WASTE FORM PERFORMANCE UNDER OPEN SITE AND PILOT REPOSITORY CONDITIONS

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ABSTRACT

Three types of matrices for LILW conditioning have been under investigation at Scientific and Industrial Association "Radon" for decades. Cemented, bituminized and vitrified waste blocks were produced in industrial and pilot scale facilities, using technologies developed at SIA "Radon", and disposed of into several pilot-scale shallow ground repositories and in the open testing area. In the latter case, waste forms are subjected to the combined action of climatic factors resulting in more rapid corrosion of waste forms, thus allowing consideration of testing under open field conditions as an accelerated method for assessment of wasteform durability.

INTRODUCTION

At present, vitrification is considered the most promising process for HLW waste immobilization. It also represents the possible method of intermediate level radwaste solidification due to maximal volume reduction, high chemical stability of the end product and capability of glass to incorporate all the major waste radionuclides. Further, low leaching rates of critical waste constituents from waste glasses are of principal importance for near-surface repositories.

Experimental evidence shows that leaching behaviour of radionuclides in waste matrices under natural ambient conditions differs strongly from that in laboratory conditions. Hence, we supposed that test procedure in open field environment could provide more correct data on long term waste form behaviour. Here we present results of long term testing of vitrified, cemented and bituminized waste products obtained from industrial and pilot-scale facilities of SIA «Radon».

EXPERIMENTAL

We used real medium level wastes from NPP reactors WWER and RBMK. The main radioactive constituents of initial liquid radwaste sludges were ^{137}Cs (40-90%), ^{134}Cs (7-17%), ^{60}Co (1-15%). The total of ^{90}Sr, ^{144}Ce, ^{106}Ru, ^{238}Pu was less than 1%. Nitrates and borates of alkali and alkali-earth elements were predominant: $NaNO_3$ (100-300 g/l), Na_2BO_2 (up to 100 g/l), Na_2CO_3 (10-90 g/l).

Samples used for the tests were prepared in industrial (cementation and bituminization) and pilot scale (vitrification) facilities with waste loading 30%, 31% and 37% for vitrified, bituminized and cemented waste blocks, respectively. For vitrification, the borosilicate glass was used that contained the main 4 oxides in proportions: 48 % SiO_2, 16 % Na_2O, 15,5% CaO and 7,5% B_2O_3. Bituminized and cemented waste blocks were prepared using portland cement and bitumen without any additives.

At the open testing site, solidified waste samples were placed on stainless steell trays for collection of contacting water (1). Waste packages in the near surface pilot repositories were disposed of at the depth of 2-2.4 m in a loamy soil. The main parameters of soil are: the freezing depth up to 0.7m, humidity 20-30%, bulk density 2.1-2.3 g/cm^3, water permeability coefficient 0.3-0.8 cm/day.

Open field testing includes visual observation, systematic sampling of water and solids (matrices, soil, container materials) and their analyses, data processing and general performance assessment of waste forms and packages (2).

RESULTS

Under the action of external factors (mixed atmospheric precipitations, air temperature fluctuations, solar irradiation, et al.) the open glass surfaces become tarnished and covered with a network of microcracks. The thickness of the alteration layer was of the order of 1.5-3 μm. The surface of bitumen blocks remained visually unchanged for years. As for cement blocks, their lifetime under conditions of open field was limited. The horizontal and vertical cracks penetrated the cement blocks, grew thick and after 6-year exposition the blocks were split into large fragments. Besides, a network of more fine threadlike cracks developed, resulting in a formation of nuciform partings.

Radionuclide release rates were estimated on base of initial radioactivity data and analytical results. As an example, leaching parameters of radwaste forms containing Kursk NPP operational waste are listed in Table. The radionuclide retention is expressed in term of coefficient f_i equal to the inventory fraction released per year (3). For the bitumen matrix, the average f_i for the exposure time was **(1-2)·10^{-3} yr^{-1}** while the average f_i for waste glass was **5·10^{-4} yr^{-1}**. Thus, the leaching rate data obtained for samples with waste loading ranging from 30% to 37% (Kursk NPP) are indicative of a high radionuclide retention of glass matrix.

Table. Waste form performance. Open-site tests

Matrix type	Initial specific radioactivity, Bq/kg		Leaching rate, g/cm^{2}day		Leaching factor cm^2/day	
	β-nuclides	α-nuclides	after 1 year	after 8 years	after 1 year	after 8 years
Glass	3.89*10^6	1.4*10^4	8.61*10^{-7}	3.72*10^{-7}	6.03*10^{-12}	1.78*10^{-11}
Bitumen	3.29*10^6	3.89*10^2	8.06*10^{-5}	5.38*10^{-5}	2.50*10^{-8}	8.03*10^{-10}
Cement	2.2*10^6	-	1.51*10^{-4}	9.28*10^{-5} max. lifetime 6 years	5.11*10^{-6}	2.67*10^{-6} max. lifetime 6 years

Figure illustrates the average annual specific radioactivity of rainfall water contacting waste blocks as function of exposure time. As can be seen, radioactivity levels decrease with time, except for cement block where an increase in radionuclide leaching rate occurred after the loss of block integrity.

Systematic monitoring of groundwater contamination in pilot shallow ground repositories, performed through borehole sampling of water in contact with waste blocks, revealed that all the waste forms exhibit low rates of radionuclide release during time intervals of about two decades. Water contamination values range from $1\cdot10^{1}$ to $1\cdot10^{2}$ Bq/l.

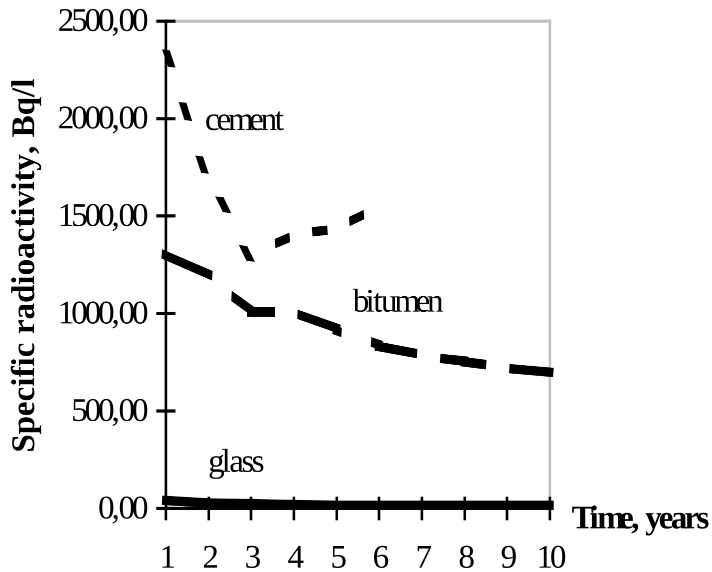

Fig. 1. Average annual specific radioactivity of water contacting waste blocks. Waste from Kursk NPP.

CONCLUSIONS

Radionuclide leaching rates, determined for glass, cement and bitumen waste forms under open site conditions, compared with data of borehole groundwater monitoring in pilot near surface repositories allows to draw a conclusion that weather ageing yields more conservative data on wasteform behaviour. Results demonstrate high radionuclide containment properties of intermediate level waste glasses prepared on pilot-scale vitrification facility of SIA «Radon».

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