CONDITIONING OF DISPERSED RADIOACTIVE WASTE WITH POWDER METAL FUEL

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ABSTRACT

A method for thermochemical conditioning of dispersed radioactive waste (ash residue, soil, soil with metal fragments) based on using of powder metal fuels (PMF) has been developed. PMF intermixed with ash residue at the appropriate ratios can sustain a glass forming reactive wave, that produces monolith glass-like blocks. The method for thermochemical conditioning of real incinerator ash residue, contaminated clay soil, and soil containing metal waste fragments is described in this paper. The conditioning technique and obtained materials are investigated in order to found the optimal processing parameters and to characterize the product materials.

INTRODUCTION

Ash residue from solid radioactive waste incineration is to be conditioned prior to transportation and disposal in order to avoid possible releases of radionuclides into the environment. Usually, contaminated clay soils have radionuclides strongly fixed to the clay particles. This makes cleaning (washing) the soil a complex task. This must also be conditioned to produce a waste form suitable for safe transportation and storage. The thermochemical treatment of such wastes (ash, clay soil) ensures their safe conditioning.

The thermochemical conditioning process can be divided onto three phases:

1. Preparation of PMF and its intermixing with waste material (ash residue, clay soil).
2. Ignition of self-sustaining heat-liberating reactive wave in the mixture.
3. Cooling of the obtained monolithic glass-like block.

The technological process is carried out directly in transportation/disposal container. Since reduction of the initial mixture volume (approximately in 2-3 times) the conditioning process is repeated a few times in each container.

EXPERIMENTS

For the thermochemical process the radioactive wastes (RAW) as follows were used:

- Ash residue from solid radioactive waste incineration,
- Soils based on soil and send contaminated by radionuclides, and
- Radioactive clay and sandy soils containing fragments of metal wastes.

Table I represents the chemical composition of the waste in oxide form and the table II represents the radioactive isotope composition of the waste.

Table 1. Chemical composition of radioactive waste

Content, weight %	**Ash residue**	**Soil**
SiO_2	32,0	75,3
P_2O_5	13,7	-
Al_2O_3	2,4	7,6
Fe_2O_3	2,6	4,9
CuO	1,3	-
CaO	19,6	0,7
MgO	1,3	1,4
K_2O	6,4	2,8
Na_2O	3,5	1,5
TiO_2	-	-
FeO	-	-
SO_3	-	-
MnO	-	-
Loss during calcination at 900 ^{0}C	17,2	5,6

Table II. Radioactive isotope composition of calcined ash residue

Isotopes content, Bq/kg						
$\Sigma\beta\,^{137}Cs$	$\Sigma\beta\,^{90}Sr$	$\Sigma\alpha\,^{239}Pu$	^{133}Ba	^{137}Cs	^{60}Co	^{90}Sr
$1,4*10^6$	$17,7*10^5$	$3,5*10^5$	$8,1*10^3$	$1,3*10^5$	$7,6*10^4$	$1,5*10^5$
Isotopes content, Bq/kg						
^{134}Cs	^{108}Ag	^{234}U	^{238}U	^{238}Pu	^{239}Pu	^{226}Ra
$6,3*10^3$	$1,8*10^5$	$1,7*10^4$	$3,2*10^4$	$2,5*10^4$	$1,4*10^5$	$2,8*10^2$

Mainly 137Cs represent radioactive isotope content of the soil. During the development of the conditioning process the scheme as follows were used:

1. Chemical analysis of radioactive waste,
2. Heat effects calculation of possible reactions,
3. The compositions selection and the reaction mixture tests under laboratory conditions,
4. Optimization of the reaction mixture compositions based on the laboratory results,
5. Determination of the process characteristics: the temperature of the reaction mixture, radionuclides and macrocomponents carry over,

6. Studying of the structure and the end monolith product properties, and
7. Realization of the experiments on the radioactive waste thermochemical conditioning bench unit.

Heat base – mixture of reactants-oxidants and reducers were selected by calculation for the radioactive waste with indicated composition. A large quantity of heat release after reaction initiating in the heat base and radioactive waste mixture as a result of the reactants interaction. The heat provides the mixture melting and glass-like material forming without external power supply.

