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THE MAIN RESULTS OF INVESTIGATION OF MODIFIED DISPERSION LEU U-MO FUEL TESTED IN THE MIR REACTOR

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ABSTRACT

The report presents the results of the tests and of post-irradiation investigations of of modified dispersion LEU U-Mo fuel tested in the MIR reactor under the RERTR program. The results of post-irradiation investigations of mini fuel elements (mini rods) with modified dispersion fuel containing 5%, 13% of silicon in the matrix and protective layers on fuel particles in the form of the oxidized layer and ZrN, tested up to the average U-235 burn-up of ~ 60% and~ 85%, have been obtained. Data proving the reduction of the interaction between U-Mo alloy and the aluminum matrix containing silicon additives were generated. It is demonstrated that the interaction with the matrix does not occur until 85% burn-up when protective ZrN coating is available on U-Mo particles. The impact of irradiation conditions on the interaction layer growth is discovered.

1. Introduction

One of the challenges of low-enriched dispersion U-Mo fuel revealed in the course of the development of high-dense fuel for research reactor conversion is the interaction between fuel particles and aluminum matrix. As fuel burns up, the amount of matrix material decreases because of this interaction and significant changes in the fuel meat physical and mechanical properties are observed, including a significant decrease of its thermal conductivity and elevation of its temperature. Paper [1] explains the generation of gas bubbles and unpredictable fuel meat swelling by an intensive interaction of U-Mo fuel with the aluminum matrix. Further, extensive research was performed under the RERTR program to find a way to decrease this interaction. The authors of paper [2] showed a possibility to decrease this interaction by adding silicon into the aluminum matrix. Under the Russian RERTR program, efforts were made to develop a protective coating for the U-Mo particles surfaces. As a result of preliminary investigations, the most promising techniques were elaborated, namely, to cover particles with zirconium nitride and to oxidize U-Mo particles surface. A decision was taken to make comparative irradiations of mini-rods with U-Mo coatings and different Si content added to the matrix. At the RERTR-2009 conference, preliminary results were presented on the investigation of modified fuel irradiated up to U-235 burnup of ~ 65%. This paper presents detailed results of the fuel meat examinations using SEM and EPMA for unirradiated mini-rods and for those irradiated up to U-235 burnup of ~ 60% and ~ 85%. Optical microscopy, SEM and EPMA were used to get data on the structure and compositions of mini-rods fuel meat. Sections of full transversal cross-sections of mini-rods were done to perform examinations. Unirradiated mini-rods were examined to have data on the initial structure and composition of fuel meats and coatings.

2. Key mini-rods characteristics and irradiation conditions

More than 70 mini-rods were fabricated to irradiate them in the MIR reactor to examine the effect of the silicon content in the matrix aluminum and U-Mo granules coatings on the interaction between fuel particles and the matrix material. Table 1 and Fig. 1 present the key characteristics of the mini-rods.

Table 1. Key characteristics of mini-rods.

Characteristics	Value
Cladding material	SAV-6, alloy 99
Fuel	U-9.4 %Mo, 1-phase and 2-phase alloys
Fuel particles coatings	no coating, oxidized particles, ZrN
Matrix material	Al(PA-4); Al+2%Si; Al+5%Si; Al+13%Si
Circumferential diameter of mini-rods, mm	4.5
Distance between edges, mm	2.6
Enrichment in ^{235}U, %	19.7
Density in U, g/cm³	~6.0
Mass of ^{235}U per rod, g	0.57
Length of fuel meat, mm	180-200

