RERTR 2009 —  $31^{st}$  International Meeting on Reduced Enrichment for Research and Test Reactors

November 1-5, 2009 Kempinski Hotel Beijing Lufthansa Center Beijing, China

## THE STATUS OF LEU U-Mo FUEL INVESTIGATION IN THE MIR REACTOR

A.L. Izhutov, V.V. Alexandrov, A.Ye. Novosyolov, V.A. Starkov,
 A.V. Sheldyakov, V.Yu. Shishin, V.V. Iakovlev
 RIAR, 433510, Dimitrovgrad-10, Ulyanovsk Region – Russia

and

I.V. Dobrikova, A.V. Vatulin, V.B. Suprun A.A. Bochvar All-Russian Scientific Research Institute of Inorganic Materials P. O. Box 369, 123060, Moscow, Russia

and

Ye. F. Kartashov, V.A. Lukichev Research and Development Institute of Nuclear Energy and Industry P. O. Box 788, 107014, Moscow, Russia

### ABSTRACT

The paper presents the results of post irradiation examinations of low-enriched U-Mo fuel irradiated in the MIR reactor under the RERTR program. At present, the reactor testing of mini fuel pins with solid U-Mo fuel and an IRT-type full size fuel assembly with fuel pins is being continued in the MIR reactor. The preliminary results of post irradiation examinations of IRT-M full size fuel assemblies with tubular fuel elements and IRT-U FA with fuel pins irradiated to an average U-235 burnup of ~ 40%, as well as mini pins with modified dispersed fuel, different Si content (2-13%) in a matrix and protective layers on fuel particles in the form of ZrN and oxidized layer were obtained.

### 1. Introduction

Under the RERTR program in the MIR reactor from the year 2003 till today have been conducted tests of the fuel with low-enriched U-Mo for the purpose of converting Russian-made pool-type reactors. During the period from 2003 to 2006 tests were performed on 72 rod-type mini fuel elements with different modifications of dispersion U-Mo fuel developed and fabricated in A.A. Bochvar institute [1]. Since 2007 the full-scale tests of IRT-type FAs and also of rod-type mini fuel elements based on modified dispersion fuel have been continued.

In the first half of 2008 two full-scale IRT-type FAs were removed from the reactor after ~ 40% burn-up of U-235 was reached, one of the FAs being with tube-type fuel elements, the second one with rod-type fuel elements. Moreover, in 2008 were completed the tests of a rod-type mini element lot based on modified U-Mo fuel meat with different Si contents in the matrix material, and also with the coatings on fuel particles with the burn-up of ~60% (device No.2). Technical characteristics, conditions and the results of tests of these full-scale FAs and of mini fuel elements were previously stated [2, 3].

In November 2008 were put to a stop the tests of the full-scale IRT-3M type FAs with tube-type fuel elements with the mean U-235 burn-up  $\sim$ 50% due to appearance of cladding failure signs as is read on the leak check system based on registration of delayed neutrons from the fission products.

In February 2009 were completed the tests of the second lot of rod-type mini elements based on modified U-Mo fuel meat after reaching the mean U-235 burn-up  $\sim$ 84 % (device No.1).

Currently in the MIR reactor are in progress the tests of rod-type mini elements based on monolithic U-Mo fuel (maximum U-235 burn-up ~21 %), and also of the full-scale FA with rod- type elements (maximum U-235 burn-up ~55 %).

## 2. Main parameters of full-scale FAs and mini fuel elements tests.

In table 1 see main parameters and the results of full-scale FAs tests.

