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Phase characteristics of rare earth elements in metallic fuel for a sodium-cooled fast reactor by injection casting

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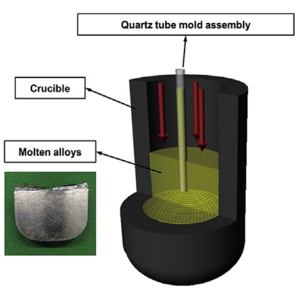
GRAPHICAL ABSTRACT



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ABSTRACT

Uranium-zirconium-rare earth (U-Zr-RE) fuel slugs for a sodium-cooled fast reactor were manufactured using a modified injection casting method, and investigated with respect to their uniformity, distribution, composition, and phase behavior according to RE content. Nd, Ce, Pr, and La were chosen as four representative lanthanide elements because they are considered to be major RE components of fuel ingots after pyroprocessing. Immiscible layers were found on the top layers of the melt-residue commensurate with higher fuel slug RE content. Scanning electron microscopy-energy-dispersive X-ray spectroscopy (SEM-EDS) data showed that RE elements in the melt-residue were distributed uniformly throughout the fuel slugs. RE element agglomeration did not contaminate the fuel slugs but strongly affected the RE content of the slugs.

1. Introduction

Uranium-zirconium (U-Zr) alloys have been considered as a nuclear fuel for sodium-cooled fast reactors (SFRs) since the 1980s because they enhance reactor safety, reduce radioactivity levels, ensure fuel cycle economy and provide a closed fuel cycle for managing minor actinides [1–3]. Metallic fuels have advantages such as a simple fabrication procedure, high thermal conductivity and excellent compatibility with sodium coolant [4–6]. The SFR needs to be suitable for remote operation and capable of mass production [7]. However, the gravity casting is not suitable to fabricate RE containing fuel slugs with a high aspect ratio and mass

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production due to the casting velocity. The gravity casting fabrication technology of high RE containing fuel slugs is still under way in several research institutes [8–11]. On the other hand, the injection casting is one of the processes that meet these needs, and it has strong advantages in terms of fabricating small-diameter castings with a high L/D ratio and a randomly oriented grain structure [7,12,13]. In a recently published paper, modified injection casting has been applied for production of metallic fuel slugs as an alternative fabrication method [7]; modified injection casting differs from conventional injection according to the atmospheric composition during operation. The modified casting method prevents the evaporation of volatile elements via Ar atmosphere during heating [7]; however, the uranium-zirconium-rare earth (U-Zr-RE) alloys are strongly reactive with the crucible, mold materials, and cladding materials, which may negatively affect the composition of fuel slugs [14–16]. Moreover, the chemical properties of RE elements are very similar to those of minor actinides in the transuranium (TRU) after pyroprocessing [7,17], and lower contents of RE is highly cost effective to the pyroprocessing progress. The RE contents in TRU is not decided in SFR program in KOREA, but the RE in the metallic fuel affects the chemical property, physical property, and FCCI with cladding [7]. Moreover, the RE elements do not react with U-Zr alloys, and an increased RE content will not necessarily distribute in the alloy. Hence, the immiscibility of RE elements in the fuel slug is a significant factor in the development of metallic fuel slugs for SFR. A previous study discussed the RE content of fuel slugs fabricated by gravity casting [10]. It was found that when the RE charging content increased, the chemical RE content in the fuel slugs was unchanged [10]. The reason for this RE deficiency has not yet been discovered. The development of pyroprocessed fuels with a high RE content requires investigation of the reason for this RE deficiency.

This study discusses the microstructure of metallic fuel slugs with various RE contents, and the melt-residue using modified injection casting. There were two main objectives in this work. The first objective was demonstration of RE deficiencies in fuel slugs. The melt-residue was investigated to verify the distribution of RE components. Information on RE deficiency may be useful in improving the purity of fuel slugs fabricated by injection casting. The second objective was to understand the effect of RE elements in fuel slugs. The RE content of a fuel slug is strongly related to the distribution of components in the slugs. The distribution of components in the fuel slug and melt-residue is also discussed, because this composition has a significant effect on the irradiation of metallic fuels due to differences in uniformity and reactivity.

