Effects of the Slag Former on the Metal Melting and Radionuclides Distribution in an Electric Arc Furnace

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Abstract

The characteristics of the metal melting and radionuclide distribution of the radioactive has been investigated in a lab-scale arc furnace. The slag former based on the constituents of silica, calcium oxide, aluminum oxide, borate and calcium fluoride additions was used for melting of the stainless and carbon steel. In the melting of the stainless steel, the amount of slag formation increased with an increase of the concentration of the slag former. But the effects of the slag basicity on the amount of slag formation showed a local maximum value of the slag formation with an increase of the basicity index in the melting of the stainless steel as well as in the melting of the carbon steel. With an increase of the amount of slag former addition, the trends of the cobalt distribution into the ingot and the slag depended on the kind of slag former used in the melting of the stainless steel while the effect of the slag basicity on the distribution of the cobalt was not clarified in the melting of carbon steel. In the melting of the carbon steel, the strontium was captured at up to 50% into the slag phase. Cesium was completely eliminated from the melt of the stainless steel as well as the carbon steel and distributed to the dust phase.

Key word : metal melting, nuclide distribution, arc furnace, basicity index

1. Introduction

As a result of the aging of nuclear research facilities as well as nuclear power plants, these facilities must be shutdown and dismantled in the immediate future. The decontamination and decommissioning (D&D) of nuclear installations results in considerably large amounts of radioactive metal wastes such as stainless steel, carbon steel, aluminum, copper etc. The reference 1000MWe PWR(pressurized water reactor) plant and 881MWe PHWR(pressurized heavy water reactor) plant will generate metal wastes of 24,800ton and 26,500ton respectively [1]. As a consequence of the D&D performance, hundreds of tons of metallic wastes such as metal, concrete, and soil will be expected from the D&D of these facilities [2]. These radioactive metal wastes will induce the problems of handling and storing these materials from the environmental and economic aspects. For this reason, prompt countermeasures should be taken to deal with the metal wastes generated by dismantling the retired nuclear facilities. As most of these steels are slightly contaminated, if they are properly treated they are able to be recycled and reused in the nuclear field [3,4]. In general, the technology of a metal melting is regarded as the most effective

method to treat radioactive metal wastes from nuclear facilities. In the technology of a melting treatment, the most important variable is the slag former because the distribution of radionuclide and the characteristics of a metal melting depend on the composition and basicity of the slag former [5].

This study is concerned with the melting of metal wastes(i.e. stainless steel and carbon steel)and the distribution behavior of the nuclides into the slag, ingot and dust phase. The melting of stainless and carbon steel was carried out with the use of surrogate nuclides such as cobalt, cesium, and strontium in a lab-scale d. c. graphite arc melting system. The characteristics of the melting itself and the nuclide distribution were investigated with a combination of slag formers and a variation of the slag former concentration.

2. Experimental

The stainless steel and carbon steel specimens(10×10×1mm sheet type) were prepared as the feed material for the melting tests. The surrogate nuclides were chosen by considering their chemical properties, the reactions among the elements and the ease of handling and a chemical detection. The slag former was added to a crucible filled with a metal specimen prior to the beginning of a melting. The composition and kinds of the slag former used in this study are given in Tables 1 and 2. Experiments of a metal melting were carried out in an electric arc furnace, as shown in Fig. 1. The d. c. graphite arc melting system consists of one graphite electrode (20mm in diameter by 150mm in long) and a copper crucible(100mm in diameter by 80mm in depth) with a graphite refractory which serves as a counter electrode. The cooling water flows in the surroundings of the graphite arc electrode, copper crucible and the chamber. The input power can be supplied by controlling the d. c. voltage from the d. c. power supplier. The stainless and carbon steel specimen contaminated with the mixed solution of the surrogate nuclides and dried in a vacuum oven at 60℃ was charged into the graphite crucible with a melting capacity of 300cc. The 500g of stainless and carbon steel charged into the graphite crucible was mixed with the prepared slag former and then melted under an argon atmosphere for 5~10minutes.

After the completion of a melting, the molten metal was poured into and cooled in acopper cast. After cooling, the ingot of stainless steel was obtained as a cylindrical type. The ingot samples were taken as chips by a drilling of the ingot to a depth of about 10mm at several locations. After a cooling, 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.

Table 1. The composition of the slag former for the melting of stainless steel

Slag composition, wt%					
Slag Former	SiO_2	CaO	Al_2O_3	Fe_2O_3	B_2O_3	Other
SF - S1	40	40	20			
SF - S2	40	30	20			CaF_2 - 10
SF - S3	30	40	10	20		
SF - S4	30	50	10	5		CaF_2 - 5

