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THE RESEARCH STATUS OF U-MO ALLOY FUEL IN NPIC

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ABSTRACT

U-Mo alloy fuel is the research focus on new-type fuel for research and test reactor. The research status and progress of U-Mo alloy fuel in nuclear power institute of China (NPIC) is produced in this paper. U-Mo alloy dispersion-type fuel was started to study since 2006. In several years, much works have been done, including the smelting of U-Mo alloy, the preparation of γ-phase U-Mo alloy powder, the fabrication of (U-Mo) - (Al-Si) dispersion fuel plate, the compatibility between U -Mo alloy and cladding or matrix materials such as pure Al, Al-Si alloy, pure Nb, Zr alloy and pure Mg. In addition, the methods of elements analysis and non-destructive examination for U-Mo alloy fuel plate were studied. Based on these research works, the improved U-Mo dispersion fuel plates have been fabricated. The NPIC plans to master the fabrication technology of improved (U-Mo) - (Al-Si) dispersion fuel plates which satisfy the technical requirements by 2010.

Key words： U-Mo alloy; research status; powder preparation; fuel plate fabrication

1. Introduction

Due to higher density of uranium, γ-phase stability, excellent irradiation performance and simply post-processing, U-Mo alloy fuel has become the focus of RERTR ("Reduced Enrichment for Research and Test Reactors") program developing low-enriched uranium fuel. From the mid-1990s, U-Mo alloy fuel has been developed in many countries: United States, France, Russia, Argentina, South Korea, Japan and so on. With international co-operation, a large number of theoretical and experimental works for U-Mo alloy fuel were carried out, such as U-Mo alloy physical metallurgy (raw materials, smelting, phase-transformation and stability of the γ-phase), fuel fabrication, irradiation examination, and fuel post-processing. However, United States and other countries found that the (U-Mo)-Al dispersion fuel plates got a serious pillowing swelling under high burn-up and high-flux irradiating conditions in 2003. Failure analysis showed that the swelling was due to unstable U-Mo/Al interaction layer. To solve this problem, several methods were proposed, including fuel and matrix chemical improving, fuel-coated coating, monolithic fuel, magnesium-based fuel, as well as zirconium cladding fuel, and so on [1]. RERTR program planed to complete qualifying identification of U-Mo alloy fuel element in order to achieve its applications in- pile by the end of 2010.

In the China, investigating data shows that 202 fuel plant and China Institute of Atomic Energy have done some works on the U-Mo alloy powder preparation, but not work on fuel fabrication. NPIC is an active participant of RERTR program and developed successfully U3Si2-Al dispersion fuel element in 1990s. And NPIC have been concerned about the latest developments of international RERTR LEU fuel. (U-Mo)-Al dispersion fuel research work was started early in the early 2006, and the modified (U-Mo) - (Al-Si) dispersion fuel mini-plates were fabricated in 2009.

2. U-Mo Alloys Smelting

The surface of pure Uranium and pure Molybdenum should be cleaned to eliminate oxid. According to the composition of U-10wt%Mo, the mass percent of Mo element is 10～11wt% in the mixture table for smelting because Mo element is easy to vaporize. Graphite mold should be coated by the mixture of Y_2O_3 powder, CMC powder water glass to prevent Carbon filter into the U-Mo alloy liquid at the high temperature. U-10Mo alloy was smelted in 5Kg vacuum-induction furnace and the liquid is quickly cooling in casting way.

The casted U-Mo ingot is showed in fig.1. Analysis shows that the ingots are compact and its shrinkage void is not deep. Chemical analysis shows that casting ingots has good uniformity, and Mo content is accurately control in less than ±1wt% and composition uniformity in less than ±0.5 %. XRD analysis showed that the casting ingot was comprised of γ-phase.

Fig. 1 The picture of casting U-Mo alloy ingot

3.γ-U-Mo Alloy Powder Preparation

HMD (Hydrogenation-Milling-Dehydrogenation) process is used for γ-U-Mo powder preparation. The γ-U-Mo powder is prepared in the HMD integral equipment. The work of hydrogenation, milling, and dehydrogenation for U-Mo alloy could be well carried out in the HMD integral equipment. At the same time of simplifying experiments works, the HMD integral equipment could effectively prevent the oxygen and nitrogen in the air to contact

U-Mo alloy because the equipment was full of 99.999% purity argon gas at any time in the process of powder preparation. The HMD integral equipment is showed in fig.2. The equipment was comprised by heating-furnace system, vacuum acquisition system, an operation glove-box, and Argon Hydrogen supply system.

