WAYS OF RESOLVING ACTIVE METAL WASTE PROCESSING PROBLEMS

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ABSTRACT

The operation and, particularly, the decommissioning of NPPs and radiochemical plants result in substantial arisings of radioactive metal waste (RAMW) having different activity levels (from 5×10-4 to ≈ 40 Ci/kg).

The long-term storage of RAMW in specially designed storage facilities is cost ineffective (the cost of 1 m^3 low activity level metal waste storage is more than 600 $ US / year at the prices of 1998). Those expenses can be reduced by RAMW decontamination and melting.

The paper reviews the specific features of the technology and equipment used to melt RAMW in electric arc and induction furnaces with ceramic or "cold" crucibles. The experimentally determined and calculated data are given on the level to which RAMW is decontaminated from the main radionuclides as well as on the distribution of the latter in the products of melting (ingot, slag, gaseous phase).

Special attention is focused on the process and the facility for the induction-slag melting of RAMW in furnaces equipped with "cold" crucibles.

The work is described that is under way at SSC RF VNIINM to master the technology of melting simulated high activity level Zr-alloy and stainless steel waste.

INTRODUCTION

All the countries that adhere to the closed fuel cycle in the nuclear power developments are engaged in processing spent nuclear fuel and treating resultant radioactive waste (RAW) [1]. The problems of conditioning for storage or disposal of low and intermediate activity liquid and combustible RAW have been resolved on the industrial level. The problem of active metal waste conditioning for storage and disposal is resolved depending on the degree of contamination with radionuclides, chemical composition and amounts [2].

The decommissioning of 1 NPP unit of 1000 MW electric power results in 15 to 42 thousand tons of RAMW. Some 1.5% of the total mass of steels used in a reactor have ≈ 40 Ci/kg and contain 99.8% of the total activity.

The radioactive contamination of RAMW from nuclear reactors and process equipment of reprocessing plants is caused by both the activated products of steel components (^{60}Co, ^{54}Mn,

^{51}Cr, ^{65}Zn, ^{55}Fe, ^{59}Fe, ^{63}Ni) and the products of nuclear fuel fission (^{134}Cs, ^{137}Cs, ^{90}Sr, ^{106}Ru, ^{144}Ce) [3].

The activity of cruds at the surfaces of the primary circuit components in nuclear power reactors can reach up to 10^{-1} Ci/m^2. The specific activity of other reactor components is much lower (10^{-8}÷10^{-6} Ci/kg).

As a rule, the main contribution to the γ-radiation dose rate is made by ^{60}Co, but radioactive contaminants may contain up to 70% ^{137}Cs and ^{90}Sr (of the overall quantity of fission products).

The reprocessing of LWR spent nuclear fuel at a radiochemical plant of the 600 t U/year will result in ~ 190 t high activity level metal waste of which chopped fuel rod claddings and end pieces of fuel assemblies constitute ~ 170 t and ~ 20 t, respectively, with the overall volume of ~ 200 m3. The waste contains long-lived radionuclides (Cs, Sr, Ru, Pu) basically at the surface layers of the claddings and activation products (Fe, Co, Ni) in the FA end pieces [4]. Currently the waste is stored in special ground storage facilities.

The long-term storage of RAMW in specially designed storage facilities is cost ineffective (the cost of 1 m3 low activity level metal waste storage is more than 600 $ US/year at the prices of 1998). Those expenses can be reduced by RAMW decontamination and melting.

The melting of RAMW can be favourable in several aspects:

- a factor of 4-6 reduction of waste and, correspondingly, of storage and burial facility volumes;
- conversion of some RAMW to intermediate- and low-level waste with the resultant simplification and lower cost of the storage;
- simplified measurement of specific and total activity of RAMW (particularly, for large sizes and volumes of waste when only random control is feasible);
- the possible radioactive contamination of the environment is almost fully eliminated due to the uniform distribution and reliable immobilization of radionuclides in the metal matrix [5].

