Results of PIE of experimental (U-Mo)-based LEU disperse fuel compositions

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Abstract

Performed in frames of Russian RERTR Program were post-irradiation examinations of 12 types of disperse (U-Mo)-based fuel compositions in Al matrix. The goal of the work was to substantiate serviceability of the new fuel compositions being developed for utilization in Russian-built pool-type research reactors. Among the methods of PIE used in this work were visual examination, gamma-scanning, optical metallography, SEM, X-ray analysis and some others.
The present paper is dedicated to an analysis of the main results of PIE completed so far. Special attention has been paid to the parameters affecting formation of interaction layer between fuel granules and matrix Al.

Introduction

The present paper is dedicated to an analysis of the results of PIE of disperse (U-Mo)-based fuel compositions in Al matrix intended for the conversion of Russian pool-type research reactors to LEU high-density fuel. In all tested in MIR reactor were 12 types of fuel compositions. On the results of PIE the most prospective fuel composition was selected for further R&D activities in frames of Russian RERTR Program. Besides, some regularities of irradiation behavior inherent in the tested fuel compositions were revealed and explained including the phenomenon of gaseous fission products accumulation at the boundary of interaction layer and matrix.

1. The make-up of irradiation vehicle and irradiation parameters.

To manufacture (U-Mo)-based LEU disperse pin-type fuel mini-elements for irradiation tests the set of U-Mo alloys with various Mo content has been smelted. Then the alloys have been reduced in size by two different methods to obtain fuel granules in the dimensional range (63÷160 μm). As a result spherical fuel granules and fuel granules of irregular shape were manufactured by atomization and hydride-dehydride processing accordingly (fig. 1a, c). And then test specimens with various types of fuel compositions were fabricated.
At present time in frames of Russian-built research reactors conversion to LEU, the development of new (U-Mo)-based dispersion composition is accompanied by the development of the novel unified pin-type design of fuel elements. Such fuel elements are supposed to replace currently used tube-type ones. Therefore the reduced-length prototype of full–sized pin-type fuel element was chosen as a test specimen design [1].
The design of irradiation vehicle [1] permitted simultaneous testing of 72 pin-type fuel mini-elements. In all tested were 12 types of (U-Mo)-based dispersion compositions differentiating on the following parameters [2]:

a) Mo content in U-Mo alloy:
   - U-9,0 wt % Mo

- U-6,5 wt % Mo .
- U-1,5 wt % Mo

b) Phase composition of fuel granules prior to FE manufacture:
- γ-phase (single phase alloy)
- (α+γ)-phase (two-phase alloy)
- α'- phase (with regard only to alloy U-1,5 wt % Mo)

c) U_{tot} concentration per volume unit of fuel meat:
- 6 g/cm^3
- 4 g/cm^3

d) Presence of alloying additions:
(combination of small additions of Al and Sn (in the range 0,2 ÷ 0,4 wt%))

e) Method of fuel granules manufacture [2] :
- Atomization (spherical shape of fuel granules)
- Hydride-dehydride processing (irregular shape of fuel granules)

Thus several goals have been pursuited: to determine the effect of phase composition, Mo content, method of fuel granules fabrication as well as uranium concentration upon the irradiation behavior of (U-Mo)-based disperse compositions. By alloying with small additions of Al and Si it was proposed to create the microstructure of fuel granules containing a lot of small disperse precipitations which would provide the uniform distribution of gaseous pores in the fuel granules interior.

The parameters of irradiation in MIR reactor were typical for Russian pool-type research reactors:

- Coolant temperature at an inlet ~ 60 °C;
- Coolant rate in the range 2,8 ÷ 3,3 m^3/h
- Peak heat flow – 0,6 ÷ 0,9 MWt/m^2
- Density of neutron flux (E>0,1 Mev) – (2,0 ÷ 7,0) 10^{13} cm^{-2} sec^{-1}

2. The methods of PIE

Irradiation tests were conducted to the average burnup ~ 65% with interim selection of test specimens of each type fuel composition after 20% and 50% burnup. PIE comprised visual examination, gamma-scanning, optical metallography, planimetry analysis (measurements of swelling), SEM, X-ray analysis.

3. Results of post-irradiation examination.

3.1 Conclusions from the results of irradiation tests as a whole.

The irradiation tests conducted in MIR reactor as a whole testify about the stable irradiation behavior of all studied types of fuel compositions up to burnup ~ 65%:

- An examination of FE appearance didn't reveal any indications of loss of leak-proofness for cladding: visible cracks or pillowing is absent.
- Gamma-scanning testifies about sufficiently uniform energy release and fission products distribution through the length of fuel meat.
- The study of microstructure didn't reveal the presence of micro-cracks or splitting in the fuel meat at all stages of PIE.
- Irrespective of the method of fuel granules fabrication interaction layers are localized around fuel particles (fig. 1) i.e. their thickness is small and thus the large areas of so-called "continuous interaction" are absent.

