Influence of a doping by Al stainless steel on kinetics and character of interaction with the metallic nuclear fuel

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Abstract. Metallic nuclear fuel is a perspective kind of fuel for fast reactors. In this paper we conducted a study of the interaction between uranium-molybdenum alloy and ferritic-martensitic steels with additions of aluminum at a temperature of 700 °C for 25 hours. The rate constants of the interaction layer growth at 700 °C is about \(2.8 \times 10^{-14} \text{ m}^2/\text{s}\). It is established that doping Al stainless steel leads to decrease in interaction with uranium-molybdenum alloys. The phase composition of the interaction layer is determined.

1. Introduction
Metallic nuclear fuel is a promising fuel for fast reactors because of its high thermal conductivity, and the ability to achieve high burnup. However, the use of this type of fuel is limited by its strong swelling and physico-chemical interaction fuel with the stainless steel, whereby the fuel element is out of order [1-4]. Currently, the main type of metal fuel is uranium-zirconium alloys. However, in the case of uranium-zirconium alloy there is a redistribution components of the alloy, migration of the uranium, zirconium, reducing the local temperature of melting [1]. A more reasonable way to improve interoperability and reduce the swelling of metallic fuel is uranium molybdenum alloying [5-7]. Molybdenum has a lower enthalpy of formation of binary mixtures with Fe (in absolute value) than Zr [6].

Also great interest are the high-chromium steel with the addition of aluminum, the main purpose of which is the confrontation corrosion in lead coolant [8-10], through the formation of interlayers of aluminum oxide. Corrosion tests of this kind of steel in lead showed good resistance. The mechanical properties of such alloys can improve by dispersion strengthening. Also noted that such steels inherent low swelling under irradiation damaging dose of 200 to dpa. In addition, tests of chromium steels alloyed with aluminum U-Zr fuels demonstrated the formation of a liquid phase at a higher temperature compared with undoped chromium steel [8]. Increase compatibility is due to the stabilization of \(\alpha\)-Zr layer at the interface reaction layer, as well as a partial substitution of aluminum by iron lattice sites, which has a low diffusion mobility of the alloy in the fuel, thereby reducing interdiffusion. Also, introduction the aluminum contributes to the suppression the influence of fission products on the interaction [8]. Also alloying aluminum allows to obtain protective coatings by nitriding the inner surface of the cladding of fast reactor [3]. In this regard, interest testing aluminum-containing chromium steel with U-Mo fuel.
2. Experiment
Alloys U-10 wt.% Mo, Fe-13 wt.% Cr-4 wt.% Al (further designated U10Mo and Fe13Cr4Al) respectively prepared by melting of pure metal chips in arc furnace with non-consumable tungsten electrode in 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 avoid possible oxidation laid getter of the chip zirconium in the ampoule. Then into the ampoule is lowered diffusion assembly and sealed ampoule, evacuated. Diffusion annealing was performed in a tube furnace, at the temperature 700 °C, the duration of excerpts of 25 hours. Upon completion of annealing time quartz ampoule was placed in a bath of cold running water. Further 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 brittle interaction layer (IL). The next stage is to pour a few pairs of diffusion into the resin. The sample was subsequently 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 of the interaction layer in the diffusion couple was determined by quantitative relations (U+Mo)/(Fe+Cr) in the phase.

3. Results
Figure 1 (a-f) shows the microstructure of the interaction layer in the diffusion couple U10Mo/Fe13Cr4Al after annealing at 700 °C for 25 hours. From the analysis of the microstructure can be seen that the eutectic UFe₂ + U₂Fe offline, and the thickness of the interaction region (X) is 50 microns. Proceeding from concepts of a parabolic law of growth of the interaction layer (X²=K·t, where t-time annealing) first determined the growth rate constant of the interaction layer K=2.8·10⁻¹⁴ m²/s. Based on the distribution of elements over the width of the interaction layer in the diffusion couple U10Mo/Fe13Cr4Al (figures 1b-f, and 2) it can be concluded that the whole interaction layer represents the layered structure comprising four areas:

1. Enriched chromium and aluminum layer Cr₂Al;
2. (U,Mo)(Fe, Cr, Al)₂;
3. Enriched molybdenum layer;
4. α-U+ U10Mo.

Areas enriched chromium and molybdenum with aluminum, is only about 10 microns and are the diffusion barriers, preventing further interaction the fuel with cladding material. Rich molybdenum layer is composed of one-third of uranium. As can be seen from the microstructure of the diffusion zone (figure 1 a), at the boundary layer interaction/U10Mo located two phase region. However, EDX analysis of this area did not reveal it in the presence of components of the material cladding. From maps of the distribution of molybdenum to the width of the interaction layer (figure 1 e) shows that in this area are observed phase with a low content of molybdenum in some places.

Molybdenum-rich layer is formed due to lower solubility of molybdenum in the γ-uranium with increasing chromium content. Also due to the low mobility of the chromium alloy is formed in the fuel rich chromium and aluminum layer, which is a Cr₂Al with some solubility of iron and uranium.

Comparison of the kinetics and the nature of the interaction FeCrAl with uranium-zirconium alloy shows the advantage of molybdenum-uranium alloys. However, co-doping with aluminum and chromium does not solve the problem of the interaction of the fuel with the cladding at a high
temperature at which a liquid phase and diffusion processes take place in a few hours. Therefore, by increasing the appropriate compatibility at high temperatures is a protective coating on the inner surface of the cladding.

![Image](https://example.com/image1.png)  
(a)  
(b)  
(c)  
(d)  
(e)  
(f)  

Figure 1. The microstructure of the interaction layer in the diffusion couple U10Mo/Fe13Cr4Al after annealing at 700 °C for 25 hours (a); map the distribution of elements across the width of the interaction layer for iron (b), aluminum (c), chromium (d), molybdenum (e), uranium (f).

One of the promising methods of coating is a chemical-thermal treatment of the inner surface of the cladding with the saturation of the gas phase with oxygen and nitrogen. [3].

As a result, the nitriding process on the steel surface is formed layer of small-disperse intermetallic compounds, whose thickness is about 25 microns. Annealing at 850 °C for 5 hours showed absence of interaction between the metal fuel and the cladding fast reactor.
4. Conclusions
In this paper, conducted a study peculiarities of diffusion interaction between uranium-molybdenum alloy and ferritic-martensitic steels with additions of aluminum at a temperature of 700 °C for 25 hours. Determined the rate constants of the interaction layer growth at 700 °C, constitutes $2.8 \times 10^{-14}$ m$^2$/s. It is established that the stainless steel with aluminum doping leads to lower interaction with uranium-molybdenum alloys. It is shown that the interaction UMo/FeCrAl at 700 °C for 25 hours in the diffusion zone formed molybdenum rich layer, due to reduced solubility of molybdenum in the $\gamma$-uranium at increase chromium content. Also, due to the low mobility of chromium in the fuel alloy, a thin intermetallic layer Cr$_2$Al formed.

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