The Use of Induction Melting for the Treatment of Metal Radioactive Waste – 13088

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ABSTRACT

The aim of the work is to assess the efficacy of induction melting metal for recycling radioactive waste in order to reduce the volume of solid radioactive waste to be disposed of, and utilization of the metal.

INTRODUCTION

Currently, sites within the Russian Federation have accumulated several thousand tons (according to some estimates, as much as 600 thousand tons) and generate annually up to 10 thousand tons more of metallic radioactive waste. Stocks measuring dozens of thousands tons of metallic radioactive waste (radwaste) contaminated with naturally occurring radionuclides have also been accumulated on oil and gas industry sites.

Processing of metallic radwaste is a very important activity as the volume of the waste is large and it must be kept under strict controls inside specifically equipped safe storage facilities, which required considerable expenses to be borne. The alternative to storage – permanent disposal – is usually associated with extremely high commercial costs.

Recycling of spent nuclear fuel, for example that discharged from the LWR or VVER reactors on a radiochemical facility with the production capacity of 600 tons of uranium a year, generates approximately 190 tons high-level of metallic radwaste, of which about 170 tons of cut-up fuel claddings and 20 tons of fuel assembly end-pieces with the total volume approximately 200 m^3. The waste chiefly contains long-lived radionuclides (U, Pu, fission products), primarily, in surface layers of fuel claddings, and activation products (Fe, Co, Ni) in the end-pieces.

The level of contamination of the fuel cladding material depends primarily upon the level of fuel burn-up during in-core operation and on the composition of the material itself. For instance, contamination of fuel claddings in fast reactor fuel assemblies is 30-130 times that of thermal reactor assemblies.

As during the final stage of nuclear sites' lifecycle – decommissioning – yet more amounts of waste are generated, the total quantity of metallic radwaste would increase even further. For example, shut-down of a single NPP unit of 1000 MW electrical capacity alone would generate 15 to 42 thousand tons of metallic radwaste. The zone surrounding the Chernobyl NPP site after the accident accommodates estimated 12 thousand tons of stainless steel and 63 thousand tons of carbon steel. Therefore, the need to perform a management of all this metallic radioactive waste becomes ever more imperative.

METHODS OF METALLIC RADWASTE MANAGEMENT

The resolution to the metallic radioactive waste management problem depends on their extent of contamination with radionuclides, chemical composition and size.

SECONDARY WASTE GENERATION BY METALLIC RADWASTE MELTING

Melting method	***Secondary waste types and amounts, kg per tonne of metallic radwaste***				
Melting method	***Slag***	***Dust***	***Replaced lining***	***Scale***	***Total***
Electric-arc	36,3	11,4	2	0,9	50,6
Air-induction	18,2	7	6,4	0,9	32,5
Vacuum-induction	2	0,3	6,4	0,9	8

Depending on contamination, several options of metallic radwaste management may be used:
– unrestricted re-use of metal purified to the maximum possible extent;
– restricted use of decontaminated metal, for example, for nuclear power applications;
– controlled storage and final isolation in geological repositories before or after processing.

The high costs associated with both storage and putting into a repository demand that the volume of the waste has to be reduced. A reliable method for metallic radwaste compaction, which is in use, for example, on the UP-3 facility in France, is compression. However, compressed waste is not a matrix that can reliably secure radionuclides and suitable for final isolation.

Another method of reducing costs for storage of metallic radwaste is decontamination and melting. If that can provide a sufficient extent of purification, controlled re-use of melted metallic radwaste will become possible, which is an advantage for the environmental and the economic point of view.

Melting of metallic radwaste can produce a positive effect in a number of areas, such as:
– reduction of waste volume, with associated reduction of required storage and repository capacities;
– reduction of radioactive contamination hazard to the environment, due to the fixation of radionuclides inside a metallic matrix;
– with slag melting – refinement (purification) of contaminated metal, with concentration of the majority of radionuclides in slag, and, as a consequence, down-grading the metallic radwaste from the high-level category to intermediate or low-level, with simplified and cheaper storage.

MELTING TECHNOLOGIES

Currently there are several proposed and evaluated technologies for metallic radwaste melting. All research performed in this field focused on factors such as type of processed radwaste, extent of contamination and origin of radioactivity, as they are the decisive circumstances for development of an efficient and safe technology and building the equipment required to effectuate the melting process.

