RADIOACTIVE METAL MELTING TEST AT JAPAN ATOMIC ENERGY RESEARCH INSTITUTE

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ABSTRACT

In order to elucidate the behavior of radionuclides during melting operations of the radioactive metal waste, melting tests were conducted using contaminated metal waste generated from the JPDR decommissioning program or simulated waste with radioactive isotopes as tracers. A series of tests indicated; 1) distribution of radionuclide in the ingot produced are fairly uniform, 2) while Mn-54, Co-60, Zn-65 likely remain in an ingot to a great extent, Sr-85 and Cs-137 move to slag or exhaust gas leaving molten metal.

INTRODUCTION

In Japan, as the standard process for the decommissioning of a nuclear power plant, the complete dismantling after 5 to 10 year's safe storage is assumed in order to reuse the site for follow-on nuclear programs. The decommissioning of the nuclear power plants currently operating in Japan will be inevitable from the 2010's and a vast volume of low and very-low level wastes will be produced within a short period of time by dismantling of the nuclear power plants. Effective recycling of these waste will be very important to carry out the decommissioning smoothly. Especially, recycling of metal waste by melting, converting metal waste to useful resources, will reduce the amount of waste and ease the problem of disposal. It also will serve the verification of the very low activity level of metal waste because it is said that consolidation by melting will be of homogeneous composition and radioactivity.

Under this background, the Japan Atomic Energy Research Institute (JAERI) has been conducting radioactive metal melting tests under the sponsorship of the Science and Technology Agency (1-4). The main objective of the tests is to elucidate the transfer behavior of radionuclides during melting operations, which will contribute to developing regulation standards on the recycling of contaminated metals in Japan in the future.

The radioactive metal melting test program began by designing test equipment in 1987. Installation of the equipment was completed by the end of March 1990, then the cold tests using non-contaminated steels were conducted. These tests provided practical operating experience with the equipment and established safe test procedures.

Following the cold tests, radioactive metal melting tests using metal waste generated from the decommissioning of the Japan Power Demonstration Reactor (JPDR) were conducted 6 times by September 1991. The RI tracer tests, which started late in 1991, are under way.

MELTING TEST

Overview of Tests

Radioactive metal melting tests were conducted using dismantled scraps from the JPDR or radioactive isotopes (RIs).

In the JPDR decommissioning material tests which used dismantled waste, contaminated pipes from the reactor primary cooling system and irradiated components related to the reactor vessel were melted. The objects melted and activities contained are summarized in Table I.

In the RI tracer tests, Mn-54, Co-60, Zn-65, Sr-85 (a substitute for Sr-90), and Cs-137 beta/gamma emitters were used as the tracers to elucidate the transfer behavior of radionuclides into the ingot, slag, dust, etc. These RIs were selected among the radionuclides which will be typically found in LWR's dismantled wastes. Considering the effects of melting temperature and slag characteristics on the transfer behavior, type of steel and the basicity (CaO/SiO_2) of flux used were varied as test parameters. Summarized in Table II are the test conditions.

Melting Equipment

Melting tests have been carried out in an existing building in the JPDR site, which was renovated to install melting equipment. Shown in Fig. 1 is the schematic view of melting equipment at the JAERI. The main component is a high-frequency (1,000 Hz) induction furnace with a loading capacity of 500 kg steel. The furnace is equipped with a ring hood on the top of the crucible as well as the usual lid, to form an air-lock during melting. Molten metal is poured directly into an ingot mold from the furnace. The furnace and related equipment are installed in the steel chamber, which is equipped with local ventilation system and monitoring glass windows. All operations are carried out from the outside of the steel chamber.

Test Method

Adding flux, removing slag, measuring temperatures, casting, and so on, were required for the operations to carry out a melting test. As an example, the procedure for the RI tracer test is schematically shown in Fig. 2 from the beginning of heating to the casting. In our tests the purpose is to elucidate the transfer behavior of radionuclides, special attention was paid to the melting procedure and the sampling method. First, to assess the influence of residual activity remained inside the furnace crucible on the activity balance, molten metal and slag samples were taken immediately after the complete melting of charge material. Second, to control the basicity of flux as possible as planned, the slag formed was removed once and then newly reproduced using chemically- adjusted flux before adding RIs into molten metal.

In current series of tests, addition of RIs was performed by charging a small vessel which contained RI-contaminated metal tips into the molten metal. Other method of RI addition will be tried in future tests, for example, addition from the commencement of the melting.

