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**Abstract**: Many nuclear power plants (NPP) facilities have aged and are being dismantled around the world. As a result, large amounts of radioactive metal waste are generated during decommissioning. Carbon steel, stainless steel, and Inconel are the most common metals used in NPP, and radioactive contaminants are mostly accumulated in the corrosion layer. There are various radionuclides, but the main ones are <sup>60</sup>Co, <sup>137</sup>Cs, <sup>54</sup>Mn and <sup>51</sup>Cr. <sup>60</sup>Co is the major activated corrosion product and responsible for high gamma radiation and longer half-life (5.27 years.) also makes it the most difficult to remove. Therefore, we investigated the melt decontamination characteristics of <sup>60</sup>Co by introducing various slags using induction melting for stainless steel and carbon steel. Cobalt plating and cobalt oxide film coatings were used as specimens instead for safety purposes. The amount of cobalt removed from the slag was analyzed by using XRF. About 11% of the cobalt was removed from the contaminated metal due to slag oxide formation. The distribution of cobalt in the melted metal was also almost homogeneous according to the XRD analysis.

Keywords: radioactive waste metal; decontamination; cobalt; slags; induction melting

### 1. Introduction

Nuclear power plants are being dismantled for reasons such as reaching the end of their design life, changes in nuclear policy, and accidents. This results in a large amount of radioactive metallic waste. The first commercial NPP, Kori Unit 1 in South Korea, has been shut down after 40 years of designed lifetime and is now being dismantled and a large number of radioactive metals will be generated and need to be decontaminated to reduce the volume of the radioactive waste which requires very high disposal cost. Of many alternatives, melt decontamination is used to reduce the volume of radioactive metals in many countries. Recycling is also possible when radioactive contamination is reduced enough through the melting process to satisfy regulatory limits. Therefore, developed countries with nuclear dismantling technologies such as the United States, Germany, Japan, and France, decontaminate radioactive metal waste using the melting process as the most suitable process for the circumstances of each country [1,2].

Basic research on melting decontamination has been conducted in South Korea. It is urgent to secure demonstration technology for melt decontamination, which is a necessary technique for dismantling nuclear power plants. Various studies and experiments for melt decontamination have been conducted since the 1980s (Table 1). The pollutant nuclides seen in a steam generator of a pressurized water reactor (PWR) are generally <sup>51</sup>Cr, <sup>54</sup>Mn, <sup>55</sup>Fe, <sup>59</sup>Fe, <sup>58</sup>Co, <sup>60</sup>Co, <sup>95</sup>Zr and <sup>63</sup>Ni, among others [3–5]. The decontamination of the main cobalt-based nuclides, <sup>60</sup>Co and <sup>58</sup>Co, is currently at a low level. Even after melt decontamination, <sup>60</sup>Co is hardly removed and remains in the metal base material [6]. The dismantling of TRIGA Mark was completed in 2001 in Korea, and related research on melting decontamination of cobalt nuclides is still in progress. Research and development



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**Copyright:** © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). for melt decontamination of radioactive metal waste contaminated with cobalt is urgent for the disassembly of Kori Unit 1.

Country/Institution	Year	Type of Furnace	<b>Types of Metal</b>	<b>Contaminant Level</b>
UK/Capenhurst	1994	Induction	aluminum, copper	-
Germany/CARLA	1989	Induction	CS, SUS, aluminum	$200 \text{ Bq/g} (\beta, \gamma),$ $100 \text{ Bq/g}(\alpha)$
France/CENTRACO	1999	Induction	CS, SUS	2000 Bq/g (β, γ), 370 Bq/g(α)
US/DURATEK	1992	Induction	CS, SUS, aluminum	<2 mSv/h
Russia/ECOMET-S	-	Induction	CS	100 Bq/g
France/INFANTE	1992	Arc	CS, SUS	250 Bq/g
Sweden	1987	Induction	CS, SUS, aluminum	-
US/Mauts	1975	Arc	CS	3.5 ppm
US/Large,SEG	1993	Induction	CS	<10 mR/h
US/Echols,SEG	1993	Induction	CS	<10 mR/h
US/Laesen	1985	Induction	CS, SUS	21 pCi/g, 27 pCi/g (Co)
Japan/JAEA	2006	Induction	CS, SUS	_