U-Mo According to U-H phase diagram and experimental works，the preparation parameters of kg-level γ-phase U-Mo alloy powder are determined. The hydrogenation of U-Mo alloy is the key process because γ-phase U-Mo alloy is difficult to be hydrogenated parameters. A kilogram of U-Mo alloy ingot was put into the Hydrogenation-Milling-Dehydrogenation integral equipment for hydrogenation at the temperature of 100 ~ 250 ℃ and the hydrogen pressure of 0.14 ~ 0.4MPa for 10 ~ 15 hours.

Fig. 2 The picture of HMD integral equipment

The appearance of hydrogenated U-Mo alloy is shown in Figure 3, the majority of the ingot was lamellar fragmentized and the hydrogenation effect is good. The U-Mo hydride was mechanically crushed into powder; then hydride powders were dehydrogenated at 300 ~ 500 ℃ for 2 ~ 6h.
Finally the coarse powder (44 μm <particle size <149 μm) and small powder (particle size <44 μm) was separated by the 110 standard sieve and 325 standard sieve. Chemical analysis showed the Hydrogen content in U-Mo powder is below 30μg/g, Oxygen content is below 3000μg/g, Nitrogen content is below 150μg/g. XRD phase analysis showed that U-Mo powder is comprised of the γ phase, as shown in Fig.4.

Fig .3 The appearance of U-Mo alloy ingot after hydrogenation

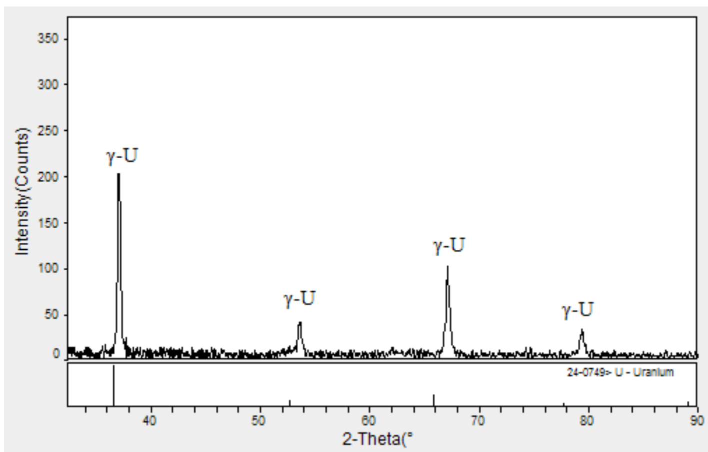

Fig. 4 The XRD pattern of U-Mo alloy powder

4 (U-Mo)-Al Dispersion Fuel Manufacturing

The international latest result showed that the improved fuel plate with Al matrix containing more than 2wt% Si behaves promising irradiation performance under higher burn-up, and this would effectively solve pillow swelling problems of the U-Mo alloy dispersion fuel. Monolithic fuel plates with the pure Al or Al alloy clad with low Si content exposed its limitations at high burn-up. Hence, NPIC has developed improved (U-Mo)-Al dispersion fuel plate fabrication technique, as shown in Fig.5, in which the meat uranium density is 5.5 ~ 7.5gU/cm^3, and the Si content added to Al matrix is 2wt % and 5wt%.

