Manufacturing and Investigation of U-Mo LEU Fuel Granules by Hydride-Dehydride Processing

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ABSTRACT

Investigations of hydride-dehydride processing for comminution of U-Mo alloys with Mo content in the range 1.9 ÷ 9.2% have been performed. Some regularities of the process as a function of Mo content have been determined as well as some parameters elaborated. Hydride-dehydride processing has been shown to provide necessary phase and chemical compositions of U-Mo fuel granules to be used in disperse fuel elements for research reactors.

Pin type disperse mini-fuel elements for irradiation tests in the loop of "MIR" reactor (Dmitrovgrad) have been fabricated using U-Mo LEU fuel granules obtained by hydride-dehydride processing. Irradiation tests of these mini-fuel elements loaded to 4 g U_{tot}/cm^3 are planned to start by the end of this year.

1. Introduction

Main efforts in frames of RERTR activities are concentrated now around studying the possibility of U-Mo alloys utilization for production disperse fuel elements on their basis. Several irradiation tests of plate type fuel elements with U-Mo fuel granules have been already conducted in USA (RERTR-1, -2 and –3) which have shown good results [1,2]. Nevertheless there are several obstacles to be overcome before such fuel can be widely used instead of dioxide and disiliside ones. One of such problems is development of method for comminution U-Mo alloys to obtain fuel granules of needed size (< 200 мкм) as mechanical crushing is not effective due to high plasticity of relevant alloys.

There already have been and are being developed several alternate methods such as centrifugal atomization with rotating disk [3] (KAERI) and with rotating electrode [4] (NZKhC, Russia). The works on studying the possibility of U-Mo size reduction by hydride-dehydride (H-D) processing are being proceeded in CNEA (Argentina) [5, 6] and in VNIINM (Russia) [7]

This paper contains some results on size reduction of U-Mo alloys with Mo content in the range 1.9 – 9.2% and manufacturing of disperse pin type mini fuel elements for irradiation tests in Dmitrovgrad (reactor "MIR").

2.Samples and equipment

Installation for conducting H-D process represented horizontal vacuum furnace with evacuating system consisting of pre-evacuation (rotary) and diffusion pumps (evacuation up to 10^{-5} mm of Hg). The furnace was equipped with inlet and outlet valves to work in hydrogen or argon atmosphere. Several separate samples could be processed to H-D at the same time and under the same conditions of experiment.

Detailed description of technological flow sheet for H-D processing of U-6.5Mo alloy has been given in reference [7]. As a whole the same process flow diagram can be employed for another U-Mo alloys with Mo content up to 10%. In general this process involves three main technological procedures:

- Preparation of needed micro-structural and phase state of the sample by appropriate heat treatment;
- Immediately size reduction by H-D process;
- Recovery of initial γ-phase state by quenching from γ-phase area of U-Mo phase diagram.

The stage of preparation is very important as it defines kinetics of hydriding and affects powder particles size distribution. Influence of chemical and phase composition of U-Mo alloy size reduction process has been studied on the samples described in table 1. Besides binary U-Mo alloys the number of U-Mo samples alloyed with small additions of Al and Sn also has been studied as well as pure U (99.8%) as a reference point.