SF - S5	80	4	2		13	K_2O - 1
SF - S6	38.1	41.4	14.1	2.6		MgO - 3.8
SF - S7	75	25				
SF - S8	35	65				

Table 3. The composition of the slag former for the melting of carbon steel

Slag composition, wt%						
Slag Former	SiO_2	CaO	Al_2O_3	Fe_2O_3	Other	Basicity Index
SF - C1	50	30	20			0.520
SF - C2	40	40	20			0.828
SF - C3	30	50	20			1.283
SF - C4	10	50	40			1.598
SF - C5	10	60	30			2.325
SF - C6	80	4	2		B_2O_3 - 13	
SF - C7	30	40	10	20		
SF - C8	40	30	20		CaF_2 - 10	
SF - C9	38.1	41.4	14.1	2.6	MgO - 3.8	
SF - C10	30	40	10		P_2O_5 - 20	

3. Results and Discussion

The slag chemistry remarkably affects the melting properties and the distribution of the radionuclides in the arc melting system. Fig. 2 shows the effect of the slag former concentration on the formation of the slag during the melting of the stainless steel. The amounts of slag generated during the melting of the stainless steel with the slag former linearly increased with an increase of the concentration of the slag former. In respect to the fluidity of the slag, the slag former containing CaF_2(SF-S2) was more effective than that of the other slag formers because the fluorides break the bonds of the silicate network present in the SiO_2 containing slag [6]. The increase rate of the slag formation with SF-S2 was lower than that of SF-S1 or SF-S3 with an increase of the concentration of the slag former. In general, the weight of the slag generated by a melting in the arc melting system was about 2 ~ 5wt% of the metal scrap charged into the graphite crucible [7]. In this experiment, the weight of the slag generated by the melting of the stainless steel was about 5 ~ 14wt% of the metal scrap. The reason why the weight of the slag was more than that of the other studies was that the weight of the metal melted was relatively low. The cobalt is an important radioactive isotope in the D&D field. Therefore, its distribution must be determined during a melting experimental study.
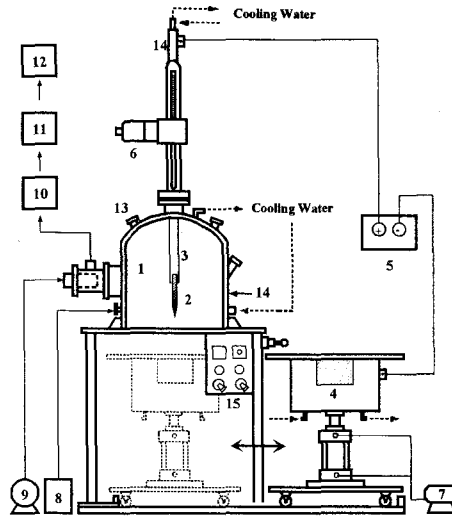
Fig. 1. The schematic of the electric arc furnace.

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

With an increase of the amount of slag former addition, the distribution ratio of the cobalt into the ingot and the slag is as shown in Fig. 3. The trend in the distribution of the cobalt in the ingot phase was different in accordance with the kind of slag former. In the SF-S1 containing Fe_2O_3, the cobalt was not transported to the slag phase with an increase of the concentration of the slag former. On the other hand, the cobalt with the slag former (SF-S3) containing CaF_2 was moved to the slag phase with a slag addition. Because of its chemical similarity to iron, the cobalt was consistently partitioned to the ingot while the cesium was captured in the slag and the fume [6].
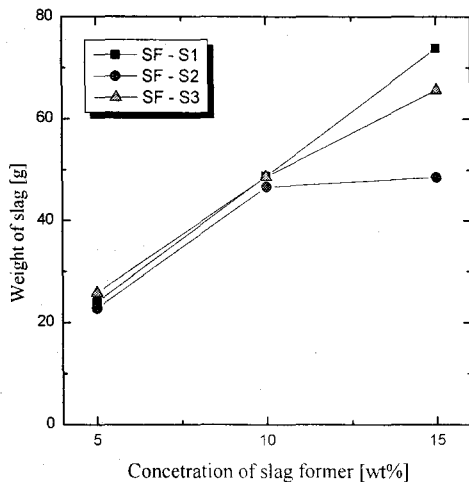

Fig. 2. Effects of the concentration of the slag former on the slag formation.
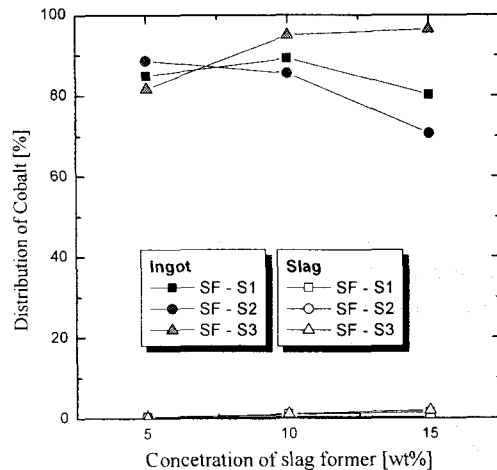

Fig. 3. Effects of the concentration of the slag former on the distribution of cobalt.

Fig. 4 shows the distribution ratio of strontium into the ingot and the slag according to the flux concentration. Strontium barely existed in the ingot in all the cases of the tests because the vapor pressure of the strontium was lower than that of the melt. In the slag phase, however, the residual strontium increased with an increase of the flux concentration but the increasing rate is very slow. Most of the strontium was eliminated from the stainless steel melt and distributed to the slag phase and to the dust phase. As shown in Fig. 5, the distribution results of the strontium showed that 20% of the strontium remained in the slag phase, 0 ~ 1% of the strontium remained in the ingot, and 80% of the strontium remained in the dust. The distribution of the strontium in each phase was similar regardless of the slag former type.

