Melting Technology for Uniformity Control of U–10Zr Alloy

Gang Zeng, Bin Su, Daoming Chen, Yuting Zhang, Jingyuan Liu and Jian Wu

Abstract U–Zr alloy has the advantages of high thermal conductivity and anti-radiation swelling performance, as a promising metallic nuclear fuel for its fast breeder reactor. In this paper, U–10Zr alloy was prepared by multi-induction melting method and chemical analysis, optical metallography, XRD and SEM were used to investigate the effects of melting technology on the distribution of Zr as well as the phase composition in U–10Zr alloy. The results show that the dissolution of Zr in U is more complete in the second smelting process. The chemical composition is homogeneous while distribution of U, Zr and C is different in different location. The first smelting by vacuum induction removes more impurity of the ingots. The distribution of zirconium and carbon in the transverse-direction of U–10Zr ingots is more uniform. U–10Zr alloy exists in an acicular martensite state along with small quantity of nonmartensitic α phase. The zirconium element is more easy to form carbide and nitride with the C and N elements than the uranium. The as-cast U–10Zr alloy is composed mainly of α phase and δ-UZr_2 phase.

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Introduction

It is known that uranium alloyed with small quantity of elements such as molybdenum and zirconium can enhance its mechanical properties and corrosion resistance without greatly reducing its density. The U–Zr binary alloy is an important subsystem of the U–Pu–Zr ternary alloy, which has been recognized as a promising metallic nuclear fuel for its fast breeder reactor [1–5]. U–Zr as alloy in the uranium alloy system, combined with effective density design, can effectively control and solve the high temperature phase transition of metal uranium fuel, radiation swelling and many other issues [6]. U–Zr alloy has the advantages of high thermal conductivity and anti-radiation swelling performance as the promising metallic nuclear fuel for its fast breeder reactor with small quantity of zirconium added to uranium to produce fine grain alloy with good dimensional stability [7, 8]. The U–10Zr alloy in U–Zr system is the most promising metal fuel. Previous studies mainly focused on physical metallurgy, thermodynamics and kinetics of fuel phase stability as well as the evolution of U–10Zr alloy [9, 10]. However, owing to the high melting point and low density of zirconium, it is difficult to ensure the sufficient dissolution of zirconium during melting process. The zirconium element is easy to form carbide and nitride. The gathered impurity will cause inhomogeneity composition of alloy. The control of homogeneity of U–10Zr alloy that is melted by vacuum induction melting has few research. The knowledge of the uniformity of U–Zr alloy is required to evaluate stability in long-term storage and disposal of spent fuels and to stabilize the metal chip waste which is generated during its melting process. Previous studies revealed some differences in oxidation behavior between the electropolished and the mechanically polished uranium samples [11]. However, they ignored the importance of chemical composition on U–Zr properties. The uranium alloys with uniform composition and high quality is a prerequisite for the study of metallic nuclear fuel [12]. Thus, it is highly important to understand the distribution of zirconium and microstructures in the as-cast U–10Zr alloys.

Materials Preparation

Depleted uranium and zirconium bars were used for the preparation of U–10Zr alloy. The alloys were melt-cast twice in a ceramic crucible by vacuum induction melting. The zirconium bars were placed under the uranium ingots in the ceramic crucible. During the first melting process, the distribution of Zr in U–10Zr alloy was nonuniform, the zirconium alloying element was added to 10.3%. The zirconium alloying element was kept to 10% at the second melting process in order to

Table 1 The major melting technological parameters

Smelt times	Crucible	Temperature (°C)	Time (min)
1st	CaO Ceramic crucible	1600	30
2nd	CaO Ceramic crucible	1600	20
		1400	10
		1600	20

eliminate macro non uniformity. In addition, the samples were heated to 1600 °C in order to ensure homogeneity before cooling down to room temperature. The major melting technological parameters were presented in Table 1. For experimental characterization, the alloy ingots were cut into suitable shapes.

Chemical Composition and Property

The chemical composition of different parts of alloy ingots was analyzed. Standard metallographic techniques were used for grinding and polishing. 5% H_3PO_4 aqueous solution was applied for electroetching and constant 2 V DC bias was used for the etching. The samples were then repolished for XRD analysis. The XRD analysis was carried out using a Philips XRD vertical goniometer with Cu Kα radiation.

