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Characteristics of a nuclides distribution during a melt decontamination of radioactive aluminum wastes

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Abstract−The characteristics of an aluminum melting and the distribution of surrogate nuclide have been investigated in a muffle furnace and a lab-scale arc furnace as part of the fundamental research for a melting of the metallic wastes generated by dismantling research reactors. The melting of aluminum was carried out with the use of surrogate nuclides such as cobalt, cesium, and strontium, and fluxes such as NaCl-KCl-Na_3AlF_6 (flux A), NaCl-NaF-KF (flux B), CaF_2 (flux C), and LiF-KCl-$BaCl_2$ (flux D). The effects of the melting temperature and the type of flux on the melting of the aluminum and the surrogate nuclide distribution in the ingot, slag, and dust phase were investigated in a muffle furnace. The addition of the flux increased the fluidity of the aluminum melt, which has a slight difference according to the type of fluxes, and the amounts of the slag generated during the melting with the flux types B and C were larger than those with flux types A and D. The results of the XRD analysis showed that the surrogate nuclides move into the slag, which can be easily separated from the melt, and then they combine with the aluminum oxide to form a more stable compound. The remaining cobalt in the ingot phase was less than 20% and the others were transferred into the slag and dust. A removal efficiency of more than 99.5% for cesium and strontium from the ingot phase could be achieved. Similar results for the slag formation and the distribution of the surrogate nuclides were obtained in a DC graphite arc melting system. Therefore, it is expected that a greater part of the aluminum wastes generated from the retired research reactors can be recycled or their volume reduced to be disposed of by melting.

Key words: Melt Decontamination, Radioactive Aluminum, Flux, Distribution

INTRODUCTION

In Korea, the decontamination and decommissioning of the retired TRIGA MARK II and III research reactors and a uranium conversion plant at the Korea Atomic Energy Research Institute (KAERI) has been under way. Hundreds of tons of metallic wastes are expected from the D&D of these facilities [1]. A commercial nuclear power plant will produce 40,000 to 50,000 tons of metal waste when dismantled. Aluminum has not been used extensively in a commercial nuclear facility. However, it has enjoyed wide used in research reactors, gaseous diffusion plants, and in fuel storage pools. The gaseous diffusion plants, or GDPs, have a significant amount of a number of metals, including aluminum, nickel, copper, and steel. Aluminum was used extensively throughout the GDPs because of its excellent strength-to-weight ratios and good resistance to corrosion. Contaminated metal waste represents a considerable storage volume as well as a significant cost since it must be maintained and monitored indefinitely in secure storage. A recycling or a volume reduction of the metallic wastes through the application of appropriate treatment technologies has the merits of an increase in a resource recycling as well as a decrease in the amount of wastes to be disposed of, resulting in a reduction of disposal cost and an enhancement of the disposal safety [2,3].

Melting has been known as one of the most effective technologies for a volume reduction and recycling of metallic radioactive wastes [4,5]. Decontamination can be accomplished during the course of the melting of metallic wastes contaminated with radioactive materials through a volatilization of the radionuclides (e.g., cesium and strontium), transportation of the radionuclides (e.g., uranium) as an oxide to the slag, and a homogeneous distribution of the radionuclides (e.g., cobalt) in the ingot resulting in a reduction of the specific radioactivity [6,7]. The radionuclides are distributed into the ingot, the slag, and the dust phases in accordance with their thermodynamic characteristics and the melting condition including the slag composition [8-10]. In a previous study, melt-refining of aluminum for removal of radioactive contaminants has been reported. Kelvin melted aluminum scraps from various AEC feed stocks. The scrap was melted at 1,400 °C in a silicon carbide crucible by using a gas-fired furnace. The ingot was solidified without dross removal and no flux was added [11]. Mautz summarizes the production-scale melting of aluminum contaminated with uranium hexafluoride. Melts were performed in an oil-fired furnace [12]. Cavendish reported laboratory-scale melting of contaminated aluminum removed from a gaseous diffusion plant, Melts were performed with an induction furnace [13]. Heshmatpour and Copeland reported melt-refining of uranium contaminate aluminum. Studies were conducted at laboratory-scale using resistance and induction heating. Melts were contained in quirt tubes and zirconia crucibles. Slag metal interactions were determined for 19 melt-refining compounds. Additives were calcium and sodium fluorides, low-melting fluoride fluxes, gas refiners, and elements which might form high-temperature intermetallic compounds (carbon, boron, sulfur, antimony, silicon) [14].