EXPERIMENTAL

To melt radioactive steel electric arc and induction facilities were basically used in different countries [6-8].

In Germany, e.g., fragmented RAMW was subjected to melting (packs of NPP intermediate steam superheater tubes); the specific β- and γ-activities being up to 200 Bq/g and the fissile element (^{235}U, ^{238}U, ^{239}Pu, ^{241}Pu) contents reaching < 100 Bq/g.

The RAMW melting facilities are installed in boxes equipped with a special gas clean-up system.

Fragmented RAMW to be melted are supplied in metallic drums. Using a tilter the waste is first discharged into a receiving hopper of a loading device and then into a crucible.

The waste fragments to be melted have to be not more than 500 mm in size, the mass of a single lump is not more than 150 kg.

For the decontamination of a metal by induction slag melting oxide and oxide-fluoride fluxes are introduced in quantities of 5-10% of the RAMW mass.

The fluxes are used in a great variety of compositions. The best results are attained with calcium alumosilicate and boron silicate fluxes (with additives of CaF_2, NiO, Fe_2O_3). However, the optimal composition of the flux has not yet been established.

The numerous experiments with actual low activity-level waste established that the process of the crucible furnace melting efficiently purifies the metal from ^{90}Sr, ^{144}Ce and other rare-earth elements, ^{65}Zn, ^{137}Cs, ^{238}U, ^{239}Pu as well as from other TRU (see table 1).

Table 1. Distribution of Radionuclides in Products Resulting from Low Activity Steel Melting.

Radionuclide	Radioactivity, %		
	Ingot	Slag	Dust and gaseous phase
^{54}Mn	95	5	–
^{55}Fe	~100	Traces	–
^{60}Co	90	10	Traces
^{63}Ni	90	10	–
^{65}Zn	Traces	10	90
^{90}(Sr+Y)	~5	~95	Traces
^{134,137}Cs	Traces	~50	~50
^{144}Ce	~50	~50	–
^{154}Eu	5	95	–
^{235,238}U	Traces	~98	Traces
^{241}Pu	Traces	~98	Traces

The residual metal radioactivity is defined by the availability of ^{60}Co that almost fully remains in the ingot.

The activity of the resultant slags is basically dictated by ^{90}Sr, ^{137}Cs, ^{144}Ce; the specific γ-activity of slags being a factor of 10-15 higher than that of the initial RAMW.

The specific activity of the ceramic lining of induction furnaces was ~ 50% of the specific activity of slags. Taking into account the limited service life of ceramic crucibles (30-50 melting runs) the furnace lining is intricate active waste that requires special conditioning to be disposed of.

The rather high temperature of the RAMW melting process (~1500°C) promotes the substantial evaporation of volatile radionuclides (^{3}H, Zn, Cs) from the surface of the liquid metal and slag to be trapped by the gas clean-up system.

In practice it has been demonstrated that the application of electrical arc and air-induction installations gives rise to serious problems in collecting and processing dust and slag; the gas clean-up system is cumbersome and expensive (20-25 million US dollar prices of 1995). However, induction vacuum furnaces also suffer from substantial disadvantages, i.e., the low service-life (≤ 50 melting runs) of melting crucibles and moulds resulting in additional non-processible secondary waste.

Their alternative is an induction furnace with a "cold" crucible (IMCC).

It is evident from the experience of Russia, France and Japan gained in melting low- and high activity level metal waste using IMCC rigs and commercial facilities that the waste volume reduction factor is 5-6, the factor of metal decontamination from Cs and Sr is 98% while that for α-emitting nuclides is >98%.

The SSC RF A.A. Bochvar VNIINM[i] together with other research institutions has designed the technology and the demonstration facility for induction-slag melting metal waste independent of its activity level (without aqueous pre-decontamination applied) using induction furnaces with "cold" crucibles.

