Fractionation According to Radioactivity during Remelting of Metallic Radioactive Waste*

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Abstract

A new process is described for the production of metallic shot and its fractionation according to residual activity during remelting of metallic radioactive waste (MRW). The process ensures reliable control of remelted and deactivated metal according to gamma-emitting radionuclides.

INTRODUCTION

Remelting of activated or surface-contaminated (with radionuclides) metalware and equipment waste formed at nuclear fuel cycle enterprises during their operation or after decommissioning has recently become a new trend in radioactive waste utilization. Using this technique for contaminated equipment of austenitic steels makes it possible to recycle to industry deficient metal with 17–19 % chrome and 9–11 % nickel contents.

Basic radionuclide contaminants in metallic radioactive wastes (MRW) are Co-60, Sr-90, and Cs-137 oxides, generally located as films on the surface of MRW. The stated radionuclides behave differently during remelting:

– Sr-90 oxides partially and uniformly dissolve in the metal melt and pass into scoria;

– Cs-137 oxide completely sublimates and is trapped by the gas purification system;

– 97 % of Co-60 oxide, whose oxygen affinity is close to or lower than that of the remelted metal, remains in the metal.

Hence residual radioactivity of remelted metal is due mainly to the presence of Co-60. This was demonstrated by studies on remelting of radioactive pipe packages of an intermediate superheater in an induction furnace performed at Karistein atomic power station (Germany) [1].

The remelted metal is intended for unrestricted industrial use; for restricted use, *e. g.*, as a construction material for Ministry of Atomic Energy (Minatom) equipment; or for ageing up the point of natural decay of radionuclides in the metal; this depends on the specific activity of the metal.

Permissible specific activities of radionuclides in metal for unrestricted use are fixed by the OSPORB-99 (Basic Sanitary Regulations

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TABLE 1

Result of remelting of low-level metallic radioactive waste

Isotope	Half-life period, years	Content in MRW		Decontamination factor	Content in the metal, Bq/kg	Permissible specific activity, Bq/kg
		%	Bq/kg			
Sr-90	29.1	18.9	832.5	20 000	0	10 000
Cs-137	30.2	75.8	3330	1000	3.3	1000
Co-60	5.3	5.3	231.3	1.03	224.5	300
Total		100	4625		227.9	

for Radiation Safety), Appendix 10 [2]. Permissible specific activities for the given radionuclides are listed in Table 1.

The OSPORB-99 requirement (paragraph 3.7.6) was adopted as a criterion for forwarding the metal for ageing [2]: radiation intensity at the surface of a shielded source intended for use in industrial conditions should not exceed 2.78 μR/s (100 μGy/h).

The result of remelting of low-level metallic radioactive waste is presented in Table 1. Radionuclide contents are taken from the data of the Zvezdochka Company.

VALIDATION OF THE TECHNOLOGICAL PROCESS OF DISPERSION

During MRW remelting, the melt is traditionally teemed into ingot moulds to form bullions, which are raw materials for iron and steel industry.

The metal melt products differ in residual radioactivity because of different degrees of surface contamination by radionuclides in MRWs forwarded for remelting. This offers an opportunity to form metal batches of varying specific activity.

Recently, production and multipurpose utilization of metallic shot have become a new promising trend in granulation metallurgy. Principles underlying this new direction are: the study of formation and crystallization of metal granules, investigations of their structure and physico-mechanical and performance characteristics, and selective modification of the properties of metalware obtained from metallic shot. New effective technologies of shot utilization are being introduced in metallurgy, mechanical engineering, foundry and welding industries. The ensuing specific requirements to the quality of granules and to the production process stimulate the development of shot casting industry. The applicability of the shot has grown considerably. Metallic shot of different grades is used as inoculator during suspension pouring in metallurgy and foundry, as granulated filler material in welding, as a reinforcing phase of the components of high-strength products formed by isostatic hot compaction in materials science, for hardening details in metal-working, for biological protection in atomic power engineering, as ballast in shipbuilding, for cutting and grinding stones in construction materials industry, *etc*.

Hence metallic shot is a commodity with high consumer properties.