Following the addition of RIs, samples were taken from slag and molten metal three times, at intervals of 5 minutes. The objectives are: 1) to reveal the relationship between the

TABLE I
Summary of Melting Tests Using JPDR Dismantled Components

Test No.	Components	Material	Activity Concentration in Components[a]
JP-1	Pipes	SUS 304	Less than 2.1×10^{-2} Bq/cm^2
JP-2	Pipes	SUS 304	Less than 2.4×10^{-2} Bq/cm^2
JP-3	Pipes	ASTM-A335	Less than 4.1 Bq/cm^2
JP-4	Pipes	ASTM-A335	Less than 6.2 Bq/cm^2
JP-5	Stud Volts (Top of PV)	ASTM-A193	6.2 Bq/g (Typical)
JP-6	Stabilizers (Attached to PV)	ASTM-A668	50 Bq/g (Typical)

[a]Radionuclides were detectable for Co-60 only.

TABLE II
Summary of Melting Tests Using Radioisotopes

Test No.	Material Used	Temperature Melted (K)	Basicity of Flux (CaO/SiO_2)	Radioisotopes Used[a]
RI-1	ASTM-A335	1,913	1.0	Mn-54,Co-60,Sr-85
RI-2	ASTM-A335	1,873	1.0	Zn-65,Cs-137
RI-3	ASTM-A335	1,903	1.0	Mn-54,Co-60 Zn-65, Sr-85,Cs-137
RI-4	SUS 304	1,823	3.0	Mn-54,Co-60 Zn-65, Sr-85,Cs-137
RI-5	SUS 304	1,843	3.0	Mn-54,Co-60 Zn-65, Sr-85,Cs-137

[a]Amount of each radioisotope are as follows (per test):
Mn-54 (2MBq), Co-60 (1MBq), Zn-65 (4MBq), Sr-85 (4MBq), Cs-137 (4MBq).

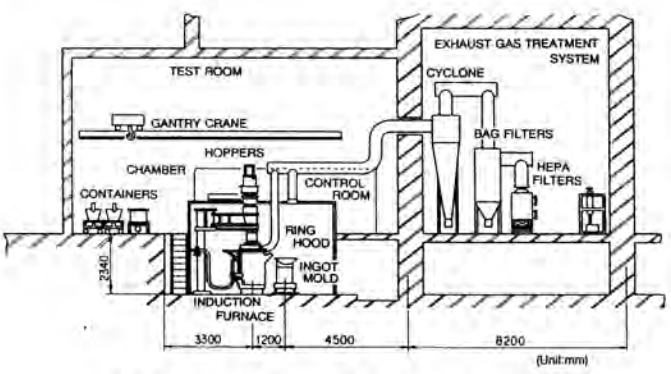

Fig. 1. Schematic view of melting equipment.

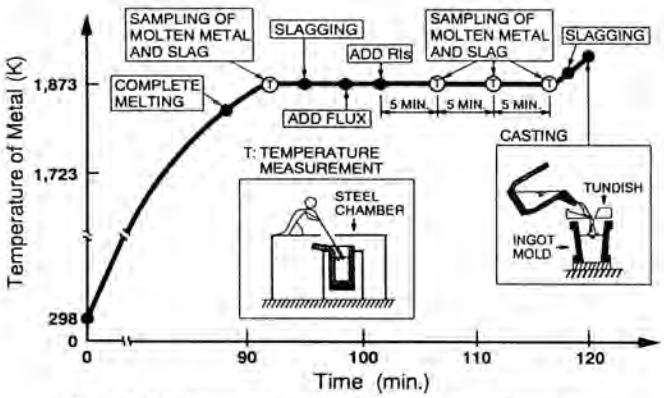

Fig. 2. Melting procedure for the RI tracer test.

activity concentrations of RIs in the slag and molten metal and that of the ingot produced, 2) to analyze the chemical compositions of samples for estimating melting atmosphere.

Melting operations are completed by pouring molten metal directly into an ingot mold after removing slag remained again. As shown in Fig. 3, twenty-six samples were taken from three cross sections of upper, middle, and lower portions of each ingot produced. These samples were used to determine the activity concentration in the ingot. Activity in exhaust gas was assessed by the activity measurements of filters, attached to the sampling lines.