Performed research melts of metallic radwaste (of both carbon and stainless steels), despite some differences in their results, have demonstrated the following typical distribution of radionuclides in the products of melting:

– Mn-54 – approximately 90% in ingot, approximately 10% in slag;
– Co-60 – 100% in ingot;
– Sr-90 – 100% in slag;
– Cs-137 – 50% in slag, 50% in exhaust gases;
– α-emitters – 98% in slag.

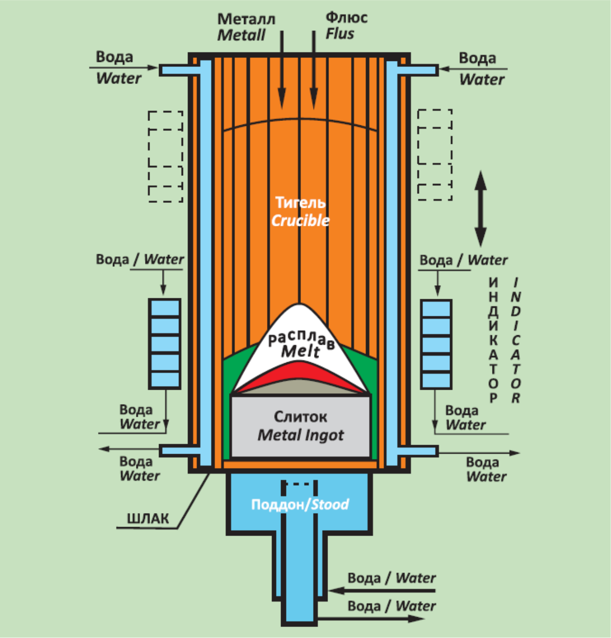

Fig. 1. Basic diagram of the "cold crucible" induction-slag melting process

The melting technologies provide for utilisation of fluxes that interact electrochemically and thermochemically with the molten metal and trap radionuclides. Contaminants that readily form oxides can be removed from the melt to the maximum extent possible and conjoin with slag, while radionuclides that are chemically homogeneous with the elements that make up steel stay in the melt practically in their entire original quantities. If the flux is correctly chosen, the output slag will be stable and small-volume "concentrators" of radionuclides.

During the 1980s, there were numerous studies (especially in Japan) performed to investigate whether electroslag melting could be used to process metallic waste. The studies produced sufficiently positive results, as it was demonstrated that many mechanisms of decontamination could be activated during melting. At the end of the process, the slag containing radionuclides would be removed in solid form, making its management far easier and safer than that of liquid radioactive waste. In 1993, effectiveness of the metallic radwaste treatment process using electroslag melting was demonstrated by studies performed by Oregon Institute of Science and Technology (OGIST), USA.

Some research melts were also performed using the electric arc method (EAM) in order to determine whether it was practically feasible to process metallic radwaste in industrial electric-arc ovens. EAM is the most common method of steel-making. This kind of melting is quite economic, as the charge of metallic radwaste and surface area for slag formation are both sufficiently large (open diameter up to 90-95 cm, capacity up to 350 tons). At the same time, the absence of a water-cooled induction coil ensures that the process is safer.

CIMI in France has conducted research to determine the distribution of radionuclides and assess potential contamination of the environment during electric-arc melting of carbon steel heat exchangers from the nuclear plant at Chinon.

Research melts have confirmed that it is indeed both possible and safe to process metallic radwaste by melting in industrial-size electric-arc ovens.

The disadvantages of the EAM process are as follow:

– generation of vast amounts of fine particles coming into the gas treatment system along with gases;
– significant metal losses to slag;
– need for controlled slag cooling in order to maintain its physical integrity;
– generation of large amounts of secondary waste (heat-proof oven lining, ladles, moulds, etc.).