Talking as a whole about the stable behavior of all studied dispersion compositions at the irradiation parameters typical to low flux research reactors, it is necessary to note some differences in their irradiation behavior as well as some negative results of PIE which nevertheless didn't result in premature failure of test specimens.

3.2 Formation of interaction layers around fuel particles

Formation of interaction layers around fuel granules as a result of interaction between (U-Mo) fuel granules and Al matrix is a typical feature for all tested fuel compositions (fig. 1b, d). During the last years this phenomenon was being thoroughly studied by the majority of

researchers being engaged in RERTR activity [3, 4, 5, 6]. In their experiments at more severe irradiation conditions the formation of large areas of interaction resulted in gross pore formation at a boundary between interaction layer and Al matrix, which in turn led to FE pillowing and loss of sealing. The most probable cause of gross pore formation apparently was the process of gaseous and highly mobile fission products diffusion through the interaction layer and their accumulation at its boundary.

3.2.1 Formation of a gaseous porosity at a boundary between interaction layer and Al matrix.

The above-mentioned process of gaseous and highly mobile fission products accumulation at the boundary between interaction layer and Al matrix was revealed in the microstructure of the dispersion composition irradiated to 50% burnup. The concentration maps for the distribution of Xe^{133} and Cs^{137} in the fuel meat after 50% burnup shown in figure 2 corroborate this approach. As can be seen the foregoing fission products outside the fuel particles concentrate mainly at the boundary between interaction layer and matrix. Also it is necessary to mark the irregular (non-uniform) distribution of fission products around an interaction layer (figure 5 Xe^{133}) and possible Xe peak concentration around interaction layer exceeding over the uniform concentration in the fuel particle interior.

Shown in figure 3 is, seemingly, the initial phase of the process of gross pore formation which has been revealed in the microstructure of the only one of twelve studied types. It was the fuel composition on the base of the uranium alloy with 9 wt% Mo and the highest uranium concentration – 6 g/cm^3.

3.2.2 The size of interaction layer.

The thickness of interaction layer forming around fuel granules is an important parameter being a characteristic of (U-Mo)-based disperse fuel irradiation behavior. Therefore its control makes it possible to estimate the prospectives of this or that dispersion composition. The result of measurements of interaction layer thickness for 50% and 65% burnups are plotted in figure 4 against fission density of U-Mo alloy. This diagram points out a good correlation between interaction layer thickness and the type of fuel composition:

-Irrespective of fission density the largest thickness of interaction layer corresponds to fuel compositions with the highest uranium concentration per volume unit of either fuel meat (6 g/cm^3) or U-Mo alloy (alloy with 1,5%Mo) (the points in the top rectangular area).

-Irrespective of fission density the lowest thickness of interaction layer corresponds to fuel compositions on the basis of two-phase ($\alpha+\gamma$) uranium alloys with 6,5% and 9% Mo (the points in the bottom rectangular area).

It's important to underline the fact that the lowest thickness of interaction layer corresponds to fuel compositions in which the fuel granules prior to fabrication of fuel elements had been converted to ($\alpha+\gamma$)-two-phase state by special heat treatment.

3.2.3 The structure of interaction layer.

The typical features of interaction layer microstructure (for all studied fuel compositions with the exception of that on the basis of alloy U-1,5 wt% Mo) are their visual uniformity and the absence of gaseous pores and any other micro-structural characteristics and peculiarities. Dispersion composition on the basis of alloy U-1,5 wt% Mo turned out to be the only one which revealed non-uniform structure of interaction layer (fig 5a). One can distinguish at least two different areas: darker internal layer containing some gaseous bubbles and bright external one devoid of gaseous bubbles and therefore similar to that of all other studied compositions. The possible explanation of this phenomenon has much to do with the microstructure of fuel compositions prior to irradiation. The pertinent composition is the only one in which the thin interaction layer around fuel granules was being formed during FE fabrication. Its thickness is comparable with that of internal one after irradiation. Apparently it signifies that the internal layer in figure 5a had been formed most probably as a result of pre-irradiation interaction between fuel particle and matrix, while the external one – under irradiation. Thus the presence or absence of gaseous pores is associated with the state of interaction layer microstructure: whether it is crystalline or amorphous.

