Fabrication Characteristics of Injection-cast Metallic Fuels

Jeong-Yong Park, Ki-Hwan Kim, Hoon Song, Jong-Hwan Kim, Seoung-Woo Kuk, Young-Mo Ko, Yoon-Myeng Woo, Seok-Jin Oh, Jung-Won Lee, Chan-Bock Lee

Next Generation Fuel Development Divison, Korea Atomic Energy Research Institute, 989-111, Daedeok-daero, Yuseong-gu, Daejeon 34057, Republic of Korea

E-mail contact of main author: parkjy@kaeri.re.kr

Abstract The fabrication process of metallic fuels for sodium-cooled fast reactors (SFR) was developed using the injection casting. U-Zr-RE(Nd-Ce-Pr-La) fuel slugs were fabricated and characterized to optimize the injection casting process. The microstructure examined by SEM showed that inclusions were uniformly distributed over the fuel slug. The reaction between the melt and the crucible was found to be significant in the fabrication of RE-containing fuel slugs compared to U-Zr fuel slugs. The pressurized injection casting method was also developed to fabricate the fuel slugs containing volatile elements. U-Zr-Mn fuel slugs were fabricated as a surrogate for Am-bearing metallic fuels under three different melting pressure conditions. From the chemical composition analysis by the ICP-AES method, no evaporation of Mn was detected in the fuel slugs fabricated under Ar atmosphere higher than 400 torr.

Key Words: Metallic fuel, Fuel fabrication, Injection casting, Microstructure

1. Introduction

Metallic fuel is one of candidate fuels for a sodium-cooled fast reactor (SFR) that is being considered as a transmutation system for long-lived minor actinides in spent nuclear fuels. When compared with alternative SFR fuels such as oxide and carbide fuels, metallic fuel has the advantages of high fissile and fertile materials density, high thermal conductivity, high compatibility with sodium, small Doppler reactivity feedback [1-3]. However, the melting point is relatively low and the propensity for fuel cladding chemical interaction (FCCI) is high. Most importantly, metallic fuel is the most suitable for using U-TRU ingots produced by the pyro-metallurgical electro-refining process of spent nuclear fuels [4].

The metallic fuel assembly to be installed in SFR, in general, consists of fuel rods, upper and lower reflectors, handling socket, nose piece and duct. The detailed design of metallic fuel assembly can be found in [5] for metallic fuel for the Prototype Generation IV Sodium-cooled Fast Reactor (PGSFR) and in [6] for Mark series fuels for the Experimental Breeder Reactor-II (EBR-II) and the Fast Flus Test Facility (FFTF). Fuel rods encapsulate fuel slugs, sodium, and helium gas inside the cladding tubes with caps welded at the both ends and wire wrapped on the outside. Sodium bonded between the fuel slug and the cladding tube enhances the thermal conduction. Cladding tubes are made of ferritic-martensitic steels such as HT9 and FC92 due to their high resistance to swelling.

The fabrication of metallic fuels has been extensively carried out to produce driver fuels and test fuels installed in EBR-II and FFTF [6]. The fabrication efforts at that time were mainly focused on U-5Fs, U-Zr and U-Pu-Zr alloys. Unlike historical metallic fuels, however, the transmutation fuel or transuranic-bearing fuel of interest (hereinafter TRU fuel) gives rise to new technical issues to the fuel fabrication [7]. Americium (Am) which is one of minor actinides to be transmuted in the fast reactor is expected to be problematic during the fuel slug fabrication due to its high volatility. Rear earth (RE) elements, which can be carried over with transuranic elements in the pyro-processing of spent fuels, are known to deteriorate the fuel integrity mainly by FCCI. RE elements also pose the challenge to the fuel fabrication primarily due to their high reactivity with the crucible and the immiscibility with U alloy [8].

Metallic fuel for PGSFR which is scheduled to be operated from 2028 is being developed, targeting the installation of U-Zr diver fuels for the initial core and the installation of lead test rods of TRU fuel in 2030. In this work, the current status of metallic fuel development was described with a focus on the fabrication and characterization of fuel slugs.