**Table 1.** Examples of melting decontamination studies [7].

The purpose of this study is to find the best slag composition and melting conditions for melting decontamination of radioactive metals. In the experiment, SUS304 and CS41 (most common metals from NPP decommissioning, Busan, Korea) were used as experimental objects, induction melting was used as the melting equipment and many compositions of slag were tested. In consideration of safety issues, non-radioactive specimens were used instead of radioactive metals. We made specimens (non-radioactive) using two methods, coating and electroplating. The process and results of the experiment will be described in detail below.

#### 2. Experimental Procedure and Results

In this study, melt decontamination experiments were conducted on two kinds of metals, SUS304 and S41C, which are the most frequently used metals in nuclear power plants, as four types of contamination replicated specimens with Co, Cs, Mn, and Cr [7]. In consideration of safety, all the samples used for the experiment were non-radioactive surrogate specimens. The SUS304 experiment was a preliminary experiment, and a melt decontamination experiment with S41C was carried out based on the results and experience obtained from the SUS304 experiment. After melting, the distribution characteristics of each isotope of nuclides were analyzed. All of the experiments were done using the same type of induction melting equipment and graphite crucible (purity 99%, height 10 cm, inner diameter 7 cm, outer diameter 9 cm). The component analysis of all the slags was done using XRF (X-ray fluorescence, Rigaku ZSX Primus II, Seoul, Korea).

#### 2.1. Decontamination Experiment and Result of SUS304

#### 2.1.1. Production of Contamination Replicated Specimens

Radioactive corrosion products are formed at the surface of metal due to direct radioactivity and adhesion of the radioactive substances in the cooling channels. Corrosion products on the metal surface are either the oxide or hydroxide of the nuclide. However, due to corrosion resistance of SUS304, contaminants can exist in the form of metal as well as corrosion products [8]. Removal of cobalt is known to be especially difficult, so simulating diversely contaminated specimens is necessary. Therefore, SUS304 specimens with cobalt were prepared by two methods: plating and oxide film coating. Others were prepared as chromium-plated specimens, manganese oxide film-coated specimens, and

Conditions	Co <sub>2</sub> O <sub>3</sub>	MnO <sub>2</sub>	CsCl	Со	Cr	
Number of coatings	3	3	3			
Solidification Temperature (°C)	200	200	Room Temperature	Tin and cobalt	Flectroplating	
Solidification Time (minute)	30	30	120	alloy plating solution, conductive plating	using Sargent solution	
Resin Ratio	50 wt %	50 wt %	50 wt %	e conductive plating		
Weight (g)	7	300	300	300	300	
Images						

cesium chloride-coated specimens. The specimens' preparation and fabrication conditions are shown in Table 2.

**Table 2.** SUS304 specimen fabrication methods.

### 2.1.2. Experimental Procedure and Conditions

Prepared specimens and reagents were placed in a graphite crucible as an order of the reagents under the specimens. The reason for doing this is that the density of the slag reagent is lower than that of the melted metal, so stirring can be performed well during melting. The crucible was then placed into an induction coil, after checking if the cooling water was flowing in or not, power was turned on, the output was gradually raised and maintained at 15 kW. At this time, the temperature inside the molten metal is about 1500–1800 °C. When the melting was completed, the graphite crucible was taken out and cooled, and the slag and ingot could easily be separated (Figure 1).



**Figure 1.** Image of molten metal and molten slag. (a) The ingot obtained after cooling; (b) the slag obtained after cooling.