3.2.4 Conglomerations of fuel granules and their microstructure at an area of contact.

The reduction of enrichment inevitably results in an increase of volume fraction and thereby in formation of large local conglomerations of fuel granules when several particles contact each other. The typical microstructures of fuel granules being in contact and differentiating on Mo content are shown in figure 6. As it can be seen the microstructure in a contact area for fuel composition with 1.5 wt % Mo significantly differs from that of all other alloys.

In the case of 6,5 % Mo and 9,0 % Mo one can observe distinct boundary between individual fuel granules both after etching and polishing and interaction layer forms only around the contact area. Supposedly, at further irradiation due to processes of radiation sintering and creep the boundary will transform into so-called "monolithic structure" when initially separate fuel granules merge into foregoing structure.

In the case of 1.5 % Mo the contact area significantly differs: the boundary between fuel particles is absent and there are traces of interaction layer inside the contact area. Presumably such difference is associated with either the α'-phase composition of this alloy or with the presence of interaction layer formed during fabrication process (see section 3.2.3)

3.4 Metallography of a cladding

Microstructure of FE cladding in a cross section after burnups ~ 20%, 50% and 65% testifies about an absence of continuous oxide film at the surface of FE cladding. At the same time several micro-cracks located at the corners of FE cross-sections near the fins were revealed in a microstructure of some mini fuel elements (figure 7). Their size was of an order from tens to hundreds of microns. They were located in diagonal direction. This notwithstanding they didn't affect the serviceability of tested fuel elements.

3.5 Planimetry analysis

The swelling of tested fuel compositions has been evaluated on the results of dimensional changes of mini-fuel elements either "on fins" or "on sides" of square (the former means measurement of FE circumferential diameter, and the latter – measurement of face-to-face distance of the cladding). With this the dimensions of either relevant un-irradiated fuel-elements or "idle" end parts of fuel element (i.e. the part of FE without fuel) were taken as zero point. To determine the main components of swelling the relationships of swelling against attained fission density and against the volume fraction of interaction layer have been plotted (figures 8 and 9 correspondingly). However in both cases no correlation was revealed. Possibly the absence of such correlation is the result of either too many variables affecting the swelling or some other reasons having nothing to do with the processes of micro-structural defects accumulation and growth of interaction layer (for example some phase transformations etc.).

So, if one should distinguish the points relating to fuel compositions alloyed with small additions of Al and Sn (white triangles) then it turns out to be that the majority of these points are located in the upper part of diagram, thus breaking the monotonic trend of swelling increase with burnup growth.

Probably this phenomenon is associated with formation of inter-metallic compounds between small additions of Al and Sn and U or Mo in fuel granules. If such compounds do form, then most probably this process has to proceed with volume increase (as in the case of UAl_2, UAl_3 and UAl_4 , as well as U_5Sn_4). In this case such compounds has to make additional contribution to FE swelling.

4. Discussion.

As it was mentioned above in section 3.1 the results of PIE as a whole testify about stable irradiation behavior of all fuel compositions tested in MIR reactor up to 65% burnup at the parameters typical to Russian pool-type research reactors. For the further investigations with regard to more severe conditions of high flux reactors, the uranium alloy U-9 wt% Mo has been selected as the most studied one and thermodynamically stable being in contact with Al matrix.

Among the positive results of PIE it is necessary to mark the stable irradiation behavior of the fuel compositions on the basis of uranium alloys with lower Mo content which are more adaptable to manufacture due to lower volume fraction of fuel. In this connection it is especially important to underline the stable irradiation behavior of fuel composition with U-1,5 wt % Mo fuel granules alloyed with small additions of Al and Sn. Introduction of small alloying additions

possibly had the positive effect upon its irradiation behavior thus escaping the formation of big gaseous bubbles inside the fuel granules. Besides, the inclination of these alloy to formation of interaction layer during manufacturing possibly turned out to be an obstacle in formation of gross pore formation at the boundary of interaction layer and Al matrix. Therefore this fuel composition also has to be considered as a perspective one under the conditions of Russian pool-type research reactors.

Talking about perspectives of fuel composition on the basis of U-1,5%Mo-(Al;Sn) it is also necessary to add that among all fuel compositions alloyed with small additions of Al and Sn the swelling of relevant one was one of the smallest (in the range from 2% to 3% for both values of burnup). Thus in contradistinction to γ-phase alloys, where the alloying resulted in the increase of swelling, in this case such effect was not observed

Another important conclusion as a result of PIE seems to be the evidence of the homogenization process of initially two–phase (α+γ)-microstructure with regard to alloy U-6,5%Mo and irradiation conditions typical to Russian pool-type research reactors. The only problem to be additionally solved in this case is the study of kinetic dependence of this transformation under irradiation.

