Effects of Operation Variables on the Distribution of Radioactive Nuclides in the Arc Furnace

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1. Introduction

The metallic dismantlement wastes will be considerably generated from the decommissioning of TRIGA MARK II and III research reactors and uranium conversion plant at Korea Atomic Energy Research Institute(KAERI). To treat these radioactive metal waste, the metal melting technology has been known as the one of the most effective technology in the volume reduction and the recycling of the metallic radioactive wastes up to now[1]. During the metal melting technologies, the electric arc furnace was preferred for the cost of maintenance, accommodation of large wastes, scale-up and the decontamination by using slag materials.

Experiment was conducted in the electric arc furnace with equipped D.C. graphite arc system, off-gas ventilation system, and vacuum system. To increase the fluidity of melt and prevent aluminum melt from oxidation, various fluxes were used up to 9wt% with the concentration of the each flux. In this study, we were investigated the effects of the various fluxes, concentration of contaminants and melting time on the distribution of radioactive nuclides and the characteristics of aluminum melting.

2. Experiment

2.1 The Electric Arc Furnace

The lab-scale d. c. graphite arc melting furnace was used to verify the melting performance of aluminum wastes with various operational conditions such as kinds of fluxes, weight of added flux and concentration of surrogate nuclides. The arc melting system consists of one graphite electrode and copper crucible with graphite refractory which serves as counter electrode. The cooling water flows in the surroundings of graphite arc electrode and copper crucible. The input power can be supplied by controlling the d. c. voltage from the d. c. power supplier. The surrogate elements of 1,000 ppm and the isotope of 500ppm were mixed with the flux and added to the aluminum wastes charged in the crucible. The mixture was melted under argon atmosphere for 3 minutes.

After the completion of melting, a molten aluminum was cooled and solidified in the copper mold. The ingot samples were taken as the chips by drilling of the ingot to the depth of about 10mm 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 nuclide distribution. The XRD analysis was carried out for slag samples to examine the slag compounds.



Figure 1. Electric arc melting apparatus.

2.2 Surrogate nuclides and Flux Composition

The surrogate elements was chosen by their chemical properties, the reaction among the elements and the ease of handling and chemical detection[2]. In order to evaluate the suitability of the elements to be used as a surrogate, an index expressed by the ratio of the Gibb's free energy can be used. The compounds of surrogate elements selected in the tests are given Table 1. The fluxes have been studied for melting of radioactive aluminum wastes, because of high thermal conductivity, high thermal capacity, low density, moderate viscosity, high electrical conductivity, mutual miscibility. Alkali metal halides are generally the major components used as the flux for aluminum melting. Four fluxes were used in this study as given in Table 1. The flux of 3-9wt% was added to the crucible filled with aluminum prior to the beginning of the melting.

Table 1. Surrogate nuclides and flux composition

Surrogate elements		CoCl ₂ ·6H ₂ O, CsCl, SrCl ₂ ·6H ₂ O				
	Flux A	NaCl(45),KCl(40), Na ₃ AlF ₆ (15)				
Flux	Flux B	NaCl(45),KCl(40), KF(15)				
Compostion	Flux C	CaF ₂ (100)				
	Flux D	LiF(14),KCl(76), BaCl ₂ (10)				

3. Results

One of the key parameters in the melting of aluminum is the composition of fluxes. It is reported that the flux affects the properties of the melt such as melting temperature, viscosity, electrical conductivity and so on. With increasing the amount of flux addition, the distribution ratio of cobalt into the ingot and the slag is shown in Figure 2. For aluminum melting with no flux, the distribution ratio in the slag and ingot phase was respectively 82.5%, and 12.8%. With adding the flux concentration, the distribution ratio of cobalt was maximized in the slag up to 60% and minimized in the ingot by 20% according to the type of fluxes. In the view point of the efficiency for removing cobalt from the ingot, the use of flux A or B is likely to be better than that of flux C or D.



Figure 2. Effect of the concentration of flux on the distribution of cobalt in the ingot and the slag phase.

Figure 3 shows that 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 increasing the flux concentration..



Figure 3. Effect of the concentration of flux on the distribution of cesium in the ingot and the slag phase.

The similar results of cesium were obtained for the distribution of strontium as shown Figure 4. The reason why the distribution ratios of strontium in the slag phase was much higher than those of the cesium was the vapor pressure of strontium compound was higher than that of cesium compound.



Figure 4. Effect of the concentration of flux on the distribution of strontium in the ingot and the slag phase.

Table 2 shows the typical result on the nuclide distribution obtained by the d. c. graphite arc melting in which 7wt% of the flux A and radioisotope nuclides such as cobalt and cesium was applied.

	Flux	Spe. Act.(Bq/g)		$C_{0}(%)$	Cc0/			
	(7wt%)	Co-60	Cs-137	CO(70)	C5/0			
Ingot	Flux A	83.53	0.00	38.38	0.00			
	Flux B	150.94	0.00	68.43	0.00			
	Flux C	96.65	0.00	43.24	0.00			
	Flux D	61.88	0.00	29.22	0.00			
	No Flux	81.89	0.00	51.17	0.00			
Slag	Flux A	1648.82	95.64	37.36	2.43			
	Flux B	480.17	135.86	11.92	3.78			
	Flux C	701.15	11.96	38.98	0.74			
	Flux D	2977.84	71.12	38.37	1.03			
	No Flux	4023.59	231.92	31.56	2.04			

Table 2. Distribution ratio of radioisotope

4. Conclusion

The distribution ratio of cobalt, cesium and strontium in the slag phase increase with increasing the weight of the flux. It is expected that the greater part of aluminum wastes generated by dismantling the retired research reactors can be effectively recycled and/or reduced their volume to be disposed by melting.

REFERENCES

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