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Study on Behavior of Radionuclides During the Melting of a Radioactive Metal Waste

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1. Introduction

During the operation or decommissioning of nuclear facilities and radioactive waste management facilities, various radioactive metal wastes are generated. In particular, according to the IAEA, it is predicted that approximately 3,500 tons of contaminated metal and approximately 650 tons of activated metal will be generated during the decommissioning of a 900-1300 MWe pressurized water reactor [1]. According to the European Commission, it is expected that the amount of metal waste generated from dismantling nuclear facilities in European countries over the next 60 years will be approximately 300,000 tons of stainless steel, 1 million tons of carbon or low-alloy steel, and around 20,000 to 30,000 tons of aluminum and copper. [2] For surface contaminated metals, if it is confirmed that the non-contaminated parts below the concentration for the free-release, they can be self-disposed after regulatory release. However, if radioactive metals are disposed of without separate treatment, they can occupy a large volume, resulting in high disposal costs. Therefore, processing is required to reduce the volume of these radioactive metal wastes. In foreign countries, a melting process for recycling these radioactive metals is being studied, and some countries have already applied this process to radioactive metal wastes. Prior to applying the melting process to radioactive metal wastes generated in domestic nuclear facilities, this study investigated the behavior of isotopes during metal melting of radioactive waste. To this end, we investigated the application of melting processes for radioactive metal waste in foreign countries and studied the distribution of isotopes during the melting of various radioactive metal wastes in each case. Finally, we predicted the existence form of radioactive isotopes in metal during melting using the HSC Chemistry program and compared it with the case studies.

2. Methods

2.1 Calculation using HSC Chemistry

The HSC Chemistry 9.0 program was used to understand the thermodynamic behavior of the radionuclides during the melting of the radioactive metal waste. The HSC Chemistry can be used to simulate chemical reactions and processes on a thermochemical basis [3]. The thermochemical database contains entalphy (H), entropy (S), and heat capacity (Cp) data for chemical compounds [3].

2.2 Simulation conditions

To confirm the behaviors of metal oxides and nuclides in the steel during the melting process of the radioactive metal were simulated using HSC Chemistry program. It was assumed that radioactive metal waste contains cobalt, uranium, and cesium. To verify whether radionuclides can be included into slag during the melting process, it was assumed that they exist within the major components of the slag composition, such as Al_2O_3, CaO, and FeO. The simulation temperature was set from 0 to 2,000 ^{o}C, including from 1,400 to 1,550 ^{o}C which is the temperature at which melting occurs. Additionally, to understand the reactions of isotopes generated during the melting process, the HSC Chemistry program was used to calculate the Gibbs free energy.

3. Results and discussion

3.1 Example of the radionuclide distribution during melting

According to NCRP data listed in table I, it has been confirmed that Mn, Co, Ni, Ag, and Sb among the nuclides contained in carbon steel and stainless steel are mainly present in the ingot. However, Sr, Sm, Eu, U, Pu, and Am are distributed in the slag. Additionally, Zn and the highly volatile nuclide Cs are included into slag or dust. Such behavior has also been observed in the case of large components composed of stainless steel and carbon steel melted using an induction furnace-type melting furnace at Studsvik (Cyclife) in Sweden. For Co-60, 88-99% was found in the ingot, 1-6% in the slag, and 0-7% in the dust. For Cs-137, less than 1% was present in the ingot, with 71-90% distributed in the slag and 10-29% in the dust. [5].

Table I: The distribution of major nuclides during melting of steel [4]

Nuclides	Ingot (%)	Slag (%)	Dust (%)	Etc. (%)
Mn-54	24-100	1-75	0-5	0
Co-60	20-100	0-1	0-80	0
Ni-63	90	10	0	0
Zn-65	0-20	0-1	80-100	0
Sr-90	0-20	95-100	0-10	0
Ag-108m	75-100	0-1	0-25	1 (bottom)
Sb-125	60-100	0-20	10-40	0
Cs-137	0	0-5	95-100	0
Sm-151	0	93	7	0
Eu-152	4	95	1	0
U	0-1	95-100	0-5	0
Pu	0-1	95-100	0-5	0
Am-241	0-1	95-100	0-5	0

3.2 Thermodynamic simulation

Figure 1 shows the Ellingham diagram obtained by the simulation using the HSC Chemistry program. It is determined that CoO has lower stability than FeO, CaO, and Al_2O_3, which make up the slag. On the other hand, UO_2 is found to be more stable than FeO and CaO. However, Cs is confirmed to exist in the gas phase even at low temperature.

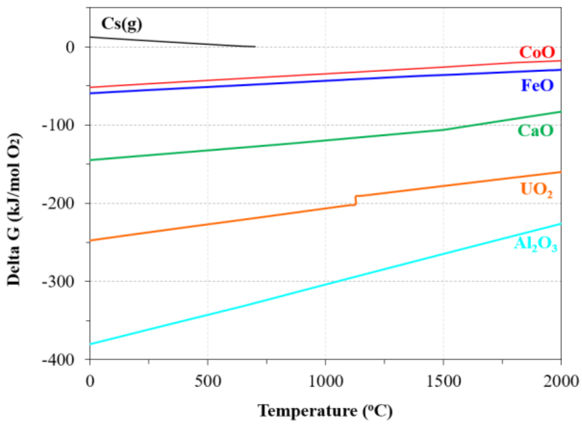

Fig. 2. Ellingham diagram simulated using HSC chemistry program.

3.3 Behaviors of radionuclides during melting

During the melting of radioactive metal waste, reduction reactions occur between radionuclides and metal oxides. As shown in Fig. 1, in the case of Co, which has a lower oxygen affinity than other composition of steel, such as Fe, Ca, and Al, at the melting temperature of about 1,500 ℃. From this reason it is expected to exist in the molten phase rather than in the slag. Therefore, it is predicted to be distributed within the ingot without moving to the slag. In the case of U, as shown in Fig. 1, it has been confirmed to have a higher oxygen affinity than Fe and Ca, which are the compositions of steel. Therefore, U, which receives oxygen from metal oxides, can be converted into UO_2 and be included in the slag in the form of an oxide Eq. (1) represents the reaction for the formation of UO_2, which was calculated using the HSC Chemistry program. It was confirmed that that oxygen is transferred from FeO and CaO to produce UO_2.

$$FeO + CaO + U \rightarrow Fe + Ca + U\,O_2$$
$$(\triangle G = -35.95\ kcal,\ 1{,}500\ ^{o}C) \qquad (1)$$

Cs, the volatile nuclide, is expected to be evaporated as vapor during the melting process and to be present in the slag or dust phase. These results are similar with the experimental results from NCRP data and Cyclife (Studsvik) mentioned in 3.1.

4. Conclusions

In this study, the behavior of radionuclides during the melting of radioactive metal waste containing cobalt, uranium, and cesium was predicted and compared with experimental cases. The results showed that:

(1) Cobalt exists in the ingot because its reactivity with oxygen is lower than FeO, CaO, and Al_2O_3, which is composed of the slag.
(2) Uranium reacts with FeO and CaO to form UO_2, which is included in the slag as an oxide.
(3) Cesium is highly volatile and evaporates during the melting process, so it is included in the slag or dust.

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