. 37 of ref. [5]).

This fact does not agree with the theory of single gas atom diffusion in the crystal matrix since the radiuses of the considered atoms are very different: Xe – 0.218 nm, Kr – 0.198 nm, Ar – 0.192 nm, He – 0.122 nm. It means that the diffusion coefficient (D) of the considered isotopes should obey the following dependences

$$D_{Xe} < D_{Kr} < D_{Ar} < D_{He} \quad (1)$$

Activation energies of xenon, krypton, argon, helium release during annealing of the considered above minerals for the three peaks in the relative temperature ranges are really constant within the accuracy of experiment (Table 1) [5].

Table 1. Activation energies of Xe, Kr, Ar, He release during annealing [5].

Uranium	Gas	200 – 400 °C	600 – 900 °C	1000 °C
		E _{ac} kcal/mole	E _{ac} kcal/mole	E _{ac} kcal/mol e
Uraninite Big Lopot	Xe	14.7 ± 4.4	42 ± 13	66 ± 20
	Kr	-	46 ± 14	110 ± 30
	Ar	14.7 ± 4.4	25 ± 14	110 ± 30
	He	14.7 ± 4.4	29 ± 8	-
Uraninite MF	Xe	13.5 ± 4.0	55 ± 16	75 ± 22
	Kr	-	64 ± 19	82 ± 25
	Ar	-	86 ± 26	110 ± 30
	He	-	59 ± 19	-
Nasturan G1	Xe	12.5 ± 3.7	29 ± 8	43 ± 13
	Kr	-	-	64 ± 19
	He	8.5 ± 2.5	37 ± 11	-

Similar experimental results are obtained [6] for the uranium oxides with different O/U ratios ranging from 2.00 to 2.62. The samples of uranium oxides were irradiated at a thermal neutron flux of 5x10¹¹ n/cm² to a total fission fluency 10¹⁵ fissions/cm³. The sample temperature under irradiation was estimated to be less than 80 °C. Release of xenon from the irradiated samples during annealing revealed also three main peaks as for the annealed minerals of uraninite and nasturan mentioned above. For instance, for the UO_{2.14} samples the three peaks lie in the temperature ranges 180 -250 °C, 600 - 650 °C and about 1000 °C. The heating curve for the xenon release from the UO_{2.14} sample is shown in Fig. 2. It is important to remember the fact that all the samples were annealed at 1050 °C prior to irradiation.

Also data of radiogenic gas release from the minerals containing uranium during annealing are of interest:

- phosphorites (xenotime, monozite, britholite),
- titanictantalousniobates-complex oxides (samarskite, khlopinte, ampargabeiite, betafite).

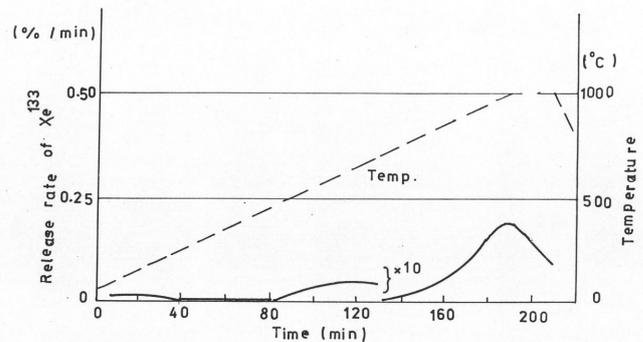


Fig. 2. Heating curve for xenon release from UO_{2.14} (copied from Fig. 5 of ref. [6]).

It is worth noting that during annealing of the mineral xenotime, 2 % of the total amount of gas in the mineral is released in the low temperature range. But it still holds about 85 % of xenon after annealing at 1325 °C. It means that to anneal the whole gas from the mineral, an activation energy above 120 kcal/mole is needed. Practically whole helium is released at temperature below 1200 °C [5].

The titanictantalousniobates minerals belong to the family of minerals where less than 5% of xenon is detained after annealing at 1300 °C. Percentage of uranium in all the minerals is of the same order. The peculiarity of these minerals is that the activation energies of krypton and xenon release during annealing at high temperature for young minerals (140 million years) as khlopinte and betafite are different according to the classical diffusion. For older minerals as samarskite (330 million years) and ampargabeiite (1950 million years) the release of krypton and xenon occurs for the same activation energies. This implies that the gas release from minerals during annealing depends on the amount of defects [5].

