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AN INFLUENCE OF POROSITY DEVELOPMENT IN U-Mo FUEL PARTICLES ON THE PERFORMANCE OF U-Mo/Al DISPERSION FUEL

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ABSTRACT

The post-irradiation examinations of the irradiated U-Mo/Al dispersion fuel for research reactors showed that its stable behavior under irradiation conditions depends on the ability of the U-Mo alloy to keep hold of gas fission products. However, the development of the porosity in U-Mo alloy particles and its relationship with the irradiation parameters were not paid the appropriate attention until the present time.
On the base of test results this work has revealed the relationship between the changes of the porosity in the U-Mo fuel particles and the irradiation parameters (temperature, fission rate and fuel burn up) and determined the limits of stable and unstable behavior of the U-Mo/Al dispersion fuel under irradiation conditions.

1. Introduction

The numerous irradiation and post-irradiation examinations revealed that a behavior of the U-Mo/Al dispersion fuel under irradiation conditions is determined by an existence of two regions. The fuel has stable and moderate swelling in the first region, whose limits are the complex function of a temperature, fission rate and fuel burn-up that determines the boundary of fuel performance. In the second region the fuel is subjected to an uncontrolled gas swelling that is inadmissible for a normal fuel elements (FEs) operation. However, the parameters of boundaries of a transition from one region to the other are not determined, that is the limiting factor for a U-Mo/Al fuel application in the FEs of high-flux research reactors. On one hand, it is due to the fact that the transition boundary is the complex function of a temperature, fission rate and fuel burn-up, on the other hand – there is no generally accepted criterion to characterize the swelling itself. Most of the investigations use the change of the thickness of a fuel plate or fuel rod diameter as a criterion. However, as the change in a thickness of FEs is a superposition of fuel swelling and thinning of fuel cladding due to corrosion, the critical value of fuel swelling is the variable quantity, that is not determined by the present time.

This work proposes to use the characteristics of the porosity, formed in fuel particles due to GFP formation during the FEs operation, as the criterion to characterize the FEs performance. This work presents test results of the changes in the porosity of the U-Mo fuel particles against the irradiation parameters (temperature, fission rate and fuel burn-up); the limits of a stable and unstable behavior of the U-Mo/Al dispersion fuel under irradiation conditions were determined.

2. Materials and test procedure

Test specimens were cut out from different locations in the fuel elements of 2 fuel assemblies (FA) designated "KM003" and "KM004" contained the U-Mo/Al dispersion fuel and which had been irradiated in the research reactor IVV-2M (Zarechny, Russia) [1, 2]. The in-pile test parameters and the characteristics of the investigated specimens regarding their location in the fuel element and positions relative to swelling in fuel claddings are shown in Table 1.

Sample #	FA #	Location in a fuel element	T_{BOL} [1)], ^{0}C	φ [2)], 10^{14} $f\cdot cm^{-3}\cdot s^{-1}$	Ψ [3)], 10^{21} $f\cdot cm^{-3}$	IL [4)], µm
101	KM003	Top	42.6	2.9	2.4	1.2
97	KM003	At a height of ¾ from FE bottom	55.0	4.1	3.4	2.5
96	KM003	Center	62.9	4.6	3.8	4.0
95	KM003	At a height of 1/2 from FE bottom	68.7	4.1	3.4	3.7
94	KM003	Bottom	69.5	2.8	2.3	3.4
135	KM004	Top	49.2	3.5	4.2	3.1
134	KM004	At a height of ¾ from FE bottom	66.7	5.1	6.1	6.4
13, 73	KM004	Center	79.3	5.9	7.1	12.4
74, 75	KM004	Near the center, swelling region	81.4	6.0	7.2	13.1
133	KM004	At a height of 1/2 from FE bottom	85.6	5.6	6.7	10.2
132	KM004	Bottom	84.4	4.2	5.0	9.1

1) – a fuel element cladding temperature at the beginning of the tests,
2) – a fuel fission rate,
3) – a fuel fission density,
4) – a thickness of the layer $(UMo)Al_x$.