Results and Discussion

The U–10Zr alloy was melt-cast by vacuum induction melting. The alloy ingots were fused into cylinder with the size of $\varphi 80$ mm × 150 mm. The samples were cut from ingots to analyze chemical composition of zirconium and carbon. Figure 1

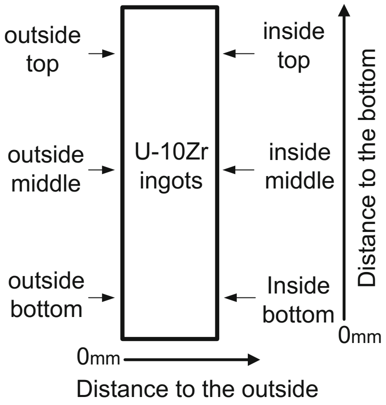

Fig. 1 Different sampling location of the as-cast U–10Zr alloys

shows the different sampling location of the as-cast U–10Zr alloy. In order to increase accuracy, the uniformity in the distribution of the alloying element was evaluated based on the different position of ingots.

Based on the analysis of constituents sampling from different locality in U–10Zr samples, the distribution of Zr and C were investigated. Figure 2 shows the chemical composition of zirconium and carbon with different distance to the bottom of U–10Zr cylinder. Zirconium was loaded on the bottom of CaO ceramic crucible, which helped to control the homogeneity of U–10Zr alloy. When the melting temperature was about 1600 °C, melting time was about 9 min, and the uranium ingots were melted totally. Zirconium bars were slowly dissolved into the liquid uranium. It showed that the general efficiency of the homogeneity controlling for Zirconium component and the carbon impurity decreasing of U–10Zr alloy were good. The first smelting by vacuum induction removes more inclusions of the ingots. The alloying effect of uranium and zirconium in the second smelting process is more complete.

Figure 3 shows the distribution of zirconium and carbon in the transverse-direction of U–10Zr ingots. The results showed that the both element Zr and C distributed evenly in the extent-direction and transverse-direction of U–10Zr ingots in the same condition. During the first melting process, the zirconium element was easy to form inclusions with carbon and nitrogen. The gathered impurity had lower density and floated up to the top of ingots. When the second melting process began, the C and N inclusions were removed. During the second melting process, the temperature should be lowered to 1400 °C after melting for 20 min at 1600 °C. Through controlling the power of induction furnace, the magnetic field had mixing effects on metal liquid. The zirconium alloying element showed no difference in distribution.

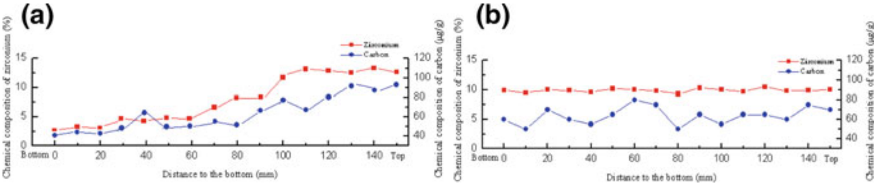

Fig. 2 Distribution of zirconium and carbon in the extent-direction of U–10Zr ingots **a** 1st smelt times, **b** 2nd smelt times

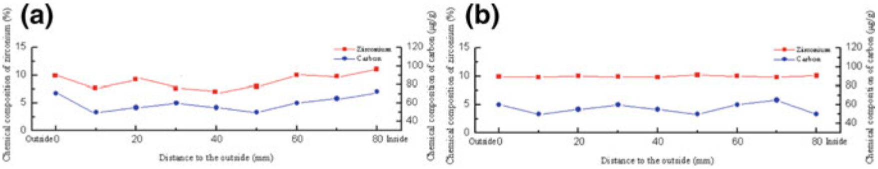

Fig. 3 Distribution of zirconium and carbon in the transverse-direction of U–10Zr ingots **a** 1st smelt times, **b** 2nd smelt times

Optical micrographs of as-cast U–10Zr alloy are presented in Fig. 4 and show that U–10Zr alloy is in an acicular martensite state along with small quantity of nonmartensitic α phase. The changes in the morphology of the U–10Zr alloy can be explained by the effects of Zr element. The martensitic plates in the samples were formed during the solidification process. In the process of alloying, uranium ingots melted slowly to avoid the rapid rise of the zirconium bars. During the first melting process, the alloy ingots were unable to dissolve zirconium bars completely. The distribution of zirconium element became uniform during the second melting process. Zirconium as alloying element caused constitutional supercooling during the cooling process, leading to the growth of dendritic microstructure. The micro-hardness of as-cast U–10Zr samples increased to 495 MPa compared with substrate uranium samples.