III. DISCUSSION

In general we can say that both the minerals containing fissile isotopes and the irradiated UO_2 fuel reveal during annealing three peaks of gas release.

In order to know the nature of the release peaks for the UO_2 fuel some samples were again heated after the ordinary experiments (the first run) were completed. No peak was observed and the releases were significantly decreased. This indicates that the elementary processes for these peaks are destroyed by the heat treatment of the first run and are of thermally irreversible nature. This behavior of the peaks is consistent with the phenomena of annealing of radiation induced defects [6].

On the basis of the conclusions of the authors studying the fission gas release from the UO_2 fuel, Morozova and Ashkinadze [5] infer that the three main peaks of release during annealing of the minerals can be explained by three different defects:

The first peak in the range of temperature 200 – 400 °C can be the result of gas migration from under surface flaws and voids what is proved by low activation energy (about 10 kcal/mole) and relatively small amount of released gas (5 – 7 %),

The second and third peaks can be caused by the annealing of the two types of irradiation defects.

The results of annealing the uranium dioxide after irradiation show three activation energies (4.6, 46, 92 kcal/mole) what is very consistent with the activation energies of the gas release from the uranium minerals during annealing (see Table 1). This confirms the dependence of gas release on the phenomena of annealing of radiation induced defects.

Thermal recovery of radiation defects and micro-structural change in irradiated UO_2 fuels studied by x-ray diffraction and transmission electron microscopy [7] supported the above interpretation. The point defects induced by irradiation begin to recover around at 450 – 650 °C and almost completely recovered above 850 °C, while defect clusters of dislocation at 1150 – 1450 °C [7].

So the gas release kinetics is determined by the kinetics of thermal recovery of the radiation induced defects.

The results suggest also that helium in the minerals is mainly detained by the point defects since almost whole helium is released in the temperature range 600 – 800 °C both from the minerals containing uranium (phosphorites, complex oxides).

However the minerals containing uranium, especially the phosphorites, beside the three peaks of radiogenic gas release still hold significant part of gas (Xe) which requires higher temperature of annealing than 1325 °C to be released [5].

We assume that for these kind minerals, even at this level of irradiation defects, some of the fission gas products are interacting chemically with the mineral.

The surmise is based on the analysis of the effect of the chemically inert fission gas products on the chemistry of the irradiated UO_2 [8].

It is assumed that the areas of atomic disorder remaining after the passage of a fission fragment can bind charged fission gas fragments with a certain probability and prevent their

mobility. There is a strong binding between gas atoms and irradiation defects [9].

The process of strong binding of the fission gas fragments with the irradiation defects is described in the work [8] as a process of chemical interaction with the UO_2 .

The alpha particles created in the process of uranium decay can also be interacting chemically with the mineral materials but probability of this process is of less significance as the charge of the particle is small and the electrical neutrality is obtained easy forming helium.

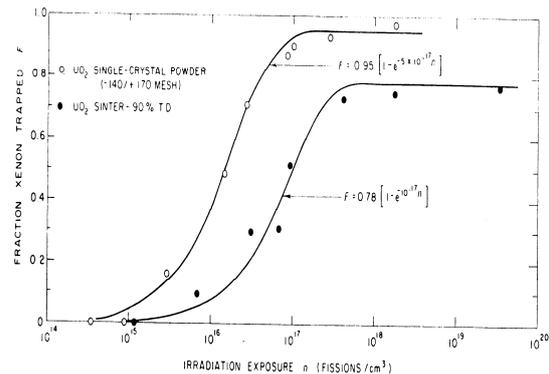


Fig.3. A plot showing how the fraction of the fission xenon trapped after annealing for a short period at 1400 °C varies as a function of the prior irradiation exposure (copied from Fig. 5 of Ref. [2]).

We can say that the probability of the chemical interaction for UO_2 fuel is dependent on irradiation damage (burn-up) and its shape versus fission fluency is like the shape of variation of a plot showing the fraction of the fission xenon trapped as a function of the prior irradiation exposure (see Fig. 3 copied from Fig. 5 of Ref. [2]).