Table 1: Characteristics of Specimens

The distribution of the GFP gas bubbles in the fuel particles was studied with a scanning electron microscope (SEM) at accelerating voltage of 30 kV and at magnification x 3000. The distribution of the gas bubbles in the volume of the fuel particles was analyzed by using the stereometric metallography methods developed by S.A. Saltykov [3].

3. Results and discussion

All data on characteristics of gas bubbles in the fuel particles, which were obtained in this work from samples ## 75, 94, 95,132 and 133 and also in works [4, 5] were analyzed and given in plots in Fig. 1. This Figure shows the curves of changing concentration of gas pores (*N*), the mean diameter (*D*) and its specific volume (*V*) against the fuel fission density (Ψ).

In accordance with the results of in-pile tests and post-irradiation examinations of fuel elements of FA KM003 and KM004 [1, 2], the whole field of fuel fission density Ψ, observed in the different FEs areas can be divided into 3 regions designated A, B and C that distinguished by a swelling rate and fuel-meat damage swelling rate. In the first two regions A and B there is moderate FEs swelling. Moreover, in the fuel meat matrix in A region there are no any defects that relate to the FEs operation conditions. While in the fuel-meat matrix in B region there were revealed the defects in the form of individual gas pores at the boundary of “interaction layer-aluminum matrix” (Fig. 2.*a*) and micro cracks of 0.2-1.2 mm long (Fig. 2.*b*). C region is characterized by a high rate of fuel element swelling with a macro cracks formation and fuel cladding swelling with no breaking their integrity (Fig. 2.*c*). Thus, B region is an intermediate region between A region with a moderate FEs swelling rate and C region with a higher, uncontrolled (abnormal) swelling rate of FEs with the U-Mo/Al dispersion fuel, inadmissible for the operation of standard FEs.

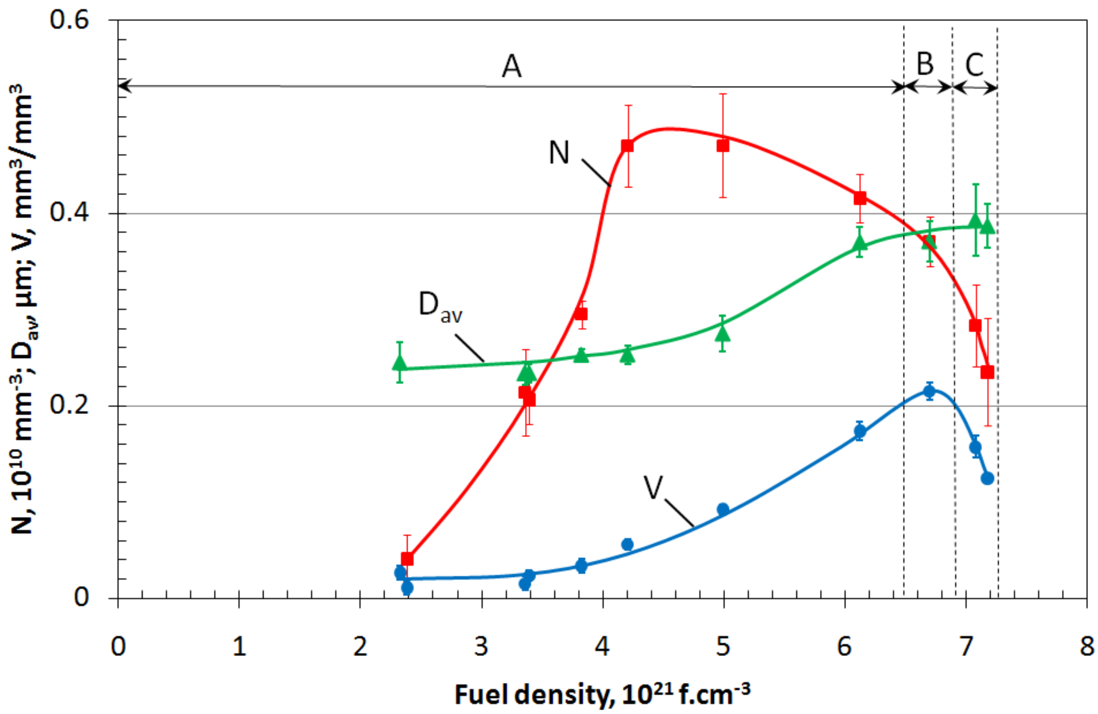

Fig. 1. The curves of changing concentration (*N*), mean diameter (*D*) and specific volume (*V*) of gas bubbles in fuel particles against the fuel fission density