Parameters	IRT-U	IRT-3M	IRT-U	IRT-3M
	№19УИ	№100МИС	№ 19УИ	№199МИС
	0012006	38606	0022006	38706
Heat power of EFA, kW				
- average	503	452	286	369
- maximum	722	800	661	800
Inlet coolant pressure, MPa	1.08	1.08	1.08	1.08
Inlet coolant temperature, ° C	40-60	40-60	40-65	40-65
Coolant velocity, m/s	5.9	6.6	5.2	6.6
Heat flux, MW/m <sup>2</sup>				
- average,	0.70	0.56	0.41	0.38
- maximum	0.99	1.00	0.96	0.99
Temperature of cladding outer surface, <sup>o</sup> C				
- average,	80-100	67-87	65-85	58-78
- maximum	95-115	86-106	95-115	86-106
Burn-up 235U of EFA, %				
- average,	40.1	40.3	39.5	50.3
- maximum	55.4	48.7	54.6	63.2
Fission rate in fuel particles, $10^{14}$ cm <sup>-3</sup> ·s <sup>-1</sup>				
- average,	~1.3	~1.2	~0.8	~0.7
- maximum	~2.6	~2.7	~2.6	~2.7
Fission density in fuel particles, $10^{21}$ cm <sup>-3</sup>				

Table 1. The main parameters and results of full-scale FAs testing.

Parameters	IRT-U	IRT-3M	IRT-U	IRT-3M
	№19УИ	№100МИС	№ 19УИ	№199МИС
	0012006	38606	0022006	38706
- average,	~2.7	~2.8	~2.6	~3.5
- maximum	~3.7	~3.3	~3.6	~4.3
Duration of testing, days				
- on power	234	257	418	394
In-reactor leak test results	No leakage	No leakage	No leakage	Leakage
PIE			To be	To be
	In progress	In progress	started	started
			in late 2010	in late 2009

In table 2 are given main parameters and the results of mini fuel elements tests.

Table 2. The main parameters and the results of mini fuel elements testing.

Parameters	Modified Modified		Monolithic fuel	
	dispersion fuel	dispersion fuel	mini elements	
	mini elements	mini elements		
	(device No.2)	(device No.1)		
Heat power of EFA, kW				
- average	66	42	39	
- maximum	102	100	54	
Inlet coolant pressure, MPa	1.08	1.08	1.08	
Inlet coolant temperature, ° C	40-65	40-65	40-65	
Coolant velocity, m/s	2.8	2.8	2.8	
Heat flux, MW/m <sup>2</sup>				
- average	1.1	1.0	0.8	
- maximum	1.7	1.7	1.1	
Temperature of cladding outer surface, <sup>o</sup> C				
- average	~110	~100	~110	
- maximum	~140	~140	~135	
Burn-up 235U of EFA, %				
- average	60.3	83.8	20.6	
- maximum	67.5	93.0	21.4	
Fission rate in fuel particles, $10^{14}$ cm <sup>-3</sup> ·s <sup>-1</sup>				
- average	~2.0	~2.0		
- maximum	~4.5	~4.5		
Fission density in fuel particles, 10 <sup>21</sup> cm <sup>-3</sup>				
- average	~4.0	~5.6		
- maximum	~4.5	~6.2		
Duration of testing, days				
- calendar,	182	455	210	
- on power	130	285	90	
PIE	In progress	To be started in early 2010	To be started in late 2010	

# 2. Preliminary results of post-irradiation examinations

### 2.1 Results of full-scale FAs investigations

The experimental full-scale IRT-3M FA consists of six tube-type fuel elements of square section with a coaxial arrangement. The fuel elements with LEU dispersion U-Mo fuel fabricated by extrusion represent a three layer tube of square section composed of the inner and the outer claddings, fuel meat, upper and lower plugs. The fuel element thickness is 1.4 mm, the thickness of the fuel layer is  $\sim 0.5$  mm. The 4<sup>th</sup> fuel element was fabricated with oxidized U-Mo particles.

The experimental full-scale IRT-U FA is a structure which consists of a bundle comprising 172 rod-type fuel elements of square section (distance between the edges is 2.6mm, the circumscribed diameter is 4.5mm), linked by means of upper and lower spacer grids. The bundle of fuel elements lies between two shrouds of square section. Fuel elements also fabricated by extrusion have a square profile with ribs and consist of a cladding, meat and two plugs. Main technical characteristics of full-scale FAs are shown in table 3.