Among the negative results of PIE the formation of micro-cracks at the corners of cladding is of special concern as it can result in a possible FE loss of leakprofness. The proposed cause of their appearance is the process of radiation ageing taking place in aluminum alloy SAV-1 of which the cladding was made.

Conclusions

PIE of twelve types of (U-Mo)-based LEU dispersion compositions showed the following:

1. All 12 tested fuel compositions with both the spherical and irregular fuel granules had demonstrated the stable irradiation behavior up to 65% burnup at the irradiation conditions typical to Russian pool-type research reactors.
2. The interaction layers were being formed during irradiation around the fuel particles. Their thickness was different for various types of fuel compositions:
   -Irrespective of fission density the largest thickness of interaction layer corresponds to fuel compositions with the highest uranium concentration per volume unit of either fuel meat (6 g/cm^3) or U-Mo alloy (alloy with 1,5%Mo).
   -Irrespective of fission density the lowest thickness of interaction layer corresponds to fuel compositions on the basis of two-phase (α+γ) uranium alloys with 6,5% and 9% Mo.
3. For all fuel compositions with full or partial decomposition of γ-phase as a result of either fabrication process or special heat treatment the process of homogenization recovering γ-phase was observed.
4. The fuel composition on the basis of uranium alloy with 1,5% Mo-(Al;Sn) which was the only one with α'-phase microstructure also had demonstrated the stable behavior under irradiation. Its microstructure showed the uniform distribution of gaseous bubbles in the fuel granules interior that is supposedly the result of alloying with small additions of Al and Sn. This composition was the only one in which the interaction layer was non-uniform: the internal layer with the visible gaseous bubbles was formed during FE manufacture and external one without visible gaseous bubbles was formed under irradiation.
5. The phenomenon of gaseous fission products preferential accumulation at the boundary between interaction layer and Al matrix was revealed using the characteristic X-ray radiation.
6. The results of planimetry analysis (measurement of FE swelling) showed that alloying of U-6,5 wt % Mo by small additions of Al and Sn hadn't resulted in swelling decrease. Moreover the swelling had increased in comparison with unalloyed one.

7. For some fuel compositions the micro-cracks measuring from tens to hundreds microns were reveled at the corners of cladding near the fins. The proposed cause of their formation is utilization of aluminum alloy SAV-1 which is prone to radiation ageing.
8. As a result of tests conducted, the uranium alloy U-9 wt % Mo was selected to be used for conversion of disperse fuel elements of Russian pool-type research reactors to LEU.

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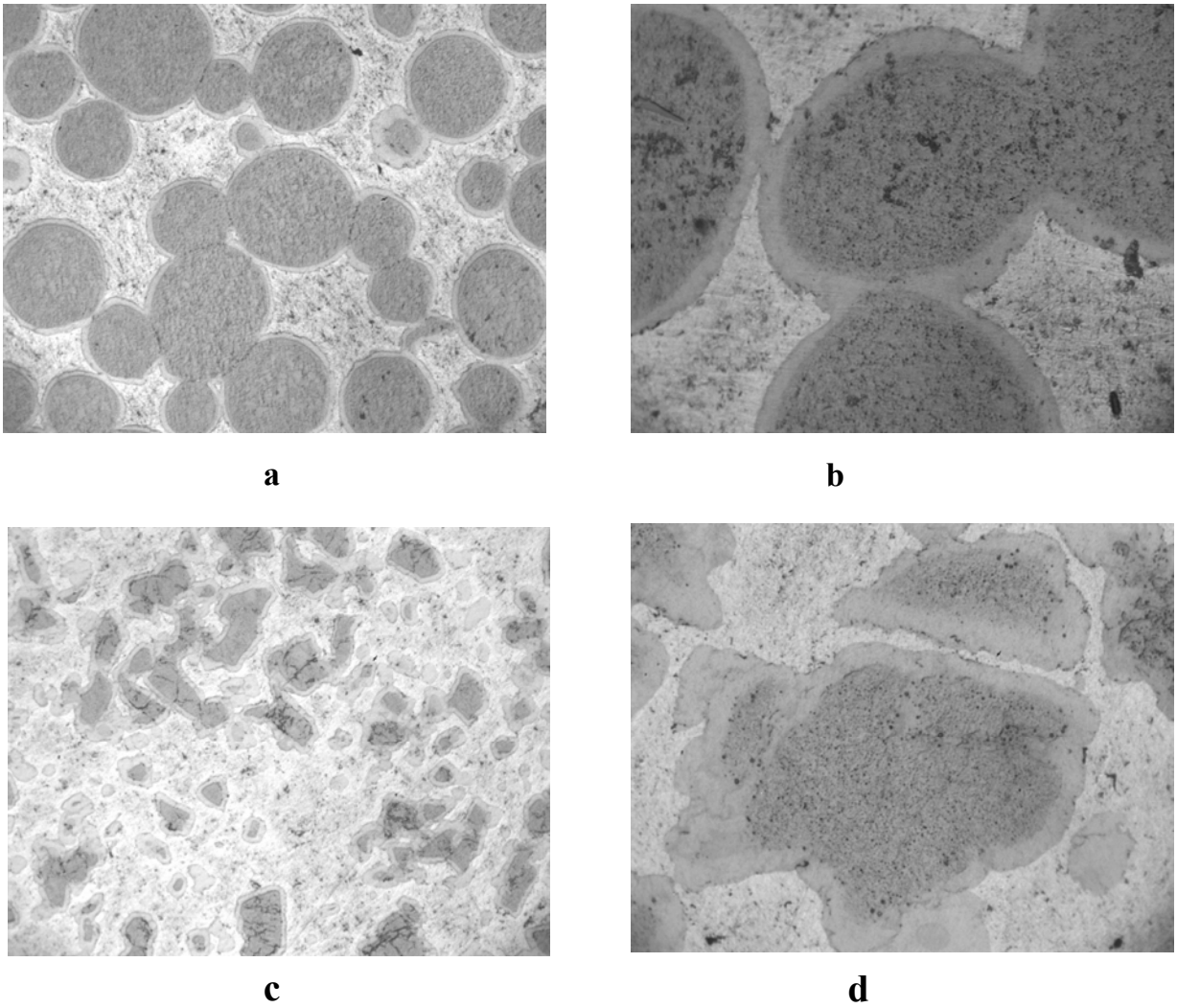


