EVOLUTION OF CONTINUOUS MELTING AND DECONTAMINATION TECHNOLOGY FOR DISMANTLED METAL BY AN INDUCTION COLD CRUCIBLE


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ABSTRACT

An induction cold crucible melting is one of the most promising technologies for the reuse and decontamination of the radioactively contaminated metallic materials generated during the dismantling of nuclear facilities, because the crucible ensures a long life operation without generating secondary wastes.

Fundamental equipment of the Melting and Recycling of Metals by Cold Crucible (MERC) process was developed in 1996 and 1997. In the MERC, surrogates of dismantled stainless steel, such as tubings, pipings, plates, bars, channels and mechanical parts, were continuously supplied together with flux for decontamination, followed by melting in the crucible. The melt was precisely withdrawn ensuring the melt dome was kept at a suitable level for the melting process. The maximum withdrawal velocity employed was 12 mm/min. Round ingots of 140 mm in maximum diameter and rectangular slabs of 50 mm x 170 mm in maximum dimension were cut in 300 mm lengths by a lubricant free mechanical saw, followed by slag delamination. They were automatically transported to the outlet of the equipment by a conveying system. The ingots proved to be suitable for manufacturing into shielding material and containers for highly active wastes.

Concerning the tracer partitioning using the simulated radioactive nuclides, the following results were obtained. More than 90% of Co and 60% of Mn remained in the ingot. While, more than 85% of Sr, 80% of Hf and 80% of Ce were partitioned to the slag, more than 80% of Cs and 55% of Zn were partitioned to the dust.

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INTRODUCTION

Melting, decontamination, formation to the suitable shape for the reuse and immobilization of radioactive nuclides are the important key technologies for the reuse of dismantled metals contaminated by low level radioactivity removed from the nuclear facilities. There are several processes offering melting and immobilization, such as plasma melting and induction hot crucible melting (1,2,3). However, there are few processes in which melting, decontamination, formation and immobilization proceed simultaneously.
An induction cold crucible technology has peculiar characteristics that the dismantled metals experience melting and solidification in the same crucible consecutively and are withdrawn continuously. This enables the melting, immobilization and formation of the metals to be reused by selecting the suitable sectional shape of the crucible. Continuous melting and withdrawal ensure sound enough quality for the reuse of the resultant ingot compared to the batch treatment.

Another characteristic of the cold crucible is that the cold wall is harm free against both high temperature metal and the slag in contact. Oxidation of the radioactive nuclides by the slag, followed by the transfer of the nuclides from the metal to the slag is promoted by the vigorous agitation of molten metal caused by electromagnetic force. These features result in the maintenance free of the crucible wall and the progression of decontamination during melting.

The induction cold crucible has long been used for the melting and casting of chemically active and high purity materials, such as titanium alloy and polycrystalline silicon ingot for photovoltaic solar cell(4,5,6,7). In 1995, fundamental technologies concerning the MERC(Melting and Recycling of metals by Cold crucible)process have been established based on the conventional induction cold crucible technology by using the cylindrical crucible of 45mm in diameter(8). In succession to the fundamental study, the testing MERC process was designed, manufactured and scaled up. Investigated items of the present study are the followings.

- Test of melting and solidifying the surrogates of dismantled metals by the scaled up MERC installed with not only circular but also rectangular sectional crucibles
- Quality of the ingot on the surface as well as inside
- Treatment rate of MERC process
- Transfer of simulated nuclides

**TESTING METHOD**

A schematic view of the testing MERC apparatus is shown in Fig.1. A water-cooled and segmented crucible made of copper is surrounded with a multi-turn induction coil. After the primary, which is a base metal for the melting, was inserted from the bottom opening to the suitable level for the melting in the crucible where the magnetic field is most effective, the atmosphere in the chamber is replaced with Ar gas. With an increase in power input from the coil through the cold wall permeable of electromagnetic energy to the primary, it melts by Joule heating to rise in a dome shape by the electromagnetic force. The molten metal experiences electromagnetic stirring and the top of the melt is out of contact with the cold wall, resulting in the increased heat efficiency.

