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# CHEMICAL COMPOSITION OF RADIOACTIVE WASTE AND THE MECHANICAL PERFORMANCE OF CEMENTED MATRIX*

L. IONASCU, M. NICU, C. TURCANU, F. DRAGOLICI, GH. ROTARESCU

"Horia Hulubei" National Institute for Nuclear Physics and Engineering, P.O.Box MG-6, RO-077125 Bucharest-Magurele, Romania,
E-mail: laura_ionascu@nipne.ro; mnicu@nipne.ro; corneliuturcanu@yahoo.com;
E-mail: fdrag@nipne.ro; gheorghe.rotarescu@nipne.ro

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*Abstract*: The mechanical characterization of the radioactive waste conditioning matrix is very important during the final disposal stage in the radioactive waste management cycle. The conditioning products should be a monolith with acceptable mechanical, chemical and physical properties that are maintained over an appropriate time such that the release of radioactivity from the waste form in the environment is minimized. The stability of matrix – radioactive waste system is an essential condition to assure the radiological safety during the final disposal and is directly connected with physical-chemical reaction between the system components and structural modifications which lead to performance parameters imposed by the waste acceptance criteria in repository. The embedding of radioactive waste in OPC matrix is the most used method, applied in the world by the countries developing nuclear energy programs. The conditioned matrix of radioactive waste must have good mechanical properties to assure the material integrity during handling, storage, transport and long term stability in the final disposal environment.

*Key words*: radioactive waste, cemented matrix.

## 1. INTRODUCTION

Solidification of radioactive wastes with hydraulic cement either with or without admixtures has been practiced for many years. Water in the waste reacts chemically with the cement to form hydrated silicate and aluminum compounds, which normally contribute towards the setting and hardening of the cement mixture. However, the wastes produced by nuclear activities are very diverse and under certain circumstances affect the rate of hydration on cement and/or reduce

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the quality of product. The waste form must have properties that provide integrity during handling, storage, transportation and disposal and the long term stability of the waste form in disposal environments [1].

One of the most relevant mechanical properties applicable to cement based material is its compressive strength. High compressive strength corresponds to superior long term chemical stability of the waste form. The compressive strength is often used to evaluate the degradation that a sample undergoes after exposure to environmental condition such as water immersion or freeze-thaw cycling [2].

The technology used for the treatment of low level liquid radioactive waste in the Radioactive Waste Treatment Plant – Magurele Romania, is a multiple precipitation based on iron hydroxide and calcium phosphate formation. The conditioning method is the embedding in Ordinary Portland Cement.

The purpose of this paper is to obtain a part of the data basis necessary for the approach the medium and long term assessment of the safety and performance for final packages disposed in Radioactive Waste National Repository Baita-Bihor [3].

To obtain the experimental data necessary for the waste form and package characterization together with the back-filling material behavior in the repository environment a medium time programme (1997–2010) was implemented. The programme provides a deeper knowledge of species and mechanism, the structure, proprieties and performances of the processes and behavior of the products:

– Study of iron precipitates obtained during LLAW treatment [5];
– Cementation of sludge chemical components [7, 8];
– Durability of cemented waste in repository and under simulated conditions [6].

For safety reasons the behavior of waste packages, which is a main important barrier, must be properly studied in terms of long term durability in real repository conditions [4]. To prove the durability of cemented forms, an experimental programme started in 1997 to establish the time alteration of mechanical and physic-chemical properties of different simulated matrices in laboratory and Radioactive Waste National Repository Baita-Bihor disposal conditions [3].

## 2. EXPERIMENTAL PART

The basic information on the waste matrix is obtained by mechanical strength tests. The selected matrices for characterization are normal mortar composition as reference samples and the conditioned matrices prepared with non-active precipitate compounds (iron hydroxides and iron phosphate) simulating the real radioactive sludge.

All samples were positioned in five points (from 1 to 5) of Radioactive Waste National Repository Baita-Bihor and kept 1, 3, 5 and 7 years before mechanical characterization. The humidity and temperature conditions for these five points are presented in Table 1.

*Table 1*

Humidity and temperature conditions in the repository conditions

| No. | The point of disposal in real condition | Temperature [◦C] | Humidity [%] |
|---|---|---|---|
| 1. | Position 1 | 11 | 98 |
| 2. | Position 2 | 10 | 92 |
| 3. | Position 3 | 10 | 92 |
| 4. | Position 4 | 11.5 | 85 |
| 5. | Position 5 | 10 | 85 |

The results for samples disposal in real conditions will be compare with the results obtained for the same type of samples kept in laboratory conditions.