Fig.1. The view of interaction layers around fuel granules of spherical (a, b) and irregular shape (c, d) irradiated to 65%:

a – alloy (U-6,5 wt% Mo), γ-phase state ×200
b – alloy (U-6,5 wt% Mo), γ-phase state ×400
c – alloy (U-6,5 wt% Mo), γ-phase state, alloyed with 0,2%Al ×200
d – alloy (U-6,5 wt% Mo),γ-phase state, alloyed with 0,2%Al ×400

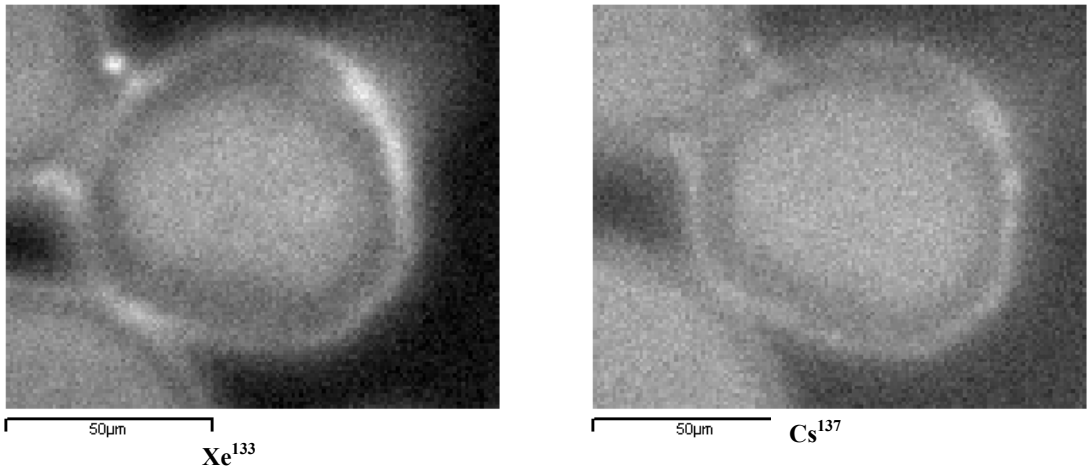
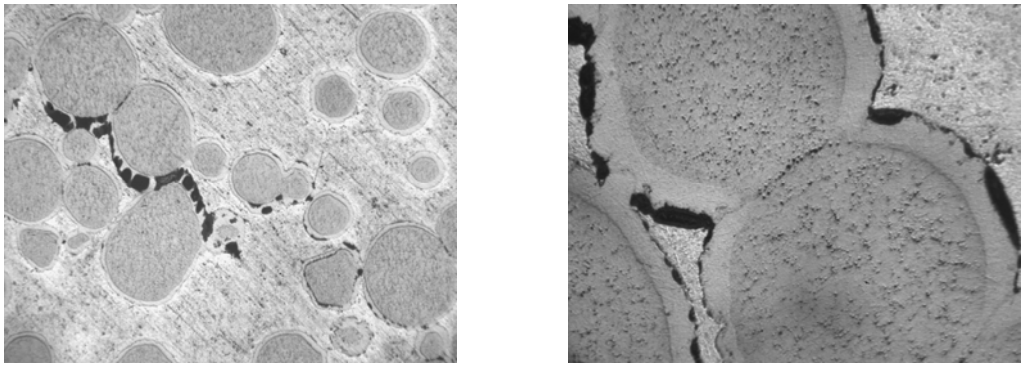
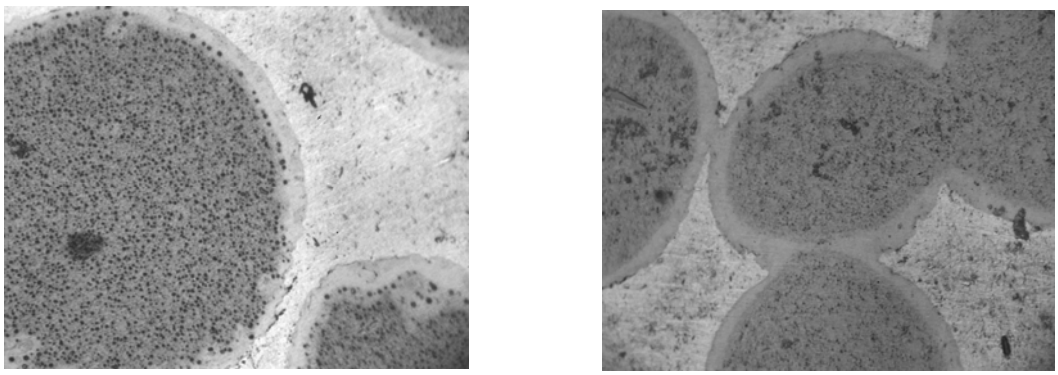


