



XA04N0649

MELTING OF CONTAMINATED METALLIC WASTE

Ying-Sheng Lee, Sheng-Yuh Cheng, Hsien-Tzu Kung, and Li-Fu Lin

Institute of Nuclear Energy Research

P.O. Box 3-14

Lung-Tan, Taiwan, R.O.C.

Fax : 886-3-4711409

ABSTRACT

Approximately 100 tons of contaminated metallic wastes were produced each year due to maintenance for each TPC's nuclear power reactor and it was roughly estimated that there will be 10,000 tons of metallic scraps resulted from decommissioning of each reactor in the future.

One means of handling the contaminated metal is to melt it. Melting process owns not only volume reduction which saves the high cost of final disposal but also resource conservation and recycling benefits.

Melting contaminated copper and aluminum scraps in the laboratory scale have been conducted at INER. A total of 546 kg copper condenser tubes with a specific activity of about 2.7 Bq/g was melted in a vacuum induction melting facility. Three types of products, ingot, slag and dust were derived from the melting process, with average activities of 0.10 Bq/g, 2.33 Bq/g and 84.3 Bq/g respectively.

After the laboratory melting stage, a pilot plant with a 500 kg induction furnace is being designed to melt the increasingly produced contaminated metallic scraps from nuclear facilities and to investigate the behavior of different radionuclides during melting.

I. INTRODUCTION

A considerable amount of contaminated metallic waste is generated at nuclear power plant during operation maintenance. Much larger quantity will result from plant decommissioning, e.g. approximately 10,000 tons of metal scrap for a 1,000 MW PWR or BWR¹. Therefore, large volumes of storage site would be occupied by the metallic scrap; this is expensive. Utilization of the metallic scrap as valuable materials not only saves the nation's resources but also decreases the waste to be disposed of.

There are two ways for the reuse of contaminated

metallic waste. One is to reuse contaminated equipment as it is in nuclear facilities. The other is to produce metallic components such as waste containers, shielding blocks, etc. by melting and to reuse them in nuclear industry or other places². By recycling the material in nuclear facilities, the limits to free release are not important. However, unrestricted release is also possible, if the residual activity concentration can be proved to be below the authorized limits.

The possible decontamination effect of melting is important with a view to the metal recycle since it concentrates the contaminants in the slag/dust and reduces their level in the metal³⁻⁴. In addition, melting also offers several advantages with regard to waste disposal⁵, i.e., volume reduction attained by converting the metal to its most dense form, reduction of the surface exposed to corrosion, immobilization of surface contamination by incorporation into the base metal, and homogenization of the radioactive nuclides for easy and accurate activity measurement.

In this report the radioactive metal melting testing program currently conducted in the Institute of Nuclear Energy Research (INER) is described. This program is expected to bring a great deal of useful fundamental data in recycling metallic wastes.

II. CURRENT STATUS IN THE WORLD

The first report of melting contaminated metallic scraps was presented at the Luxembourg conference, the 1984 status report of the EC decommissioning project⁶. The progressive importance of melting as an alternative for decontamination and volume reduction of scraps was later realized by a number of countries. Among these, Germany, Sweden, France, USA and Japan have all built up the exclusive melting furnace and cutting techniques for metal sizing. In addition, reuse criteria in terms of activity limits are also established in several countries, e.g. Germany.

Germany started melting decontamination and recycling program at Karlsruhe Nuclear Research center (kfk) in 1981 and

Siempelkamp, a private company, using a 20 tons induction furnace with an off-gas system including cyclone, bag filter and HEPA filter, had melted 1500 tons steel scraps of activities less than 74 Bq/g. In 1990, a 3.2 tons capacity of induction furnace was built up. More than 3000 tons of steel and copper scraps have been melted and the recovered ingots were used to manufacture test weight, shielding plate, type A and type B casks, etc^{7,8}.

Studsвик in Sweden built up a 3 tons capacity of induction furnace in 1987⁹. The behavior of radionuclides migration upon melting was similar to that of Germany. There was only one person receiving 0.5 mSv of radiation dosage, and no body had internal contamination upon 450 tons of steel melting. Sweden also conducted the melting of 1500 tons copper condenser tubes and used the recovered copper to manufacture ship propellers².