The reagent composition, basicity (B ratio, the ratio of CaO to SiO<sub>2</sub> in slag), weight change of the slag and metal specimens, and the melting time of each experiment are shown in Table 3. The composition of basic slag is SiO<sub>2</sub>, CaO, and Al<sub>2</sub>O<sub>3</sub> [9]. In order to find the best composition for slag, Fe<sub>2</sub>O<sub>3</sub>, CaF<sub>2</sub>, or other substances were added to adjust the flow basicity of the slag [9]. The standard melting time is 1800 s. Experiments Co (No.3), Cr (No.3), and Mn (No.3) were carried out for 2 times longer than the standard

melting time. Co (No.5-2) was a remelting experiment that uses the obtained ingot through experiment Co (No.5-1). In experiment Co (No.8), we supplied a small amount of oxygen to accommodate more cobalt oxide and make it easier to transport into the slag.

N	No.		ag Con	nposition	(wt %)	Basicity	Weight before/after (g)		Type of Specimen	Melting Time	Remarks
		$SiO_2$	CaO	$Al_2O_3$	Others		Specimen	Slag			
	1	10	60	30		6.00	299.73/322.19	30/29.72	Plating	1800 s	
	2	50	30	20		0.60	299.70/319.31	30/27.55	Plating	$1800 \mathrm{~s}$	
	3	10	60	30		6.00	299.70/319.31	30/27.55	Plating	3600 s	
	4	30	40	10	$Fe_2O_3(20)$	1.33	299.70/321.93	30/25.24	Plating	$1800 \mathrm{s}$	Remelt
	5-1	20	50	20	$CaF_{2}(10)$	2.50	299.49/-	30/26.38	Plating	$1800 \mathrm{s}$	Remelt
	5-2	40	10		$CaF_{2}(50)$	0.25	-/319.83	30/25.77	Plating	$1800 \mathrm{s}$	
Ca	6	75	25			0.33	299.68/317.20	30/19.80	Plating		
Co	7	30	50	10	Fe <sub>2</sub> O <sub>3</sub> (5), CaF <sub>2</sub> (5)	0.17	299.80/314.82	30/29.25	Plating	1800 s	O <sub>2</sub> Input
	8	50	30	20	_ ` `	0.60	299.11/325.55	30/24.51	Plating	$1800 \mathrm{s}$	
	9	10	60	30		6.00	70.78/78.92	30/27.91	Coating	$1800 \mathrm{s}$	
	10	50	30	20		0.60	70.03/76.98	30/29.33	Coating	1800 s	$Slag \times 2$
	11	50	30	20		0.60	299.63/317.44	60/58.82	Plating	1800 s	Specimen $\times 2$
	12	50	30	20		0.60	599.28/633.49	60/57.71	Plating	1800 s	
	1	10	60	30		6.00	299.37/319.19	30/29.40	Plating	1800 s	
	2	50	30	20		0.60	300.11/322.65	30/23.45	Plating	$1800 \mathrm{s}$	
	3	10	60	30		6.00	299.29/320.68	30/27.87	Plating	3600 s	
	4	30	40	10	Fe <sub>2</sub> O <sub>3</sub> (20)	1.33	299.57/322.65	30/24.58	Plating	$1800 \mathrm{~s}$	
Cr	5	65	15	10	Fe <sub>2</sub> O <sub>3</sub> (5), CaF <sub>2</sub> (5)	0.23	299.35/318.71	30/28.42	Plating	1800 s	
	6	75	25		_ , ,	0.33	299.37/324.09	30/15.71	Plating	1800 s	
	7	30	50	10	Fe <sub>2</sub> O <sub>3</sub> (5), CaF <sub>2</sub> (5)	1.67	299.51/328.10	30/29.53	Plating	1800 s	
	1	10	60	30		6.00	299.46/314.36	30/28.21	Coating	1800 s	
Mn	2	50	30	20		0.60	301.90/323.33	30.24.06	Coating	$1800 \mathrm{s}$	
	3	10	60	30		6.00	299.81/313.85	30/22.36	Coating	3600 s	
	1	10	60	30		6.00	302.18/317.98	30/29.00	Coating	1800 s	
Cs	2	50	30	20		0.60	301.53/309.31	30/25.41	Coating	1800 s	

Table 3. Experimental conditions and weight change for SUS304.

