Role of Radiation-Enhanced Diffusion in Radial Distribution Formation of UO_2 Fuel Fission Products over the Pellet

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Abstract – It is shown, that the radial distributions of fuel fission products are formed basically as a result of radiation-enhanced diffusion, which in insulators does not depend on temperature (as against metals and semiconductors), see review [1]. Though the absolute value of radiation-enhanced diffusion factors are usually about 10^{-11} cm²/s, the strong migration takes place in accordance to mechanism of quasi-chemical reactions with ratios determined by a current of stimulating particles (products and neutrons), and also radiations of a wide spectrum (from sound up to γ). Possible application of our considerations on the mechanism of products radial distribution formation gives an opportunity of products structure control (with the purpose of fuel burning increase) by a method of the special impurities (together with burning absorbers).

1. Introduction

Creation of a fuel pellet for BWR, capable to keep products in own volume and having the large burning and power uniformity both on volume, and on time of a fuel cycle, is of significant ecological and economic importance. Burning absorbers are usually used to solve a part of these problems. However the tendency of products to concentrate at the border of a UO_2 pellet [2–4] is a limiting factor. The important problem is also increase of UO_2 fuel heat conductivity. In our previous paper [1], the radiation-enhanced diffusion laws in various materials are described, understanding of which is a key for the solving of above problem.

2. Theoretical model

The modified description [1] of process radiation-enhanced diffusion of isotopes, created on sufficiently long and intensive irradiation of solids by neutrons, have been presented in this paper. Along with diffusion transfer, the capture of an active impurity (index a) by vacancies (v) in a condition (c) and its activation by defects (d) is assumed. The important issue of the model is that diffusion occurs in the presence of intensive creation and disappearance of (d–v) pairs. These processes are described by the following system of quasi chemical reactions: a+v=c+phonon, d+c=a+phonon, d+v=2 phonons, and the connected system of kinetic equations written in cylindrical coordinates with a volumetric source. The particular approached solution of this system looks like: $(Na+Nc)=A_0+A_1r+A_2r^2+A_3r^3$, where $|A_2|=C_0/(4Dt)$, and $C_0=A_0+2A_1r_0/3+A_2r_0^2/2+2A_3r_0^3/5$, r_0 – pellet radius. The sense of other parameters is less obvious.

3. Results of the calculations for BWR fuel tests

We processed experimental data [5] on Xe concentration from pellet radius distributions as results of tests FK–1, FK–2 and FK–3. Initial burning and loading energy are given in the Table 1. The results of fit are given in the Table 2 and shown on the Fig. 1.

Table 1. Initial tests conditions

Test	GWd/tU	MJ	C_0, %
FK–1	45	111.2	0.9077
FK–2	45	62.2	0.9597
FK–3	41	111.2	0.9839

Table 2. Parameters fit results

Test	A_0	A_1	A_2	A_3
FK–1	0.5249	0.3864	5.1158	–7.7845
FK–2	0.6511	–0.0234	5.1158	–6.5524
FK–3	0.8340	1.0451	–5.1158	8.6977

From these data, the radiation-enhanced diffusion nature of products depth profiles is obvious.

4. Basis for a migration direction control

We processed experimental data of paper [6] on 300 keV Au^+ ions depth profiles determination when implanted into pure Cu and Cu–1 at.%Ni alloy at room temperature with dose $2\cdot10^{16}$ cm^{-2}, current density 10^{12} cm^{-2}s^{-1} under the angle of 80 to normal direction (see Fig. 2). From Fig. 2 the change of gold migration direction with respect to R_p is clearly seen at transition from pure Cu to Cu–1 at.%Ni alloy. The results of our calculations on model [1] basis are given in the Table 3. Proposed direction control rules are given in Table 4.