After the primary was melted, metallic materials, such as piping or fragments of metals, are supplied from the crucible top opening onto the melt dome either by horizontal vibratory feeder or vertical linear feeder, depending on the shape of the material. Some amount of the decontaminating flux is also supplied through the vibratory feeder. Precise control on the withdrawal velocity and the supply of the metallic materials preserves the dome height at a certain fixed level during operation. As a result, an ingot which experiences melting and successive solidification is obtained.
Ingots are cut in every 300mm length by the mechanical lubricant-free rotating saw followed by slag delamination and conveyance to the outlet of the chamber. The operation is performed automatically except for attaching materials to the feeder.

![Schematic view of the testing apparatus MERC.](image)

**Table I. Specification of the testing apparatus MERC.**

<table>
<thead>
<tr>
<th>Specification</th>
<th>Details</th>
</tr>
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<tbody>
<tr>
<td>Output of the electric power</td>
<td>Max. 150 kW</td>
</tr>
<tr>
<td>Frequency</td>
<td>25 kHz</td>
</tr>
<tr>
<td>Dimension of the ingot</td>
<td>φ100x300 mm, φ140x300 mm, 110x50x300 mm, 170x50x300 mm</td>
</tr>
<tr>
<td>Withdrawal velocity</td>
<td>1~12 mm/min (automatic), 50 mm/min (manual)</td>
</tr>
</tbody>
</table>

Specification of the testing MERC apparatus is shown in Table I. The electric power source is 25kHz in frequency and 150kW in the maximum output. Four different kinds of the crucible shape were prepared to investigate the forming function of the crucible.

Either fragments of stainless steel (5mm in diameter and 10mm in length) or tubings (20mm in diameter and 500mm in length) are supplied from the top opening of the crucible onto the melt dome.
Major testing parameters are the electric power, withdrawal velocity and dimension of the ingot to obtain the design parameter for the future scale up. In the test, some amount of flux and tungsten powder, which are the oxidation agent for the decontamination and the tracer of identifying the solidification front, respectively, were added to the melt dome. The flux is an admixture of SiO$_2$, Al$_2$O$_3$, CaO and MgO. Another kind of admixed fluoride flux, CaF$_2$ and MgF$_2$, was also added. In the present test, radioactive nuclides were not added and surrogates of them, such as Co, Mn, Cs, Zn, Sr, Hf and Ce were added to investigate the partition of nuclides among ingot, slag and dust during casting either round ingot of 100mm in diameter or rectangular slab of 110mmx50mm in cross section. The surrogates play the tracer of nuclides. Measured amounts of tracers are homogeneously filled in the vacancy of the tubes before melting.

The melt is almost covered with the flux to enhance the reaction between melt and slag. Major parameters of the tracer addition test are the basicity of the oxide flux, kind of flux (addition of fluoride flux) and the shape (cross section) of the ingot. Basicity of the flux, which is the dimensionless number of mass of lime divided by that of silica, was changed from 0.3 to 1.8 during the test.

TESTING RESULTS

Melting Behavior
Generally, after the electric power reached some fixed value of the test, the primary melted in a few minutes. Continuously supplied fragments of metallic materials floats on the dome surface on account of surface tension and falls down into the narrow gap between the periphery of the dome and the cold wall, followed by melting by Joule heating. In the case of melting long shaped materials, such as tubings and pipings, vertical linear feeder is used. At the end of the cyclic supply by the linear feeder, the materials being put between the hook of the feeder are released onto the melt dome. However any trouble such as splashing of the melt and melting of the tip of the hook did not take place.

Molten metal dome elevated by the electromagnetic pinch force could be stabilized by selecting the suitable dome height and the addition of the flux. Infiltration of the molten metal into the narrow slit gap did not take place because of the precise manufacturing technology for the slit gap less than 0.2mm and selective infiltration of the slag into the gap.