Experiments Co (No.9) and Co (No.10) used cobalt oxide film-coated specimens, and the remaining experiments used cobalt-plated specimens. In experiment Co (No.11), the slag input was doubled and in experiment Co (No.12), the standard specimen weight was doubled. All details and experimental conditions are shown in Table 3. After melting, we crushed the obtained slag into a powder and analyzed its ingredients using XRF. The ingot was also cut in an arbitrary place, analyzed using a scanning electron microscope (Pusan National University, Busan, Korea) on each surface, and the distribution of nuclides was observed.

#### 2.1.3. Experimental Result of SUS304

The analysis results of slag using XRF are summarized as follows. Cobalt and chromium turned into oxide forms and moved onto the slag. The XRF results are shown in mass % (Table 4). A comparison of the weight percent of nuclides trapped in each slag is shown in Figure 2.

N		Metal Oxide wt %			
N0.	Co <sub>2</sub> O <sub>3</sub>	Cr <sub>2</sub> O <sub>3</sub>	MnO	Cs <sub>2</sub> O	
1	0.017	0.268	0.260	-	
2	0.069	0.438	1.580	0.332	
3	0.009	0.099	1.080	-	
4	0.044	0.246	0.332	-	
_ 5-1	0.005	0.279	-	-	
5 5-2	0.002	-	-	-	
6	0.042	0.332	-	-	
7	0.005	0.102	-	-	
8	0.020	-	-	-	
9	0.030	-	-	-	
10	0.051	-	-	-	

Table 4. XRF analysis results of weight percent of metal oxide.



Figure 2. XRF analysis result. (a) Result of Co; (b) result of Cr; (c) result of Mn; (d) result of Cs.

Using Equation (1), it is possible to calculate the absolute number of surrogate nuclides that migrated onto the slag after melt decontamination through the XRF analysis results in Table 4. The calculated results are summarized in Table 5.

$$\mathbf{D} = (\mathrm{wt} \ \% \times \mathbf{m})\mathbf{M} \tag{1}$$

D: Removal rate of nuclides

# wt %: Percentage of nuclides in slag M: Weight of plated or coated nuclide m: Weight of slag after melting

**Table 5.** Percentage of the surrogate nuclides captured in the slag.

	Metal wt %				
No.	Со	Cr	Mn	Cs	
1	1.00	14.04	12.80	-	
2	3.88	19.72	70.46	9.67	
3	0.49	4.89	59.11	-	
4	2.35	11.05	-	-	
_ 5-1	0.39	10.00	-	-	
5 5-2	0.18	13.62	-	-	
6	1.83	8.95	-	-	
7	0.31	5.13	-	-	
8	1.20	-	-	-	
9	5.92	-	-	-	
10	10.68	-	-	-	
11	8.61	-	-	-	
12	6.82	-	-	-	

Based on the calculation results shown in Table 5, No.2  ${SiO_2(50\%)-CaO(30\%)-Al_2O_3(20\%)}$  slag has the best removal effect on each nuclide. The experimental results show that there is a large difference in the removal rate between the plated Co specimen (3.88%) and the coated cobalt specimen (10.68%). The pollutant nuclides, which are metallic forms, are much more difficult to decontaminate by melting. Although the removal rate of plated chromium was up to about 20%, we could not confirm how accurately the plated chromium was moved on the slag, since the SUS304 contained about 18% chromium. These uncertainties were confirmed in experiments with carbon steel. The decontaminating rate of manganese was about 70%, and cesium was hardly detected in the slag due to its high volatility. Furthermore, a better decontamination effect could not be obtained by supplying remelting and a little oxygen.