This process (its abbreviated form is "ISMW-CC") consists in melting down waste lumps 300-500 mm in size in a "cold" crucible with < 5% oxide-fluoride flux, monolithic decontaminated ingot production and ejection out of the crucible, cementation or vitrification of the resultant slag containing the main part of long-lived radionuclides (Cs, Sr, Pu).

The similar process of RAMW melting that is under development in France involves the melting of cut-off fuel rod claddings in a furnace having a cold crucible and the pulling of the resultant ingot out of the crucible. The fuel essembly (FA) end pieces are processed in an induction steel-making tilting furnace having a ceramic crucible and die cast ingots [9].

The SSC RF VNIINM has adopted the concept that is currently under way to jointly melt down cuts of fuel rod claddings and FA end-pieces in a single induction furnace equipped with a "cold" crucible. Another distinction of the process that is under development in RF lies in the way an ingot is formed, namely, not by pulling out but by building it up. Accordingly, the design and arrangement of the equipment were changed.

In essence, the "ISMW-CC" process (fig.1) as applied to RAMW processing consists in the following. The chopped claddings dried in the hopper-collector are supplied to a batchmeter from which they are charged in portions into the "cold" crucible.

The melting of RAMW is implemented together with salt fluxes that are in portions charged together with the chopped claddings in the quantity of 3-5 % of the RAMW mass and serve to decontaminate the metal from radionuclides and to insulate thermally the molten metal pool and eliminate its intensive cooling. The main decontamination is achieved due to the chemical interaction between the flux components and fission product oxides available in the surface layers of the fuel rod claddings. Radionuclides transfer to a slag that is accumulated at the outer surface of the metal ingot.

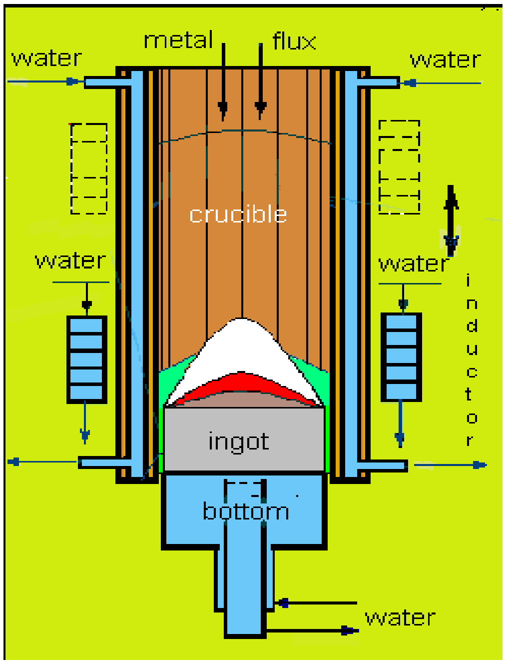


Fig. 1 Schematic of ISMW-CC process

The resultant slag containing the main mass of long-lived radionuclides (U, Pu, Sr, Cs, Ru) is separated from the ingot and additionally processed to be stored or disposed of [10].

The RAMW decontamination by induction-slag melting is accomplished via the re-distribution of radionuclides between the metal and the salt flux. The metal decontamination factor is the higher the higher is the chemical reactivity of the molten salt in relation to radionuclides contained by the fuel rod claddings. Two salt compositions were used as fluxes:

- for stainless steel [CaF_2(22,5%)-MgF_2(6,5%)-CaO(27,5%)-SiO_2(20,0%)-Al_2O_3(6,0%)-Fe_2O_3(9,0%)-B_2O_3(6,0)%)-Na_2O(2,5%)];
- for Zr-alloys [CaF_2(60,0%)-MgF_2(20,0%)-CaO(13,5%)-SiO_2(6,5)].

The technological parameters of the "ISMW-CC" process were tried in the course of testing the "ISMW-CC-2" demonstration facility (fig.2).

