

VITRIFICATION OF ASH RESIDUE IN CONTAINERS

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ABSTRACT

Vitrified ash residue, left after incineration of solid radioactive waste, is the most sustainable waste form for safe storage and disposal. Use of conventional vitrification technologies such Joule heated or induction cold crucible melters for this is discussed.

The thermo-chemical technology looks like promising one in order to obtain the glass-like products. This technology uses the exothermic reduction and oxidation reactions in order to produce heat required for vitrification of ash. As a result, up to 60wt.% of ash residue can be immobilized in glass-like matrix. This matrix meets the requirements of storage for solidified radioactive waste.

Technological process was conducted in the specially designed crucible-container. The vitrified ash is disposed in the same container.

The process of taking away the radionuclides and macro-components during the vitrification process was studied.

The main parameters of process as well as the chemical and physical characteristics of glass-like product are presented.

The possibility of use of the developed technology for vitrification of contaminated soil was also studied.

INTRODUCTION

Moscow SIA "Radon" develops different thermal technologies for vitrification of ash residue, left after incineration of solid radioactive waste. These technologies include the technologies of ash residue vitrification into borosilicate glass in Joule heated melters (1) and in induction «cold» crucible melter (2,3), as well as the obtaining of the melted glass-ceramic materials using plasma furnace (4).

As an alternative, more simple and cheaper technology based on exothermic thermo-chemical treatment of ash residue is developed. This technology doesn't require an external source of heat for vitrification process. Both reduction and oxidation exothermic reactions between the components of the heating batch provide heat. Mixture of heating batch and ash residue is placed in the crucible-container. Because of solid-phase exothermic chemical reactions between components of heating batch and ash residue, the temperature in the reaction zone reaches 1200-1800°C (depending on composition of the heating batch). Reaction is developed in the relatively thin layer and spread in the batch in form of combustion wave. As a result, the content of the crucible-container melts. After cooling, the monolithic glass-like product, which is ready for disposal, is obtained.

Composition of heating batch is computed in a way to generate heat in amounts required for ash residue melting. In case of melting of ash residue no less than 2,000-4,000 kJ/kg of heat has to be provided by thermo-chemical process. The heating batch developed by Moscow SIA

“Radon” based on K, Mn, Si, Al, Ca, Fe and other components isolates about 5 MJ/kg of energy. The temperature of reacting mixture reaches 1600 – 1800°C.

The similar heating bath with special additives was used for vitrification of the soil, containing clay and sand, and contaminated by radionuclides.

EXPERIMENT

The experiments on vitrification of ash residue were conducted in the crucible-containers from 1 to 10 liters in volume.

The process contains the steps as follows:

1. Ash residue is crushed (particles size bellow 0.3mm),
2. Crashed ash residue is mixed with the heating batch,
3. Mixture is filled into container,
4. Exothermic reaction is initiated by electric ignition device.

At the end of exothermic reaction, the glass-like monolith is developed in the container with the volume reduction by 2-4 times. Then, the filling of container with the mixture of ash residue and heating materials followed by exothermic reaction are repeated as many times as it is necessary in order to fill the container completely with the final product.

The release of radionuclides and macrocomponents during the vitrification process was studied. The temperature of melt T , °C and the speed of release of particles (by mass) U , g/min as a function of time are represented at the Fig. 1.