Table 1

Chemical and phase composition of U-Mo alloys to be comminuted

№	Chemical composition of alloy (mass %)				Phase composition of alloy, type and parameters of lattice (A°)	
	U	Mo	Al	Sn	As-cast	Before H-D processing
1	99,8	-	-	-		**α-phase**; orthorhombic (a=2,859, в=5,879, c=4,971)
2	basis	1,9	-	-	**(α+γ)-phases;** orthorhombic (a=2,858, в=5,857, c=4,964) cubic, a=3,431	**α'-phase** orthorhombic (a=2,874, в=5,818, c=4,983)
3	basis	1,9	-	-	**(α+γ)-phases;** orthorhombic (a=2,856, в=5,857, c=4,964) cubic, a=3,431	**(α+γ)-phases;** orthorhombic (a=2,856, в=5,857, c=4,964) cubic, a=3,431
4	basis	1,9	0,15	0,2	**(α+γ)-phases;** orthorhombic (a=2,858, в=5,860, c=4,965) cubic, a=3,429	**(α+γ)-phases;** orthorhombic (a=2,858, в=5,860, c=4,965) cubic, a=3,429
5	basis	6,5	-	-	**γ-phase** cubic, a=3,442	**(α+γ)-phases;** orthorhombic (a=2,859, в=5.856 , c=4,968) cubic, a=3,423
6	Basis	6,5	0,37	-	**γ-phase** cubic, a=3,443 (traces of UAl_2; a=7,789)	**(α+γ)-phases;** orthorhombic (a=2,861, в=5,856, c=4,966) cubic, a=3,427 **phase UAl_2**; a=7,723
7	basis	6,5	0,15	0,2	**γ-phase** cubic, a=3,445	**(α+γ)-phases;** orthorhombic (a=2,862, в=5,854, c=4,965) cubic, a=3,429
8	basis	9,2	-	-	**γ-phase** cubic, a=3,421	**(α+γ+γ')-phases;** orthorhombic (a=2,865, в=5,851, c=4,969) cubic a=3.426; tetragonal a=3,425, c=9,872

3. Powder particles size distribution

All above mentioned U-Mo alloys weighing 140–160 g were put into separate cells of sample holder and hydrided at hydrogen pressure 2 atm and temperature 250˚C. The end of hydriding process was determined on saturation of samples with hydrogen i.e. on ceasing hydrogen absorption. In general the duration of the process depends upon the total mass of samples and in relevant test has made 8.5 hours. After hydriding the samples were dehydrided and then – γ-quenched. Powder particles size distribution has been determined by sieve analyses (table 2) [7].

Table 2

Size distribution of U-Mo powder particles obtained by H-D processing

№ of sample	Mass fraction (%)				
	0 – 63 μm	*63 – 200 μm	200 – 315 μm	315 – 1000 μm	> 1000 μm
1	86,4	11,3	2,3	-	-
2	67,2	25,7	5,2	1,9	-
3	34,7	54,6	6,9	2,9	0,9
4	53,2	32,0	11,5	3,3	-
5	12,8	56,1	17,6	10,4	3,1
6	14,4	65,3	16,1	3,0	1,2
7	18,2	66,1	13,8	1,3	-
8	2,9	21,2	27,8	39,4	8,7

* - This column corresponds to the weight fraction of fuel granules suitable for fuel element manufacturing

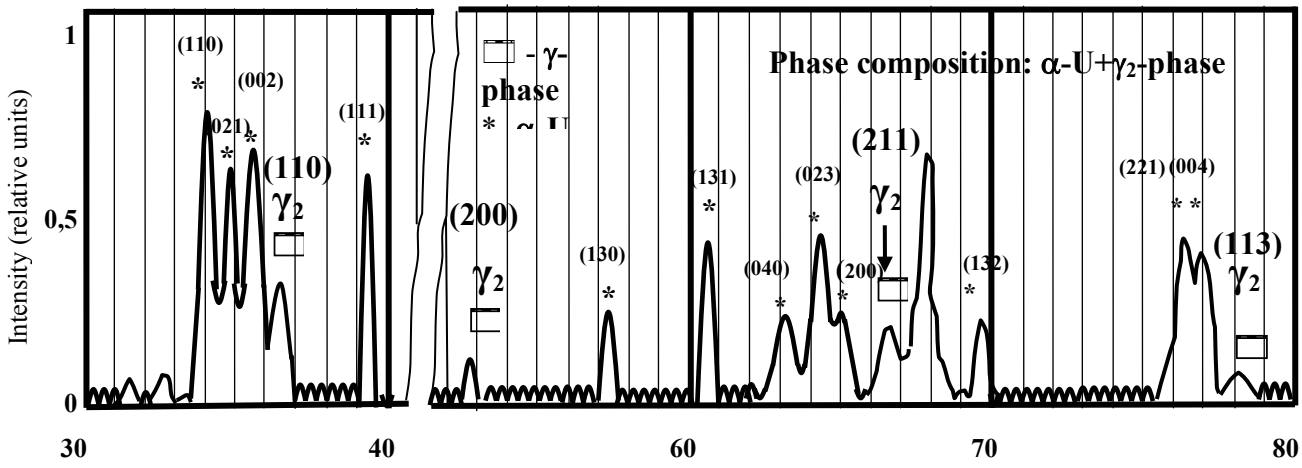
As it can be seen in the table 2, there is substantial dependence of powder particles size distribution upon the phase and chemical composition of U-Mo alloy comminuted by one-cycle H-D processing. It is obvious that increase of Mo content in the case of binary U-Mo alloys (samples № 3, 5, 8) results in significant rise of coarse particles fraction. Increase of coarse particles fraction for binary U-9,2Mo alloy is so high that to raise the amount of needed fraction it was necessary to conduct several H-D cycles (3-5 cycles). As a result it makes it possible to increase ingot-to-product yield up to the level of alloys with less Mo content. Addition of small amounts of Al and Sn into binary U-6.5Mo and U-1.9Mo alloys on the contrary promotes to easier size reduction i.e. to the growth of fine powder fraction (samples № 4, 6 and 7). Therefore in this case it is very important to conduct H-D process in such a way which will not result in excessive size reduction.