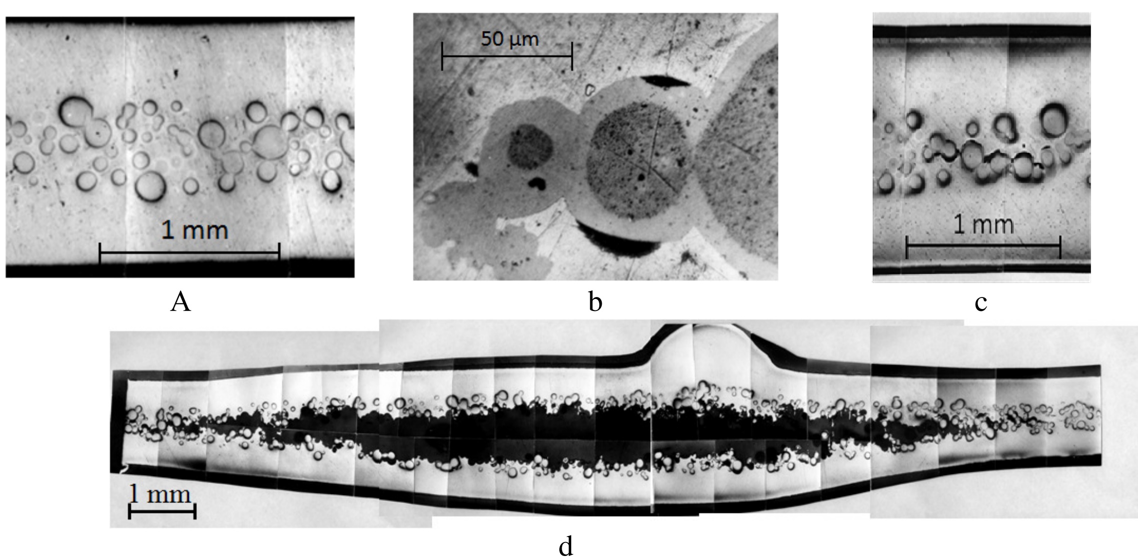

Fig. 2. The fuel-meat structure of the FEs in FA KM004 in the regions of fuel fission density A (*a*), B (*b, c*) and C (*d*)

The boundaries of each region are the complex function of a temperature, fuel fission rate and in – pile exposure time. For the exposure conditions typical for the FEs of FA KM003 and KM004, the following boundaries of the fuel fission density can be defined for each region:

- $0<\Psi<\sim 6.5\cdot 10^{21}$ f.cm^{-3} for A region;
- $\sim 6.5\cdot 10^{21}<\Psi<\sim 6{,}9\cdot 10^{21}$ f.cm-3 for B region;
- $\Psi>\sim 6.9\cdot 10^{21}$ f.cm^{-3} for C region.

In the range of values from 0 to $\sim 4.2\cdot 10^{21}$ f.cm^{-3} there is a steady accelerated increase in the concentration of pores in the fuel particles up to $N = \sim 5\cdot 10^{9}$ mm^{-3} (Fig. 1). The mean velocity of the gas bubbles formation in the range of changing Ψ from $\sim 2.4\cdot 10^{21}$ to $\sim 4.2\cdot 10^{21}$ f.cm^{-3} equals to 2.4 pores per $1\cdot 10^{12}$ f.cm^{-3}. The increase Ψ from $\sim 4.2\cdot 10^{21}$ to $\sim 5\cdot 10^{21}$ f.cm^{-3} does not influence the concentration of pores (Fig. 1), while the further increase of the fuel fission density leads to the decrease of the pores concentration. In addition, in the region of the values $\Psi>\sim 6.7\cdot 10^{21}$ f.cm^{-3} there is a rather steep curve of the decrease in the pores concentration. The mean velocity of gas bubbles concentration in the range of changing Ψ from $\sim 5\cdot 10^{21}$ to $\sim 6.7\cdot 10^{21}$ f.cm^{-3} equals to 0.6 bubbles per $1\cdot 10^{12}$ f.cm^{-3} and in the range $\Psi=(6.7\div 7.2)\cdot 10^{21}$ f.cm^{-3} – 2.8 pores per $1\cdot 10^{12}$ f.cm^{-3}.