Characteristic	IRT-3M	ИРТ-У
Cladding thickness, mm	0.31÷0.46	0.31÷0.46
Length of active part, mm	561÷609	620-÷635
Fuel material	U-9.4%Mo, U-8.8% Mo oxidized	U-9.4% Mo
Matrix material	PA-4 (Si≤0.3%)	PA-4 (Si≤0.3%)
U density in the fuel meet, $g/cm^3$	5.3-5.4	~ 6.0
Enrichment of <sup>235</sup> U, %	19.3÷19.9	19.7
Number of fuel elements, pcs	6	172
Cladding material	SAV-1	SAV-1, AMg2

Table 3. The main technical characteristics of full-scale FAs

Destructive investigations of IRT-3M FAs were performed on the samples with maximum burn-up – height-mean cuts of the external fuel element No.1, besides were analyzed the samples from fuel element No.4 with the oxidized fuel particles. For the destructive tests of IRT-U were also chosen fuel elements with the maximum burn-up value.

The macro sections of the fragments of fuel element No.1 of IRT-3M with the burn-up  $\sim 52$  %, cut at the height of  $\sim$ 335mm from the fuel element bottom, are shown in figure 1a. The cross-section of fuel element No.97 of IRT-U FA at the elevation of 325 mm with the burn-up  $\sim 56$  % is shown in fig.1b.

As a whole the fuel condition is satisfactory; there is a tight contact between the cladding and the meat; no cracks or any significant defects were revealed on the cladding. The thickness of the oxide layer on the outer surface of the cladding of fuel element No.1 of IRT-3M FA reaches  $\sim 30$  µm on the edges and  $\sim 14$ µm on the rounding's in the corners of the square. The oxide film on

the surface of fuel element No.97 of IRT-U FA constitutes  $\sim 23 \mu m$  on the ribs and  $\sim \!\! 16 \mu m$  on the edges.



Fig. 1. The macro sections of the fragments of fuel element No.1 of IRT-3M at the elevation of  $\sim$ 335mm, burn-up  $\sim$  52 % (a) and the cross-section of fuel element No.97 of IRT-U FA at the elevation of  $\sim$ 325 mm, burn-up  $\sim$  56 % (b).

In fig. 2 are shown the images made by scanning electronic microscopy (SEM). As is seen the fuel elements condition is satisfactory – there is a tight contact between fuel particles and the matrix, however in the matrix material some process porosity was noticed.





Fig. 2. SEM images of macro- and microstructures of fuel meat: a and b (element No.1 of IRT-3M, burn-up  $\sim$  52 %), c and d (element No.97 of IRT-U FA, burn-up  $\sim$  56 %).



Fig. 3 . Fuel particle-matrix interaction layer: a (element No.1 of IRT-3M, burn-up  $\sim$  52 %), b (element No.97 of IRT-U FA, burn-up  $\sim$  56 %).

The thickness of interaction layer (IL) between U-Mo alloy and the matrix is not regular along the fuel particles perimeter (fig.3). Its mean value constitutes (6-7)  $\mu$ m in the meat of the first fuel element of IRT-3M FA and (9-11)  $\mu$ m in fuel element No.97 of IRT-U FA.

The structure of the interaction layer is homogeneous, no pores are revealed (fig.4). On the joints of fuel particles there are pores of a larger diameter up to 1 $\mu$ m than inside the particles. On the cut of U-Mo granules the distribution of porosity is irregular. The pores are located mainly along the grain boundaries (see fig. 5 a, 6). The areas where porosity is absent are visible. It could be due to existence of U-Mo alloy grains of different size.



Fig. 4. The microstructure of the interaction layer (a) and the porous formation on the joint of U-Mo particles (b).



Fig. 5. The porous formation inside of U-Mo particles: a (element No.1 of IRT-3M, burn-up  $\sim$  52 %), b (element No.97 of IRT-U FA, burn-up  $\sim$  56 %)..