In USA, Scientific Ecology Group Inc. (SEG), has a 20 tons, 7200 kilowatt, high-efficiency induction furnace. With this furnace, SEG has made shielding blocks out of slightly contaminated scraps for use in a high energy physics program.

Japan Atomic Energy Research (JAERI) has completed the construction of a 500 kg capacity induction furnace in 1990 and has been conducting basic melting and casting test. This is the part of the first stage of decommissioning material research in JAERI¹⁰.

III. MELT DECONTAMINATION AND RECYCLING IN TAIWAN

3.1 ROCAEC Reuse Technology Development

To cope with the increasingly produced metallic waste, the ROCAEC has completed a research program in 1992 titled "Feasibility study of reuse criteria for low-level contaminated metallic waste". The work scope included:

1. Evaluation of release criteria in European countries (Radiation Protection No.43 of the Commission of the European Communities).
2. Survey of the radiation monitoring techniques for compliance with release criteria.
3. Survey of the metallic scrap which can be recycled or reused, including the types, quantities, specific activities etc.
4. Evaluation of the possibilities for implementing the EC recommended reuse criteria in Taiwan.
5. Evaluation of decontamination of metals by melting.

The major conclusions from this research can be drawn as follows:

- (1) TPC's low level metallic wastes are mainly steels and

copper; copper could be melted by INER's induction melting facilities as a pilot research to establish melting decontamination related techniques.

- (2) Steel scraps could be melted in the domestic foundry industries, which should be licensed by the authorities. In order to reduce social as well as political impact, recycling should be restricted within nuclear facilities.

- (3) Recycling and reuse criteria should be built up as legal bases.

- (4) It's beneficial, based on financial analysis, to build up a large melting facility to handle low level metallic wastes.

3.2 Melting Practice in INER

3.2.1 Melting facility

Melting tests were performed in the fuel fabrication Laboratory of INER using a 250 KVA, 3000 Hz Vacuum Induction Furnace (VIM) which was originally designed for uranium melting.

The main parts of the furnace unit consist of a quartz tube surrounded by induction coils, a pushing rod device for bottom pouring, a graphite crucible, and a vacuum pumping system with filters (FIG.1). Scraps were charged into graphite crucible with a bottom pour orifice, below which was a graphite mold for receiving the melt. After scraps were completely melt down, which could be seen from the viewport, the graphite rod was then pushed down to perforate the bottom orifice of the crucible. The temperature of the melt was measured by an optical pyrometer. The maximum temperature which can be reached by the furnace was 1300°C.

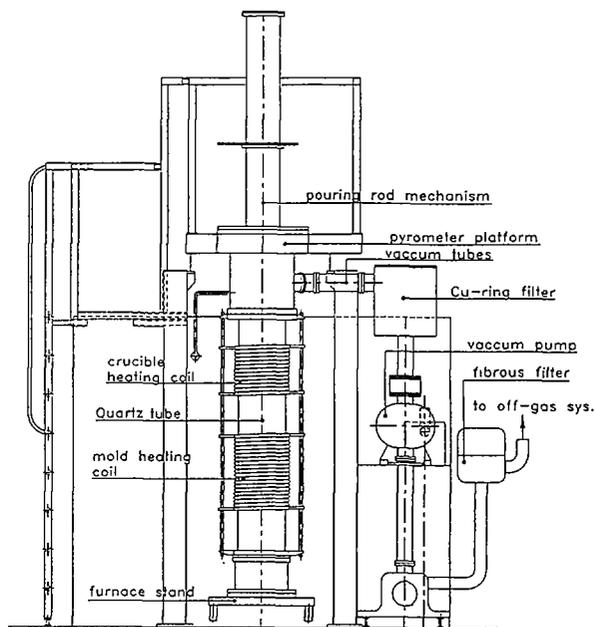


FIG.1. Schematic Diagram of the Furnace Assembly.