Such a behavior of gas pores concentration in the fuel particles can be explained as follows. With the increase of the fuel fission density, the formation of new gas pores, a growth of already formed pores and an absorption of small gas pores by the large ones due to a dissolution of small bubbles and also a coalescence of 2 and more bubbles in one large pore occurred. Moreover, in the region of abnormal swelling the GFP release from the fuel particles into the interaction layer and aluminum matrix is possible.

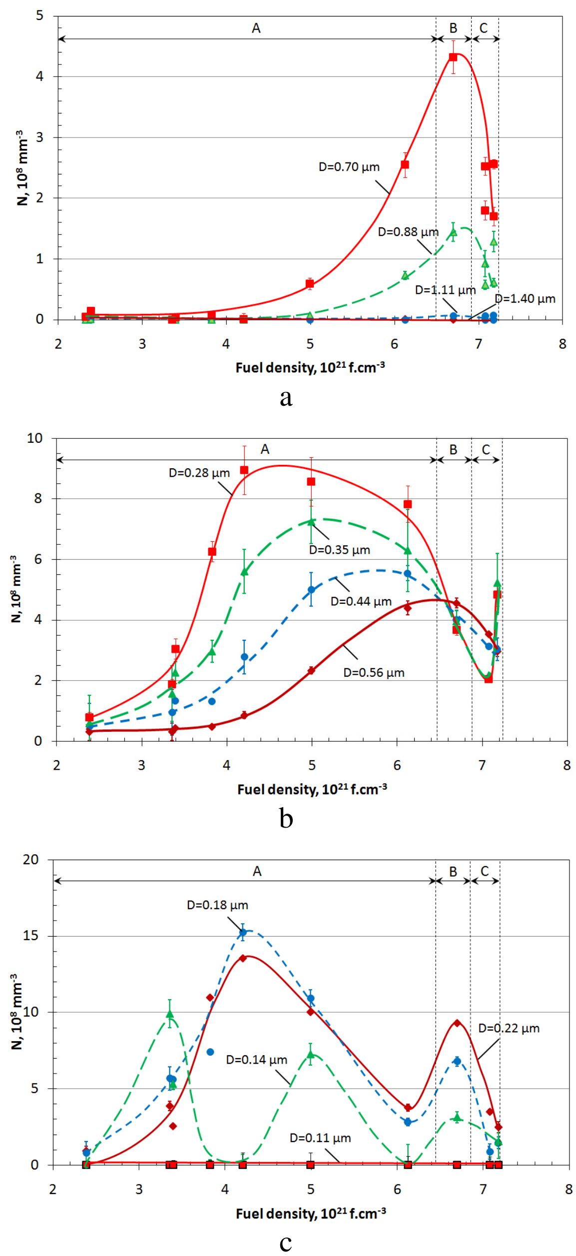

Fig. 3. The curves of changing concentration $N(D_i)$ of the gas bubbles of a particular size D_i in the fuel particles against the fuel fission density

The total amount of pores in the fuel particles is a poly-dispersed system consisted of 12 groups of pores with different diameter D_i and each of them has its own dependence of changing gas bubbles concentration on the fuel fission density $N(D_i)=f(\Psi)$ (Fig. 3). A typical feature of all groups of gas bubbles is an extreme behavior of the dependences $N(D_i)=f(\Psi)$ with the maximum position, depending on the bubbles' sizes, in *Ψ* direction. (Fig. 2). It was revealed that with the increase of the bubble diameter D_i there is the shift of the maximum $N(D_i)=f(\Psi)$ to the larger values of *Ψ*. Moreover, for the bubbles of $D_i<0.27$ there is a poly-extreme behavior of the dependences $N(D_i)=f(\Psi$, *i.e.*: with 2 maxima for the bubble diameters of 0.18 and 0.22 µm and 3 maxima for the bubble size of 0.14 µm (Fig. 3.*в*).