#### 2.2. Decontamination Experiment and Result of S41C

Based on the experimental results of SUS304, it was found that slag No. 2 is the best. Therefore, experiments with carbon steel were conducted based on slag No. 2, and  $Fe_2O_3$  was used to improve fluidity.

#### 2.2.1. Decontamination Experiment and Result of S41C

Compared to SUS304, S41C is easier to corrode, and most radioactive contaminants are present in the form of a corrosion product. The corrosion products are composed of oxides or hydroxides on the corrosion layer [8]. Therefore, it is preferable to coat the oxide or hydroxide of the nuclide isotope of the corrosion layer after corroding S41C. In order to maximally match the thickness of actual contamination metal, the corrosion thickness was measured. For the measurement, we used 20% hydrochloric acid to corrode S41C at room temperature at 2-h intervals and analyzed the corrosion thickness. The surface condition of the corrosion layer was observed through photographs taken with an optical microscope (Figure 3). The average value of the corroded depth was obtained, and the functional relationship between the corrosion time and the thickness of the corrosion layer was found (Figure 4).



(**d**)

**Figure 3.** Observation of corrosion thickness using optical microscope. (**a**) The corrosion time is 2 h; (**b**) the corrosion time is 4 h; (**c**) the corrosion time is 6 h; (**d**) the corrosion time is 8 h; (**e**) the corrosion time is 10 h; (**f**) the corrosion time is 12 h.

(e)



Figure 4. Changes in corrosion thickness according to time. After 12 h of corrosion, the corroded thickness can reach  $800 \ \mu$ m.

The corrosion test shows that the corrosion rate of the surface of carbon steel S41C is about 80  $\mu$ m/h (Figure 4). In order to simulate the corrosion surface that the thickness is about 700–1000  $\mu$ m, corrosion was carried out for 12 h and the corroded specimens were coated with radionuclides. Table 6 summarizes the working conditions and the prepared specimen images.

(**f**)

Coating						
Conditions	Co <sub>2</sub> O <sub>3</sub>	Cr <sub>2</sub> O <sub>3</sub>	MnO <sub>2</sub>	CsCl		
Number of coatings	3	3	3	3		
Solidification Temperature (°C)	200	200	200	Room temperature		
Solidification Time (minute)	30	30	30	120		
Resin Ratio	50 wt %	50 wt %	50 wt %	50 wt %		
Weight (g)	70	70	70	70		
Corrosion Thickness	800 µm	800 µm	800 µm	800 μm		
Images						

Table 6. S41C specimen fabrication methods.

## 2.2.2. Experimental Procedure and Conditions

The experimental procedure and working method for carbon steel are the same as those of the SUS304 experiment and the experimental conditions are shown in Table 7. The melting time is 1800 s. In the Co (No.3) and Cr (No.3) experiments,  $Fe_2O_3$  was used in place of 10% of  $Al_2O_3$ .

No.		Slag Composition (wt %)			Basicity	Weight before	/after (g)	Melting Time	Type of Specimen	
		SiO <sub>2</sub>	CaO	$Al_2O_3$	Others		Specimen	Slag		
	1	50	30	20		0.60	355.81/351.51	72/84.42	1800 s	Coating
Co	2	50	30	20		0.60	355.05/346.23	36/54.20	1800 s	Coating
	3	50	30	10	Fe <sub>2</sub> O <sub>3</sub> (10)	0.60	356.49/315.99	36/50.73	1800 s	Coating
	1	50	30	20		0.60	357.92/340.32	72/90.36	1800 s	Coating
Cr	2	50	30	20		0.60	355.41/330.43	36/54.54	1800 s	Coating
	3	50	30	10	Fe <sub>2</sub> O <sub>3</sub> (10)	0.60	356.08/311.98	36/46.03	1800 s	Coating
Ma	1	50	30	20		0.60	357.58/345.79	72/80.97	1800 s	Coating
Min	2	50	30	20		0.60	356.17/329.56	36/56.61	1800 s	Coating
6	1	50	30	20		0.60	360.81/335.70	72/83.22	1800 s	Coating
ĊŚ	2	50	30	20		0.60	360.79/313.45	36/6182	1800 s	Coating

Table 7. Experimental conditions and weight change for S41C.