So the fission fluency threshold when the more intensive process of chemical interaction occurs is evaluated to be about 10^{15} fissions/cm³. The second fission fluency threshold when the diffusion of single gas atoms is insignificant, it means when practically all the gas comes into chemical interaction is obtained for the burn-up above 10^{18} fissions/cm³. Above this burn-up the probability is independent of burn-up until saturation is obtained. It is expected that the gas being chemically bound can be released during annealing at a temperature of re-crystallization of UO_2 in a form of a large burst release. The higher burn-up the higher burst release should be observed.

Out-of-pile experiments support the assumptions [10]. The critical temperature of burst release is about 1800 °C for low burn-up and decreases to about 1500 °C for high burn-up (30 MWd/kgU). This that the critical temperature decreases suggests that the re-crystallization temperature of UO_2 is changed by the process of chemical interaction (see Fig.4 copied from Fig. 4 of Ref. [10]).

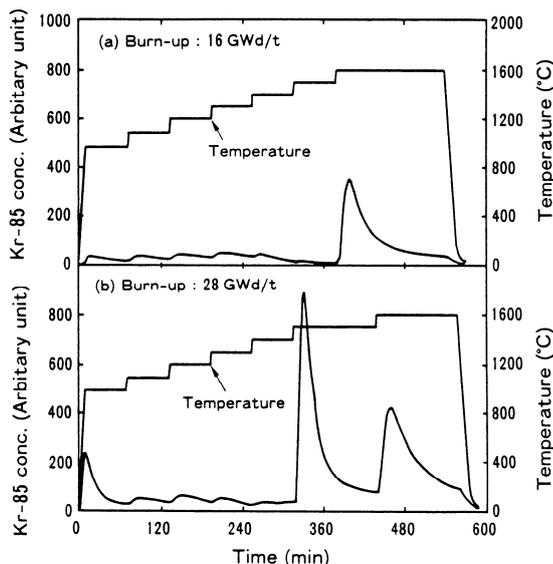


Fig. 4. Kr^{85} release on stepwise temperature ramp (copied from Fig. 4 of Ref. [10]).

Placing side by side the two figures Fig. 2 and Fig. 4 which show the fission gas release from the UO_2 fuel during annealing we can infer that in fact measuring precisely the release we should observe four peaks of release. The first three peaks are determined by the kinetics of thermal recovery of defects: inherent surface flaws and voids—first peak, radiation induced point defects—second peak and defect clusters of dislocation—third peak. While the fourth peak caused by destroying the radiation induced chemical bonds requires higher annealing temperature.

The fact that during annealing of the mineral xenotime at 1325°C it still holds about 85 % of xenon let us infer that the noble gases behavior in the minerals is alike in the irradiated fuel— is expected four peaks.

The temperatures ranges of the first three peaks can be conjoined with the experimental data pertaining thermal recovery of the irradiation-induced lattice defects in UO_2 single crystals that have been irradiated to 3×10^{20} alpha-particles/ m^2 [14]. Three recovery stages were observed in the temperature range between 50° and 1050°C . The isochronal recovery of the change in lattice parameter of a UO_2 single crystal is shown in in fig. 5. (copied from Fig. 3 of Ref. [14]).

The lattice expansion is almost completely recovered by 1050°C in three stages. Although there appears to be an equal amount of recovery at each stage, the stage II appears much sharper than stage I or stage III. From Fig. 5b the average temperature associated with the recovery stage, under these annealing conditions, can be determined from the position of the peak in the differential recovery curve. These characteristic temperatures are as follows: stage I (300°C), stage II (575°C) and stage III (925°C).

