Influence of the Sludge Content on the Mechanical Properties of the Cemented Based Radioactive Waste Form

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The paper describes the results obtained in the laboratory investigations on the mechanical properties of the cement-based radioactive waste form containing two kinds of sludge in different contents. One of them simulates the sludge obtained by treatment of liquid radioactive effluents using a PA-type anionic polyelectrolyte, *and the other one simulates the sludge obtained by decontamination of contaminated surfaces using a pNaAc-* type *hydrogel. The influence of the content of sludge on the setting time of the mortar paste as well as on the compressive and bending strength of the cement-based radioactive waste forms, were studied.*

Keywords: waste form, radioactive waste, mechanical properties, sludge

The nuclear energy production generates a large variety of low and intermediate level radioactive waste. It contains mainly contaminated materials, sludge and concentrate, ash, ion-exchange spent resins, contaminated oils and organic solvents.

The sludge is secondary radioactive waste resulted both from the treatment of aqueous radioactive waste and from the decontamination of radioactive contaminated surfaces.

The sludge resulted from the treatment of aqueous radioactive waste as well as the one resulted from the decontamination of contaminated surfaces need to be immobilised, through solidification, into various materials in order to be suitable for handling and disposal. There are lots of materials, known under the name of waste forms, used for the solidification of sludge, such as: concrete, bitumen, polymers, ceramic materials and glass, which ensure mechanical strength for handling, as well as durability of long term retention of contained radionuclides. The most common material used for the solidification of radioactive sludge for subsequent disposal is concrete. Typical concrete consists of a mixture of Portland cement, sand and water in various proportions and it possesses many practical advantages: good mechanical properties, low cost, easy operation, and radiation and thermal stability. The chemical composition, as well as the sludge content, of the waste forms affects its properties in the sense of diminishing its quality. The mechanical properties of the cement-based radioactive waste forms affected by the chemical composition of the radioactive sludge are the following: the setting time, the compressive strength and the binding strength [1, 2].

Many techniques used for the treatment of aqueous radioactive waste, as well as for the decontamination of radioactive contaminated surfaces, have been developed, mainly with a view to obtaining a higher decontamination factor, without considering the content and quantity of secondary radioactive waste and their influence on the strength and long term durability of waste forms for disposal.

In Romania, techniques for the treatment of aqueous radioactive and for the decontamination of radioactive contaminated surfaces, using polymeric materials obtained through gamma irradiation, have been developed [6]. These techniques aim at obtaining a similar decontamination factor as those obtained through classical methods and at reducing the amount of radioactive sludge. The resulted sludge is characterized by a lower content of chemical compounds than the sludge resulted from the classical treatment and decontamination methods [5, 6].

The purpose of this paper is to experimentally determine the influence that the content and chemical composition of the following kinds of sludge have on the mechanical properties of the cement-based radioactive waste forms:

- sludge resulted from the treatment of aqueous radioactive waste using a PA-type anionic polyelectrolyte obtained through gamma-irradiation polymerization of an acrylamide and sodium-acrylate aqueous solution;
- sludge resulted from the decontamination of radioactive contaminated surfaces using a pNaAc-type hydrogel obtained through gamma-irradiation polymerization of a sodium-acrylate aqueous solution.

This paper presents the results of the experimental study on the influence of the content of two different kinds of sludge on the setting time, the compressive strength and the bending strength of the cement-based radioactive waste forms.

Experimental part

Materials and methods

Two kinds of sludge were simulated to prepare samples of cement-based waste forms:

- the sludge named hereinafter CF is simulated by preparation of a dilute effluent obtained by dissolution in demineralised water of salts in the following concentrations: $Al_2(SO_4)_3$ 5 g.L^{-1}, PA-type polyelectrolyte 1 g.L^{-1}, NaCl 20%.
- the sludge named hereinafter HG is simulated by preparation of an effluent obtained by dissolution of 25 g of pNaAc-type hydrogel in 1 L of demineralised water.

The samples of waste form were prepared by mixing of cement, sand, water in a constant ratio, and CF and HG in different concentrations as they are presented in table 1. In order to compare the results, a reference sample with a content of cement, sand and water was prepared (sample 1, table 1).

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Table 1
COMPOSITION OF CEMENT BASED WASTE FORM SAMPLES

Identification number of sample	Portland cement [g]	Sand [g]	Water [mL]	HG [g]	CF [mL]
1	300	300	120	0	0
2	300	300	120	30	0
3	300	300	120	60	0
4	300	300	120 +12	90	0
5	300	300	120 +18	120	0
6	300	300	120 +25	150	0
7	300	300	80	0	40
8	300	300	60	0	60
9	300	300	40	0	80
10	300	300	20	0	100
11	300	300	0	0	120
12	300	300	80	30	40
13	300	300	80	60	40
14	300	300	80+11	90	40
15	300	300	80+18	120	40
16	300	300	80+25	150	40
17	300	300	80	30	40
18	300	300	60	30	60
19	300	300	40	30	80
20	300	300	20	30	100
21	300	300	0	30	120

The ratio of cement: sand: water was constantly maintained 1:1:0.4 in all samples.