2. Fabrication of Fuel Slugs

The fabrication technology of fuel slugs has been developed using an injection casting which is considered to be of relatively simple fabrication sequence, cost efficient, suitable for remote operation and capable of mass production. Fuel slugs fabricated by an injection casting have a non-textured microstructure that is desirable for reducing the anisotropy in growth behaviour. The schematic of injection casing furnace was shown in Fig. 1. We have been improving the fuel slug fabrication technology to make multiple fuels simultaneously by increasing the casting furnace capacity as well as the amount of feedstock per batch. The current injection casting furnace can produce 13 fuels slugs, 5.54 mm in diameter and 300 mm in length, using 2.5 kg feedstock per batch. Fig. 1 shows U-10wt.%Zr fuel slugs fabricated simultaneously by the injection casting. γ ray radiographs of the fabricated fuel slugs were also shown in Fig. 1. The fabricated fuel slugs were found to have identical quality based on the characterization results. Recently, high-capacity casting furnace shown in Fig. 1 was manufactured to develop the remote fabrication process for TRU fuel that needs to be fabricated in hot cell owing to its high radioactivity.

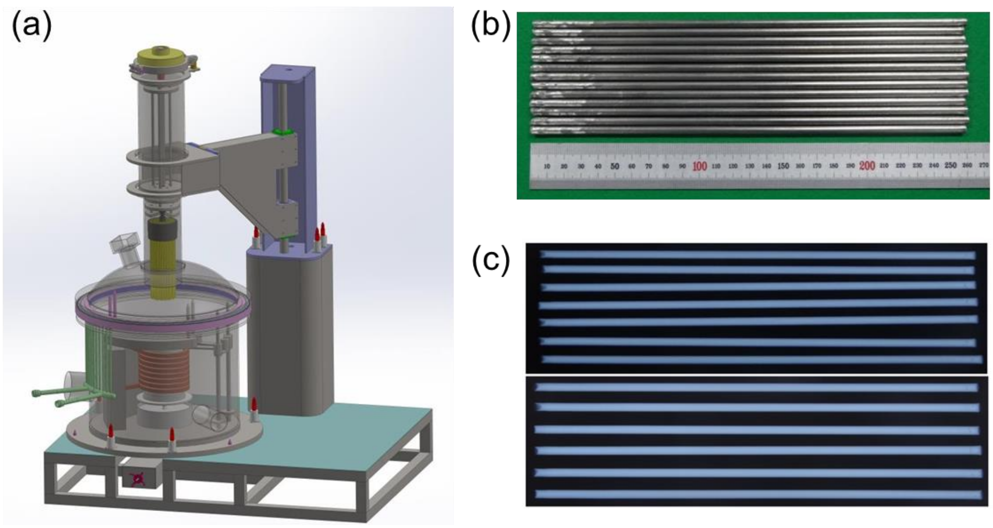

FIG. 1. (a) Schematics of injection casting furnace, (b) U-10wt.%Zr fuel slugs fabricated by the injection casting and (b) γ ray radiographs for the fabricated fuel slug.

The optimal injection casting conditions changed depending on the fuel slug composition. In the beginning, the fabrication process for U-Zr fuel slug was developed, and then U-Zr-RE and U-Zr-Mn fuel slugs were fabricated to develop the fabrication process for TRU fuel. The feedstock used in the injection casting include depleted U, low-enriched U, Zr, RE alloy consisting of 53wt.% Nd, 25wt.% Ce, 16wt.% Pr and 6wt. % La. The feedstock materials prepared according to the fuel composition were put in the graphite crucible coated with Y_2O_3 by the plasma spray method. Temperature, which was measured by the thermocouple at the bottom outside of the crucible, was increased up to the target value under vacuum or Ar gas atmosphere to melt the feedstock by high frequency induction heating using the Cu coil surrounding the graphite crucible. Melting temperature was controlled differently depending on the fuel slug compositions. The temperature was held for a certain time at the target value to homogenize the melt. Molds, which are quartz tubes bundled to the pallet, were lowered into the melt after pre-heating right above the melt. The chamber was pressurized by Ar gas to drive the melt into molds so that the melt was solidified to become fuel slugs. The pressurization process was controlled sophisticatedly depending on alloy compositions and dimensions of fuel slugs.