The vacuum of the furnace was usually maintained at 10^{-1} torr during melting. The filter device consists of a Cu-ring filter and a fibrous filter for collecting metal vapors and dusts. In several cases, it was shown that no radioactivity could be detected behind the fibrous filter.

Approximately 546 kg of copper condenser tubes from TPC and 200 kg of aluminum alloys from INER were melted with this VIM.

The specific activities of the materials before and after melting were measured by a multichannel analyzer, MCA (Canberra Series 90), with a high purity germanium detector. Eu-152 was used as the standard source.

3.2.2 Results of the melting

Generally speaking, the quantities of secondary wastes (slag and dust) depend on the scraps quality and impurities, also relate to the melting parameters of the melting facility, for example pumping speed, melting temperature.

(1) Melting of condenser tubes

Melting operation is shown in FIG.2. Two kinds of copper alloys were melted, one was aluminum-brass which contained about 22% of Zn and the other was cupronickel (copper-30% nickel). Table I shows the typical weight distribution after melting. For aluminum brass, since large quantity of Zn were vaporized and pumped out by the vacuum system to deposit on the internal surfaces

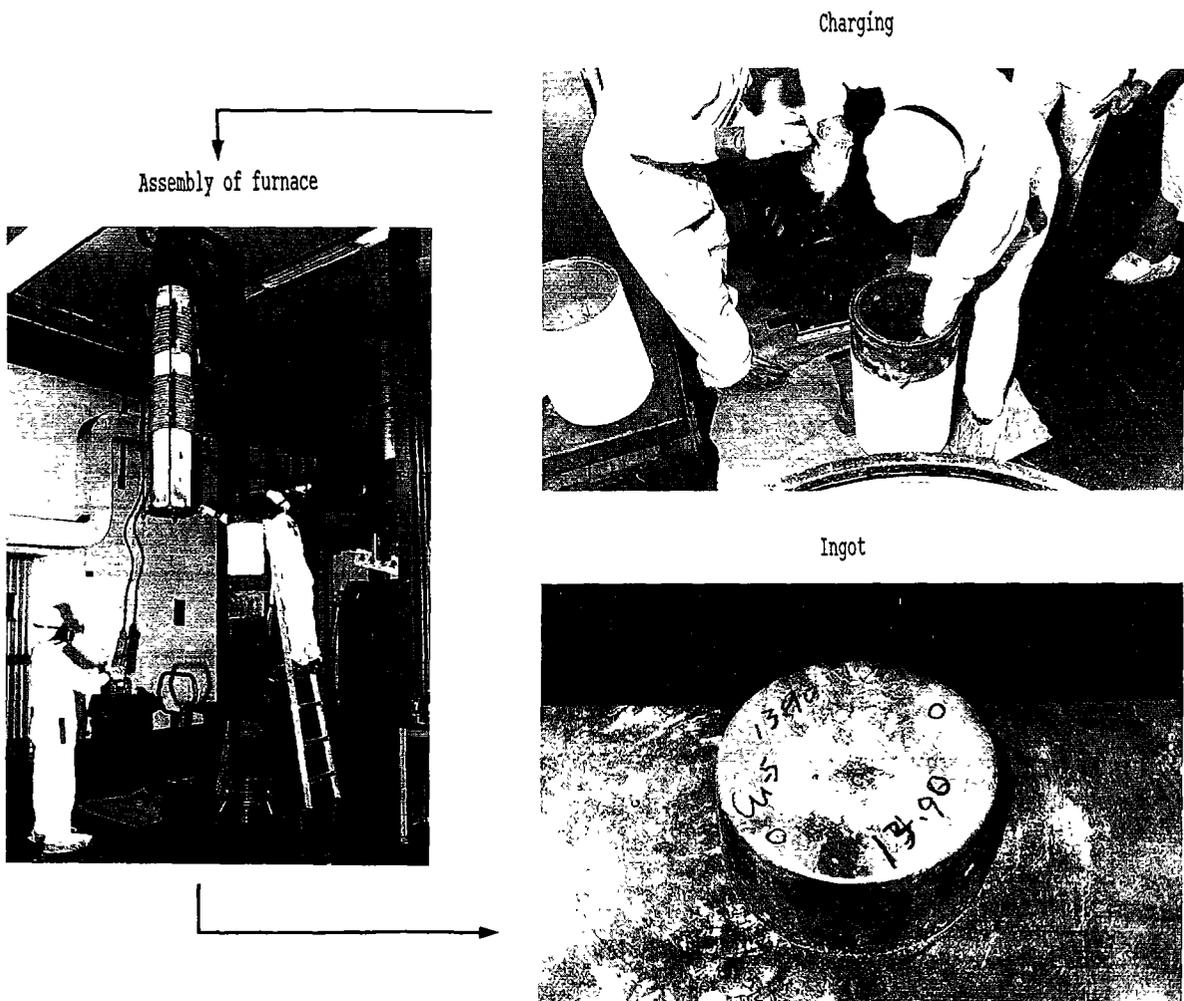


FIG.2. Melting of Condenser Tubes.

of the vacuum lines, which were collected as slags, the ingot recovery was merely 76.4 wt.% and slags amounted up to 22%; i.e. almost all of Zn was vaporized. Some dust and Zn vapor were also captured by the off-gas system connected to the vacuum lines; the amount was 1.6 wt.%.

For copper-nickel, there existed no elements with low boiling point, so that we have high recovery (98.6%) of ingot and apparently small quantity of slag (1.1%) and dust (0.3%) compared with aluminum-brass.

Table I Typical Weight Distribution after a Melting Run

Scraps	Charging	Products		
		Ingot	Secondary Wastes	
			Slag	Dust
Aluminium-brass	24.5 kg	18.37 kg	5.3 kg	0.38 kg