Fig. 5 shows the effects of the kinds of slag former on the distribution of the surrogate nuclides during the melting of the carbon steel. Results showed that the range of the cobalt distribution in the ingot was between 82% and 96% in a lab-scale arc furnace testing. The slag former of SF-C9 was found to be more effective than the other slag formers for the decontamination of the strontium and cobalt in the melting of the carbon steel. The results of the cesium distribution into the ingot, slag and dust phase were the same as the results of the stainless steel melting.
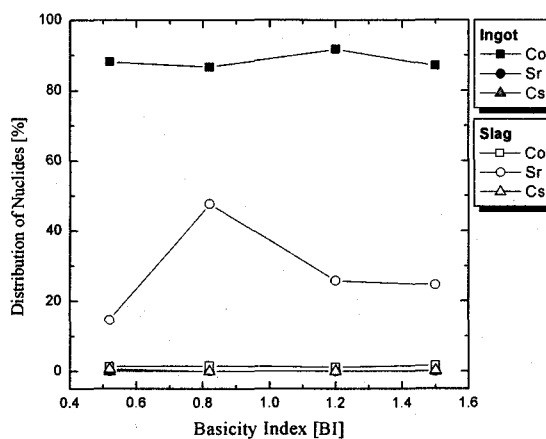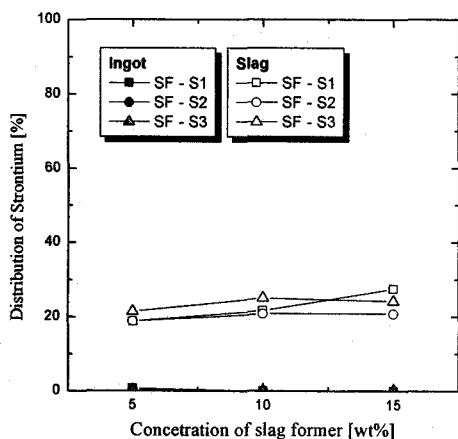

Fig. 4. Effects of the concentration of the slag former on the distribution of strontium.

Fig. 5. Effects of the kinds of slag former on the distribution of the nuclides.

4. Conclusions

The results of the melting experiments in the arc furnace have fostered the following conclusions. In the melting of the stainless steel, the amount of slag formation increased with an increase of the concentration of the slag former. But the effects of the slag basicity on the amount of slag formation showed a local maximum value with an increase of the basicity index in the melting of the stainless steel as well as in the melting of the carbon steel. The optimum conditions of the stainless steel melting in this study were 15wt % of the added slag former and SF-S2. The trends of the cobalt distribution into the ingot and slag phase depended on the kinds of slag former used in the melting of the stainless. In the melting of the stainless steel, only 20% of the strontium remained in the slag phase and it was barely present in the ingot. But, strontium was captured at up to 50% into the slag

phase in the melting of the carbon steel. Cesium was completely eliminated from the melt of the stainless steel as well as the carbon steel and distributed to the dust phase. The slag former of SF-C9 was verified to be a more effective slag former than the other slag formers for the decontamination of the surrogate nuclides in the melting of the carbon steel.

References

1. L.A. Nieves and R. W. Tilbrook, Radwaste magazine, "Potential Radioactive Scrap Metal Quantities from Nuclear Power Plants Worldwide", January, pp. 45-53 (1996).

2. W.K. Choi, P.S. Song, B.Y. Min, H.I. Kim, C. H. Jung, and W. Z. Oh, "A State of the Art on the Technology for Melting and Recycling of the Radioactive Metallic Wastes", KAERI/AR-716/2004, Korea Atomic Energy Research Institute (2004).

3. V. Adams, W. Murphie, and M. Gresalfi, "Recycle and Reuse of Radioactive Scrap Metals within the Department of Energy", Safewaste 2000, 1, 115-124 (2000).

4. K. L. Hopkinson, A. Bishop, M. T. Cross, J. Harrison, and Selgas, Nuclear safety and the environment, European Commission, "Decontamination of Metals by Melt Refining/Slagging", EUR 18041 EN (1998).

5. S. A. Worcester, L. G. Twidwell, D. J. Paolini, T. A. Meldon, and R. E.Mizia, WINCO-1198, Westinghouse Idaho Nuclear Company, Idaho Falls, Idaho (1994).

6. M. B. Joanna, K. D. Brian,and M. Eric Schlinenger, SAND 96-0902, Category UC-704 (1998).

7. M. Sappok, G. Lukacs, and A. Ettemeyer, "Melting of Radioactive Metal Scrap from Nuclear Installations", in Proc. Decommissioning of Nuclear Installations, K. Pflugarad, R. Bisci, B. Huber, and E. Skupinski Eds., pp. 482-493 Brussels, Belgium (1989).