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FABRICATION OF U-10WT.%Zr-RE FUEL SLUGS BY RECYCLING OF METALLIC FUEL SCRAPS

U-10wt.%Zr-5wt.%RE fuel slugs for a sodium-cooled fast reactor (SFR) were conventionally prepared by a modified injection casting method, which had the drawback of a low fabrication yield rate of approximately 60% because of the formation of many metallic fuel scraps, such as melt residue and unsuitable fuel slug butts. Moreover, the metallic fuel scraps were classified as a radioactive waste and stored in temporary storage without recycling. It is necessary to develop a recycling process technology for scrap wastes in order to reduce the radioactive wastes of the fuel scraps and improve the fabrication yield of the fuel slugs. In this study, the additive recycling process of the metallic fuel scraps was introduced to re-fabricate the U-10wt.%Zr-5wt.%RE fuel slugs. The U-10wt.%Zr-5wt.%RE fuel scraps were cleaned on the surface impurity layers with a mechanical treatment that used an electric brush under an Ar atmosphere. The U-10wt.%Zr-5wt.%RE fuel slugs were soundly re-fabricated and examined to evaluate the feasibility of the additive process compared with the metallic fuel slugs that used pure metals.

Keywords: Fuel slug, Scrap recycling, Mechanical cleaning treatment, Fabrication yield rate, Radioactive wastes

1. Introduction

The transuranic element (TRU) recovered through the pyroelectrochemical processing of spent light water reactor (LWR) fuels is used to fabricate metallic fuels [1]. The extracted TRU, including Pu and long-lived minor actinides (MA) such as Np, Am, and Cm, are used to fabricate metallic fuels [2,3]. This fuel recycling can solve the problem of pressurized water reactor (PWR) spent fuel accumulation by reducing the overall volume of PWR spent fuel and increasing the utilization of uranium resources while maintaining a high proliferation resistance [4]. U-Zr metallic fuels generally have excellent reactor safety and fuel cycle economy [5,6]. A U-TRU-Zr alloy is being developed for a sodium-cooled fast reactor (SFR) to be built in Korea [7-9].

U-TRU-Zr-RE (RE is a rare-earth alloy consisting of 53% Nd, 25% Ce, 16% Pr, and 6% La by weight) fuel slugs have been prepared with a modified injection casting using pure metals [10]. Unlike the conventional injection casting that operates under atmospheric pressure, a modified injection casting method can prevent the evaporation of volatile elements in a pressurized Ar atmosphere during the melting of metallic fuels [11]. A considerable amount of fuel scraps up to 60% of the charge amount for the injection casting has been made consisting of fuel scraps such as melt residue and unsuitable fuel slug butts [12]. The fuel scraps have been classified as radioactive waste and stored in temporary storage without recycling. It is important to reduce radioactive waste worldwide. Thus, it is worthwhile to consider the additive process of the fuel slugs [13]. Hence, the recycling process of the fuel scraps is necessary to reduce the amount of radioactive wastes and improve the utilization of the uranium resources [13].

In this study, we used a mechanical cleaning treatment method that included an electric brush to remove various impurity layers on the surface of the metallic fuel scraps. The U-10wt.%Zr-5wt.%RE fuel slugs were re-fabricated and characterized to examine the feasibility of the additive recycling process of the fuel scraps and compared with the metallic fuel slugs prepared using pure metals as raw materials.

2. Experimental

U-10wt.%Zr-5wt.%RE fuel scraps were formed after a modified injection casting of the metallic fuel slugs [13].

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Various impurity elements such as RE and Y_2O_3 coating layer existed on the surface of the fuel scraps. In order to reuse the fuel scraps to re-fabricate the fuel slugs, the fuel scraps were cleaned on the contaminated irregular surface layer with impurities. This was done by a mechanical treatment that used an electric brush variable in a rotation speed under an Ar atmosphere in a glove box system to prevent the oxidation of the fuel scraps [14]. Most of contaminated layers of the fuel scraps were removed from the surface after the mechanical cleaning treatment.

The graphite crucible was coated with Y_2O_3 plasma-spray coating layer to hinder interaction between the molten uranium alloy and the graphite crucible [15-17]. Quartz tube molds were also slurry-coated with yttrium oxide on the inner surface. Two types of charge materials composed of pure metals and the fuel scraps, respectively, were loaded into the graphite crucible. The U-10wt.%Zr-RE alloys were heated to a temperature of 1470°C before the injection casting into the quartz. Then the pressure of the melting chamber was maintained in an Ar atmosphere of about 400 torr during the heating to prevent the volatilization of the molten materials. At a predetermined superheat, the quartz tube molds were lowered in the molten alloys with the pressurization of atmospheric gas immersing the open tip into the metal melt. Major fabrication process parameters such as casting temperature, pressurization pressure, and mold preheating temperature were optimized through the casting experiments [18].