Inclination of binary U-1,9Mo alloy to the size reduction by H-D processing can be further raised by means of transformation two-phase (α+γ) state inherent to as-cast alloy into single α'-phase samples 2 and 3). Such transformation may be realized by γ-quenching U-1.9Mo into water. As a result of martensite-type transformation taking place under such treatment, α'-phase is forming, which is prone to size reduction by H-D process almost as well as pure U in equilibrium α-phase state (samples № 1 and 2).

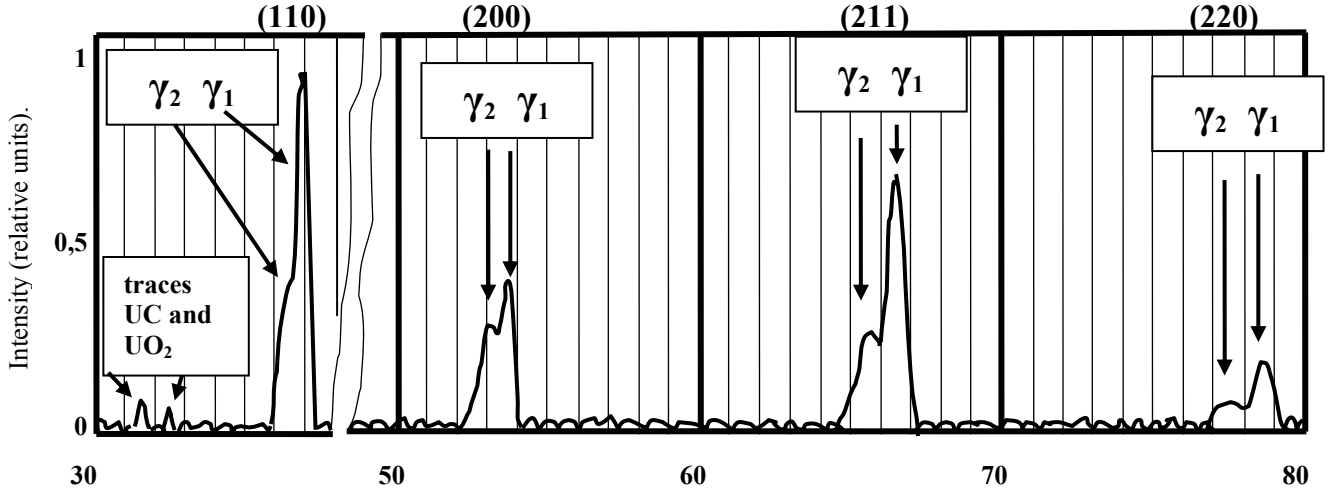
4. Phase composition of fuel granules obtained by H-D processing

Original as-cast phase state of U-Mo alloys has been – γ-phase (table 1, samples № 5, 6 7 and 8) and (α+γ)-phase (table 1, samples № 2 3 and 4). During H-D comminution the samples of U-Mo alloys were being subjected to various heat treatments affecting phase composition of fuel granules. Therefore the control of phase composition is of particular importance for characterization finished product.