The dependence of a change in a specific volume of gas bubbles in the fuel particles on the fuel fission density is the similar to the dependence $N=f(\Psi)$ with the only difference that the maximum of the dependence $V=f(\Psi)$ is shifted to the region of higher values of the fuel fission density $6.7\cdot10^{21}$ f.cm^{-3} (Fig. 1). With the further increase of the fuel fission density, the specific volume of the gas bubbles in the fuel particles decreases, that is associated with a partial GFP release into the interaction layer and Al matrix to form discontinuities as large pores, micro and macro cracks. The maximum of the dependence $V=f(\Psi)$ equals to 0.215 mm^3/mm^3 and corresponds to the value *Ψ* in the intermediate region B of value *Ψ*.

The mean diameter of bubbles steadily increases practically in the whole range of fuel fission density values (Fig. 1), and becomes the plateau at the values *Ψ* corresponding to the boundary of transition from A region to B region.

One of the typical features of the changing porosity of the fuel particles is the manner of the concentration distribution curves and the specific volume of the gas bubbles against their size (Fig. 4). A form of the dependences $N=f(D_i)$ and $V=f(D_i)$ correspondent to a normal Gauss distribution with only one extremum (Fig.4.*a*) is typical for the region of FEs with normal swelling of the fuel meat. It means that the bubbles mostly related to one and the same poly-dispersed system are formed in the region with the normal swelling of fuel meat in the fuel particles.

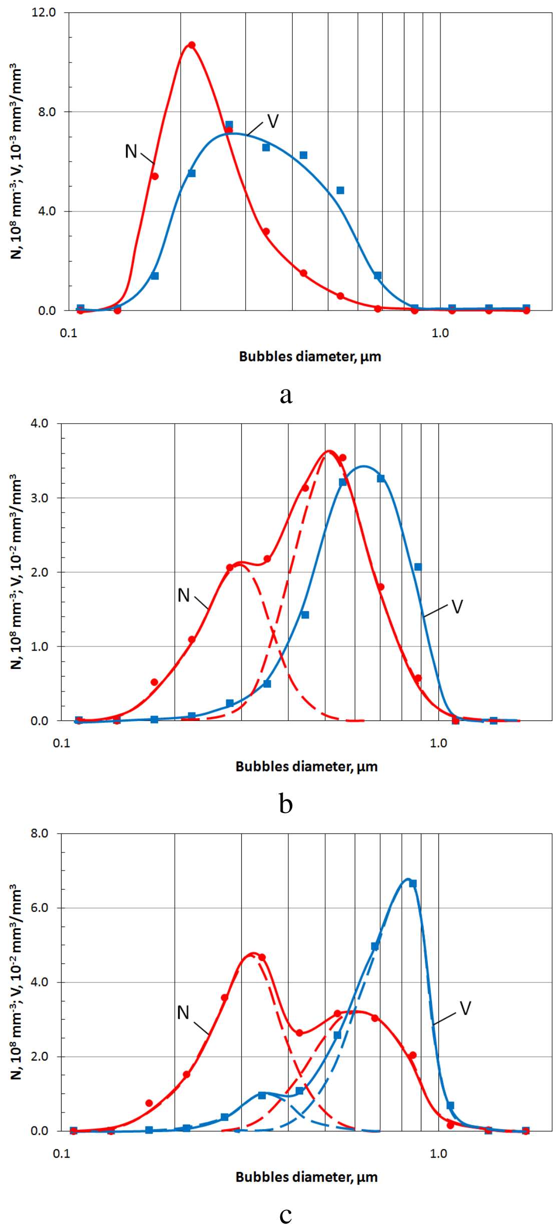

Fig. 4. The plots of dependences $N=f(D_i)$ and $V=f(D_i)$ of gas bubbles in the fuel particles of fuel meat in the regions A (*a*), B (*b*) and C (*c*)

The shift of the maximum of the dependences $N=f(D_i)$ and $V=f(D_i)$ to the lager sizes of the bubbles with the increasing fuel fission density is a typical feature of the dependence of bubbles distribution on the diameter in the A region (Fig. 4.*a*). In the intermediate B region the deviations from a normal Gauss distribution on the curve begin to reveal themselves and mostly do in C region, that refers the dependence $V=f(D_i)$ as well. Such changes in the forms of the curves $N=f(D_i)$ and $V=f(D_i)$ are stipulated by the formation of the second system of the bubbles with smaller sizes (Fig. 4.*b,c*).