	100 %	76.4 %	22 %	1.6 %
Copper-Nickel	24.67 kg	24.32 kg	0.27 kg	0.08 kg
	100 %	98.6 %	1.1 %	0.3 %

The overall results for melting brass and Cu-Ni is shown in Table II. Before melting, Co-60 and Cs-137 were the dominant radionuclides with specific activities 2.58 and 0.18 Bq/g respectively. The average activity of the ingot was 0.1 Bq/g with Co-60 dominant and no measurable level of Cs-137. This is probably due to the low boiling point of Cs which evaporizes during melting process. There were about 78% of total activities enriched in the 2.6 wt.% of dusts, within which Co-60 and Cs-137 dominant, so that specific activity in the dust was increased by a factor of 30.

Table II Total Recovery and Activity Distribution of Melting for Condenser Tubes

	Charging	Products		
		Ingot	Secondary Wastes	
			Slag	Dust
Recovery (aluminium-brass and copper-nickel)	546 kg	472 kg	60 kg	14 kg
	100 %	86.4 %	11.0 %	2.6 %
Specific Activity	2.76 Bq/g	0.1 Bq/g	2.33 Bq/g	84.3 Bq/g

(2) Melting of aluminum scraps

There were many kinds of contaminated aluminum scraps from different nuclear systems, e.g. anodized aluminum and painted plates which increased the quantities of slags and dusts. The scraps were cut, packed into 55 gallon drums, and shipped to the melting facility. Table III indicates the weight percentage of ingot, slag and dust are 93.5%, 5.3% and 1.2%, respectively. Since these aluminum scraps contained little elements with low boiling point, slags and dusts mainly came from anodized oxide layer and paints. Table IV shows the specific activities of these ingots. It is hard to estimate the decontamination factor since these scraps came from different sources which had different activities of radionuclides and were mixed together in the 55 gallon drum; gross counting technique for the whole drum is not available in our laboratory.

However, It can be noted that Cs-137 is the dominant radionuclide left within the ingots, in contrast to the melting of steel and brass which showed most of Cs vaporized during the melting process.