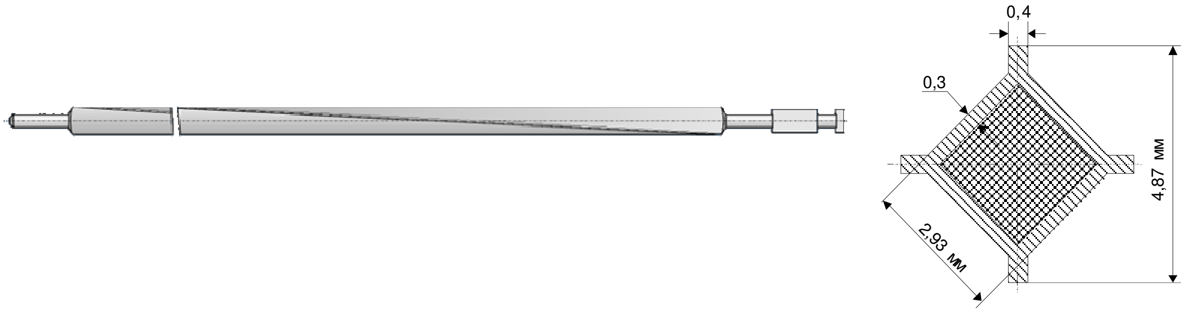

Fig. 1. Appearance and cross-section of a mini rod.

Mini-rods were irradiated in two irradiation rigs in cell 1-3 of the MIR reactor. Table 2 presents the irradiation conditions and results of mini-rods tests in rigs 1 and 2.

Table 2. Irradiation conditions of mini-rods.

Parameters	Irradiation rig 1	Irradiation rig 2
Inlet coolant pressure, MPa	1.08	1.08
Inlet coolant temperature, ºC	40-65	40-65
Coolant velocity, m/s	3.1	3.1
Thermal flux density, MW/m^2		
- average	~ 0.6	~ 1.0
- maximal	~ 1.2	~ 1.8
Outside surface temperature, ºC		
- average	~ 90	~ 120
- maximal	~ 140	~ 180
235U burnup, %		
- average	~ 84	~ 60
- maximal	~ 93	~ 68

Parameters	Irradiation rig 1	Irradiation rig 2
Fission rate in fuel particles, 10^{14}cm^{-3} s^{-1}		
- average	~ 2.3	~ 3.6
- maximal	~ 4.6	~ 6.8
Fission density in fuel particles, 10^{21} cm^{-3}		
- average	~5.6	~4.0
- maximal	~6.2	~4.5
Test duration, days		
- total	455	182
- at power	285	130

3. The results of investigations

3.1. State of fuel meats of unirradiated mini-rods

The examination of unirradiated fuel compositions showed no interaction between fuel particles and matrix during manufacturing of mini-rods of all types. As for mini-rods with 5% Si, there is an interaction between silicon and U-9%Mo alloy. A silicon-saturated layer is observed on the fuel particles surfaces. This layer is not continuous and its thickness is different for different particles and non-uniform over the perimeter of one and the same particle.

EPMA was used to plot the distribution maps of silicon, uranium, molybdenum and aluminum (Fig.2). A decreased content of molybdenum and increased content of uranium were found in the grain boundaries of the U-Mo alloy, as it was found in Ref. [4]. The analysis of the distribution maps shows that silicon diffuses inside the U-Mo particles at the grain boundaries.

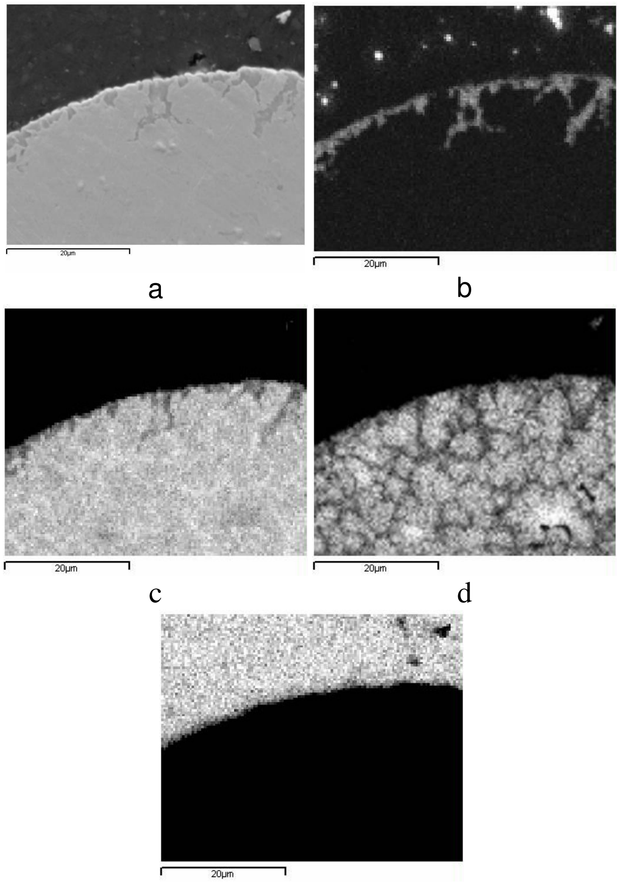

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Fig. 2. A fuel meat section of a mini-rod with 5% Si in the matrix (a) and distribution maps of Si (b), U (c), Mo (d) and Al (e).