3. Characterization of Fuel Slugs

Characteristics of fuel slugs were significantly affected by the fuel composition as well as the injection casting condition. Specific heat, coefficient of thermal expansion and thermal diffusivity were measured to clarify the physical properties of fuel slugs. The composition of fuel slug was analysed by an inductive coupled plasma atomic emission spectrometry (ICP-AES). The density determination was performed by means of Archimedes' principle. Phases of fuel slugs were identified by X-ray diffraction (XRD) method. Microstructure of fuel slug was observed by a scanning electron microscopy (SEM) equipped with an energy dispersive X-ray spectroscopy (EDS). In this work, the characterization was focused on the microstructure observation for the different types of fuel slugs fabricated by injection casting.

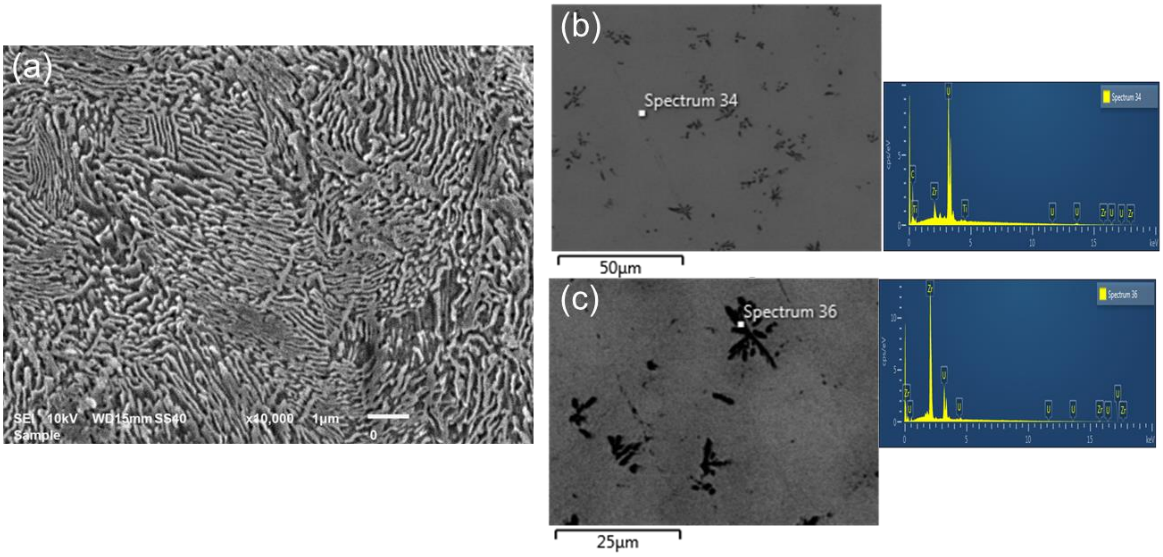

FIG. 2. (a) Scanning electron micrograph showing a lamellar structure of U-10wt.%Zr fabricated by the injection casting and EDS results on (b) the matrix and (c) inclusions.

3.1. U-10Zr Slugs

Fig. 2 shows the microstructure of the radial cross-section of U-10wt.%Zr fuel slugs fabricated by the injection casting. The morphology of as-cast U-10wt.%Zr alloy showed a lamellar structure which was expected to consist of α-U and δ-UZr_2 according to the equilibrium U-Zr binary phase diagram. Kim et al. reported that the lamellar structure of U-10wt.%Zr fabricated by the gravity casting was identified as α-U and δ-UZr_2 based on the XRD analysis [9]. In many previous works [10,11], however, δ-UZr_2 phase was not able to be identified by XRD method in U-Zr alloys consisting of U-rich and Zr-rich phases. The formation of phases in U-Zr alloys depends on the alloy composition and cooling rate, which could leads to the formation of the metastable phases and γ phase retention other than stable phases.

Another typical feature together with the lamellar structure in U-10wt.%Zr alloys is the formation of inclusions which were distributed throughout the microstructure as can be seen in Fig. 2 (b) and (c). EDS results shows that the inclusions are Zr-rich phases with the compositions being varied from inclusion to inclusion. According to U-Zr phase diagram, these Zr-rich inclusions are not the equilibrium phases. However, the formation of Zr-rich inclusion was known to be typical in the microstructure of as-cast U-Zr alloys presumably due to the fact that impurities such as C, N, O or Si having strong tendencies to react with Zr stabilized Zr-rich inclusions. Irukuvarghula et al. also reported the formation of Zr-rich inclusions decorated along the grain boundaries in as-cast U-10Zr alloys [10]. The formation of Zr-rich inclusions was also known to be effective to purify the bulk materials by gathering impurities into the inclusions