In fig.6 are shown SEM images of macro- and microstructure of fuel element No/4 of IRT-3M FA with oxidized U-Mo particles (burn-up U-235 ~ 37 %). The thickness of the interaction layer on the oxidized particles is within the range  $(4 - 6) \mu m$ . It is noteworthy that considering the smaller value of the fuel burn-up in this fuel element the thickness of the interaction layer is correspondingly smaller than the thickness of the interaction layer in the fuel meat of the fuel element No.1 with the normal U-Mo fuel. Thus, the oxidizing of fuel particles has no significant impact on the interaction layer forming.



Fig. 6. SEM images of macro- and microstructure of fuel element No4 of IRT-3M FA with oxidized U-Mo particles (burn-up U-235  $\sim$  37 %).

In fig 7. are given the results of EPMA in terms of distribution of different elements in the fuel meat; the scanning trace is shown in fig. 7a. The notable result of these investigations is the fact that on the boundary of the matrix interaction layer the increased contents of fission products is detected fig.7 (e -Xe, f-Cs, g-Zr). The most vividly this effect is being expressed for Zr (fig.7, g). Subsequently it seems reasonable to perform additional investigations and to conduct more thorough analysis of the revealed facts.



а



Fig. 7. The results of EPMA (element No.1 of IRT-3M, burn-up ~ 52 %): a-the scanning trace, b - Al, c - U, d - Mo, e - Xe, f - Cs, g - Zr.

### 2.2 Post-irradiation investigations of the mini fuel elements

Reactor tests of the mini fuel elements were performed to investigate the influence of Si contents in the matrix Al and of protective coatings on U-Mo granules on the degree of interaction between the fuel particles and the matrix material. To decrease the intensity of interaction between the fuel and the aluminum matrix in some fuel elements U-Mo alloy granules with protective coatings were used. Two processes were used – oxidizing and ZrN layer application. The mini fuel elements characteristics are given in table 4, parameters of their irradiation are shown in the 1<sup>st</sup> column of table 2.

 Table 4. The main technical characteristics of mini elements

Characteristic	Value
Cladding material	SAV-6, alloy 99
Fuel material	U-9.4 %Mo, 1-phase and 2-phase alloys
Coatings on U-Mo granules	no coated, oxidized particles, ZrN
Matrix material	Al(PA-4); Al+2%Si; Al+5%Si; Al+13%Si
Circumscribed diameter, mm	4.5
Distance between the edges, mm	2.6
Enrichment of <sup>235</sup> U	19,7
U density in the fuel meet, $g/cm^3$	6.0.
Length of active part, mm	180-200



а



b



Fig. 8. The cross-sections of non irradiated (a and b) and irradiated (c – burn-up 64%, and d burn-up 67%) of mini elements.

In fig.8 are shown the typical views of cross-sections of non irradiated (a and b) and irradiated (c and d) of mini fuel elements. Similar to the earlier investigated mini fuel elements [1], in some cuts the thickness of the claddings seems irregular being a result of fuel particles implantation into the cladding. The state of the irradiated fuel elements is satisfactory – in metallographic specimen no porosity inside fuel meat is detected, neither is defects forming on the claddings. Swelling of the fuel meat appearing as a transformation of the square into a certain round shape can be seen, and the obtained deformations are not resulting in formation of cracks on the cladding.

On the outer surfaces of the claddings of the mini fuel elements there is a rather thick oxide film up to ~ 70  $\mu$ m. This fact is an evidence of much bigger heat flows on the surface than during the previous tests of mini fuel elements of such design [1], when the mean thickness of the oxide film did not exceed ~ 12  $\mu$ m at reaching burn-up ~ 67 %, but at heat flows not more than 1 MW/m<sup>2</sup>. The authors are planning to carry out an additional analysis of conditions and parameters of the tests to clarify the heat loads and temperatures of the fuel elements' surface during these tests performance. Fig.9 illustrates the microstructure of the oxide film on the surface of fuel element No.3297.