No traces of aluminum penetration into the U-Mo alloy were revealed. When 13% of Si is added into the matrix, no qualitative changes in the interaction between silicon and fuel particles are observed as compared to the 5% Si content. Silicon penetrates deeper into the U-9%Mo alloy diffusing over the grain boundaries (Fig.3). A silicon-saturated layer becomes wider on the fuel particles surfaces. It covers the majority of fuel particles but does no form a continuous film. As for U-Mo alloy particles with a silicon-saturated layer, matrices with 5%Si and 13% Si have silicon-free areas about 5μm thick around these particles. In other areas, where silicon particles are located very close to the U-Mo alloy, the U-Mo particles surfaces do not have a Si-saturated layer (Fig.4).

As for mini-rods with Zr-N coating, this layer thickness makes up 2-3μm (Fig.5). Some U-Mo particles do not have the Zr-N coating either partially or completely. Oxidized U-9%Mo alloy was used for the other type of mini-rods. The oxide layer thickness of the fuel particles surfaces made up 0.3µm (Fig.6).

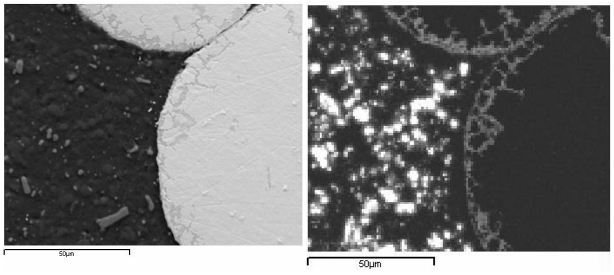

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Fig. 3. Fuel meat section of a mini-rod with 13% Si in the matrix (a) and Si distribution map plotted for this section (b).

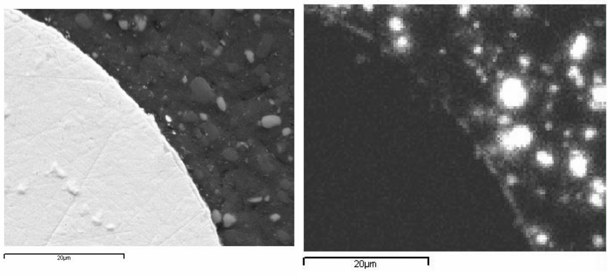

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Fig. 4. SE image of a U-Mo particle fragment with no silicon-saturated layer (a) and Si distribution map (b), 13% Si in the matrix

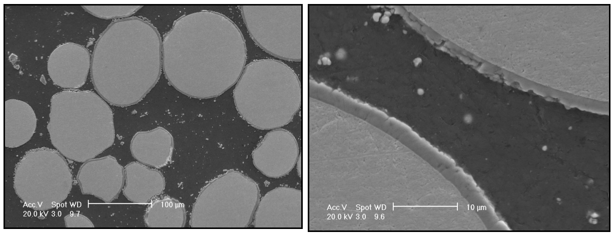

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Fig. 5. A fragment of mini-rod fuel meat with ZrN coating on U-9%Mo particles (a). ZrN coatings in SE (b).

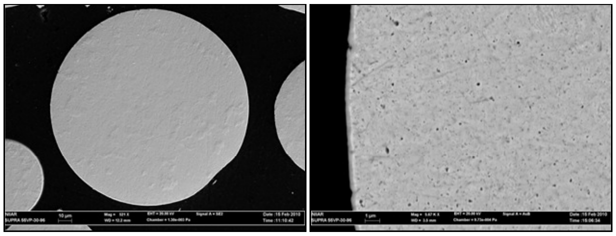

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Fig. 6. SE images of oxidized U-9%Mo particle before irradiation (a). Oxide layer on the particle surface is ~ 0.3µm. Image (b) is in backscattered electrons in the composition contrast mode.

