High-frequency melting technology for radioactive metal waste

Nobutake Horiuchi, Takeshi Nishikawa, Naoto Sasaki, and Hirokazu Tanaka

Energy Project & Technology Center

Mitsubishi Materials Corporation

1002-14 Mukohyama, Naka-city, Ibaraki-pref.

Key words; Melting, radioactive metal scrap, decontamination, volume reduction, recycling

Abstract

Mitsubishi Materials Corporation has been developing the melting technology for metal waste by high-frequency melting, for volume reduction, decontamination and recycling of metal scrap contaminated with radioactive materials from nuclear power plants and nuclear facilities. By the melting treatment, high volume reduction of metal waste and decontamination of radionuclides such as cesium, strontium and α nuclides are expected. In addition, the melting process is also suitable for the metal recycling, because the process can homogenize the waste and cast it into any shape.

Through development, we confirmed the followings; - the volume of metal waste can be reduced from 1/5 to 1/3 by melting, - the radioactivity concentration of Cs of metal that actually contaminated due to a nuclear accident can be set below the detection limit, - demonstration scale test with simulated substances can decontaminate Cs, Sr and U. In addition, we have been developing manufacturing technology to make the metal waste containers by centrifugal casting method, which was applied to advanced volume reduction facilities of Japan Atomic Energy Agency (JAEA).

1. Introduction

Reducing the final disposal amount of radioactive waste in decommissioning nuclear facilities is an important issue. In order to reduce the amount of waste, decontamination and volume reduction are effective. In addition, when reducing the amount of waste by recycling, it is necessary to measure the radioactivity concentration and establish a reuse method.

Mitsubishi Materials Corporation (MMC) has been developing a melting decontamination technology for metal waste by high-frequency melting. By using melting decontamination technology, the radionuclide will be removed, and it will be possible to collect metal materials and recycle it. Also, the amount of secondary waste to be final disposal can be greatly reduced compared to the amount of metal waste before decontamination. Furthermore, since the composition of the metal becomes uniform, the radioactive concentration measurement becomes easy.

Below we explain the mechanism of melt decontamination, the experimental equipment owned by Mitsubishi Materials, and the results of development. In addition, we will also introduce the development of the ultra-high frequency furnace that can be used for the treatment of waste other than metals such as ceramics

2. The mechanism of melting decontamination

The melting decontamination technology is based on the difference in free energy of formation of metal oxide or the difference in boiling point between metal and radionuclide (Fig.1). Since the boiling point of Cs and its compounds is lower than the melting temperature of iron (1,600 ° C.), when iron-based metal waste is melted, Cs is volatilized and removed from the metal. The part of Cs is supplemented by slag during the transition from the metal phase to the gas phase.

Sr and α nuclides (such as U, Pu) exist stable as oxide as compared with iron (Fig.2). In the electromagnetic field of the high-frequency induction furnace, the metal is move to the center of the furnace by electromagnetic force, on the other hand, the oxide, which is not affected by electromagnetic force, moves relatively to the outside of the furnace. In addition, the metal has good homogeneity because molten metal is stirred by electromagnetic force.