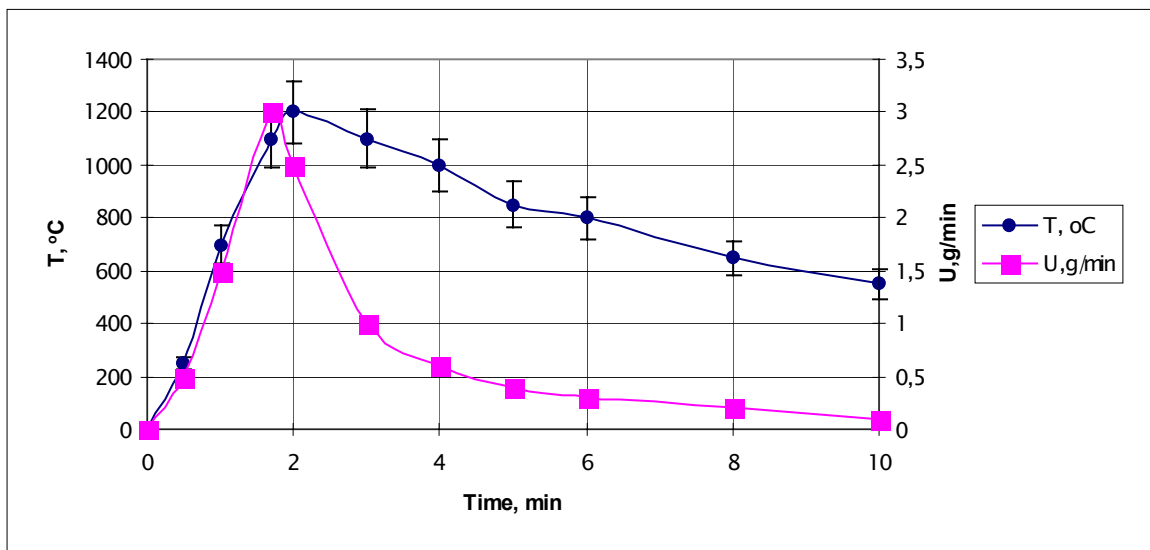


Fig 1. Melt temperature and release of particles (by mass) in function of time

The lost of particles was between 0.6 and 3%. The chemical analysis of particles has shown that they contained, mainly, the oxides of potassium, silica, calcium, and aluminum. As it was shown before (5), the release of particles depends on process temperature. The more is the process temperature and the lower is the content of ash residue in the mixture the higher is the release of particles during thermo-chemical process.

The size of particles did not exceed 0.5μm. The off-gas volume was 10-20cm³ per 1g of ash residue/heating materials mixture.

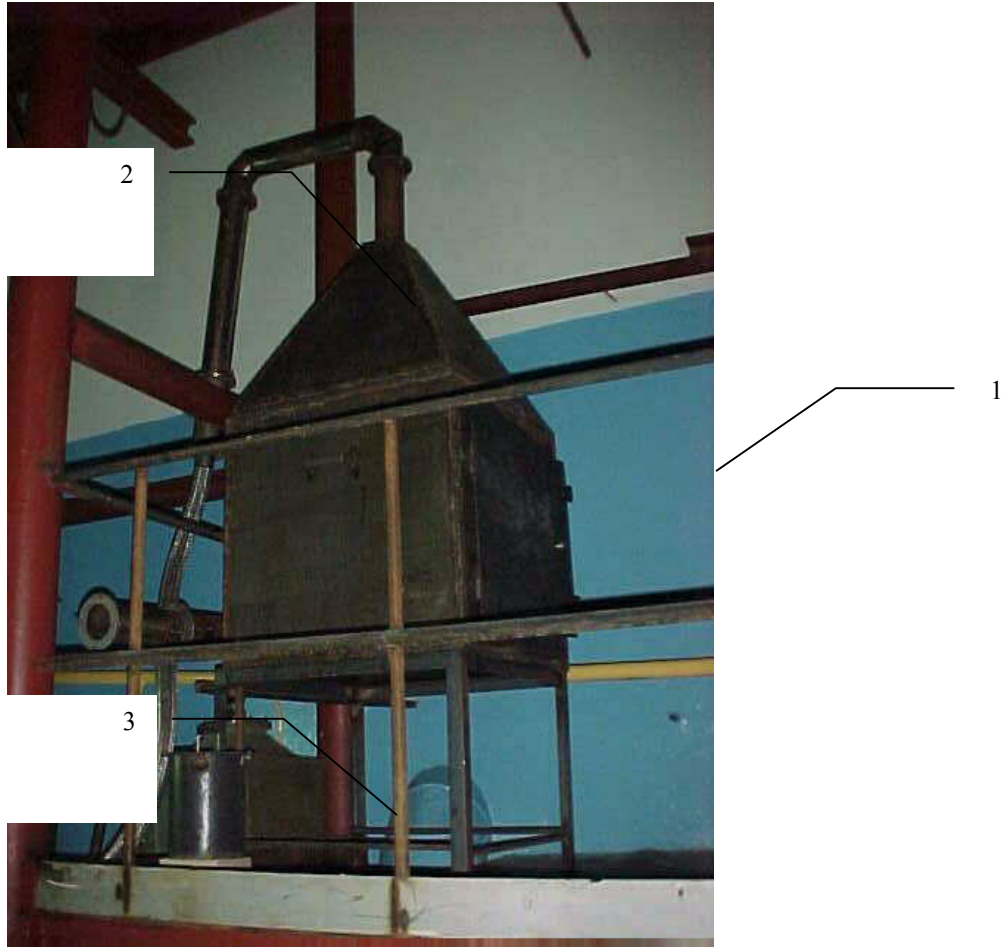


Fig. 2. Stand for vitrification of ash residue:
1 - technological box, 2 – outlet to the gas cleaning system, 3- crucible-container (shown out of technological box)