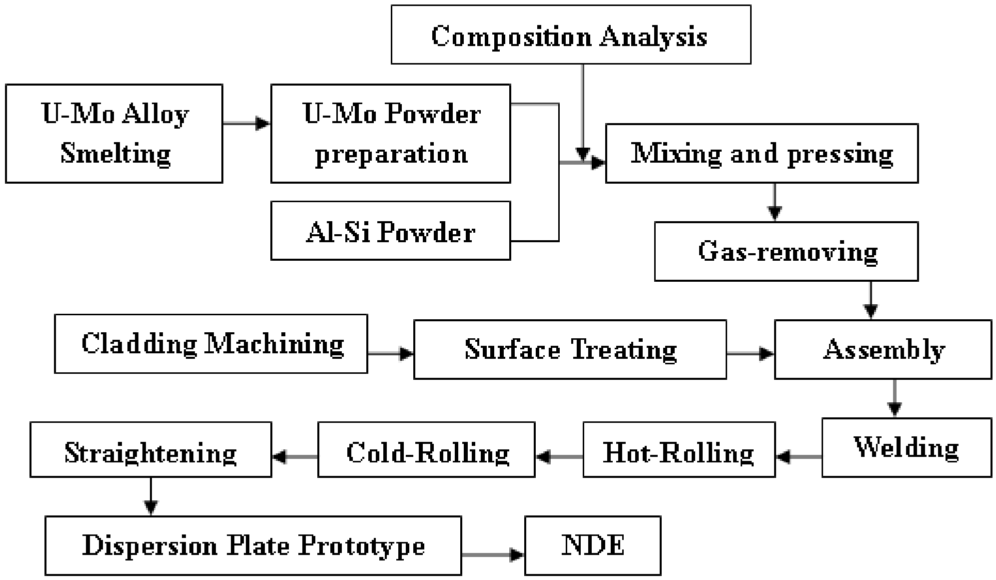

Fig. 5 Fabrication technique of improved（U-Mo）-Al dispersion fuel plate

4.1. Burden calculation

Meat material includes U-Mo powder, Al powder, Si powder (used to prevent U-Mo and Al over-reaction), and the weight ratio of different powders are determined by the meat uranium density(ρ_{mu}), the amount of uranium loading(m_{mu}), and meat volume(v_{mu}) , and needs to take into account the porosity(p) . Its calculation formula is as follows:

$$m_{umo} = \rho_{mu} * v_{mu} / w_u$$

$$m_{Al} = (\rho_{Al} w_{Al} + \rho_{Si} w_{Si}) * p * (v_{mu} - m_{umo} / (\rho_U w_u + \rho_{mo} w_{mo}) * p) * w_{Al}$$

$$m_{Si} = (\rho_{Al} w_{Al} + \rho_{Si} w_{Si}) * p * (v_{mu} - m_{umo} / (\rho_U w_u + \rho_{mo} w_{mo}) * p) * w_{Si}$$

Among the formula, m_{umo}: U-Mo powder weight; m_{Al}: Al powder weight ; m_{Si}: Si powder weight; w_u: U weight ratio in U-Mo alloy; w_{Al}: Al powder weight ratio in (Al, Si) powder; ρ_u: U density; ρ_{mo}: Mo density; ρ_{Al}: Al density; ρ_{Si}: Si density.

4.2. Mixing

U-Mo powder, Al powder, Si powder should be mixed evenly before pressing. Three kinds of powder mixing uniformity is very important to improve the uniformity of dispersion fuel plate. In order to ensure uniform distribution of uranium density, U-Mo powder should be distributed in the Aluminum matrix as evenly as possible, otherwise it will seriously affect the performance of the fuel plate.As the density of the U-Mo powder (ρ= 17.2g/cm^3), Al powder (ρ= 2.7g/cm^3) and Si powder (ρ= 2.3g/cm^3) has large differences, three kinds of powder is more difficult to mix evenly.The particle size of three kinds of powder is required as follows: U-Mo coarse powder is accounted for 80%, U-Mo small powder is accounted for 20%; Al powder is all less than 149µm, Si powder is all less than 44μm, in which particle size less than 23µm should exceed 80%.

According to powder metallurgy principle, it is best that mixed powder volume in the mixing tank should be one-third of total tank volume, so the mixing tank volume is calculated by the loose powder volume, and the mixing tank is specially designed. Three kinds of powders are put into mixing tanks which are fixed in the mechanical mixer, the mixing time is 2 ~ 3h, the rotational speed is 100 ~ 150 turn / min. In order to prevent powder oxidation, mixing is carried out in the glove box filled with argon gas.