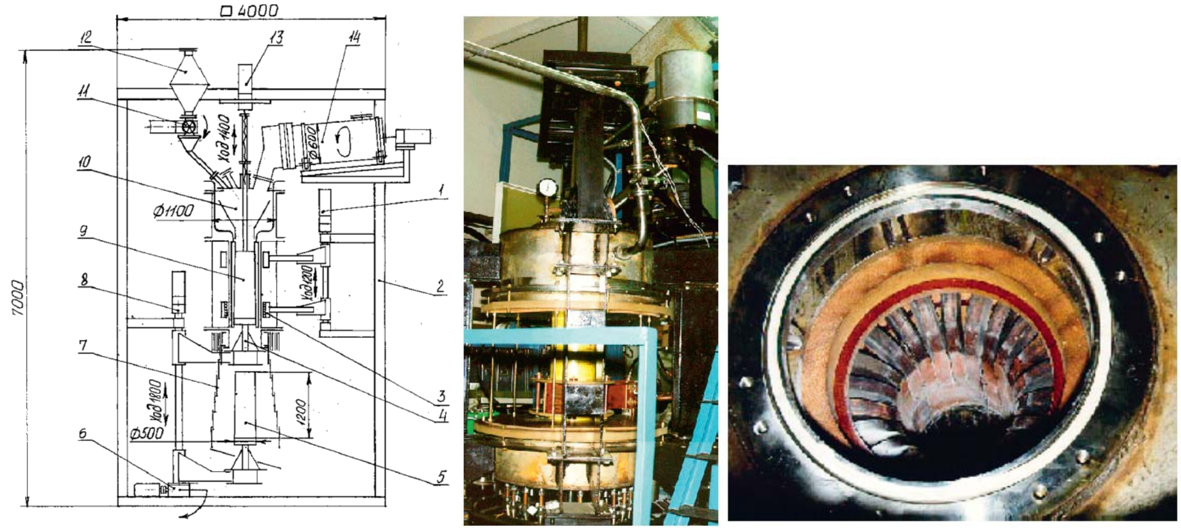

Fig. 2. Pilot "cold crucible" induction-slag smelter at Bochvar VNIINM:
1 – inductor drive; 2 – frame; 3 – inductor; 4 – "cold" tray; 5 – metal ingot; 6 – turntable drive; 7 – protective shroud; 8 – "cold" tray drive;
9 – crucible; 10 – smelter unit cover; 11 – flux feeder; 12 – flux batcher; 13 – pusher drive; 14 – melting metal feeder-batcher with drive

Using induction ovens can reduce generation of fine particles and provide for higher-intensity mixing (thanks to dynamic contact between metal phase and slag), ensuring uniform temperature distribution and homogeneous composition of the melt. Induction melting also provides for fast heat-up, high melting rate and simpler control of exhaust gases.

Studies of induction melting of metallic radwaste in air and inert environments have been performed in Germany, United States, Japan and other countries. It was demonstrated that induction melting in an air atmosphere, in addition to problems associated with fine particles carry-over, also necessitates creation of a complex gas treatment system, producing additional radioactive waste to be disposed of. Therefore, for development of a strategy for refinement melting in an air-induction oven, provisions must be made, just like in the case of EAM, for a facility to trap any radionuclides borne by dust and smoke – a sophisticated and expensive task. In addition to that, when stainless steel is recycled in an air atmosphere, the alloying component (Cr and Ni) are partly lost.

Vacuum or inert atmosphere prevent oxidation of the melt, significantly reducing production of secondary waste during melting. In addition, this technology can provide for forced entrapment of volatile radionuclides and treatment of small exhaust gas quantities to ensure that any radioactive components are caught, which influences the overall layout of the smelter and its cost.

Radiologically, vacuum induction melting appears to offer the best strategy for removal of volatile radionuclides. But, as it is slag-free, so far there has been no mechanism developed for removal of non-volatile radioactive contamination, with the exception of flotation and agglomeration methods.

Metallic waste *Flux* Slag Ingot

Fig. 3. Input and output products of the "cold crucible" induction-slag melting process

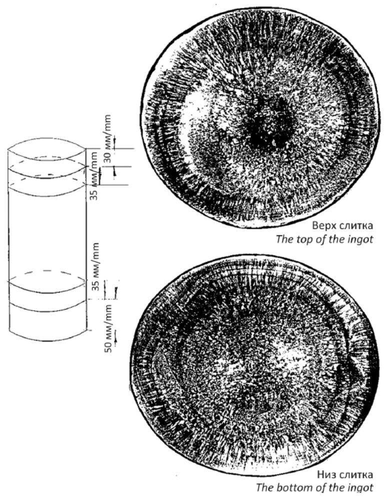

Fig. 4. Steel ingot cross-section macrostructure

In order to address these issues, scientific organisations and industrial companies in Russia, France and Japan are performing research and development work to devise methods for melting of metallic radioactive waste with refining fluxes in induction ovens with "cold crucible" smelters ("cold crucible" induction-slag waste melting technology). As known from processing of low and high-level metallic waste in experimental and industrial-scale "cold crucible" induction-slag smelters, the waste volume reduction factor is 5-6, Cs and Sr removal factor is up to 98%, and a-emitters removal factor yet even higher.