3.2. U-Zr-RE Slugs

Fig. 3 shows the scanning electron micrographs of U-Zr-RE alloys fabricated by the injection casting. Microstructures of U-Zr-RE alloys show that a lot of inclusions were dispersed throughout the microstructure. Based on the EDS analysis, the characteristics of the inclusions could be categorized into two groups which were Zr-rich inclusions and RE-rich inclusions as can be seen in Fig. 4. The types of inclusions were found to be similar even though the fabrication method was changed from the injection casting to the gravity casting. The EDS results obtained from U-10wt.%Zr-5wt.5RE fuel slug fabricated by the gravity casting [8] also showed that Zr-rich inclusions was formed together with RE-rich inclusions in the microstructure. However, the formation of inclusion in U-Zr-RE alloys is anticipated to be even more complicated compared to U-Zr binary alloys considering the immiscibility RE elements in the U melt. More precise identification on the inclusion is required to clarify the microstructure and morphology of RE-containing fuel slugs. The overall fraction of inclusions was increased with an increase of RE content as shown in Fig. 3. However, the fraction of inclusions was not greatly changed in 5wt.% and 7wt.% RE-containing alloys even though the amount of RE charged in the feedstock was increased.

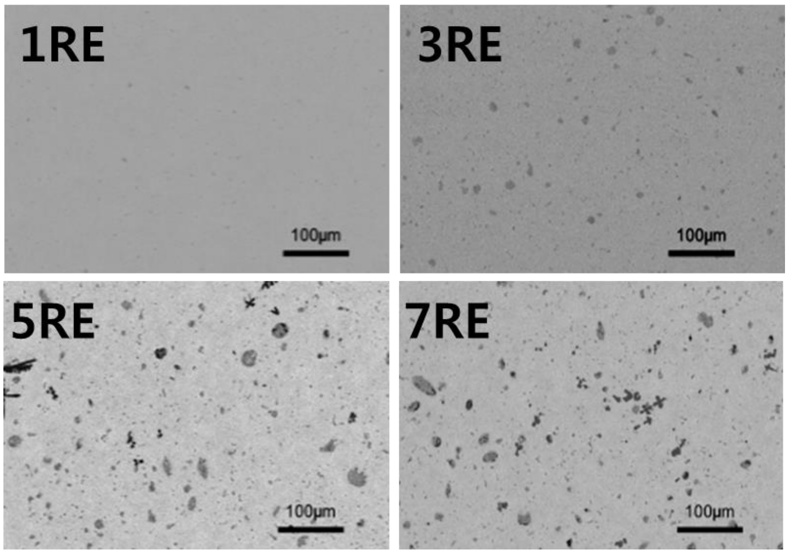

FIG. 3. Scanning electron micrographs of U-10wt.Zr-(1,3,5,7)wt.%RE alloys fabricated by the injection casting

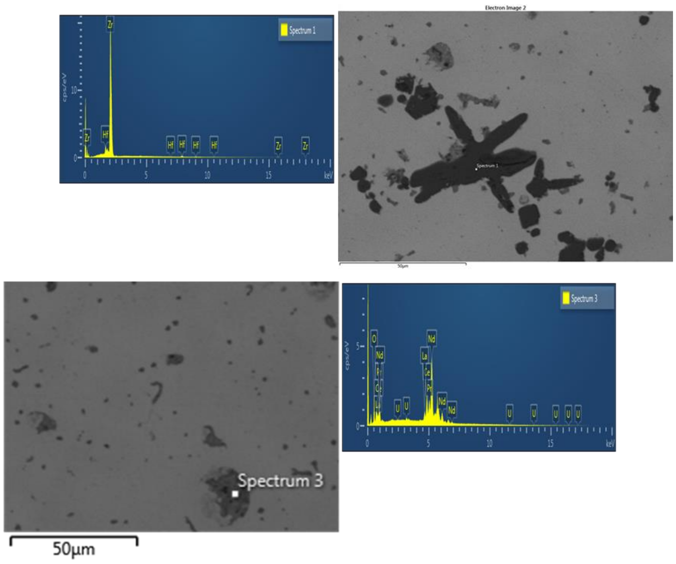

FIG. 4. EDS results on inclusions formed in U-10wt.%Zr-5wt.%RE alloys fabricated by the injection casting