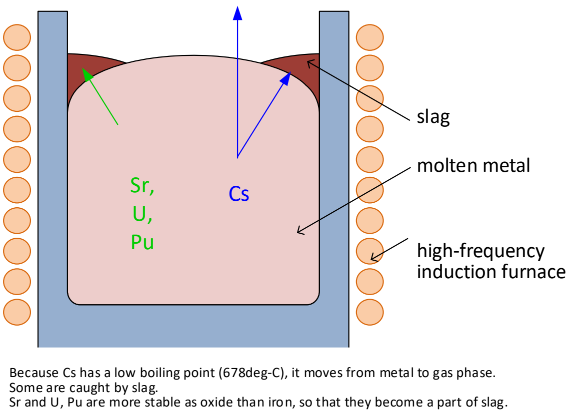


Fig.1 Image of nuclide behavior in high frequency induction furnace

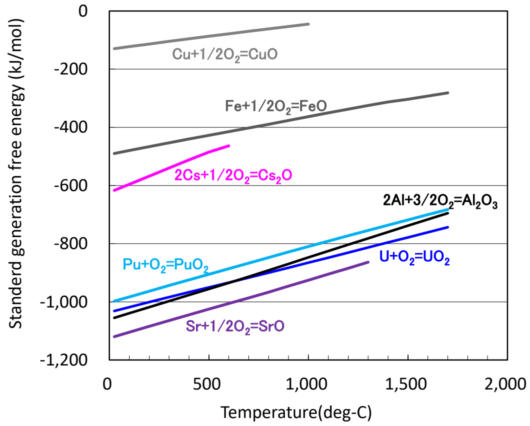


Fig.2 Free energy of formation of metal oxide
*The Japan Society of Thermometry Calculated by thermodynamic data calculation software “MALT2”

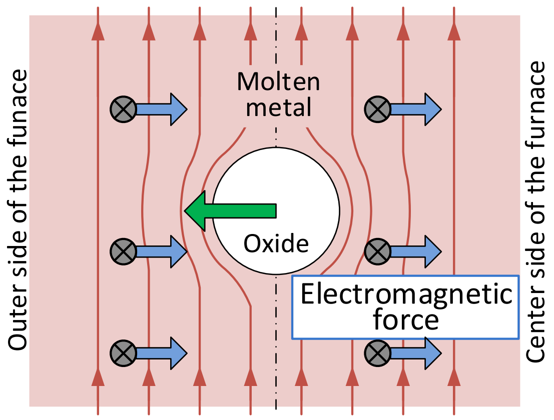


Fig.3 Image of oxide behavior in high frequency induction furnace.

3. Experimental apparatus and facilities

MMC owns three experimental furnaces (Table 1). One is a basic test apparatus that measures material balance and mass transfer rate in a small test furnace (capacity: 1kg-Fe, U available). The other two are for demonstration tests and the applicability of melt decontamination on a real scale was confirmed.

Table 1 Specs of the experimental apparatus and facilities

	No.1	No.2	No.3
Heating Method	High frequency induction heating		
Capacity (For Iron)	1kg	1ton	430kg
Input Power	30kW	600kW	250kW
Frequency	3kHz	500Hz	1kHz
Furnace Size	ϕ 60 × 150mm	ϕ 490 × 900mm	upper ϕ 722 bottom ϕ 400 × 600mm
Remark	U available	Using simulated substances	U available
Fig.	Filg.4	Fig.5	Fig.6

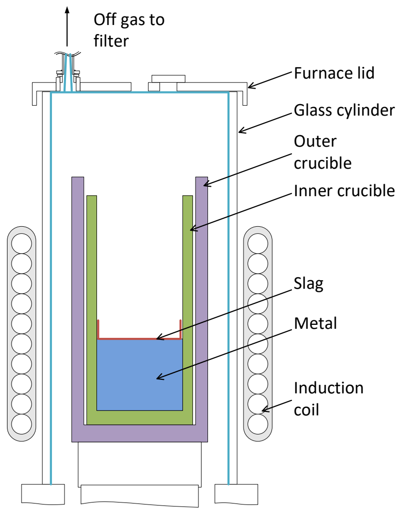


Fig.4 Overview of No. 1 basic test apparatus

Fig.5 No.2 demonstration scale furnace (Capacity: 1ton-Fe/batch)

Fig.6 No.3 engineering scale furnace (Capacity: 430kg-Fe/batch)

4. Development Results

4-1 Volume reduction

Because of the wide variety of shapes of metal waste, when metal waste is put into a disposal container as it is, many useless voids are formed. On the other hand, when the metal waste is melt-processed, unnecessary voids can be eliminated since the metal is made into a uniform ingot.It was confirmed by the test that the volume of metal waste can be reduced from about 1/5 to 1/3. Furthermore, we confirmed that the amount of secondary waste such as refractories, slag, filters, etc. generated by the melting process is about 5% of the treated material [1].

4-2. Decontamination of Cs and Sr

Due to the accident at Fukushima Daiichi Nuclear Power Station, a large amount of radioactive substance, Cs, Sr, etc. was released into the atmosphere. For this reason, a large amount of metallic rubble contaminated with radioactive Cs and Sr has been generated, and their decontamination and reduction has become a subject. Therefore, we confirmed the decontamination effect of radioactive Cs and Sr by melting.