Thus, the analysis of the dependences $N=f(\Psi)$ and $V=f(\Psi)$ shows that the specific volume of gas bubbles in the fuel particles can be a generalized parameter, the values of which can be used for determining the safety limits on the fuel-element operational parameters such as a operation time, temperature, fuel fission rate and density (or burn-up).

The gas in the bubbles is under the pressure induced by the forces of surface tension in the U-Mo alloy, the forces tend to decrease the size of the bubbles. The condition of the gas in the bubbles is determined by the equilibrium between the surface tension forces as well as the forces related to the elastic deformation of the alloy U-Mo and the internal pressure and is described by the following expression

$$(p + p_m + 2\sigma/r)V = nkT, \qquad (1)$$

where r – the radius of a pore; p_m – the pressure, induced by a resistance of matrix; σ – the surface tension; n – the number of gas atoms in a bubble; k, T – the Boltsman constant and the absolute temperature, respectively.

Fig. 5 illustrates the fuel fission density dependencies of the characteristics as follows:

- the specific volume of gas pores (V_B) formed in 1 mm^3 of the fuel particles;
- the gas volume (V_{GP}) in pores;
- the total volume of the gas fission products (V_{GFP}) formed in fission of uranium - 235 in 1 mm^3 of the U-Mo alloy.

The V_{GFP} - values were calculated by using the estimated value of the volume of the GFP formed in fission of 1 g ^{235}U, the volume being equal to 24.7 cm^3 [6]. The gas volume in the bubbles was calculated from expression (1) using the values p_m and σ for uranium, which equal to 1 Pa and 7 MPa, respectively, in a temperature range of 500 to 600 ^{o}C [7].

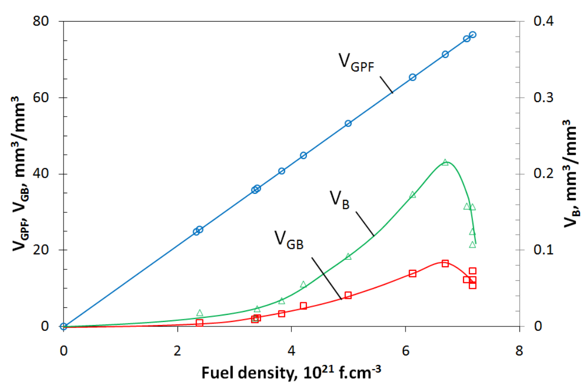

Fig. 5. Changes of the total GFP volume (V_{GFP}), formed from a fission of 1 mm^3 - volume fuel, the GFP volume in gas bubbles (V_{GP}) in fuel particles of 1mm^3 - volume and specific volume of bubbles (V_B) against the fuel fission density

These estimates show that the GFP fraction in the gas bubbles of the fuel particles reaches its maximum value of 20-23 % at Ψ=~$6.7\cdot10^{21}$ f.cm^{-3} with increasing fuel fission density. However in C region of abnormal swelling with the further increasing of Ψ value (at Ψ>~$6.7\cdot10^{21}$ f.cm^{-3}) the fraction decreases to 12-15 %.

Thus, the specific volume of the gas bubbles in the fuel particles can be the generalized parameter, whose values can be used for determining the safety limits on fuel-element operational parameters such as a temperature, fission rate and fission density (burn up) of fuel.

A mathematical treatment of the data on changes in the specific volume of gas pores in the fuel particles for the region of normal fuel swelling gives the following expression:

$$V = 2{,}217\cdot10^{-37}\cdot\varphi^{0.995}\cdot t^{3.408}\cdot\exp(-2365/T_{BOL}), \quad (2)$$

where V - the specific volume, mm^3/mm^3; φ – the fuel fission rate, $f.cm^{-3}\cdot s^{-1}$; t – time, sec; T_{BOL} – fuel cladding temperature at the beginning of tests, Kelvin.