Scanning electron microscope (SEM) examinations of as-cast U–10Zr alloys are shown in Fig. 5. It is known to all that the microstructure contains martensitic α phase, and the dark places in Fig. 5 are composed of zirconium and its inclusions. During the melting process, zirconium as a kind of alloying element compounds with carbon inclusions. When the melting temperature rises to 1600 °C and melting time is extended to over 30 min, the zirconium inclusions are reduced.

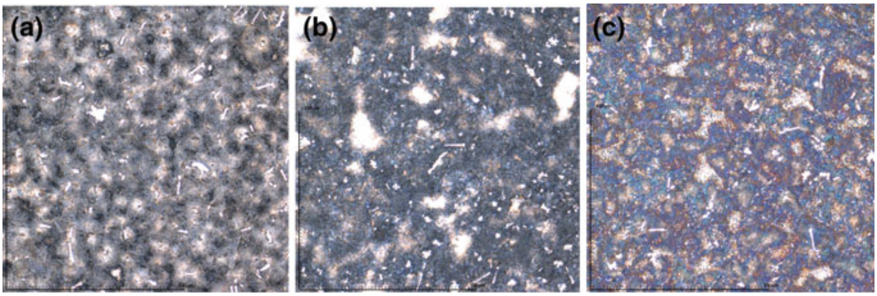

Fig. 4 Different sampling location of optical photomicrograph of the as-cast U–10Zr alloys; **a** top, **b** middle and **c** bottom

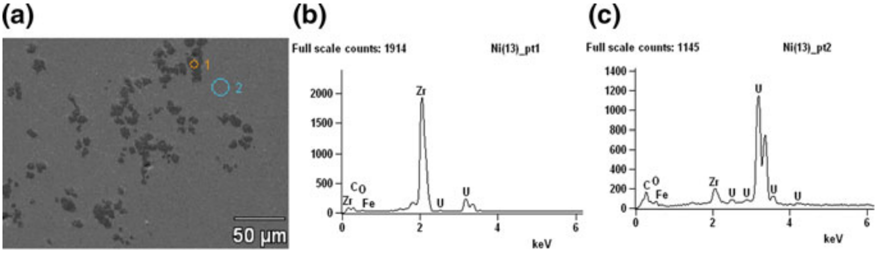

Fig. 5 SEM and EDS images of the as-cast U–10Zr alloys **a** SEM images, **b** EDS images of pt1 and **c** EDS images of pt2

Fig. 6 XRD data of the as-cast U–10Zr alloys

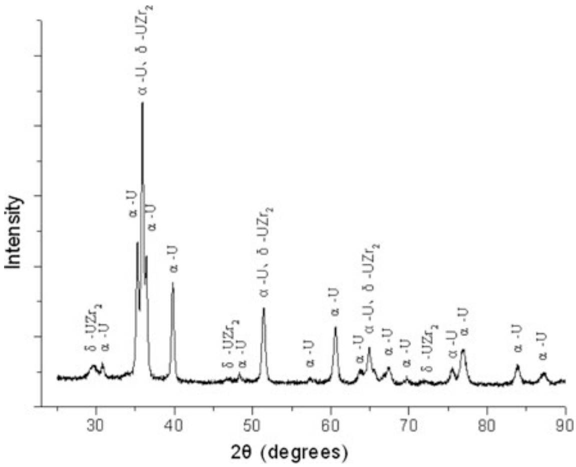

Figure 6 presents the results of XRD measurements of as-cast U–10Zr alloy. It indicates that the U–10Zr alloy is mainly composed of α phase. According to Fig. 6, three weak peaks appear in the XRD experimental spectra at 29.6°, 46.9° and 72.0°. These peaks corresponding to the δ-UZr_2 phase indicates that the δ-UZr_2 phase exists in as-cast U–10Zr alloy. Previous studies have already found the formation of δ-UZr_2 phase in as-cast U–10Zr alloy by arc melting [9]. The present study reveals that the δ-UZr_2 phase still exists in the as-cast U–10Zr alloy by vacuum induction melting. The uniform distribution of zirconium element is propitious to the stabilization of δ-UZr_2 phase.

Summary

As-cast U–Zr alloys with a Zr content of 10 wt% have been characterized by chemical analysis, optical metallography, X-ray diffraction and scanning electron microscopy. The chemical composition of ingots by the multi-induction melting method is homogeneous. The element distribution of the transverse-direction of the ingot is more uniform. As-cast U–10Zr alloy predominately consists of α phase supersaturated with Zr and δ-UZr_2 phase. Moreover, the uniform distribution of zirconium element is propitious to the stabilization of δ-UZr_2 phase.

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