The determination of setting time was performed according to the standard procedure SR ISO 196-3:2006 “Methods of cement testing. Part 1. Determination of setting time” [7]. The compounds in the composition mentioned in table 1 were manually mixed for 5 min and the resulted mortar paste was poured in a plastic ring. For determining the setting time, a device for measuring of the hardening of the mortar pasta was used (Vicat device). The setting time represents the time passed from the preparation of mortar pasta to the hardness of the pasta.

The determination of compressive strength as well as the bending strength was performed according to the standard procedure SR EN 196-1:2006 “Methods of cement testing. Part 1. Determination of mechanical strength” [6]. The compounds were mixed using a mortar mixer device according to the following procedure: (1) water and sludge were mixed at low speed (2) powdering materials cement and sand were added while maintaining a low stirring speed, (3) mixing at high speed. The total mixing time is about 15 minutes. The obtained mortar paste was poured in moulds to get samples of 4 . 4 . 16 cm dimension. After the preparation, the samples were kept in the moulds, in normal atmospheric laboratory conditions, for 24 h, and then they were removed from the moulds and kept in tight plastic bags, in wet conditions, for 28 days.

The compressive and binding tests were performed on five specimens of the same sample, the recorded value of the compressive strength as well as of the bending strength being the average value. The compressive and binding tests were performed using a digital pressing device, MATEST-type, with loading rates of 1.2 MPa/s.

Results and discussions

The mechanical properties are strongly dependent on the water/cement ratio, the degree of compressing of mortar paste, the hardening time of cement. The samples were prepared in the same condition in order to emphasis the influence of content of HG as well as CF on the mechanical properties of the samples.

The mortar paste showed a good plasticity and workability except the mortar paste where the concentrations of HG were 90g, 120g and 150g. Low workability was noted regarding the samples 14, 15 and 16 with content of CF and content of 90g, 120g and 150 g of HG. For these mixture supplementary water had to be added in order to improve the workability of the mortar paste. The added water quantity is mentioned in the table 1(added values with “+”).