Table III Weight Distribution after Melting for Aluminum Scraps (18 batches)

Scraps	Charging	Products		
		Ingot	Secondary Wastes	
			Slag	Dust
Aluminium alloy	194.8 kg	182.14 kg	10.36 kg	2.3 kg
	100 %	93.5 %	5.3 %	1.2 %

(3) Occupational exposure

During the melting experiments conducted from May 1992 to April 1993, only two persons received 0.12 mSv and 0.09 mSv of radiation exposures, respectively, among seven persons participated in the work. Whole body measurements didn't reveal any internal contamination in any person.

IV FUTURE PROSPECT

(1) A pilot plant with an 500 kg induction furnace is being designed to melt the increasingly produced contaminated scraps including ferrous and nonferrous metals. The furnace and its pouring system will be installed within a containment chamber which will be kept at a pressure 25 mmW.G. below atmosphere to prevent the leaking of radionuclides while melting and pouring are being done. Operation of the facility will be performed

by remote control to reduce the possibility of radiation exposure(FIG.3).

(2)Further researches about the radionuclides migration during melting will be studied quantitatively to extend the melting decontamination to deal with scraps of higher specific activity. Data on the material balance, radioactivity balance and environmental condition will be collected. The melting parameters are materials (steel, copper, aluminum), radioactivity, flux, melting temperature etc. The assessment items of interest include transport of elements from charging materials to ingot, slag, dust, etc., transport of radionuclides to ingot, slag, dust, etc., radionuclides distribution within an ingot and radiation exposure within the plant.

(3)Environmental evaluation on the reuse of the recovered ingots in restricted as well as unrestricted recycling are being performed. Hopefully acceptable activities criteria can be set up by the regulatory authorities for legal reuse in the near future.

Table IV Activities of Aluminium Ingots after Melting

Nuclides Batches	Co-60	Cs-137	Cs-134	Surface Dose rate
	(Bq/g)	(Bq/g)	(Bq/g)	(mR/h)
Al-I1	< 0.09	< 0.04	—	0.05~0.08
Al-I2	< 0.15	< 0.16	—	0.05~0.08
Al-I3	< 0.11	< 0.13	—	0.05~0.08
Al-I4	< 0.09	4.81	< 0.07	0.05~0.
Al-I5	< 0.27	3.28	< 0.15	0.05~0.08
Al-I6	< 0.40	9.07	< 0.25	0.1 ~0.22
Al-I7	< 0.23	5.88	< 0.15	1 ~2
Al-I8	< 0.45	10.32	< 0.	0.05~0.15
Al-I9	< 0.23	84.73	< 0.15	0.1 ~0.15
Al-I10	< 0.25	0.93	< 0.16	0.04~0.
Al-I11	< 0.30	2.2	< 0.22	0.03~0.05
Al-I12	< 0.41	5.07	< 0.27	0.1 ~0.25
Al-I13	< 0.32	1.49	< 0.23	0.1 ~0.2
Al-I14	< 0.22	0.75	< 0.14	0.02~0.03
Al-I15	< 0.24	3.92	< 0.15	0.05~0.08
Al-I16	< 0.25	9.44	< 0.17	1 ~2
Al-I17	< 0.21	9.32	< 0.19	0.15~0.25
Al-I18	< 0.32	26.38	< 0.25	0.1 ~0.18
AVE.	< MDA	35.44	< MDA	