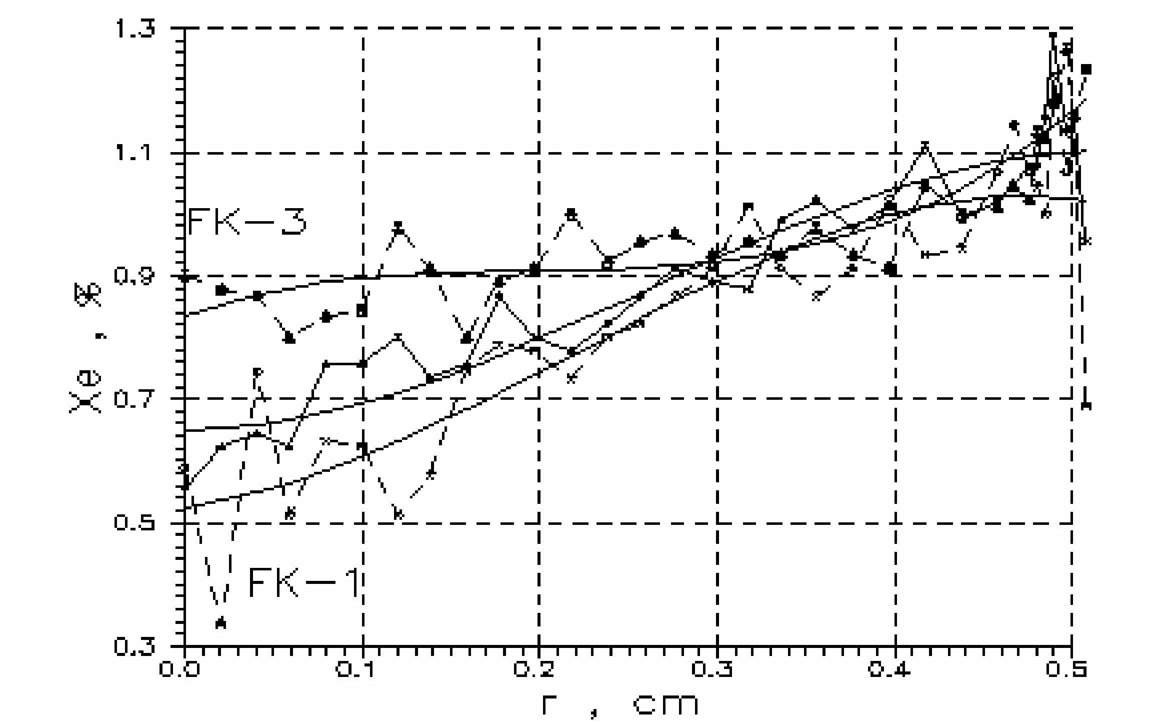

Fig. 1. Experimental points [5] and fit for Xe concentration position dependence after BWR fuel tests

Table 3. Depth profiles [6] by model [1] fit results

Parameter	Value unit	Value (for pure Cu)	Values ratio (Cu to alloy)
D_{dv}	10^{-11} cm^2/s	0.25126	1/2
D_a	10^{-11} cm^2/s	0.00283	5
k_{cap}	10^{-23} cm^3/s	0.69111	2/5
k_{ann}	10^{-23} cm^3/s	9232.99	5
k_{act}	10^{-23} cm^3/s	0.80218	20

Table 4. Direction management rules

Substance A and Z	Impurity A and Z	Ions A and Z	Direction
odd–odd	even–even	odd–odd	outside
		even–even	inside
even–even	odd–odd	odd–odd	outside
		even–even	inside
odd–odd	no	odd–odd	inside
		even–even	outside
even–even	no	odd–odd	inside
		even–even	outside

5. Discussion

All products are formed at fission of a compound nucleus (even even) and inherit its properties. In Fig. 3 the examples of products distributions from paper [3] are given. From a Fig. 3 the similarity of various products distributions to each other can be seen. Difference of structures at $r=r_0$ is due to surface of the sample that is well known for ion implantation into semiconductors. Data in Table 4 clearly shows the necessity of application of (odd–odd) impurity (more or near 1 at. % Al, V, Nb, Ta etc. or its mixtures) to change of a migration direction into (even–even) fuel pellet (UO_2 or U-Mo)