2.2.3. Experimental Result of S41C

The XRF analysis results of slag are summarized as follows, Figure 5. The XRF results are shown in mass %.

Using Table 8 and Equation (1), we calculated the exact weight of each replicating nuclide used on the slag, as shown in Table 9. The removal rate of cobalt, which is the least difficult to remove is about 10%, which is similar to the experimental result of SUS304. Since there was almost no chromium component of S41C, it was certain that all the chromium moved onto the slag was the coated chromium, and the maximum removal rate was better than that of the plated SUS304, which was about 90%. The removal rate of manganese and cesium is similar to the experimental result of SUS304.



Figure 5. XRF analysis result. (a) Result of Co; (b) result of Cr; (c) result of Mn; (d) result of Cs.

No.	Metal Oxide wt %						
	Co <sub>2</sub> O <sub>3</sub>	Cr <sub>2</sub> O <sub>3</sub>	MnO	Cs <sub>2</sub> O			
1	0.318	2.161	7.113	0.599			
2	0.308	5.050	7.131	0.613			
3	0.097	5.865					

Table 8. XRF analysis results of weight percent of metal oxide.

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Table 9. Percentage of the surrogate nuclides captured in the slag.

N	Metal wt %					
No	Со	Cr	Mn	Cs <sub>2</sub>		
1	10.89	56.84	79.73	10.01		
2	8.17	88.76	58.97	9.97		
3	1.69	68.70				

## 2.3. Behavior of Slag and Metal in Melting Process

Figure 6 shows the melting process of slag and metal from start to finish. The density of the molten slag is lower than that of the metal, and when melted completely, the slag

moves from the bottom to the top. A reagent to sufficiently enable the metal to oxidize and capture the transfer nuclide on the metal surface is positioned under the metal and then melted. The metal of the crucible is a substance with high electric resistance that receives much energy from induction heating and indirectly heats the surrounding slag. The slag continues to receive heat, it reaches melting point earlier than the metal and begins melting. Liquid slag surrounds the solid metal and begins collecting nuclides on the metal surface. When the temperature reaches the melting point of the metal, the slag moves to the surface of the molten metal and when in contact with the air forms a slag layer composed of solid and liquid.



**Figure 6.** The whole process of melting and slag formation. (**a**) Prepared specimens and reagents were placed in a graphite crucible as an order of the reagents under the specimens; (**b**) to preheat the machine, the power is controlled within 2 kw; (**c**) gradually increase the power; (**d**) when the power reaches 15 kw, the metal is melted and slag is also formed; (**e**) liquid metal can be seen after breaking the solidified slag cover; (**f**) picture after cooling.

It is possible to check the molten metal underneath the slag layer. Once fully cooled, if the slag and metal are solid again and there is enough slag, it will form on the metal surface like a lid. The melting process has a major impact on the performance and efficiency of the final slag. Therefore, the melting process should be optimized and improved. For example, it is necessary to increase the stirring force between the molten slag and the molten metal and to control the amount of slag generated.

#### 2.4. Analysis of Nuclide Distribution

The distribution characteristics of nuclides in the ingot after melting are very important in the treatment of radioactive waste. Complete decontamination is impossible in any decontamination process, and decontaminated metals will ultimately be stored in a radioactive waste disposal plant. If the distribution of the nuclides present in the metal via melting is not homogeneous, a very large economic benefit can be obtained by removing specific sites with high nuclide density and storing them separately. However, waste storage entails considerable cost [10].