This study is concerned with the melting of aluminum and the distribution behavior of the surrogate nuclides into the slag, ingot and dust phase. As part of the fundamental research for a melting

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Table 1. The surrogate nuclides

Surrogate nuclide	Chemical form	Amount used (ppm)
Co	$CoCl_2 \cdot 6H_2O$	500
Cs	CsCl	500
Sr	$SrCl_2 \cdot 6H_2O$	500

Table 2. Flux composition

Flux type	Composition
Flux A	NaCl (45%), KCl (40%), Na_3AlF_6 (15%)
Flux B	NaCl (45%), KCl (40%), KF (15%)
Flux C	CaF_2 (100%)
Flux D	LiF (14%), KCl (76%), $BaCl_2$ (10%)

of the aluminum wastes generated by dismantling research reactors, the melting of aluminum was carried out with the use of surrogate nuclides such as cobalt, cesium, and strontium in a muffle furnace and in a lab-scale D. C graphite arc melting system. The characteristics of the melting itself and the surrogate nuclide distribution were investigated with a combination of fluxes and a variation of the flux concentration.

EXPERIMENTAL

Aluminum specimens (10×10×1 mm sheet type) were prepared as the feed material for the melting tests. The surrogate nuclides were chosen by considering their chemical properties, the reaction among the elements and the ease of handling and chemical detection [15]. The chemical forms of the surrogate nuclide selected in the tests are given in Table 1.

In the melting of radioactive aluminum scrap, fluxes have been widely used for high thermal conductivity, high thermal capacity, low density, moderate viscosity, high electrical conductivity and mutual miscibility [10,16]. Alkali metal halides are the major components used as the flux for an aluminum melting. Four combinations of fluxes were used in this study as given in Table 2. The flux was added to a crucible filled with an aluminum specimen prior to the beginning of the melting.

A muffle furnace (Nabertherm Co.) was used in the basic melting experiments to obtain basic data on the melting characteristics of the aluminum and the surrogate nuclide distribution into the ingot and slag phase. The aluminum specimen contaminated with a mixed solution of the surrogate nuclides as shown in Table 1 and dried in vacuum oven at 60 °C was charged into the alumina crucible. The melting tests were carried out by introducing the alumina crucible filled with 50 g of aluminum to the furnace in which the desired melting temperature was being maintained.

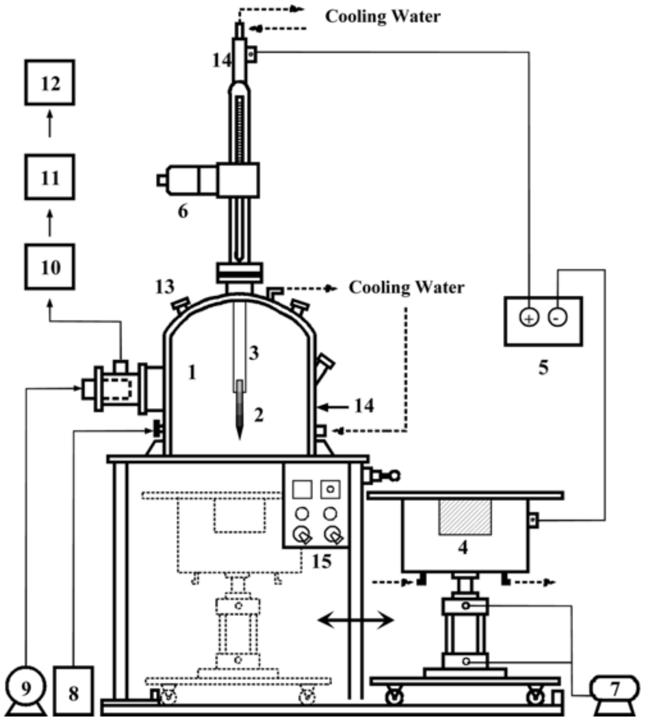

Fig. 1. The schematic of the electric arc furnace.