The tests with the volume of mixture up to 10 liters were performed at the stand shown at Fig.2. Stand was connected to the gas cleaning system of Radon's industrial radioactive waste incineration plant "Fakel". The vacuum not less than 200Pa was sustained in the technological box. The device for taking samples of gas was mounted into the off-gas pipeline.

The vitrification of ash residue and its simulator in the container of 10 liters was performed. The ratio of ash residue/heating materials was 60/40.

The speed of gas in gas pipelines was measured by thermo-anemometre. The average linear speed of gas in the outlet pipeline was 2.4 m/sec and the volumetric speed of the air coming to the technological box was around 68 m³/hour.

Electric aspirator took the aerosol samples to the analytical filter. The content of oxygen, hydrogen, carbon oxides and hydrocarbons was measured by chromatographs. The results are shown at the Table 1.

Table1. Spread of radionuclides and aerosols during vitrification process.

Ash residue	Content in off-gas, % volume	Spread, %			
		Aerosols	$\Sigma \beta$ Cs ¹³⁷	$\Sigma \beta$ Sr ⁹⁰	$\Sigma \alpha$ Pu ²³⁹
Simulator of ash residue	CO - 0.02 CO ₂ - 0.14 H ₂ - 0.01 O ₂ - 21 Hydrocarbons - not found	0.2	-	-	-
Real ash residue	CO -- 0.001 CO ₂ - 0.1 H ₂ - 0.001 Hydrocarbons - not found	1.0	1.1	1.1	0.002

The results of tests have also shown that the spread of radionuclides increases with the increase of volume of mixture of ash residue/heating materials. For the volume of 10 liters the spread of radionuclides was 1.1% against 0.3-0.5% for the volume of 1 liter. Probably it may be explained by the slower cooling of the block of glass-like material having a bigger mass. The glass-like monolith, manufactured in 10 liters container, had more homogenous and less porous structure than the monolith, prepared in 1-liter container.

XRD analysis of final product shows that it consists mostly of the amorphous phase. The fraction of amorphous phase in the final product increases with decreasing of the content of ash residue. Quartz and cristobalite are mostly present in the crystal phase.

Up to 3-5% by mass of the metallic inclusions of spherical form were found in the monolith structure. These inclusions are well visible at images of surface made by electronic microscopy (Fig. 3) and spectroscopy (Fig. 4).

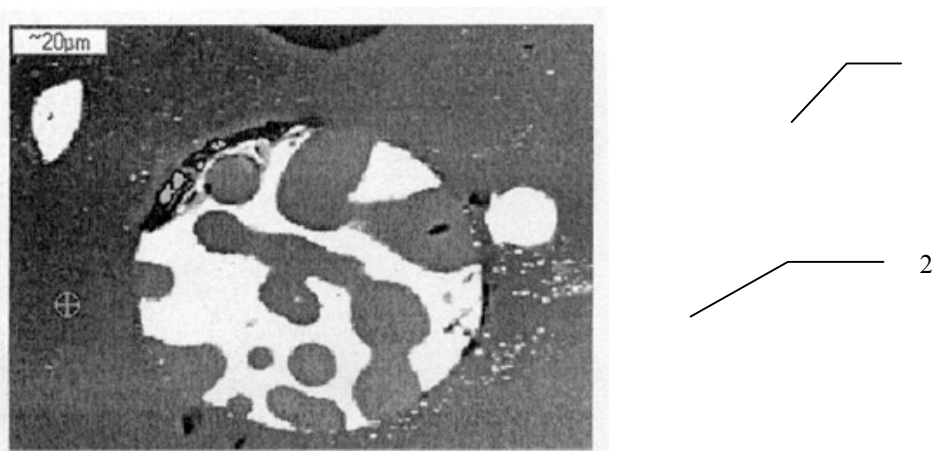


Fig. 3. Electronic microphotography of the surface of the vitrified product: 1- matrix, 2 – inclusion