By means of thermodynamic analysis, the PMF to be used for ash immobilization with the empirical formula $K_{2.28}Ca_{1.70}Al_{3.37}Si_{10.52}Fe_{1.63}Mn_{2.87}O_{11.50}$ and the PMF to be used for clay soil vitrification with the empirical formula $K_{3.16}Ca_{1.23}Al_{6.30}Si_{7.64}Fe_{1.18}Mn_{3.16}O_{12.66}$ were chosen. The reactions between components of this PMF generate a large quantity of energy: the equilibrium temperature reaches 1900-2000 ^{0}C.

Laboratory experiments were carried out using 1L-alunde crucibles. The reacting mixtures that involve radioactive waste and heat base at the appropriate ratio were placed in a crucible. Further, the mixture was initiated with fire-supply cord or electric pulse. The combustion wave develops and spontaneously propagates from top to down on initial material (substance) with a rate up to 5-10mm/s. High temperature necessary for melt process and new junctions formation develops in the combustion wave.

Radioactive wastes before intermixing with heat base were grind for exception of large particles by a size more than 0,5 mm.

The process temperature was measured by thermocouples placed within the reacting mixture bed depth. Fig.1 shows typical thermograms obtained during monolith soil forming process.

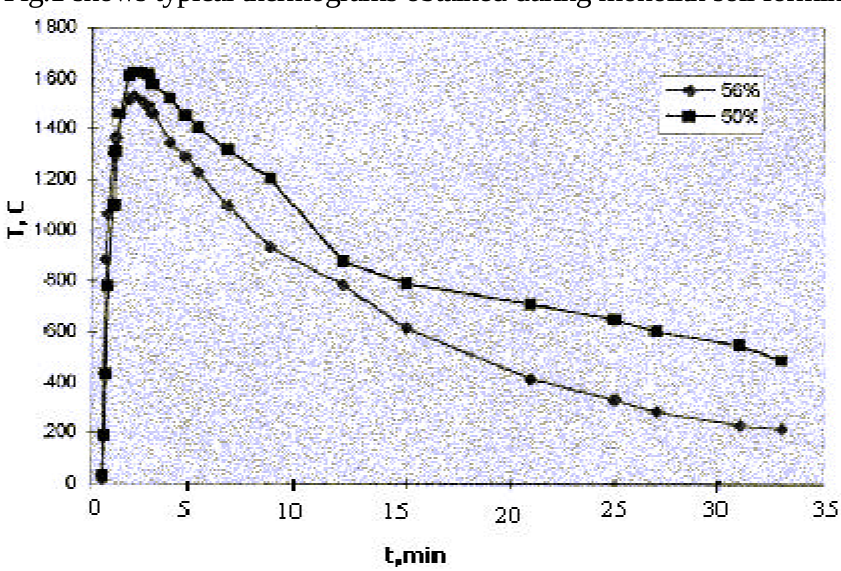

Fig. 1. Thermograms for the monolithic soil forming process

For determination of macrocomponents and radionuclides carry over from reaction region the process was performed in a special device mounted in exhaust cabinet (Fig.2).

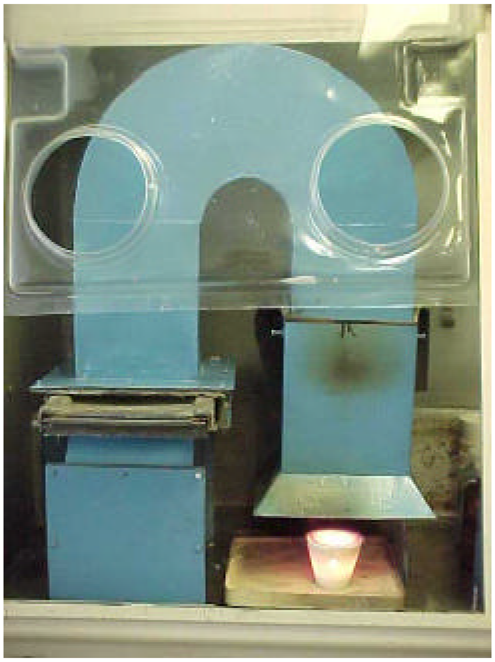

Fig. 2. Device for the performance of the process: 1 - crucible-container, 2 - gas collecting hood, 3-cassette with aerosol filters.