Fig. 9. The microstructure of the oxide film on the surface of fuel element No.3297, burn-up 67 %

Examining the images of the fuel meat microstructure of various modification mini fuel elements (fig.10) a significant distinction in the thickness of the interaction layer in the fuel elements of different types could be seen. It is noteworthy that the depth of the fuel burn-up (the relative value of which was determined by the results of gamma-scanning) in the fuel elements is different and it is due to the irregularity of the neutron flow in cell 3-1 of the MIR reactor during the period of testing these mini fuel elements. The assessment of the interaction layer volume fraction was carried out by determination of the interaction layer area on the metallographic section surface and ratio of this area to the total area occupied by fuel composition. Method of quantitative assessment of material microstructure is based on principles of quantitative metallography allowing us to determine quantitative parameters of a three-dimensional object while investigating its two-dimensional sections which presented on metallographic sections of the material specimens [4].



Fig. 10. The images of the fuel meat microstructure of various modification mini elements:

- a mini element № 3496. Atomized, matrix PA-4 (Si<0.4%), cladding alloy-99
- b mini element № 2494. 2-phases U-Mo, matrix PA-4, cladding SAV-6
- c mini element № 3297. Atomized, matrix A1+2%Si, cladding alloy-99
- d mini element № 3597. Atomized, matrix A1+5%Si, cladding alloy-99
- e mini element № 3396. Atomized, matrix A1+13%Si, cladding alloy-99
- f mini element № 5495. Oxidized atomized particles, matrix- PA -4, cladding-alloy 99
- g mini element № 4496. Atomized with protective ZrN layer, matrix-PA-4, cladding alloy-99

For comparative assessment of the interaction layer of different pins, volume fraction of the interaction layers in the fuel meats was determined on the metallographic sections (table 5).

Mini element	3496	2494	3297	3597	3396	5495	4496
No.							
Version	1-phase	2-	A1+2%Si	A1+5%Si	A1+13%Si	Oxidized	(U-
	Si<0.4%	phases,				U-Mo,	Mo)+
		Si<0.4%				Si<0.4%	ZrN,
							Si<0.4%
Burnup, %	59	64	67	59	60	62	60
Volume	44	51	49	25	21	53	6
fraction of IL,							
%							

Table 5. Assessment of IL volume fraction in the different versions of fuel meats

The results demonstrated in table 5 can be interpreted as follows:

- for 1-phase ( $\gamma$ ) and 2-phases ( $\gamma$ ,  $\alpha$ ) status of U-Mo particles with no Si in the matrix material the IL fractions by volume differ slightly considering the difference in the burn-up and are within the limit (44-51) %;

- adding 2 % Si to the aluminum matrix does not result in a considerable reduction of intensity of interaction between the fuel and the matrix, the IL volume fraction  $\sim 49\%$ ;

- 5% content of Si considerably reduces the IL fraction in the meat volume, approximately 2 times less compared to the matrix without Si ( $\sim$ 25% against  $\sim$ 50%);

- considerable increasing up to 13% of Si contents in the Al matrix does not bring to an adequate reduction of the interaction layer: ~ 21% fraction by volume of the interaction layer compared to ~ 25% at 5% contents of Si. Probably, the effect of high contents of Si will be revealed at reaching the values of burn-up higher than ~ 60%;

- oxidizing of U-Mo particles does not affect the interaction between U-Mo alloy and the aluminum matrix, the fraction of the IL is on the level of particles without oxidizing ~ 53%;

- the application of ZrN coating on U-Mo granules leads to an essential reduction of the interaction layer, the fraction by volume in the fuel element meat constitutes only ~ 6%, being 8-9 times less than in the Si-free matrix. It is noteworthy that such value could be less, as the interaction between U-Mo particles and the aluminum matrix occurs mostly on the surface of the particles where the ZrN layer is absent.