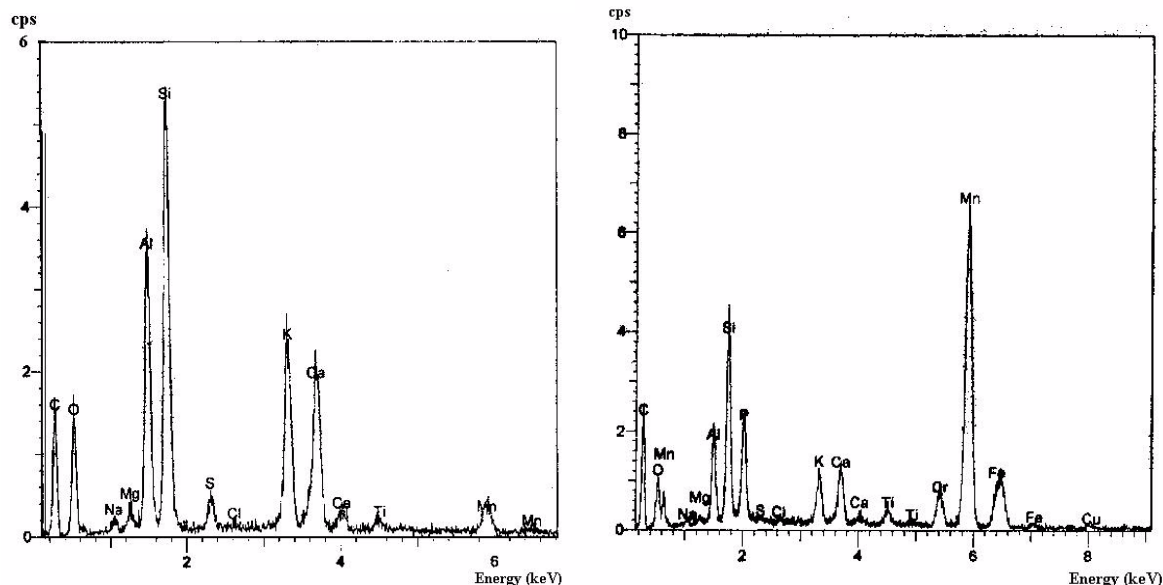


Fig. 4. Spectrum of glass-like matrix (a) and inclusion (b).

The content of elements in matrix and inclusions is shown in the Table 2.

Table 2. Content of elements in matrix and inclusions

Phase	Content of element, % at									
	O	Si	P	Cr	Mn	Fe	Al	Ca	K	Na
Matrix	43.13	16.7	0.09	0.05	2.50	0.20	15.25	7.61	10.77	1.29
Inclusions	-	9.32	4.86	0.23	60.64	12.18	-	-	-	-

It is evident that the macroelements-analogues of radionuclides, potassium and calcium, are localized in the glass-like matrix.

The study of chemical and physical properties of the final product shows that the density of glass-like monolith is 2.5-3.0 g/cm³, the compression strength – 50-100Mpa.

Leaching rates of ²³⁹Pu and ¹³⁷Cs from samples with the different content of ash residue were measured. Standard IAEA leaching test procedure was used. Leaching rates of 10⁻⁶-10⁻⁵ g/cm²*day for ¹³⁷Cs, and 10⁻⁸-10⁻⁷ g/cm²*day for ²³⁹Pu were obtained. So, the leaching characteristics of final glass-like product are similar of those for glass, obtained by vitrification of ash residue in conventional melters.

CONCLUSION

1. The technology, which allows to vitrify the ash residue directly in containers, was developed
2. The solidification process is going on directly in containers. After the end of the process the containers are ready for storage or disposal.
3. The spread of radionuclides and macrocomponents in the course of vitrification of ash residue do not exceed the admissible level. Variation of content and volume of ash residue/heating materials mixture could optimize it.

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4. The content of off-gases remains in the admissible limits.
5. The final product meets the requirements for solidified radioactive waste to be disposed.
6. Up to 60 wt. of ash residue can be immobilized in glass-like matrix.
7. The technology can be used for vitrification of ground, based on clays and sand, and contaminated with the radionuclides.

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