1. Reactor chamber
2. Graphite electrode
3. Copper electrode
4. Copper crucible
5. Power supply
6. Stepper motor
7. Air compressor
8. Argon gas
9. Vacuum pump
10. Filter bank
11. Emission monitor
12. Radiation monitor
13. Viewport
14. Cooling water jacket
15. Control box

The lab-scale DC graphite arc melting system was used to verify the melting performance with the operational conditions obtained through basic experiments on the melting of aluminum in a muffle furnace. The melting system consisted of one graphite electrode and a copper crucible with a graphite refractory which served as a counter electrode, as shown in Fig. 1. The cooling water flows in the surroundings of the graphite arc electrode and the copper crucible. The aluminum specimen was charged and melted in the graphite crucible which was placed in the copper crucible. The melting capacity of the graphite crucible was 300 cc. Input power can be supplied by controlling the DC voltage from the DC power supplier. The range of the electric power used in the melting test was 7-20 KW. In the DC graphite arc melting, the surrogate elements of 500 ppm were mixed with the flux and added to the metal in the crucible. The mixture was melted under an argon atmosphere for 5-10 minutes.

After the completion of melting, the molten metal was cooled and solidified in the crucible. The ingot samples were taken as chips by drilling of the ingot to a depth of about 10 mm in two different locations. The slag was easily separated from the ingot. The ingot and slag samples were analyzed with ICP-AES (IRIS DUO, Thermo Elemental Co.) for cobalt and ICP-MS (X-7, Thermo Elemental Co.) for cesium and strontium to evaluate the surrogate nuclide distribution. An XRD analysis was carried out for the slag samples to examine the in slag compounds.

RESULTS AND DISCUSSION

One of the key parameters in the melting of aluminum is the composition of the fluxes. It is reported that the flux affects the properties of the melt such as the melting temperature, viscosity, electrical conductivity and so on [17]. The term "fluxing" is used to represent all additives to, and treatments of, molten aluminum in which chemical compounds are used. These compounds are usually inorganic and may perform several functions, such as degrassing, demagging, cleaning, and alloying. Fluxing also includes the treatment by inert or reactive gases to remove inclusions or gaseous impurities. Fluxing is temperature dependent. It must be high enough to provide for good contact and reactivity and achieve good physical separation. The choice of specific compounds or chemical reagents in fluxes depends on the specific purpose of the flux [18].

During the course of an aluminum melting, the flux builds up in the slag and it is combined with the components of the fluxes and aluminum oxides. The slag forms more stable compounds by including the impurities contained in the aluminum or the surrogate nuclides.

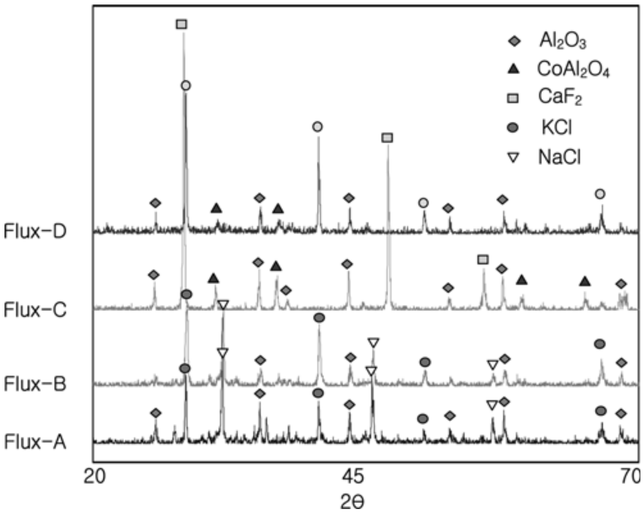

Fig. 2. The results of the XRD analysis for the slag.

The XRD analysis results of the slag generated from the melting of aluminum with the fluxes and surrogate nuclides are shown in Fig. 2. The results showed the formation components between the aluminum oxide and surrogate nuclides. Therefore, the results of the XRD analysis mean that the surrogate nuclides move into the slag phase, which can be easily separated from the melt, and then they combine with the aluminum oxide to form a more stable compound [19].

The effect of a flux addition on the melting of the aluminum was observed with the type of fluxes as given in Table 2. The addition of the flux increased the fluidity of the aluminum melt, which has a slight difference according to the type of fluxes. It was observed that the fluidity of the aluminum melt with fluxes A and D is better than that with fluxes B and C.

Fig. 3 shows the effect of the flux concentration on the formation of the slag. The amounts of slag generated during the melting with fluxes B and C were larger than those with fluxes A and D. In the separation of the slag from the ingot, the use of fluxes B and C was easier than that of types A and D. Through the XRD analysis for the slag sample, the components of the fluxes, the surrogate nuclides and the aluminum compound were detected. It shows that the surrogate nuclides move into the slag and then they combine with the aluminum oxide to form a more stable compound.