Fig. 2 Concentration maps for distribution of gaseous and highly mobile fission products in a fuel granule and interaction layer. FE № 0169 (burnup 49,5%)



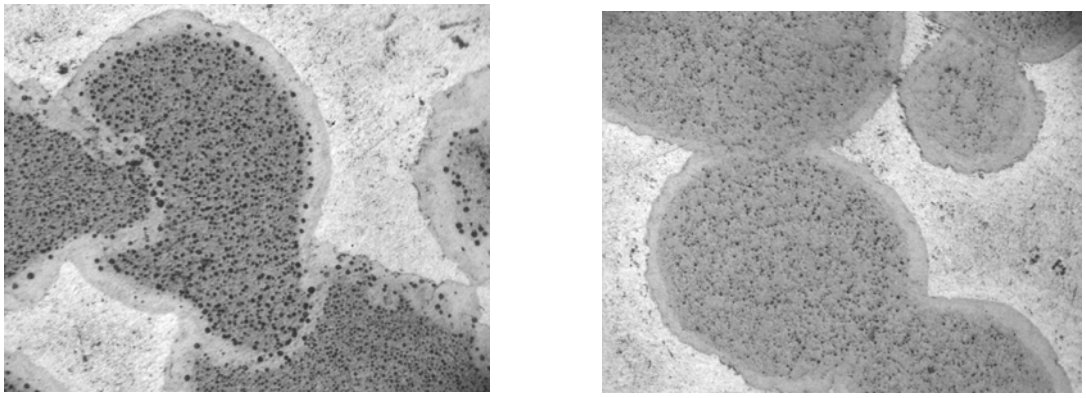
a **b**

Fig. 3 Gross pore formation at the boundary of interaction layer and matrix (FE №0165) (burnup 64,5%)
a – × 200
b – × 400



a **b**

Fig 5 Microstructure of fuel compositions (burnup ~ 65%).
a – fuel composition on the basis of alloyU-1.5%Mo
b – Typical microstructure of fuel compositions on the basis of alloys with 6,5 and 9,0% Mo.



a **b**

Fig 6 The areas of contact between fuel granules after burnup ~ 65%.
a – fuel composition on the basis of alloy 1.5%Mo
b – typical view of contact areas for the fuel compositions on the basis of alloys with 6,5 and 9,0% Mo.