The U-10wt.%Zr-5wt.%RE fuel slugs were fabricated using clean fuel scraps and pure metals by the injection casting method, respectively [19]. The density of the metallic fuel slugs was measured using an Archimedean immersion method. The microstructure and composition of the fuel slugs were analyzed using scanning electron microscopy (SEM) with an energy-dispersive spectroscopy (EDS) to examine the microstructure and composition [20]. Chemical analyses were carried out to assess the alloy compositions of the fuel slugs using inductively coupled plasma atomic emission spectroscopy (ICP-AES).

3. Results and discussion

Fig. 1 shows the back-scattered electron micrographs of the cross-sectional U-10wt.%Zr-5wt.%RE fuel slug re-fabricated using melt residue and fuel slug butts, as well as the metallic

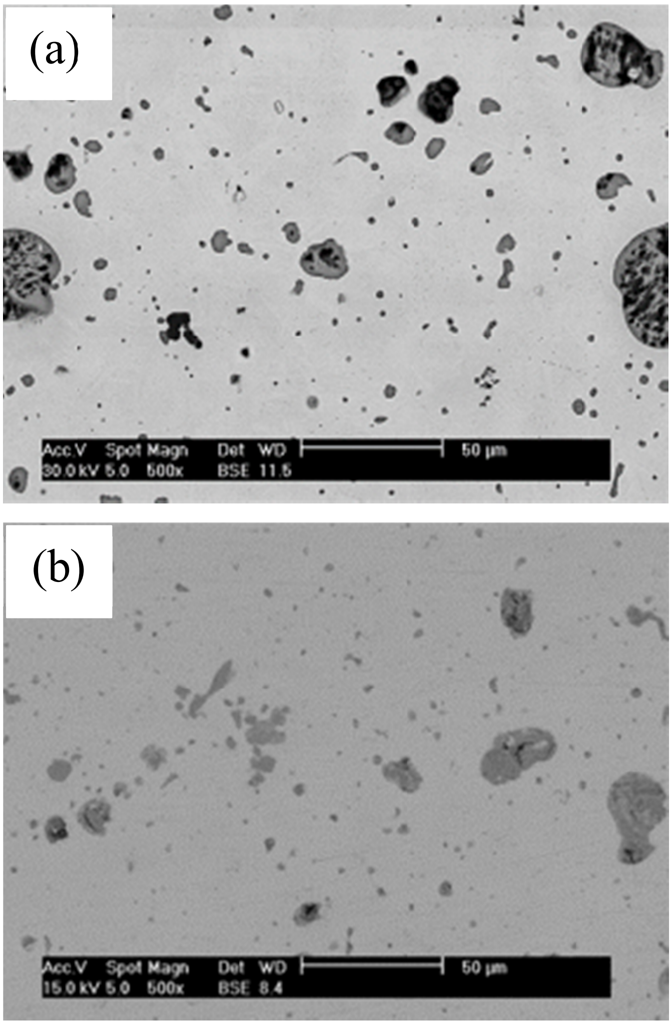

Fig. 1. Back-scattered electron micrographs of the cross-sectional of U-10wt.%Zr-5wt.%RE fuel slugs; (a) re-fabricated using fuel scraps, (b) prepared using pure metals