The amount of slag formation increased with an increasing flux concentration. It became slow at a flux concentration above 5 wt%. Considering the amount of flux addition, it means that the compounds included in the aluminum oxide, which is able to capture the surrogate nuclides, are produced effectively at 5 wt% of a flux concentration.

By increasing the amount of flux addition, the distribution ratio of cobalt into the ingot and the slag phase is shown in Fig. 4. At 5 wt% of a flux concentration, the distribution ratio of cobalt was maximized in the slag phase up to 80% and minimized in the ingot phase by 10% according to the type of fluxes. From the viewpoint

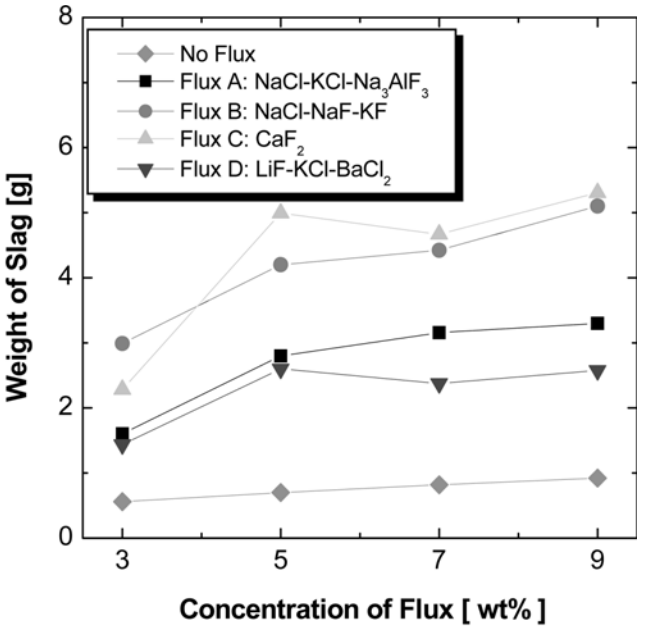

Fig. 3. Effects of the concentration of the flux on the slag formation in the arc furnace (800 °C, 20 min).

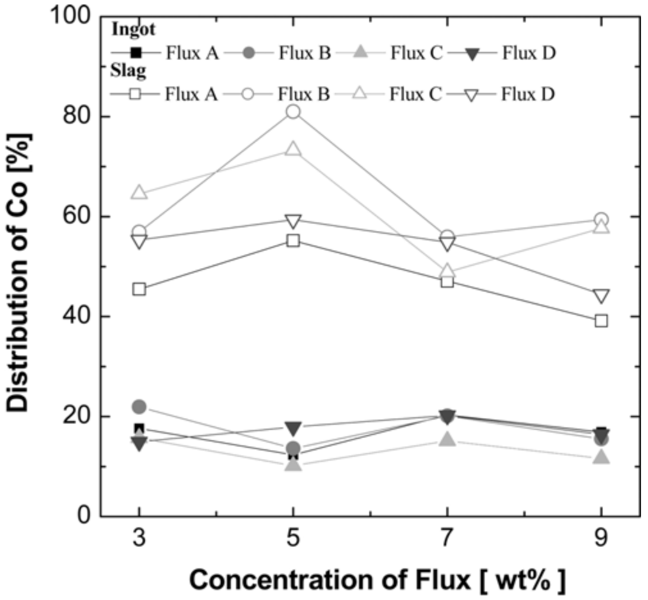

Fig. 4. Effects of the concentration of the flux on the distribution of cobalt in the arc furnace (800 °C, 20 min).

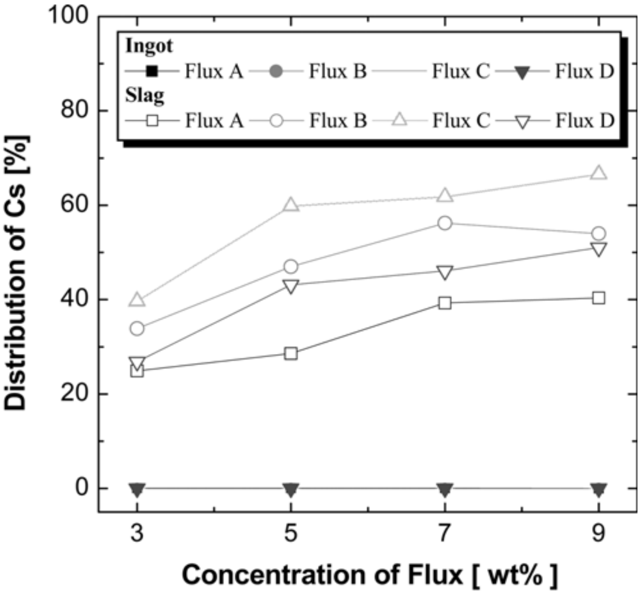

Fig. 5. Effects of the concentration of the flux on the distribution of cesium in the muffle furnace (800 °C, 20 min).

of the efficiency for removing the cobalt from the ingot, the use of fluxes B or C is likely to be better than that of A or D.