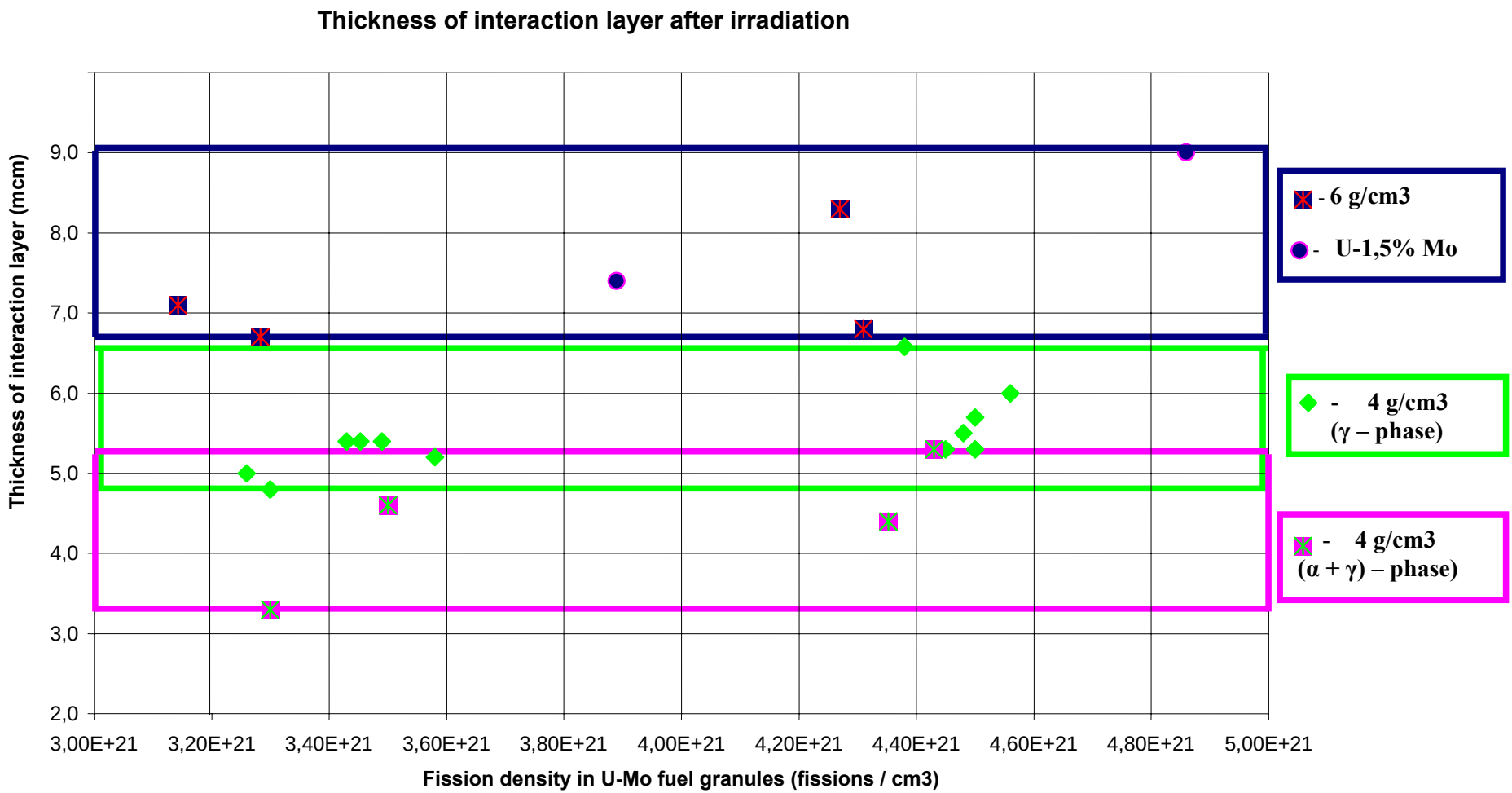

Thickness of interaction layer after irradiation
Thickness of interaction layer (mcm)
9,0
8,0
7,0
6,0
5,0
4,0
3,0
2,0
3,00E+21
3,20E+21
3,40E+21
3,60E+21
3,80E+21
4,00E+21
4,20E+21
4,40E+21
4,60E+21
4,80E+21
5,00E+21
Fission density in U-Mo fuel granules (fissions / cm3)
- 6 g/cm3
- U-1,5% Mo
- 4 g/cm3 (γ – phase)
- 4 g/cm3 (α + γ) – phase)