2. Experimental

U-Zr alloys containing RE elements were fabricated by modified injection casting. Uranium and Zr elemental lumps, as well as RE alloy lumps, were cast in a graphite crucible to simulate the RE element of a transuranium (TRU) bearing metallic fuel. The RE alloy lumps were fabricated using four different lanthanide elements, with an overall composition of 53, 25, 16 and 6 wt% of Nd, Ce, Pr, and La, respectively. The composition of the RE in this manuscript was set according to the expecting condition of RE in TRU ingots; after pyroprocessing of the Light-Water Reactor (LWR) spent fuel [18]. The weight ratios of the RE elements in the fuel slugs were controlled to verify the effect of RE elements in fuel slugs by varying the amount of RE alloy lumps to the melt. Fuel slugs were fabricated with a Zr content of 10 wt% at a purity of 99.5 wt% (U-10Zr). The graphite crucible was coated with yttrium oxide using a high-temperature ceramic plasma spray to prevent carbon contamination from the crucible. A quartz tube mold assembly was also coated with yttrium oxide using a slurry coating method. The U-Zr-RE alloys were overheated to a temperature of 1,600 °C and cooled to 1,450 °C before injection casting into the mold assembly. While the melt was ununiform and not fully molten at the casting temperature, 1450 °C, the overheating improved uniformity of melts and soundness of fuel slugs. The quartz tube mold assembly was preheated to 600 °C before casting. Heating during casting was performed using an induction heater at a frequency of 3 kHz and a maximum power of 30 kW. The quartz tube mold assembly was immersed in the molten alloys at 1,450 °C, and Ar gas was infused to inject the U-Zr-RE alloys into the quartz tube mold assembly (Fig. 1). The initial atmosphere of the chamber began at a vacuum of less than 1.5×10^{-2} Torr. In order to prevent the evaporation of the molten elements, the atmosphere was maintained in an Ar atmosphere of 400 Torr during the heating. The injection casting was carried out under a much higher injection pressure of 200 kgf/cm^2 to overcome the Ar atmosphere during the injection casting. The diameter of the cast U-10Zr-RE fuel slugs was 5 mm, with a length of 250 mm. The cast fuel slugs were cut into suitable thicknesses using a slow-speed SiC abrasive wheel, to measure their density, chemical composition and microstructure. The density of the

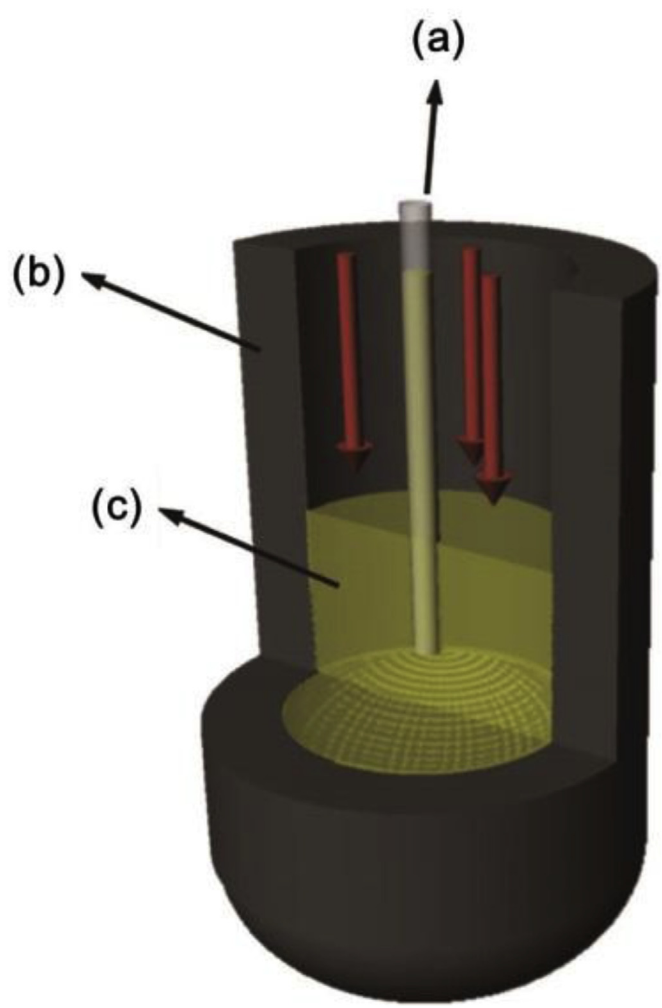

Fig. 1. Schematic of the injection casting. Red arrows in the crucible show the injection pressurization for injecting molten U-Zr-RE alloys into the quartz tube mold assembly using inert gas. (a) shows the quartz tube mold for fuel slug fabrication, (b) is the graphite crucible, and (c) is the molten U-Zr-RE alloy. Molten residues are produced from the remaining molten alloy after injection casting. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)