The selection of the all aerosols released during the reaction in the crucible-container 1 was performed using aerosol filters placed into the cassette 3.

The dynamics, quantity, chemical and radionuclides composition of the aerosol release was defined. Oxides of the main part of the metals in the initial reagents represent the chemical composition of the aerosols. The aerosols carry over increases with process temperature raising as during ash residue monolith producing and with decreasing of radioactive waste content in the reaction mixture accordingly. The Fig.3 shows the aerosols and radionuclides (^{137}Cs) carry over dependence of radioactive waste (ash residue, clay soil) content in the reaction mixture.

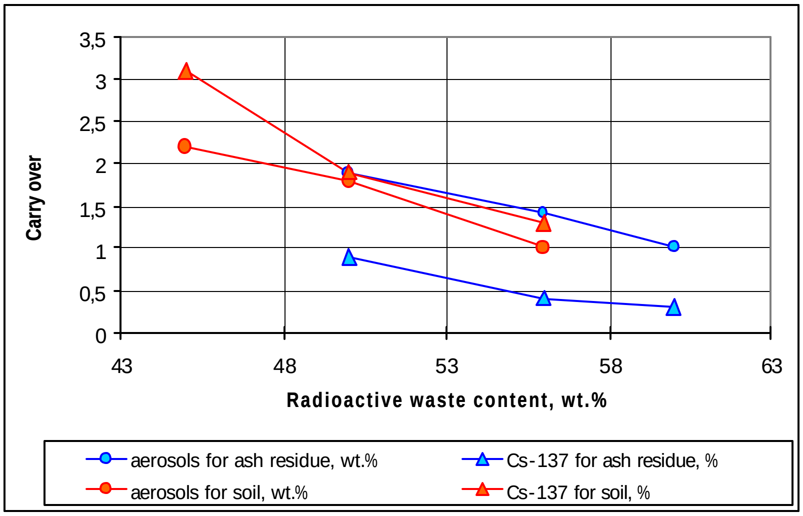

Fig. 3. The melt temperature and aerosol release mass rate dependence on time.

As a result of the thermochemical process of the all wastes described above end products in the form of green-black glass-like materials which have dense structure or structure with finest pores are produced. During soil conditioning a layer of metal alloy in a one monolith form with a glass-like end product on the top is formed on the crucible bottom.

The results of the end product treatment study are represented in the Table III.

Table III. Characteristics of end product

RAW content, %	Density, g/cm^3	Comprehensive strength, MPa	Leachability, g/cm^{2}day		Volume reduction of RAW
			^{137}Cs	^{239}Pu	
Ash residue					
50	2,7	20	$4,4 \cdot 10^{-6}$	$2,6 \cdot 10^{-7}$	2,5
56	2,7	12	$6,4 \cdot 10^{-7}$	$6,0 \cdot 10^{-8}$	3,0
60	2,9	17	$5,2 \cdot 10^{-6}$	$1,4 \cdot 10^{-7}$	3,0
Contaminated soil					
45	2,4	8	$7,5 \cdot 10^{-6}$	-	1,2
50	2,6	9	$6,5 \cdot 10^{-6}$	-	1,5
56	1,8	9	$5,4 \cdot 10^{-6}$	-	1,0

CONCLUSIONS

The samples of glass-like materials produced during the suggested process comply with the requirements for intermediate activity solidified radioactive waste.

It should be noted that with samples amorphousity increasing i.e. with increasing of glass phase content and homogeneity of the samples their water stability raises. At the same time the process temperature and correspondingly the radionuclides carry over raise with decreasing of ash residue and soil content in the reaction mixture. Therefore choosing the heat base and the reaction mixture compositions for both ash residue and other radioactive wastes treatment the optimal method seems to be the method ensuring the parameters as follows:

- sufficient volume reduction of a solidified product in respect with initial radioactive waste volume;
- safety process i.e. maximal reduction of the radionuclides carry over during radioactive waste monolith producing; and
- sufficient quality of the end product for the safe long-term storage.

REFERENCE

1. I.A.Sobolev, O.K.Karlina, G.A.Varlaova, V.M.Tivansky, M.I.Ojovan, K.M. Efimov. Vitrification of ash residue in containers // Waste Management`2000. Proc.Int.Symp., Tucson, AZ, 2000. (on CD Rom).