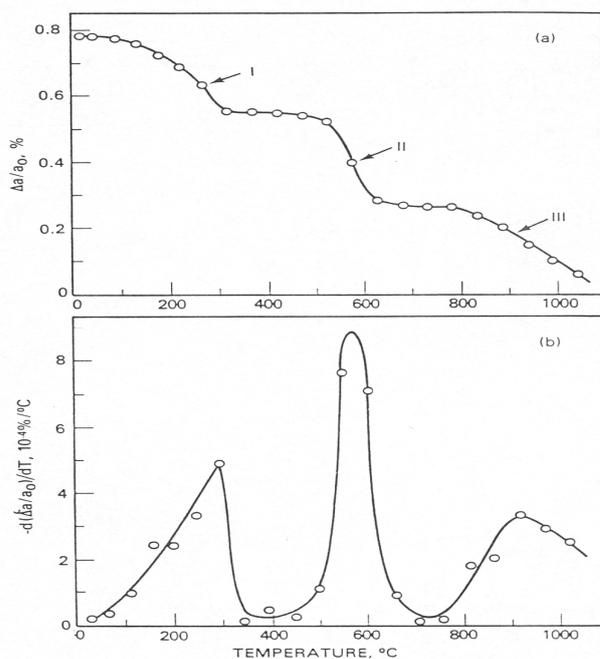


Fig. 5 Isochronal (a) and differential isochronal (b) recovery in UO_2 single crystal irradiated to 3×10^{20} alpha-particles/ m^2 . (copied from fig. 3 of Ref. [14]).

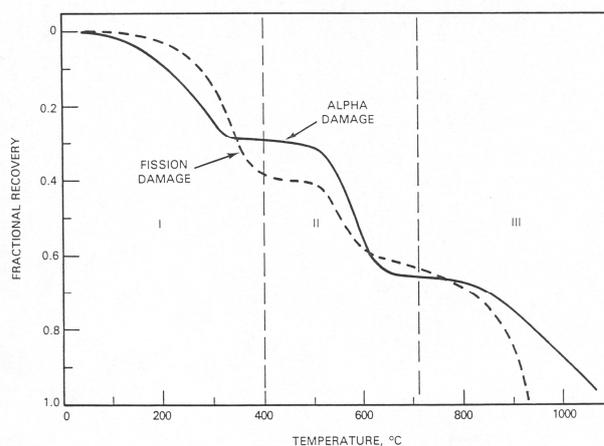


Fig. 6. Fractional recovery for both alpha-damaged and fission-damaged single crystals. (copied from fig. 7 of Ref. [14]).

The three isochronal recovery stages observed in the alpha-irradiated UO_2 single crystals correspond very well with the recovery stages observed in fission-damaged UO_2 single crystals. This is illustrated in Fig. 6. which compares the fractional recovery, after 24-hour anneals, of fission-damaged UO_2 single crystal irradiated to 5×10^{22} fissions/ m^2 with the fractional recovery of alpha-damaged crystals. These results clearly show that, although the magnitude of the lattice expansion in alpha and fission-damaged UO_2 is different, the recovery of the lattice expansion occurs by similar mechanisms independent of any difference in irradiation induced microstructures which may be present [14].

Presented above the three temperature stages of the isochronal recovery of the change in lattice parameter of a

UO₂ single crystal which correspond to the three temperature stages of noble gas burst release during annealing of both the minerals and the irradiated UO₂ samples, additionally supports the assumption that the gas release kinetics is determined by the kinetics of thermal recovery of the radiation induced defects.

Spontaneous fission fluency for the mineral uraninite of about 2 billion years old is about 10¹⁵ fissions/cm³ [5].

Using the data of spontaneous fission fluency for the mineral uraninite of about 2 billion years old we can make an assessment of irradiation damage in the mineral material containing fissile atoms by spontaneous fissions and by alpha decays of ²³⁸U in order to compare them.

The maximum specific amount of Frenkel pairs created by fission events (D_{fFr}) for the mineral uraninite of about 2 billions years old under simplified assumption that the damage is an additive process is:

$$D_{fFr} = F_1 N = 10^{15} \cdot 2 \cdot 10^5 = 2 \cdot 10^{20} \text{ Frenkel pairs/cm}^3 \quad (2)$$

where

F_1 - spontaneous fission fluency (F_1) for the mineral uraninite of about 2 billion years old,

N - maximum number of Frenkel pairs created by one fission event is $N = 200000$.