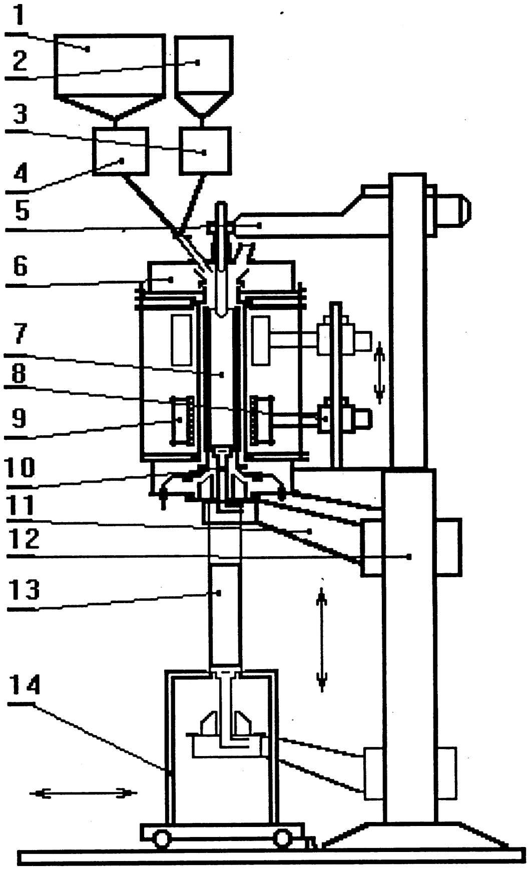
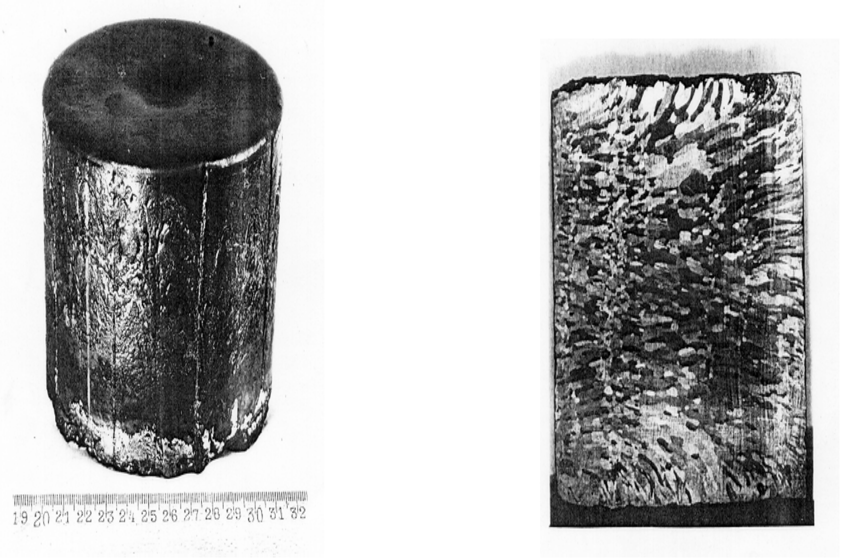


Fig. 2. Schematic of demonstration facility
1. Bin for fuel rod cladding simulators. 2. Bin for flux. 3. Flux feeder.4. Simulator feeder. 5. Pusher drive. 6. Lid of melting unit. 7. Crucible. 8. Inductor travel drive. 9. Inductor. 10. "Cold" bottom plate. 11. "Cold" bottom plate drive. 12. Frame post. 13. Metal ingot. 14. Transport truck.

The simulators of chopped stainless steel and Zr-alloy claddings 15-30 mm in size and cylindrically shaped ingots were used as initial materials.

The experimental melting runs resulted in monolithic ingots 120-280 mm high (figs. 3a and 3b) having a dense, gas void - free microstructure. It is revealed that the process of ingot building-up is most stable in argon at the rate of lump charging of ~ 50 kg/h.



a b

Fig. 3. Appearance (a) and microstructure (b) of metal waste ingot

Based on the experimental and design data the distribution of the main radionuclides was assessed in the products resulting from induction furnace with "cold" crucible melting Zr alloys and stainless steel (table 2).