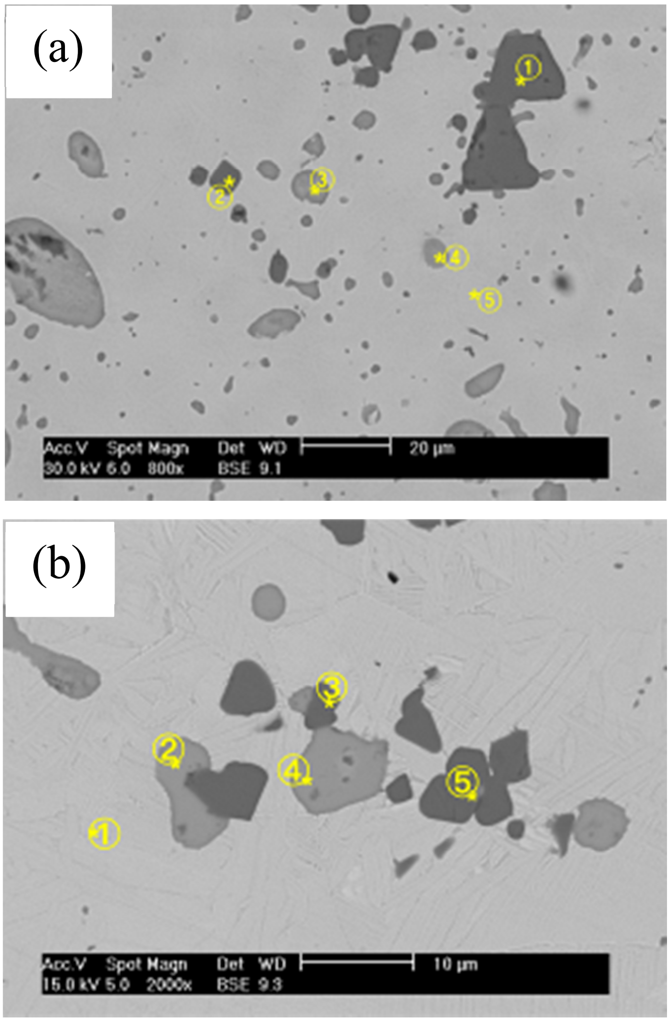

Fig. 2. A higher magnification region of the images shown in Fig. 1. The composition analysis points are marked with numbers; (a) re-fabricated using fuel scraps, (b) prepared using pure metals

fuel slug prepared using pure metals. A considerable amount of precipitates composed of RE-rich and Zr-rich phases less than 50 μm in size were uniformly dispersed in the U-10wt.%Zr alloy matrix, irrespective of raw materials. Fig. 2 shows the high magnification images of Fig. 1 with the EDS results listed in Table 1. The shape of the bright-gray RE precipitate was spherical, whereas the dark-grey Zr precipitate had an angular shape. The dark-grey phases in Fig. 2 show a higher content of Zr and a lower content of U, and the bright-grey phases in Fig. 2 show a higher content of RE, including La, Pr, Ce, and Nd, and a lower content of U and Zr, as shown in Table 1. The dark-grey Zr-rich phase with a shape similar to the one shown in Fig. 2(a) was also observed in Fig. 2(b). Small amounts of U and O elements were detected in this Zr-rich phase (Table 1). The Zr-rich phases in the U-10wt.%Zr alloy were not found in the phase diagram of the U-Zr alloy [21]. It was inferred that trace impurity elements from the charge materials were involved in the formation of the precipitates [22,23]. The bright-grey RE precipitates were located in the matrix shown in Fig. 2(a) and Fig. 2(b). The composition analysis revealed that the compositions of the RE precipitates were similar to the input contents of the RE elemental lump, as shown in Table 1. This indicates that the RE elements did not dissolve in the U-Zr alloys, and the elements did not separate among them [12]. The charged RE elements did not make precipitates with the molten U-Zr alloy due to their immiscibility [24].

TABLE 1

Composition analysis of the U-10wt.%Zr-5wt.%RE fuel slugs analyzed using EDS in Fig. 2. Values in atomic %

Melting materials	Location	U	Zr	Nd	Ce	Pr	La	O
Metallic fuel scraps (a)	#1	0.6	81.5	—	—	—	—	17.8
	#2	1.9	81.1	—	—	—	—	16.8
	#3	14.7	11.1	32.5	14.7	10.2	5.0	11.5
	#4	18.9	16.8	26.7	11.6	8.4	4.4	12.7
	#5	49.1	23.8	0.2	0.09	0.2	—	26.5
Pure metals (b)	#1	80.1	19.9	—	—	—	—	—
	#2	—	—	33.6	37.9	15.5	13.1	—
	#3	—	84.9	—	—	—	—	15.1
	#4	—	—	36.1	35.5	15.9	12.5	—
	#5	—	83.9	—	—	—	—	16.1