INDUCTION-SLAG MELTING IN A "COLD CRUCIBLE"

A.A. Bochvar High-technology Research Institute for Inorganic Materials (Bochvar VNIINM) has developed the technology and pilot installation for induction-slag melting of any kind of metallic waste of all levels of radioactivity (without prior or concurrent decontamination by liquids) in 'cold crucible' induction ovens.

The "cold crucible" induction-slag melting process (Fig. 1) provides for melting metallic radwaste fragmented into 300-500 mm pieces in a "cold crucible" in the presence of up to 5 weight % of oxyfluoride flux, formation (shaping) of a monolithic decontaminated metal ingot and its recovery from the crucible, vitrification processing of the resulting slag containing the bulk of long-lived radionuclides (Cs, Sr, Pu).

Bochvar VNIINM has produced and operates a pilot "cold crucible" induction-slag smelter 130 mm in diameter and 500 mm tall, capable of putting out ingots up to 50 kg of weight (Fig. 2). An ingot of cleaned metal comes out of the bottom of the oven when the delivery table is lowered.

The smelter has been used to fine-tune the technological and electrical melting modes of fragmented metallic waste, shape the decontaminated metal ingots, and improve the remote control and monitoring facilities. Studies were also performed of the properties of ingots made.

Figure 3 illustrates the input and output products of the "cold crucible" induction-slag melting process.

DISTRIBUTION OF RADIOACTIVITY IN METALLIC RADWASTE MELTING PRODUCTS (% OF WEIGHT)

Radionuclide	***Ingot***	***Slag***	***Gases***
Cs-137	0,3	39,5	60,2
(Ru+Rh)-106	95,0	4,5	0,5
(Sr+Y)-90	3,5	90,5	6,0
Co-60	99,97	0,02	0,01
Pu -239	0,1	99,5	0,4

As a result of test melts performed by Bochvar VNIINM, monolithic ingots were produced with a dense, gas pores-free microstructure (Fig. 4). It was noted that the ingot production process was most stable in an argon atmosphere with the furnace-charge feeding rate of 50 kg/hour.

It was demonstrated that the process can successfully purge the metal of the main radionuclides (Pu, U, Cs, Sr, Ru), reducing their concentrations by two or three orders of magnitude.

Therefore, from the viewpoint of metallic radwaste processing, the "cold crucible" melting method offers the following advantages:

– maximum reduction of volume and effective surface area;
– production of ingots suitable for disposal or metal re-use;
– costs insignificant compared to those associated with metallic radwaste storage;
– insignificant costs associated with metals preparation;
– no generation of secondary waste in the form of high-melting metal crucibles and moulds;
– possibility of deploying a compact set of remotely-controlled equipment in an isolated chamber (cell).

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CONCLUSION

Therefore, modern approaches to the metallic radwaste management problem provide for the following.

Major efforts associated with creation and improvement of melting facilities and fine-tuning of their technological parameters should focus on reduction of the volume of waste (slag, dust, smoke) produced during metallic radwaste conditioning that require additional processing and disposal.

The preferred choice to ensure observance of this requirement is the induction-heating melting technology, which is also easy to implement, offers a long design life and simplicity of equipment replacement. There is no need for interim vessels during metal casting, which further reduces the amount of secondary waste.

Unification is needed to make sure a single oven can be used for melting all types of metals contaminated with radionuclides as this approach is most economic. This can be achieved by using the vacuum-induction melting technology based on "hot" and "cold" crucibles.

Exposure to radiation during melting of metallic radwaste can be reduced by optimisation of the smelters. The process needs to be made as mechanised as possible, with further improvements to the loading system and smelter monitoring instruments.

Better decontamination of metal with increased output suitable for re-use is achieved by optimal choice of refining fluxes. Introduction of the fluxes in itself improves the extent to which the mix can be purified.

Practically all of the features described above can be achieved through the application of the 'cold crucible' induction oven technology.