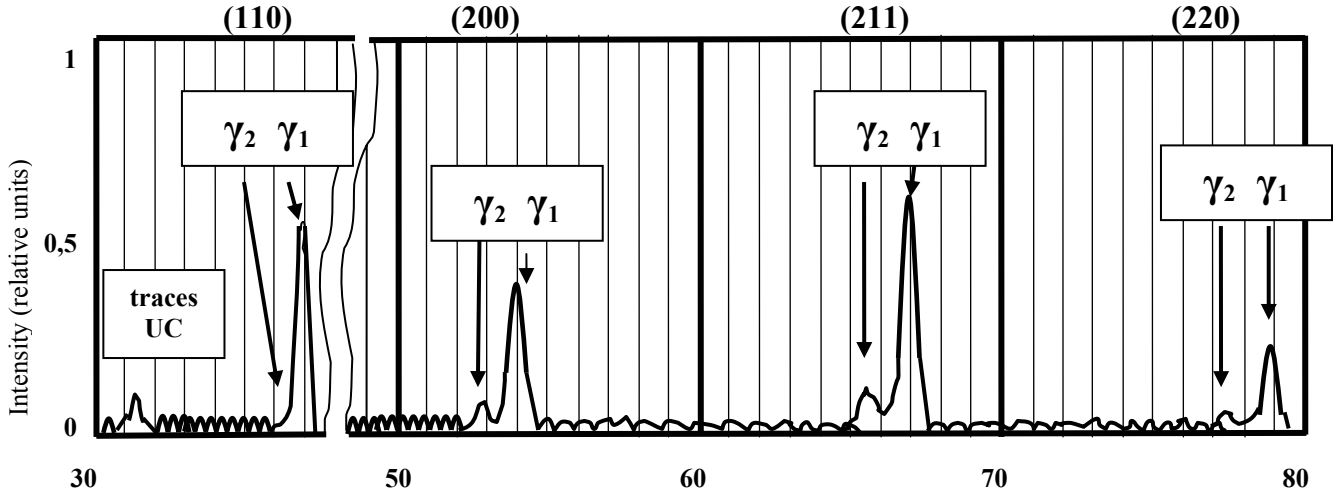
Results of X-ray analysis of fuel granules manufactured by H-D process are shown in figure 1 [6,7].