U-10wt.%Zr-5wt.%RE fuel slugs with a diameter of 5.5 mm were prepared with the full length of the quartz mold using the fuel scraps and pure metals per batch, as shown in Fig. 3. They were soundly fabricated with no evidence of large cracks and thin sections formed during solidification, irrespective of raw materials. Density generally provides an indirect indication of the internal defects and information concerning melting or alloying conditions of the charged materials. The density becomes lower when the cast products have pores or shrinkage defects, and when the charged materials melt well and are homogeneously alloyed, the density becomes constant at all positions of the cast. The density of the U-10wt.%Zr-5wt.%RE fuel slugs was generally uniform, irrespective of the position (Fig. 4). The densities of the U-10wt.%Zr-5wt.%RE fuel slugs decreased with the addition of RE content because the atomic weight of the RE alloy is lower than that of the matrix element, U [19]. The results showed that the density of the U-10wt.%Zr-5wt.%RE fuel slugs fabricated using melt residue and fuel slug butts was somewhat lower than that of the fuel slugs prepared using the pure metals of uranium, zirconium, and RE elements [13]. It is thought that this can be explained in relation to the alloy content of the U-10wt.%Zr-5wt.%RE fuel slugs [18]. This is discussed further with the results in the alloy compositions of the fuel slugs.

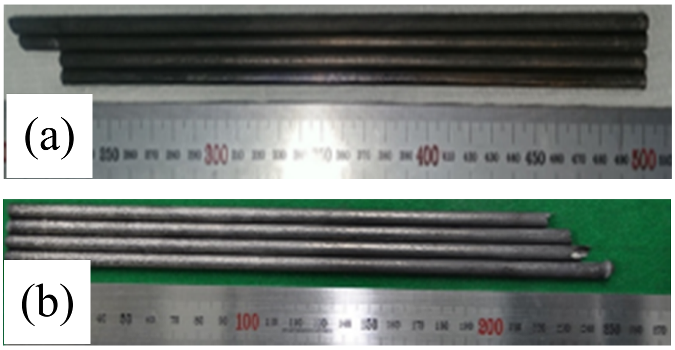

Fig. 3. U-10wt.%Zr-5wt.%RE fuel slugs fabricated by a modified injection casting method; (a) re-fabricated using fuel scraps, (b) prepared using pure metals

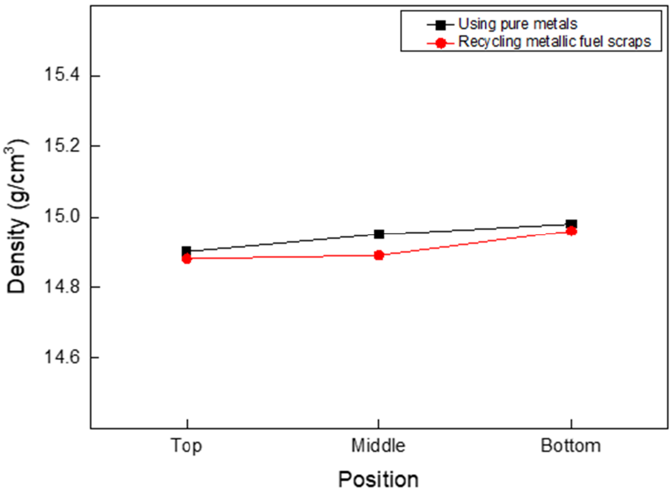

Fig. 4. Measured density of the U-10wt.%Zr-5wt.%RE fuel slugs relative to raw materials

Table 2 shows the alloy compositions of the U-10wt.%Zr-5wt.%RE fuel slugs prepared using pure metals and re-fabricated using the metallic fuel scraps. The RE content of the U-10wt.%Zr-5wt.%RE fuel slugs prepared using pure metals and re-fabricated using the fuel scraps was 2.6wt.% and 4.4wt.%, respectively. The RE content of the fuel slugs was slightly different relative to the melting materials. The RE content of the U-10wt.%Zr-5wt.%RE fuel slugs fabricated using the fuel scraps

TABLE 2

Chemical composition of the U-10wt.%Zr-5wt.%RE fuel slugs prepared using the fuel scraps and pure metals