The maximum number of Frenkel pairs created by one fission event is $N = v/(2E_d)$, where E_d is the displacement threshold energy and v is the energy that each fission event delivers directly to the lattice of the irradiated material via atomic collisions which does not exceed 10 MeV. If $E_d = 25$ eV, then $N = 200000$.

However the specific amount of Frenkel defect pairs produced per alpha particle ($D_{\alpha Fr}$) for the mineral uraninite of about 2 billion years old under simplified assumption that the damage is an additive process:

$$D_{\alpha Fr} = 8 N_{\alpha} N_d = 8 F_1 / (Pr_f \cdot 10^{-2}) N_d = 8 \cdot 10^{15} / (5.4 \cdot 10^{-7}) \cdot 80 = 1.2 \cdot 10^{24} \text{ Frenkel pairs/cm}^3 \quad (3)$$

Where

- N_{α} is a number of alpha decays of ²³⁸U and is evaluated by dividing the spontaneous fission fluency (F_1) per fission probability per decay (Pr_f). So $N_{\alpha} = F_1 / (Pr_f \cdot 10^{-2})$.
- N_d is the average number of Frenkel defect pairs produced per alpha particle of energy 5.5 MeV. The value of N_d was experimentally determined to be 80 [14].
- (Pr_f) is fission probability per decay. Pr_f is equal to - 5.4 10⁻⁵ %.

In fact number of alpha decays is eight times as much as number of alpha decays of ²³⁸U because we have to add the alpha decays in the alpha chain decays of ²³⁴Th₉₀ of which half lives is far shorter than the 238 uranium half life.

So the irradiation damage for the uraninite mineral of two million years old formed in the result of alpha decay of uranium is about 10⁴ times higher than the irradiation damage of UO₂ fuel formed in the result of fissioning for the irradiation exposure 10¹⁵ fissions/cm³ under simplified assumption that the damage is an additive process.

Equalizing the amount of Frenkel pairs (2 10²⁰) formed by the fission fluency of 10¹⁵ fissions/cm³ to the equation of Frenkel pairs formed by alpha particles we can evaluate the mineral age for which we should expect to occur the peculiarity of equal diffusion of different noble gases during annealing.

$$D_{\alpha Fr} = 8 N_{\alpha} N_d = 8 C_{238U} [\exp(\lambda_{\alpha} t) - 1] \cdot 80 = 2 \cdot 10^{20} \text{ Frenkel pairs/cm}^3$$

$$t = \ln[2 \cdot 10^{20} / (8 N_d C_{238U}) + 1] / \lambda_{\alpha} \text{ years} \quad (4)$$

where

N_{α} is number of alpha decays of ²³⁸U:

$$N_{\alpha} = C_{238U} [\exp(\lambda_{\alpha} t) - 1]$$

C_{238U} - actual concentration of uranium atoms in the mineral, - uranium atoms/cm³,

λ_{α} - decay constant of uranium 238- 1.55 10⁻¹⁰ y⁻¹,

t - age of the mineral assumed according to the uranium-lead method - in years,

Considering the above equation for the uraninit K10y, 2 billion years old containing 71 wt% uranium [5] what equals 80.59 wt% UO₂, the actual concentration of uranium atoms (C_{238U}) is about 2.0 10²² uranium atoms/cm³. For this uraninite (K10y) the spontaneous fission fluency (F_1) is about 10¹⁵ fissions/cm³ (this will be proved later). Placing the presented here data into the equation (4) we get the hypothetical time of 100 800 years for which the amount of Frenkel pairs formed by alpha particles is equal to the amount of Frenkel pair formed by fission fragments.

Now then, the equivalent irradiation damage in the uraninite formed by alpha particles comparing with the fission damage in the UO₂ after irradiation of 10¹⁵ fissions/cm³ should be obtained after about 100 800 years what means that the radiogenic gas behavior in the minerals of 140 million years old (1400 times older) should be alike as the fission gas behavior in the fuel irradiated to the fluency of 4 10¹⁷ fissions/cm³. But this does not happen. Still the activation energies of krypton and xenon release during annealing at high temperature for young minerals (140 million years) as khlopinte and betafite are different according to the classical diffusion. It means that the same amount of damage caused by alpha decay of uranium is much less effective than caused by fission of uranium for the immobilization of noble gases. Both the alpha particles and the fission fragments form point defects and defect clusters of dislocation but exclusively the fission fragments create circumstances for irradiation induced chemical bonds of noble gases with the UO₂ matrix. It means further that the radiogenic gas release during annealing from the uranium minerals (where spontaneous fission fluency is about 10¹⁵ fissions/cm³) is alike the fission gas release from UO₂ fuel irradiated to about 10¹⁵ fissions/cm³. It implies further that the same mechanism both in the minerals containing fissile isotopes and in the UO₂ fuel is responsible for the attachment of the gas in the matrix.