Addition of the surplus amount of slag or poor control of the molten metal height gradually results in the narrower cross section of the top opening, on account of the accumulation of the molten slag on the cold part of the crucible, followed by solidification. Precise control concerning the supply rate of the flux and dome height of the metal is important for the stable operation of the MERC system.

Quality of the Ingot
Quality of the ingot plays an important role in the reuse of dismantled metals. This is not only due to the requirement from the manufacturing of materials but also from the decontamination of the cast ingot. It would be difficult to remove the slag from the ingot if the slag infiltrated into the crack gaps that were made during withdrawal. Selection of the optimal casting condition concerning dome height, withdrawal velocity and power input resulted in the sound
ingot of about 37kg. Surface and longitudinal section after etching along the withdrawal direction for the round ingots of 100mm and 140mm in diameter and cut in 300mm length are shown in Fig.2A and Fig.2B, respectively. Figure 3 shows those of rectangular slabs of 110mmx50mm and 170x50mm, respectively.

The surfaces are smooth and crack free, promising that the ingot surface will be free of adhered slag. Actually some of the slag was stripped off automatically by thermal shrinkage of the ingot during cooling down and some was removed by simply thrashing. Dark parts on Fig.2A and Fig.2B observed at the upper part of the ingot surface are the slag that could not be removed by the rotary brushing device for slag delamination. Slag adhered on the surface is observed for rectangular slabs in Fig.3, because we have no slag-delamination device, in the moment. Soundness of surface quality facilitates decontamination by the removal of slag concentrated in radioactive nuclides.

Figure3A and Fig.3B show the surface and the etched macro structure pattern on the longitudinal section of the rectangular slab. Supplied metallic materials are completely melted and none of the slag is included in the metal. As, the rotary slag-delamination device cannot be applied to the rectangular slabs, most of the surface is covered with the slag.

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Fig. 2. Surface and etched macro structure pattern of round ingots continuously cast by MERC. A: φ100mmx300mm, B: φ140mmx300mm.
Direction of the structure lines represents the trajectories of the removed heat flow. They are horizontal near the surface, bend to upward in the bulk and become vertical on the axis, except for the middle height of the ingot, to which starting and ending of the operation corresponds and the melting followed by solidification there is not steady. In the testing, length of the ingot is limited to 300mm. However, in the future commercial plant, ingot length can be prolonged and unsteady length of solidification can be minimized.

There was no macro segregation in horizontal as well as in longitudinal directions for the elements of Fe, Cr, Ni and Mn contained in the ingot. Cu concentration was the same as that contained in the materials before melting. This means that the crucible is harm free against any corrosion.

**Treatment Rate**

Relations between applied electric power and withdrawal velocity for the round ingots as well as the rectangular slabs are shown in Fig. 4 A and Fig. 4 B, respectively. Closed marks and open ones represent the testing condition that the melting is possible and impossible, respectively. With an increase in applied electric power, upper limit of withdrawal velocity where the melting can be maintained increases as shown by the dotted lines. Inclination of the dotted line is larger for the rectangular slabs rather than for the round ingots. The inclination seems to become large when the cross section of the ingot is small and side area of the ingot responsible for heat removal by the cold wall is large.

Withdrawal velocity in the present MERC apparatus is limited below 12mm/min. So, we couldn’t find the upper limit of withdrawal velocity that can be reached by MERC at the electric power of 150kW for the round ingot of 100mm in diameter and the rectangular slab of 110mmx50mm in cross sectional dimension. From Fig. 4 A and Fig. 4 B, we are able to deduce the treatment rate of stainless steel around the maximum out put of the electric power.
The results are shown in Table II. With the decrease in cross section of the ingot and side surface area adjacent to the cold wall, which is responsible for the heat removal from the ingot, treatment rate increases up to 0.35kg/kWh in the case of round ingot of 100mm in diameter.