Dispersion of a liquid metal melt to form a granulate from remelted MRW makes it possible to fractionate the granulate according to the degree of its activity. This increases the yield of the metal with standard permissible activity and accordingly decreases the yield of the metal directed for natural decay of radionuclides during ageing.

The advantages of shot production such as reliable process, easy automation, and remote control and monitoring make it preferable to ingot production, which requires that the operating staff should attend to the knock-out from ingot moulds, prepare the latter to casting, *etc*.

The process of shot production from the metal melt is well studied. A production process and a “Grad” set of equipment with an output of 400–5000 t per year [5] were designed in the late 1970s at the Institute of Foundry Problems, Academy of Sciences, Ukrainian SSR. The basic technological principle of shot production is spraying a flat jet of melt by power supplies (water under pressure or compressed air) into water.

TECHNOLOGICAL PROCESS OF METAL DISPERSION AND FRACTIONATION ACCORDING TO RADIOACTIVITY

Using equipment complexes for shot production by spraying the metal melt with water or compressed air demonstrated that the shot is characterized by low quality:

– high degree of deformation;

– large quantities of particles varying in size;

– significant porosity and surface cavities; finally,

– the shot is oxidized by interaction with the power supply.

In addition, the metal particles (spray) freeze onto the spray unit, main collectors and sprue hole, forming wall accretions. This results in freezing of the sprue hole and quick failure of the spray unit.

The centrifugal metal dispersion process is free from these limitations. According to the data of [6], the shot is more uniform in size distribution, is dense enough, and has lower porosity. The sprue hole is in more favourable temperature conditions, and the spray unit is a mechanical centrifugal granulator made from heat resistant material. A flow chart for shot production using a centrifugal shot casting machine has been developed at the “New Technologies in Metallurgy” Company. The pilot tests of the machine have demonstrated reliable work and fairly high quality of the granulate product.

The metal is poured through a funnel. Having passed the runner gate of a tray, the molten metal gets onto the rotating granulator and breaks down into drops, which are thrown to the walls of the case, into the water layer formed by rotation of the impeller. Meanwhile, the molten metal drops mould into pellets. The cooled shot rolls along the tapered part of the case into the elevator, which is the unloading site. The water surface involved in rotation induced by the impeller takes paraboloid form. The diaphragm mounted in the cover ensures the formation of a fixed water layer no less than 225 mm thick. The spill part of the water flows over the diaphragm and goes away through the connecting pipe into the turnaround system for refrigeration. Hence, continuous water circulation and cooling are maintained. The steam formed in the course of refrigeration is sucked off through the connecting pipe.

A project of a pilot complex for radioactive waste processing has been worked out at VNIPIET (All-Russian Project and Research Institute of Diversified Power Technology) for “DalRAO”. The complex is intended for remelting radioactive metal waste in the form of fragments in a special gas-lift electric furnace designed at the “Crystal” Scientific Research Centre (GU NIITS).

A flow diagram has been developed (Fig. 1) for granulate production with direct draining of liquid steel from the furnace complex and