3.2. State of fuel meats after irradiation

The optic microscopy showed that there was rather thick corrosion layer on the mini-rods claddings. Table 3. presents the corrosion layer measurement results.

The degree of interaction between the U-9%Mo particles and matrix from aluminum powder PA-4, 5% Si and 13% Si and ZrN coating was evaluated based on the volume fraction of the interaction layer. The volume fraction of the interaction layer was determined by SEM. Photos were made for the whole cross-section of a mini-rod. Then, by means of VideoTest software, the fuel meat area was evaluated as well as the interaction layer in this cross-section for each mini-rod. These data were used to evaluate the volume fraction of the interaction layer. The results are presented in Table 4.

It follows from the table 4. that the interaction layer decreases significantly if silicon is added into the matrix. A silicon content increasing from 5% to 13% decreases significantly the interaction layer volume fraction.

Higher fission rates and, correspondingly, higher temperature of fuel under irradiation increase the rate of U-Mo alloy interaction with the aluminum matrix. At U-235 burnups of ~ 60 % and ~ 84%, the volume fractions of interaction layers in the fuel meat with aluminum matrix are equal. In the first case, the average fission rate made up 3.6×10^{14}cm^{-3} s^{-1}, in the second case it was 2.3×10^{14}cm^{-3} s^{-1}.

Table 3. Maximal thickness of the corrosion layer of the mini-rods surfaces.

Clad material	Type	Time of irradiation at power, days	Clad T, av/max., ºC	U-235 burnup, %	Corrosion layer thickness, µm
Alloy 99	Si<0.4%	130	120/180	~ 60	71
	A1+5%Si	130	120/180	~ 60	69
	A1+13%Si	130	120/180	~ 60	64
	ZrN coating, Si<0.4%	130	120/180	~ 60	64
	Si<0.4%	285	90/140	~ 84	64
	A1+5%Si	285	90/140	~ 84	62
	A1+13%Si	285	90/140	~ 84	73
	ZrN покрытие, Si<0.4%	285	90/140	~ 84	55
SAV-6	Si<0.4%	130	120/180	~ 60	60
	A1+5%Si	130	120/180	~ 60	64
	A1+13%Si	130	120/180	~ 60	71
	ZrN coating, Si<0.4%	130	120/180	~ 60	60
	Si<0.4%	285	90/140	~ 84	52
	A1+5%Si	285	90/140	~ 84	52
	A1+13%Si	285	90/140	~ 84	49
	ZrN coating, Si<0.4%	285	90/140	~ 84	52

Table 4. Measurement of the interaction layer volume fraction in mini-rods irradiate up to average burnups ~ 60% and ~ 84%.

Modification	Rig 1			Rig 2		
	rod No.	Burnup, %	IL volume fraction, %	rod No.	Burnup, %	IL volume fraction, %
Si<0.4%	3496	~ 60	40±4	3492	84	40±4
A1+5%Si	3597	~ 60	25±3	3596	84	10±2
A1+13%Si	3396	~ 60	10±2	3395	84	6±1
ZrN coating, Si<0.4%	4496	~ 60	6±1	4491	84	6±1

Under the same irradiation conditions, but with the presence of silicon, the increase of U-235 fission rate and, correspondingly, higher fuel meat temperature, have higher effect on the acceleration of the fuel particles-to-aluminum interaction. Here, at U-235 burnup of ~ 84%, the volume fraction of the interaction layer is significantly less as compared to that of ~ 60%. Paper [3] gives a preliminary and overestimated value of the volume fraction of the interaction layer in a mini-rod with 13%Si in the matrix and ~60% burnup. This is because the interaction layer volume fraction was evaluated by images that did not reflect the whole area of the fuel meat cross-section. Since there is a high non-uniformity of the interaction layer thickness in this type of mini-rods, there can be a great scattering in the volume fraction values on different fuel meat areas. To have an average estimation, the interaction layer volume fraction is to be evaluated for the whole fuel meat cross-section as it has been done in this paper. As for mini-rods with the ZrN coating, the interaction layer volume fraction remains at a level of 6% under the above irradiation conditions. The mini-rods, which contain oxidized U-9%Mo particles in the fuel meat, irradiated up to a burnup of 61.6% had the interaction layer volume fraction 50±5% [3]. Figure 6. presents the microstructure of the fuel compositions.