TEST RESULTS

Balance of Material

The total weight of products was compared with that of charge material in each test of the JPDR decommissioning material tests (6 times) and the RI tracer tests (5 times). The balance of material from a total of eleven tests was 99.5% on the average. Although some of molten metal remained on the

TABLE III
Activity Concentration in Ingot

Sample No.	Mn-54			Co-60			Zn-65		
	A	B	C	A	B	C	A	B	C
No.1	4.07	4.04	3.70	2.00	2.28	2.16	6.13	6.60	6.34
No.2	3.94	4.02	4.16	2.13	2.71	2.29	5.93	7.27	7.21
No.3	4.35	3.86	4.20	2.55	2.22	2.03	7.58	7.06	6.42
No.4	3.96	4.25	4.07	2.44	2.23	2.10	6.77	7.05	6.21
No.5	4.10	4.00	4.22	2.35	2.04	2.40	6.79	6.48	6.01
No.6	3.91	4.23	4.22	2.05	2.17	2.27	6.64	7.66	5.53
No.7	4.07	3.91	-	2.35	2.01	-	8.27	5.85	-
No.8	3.90	3.70	-	1.94	2.28	-	7.03	6.97	-
No.9	4.22	3.97	-	2.24	1.75	-	7.01	5.73	-
No.10	4.40	4.07	-	2.26	2.25	-	6.91	6.02	-
Mean	4.06 Bq/g			2.21 Bq/g			6.67 Bq/g		
S.D./Mean	4.4 (%)			9.0 (%)			8.7 (%)		

Notes: 1) A,B,C: Cross sections at upper, middle, and lower portions of an ingot (see Fig.3.)
2) S.D.: Standard deviation

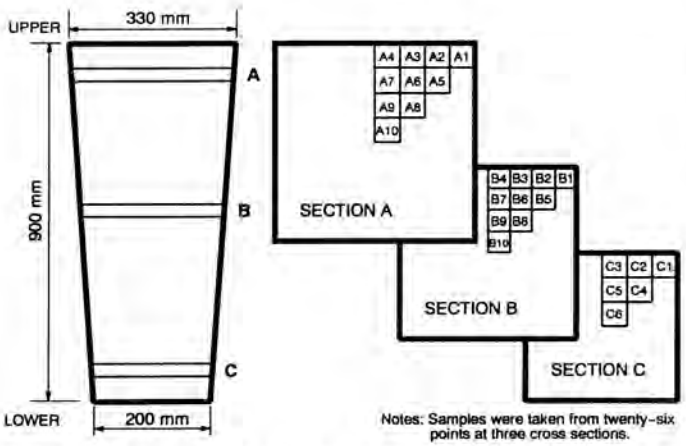

Fig. 3. Sampling method for activity measurement of ingot.

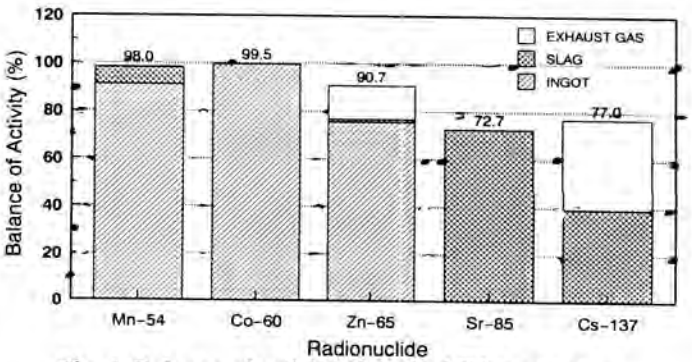

Fig. 4. Balance of activity of the 3rd RI tracer test.

crucible wall of the furnace, the maximum difference in the material balance was within about 3% through all the tests.

The weight proportions of the products were about 95% for an ingot, 2 to 3% for slag, less than 0.1% for dust, and 1 to 2% for others (residual metal on the tundish and metal splash), to the total weight of charge material.

Balance of Activity

The balance of activity in each test was assessed based on the activity measurement of the ingot, slag, etc. The total activities of the products were calculated by multiplying their measured activity concentrations by the total weight of each product. The activity in exhaust gas was estimated through the activity measurement of filter papers, which were attached to the off-gas lines to trap radionuclides, considering the sampling flow rate of exhaust gas.

As an typical example, the activity balance of the 3rd RI tracer test is shown in Fig.4. As can be seen in this figure, Mn-54, Co-60, and Zn-65 remained in the ingot to a great extent. A total of 90 to 100% of their activity was detected in the ingot. However, a little of Mn-54 was detected in slag. A little of Zn-65 were also detected in exhaust gas.