Taking V_{cr}=0.215 mm^3/mm^3 as a critical value that equals to the maximum value of the specific volume of bubbles in the intermediate B region and substituting it into expression (2), one obtains the expression to estimate the critical value of fuel fission density (or burn-up fuel) corresponding to the upper safety limit on the fuel-element operational parameters such as a fuel temperature and fission rate.

$$\Psi_{cr}=3{,}61\cdot10^{10}\cdot\varphi^{0.71}\cdot\exp(695/T_{BOL}). \quad (3)$$

The fuel-element operation with the intermediate swelling rate is possible and do not have an effect on this operation with the calculated values of $\Psi_{calc.}<\Psi_{cr}$. At $\Psi_{calc}\geq\Psi_{cr}$ there is a strong possibility of FEs abnormal swelling that can provoke the negative effects.

It should be noted that the expression (3) was obtained for the parameters of in-pile tests of FEs FA KM003 and KM004, so it is possible to use it within the limits: from 40 to 90 °C for the fuel cladding temperature at the beginning of tests; $2.5\cdot10^{14}$ to $6.0\cdot10^{14}$ $f.cm^{-3}\cdot s^{-1}$ for the fuel fission rate; $2.3\cdot10^{21}$ to $7.2\cdot10^{21}$ $f.cm^{-3}$ for the fuel fission density.

The applicability of expression (3) was checked for the IRIS-1 test, where the abnormal fuel-element swelling with fuel fission density ~$5.0\cdot10^{21}$ $f.cm^{-3}$ has been revealed at the temperature T_{BOL} = ~75 °C and fuel fission rate $3.4\cdot10^{14}$ $f.cm^{-3}\cdot s^{-1}$. The calculated value Ψ_{cr} from expression (3) for the IRIS-1 test equals to ~$5.17\cdot10^{21}$ $f.cm^{-3}$, i.e. there is a good fit between the observed and the calculated values Ψ_{cr}.

4. Conclusion

The volume bubble distribution of gas fission products in the U-Mo fuel particles, irradiated in the range of fuel fission density from ~$2.3\cdot10^{21}$ to ~$7.2\cdot10^{21}$ $f.cm^{-3}$ in the regions with normal and abnormal fuel meat swelling, was investigated.

It has been revealed that the changes in the concentration, mean diameter and specific volume of gas bubbles against a fuel fission density (burn up) have extreme dependencies and the mean diameter of the gas bubbles increases practically linearly from 0.17 to 0.45 µm till the transition to the abnormal swelling region, where it keeps practically the same or increases.

Gas pores concentration increases in a linear fashion by an order of magnitude from ~$4.1\cdot10^{8}$ to $4.9\cdot10^{9}$ mm^{-3} with increasing fuel fission density from ~$2.4\cdot10^{21}$ to ~$4.2\cdot10^{21}$ $f.cm^{-3}$ and mean velocity of 2.4 pores per $1\cdot10^{12}$ $f.cm^{-3}$. In the region of fuel fission density values from ~$4.2\cdot10^{21}$ to ~$5\cdot10^{21}$ $f.cm^{-3}$ pores concentration keeps practically the same. If Ψ>~$5.0\cdot10^{21}$ $f.cm^{-3}$, the bubbles concentration decreases with the mean velocity of 0.6 pores per $1\cdot10^{12}$ $f.cm^{-3}$ in the range of Ψ from ~$5\cdot10^{21}$ to ~$6.7\cdot10^{21}$ $f.cm^{-3}$ and 2.8 pores per $1\cdot10^{12}$ $f.cm^{-3}$.

The specific volume of gas pores in the normal swelling region of the fuel meat increases with some acceleration and reaches the maximum value of ~0.215 mm^3/mm^3 at the boundary of transition to the region of abnormal swelling. In this region the specific volume of gas pores decreases and the degree of this reduction depends on the fuel particles position relative to defects in the fuel meat.

The experimental expression has been obtained to estimate the critical value of fuel fission density corresponding to the upper safety limit on fuel-element operational parameters such as fuel temperature and fission rate.

5. References

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