Melting decontamination test using real metal waste contaminated with radioactive Cs caused by the accident was carried out using our No. 1 basic test furnace. As a result, it was confirmed that real metal waste can be decontaminated to less than the detection limit (0.2Bq/kg) even when the metal is contaminated with soil contaminated with Cs (Table2) [2].

To understand the behavior of Cs and Sr, melting decontamination test using stable nuclides of Cs and Sr were carried out (Fig.7). As a result, we confirmed the followings. 1) It is possible to decontaminate Cs and Sr from the metal, 2) As the added amount of the slag material increases, the amount of migration of Cs and Sr to the slag increases, 3) Since Cs and Sr concentrations in the ingot could be confirmed with representative sample, radioactivity concentration of metal after melting process can also be measured by representative sample measurement.

To confirm those phenomenon in the demonstration scale, we carried out the test using No.2 demonstration scale furnace (Fig.8). As a result, it was confirmed that there is no difference between basic test and demonstration test on decontamination performance and uniformity by representative sample (Table 3).

Table 2. Results of melting decontamination test using real metal waste contaminated with fall-out radioactive Cs

		Radio Activity[Bq/kg]	
		Run1	Run2
Before	Metal Scrap	5.6	5.6
	Soil	—	10,500
After	Ingot	<0.2	<0.2

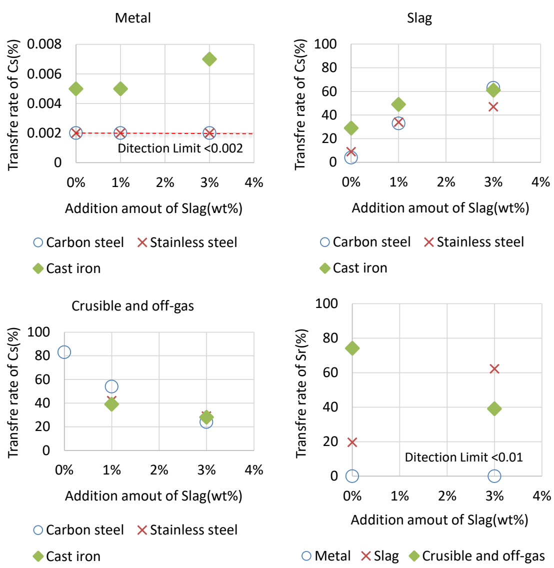


Fig.7 Behavior of Cs and Sr in the basic test.

Table 3. Concentration of Cs and Sr in the demonstration scale test.

		Concentration [ppm]	
		Cs	Sr
Before	Simulated Sample	501	500
After	Ingot 1	<0.02	<0.1
	Ingot 2	<0.02	<0.1
	Representative sample	0.04	<0.1

Lower detection limit: Cs 0.02ppm, Sr 0.01ppm

Fig.8 Implementation status of demonstration scale test (casting)

4-3 Decontamination of U

In nuclear fuel cycle facilities such as nuclear fuel processing facilities, a large amount of metal waste contaminated with U is generated. We confirmed the applicability of melt decontamination to metals contaminated with U.

To know the behavior of U, we carried out engineering scale test using No.3 engineering scale furnace (U available) [3], [4]. As a result, the followings were confirmed (Table 4). 1) The uranium concentration in the metal can be reduced to several ppm or less, in terms of natural U <0.1 Bq/g by melt decontamination. 2) U concentration measurement is possible with representative sample. 3) The metal after treatment has a uniform composition.

Table 4, Concentration of U in the engineering scale test.

		Concentration of U [ppm]	
		Run1	Run2
Before	Simulated Sample	1,000	1,000
After	Ingot 1	1.5	2.3
	Ingot 2	2.1	2.3
	Ingot 3	1.4	4.6
	Representative sample	1.3	3.2

Lower detection limit: U 0.5ppm

4-4 Recycle technology and deployment to actual facility

In order to promote recycling, it is necessary to create a method of reusing metals. In the case of recycling metal contaminated by activation, it is not possible to decontaminate radionuclides such as Co - 60 by melt decontamination, so that the treated metal needs limited reuse in nuclear facilities.