4.3. Meat pressing

The mixed powders were pressed in a die withstand a certain degree of suppression of the pressure and became a meat of certain density, shape, and intensity. Because there was a larger density difference between U-Mo powder, Al powder, and Si powder, if loading the mixed powder into die in the ordinary way, then the heavy particles and large particles were easy to fall into the bottom of the die which could result in re-distributed unevenly. So the special loading tool was designed to achieve non-drop loading method. U-Mo alloy dispersion fuel meat was pressed in a one-way hydraulic machine at the molding pressure of 6 ~ 10MPa for 5 ~ 10s. In order to avoid the "Hugging" phenomenon between die and meat, the die surface should be brushed the reasonable thickness of the lubricant (zinc stearate). The size of pressed meat is $30\times60\times4.05mm^3$

4.4 .Gas-removing

The meat contains a large number of gases in the sealed pore. The meat will exhaust gas during rolling process, which will affect joint between meat and cladding, therefore the gas is necessary to be removed. Gas-removing was carried out in a vacuum heat treatment furnace at 350 ~ 500 ℃ for 2 ~ 4h. The zinc stearate on the surface of meat was completely decomposed, and the gas could be considered completely removed when the vacuum in the furnace decreased down to 10^{-2}Pa.

4.5. Cladding treating

It is necessary to carry out a specially cleaning process for Cover and framework after machining processing, aiming at facilitating follow-up joint between the meat and cladding. Acid and alkaline washing method is used to removing oxide, and ultrasonic cleaning with deionized water is used to cleaning acid and alkaline.

4.6. Assembly and welding

The meat and cladding was assembled in cleaning circumstance, and then it was welded for combination in glove-box filled argon with argon-arc welding.

4.7. Hot-rolling, cold-rolling and straightening

Rolling is to make a metallurgical combination between fuel meat and cladding. After hot-rolling at 480 ℃, the plate would be cold-rolled into approximate 1.27mm thickness. The nominal thickness of the meat and cladding is respectively 0.51mm and 0.38mm. The plate was annealing to release stress for several hours in furnace. At last, the plate is straightened by straightening machine. The prototype fuel plate was shown in Figure 6.

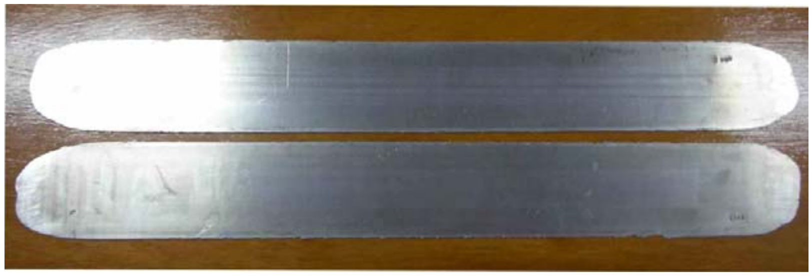

Fig. 6 （U-Mo）-（Al-Si） dispersion mini-plate

5. Fuel plate performance examination

The technical requirements of (U-Mo)-Al dispersion fuel plate fuel mainly includes: fuel plate size, Uranium uniformity in the fuel meat, and the combination quality between fuel meat and cladding. The fuel plate with uranium density of 6.5gU/cm^3 was destructed to use optical microscope to observe the microstructure, as shown in Figure 7.10 different parts were obtained from Fig.7, and the fuel plate thickness, cladding thickness and meat thickness were measured and averaged. The measured average size of the fuel plates as shown in Table 1, the fuel plate sizes satisfied the technology requirements. There are no visible cracks, voids, and no obvious reaction layer in the interface of cladding and the meat. The cladding and meat are linked closely together. Ultrasound examination also shows that the combination quality between fuel meat and cladding meets the technical requirements.

Uranium uniformity of the (U-Mo)-Al fuel plate is measured by γ-ray absorption method. The status of the Uranium distribution in uniformity zone is relatively ideal, and within ±20%, as shown in Figure 8.

The above examination results show that, (U-Mo)-Al dispersion fuel plate can basically meet the technical requirements and behave good quality.