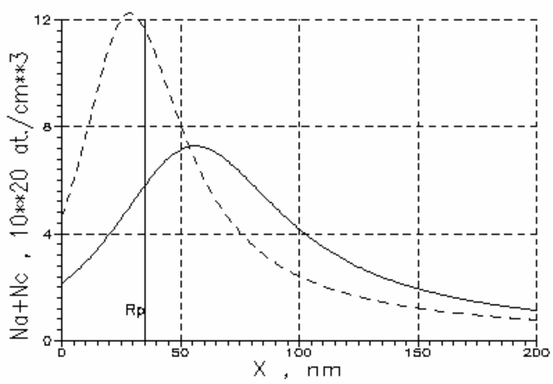

Fig. 2. Depth profiles of Au^+ ions implanted [6] into pure Cu (solid) and Cu–1 at.%Ni alloy (dashed)

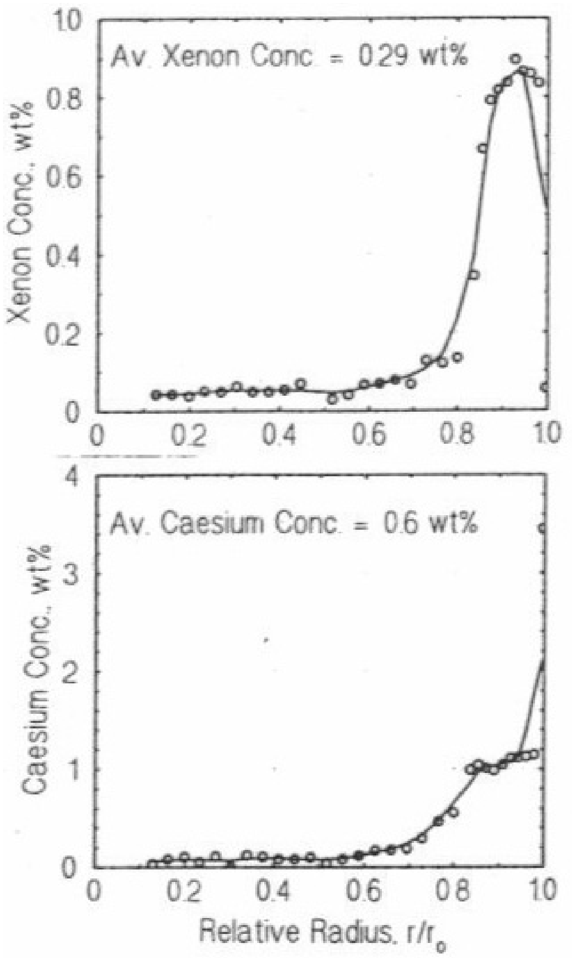

Fig. 3. Depth profiles of Xe and Cs products [3]

6. Conclusion

The radiation-enhanced diffusion nature of products depth profiles are proved. In our paper the question of products migration direction control is discussed. We propose to create BWR fuel pellet based on UO_2 matrix with the increased ecological safety and greater burning by means of modification by ~1 at.% (odd–odd) impurity (Al+V+Nb+Ta+etc.).

References

[1] V.A. Starostin, Nucl. Instrum. Meth. in Phys. Res., B, 242, 402 (2006).

[2] C.T. Walker et al., J. Nucl. Mater., 228, 8 (1996).

[3] C.T. Walker et al., J. Nucl. Mater., 129 (1995).

[4] C.T. Walker et al., Nucl. Eng. and Des., 1 (1991).

[5] T. Sugiyama, T. Nakamura, K. Kusagaya et al., *Behavior of irradiated BWR fuel under reactivity-initiated-accident conditions results of tests FK-1,-2 and -3., JAERI-Research* 2003-033.

[6] Y. Fujino, Y. Igarashi, and S. Nagata, Phys. Rev. B 63, 100101(R)(4) (2001).