Fig. 5 shows the distribution ratio of cesium into the ingot and the slag phase according to the flux concentration. Cesium barely existed in the ingot. In the slag phase, however, the residual cesium increased with an increase of the flux concentration, but the increasing rate was mitigated at a flux concentration of more than 5 wt%. Similar results were obtained for the distribution of strontium. Therefore, the addition of 5 wt% of a flux is likely to be adequate enough with a consideration of the surrogate nuclide distribution.

The reaction temperature has a considerable effect on the transfer phenomena and the chemical reaction. The amount of slag formation increased with an increase of the melting temperature regardless of the flux type. This increasing tendency is likely to be due to an increase in the oxidation rate of the aluminum. With an increase in the melting temperature, the concentration of cobalt in the ingot phase decreased while that in the slag phase increased due to a reduction in the viscosity and an increase in the fluidity resulting in an enhancement of the mass transfer and the mobility of the surrogate nuclides by a diffusion and a convection [20]. On the other hand, the concentration of cesium in the ingot phase was very low, while that in the slag phase was quite high because of its volatile property. With increasing temperature to 900 °C, the concentration of cesium in the slag increased slightly due to an increase in the amount of slag formation. Above 900 °C, however, the residual cesium in the slag decreased again, which is attributed to the cesium being exhausted as a form of dust or vapor due to a higher temperature. Similar results were obtained for the distribution of strontium.

On the basis of the results obtained through the basic melting experiments in a muffle furnace, DC graphite arc melting tests were carried out to verify its applicability to the melting of an aluminum waste. In a comparison with the results obtained from the previous basic melting experiments in a muffle furnace, similar tendencies for the slag formation and the distribution of the surrogate nuclides were shown.

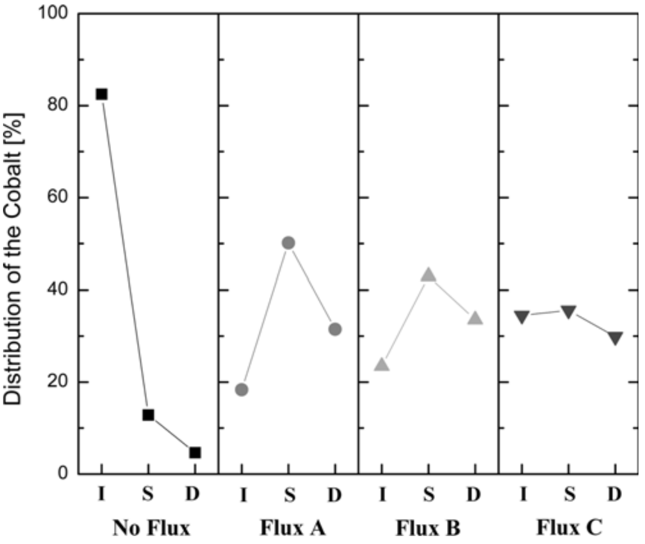

Fig. 6. Effects of the fluxes on the distribution of the cobalt in the arc furnace (I: Ingot, S: Slag, D: Dust).

The distribution of the surrogate nuclides in each phase such as the ingot, slag, and dust phase was dependent on the characteristics of the flux added during the melting experiment. Fig. 6 shows the effects of the kind of flux on the distribution of the cobalt during a melting of the aluminum in the arc furnace at 5 wt% of a flux concentration. During aluminum melting without a flux, 82% of the cobalt remained in the ingot phase and the others were transferred into the slag and dust phase. But in a melting with a flux, most of the cobalt was eliminated from the aluminum melt and distributed to the slag phase and to the dust phase. In respect of the decontamination of materials contaminated with surrogate nuclides, the fluxA type was more effective than the other fluxes in this study.