For this purpose, the ingot was cut after melting into three equal parts and the distribution characteristics of the Co, Cr, Mn, and Cs remaining in the ingot were studied using a scanning electron microscope. The results of the analysis are shown in Table 10. The

reproductive nuclides present inside each ingot are evenly distributed without deviating from the error range. However, the distribution of chromium is 10.37 wt %, 21.08 wt %, and 10.44 wt % in the top, middle, and bottom, respectively. The content of chromium grew rapidly in the middle part, which can be regarded as the segregation of the SUS304 ingot during cooling. The distribution of chromium is still homogeneous.

 Table 10. Analysis results of distribution.

Nuclides	Location	Images	Element wt	%
	Тор			3.39
Со	Middle		The second secon	3.18
	Bottom	10 10		3.17
	Тор	-		10.37
Cr	Middle			21.08
	Bottom			10.44
Mn	Тор			2.35
	Middle			2.24
	Bottom			2.65



Table 10. Cont.

# 3. Discussion

# 3.1. Decontamination Characteristics of Co and Characteristics of Specimens

In this study, two types of cobalt specimens were made and tested to improve the reliability of decontamination of <sup>60</sup>Co and <sup>58</sup>Co. Most of the <sup>60</sup>Co and <sup>58</sup>Co are present on the metal surface in the form of corrosion products. SUS304 itself may also be exposed to radiation and <sup>60</sup>Co may be formed. In such a case, <sup>60</sup>Co exists in the form of metal. Therefore, to completely deal with these two forms of cobalt contaminants, cobalt plating and cobalt oxide film-coated specimens were studied. The principle of melt decontamination is to melt the metal and utilize slag or oxygen to form an oxide of the nuclide and then move it to the slag [8].

In the case of corrosion products, the nuclides already exist in the form of oxides or hydroxides but nuclides present in the form of metals need oxidation processes, and it is difficult to decontaminate them sufficiently. The experimental result of the cobalt oxidation-coated specimens was 2.07% higher than that of the cobalt plated specimens due to slag No.2 having the highest ability to collect nuclides. In the case of S41C, the decontamination result of cobalt was about 10%, which was not significantly different from the experimental result of SUS304. The decontamination effect of the <sup>60</sup>Co and <sup>58</sup>Co contamination specimens is about 8–11%. However, the cobalt removal rate is still insufficient.

According to the Ellingham diagram (Figure 7) [11], the Gibbs free energy of cobalt is highest, and the stability of cobalt oxide is the lowest. Therefore, at the time of melting, oxygen first bonds with a stable element to form an oxide, and it is difficult to form stable cobalt oxide and decontaminate it. Hence, we were not satisfied with the result of cobalt decontamination. However, based on the experience and knowledge gained from this research, we are planning to increase the decontamination effect of cobalt, such as by supplying oxygen and carburizing.



Figure 7. Gibbs free energy in Ellingham diagram [11].

#### 3.2. The Performance of the Slag

Melted slag is a liquid to which flux has been added for the purpose of removing impurities and radionuclides in the metal through melting. The roles of slag in melt decontamination are very important. The first role is to supply oxygen to the radionuclide during melting to make nuclide oxide. This is the reason why the removal rate of the coated specimen was higher than the removal rate of the plating specimen in the cobalt experiment. If the oxidizing power of the slag is not enough, we can supply oxygen to produce more cobalt oxide. However, although we supplied a small amount of oxygen in the experiment (experiment No.8), we did not obtain a large effect, and we plan to conduct additional experiments after the design of equipment.

As the basicity increases, the fluidity of the slag declines, which is disadvantageous for decontamination. The higher the basicity of slag, the higher the amount of oxygen that can be supplied. The B ratio of slag No.1 is 6, which is higher than the B ratio of 0.6 of slag No.2. However, as a result of decontamination, the nuclide capture power of slag No.2 is far stronger. Therefore, design optimization of the slag of each nuclide is very important.

The second role of the slag is to stabilize the oxide of the radionuclide, which moves in the slag while coming into contact with the molten metal. Therefore, if the thermodynamic stability of the slag is lower than the thermodynamic stability of the radionuclide oxide, the radionuclide oxide will not return to the metallic form again and will remain in the form of a stable oxide. However, if the slag with high thermodynamic stability is stabilized, the oxide of the radionuclide becomes relatively unstable and is finally reduced again in the form of metal.

