Interaction UMo fuel with Fe and FeCr

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Abstract. Uranium-molybdenum alloys are perspective nuclear fuel for fast reactors. In this work, a study was conducted of the interaction of uranium-molybdenum alloy with iron and chromium steel at an elevated temperature of 750 °C for 5 hours. It was found that the constant rate of the interaction layer growth for diffusion couple UMo/FeCr is about $5.4 \times 10^{-12}$ m$^2$/s at 750 °C. The phase composition of the interaction layers for the both diffusion couples was determined. The interaction comes along the grain boundaries, there are not interacts UMo alloy grain in the structure of the diffusion zone.

1. Introduction

During the operation in a nuclear reactor metallic fuel comes into close contact with the cladding of stainless steel, because of swelling which contributes to diffusive reacted fuel cladding at a high temperature to form a liquid phase and results in the destruction of the fuel element [1–4]. One of the possible ways to improve compatibility and reduce the swelling of metallic fuel is uranium molybdenum alloying [5–7]. In the case of uranium-zirconium alloy there is a redistribution of the alloy components, migration of the uranium, zirconium, reducing the local temperature of the melting point [3]. Based on the enthalpy of formation of binary mixtures of Mo and Zr with Fe, which is an approximate measure of the reactivity between the two elements shows that Mo has a lower enthalpy (in absolute value) than Zr. That said that Mo is less active in the interaction with a cladding compared to Zr [4]. However, at present there is no clear understanding of the processes occurring in the interaction of uranium-molybdenum fuel with material cladding of fast reactor made of stainless steel. Therefore, the main objective of the work is the experimental determination of the effect of alloying molybdenum nuclear fuel on the nature and kinetics of its interaction with iron and chromium steel.

2. Experiment

Alloys U10 wt.% Mo, Fe, Fe13 wt.% Cr hereinafter U10Mo, Fe13Cr respectively prepared by melting a batch of metal chips in an arc furnace with non-consumable tungsten electrode in an atmosphere of purified argon. To prepare the alloys used pure uranium (99.98), distilled molybdenum, iodine chromium, carbonyl iron and high purity Al (99.999). The batch melted several times to improve the homogeneity of the ingot. All alloys were annealed in vacuum at 1000 °C for one day for homogenizing. The obtained alloy was cut into tablets of 3 mm thickness at electric discharge machine, then subjected to grinding followed by polishing with diamond paste to 1 µm. Follows from the obtained "tablets" samples were taken for analysis of homogeneity and to determine the exact composition of the obtained alloy. To carry out the diffusion annealing, uranium alloy tablets and stainless steel were clamped together between the molybdenum discs, followed by annealing in a
quartz ampoule. Before annealing to prevent possible oxidation of the getter vial laid chip zirconium iodide. Then the vial drops diffusion assembly and vial is sealed, evacuated. Diffusion annealing is conducted in a tube furnace at temperature 750 °C, for 5 hours. After completing annealing time the quartz ampoule is placed in a bath of running cold water. Then diffusion couples were cut perpendicular to the direction of diffusion in the electric spark machine, the use of which will allow to cut the diffusion couples without destroying the fragile interaction layer (IL). The next step is to pour a few of diffusion couples into the resin. The resulting sample was further passed metallographic preparation. The microstructure of the surface and the elemental composition of the samples was investigated by scanning electron microscope JEOL 6610LV using energy-dispersive spectrometer Oxford Instruments INCA x-ACT. Phase composition layer interaction in diffusion pairs was determined by quantitative relations (U+Mo)/(Fe+Cr) in phase.

3. Results

3.1. The interaction between U10Mo and Fe
Analysis of the structural-phase state of the interaction layer in diffusion couple U10Mo/Fe after annealing at 750 °C for 5 hours showed that the reaction layer may be divided into the following areas (Figure 1 a-d):
1. (U,Mo)Fe2;
2. (U,Mo)Fe2 + (U,Mo)6Fe;
3. binary eutectic (U,Mo)6Fe + (U,Mo)Fe2;
4. U6Fe + 90wt.% Mo + binary eutectic (U,Mo)6Fe + (U,Mo)Fe2;
5. binary eutectic {(U,Mo)6Fe + (U,Mo) Fe2} + U6Fe;
6. binary eutectic {(U,Mo)Fe2 + (U,Mo)6Fe} + grain U10Mo + U6Fe.