Fig. 7 shows the effects of the kinds of fluxes on the distribution

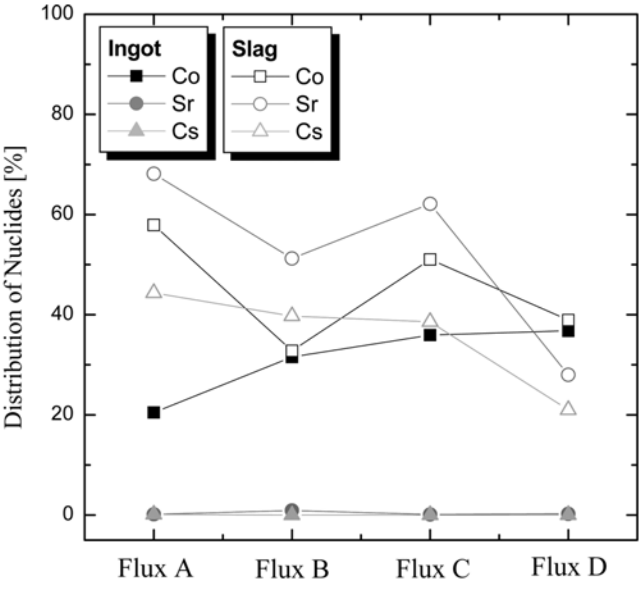

Fig. 7. Effects of the fluxes on the distribution of the nuclides in the arc furnace (9 wt%).

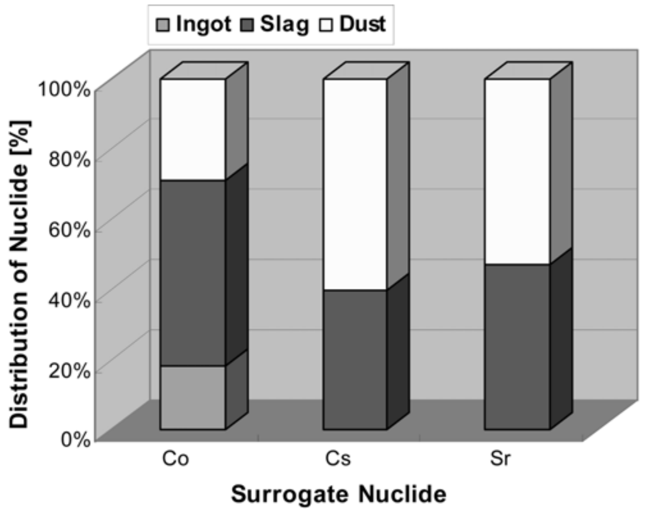

Fig. 8. The distribution of the nuclides in the arc furnace (5 wt% of flux A).

of the surrogate nuclides during the melting of the aluminum waste. Results showed that the cobalt distribution was considerably scattered according to the variations of the fluxes. Flux A and flux C types were shown to be more predominant than the other fluxes for the decontamination of the cobalt in the melting of the aluminum. The results of the cesium distribution into the ingot, slag and dust phases showed that most of them were eliminated from the melt of the aluminum and captured in the slag and dust phases.

Fig. 8 shows a typical result for the surrogate nuclide distribution obtained by the DC graphite arc melting system in which 5 wt% of flux A was applied. The distribution of the cobalt in the ingot phase was less than 20%. A removal efficiency of more than 99.5% for the cesium and strontium from the ingot phase could be achieved for the volatile nuclides such as cesium and strontium due to their transportation from the ingot to the slag and dust phases.

Summarizing the results from the melting experiments in a muffle furnace and a DC graphite arc melting system, it is expected that a greater part of the aluminum wastes generated by dismantling the retired research reactors can be effectively recycled and/or their volume reduced to be disposed of by a melting.

CONCLUSIONS

The melting of aluminum was carried out with the use of surrogate nuclides such as cobalt, cesium, and strontium in a muffle furnace and in a lab-scale DC graphite arc melting system. The addition of the flux increased the fluidity of the aluminum melt and there was a considerable difference in the amounts of slag generated during a melting according to the type of fluxes. The amount of slag formation increased with an increase of the melting temperature due to an increase in the oxidation rate of the aluminum regardless of the flux type. The addition of a 5 wt% flux is likely to be adequate enough for a melting of aluminum because the distribution of the cobalt into the slag was maximized at 5 wt% of a flux and the increasing distribution tendency of the cesium and strontium into the slag phase was mitigated at above 5 wt% of a flux. The remaining cobalt in the ingot phase was less than 20% according to the type of fluxes, and a removal efficiency of more than 99.5% for the cesium and strontium from the ingot could be achieved.

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