It has to be remembered that the defects created by the fission fragments along the fission fragment trajectories and the strong thermal spikes, both in the UO₂ fuels and in the

minerals containing fissile isotopes, are of the same order. This is the link between fission gas behavior in the UO₂ fuels and in the minerals.

The examples of rare gas compounds presented in the ref. [11] show that noble gas chemistry is much richer than it would be expected. New chemical bonds between strange bedfellows, like noble metals, actinides and noble gases, can still be found.

Since the examples of rare gas compounds are formed by applying the classical chemical methods, the more the noble gas species in the conditions of neutron and fission fragments irradiation of the UO₂ fuel type can be expected [11].

This assumption is suggested by the fact that the ClXeCl has been found to form after irradiation with 501.7 nm laser light of Cl₂-doped xenon matrices. It appears that after excitation of the Cl₂ there is little or no barrier for the rearrangement to ClXeCl [12].

Knowing the fission probability per decay (Pr_f) and the number of alpha decays of ²³⁸U (N_α), the spontaneous fission fluency (F_i) is:

$$F_i = N_\alpha \times Pr_f \cdot 10^{-2} = C_{238U} [\exp(\lambda_\alpha t) - 1] \times Pr_f \cdot 10^{-2} \quad (5)$$

where

t - age of the mineral assumed according to the uranium-lead method – in years,

In order to evaluate the spontaneous fission fluency range for the minerals of about the same specific weight but of different age let us consider two following minerals:

- the uraninite from Webb Mine, North Carolina, 399 millions years old, of density 9.42 g·cm⁻³ contains 87.39 wt% UO₂ [15], what means that in cubic centimeter are about 2.17 10²² uranium atoms
- the uraninit K10y, 2000 millions year old contains 71 wt% uranium [5] what equals 80.59 wt% UO₂. This means that in cubic centimeter of the uraninite are about 2.0 10²² uranium atoms.

So after calculation it was found that the spontaneous fission fluency (F_i) for the uraninite of 400 million years and 2 billion years lies in the range from 7 10¹⁴ fissions/cm³ to 5 10¹⁵ fissions/cm³.

According to the assumption that Fig. 3 describes the probability of chemical interaction for UO₂ fuel in function of fission fluency which starts to be perceivable from about 7 10¹⁴ fissions/cm³ we can surmise that the minerals of less than 400 million years old are the minerals where the inert gases behave according the classical diffusion. This surmise agrees with the experimental data that for the young minerals (140 million years) as khlopinite and betafite the gas behavior during annealing obey the classical diffusion [5].

Research of irradiation induced chemical bonding process of noble gases in the irradiated UO₂ fuel after irradiation exposure in excess of 10¹⁵ fissions/cm³ is difficult for high radioactivity. This difficulty can be avoided by researching the samples of the minerals containing fissile atoms of about 2 billion years old of low radioactivity instead of the irradiated UO₂ fuel samples, which are highly radioactive. It gets out of the fact that there occurs an identical nature of noble gas

accumulation mechanisms both in the irradiated UO₂ fuel and in the old mineral materials containing fissile atoms.

Spontaneous fission fluency for the minerals containing fissile atoms of about 2 billion years old is above 10¹⁵ fissions/cm³. For example the spontaneous fission fluency of the uraninit K10y, 2000 million years old containing 80.59 wt% UO₂ is equal approximately to 5 10¹⁵ fissions/cm³.

Annealing the samples of minerals containing fissile atoms of about 2 billion years old at about 1400 °C we carry out the thermal recovery of radiation induced defects what actuates release small part of fission gas products from the sample but still holding significant part of the gas being chemically bound there. So prepared test samples permit the chemists to research the fission gas products chemistry in the UO₂ fuel after irradiation exposure in excess of 10¹⁵ fissions/cm³.