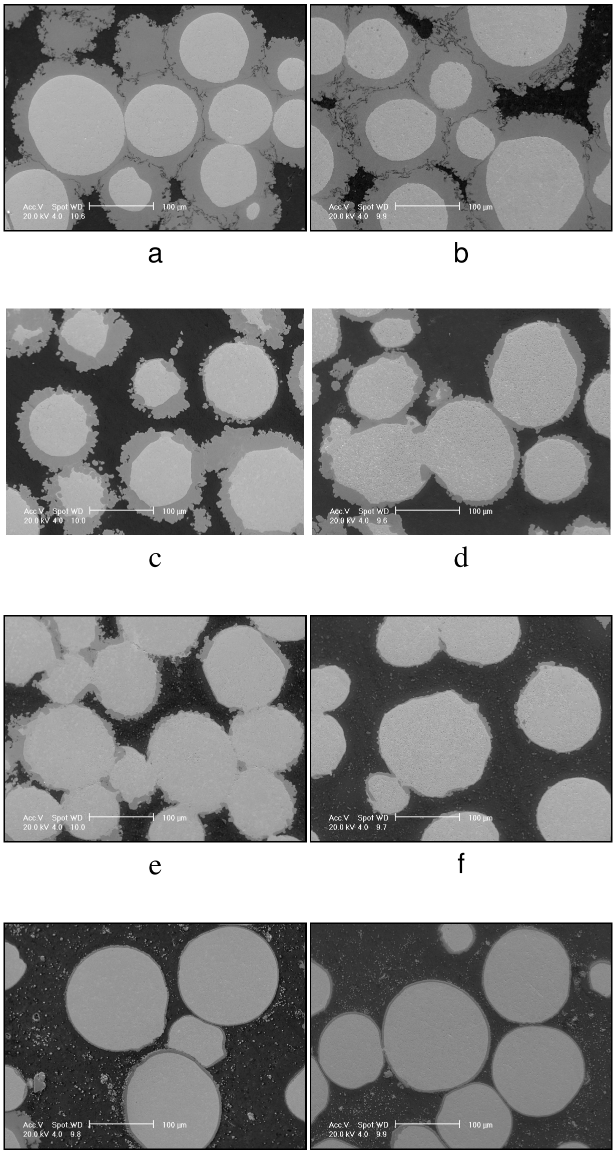

Fig. 6. SEM images of the fuel meat microstructure
- U-9%Mo particles in the aluminum matrix. Burnup 60% (a) and 80% (b);
- U-9%Mo with 5% Si in the matrix. Burnup 58.8% (c) and 84 % (d);
- U-9%Mo with 13% Si in the matrix. Burnup 60.2% (e) and 84 % (f);
- U-9%Mo with ZrN coating. Burnup 60.2% (g) and 84 % (h);

Analysis of the fuel meat state shows that the IL thickness is equal for all U-Mo particles of the U-9%Mo/Al (without Si) composition and makes up 25-30μm.

The Si added matrix IL thickness becomes very uneven over the fuel particles perimeter. For 5 % Si added matrix IL values range from 2 to ~30μm for more high fission rate and burnup of 60% and from 2 to ~20μm for relative low fission rate and burnup of 84%. For 13% Si added matrix the major area of the U-Mo particles surface has IL ~ (1-2)μm thick. At more high fission rate and burnup of 60% this layer achieves ~15μm, at relative low fission rate and burnup of 84% IL achieves ~10μm.

The ZrN coating prevents completely the interaction of U-Mo alloy with the aluminum matrix. The interaction layer is observed in the areas with no coatings on the fuel particles. It is true both at a burnup of ~ 60% and ~ 84%.

EPMA was used to determine the effect of silicon on the interaction layer size. Silicon distribution maps were plotted. The quantitative EPMA shows the silicon content in the interaction layers to range from 7 to 15% at. Close to the silicon-saturated interaction layer, there is 3-5μm wide zone with no silicon particles in the matrix (Fig.9). No silicon was revealed in the wide interaction layer. Silicon concentrates on the boundary of the wide interaction layer with the matrix.