To formulate final conclusions in terms of influence of Si contents in the matrix aluminum and matrix coatings applied to U-Mo granules on the processes of interaction between the fuel and the matrix it is necessary to perform additional investigations using SEM and EPMA and probably other procedures.

# 3. Conclusions

1. The results of post-irradiation examination of the full-scale experimental FAs based on low enriched U-Mo fuel of IRT-3M, consisting of six tube-type fuel elements of square section with a coaxial arrangement, and also of IRT-U FAs, consisting of a bundle comprising 172 rod-type fuel elements of square section irradiated in the MIR reactor up to the mean burn-up U-235  $\sim$  40% at parameters typical for fuel application in Russian-made pool-type reactors demonstrated as follows:

- the state of transversal sections of the fuel elements' segments with the maximum power is satisfactory;

- no cracks or other defects evidencing breaking of the claddings were discovered;

- in the fuel meat structure there is no evidence of porosity forming being a result of gas fission products accumulation;

- U-Mo granules' impact on the process of interaction with the matrix was not revealed.

2. During the test of the  $2^{nd}$  full-scale FA with the tube-type IRT-3M fuel elements after reaching the mean burn-up U-235 ~ 50% were discovered the signs of fuel elements' cladding failure by the readings of the standard system of the fuel element claddings leak test which is based on delayed neutrons registration. Following the requirements of the program testing of that FA was stopped, and it is planned to start the post-irradiation FA examination in late 2009.

3. The post-irradiation examination of mini fuel elements with one- and two-phase U-Mo fuel with different contents of Si in the matrix aluminum and with protective coatings on the granules tested up to  $\sim 65\%$  burn-up of U-235 to examine the interaction between the fuel and the matrix material proves as follows:

- both at 1-phase ( $\gamma$ ) and 2-phases ( $\gamma$ ,  $\alpha$ ) status of granular increase particles there occurs an intensive interaction between the fuel and the matrix, the interaction layer fraction by volume reaches ~ 50%;

- 5% contents of Si in the matrix AL is optimum for reduction of the intensity of interaction between the fuel and the matrix, along with that a 2-time reduction of the interaction intensity occurs. The increase of Si contents in the matrix up to 13 % does not bring to an essential reduction of intensity of (U-Mo) - Al interaction;

- the oxidizing of particles does not reduce the intensity of interaction between U-Mo granules and the Al matrix;

- there is a significant reduction up to (8-9) times of the intensity of interaction between U-Mo granules and the matrix material with ZrN protective coating applied to their surfaces. This process could solve the problem of interaction between the U-Mo alloy and the Al matrix without Si adding.

4. The tests of the full-scale IRT-U with rod-type fuel elements and of rod-type mini fuel elements with solid fuel are in progress in the MIR reactor.

### References

1. A.L. Izhutov, V.V. Alexandrov, A.Ye. Novosyolov, Starkov V.A. et al., "Results of PIE pin type LEU U-Mo fuel elements tested in the MIR reactor", The 2006 International Meeting on Reduced Enrichment for Research and Test Reactors, Cape Town, Republic of South Africa, October 29-November 2, 2006.

2. A.L. Izhutov, V.A. Starkov V.A., V.V. Pimenov V.V. et al. "The status of tesning LEU U-Mo full-size IRT type fuel elements and mini-elements in the MIR reactor", The 2008 International Meeting on Reduced Enrichment for Research and Test Reactors, October 5-9, 2008, Washington D.C., USA.

3. A.L. Izhutov, V.A. Starkov V.A., V.V. Pimenov V.V., V.E. Fedoseev et al. "Preliminary results of testing performed in the MIR reactor for full-size IRT-type fuel assemblies with low-enriched U-Mo fuel", Proceedings of the 2007 International Meeting on Reduced Enrichment for Research and Test Reactors, Prague, Czech Republic, September, 23-27, 2007.

4. S.A. Saltykov, "Stereometric metallography" (stereology of metal materials). M.: Metallurgy, 1976.