Proving existence of the irradiation induced chemical bonding process of noble gases we can replace the term <re-solution> with the term <irradiation induced chemical bonding process>.

IV. CONCLUSION

Both the old mineral materials containing fissile isotopes and the neutron irradiated uranium dioxide fuel hold fairly a lot of noble gases when the fission fluency of the minerals and the irradiated uranium dioxide fuel is in excess of 10¹⁵ fissions/cm³.

Alike the old minerals containing fissile isotopes and the irradiated UO₂ fuel reveal during annealing three peaks of gas release in the low temperature range. In the temperature range only several percent of the total amount of gas as well from the mineral materials and from the uranium dioxide fuel is released. It still holds significant part of noble gases after annealing at a temperature about 1400 °C. So, we can infer that increasing the temperature of old minerals during annealing over 1400 °C, we should observe the fourth peak of release as for the irradiated UO₂.

Noble gas atoms can be trapped by the point defects, clusters of the point defects, clathrates and small bubbles in the irradiated material.

It is suggested that above a fission fluency threshold about 10¹⁵ fissions/cm³ occurs an additional trapping process based on the irradiation induced chemical bonding process.

It is inferred further that as well in the minerals and in the irradiated UO₂ fuel the gas release kinetics is determined by the kinetics of thermal recovery of the radiation induced defects in the low temperature range.

It is surmised that the same amount of damage caused by alpha decay of uranium is much less effective than caused by fission of uranium for the immobilization of noble gases since only during fissioning there are formed conditions for strong chemical binding between the noble gas atoms and the mineral materials.

Finally it is concluded identical nature of noble gas accumulation mechanisms in the irradiated UO₂ fuel and in the old mineral materials containing fissile atoms.

This conclusion enables to avoid high radioactivity of the irradiated UO_2 fuel by using the old minerals containing fissile atoms in the research of the fission gas product chemistry.

REFERENCES

- [1] W.B. Lewis; Nuclear Applications, Vol. 2(1964)171 – 181.
- [2] J.R. MacEwan, H.W. Stewens; J. Nucl. Mater. 11, 1(1964)77
- [3] W.B. Lewis, J.R. MacEwan, W.H. Stevens, R.G. Hart; Proceedings of the 3rd U.N. Conference on the Peaceful Uses of Atomic Energy, Geneva, May 1964.
- [4] J.A. Turnbull; Radiation Effects, Vol. 53(1980)243-250.
- [5] I.M. Morozova, G.Sh. Ashkinadze; Migratsiya atomov redkikh gazov v mineralakh, Izdatelstvo "Nauka", Leningrad 1971.
- [6] K. Shiba; J. Nucl. Mater. 57(1975)271-279.
- [7] Y. Nogita, K. Une; J. Nucl. Sci. Tech. 30[9](1993)900.
- [8] D.A. MacInnes, P.W. Winter; J. Phys. Chem. Solids, Vol. 49, No 2(1988)143.
- [9] M. Szuta; J. Nucl. Mater. 210(1994)178.
- [10] K. Une, Sh. Kasibe; J. Nucl. Sci. Tech. 27(11)(1990)1002.
- [11] M. Szuta; Paper presented in the 6th International Conference on WWER Fuel Performance, Modelling and Experimental Support, 19 -23 September 2005, Albena Congress Center, Bulgaria.
- [12] C.R. Bieler, K.E. Spence, K.C. Janda; J. Phys. Chem. 95, 1992, p. 5058-5064.
- [13] J. Leteurte and Y. Quéré, Irradiation Effects in Fissile Materials; North-Holland Publishing Company – Amsterdam – 1972.
- [14] W.J. Weber; Thermal recovery of lattice defects in alpha-irradiated UO_2 Crystals; J. Nucl. Mater. 114(1983) pp.213-221.
- [15] J. Janeczek; Krystalochemia uraninitu I jego trwałość w warunkach redukcyjnych, Wydawnictwo AKAPIT-DTP, Sosnowiec 1993.