Fig.4

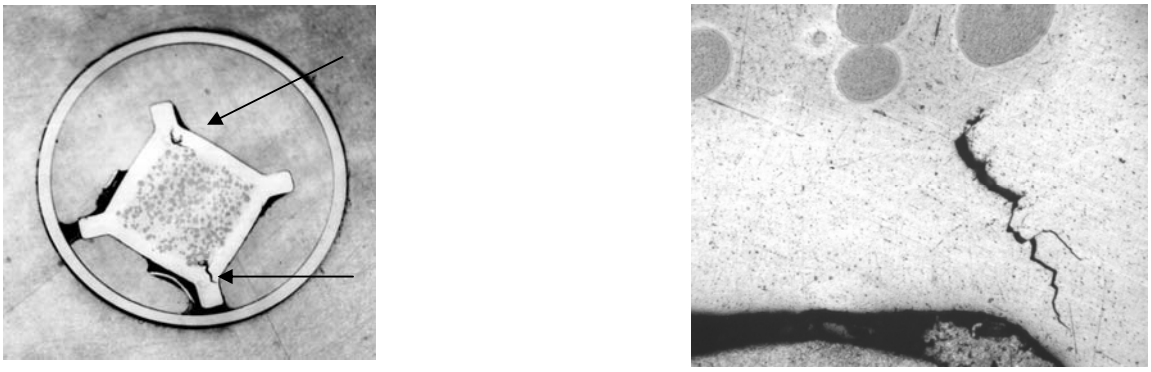


Fig 7. Macrostructure of FE cross-section with twomicro-cracks at the corners of cladding after burnup 65% (to the left) ((× 10) and typical view of micro-crack at bigger magnification (to the right) (×100)

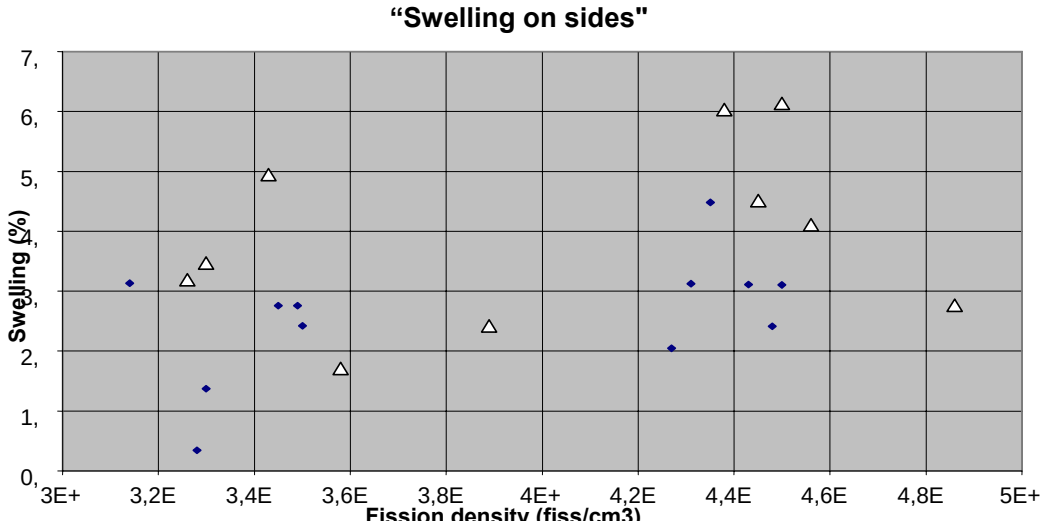


Fig 8 FE swelling against fission density in U-Mo alloy of fuel granules

△ **- fuel elements alloyed with Al and Sn**

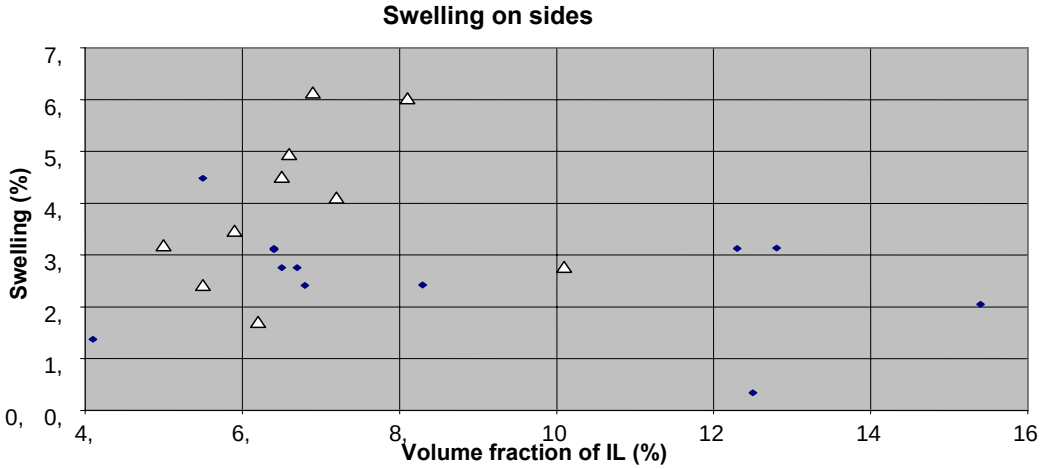


Fig.9 FE swelling against volume fraction of IL in fuel meat.

△ **- fuel elements alloyed with Al and Sn**