※ MDA : Minimum detective activity

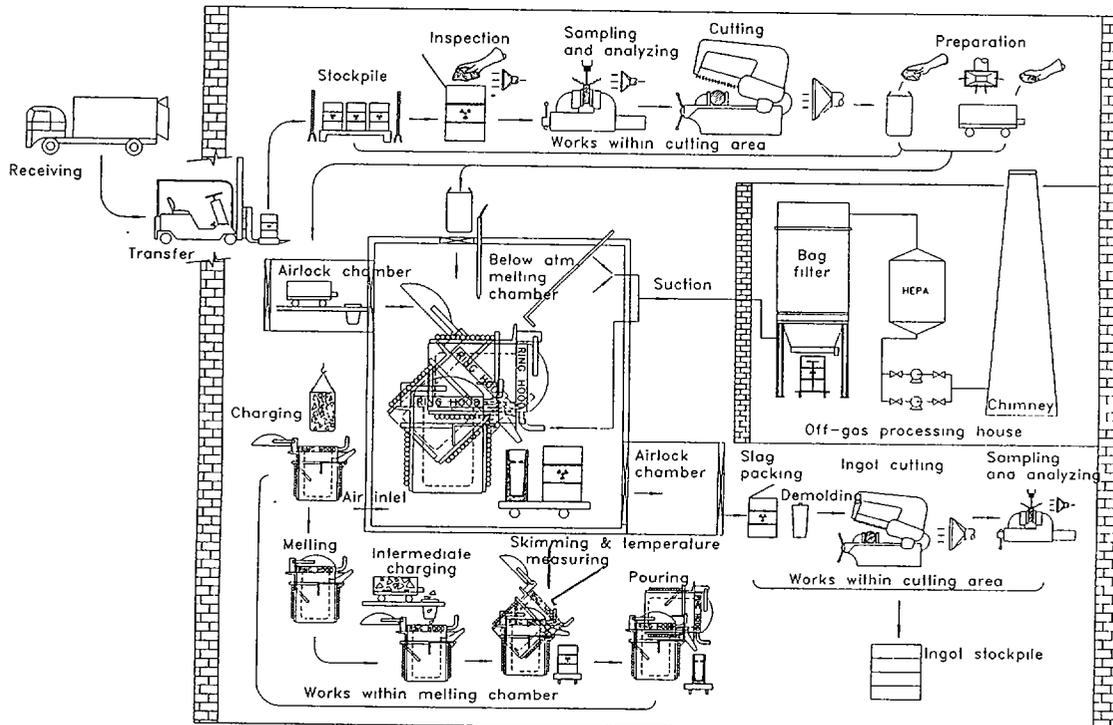


FIG.3. Schematic Flow Diagram of Melting and Casting for Metallic Wastes

V. REFERENCES

1. "COMMISSION OF THE EUROPEAN COMMUNITIES, Radiological Protection Criteria for the Recycling of Materials from the Dismantling of Nuclear Installations", COMMISSION OF THE EUROPEAN COMMUNITIES, Luxembourg, November 1988.
2. INTERNATIONAL ATOMIC ENERGY AGENCY, "Factors Relevant to the Recycling or Reuse of Components Arising from the Decommissioning and Refurbishment of Nuclear Facilities", Technical Reports Series No.293, IAEA, Vienna (1988).
3. T. UDA, H. IBA and H. TSUCHIGA, "Decontamination of Uranium-Contaminated Mild Steel by Melt Refining", Nucl. Tech., 73 (1986) 109.
4. M. SAPPOK, "Recycling of Metallic Materials from the Dismantling of Nuclear Plants", Kerntechnik 56 (1991) 376.
5. B. HUBER, "The European Community's Research and the Development Activities on the Management of Radioactive Waste from Decommissioning", Proc. Int'l Conf. on Radioactive Waste Management, pp.497-5-5, Seattle, 16-20 May 1983, IAEA.
6. "Decommissioning of Nuclear Power Plants", Proc. Conf. Luxembourg, 22-24 May 1984, Commission of the European Communities.
7. A. THOMA, "First Results of the Melting of Radioactive Waste in the EIRAM Plant", Proc. Int'l Conf. on Decommissioning of Nuclear Installations, pp. 494-496, Brussels, Belgium (1989).
8. M. SAPPOK "Results of Melting Large Quantities of Radioactive Steel Scrap", Nucl. Tech. 86 (1989) 188.
9. S. MENOR, G. HERNBORG AND L. ANDERSSON, "Melting of Low-Level Contaminated Steels", Proc. Int'l Conf. on Decommissioning of Nuclear Installations, pp.497-503, Brussels, Belgium (1989).
10. M. TANAKA AND N. NAKAMURA, "Research Program of Decommissioning Material Reuse in Jaeri", pp.213-217, Proc. of 1989 Joint Int'l Waste Management Conference, Kyoto, Japan (1989).