At the initial stage of interaction comes at the expense iron diffusion along grain boundaries U10Mo, which is confirmed by unreacted grains of uranium-molybdenum alloy (Figure 1 d). However, due to the fact that the temperature of isothermal annealing at 750 °C which is 25 °C above the temperature of formation of eutectic U6Fe + UFe6 for binary system uranium-iron character of interaction is different from below the eutectic to form a eutectic phase of rapid interaction begins. Maximum width of the interaction layer region from 200 to 300 µm is U6Fe + 90 wt.% Mo + binary eutectic (U,Mo)6Fe + (U,Mo)Fe2. A special feature of this area are the particles enriched more 90wt.% Mo, the amount of which increases gradually with the advance deep into the alloy fuel. One of the main characteristics of the interaction of the shell material with fuel is a penetration depth of the fuel cell in cladding which in of diffusion couple U10Mo/Fe is about 200 µm at 750 °C for 5 hours.

3.2. The interaction between U10Mo and Fe13Cr
As can be seen from the microstructure of the interaction between the layer of chromium steel and uranium-molybdenum fuel (figure 2 a-d), occurs due to diffusion of elements of the shell U10Mo grain boundaries to form ternary eutectic (U,Mo)Fe2 + μ + UFe6, where μ 80% U-6% Mo-11% Fe-3% Cr. However, from the fuel alloy there is a change in the mechanism of grain boundary volume, as a result we observe the so-called "feathers" (figure 2 d), while in the border zone is observed only UFe6. All interaction layer can be divided into five areas:
1. (U,Mo)(Fe,Cr)2;
2. (U,Mo)(Fe,Cr)2 + μ;
3. ternary eutectic UFe6 + (U,Mo)Fe2 + μ;
4. grain U10Mo + ternary eutectic (U,Mo)Fe2 + μ + UFe6;
5. UFe6 + U10Mo.

Chromium rich layer is not formed due to the high temperature annealing. The thickness of the reaction layer in a diffusion couple is about 700 microns at a temperature of 750 °C for 5 hours of isothermal annealing.
Figure 1. (a) The microstructure IL for diffusion couple U10Mo/Fe after annealing at 750 °C for 5 hours; (b) limit of Fe/IL; (c) the boundary between the areas 4 and 5; (d) between the boundary areas 5 and 6.

The depth of penetration of the fuel components in the shell of diffusion couple U10Mo/Fe13Cr is about 60 microns. Based on the concepts of a parabolic law of growth of the interaction layer ($X^2 = Kt$, where $X$ - thickness interaction, $t$ - annealing time) for the first time defined the growth rate constant ($K$) for diffusion couple U10Mo/Fe13Cr, which is about $5.4 \times 10^{-12}$ m$^2$/s at 750 °C. Comparison of the results is impossible due to lack of data on high-temperature interaction U and UZr with FeCr.
4. Conclusions
In this work, research was conducted of the interaction of uranium-molybdenum alloy with iron and chromium steel at an elevated temperature of 750 °C for 5 hours. It is found that the rate constant of the interaction layer growth for diffusion couple UMo/FeCr is about $5.4 \times 10^{-12} \text{m}^2/\text{s}$ at 750 °C. The phase composition of the interaction layers for the both diffusion couples was determined. The interaction goes along the grain boundaries, as evidenced not interacts UMo alloy grain in the structure of the diffusion zone.

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References
[1] Otsuka S, Kaito T, Ukai S and Inoue M 2011 J. Nucl. Mater. 441 286
[2] Nikitin S N, Shornikov D P, Tarasov B A and Baranov V G 2015 Tsvetnye Metally 3 36 (in Russian)
[3] Hofman G L, Walters L C and Bauer T H 1997 Progress in Nuclear Energy 31 (1/2) 83
[4] Huang K, Park Y, Zhou L, Coffey K R, Sohn Y H, Sencer B H and Kennedy J R 2014 J. Nucl. Mater. 451 372
[5] Cohen A B, Tsai H and Neimark L A 1993 J. Nucl. Mater. 204 244
[6] Aitkaliyeva A, Madden J W, Miller B D, Papesch C A and Cole J I 2015 J. Nucl. Mater. 464 28
[7]  Jia Jian-ping, Wang Zhi-gang, Chen Miao, Wang Xi-sheng, Zhang Peng-cheng and WU Sheng 2013 *Atomic Energy Science and Technology* **47** (2) 295

[8]  Kim Y S, Hofman G L and Yacout A M 2013 *J. Nucl. Mater.* **441** 520