1.1 X-ray pattern of fuel granules obtained by H-D process of U-1,9Mo alloy after γ-quenching



1.2 X-ray pattern of fuel granules obtained by H-D process of U-6.5Mo alloy after γ-quenching



1.3 X-ray pattern of fuel granules obtained by H-D process of U-9.2Mo alloy after γ-quenching

As a result of comminution by H-D processing it is possible to manufacture fuel granules in metastable γ(U;Mo)-phase. But in contradistinction to γ-state of as-cast alloy, there were two γ(U;Mo)-phases with different Mo content which had formed in the powder obtained by H-D processing - γ_1-phase and γ_2-phase (fig. 1). For instance in the case of U-6,5Mo-0,2Al-0,2Sn γ_1- and γ_2-phases formed with lattice parameters a_1= 3,430 and a_2=3,467 that is with Mo content ~ 6,7% and ~ 1,6% correspondingly. Such result was confirmed with SEM equipped with X-ray spectral analyzer. These investigations of polished cross sections of fuel granules revealed the areas with different Mo contents close to that determined by X-ray analysis.
Similar X-ray pattern has been inherent to fuel granules of U-9.2Mo alloy (fig. 1.3) The only difference was in lattice parameters of γ_1- and γ_2-phases (a_1=3,414 and a_2=3,465) and in the ratio of intensity of X-ray peaks with identical Miller indexes relating to both phases. The latter means that volume ratio of these phases also is different. Maximum amount of γ_2-phase with Mo content ~ 2,1%, was in alloy with 1,9%Mo and decreased with Mo content increase.
These regularities can be explained analyzing all stages of H-D process as applied to the position of each alloy on the phase diagram. It is obvious that maximum amount of equilibrium α-phase (with Mo content < 0,3%) is forming during decomposition of U-1.9Mo, and minimum – in U-9.2Mo alloy. As hydriding takes place through α-phase then some spatial splitting of α- and γ-phase apparently has to occur during powder formation and therefore α-phase should transform into appropriate γ_2-phase at repeated γ-quenching
Thus on one hand to increase irradiation stability of U-Mo fuel granules manufactured by H-D processing it is necessary to raise Mo content. This results in increase of γ_1-phase amount with high Mo content and decreases γ_2-phase amount with significantly smaller Mo content which is apparently less stable under irradiation. On the other hand to improve manufactureability and efficiency of H-D processing and to increase the density of fuel requires decreasing of Mo content in the alloy.
Taking into account foregoing, some "compromise" composition can be probably chosen as an "optimal" one.

5. Manufacturing of mini fuel elements for irradiation tests.
Low enriched U-Mo alloy (enrichment-19.7%) with Mo content 7.3mas%, alloyed with small additions of Al and Sn (~ 0,2% each) has been comminuted by H-D processing to manufacter fuel granules. More than 72 disperse pin-type mini-fuel elements (fig. 2) have been fabricated for lifetime tests in the reactor "MIR" up to burn-up 45% and 70%. Irradiation tests will have started by the end of this year.

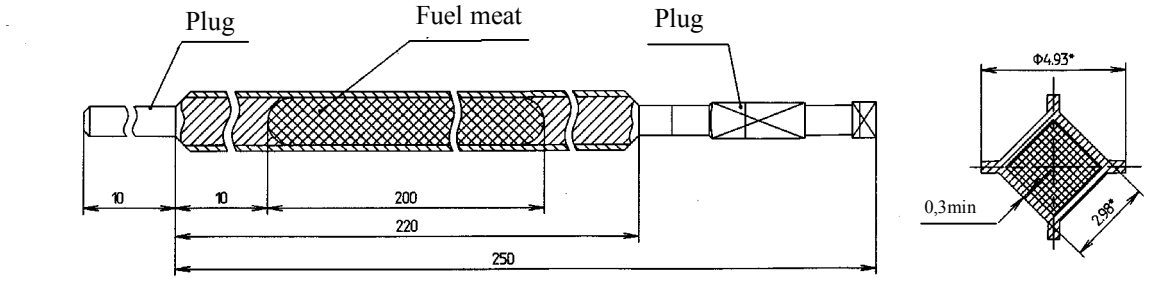


Fig. 2.Design of mini-fuel element for irradiation tests in "MIR" reactor.

Conclusions

1.Some regularities of U-Mo alloys size reduction by H-D processing has been studied. As a result it has been shown that:

- Chemical and phase composition of U-Mo alloy significantly affects the efficiency of size reduction process;
- Introduction of small additions of Al and Sn (~ 0,2% each) into binary U-Mo alloys raises inclination to size reduction (growth of fine particles fraction) after singly cycle of H-D processing;
- Transition of U-1,9Mo alloy into α'-phase state by martensite transformation increases an capability to size reduction by H-D processing;
- Phase composition of U-6,5Mo and U-9,2Mo fuel granules represents two-γ-phase microstructure consisting of γ_1- and γ_2-phases with different Mo content.

2.LEU U-Mo fuel granules with 7,3mass% Mo and small alloying additions of Al and Sn have been manufactured by H-D processing. Disperse pin-type mini-fuel elements on the basis of these fuel granules have been manufactured for irradiation tests in "MIR" reactor.

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