![Graph A](image1)

**Fig. 4.** Relation between applied electric power and withdrawal velocity.
 A: round ingots, B: rectangular slabs

<table>
<thead>
<tr>
<th>Ingot dimension</th>
<th>Withdrawal velocity (mm/min)</th>
<th>Applied electric power (kW)</th>
<th>Treatment rate (kg/kWh)</th>
</tr>
</thead>
<tbody>
<tr>
<td>φ100mmx300mm</td>
<td>12</td>
<td>130</td>
<td>0.35</td>
</tr>
<tr>
<td>φ140mmx300mm</td>
<td>4</td>
<td>150</td>
<td>0.25</td>
</tr>
<tr>
<td>110 mm x50 mmx300mm</td>
<td>12</td>
<td>120</td>
<td>0.26</td>
</tr>
<tr>
<td>170 mm x50 mmx300mm</td>
<td>9</td>
<td>150</td>
<td>0.22</td>
</tr>
</tbody>
</table>

**Transfer of Tracer**

Elements transferred to the ingot are obtained by the mass of ingot identified from the solidification front using added tungsten powder multiplied by concentration of the elements evaluated by chemical analysis. In the same way, elements transferred to the slag are obtained by the mass of vitrified slag collected through the bottom opening of the airtight chamber and chemical analysis of it. It is difficult to capture all dust. Some of the dust could be captured by an off gas filter. However this is not the total amount. Hence, in the present analysis total amount of the added elements as tracer subtracted by those transferred to the ingot and the slag were identified those transferred to the dust.
Partitions of the simulated nuclides for cases from A through D among ingot, slag and dust are shown in Fig.5. Oxide flux was added in both cases A and B during casting of round ingot of 100mm in diameter, though its basicity is different. Fluoride flux was added in case C and rectangular slab of 110mmx50mm in cross section was cast with the addition of oxide flux in case D.

Fig. 5. Partition of simulated nuclides.
In the case of addition of oxide flux during the casting of round ingot, more than 90% of Co and 60% of Mn remained in the ingot. While, more than 85% of Sr, 80% of Hf and 80% of Ce were partitioned to the slag, more than 80% of Cs and 55% of Zn were partitioned to the dust. Effect of basicity on the partition was insignificant. Amount of Ce remained in the ingot was 1%-5%, showing the decontamination effect of U by the oxide slag. Partition of tracer elements among ingot and slag is responsible for the difference in oxidation energy.

In the cases of addition of fluoride flux, C, and casting of rectangular slab with the addition of oxide flux, D, small splashing of the melt took place during the release of tubings and pipings from the linear feeder followed by free fall onto the melt dome because the tracers with the low boiling temperature such as Cs and Zn were mixed with the molten metal. When the small splashing of the melt takes place, although the amount of elements partitioned to the dust becomes large, the amount partitioned to the ingot is almost the same as that in splashing free cases, A and B.

CONCLUSION

The MERC (Melting and Recycling of Metals by Cold Crucible) process was designed, manufactured and scaled up to 100-140mm in diameter for the reuse and decontamination of the radioactively contaminated metallic materials generated during the dismantling of nuclear facilities. Not only circular sectional crucibles but also rectangular slab sectional crucibles of 110mmx50mm and 170mmx50mm were developed. The maximum power of the high frequency generator is 150kW and the frequency is 25kHz. In the MERC, either continuously supplied fragments of stainless steel or pipings, which was the surrogates of contaminated metallic materials was successfully melted, followed by solidification, pulling down, cutting and transportation. The following results were obtained.

1) The ingot surface was smooth and crack free, facilitating the removal of radioactive elements concentrated in a slag stuck on the ingot surface. Solidification structure was regular and macro segregation was not observed. The ingot proved to be suited for the reuse.
2) Treatment rate of the MERC depends on the dimension and the shape of the ingot. It was increased up to 0.35kg/kWh in the continuous casting of round ingot of 100mm in diameter.
3) Amount of Ce partitioned to the ingot was 1%-5%, showing the decontamination effect of U by the slag.

REFERENCES