Therefore, we have been developing technology to manufacture metal waste containers by centrifugal casting method, because this method has few secondary waste, is easy to manufacture, and is suitable for remote operation. By using the centrifugal casting method, it is possible to manufacture a cylindrical container without a core mold. It was adopted for advanced volume reduction facilities (AVRF) of JAEA. A part of AVRF, the melting furnace and related facilities were designed and built by the MMC group.

If metal waste is made into ingots, they can be processed into various products later. Melting technology may be effective as a means to reduce and recycle radioactive metal waste generated at nuclear facilities such as TEPCO's Fukushima Daiichi NPP.

Fig.9 Metal Container made by centrifugal casting method

5. Development of Ultra-high frequency induction furnace.

In a high frequency induction furnace, since only the metal is heated, it is not possible to make miscellaneous solid waste that contained nonmetals such as concrete and glass by direct induction heating. Therefore, we developed an ultra-high frequency furnace that can directly heat melt of miscellaneous solid waste by induction heating. The developed furnace uses an induction frequency of about 100 kHz (Fig. 10).

As a result of melting several sample mixed with carbon steel and glass using a ultra-high frequency furnace, we confirmed that it is possible to make solidified bodies with low porosity composed of slag and metal, and the compositions of the metal layer and the slag layer are respectively uniform (Fig. 11) [5].

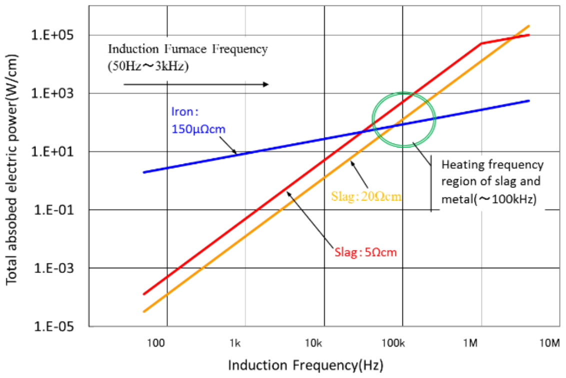
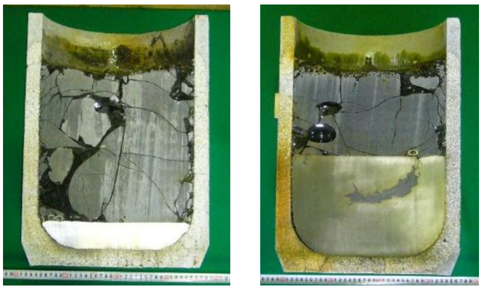


Fig.10 Relation between total absorbed electric energy and induction frequency



Metal / Slag =1/9, 5/5

Fig.11 Example of a molten and solidified product produced in an ultra-high frequency furnace

6. Conclusion

High frequency melting technology for radioactive metal waste developed by MMC was summarized. It was confirmed that Cs, Sr, U can be decontaminated from metal, radioactive concentration measurement can be easily performed by a representative sample. We also confirmed the applicability of the ultra-high frequency furnace. We are planning to continue to develop the melting technology, with the aim of contributing to the decommissioning of nuclear power plant including Fukushima Daiichi NPP.

7. Acknowledgments

The development of melting technology of radioactive metal waste introduced here was carried out under the guidance and cooperation of many people, demonstration project of MOE, Basic research programs of vitrification technology for waste volume reduction of METI, JAEA, RWMC, effective use project of METI, and so on. I deeply appreciate it here.

References

[1] MMC, AESJ 2009 Fall Meeting, L19
[2] 2015, MOE, FY2017 Demonstration Project for Decontamination Technology.
[3] 2002, JNC, JNC technical report No.14
[4] 2004, RWMC, Uranium waste decommission technology investigation.
[5] 2008, MMC et al, Innovative and Variable Nuclear energy Technology (IVNET) Development Project, Development of the System of the Volume Reduction Treatment System of Waste by the Ultra-High Frequency Induction Furnace.