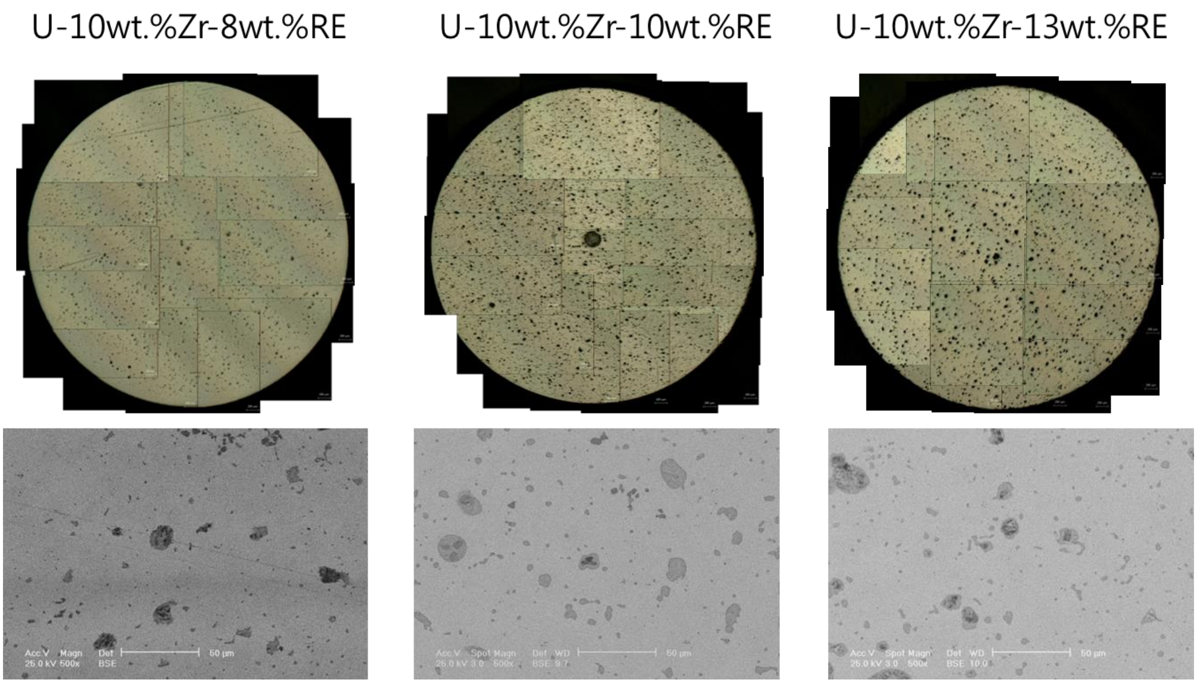

FIG. 5. Optical and scanning electron micrographs of U-10wt.Zr-(8,10,13)wt.%RE alloys fabricated by the injection casting

The fabrication characteristics of fuel slugs with high content RE elements were investigated to clarify the RE behaviour during the injection casting. Fig. 5 shows the optical and scanning electron micrographs of the radial cross-section of U-10wt.%Zr-(8,10,13)wt.%RE alloys. The inclusions were distributed throughout the fuel slug. Such microstructural characteristics were not significantly changed depending on the amount of RE charged. It was found from Figs. 3 and 5 that the fraction of inclusions was shown to be similar in the alloys containing more than 5wt.% RE.

TABLE I: CHEMICAL COMPOSITIONS MEASURED BY ICP-AES FOR U-10wt.%ZR-(8,10,13)wt.%RE FBARICATED BY THE INJECTION CASTING.

Alloys	Position	U	Zr	RE				
				Nd	Ce	Pr	La	Total
U-10Zr-8RE	Upper	86.3	10.5	1.61	1.17	0.54	0.14	3.46
	Middle	87.4	10.3	1.33	1.05	0.45	0.1	2.93
	Lower	86.7	10.6	1.4	1.09	0.49	0.12	3.10
U-10Zr-10RE	Upper	86.2	10.1	1.77	1.25	0.55	0.12	3.69
	Middle	86.0	10.2	1.73	1.24	0.55	0.11	3.63
	Lower	84.8	10.8	2.22	1.41	0.7	0.17	4.50
U-10Zr-13RE	Upper	86.1	10.2	1.95	1.46	0.62	0.13	4.16
	Middle	87.5	10.0	1.86	1.41	0.6	0.13	4.00
	Lower	86.0	10.1	1.92	1.19	0.63	0.14	3.88