Sr-85 and Cs-137 moved to the products other than ingot. Sr-85 was detected in slag only. Cs-137 was detected approximately by half in both slag and exhaust gas. However, some portion of these radionuclides were missing. Activities detected were only 73% for Sr-85 and 77% for Cs-137 compared with the each activity used in the test. Though the causes of it were not identified exactly at this stage, possible reasons are described as follows:

1. Activity of slag was estimated based on that of a lump of slag without measuring all of slag.
2. Activity assessment in exhaust gas is based on a small volume of sampling (about 1/2500 of the total exhausted volume).
3. It is difficult to assess the activity accumulated within the off-gas system owing to the very low level of activity.

The activity balance in other four RI tracer tests also show the same tendency with that of the 3rd test.

Activity Concentration Changes in Molten Metal

The activity concentrations of molten metal samples and the produced ingot were compared to understand their relationship. They are shown in Fig. 5 every radionuclides. With regard to Mn-54 and Co-60, the activity concentrations of molten metal samples approach to that of ingot after about 10 minutes and a little concentration changes are observed after that. This suggests that Mn-54 and Co-60 diffused homogeneously in molten metal within a short time and remained without leaving during melting.

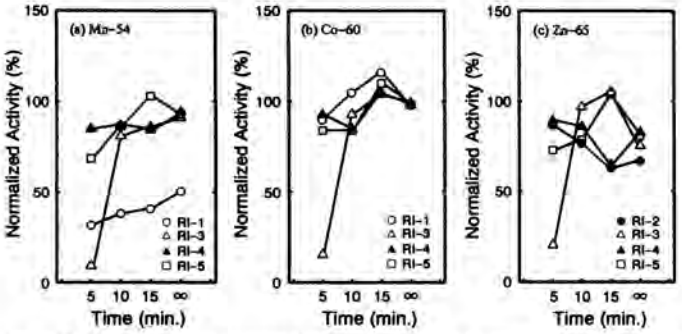

Fig. 5. Activity concentrations in molten metal as function of time. Notes: 1) ∞ stands for a solid state (ingot). 2) activity is normalized to added activity of each nuclide.

The activity concentration of Zn-65 in molten metal changes largely after the addition of RIs. But, the activity concentration of the produced ingot showed almost a constant value each test. Because sampling of molten metal is much easier than that of the produced ingot, the result described above implies that the activity concentration in the ingot can be easily estimated by using molten metal samples, if it is confirmed that radionuclides in the produced ingot are homogeneously distributed.

The activity concentrations of both Sr-85 and Cs-137 in the molten metal and in the produced ingot were less than the detection limit. It is considered that these radionuclides do not remain in molten metal and move to slag or exhaust gas within a short time.

Activity Concentration in Ingot

As described in the previous section, three radionuclides of Mn-54, Co-60, and Zn-65 were detected in the produced ingot. Activity concentrations at three cross sections of the ingot were, therefore, examined to confirm its activity homogeneity.

Activity concentrations at the upper, middle, and lower cross sections of the ingot, which were obtained from the 3rd RI tracer test, are shown in Table III as an typical example. Standard deviations calculated from twenty-six measured values were 4.4% for Mn-54, 9.0% for Co-60, and 8.7% for Zn-65 compared with each average. The activity concentrations at three cross sections along the longitudinal direction of the ingot can be said to be almost uniform. This characteristics of activity homogeneity did not depend on the type of nuclides remained in the ingot.

CONCLUSIONS

In order to investigate the behavior of radionuclides during the melting operations, melting tests using JPDR decommissioning waste or RIs were conducted. By reviewing the balances of material and activity through the test results to date, the basic transfer behavior of radionuclides has been understood. Though the data obtained are not enough to conclude yet, main accomplishments would be described as follows:

1. Balance of material related to melting was kept well.
2. Three nuclides, Mn-54, Co-60, and Zn-65, were transferred into the produced ingot by 90-100 % after melting. But, a part of Mn-54 was detected in slag and a part of Zn-65 was also detected in exhaust gas.
3. Sr-85 and Cs-137 were transferred into slag or exhaust gas without remaining in the ingot. Especially, Sr-85 was detectable in slag only.
4. Activity concentrations in the produced ingot was essentially homogeneous. It indicated a few differences at three cross sections along the longitudinal direction of the ingot. This was observed for all the RIs remained in the ingot.

In order to accumulate much data, the RI tracer tests are being continued at present, by changing test conditions such as the basicity of flux and the melting method of RI-contaminated steels (charging RI- contaminated steels at the beginning of melting). In addition, to identify the melting atmosphere, samples taken from molten metals, slags, and ingot have been analyzed on their chemical compositions.

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