The summary of the EPMA results shows that the interaction layer becomes larger until it meets silicon particles. Here, it stops growing and further interaction takes place on the neighboring silicon-free areas. If there is a large amount of silicon particles, the interaction layer growth is blocked. It can be clearly seen in Figure 10, where the interaction layer growth is blocked by silicon particles in all directions. No silicon particles were

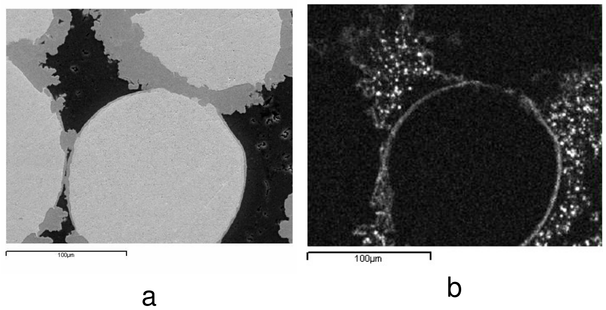

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Fig. 7. Non-uniform in thickness interaction layer in the matrix with 5%Si. U-235 burnup - 58.8%. Si distribution around fuel particles. No Si in the wide interaction layer.

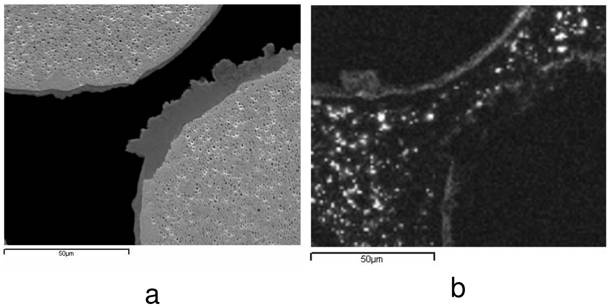

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Fig. 8. Si distribution (b) around fuel particles (a). Matrix with 5% Si. U-235 burnup - 80%.

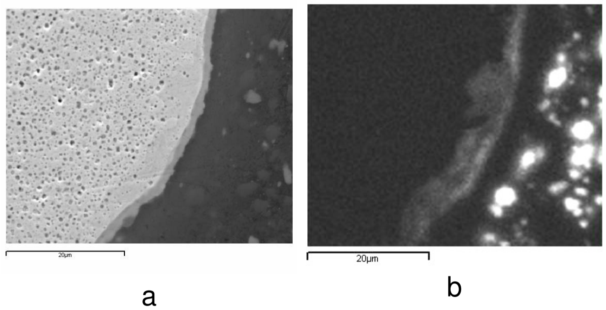

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Fig. 9. A fragment of U-Mo particle in the matrix with 13% Si (a). U-235 burnup - 80%. Si distribution map (b) with a Si-free layer 3.5µm wide located over the fuel particle perimeter.

found inside the interaction layer. In the U-Mo particles near-surface area, the silicon distribution becomes more uniform. There is no pronounced Si distribution over the U-Mo alloy grains as it was observed in the unirradiated state.

4. Discussion

The results of examination of mini-rods irradiated in two rigs, irradiation conditions being different in thermal power (fission rate), show a significant influence of irradiation conditions on the intensity of the interaction layer generation. In irradiation rig 1, the average U-235 burnup of ~80% (average fission density in fuel particles ~5.6×10^{21}cm^{-3}) was achieved for 258 days of operation at full power and at an average fission rate of 2.3×10^{14}cm^{-3}s^{-1}. In irradiation rig 2, the U-235 burnup of ~60% (average density in fuel particles ~4.0×10^{21}cm^{-3}) was achieved for 130 days of operation at full power and at an average
fission rate of 3.6×10^{14}cm^{-3} s^{-1}. The following results were obtained (Table 3):

- nearly the same volume fractions of the interaction layer were obtained for the matrix with no Si;- volume fraction of the interaction layer in the mini-rod with 5% Si in the matrix is 2.5 times higher that that one in the mini-rod with 13% Si irradiated in rig 2 up to ~60% burnup at a fission rate of 3.6×10^{14}cm^{-3} s^{-1};
- volume fraction of the interaction layer in the mini-rod with 5% Si in the matrix is 1.7 times higher that that one in the mini-rod with 13% Si irradiated in rig 1 up to ~80% burnup at a fission rate of 2.3×10^{14}cm^{-3} s^{-1}.