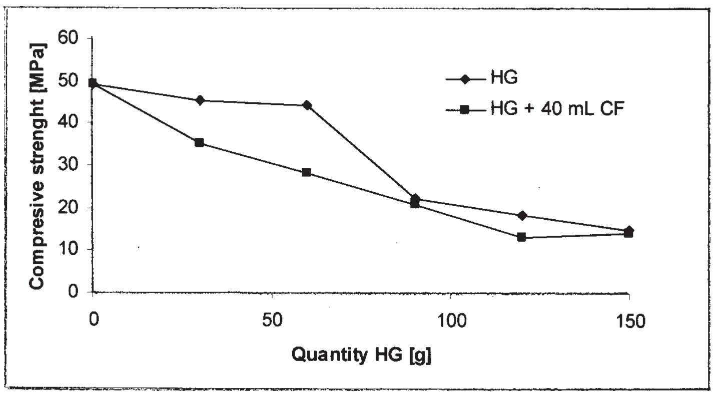

Fig. 1. Variation of compressive strength with the quantity of HG

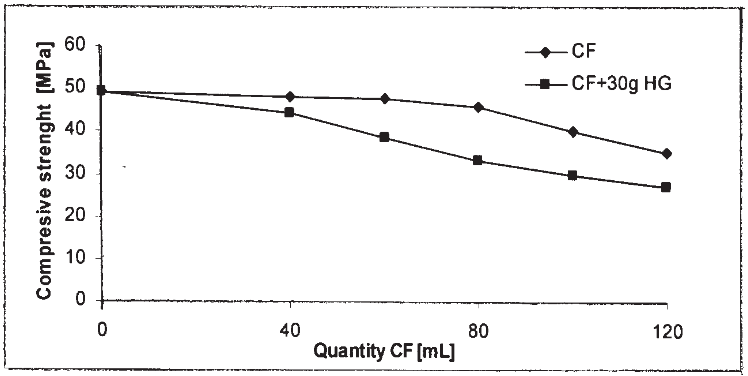

Fig. 2. Variation of compressive strength with the quantity of CF

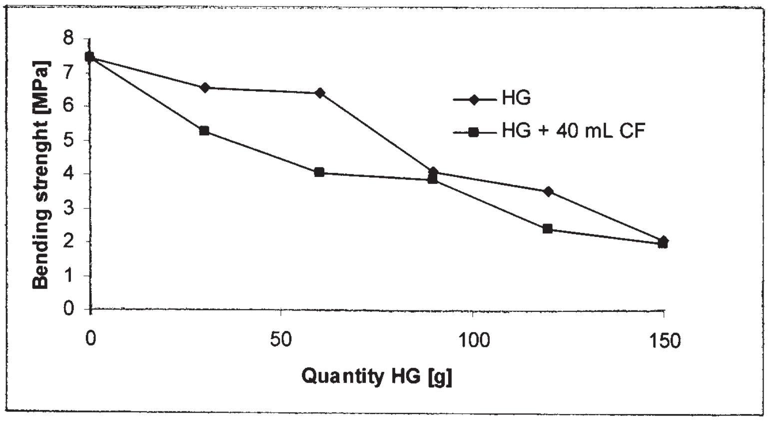

Fig. 3. Variation of bending strength with the quantity of HG

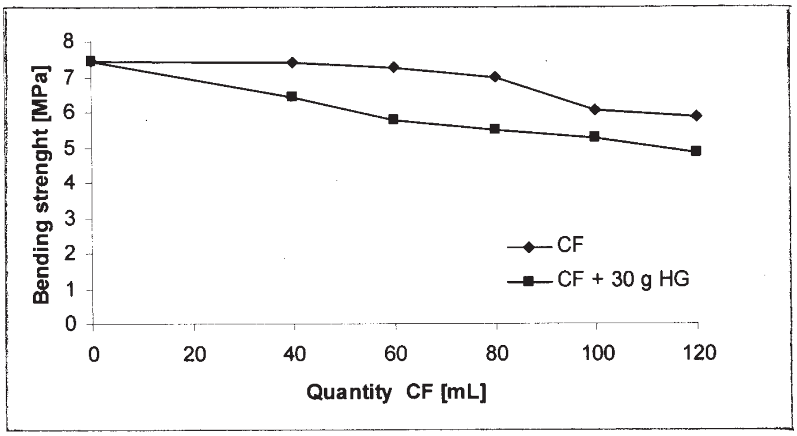

Fig. 4. Variation of bending strength with the quantity of CF

The setting time of the mortar paste is strongly dependent on the quantity of HG and it varies in the range of 3-6.8 h. The setting time is low dependent on the quantity of CF and it varies in the range of 2.5 -3 h. High concentration of HG leads to the increase of the setting time, mainly, due to the increase of the water quantity. Higher values of the setting time were noted for the samples 14, 15 and 16 with content of CF and content of 90g, 120g and 150 g of HG, mainly due to the high quantity of water. The increase of quantity of CF in the samples with the same content of HG leads to similar setting times as the ones observed for the samples with content of CF, but lower than the ones observed for the samples with a content of HG. For the cement the recommended setting time is ranging from 2 to 10 h.

The compressive strength is dependent on the concentration of HG and CF. High content of HG leads to the decreasing of the compressive strength of up to 3 times than the reference sample. A strong decrease in the compressive strength was obtained for samples 14, 15 and 16 with content of 90, 120 and 150g of HG.

The variation of compressive strength according to the quantity of HG is presented in figure 1.The CF content leads to a compressive strength of samples lower than the reference sample to 35.1 MPa for 120 mL CF, but higher than the samples with content of HG. The variation of compressive strength with quantity of CF is presented in figure 2. The lower values of compressive strength were noted for the samples with content of CF and HG.

The bending strength is dependent of the concentration of HG and CF. High content of HG leads to the decreasing of up to 2 times of the bending strength than the reference sample.

A strong decreasing of bending strength was obtained for samples 14, 15 and 16 with quantity of 90, 120 and 150g of HG.

The variation of bending strength with quantity of HG is presented in figure 3.

Increasing of CF content leads to a bending strength of samples lower than the values for reference sample to 5.85 MPa for 120 mL CF, but higher than the samples with content of HG. The variation of bending strength with quantity of CF is presented in figure 4.

Lower values of bending strength were noted for the samples with content of CF and HG as compared to the values obtained from the reference sample.

The compressive strength is not necessarily proportional with the bending strength, but for the studied samples a proportional variation of these two properties with the quantity of HG as well as CF can be noticed.

Conclusions

The mortar paste showed a good plasticity and workability except for the mortar paste where the content of HG was 90g, 120g and 150g. For these mixtures, supplementary water had to be added in order to improve the workability of the mortar paste. The setting time of the mortar paste is strongly dependent on the quantity of HG and it varies in the range of 3-6.8 h. The setting time is low dependent on the quantity of CF and it varies in the range of 2.5 -3 hours.

The compressive strength is dependent on the quantity of HG and CF. Increasing of HG quantity more than 14% leads to the decreasing of up to 3 times of the compressive strength than the reference sample.

The increasing of CF quantity to 120 mL does not affect significantly the compressive strength of the samples than the reference sample, recording 35.1 MPa for the maximum studied quantity of 120 mL CF. Adding up to 14% of HG to the samples with content of CF leads to not significant decreasing of the compressive strength of the samples.

The bending strength is dependent on the quantity of HG and CF. Increasing of HG quantity more than 14% leads to the decreasing of up to 2 times of the bending strength than the reference sample. Lower values of bending strength were noted for the samples with content of CF and HG.

The variation of compressive strength is proportional with the variation of bending strength for the studied samples.

Based on the conclusions on the mechanical tests it is recommended limitation of the quantity of HG up to 14% for preparation of both waste forms with HG content and HG and CF content.

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