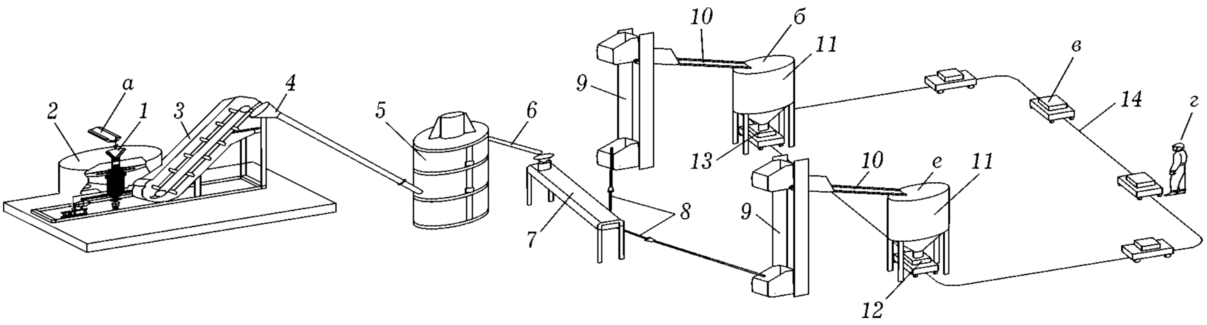

Fig. 1. Plant layout for shot production and fractionation according to radioactivity: *1* – funnel; *2* – shot casting machine; *3* – inclined elevator; *4* – receiving cone; *5* – vibrating dryer; *6* – tray; *7* – radiometric dividing box of RS type; *8* – final chutes; *9* – skip hoist; *10* – tray; *11* – receiving bin; *12* – container for the separated “pure” shot; *13* – container for the shot of limited application; *14* – wagon towing conveyor; *a* – draining tray for the tapped-out metal, *b* – bunker for the limited application shot, *c* – shot for temporary storage or for sale for articles of the Ministry of Atomic Energy, Russian Federation, *d* – shot for sale in the general industrial complex, *e* – bunker for “pure” shot.

subsequent fractionation of the granulate according to the degree of gamma-activity.

The flow diagram affords increased yields of the metal with a permissible level of radioactivity. The end product is of higher market value compared to ingot metal.

The granulate production line functions as follows. The molten metal coming from the overflow device of the gas-lift furnace is fed to funnel *1* of the shot casting machine *2*. The flat jet of metal shaped by the sprue hole is granulated under the action of the centrifugal forces of granulators and enters the tapered refrigeration chamber filled with water, from which granules are delivered by inclined elevator *3* to vibrating dryer *5*, where they are dried, while in motion, with electroheaters (drying temperature 180–250 °C). From the vibrating dryer the granules are discharged on the belt of radiometric dividing box 7, on which the shot is fractionated according to gamma-activity. If the exposure dose rate is exceeded, the direction of shot movement is altered by means of a mobile chute. The chute is operated by a gamma-gauge adjusted to an exposure dose rate of 2.78 μR/s or to any other value.

The granulate divided into two streams is loaded by skip hoists *9* into associated storage hoppers *10*, *11*. From the storage hoppers the granulate is released into containers and goes for ageing or to the consumer.

Radiometric separators of RRS type are produced by the "Rados" Company and have been in successful operation in sorting of various ores and slags.

Additional fractionation of granules according to gamma-activity can be performed if necessary because gamma-radiation intensity of the remelted metal correlates well with the Co-60 content in the metal. This affords a fraction with a Co-60 content of less than 300 Bq/kg.

Containers with granules characterized by dose rates higher than 2.78 μR/s (100 μGy/h) are forwarded for ageing until they have reached the permissible level of decay of radionuclides; those with a limiting activity of more than 300 Bq/kg but with dose rates lower than 2.78 μR/s (100 μGy/h) are supplied for the needs of atomic industry; and those with a specific activity of less than 300 Bq/kg are subjected to fractionation of granules according to size, to thermal treatment, weighing, and packing, and dispatched for general-purpose use in industry.

Automatic control of β-ray emitters is not performed because the specific activity of the source metal and high Sr-90 and Cs-137 decontamination factors guarantee low residual contents of these isotopes in the remelted metal.

After radiometric fractionation, manual random inspection for β-ray emitters is made.

Shot production and fractionation according to gamma-activity are completely automated processes. The technological line is computerized and man-operated. The gamma-activity of the processed metal is also recorded by a computer.

Technical data for equipment for metallic granulate dispersion and fractionation are presented below:

Output, t/h	0.4–1.0
Shot material	Stainless steel
Size distribution, mm	0.4–6
Shot density, kg/m^3	7200
Melted metal temperature during pouring, °C	1540–1580
Refrigerant	Process water
Water discharge in the closed cycle, m^3/h, no more than	3–5
Finished product, t/year	2000
Including:	
the shot of general industrial use (less than 300 Bq/kg for Co-60)	1200
the shot for Ministry of Atomic Energy needs (more than 300 Bq/kg for Co-60, but less than 2.78 μR/s)	600
the shot for ageing (more than 2.78 μR/s)	200

CONCLUSIONS

The technological process of dispersion of the metal purified from radionuclides by remelting and its fractionation according to gamma-activity make it possible to obtain metallic shot which is economically preferable to metal ingots moulded by conventional technology. In addition, fractionation according to gamma-activity enables one to ensure reliable radioactivity control according to the residual value, which precludes accidental release of radionuclides into environment.

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