Thus, as Si is added to the matrix, the effect of interaction suppression depends on the Si concentration and it is lower at a relatively low fission rate of 2.3×10^{14}cm^{-3} s^{-1} as compared to a fission rate of 3.6×10^{14}cm^{-3} s^{-1}. The fuel meat temperature can have a significant effect on the above phenomena as well. Therefore, the mini-rods irradiation temperature should be calculated for two rigs with the account of the irradiation background, generation of corrosion film on the claddings and degradation of the thermal conductivity under irradiation. When manufacturing mini-rods with Si addition in the matrix, the silicon dissolution near the U-Mo particles surfaces is not complete. Due to this, either fuel particles or areas of their surfaces are not saturated with silicon. If there is a silicon-saturated layer on the U-Mo particles surfaces, then it slows down sharply the diffusion of U and Mo atoms and Al ones as well. As a result, the width of the interaction layer generated under irradiation makes up 1-3µm. It has been determined that a silicon concentration of 7%at is enough to prevent an intensive growth of the interaction layer under this irradiation in the MIR reactor. If, in the unirradiated state, the Si concentration on the fuel particle surface is not enough, then wide interaction layers generate under irradiation. There is no silicon inside the wide interaction layer. It is displaced to the boundary of the interaction layer with the matrix. As this boundary moves inside the matrix, the silicon concentration in it increases. If there can be large silicon particles, the interaction layer stops growing while it goes on growing on the silicon-free areas. It explains the non-uniform thickness of the interaction layer in the mini-rods matrices that contain silicon additions. Thus, silicon particles are the effective barrier to prevent the interaction between the U-Mo alloy and aluminum. The increase of the fuel meat temperature contributes to more intensive diffusion. It leads to the increase of volume fraction of the interaction layer as the fission rate increases.

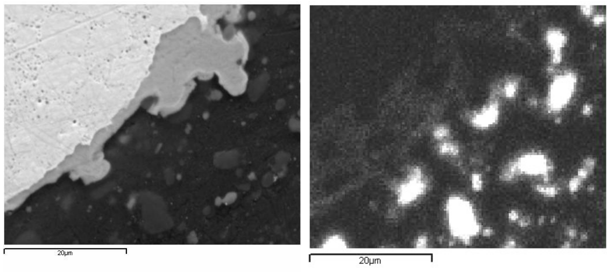

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Fig. 10. Matrix with 13% Si, U-235 burnup - 58,8%. All directions of the interaction layer growth are blocked with silicon particles (b).

Paper [3] states that if there is an oxide layer on the unirradiated U-9%Mo particles, the volume fraction of the interaction layer at a burnup of 61.6% is higher than that one of the U-9%Al fuel meat composition. Consequently, the presence of a 0.3µm thick oxide layer on the U-Mo alloy does not prevent the interaction of fuel particles with the aluminum matrix. The most effective way to stop the interaction between the U-Mo alloy and aluminum is the ZrN coating on the U-Mo particles. If the width of the coating is 2-3µm, there is no interaction with the aluminum matrix under the above irradiation conditions up to a burnup of 85%.

Conclusion

The post-irradiation examinations of the mini-rods with modified U-Mo fuel irradiated in the MIR reactor up to average burnups of U-235 of 60% and 85% showed the following:

- 2-3µm thick ZrN coating on U-Mo particles prevent their interaction with the aluminum matrix up to high burnups 85% at fission rates up to ~ $7\times10^{14}cm^{-3}\ s^{-1}$ without Si additions;
- 0.3µm deep oxidation of the U-Mo particles surfaces does not effect the interaction between the matrix and fuel particles. Probably, the oxidation should be deeper;
- silicon added into the matrix decreases significantly the interaction between U-Mo particles and the matrix, both silicon content and fission rate being effective.

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