In experiment Co (No.11), the weight of the specimen was 299.63 g and the amount of slag added was 20% of the specimen weight (60 g). In experiment Co (No.12), the weight of the specimen was 599.28 g and the amount of slag introduced was 10% of the weight of the specimen (again 60 g). Through the decontamination effect, the removal rate of nuclides

in the Co (No.11) experiment was 8.61% and the nuclide removal rate in the Co (No.12) experiment was 6.82%. When the amount of slag was doubled in the experiment with carbon steel, the removal rates of the nuclides all increased except for chromium. The more slag that was produced, the better the decontamination effect was. However, it is limited to the amount of reactive slag that is effective in the molten slag. Only some slag in contact with the metal surface will react with the metal. However, most slags are limited in flow ability or cool quickly, so they do not play a role in decontamination. Therefore, research to increase the effectiveness of slag is necessary.

Considering the characteristics mentioned, it is difficult and complicated to construct the composition of the slag. We need to make a ternary-system equilibrium-state diagram of slag, molten metal, and simulated nuclide at the melting point and study the characteristics of slag. In equilibrium, there are three variables: temperature, pressure, and time. High temperature, very long time, and low pressure are important. Therefore, the composition of the slag was determined in consideration of the state diagram under the experimental conditions of 1500–1700 °C, 1 atm, and 10–30 min.

## 3.3. Decontamination Characteristics of Cr, Mn, and Cs

The chromium decontamination experiment used a chromium-plated specimen on the surface of SUS304 and a surface coating on S41C. The highest removal rate of the plated specimen was 19.72% and the removal rate of the coated test piece was 88.76%. The reason for such a big difference is that it is much more difficult to transfer the plated chromium. Fortunately, most radioactive contaminated metals are contaminated with corrosion products on the surface and decontamination can be sufficiently done by the melt decontamination method.

The half-life of <sup>51</sup>Cr is about 27 days, and the half-life of <sup>54</sup>Mn is relatively long at 312 days, but the decontamination effect of manganese was very good (about 80%). After melting, manganese is distributed almost uniformly in the ingot. <sup>137</sup>Cs has a very long half-life of about 30 years, it releases beta rays upon collapse, and it is very dangerous to the human body. Fortunately, using the low volatility point (671 °C) of cesium, complex decontamination processes can easily separate cesium from the metal matrix. However, an excellent filter system must be constructed.

### 4. Conclusions

In this study, metal samples were plated or coated with non-radioactive isotopes of <sup>60</sup>Co, <sup>58</sup>Co, <sup>51</sup>Cr, <sup>54</sup>Mn, and <sup>137</sup>Cs, which are typical radionuclides generated during nuclear dismantling. Samples were melted by induction heating, the decontamination characteristics of each element were studied, and the following conclusions were obtained. The removal rates of chromium (88.76%), manganese (79.73%), and cesium (>90%) were excellent and will be useful for dismantling nuclear power plants and the treatment of radioactive metal waste. The removal rate of cobalt (about 10%) is not good enough yet, but there is little difference compared to the research results from developed countries.

After melting, each nuclide is uniformly distributed in the ingot.  ${SiO_2(50)-CaO(30)-Al_2O_3(20)}$  has the highest capture power and removal efficiency for each simulated nuclide element in SUS304 and S41C. The melting time has no significant influence on the final decontamination rate, even if we increase the melting time if the metal piece or the slag is completely melted. The basicity of slag is not proportional to the decontamination rate. When the basicity increases, the oxidizing power becomes strong, and the decontamination effect is excellent. However, at the same time, the fluidity of the slag decreases, which is disadvantageous for decontamination. By increasing the amount of slag formation, decontamination is more easily restricted to the effective amount of slag. As the area of molten metal and slag increases, the decontamination performance is improved.

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