Table 1 shows chemical compositions of U-10wt.%Zr-(8,10,13)wt.%RE fuel slugs analysed by ICP-AES. Samples for ICP-AES were taken out from the middle of as-cast fuel slugs. The analysis result revealed that the analysed RE content of fuel slugs was much less than the content changed. The composition analysis is consistent with the microstructure observation where the morphology of RE-containing fuel slugs showed a similar morphology when RE content was above 5 wt.%. This implies that it is hard to fabricate fuel slugs containing more than 5 wt.% RE by the injection casting.

In order to examine the behaviour of RE during melting, U-10wt.%Zr-10wt.%RE was melt in the graphite crucible coated with Y_2O_3 and then solidified without performing the injection casting. The scanning electron micrograph on the cross-section of melt residue showed that the RE-enriched slag was formed on the surface of melt residue. It was anticipated that a large fraction of RE charged rose up to make the RE-rich slag on the surface. This in turn leads to the lack of RE content in the middle of melt where the molds were immersed for the injection casting. This could be suggested to be the main reason why the fuel slugs have RE content much less than the content charged in this work. The behaviour of RE during melting and casting is now under more precise examination.

From the fabrication of U-Zr-RE fuel slugs, it was also found that the reaction between melt and crucibles occurred significantly when compared with that observed in the injection casing of U-Zr alloys. The reaction of melt with crucibles and molds is considered as one of the biggest concerns regarding TRU fuel because the reaction can be a source of contamination, fuel loss and radio-active waste. It is suggested that reusable crucibles and molds need to be developed to tackle these technical issues before the installation of TRU fuel in SFR.

3.3. U-Zr-Mn Slugs

U-Zr-Mn fuel slugs were fabricated by the injection casing to develop the fabrication process of fuel slugs containing Am. Minor actinides like Am need to be retained in metallic fuel during the fabrication process before being transmuted in the fast reactor. Mn was selected as surrogate of Am because the volatility of two elements was expected to be similar based on the saturated vapor pressure [12]. We investigated the effect of the chamber system pressure on the volatility of the element by performing the injection casting of U-10wt%Zr-5wt.%Mn with varying Ar cover gas pressure from 400 to 600 torr [13]. The fabricated fuel slugs were subjected to the composition analysis by ICP-AES. The slugs fabricated by the injection casting under 400 and 600 torr retained Mn whereas Mn content was reduced to less than 2 wt.% in the slugs fabricated under vacuum [13]. It implies that the volatility issue could be resolved by increasing the cover gas pressure during melting.

The fabrication of U-10wt%Zr-5wt.%Mn fuel slugs was intended to see whether or not Mn, as surrogate of Am, can be maintained in the fuel slugs under the varying chamber atmosphere. The microstructure of Mn-containing fuel slugs was investigated even though it was not directly relevant to analysing the microstructure of TRU fuels [13]. The scanning electron micrographs of U-10wt.%Zr-5wt.%Mn was shown to be quite different from the microstructure of U-Zr and U-Zr-RE. In the microstructure of Mn-containing fuel slug, the dendritic morphology was prominent, which is different from the typical microstructure of U-Zr and U-Zr-RE. Fabrication of metallic fuel with high volatile Am should be demonstrated by developing the advanced casting technology utilizing higher pressure system, shorter heating time and removing conditions that would promote Am vapor deposition [7].

4. Conclusions

The fabrication process of metallic fuels for sodium-cooled fast reactors (SFR) was developed using the injection casting. U-Zr-RE(Nd-Ce-Pr-La) fuel slugs were fabricated and characterized to optimize the injection casting process. The microstructure examined by SEM showed that inclusions were uniformly distributed over the fuel slug. The reaction between the melt and the crucible was found to be significant in the fabrication of RE-containing fuel slugs compared to U-Zr fuel slugs. The pressurized injection casting method was also developed to fabricate the fuel slugs containing volatile elements. U-Zr-Mn fuel slugs were fabricated as a surrogate for Am-bearing metallic fuels under three different melting pressure conditions. From the chemical composition analysis by the ICP-AES method, no evaporation of Mn was detected in the fuel slugs fabricated under Ar atmosphere higher than 400 torr.

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