Chemical composition	Re-fabricated fuel slugs	Metallic fuel slugs (pure metals)
U (wt.%)	86.4	85.6
Zr (wt.%)	11.0	10.4
RE (wt.%)	2.6	4.4
C (ppm)	197	40
N (ppm)	13	17
O (ppm)	237	387
Si (ppm)	467	300
C + N + O + Si (ppm)	917	744

was somewhat higher than that of the fuel slugs prepared using the pure metals. This is consistent with the slight difference between the density of the fuel slugs re-fabricated with the fuel scraps and that of the fuel slugs prepared with the pure metals. The RE elements in the re-fabricated fuel slugs were distributed more with spherical RE precipitates as an immiscible state in the U-Zr alloy matrix compared with the fuel slugs prepared using pure metals. It is thought that the fuel scraps restrained the upward floating and separation of the RE elements from the U-Zr alloy melt during remelting due to the alloyed state of the fuel scraps [12,24]. In fact, the U-10wt.%Zr-5wt.%RE remelt residue after the re-fabrication of the fuel slugs showed RE-rich layers with a small thickness of about 50 μm in the top surface of the remelting state. In contrast, the U-10wt.%Zr-5wt.%RE melt residue after the fabrication of the fuel slugs showed an RE-rich layer with a large thickness of about 1.7 mm in the melting state [25]. Hence, the RE content of the recycled fuel slugs became slightly higher than the fuel slugs prepared using pure metals [13]. From the alloy composition of the re-used fuel slugs, the majority of the impurity content of the C, N, O and Si elements was satisfied according to the specification criteria (C + N + O + Si < 2000 ppm) [25]. The XRD patterns for the re-fabricated fuel slugs compared with the fuel slugs prepared with pure metals are shown in Fig. 5. The major phases of the fuel slugs were the α-U phase and the δ-UZr_2 phase, irrespective of the melting materials [13]. The XRD patterns that originated from the contamination of the impurity elements were not observed further because most of the impure surface layers were removed. Thus, we believe that the re-fabricated U-10wt.%Zr-5wt.%RE fuel slugs were soundly fabricated during a modified injection casting process.

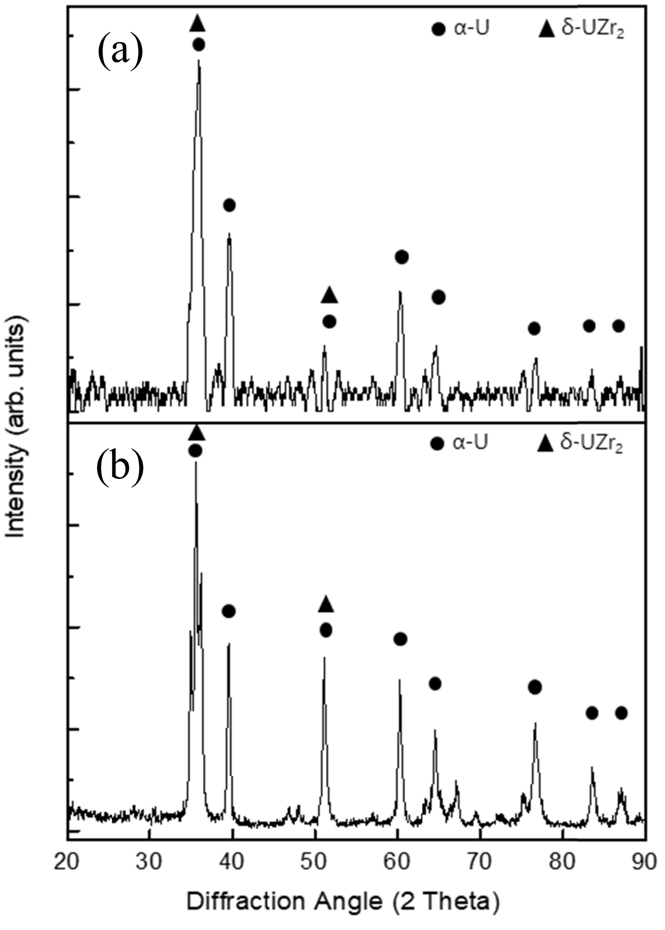

Fig. 5. X-ray diffraction patterns of U-10wt.%Zr-5wt.%RE fuel slugs fabricated by a modified injection casting; (a) re-fabricated using fuel scraps, (b) prepared using pure metals

4. Conclusions

The additive recycling process of the metallic fuel scraps is important to improve the fabrication yield rate of the metallic fuel and the utilization of the uranium resources. U-10wt.%Zr-5wt.%RE fuel slugs were soundly re-fabricated to the full length of the quartz mold using recycled fuel scraps by a modified injection casting. The re-fabricated fuel slugs were almost the same as the metallic fuel slugs prepared with pure metals from the viewpoint of the microstructure, alloy phases, density, and alloy composition of the fuel slugs. This study demonstrated the feasibility of the additive process of the fuel scraps by re-fabricating the fuel slugs.

Acknowledgments

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