Table1. The measured averaged size of fuel plate

尺寸	板厚度/mm	芯体厚度/mm	包壳厚度/mm	最小包壳厚度/mm
实测值	1.263	0.495	0.384	0.374

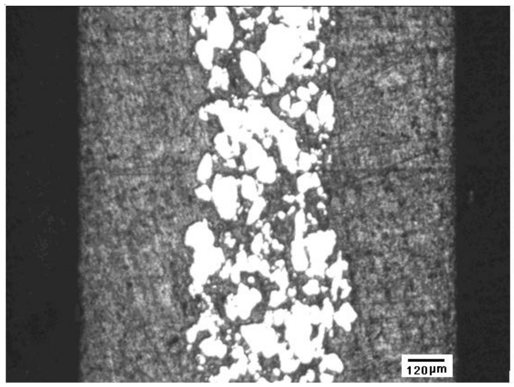

Fig. 7 The micrograph of the U-Mo dispersion plate

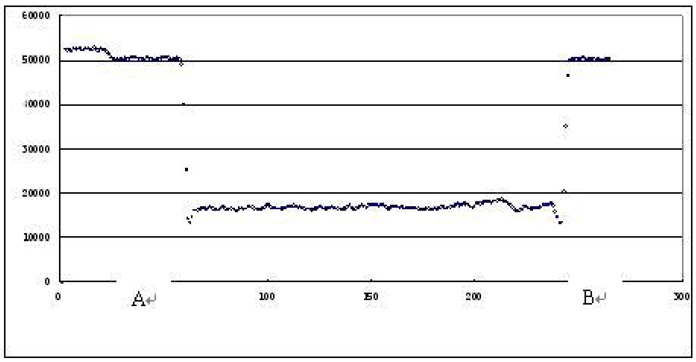

Fig .8 The Uranium distributing curve of U-Mo dispersion plate

6. Compatibility between Fuel and Matrix, Cladding, Barrier Materials

Using diffusion-couple technique, compatibility study was carried out between U-10Mo and LT24Al alloy. U-Mo/LT24 Al diffusion samples with sandwich structure was annealed in hot vacuum-pressing furnace in different conditions. Results showed that the compatibility between U-10Mo/LT24Al is not perfect. Si impurities in Al matrix enriching in reaction layer, which have given a apocalypse about improving U-Mo/Al compatibility.

To research barrier effect of Nb element between U-Mo/Al reactions, the compatibility study is carried out with U-Mo/Nb. Heat treatment temperature is respectively at 760 and 790 ℃ in different time. The study show that U-Mo/Nb compatibility below 700 ℃ is well; for 760 ℃ and 790 ℃it shows short-time compatibility. After a long time heat treatment there will be holes and cracks on the side close to U-Mo alloy. This is possibly as a result of "Kirkendall phenomenon" that unequal atomic exchange between U and Nb.

The U-Mo/Zr-4 diffusion couple was sealed in the steel frame plate by welding. The frame plate was rolled at 760～800℃ for several passes. Then the U-Mo/Zr-4 couple was annealed at 760～800℃ for 10～66h as soon as the termination of rolling. Experimental and analytical results show that, the compatibility of U-Mo/Zr-4 was good because U-Mo/Zr-4 couple was difficult to interdiffuse even at high temperature 800℃, and there were the cracks in the middle of the U-Mo/Zr-4 diffusion layer whose thickness was only about 10 μm. The components of diffusion layers on both sides of the crack were obviously different. The diffusion layer close to the side of U-Mo alloy mainly includes a Mo-rich phase which is identified as the $ZrMo_2$-based solid solution. The diffusion layer close to the side of Zr-4 alloy mainly includes a Zr-rich phase which is identified as the UZr_2-based solid solution. The crack was formed as a result of the unequal exchange of U atom and Zr atom.

7. Conclusions and Summary

In past two years, we have done such works: U-Mo alloy smelting, γ-phase U-Mo alloy powder preparation, (U-Mo)-Al dispersion fuel plate fabrication, compatibility studies of U-Mo/Al, Nb, Zr, and U-Mo/Al-Si inter-diffusion reaction, and so on. Based on this research work, some results are got, including: mastering the U-Mo alloy smelting process parameters; mastering the U-Mo alloy powder preparation process and parameters, and establishing a control method of powder particle size; manufacturing (U-Mo) - (Al-Si) dispersion fuel mini plates; carrying out compatibility studies between U-Mo and Al, Nb, Zr, and other elements, and Si added to Al matrix impacting on U-Mo/Al reaction layers. All of this will provide a support for U-Mo alloy fuel development and the ultimate success of this technology in China.

In the field of developing U-Mo alloy fuel, NPIC is latter than some countries. Basing on worldwide research results as well as self-experiences in research of dispersion fuel, NPIC plans to completely master manufacturing technique of improved (U-Mo)-Al fuel plate which meets technical requirements. The irradiation work for U-Mo fuel plate will begin at 2010-2011.

References

[1] M.K.Meryer, R.Ambrosek. Progress in the RERTR fuel development program. The 29th International Research Reactor Fuel Management (RRFM) Meeting. 2007