Table 2. Activity Distribution in Products of High Level RAMW Melting (mass %)

Nuclide	Ingot	Slag	Gaseous phase
^{137}Cs	0.3	39.5	60.2
^{106}(Ru+Rh)	95.0	4.5	0.5
^{90}(Sr+Y)	3.5	90.5	6.0
^{60}Co	99.97	0.02	0.01
^{239}Pu	0.1	99.5	0.4

The investigations to study the "ISMW-CC" process and the tests of the pilot equipment corroborated the correctness of the technological and design solutions.

The basic advantages of the "ISMW-CC" facility compared to the electric arc and induction furnaces with ceramic crucibles are:

- its compactness and the low material intensity of the equipment; its long service life (up to ten years);
- the compact and simple gas-clean-up system (the process is carried on in a small furnace volume filled with an inert gas); low amounts of the flux (3-5% of the metal mass) and the resultant slag; no dies or other casting equipment;
- the high quality of the metal produced; the feasibility of remote control and management of the melting and equipment disassembly processes;
- the possible cementation and vitrification of the resultant slag in IMCC furnaces.

Based on the experience gained by SSC RF VNIINM a semicommercial facility is under development that will be used for melting various metal waste arising at NPP and radiochemical plants (table 3 and fig. 4)

Table 3. Basic Technical Characteristics of Semi-Commercial Facility

Parameters	Value
Throughput	up to 3000 kg/d
Type of heater	Induction
Power	up to 1000 kW
Current frequency	up to 2400 Hz
Molten pool temperature	up to 2000 °C
Type of melter	"cold"
Crucible diameter	up to 500 mm
Crucible height	up to 1500 mm

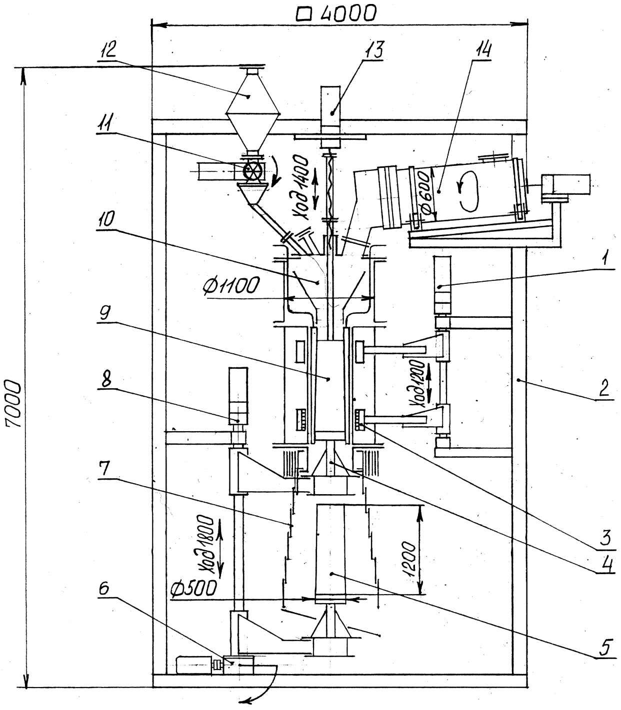


Fig.4. Schematic of semi-commercial "ISMW-CC"

1.Inductor drive. 2. Frame. 3. Inductor. 4. "Cold" bottom plate. 5. Metal ingot. 6 Swivel table drive. 7. Protective jacket. 8. "Cold" bottom plate drive. 9. Crucible. 10. Lid of melting unit. 11. Flux feeder. 12. Bin for flux. 13. Pusher drive. 14. Bin-feeder for metal to be melted and drive.

CONCLUSION

The acquired results of the combined research and experimental-design work allow the technology and equipment based on the induction melter with the "cold" crucible to be recommended for the extensive application in the atomic industry, specifically, for conditioning active metal waste to be stored or disposed of.

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FOOTNOTES

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