For Romanian Radioactive Waste National Repository Baita-Bihor the following steps are assumed for the conditioned waste matrix characterization in simulated and real conditions:

– preparation and mechanical characterization of reference cemented matrices:

- Ordinary OPC(OPC)+ water, with a cement/water ratio 2:1;

– preparation of iron precipitates such as to:

- addition of the neutralization agents, NaOH(20%), to the diluted solution of $FeCl_3$(40%);
- addition of $Na_3PO_4$ to the diluted solution of $FeCl_3$(40%);

– preparation and mechanical characterization of simulated sludge cemented matrices containing iron precipitates:

- OPC+ iron phosphate precipitate with cement / iron phosphate ratio 1:0.5;
- OPC+ iron hydroxide precipitate with cement / iron hydroxide ratio 1:0.5

– selection of real and simulated disposal conditions for cemented matrices;

– emplacement of cemented matrices in Radioactive Waste National Repository Baita Bihor for real disposal characterization.

After the preparation, the samples of cemented matrices were kept 24 hours in pattern, then in tight plastic bag for 7, 14 and 28 days for reinforcement.

After 28 days, the samples of cemented matrices were introduced in distillate water (laboratory conditions) and in the Radioactive Waste National Repository Baita-Bihor (real condition).

For the mechanical characterization tests the size of the samples was 20×20×20 mm (tested at compression).

The compression tests, performed on five samples, were done in a construction material laboratory using an digital press machine type MATEST, with loading rates 1.2 MPa/s.

## 3. DISCUSSION OF RESULTS

The compression tests were done on the samples of cemented matrices with size 20×20×20 mm. The samples were submitted to mechanical tests after 28 days and 1, 3, 5, 7, 10 years disposal in real and laboratory conditions.

Preliminary results showed that iron hydroxide and iron phosphate precipitate have a negative influence on the mechanical performances of cemented matrices.

In Table 2 are presented values obtained for the compressive strength on the cement: water matrices after 1, 3, 5 and 7 years of disposal in real and laboratory conditions.

*Table 2*

Compressive strength for cement-water matrices

| No. | Sample | $S_{comp.}$ (N/mm$^2$) | | | |
|---|---|---|---|---|---|
| | | 1 year | 3 years | 5 years | 7 years |
| 1 | (C:W-lab), Cement-Water, laboratory conditions | 28.3 | 51.3 | 61.0 | 64 |
| 2 | (C:W-1), Cement-Water, real conditions, position 1 | 39.5 | 55.3 | 63.5 | 69 |
| 3 | (C:W-3) Cement-Water,real conditions, position 3 | 43.5 | 59 | 72 | 73 |
| 4 | (C:W-5), Cement-Water, real conditions, position 5 | 37.6 | 56 | 62 | 66.7 |

The behavior in time of the compressive strength for cement-water matrices is presented in Fig. 1.

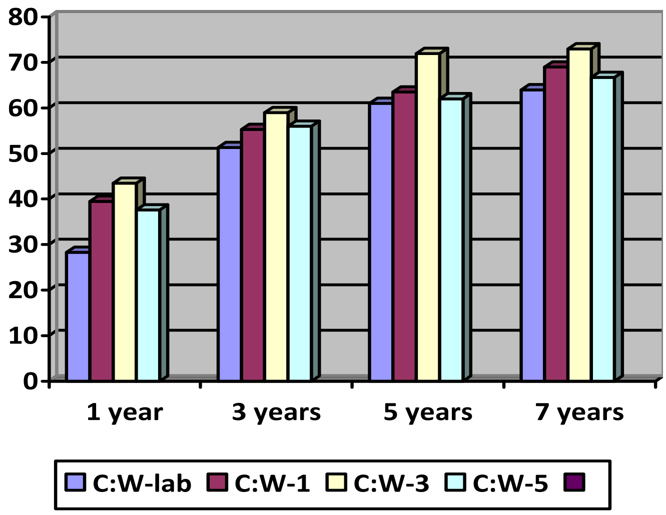


Fig. 1 – Behavior in time of the compressive strength for cement-water matrices.

In Table 3 are presented values obtained for the compressive test on the cement: iron hydroxide precipitate matrices after 1, 3, 5 and 7 years of disposal in real and laboratory conditions.

*Table 3*

Compressive strength for cement-iron hydroxide precipitates matrices

| No. | Sample | $S_{comp.}$ ($N/mm^2$) | | | |
|---|---|---|---|---|---|
| | | 1 year | 3 years | 5 years | 7 years |
| 1 | (C:Fe(OH)3-lab),<br>Cement-Iron hydroxide precipitate,<br>laboratory conditions | 23.41 | 25.4 | 29.1 | 37.6 |
| 2 | (C:Fe(OH)3-1),<br>Cement-iron hydroxide precipitate,<br>real conditions, position 1 | 25.3 | 27.45 | 31.37 | 54.95 |
| 3 | (C:Fe(OH)3-3),<br>Cement-iron hydroxide precipitate,<br>real conditions, position 3 | 26.3 | 28.65 | 33.5 | 42.32 |
| 4 | (C:Fe(OH)3-5),<br>Cement-iron hydroxide precipitate,<br>real conditions, position 5 | 24.2 | 23.53 | 28.0 | 30.5 |

The behavior in time of the compressive strength for cement-iron hydroxide precipitate matrices is presented in Fig. 2.

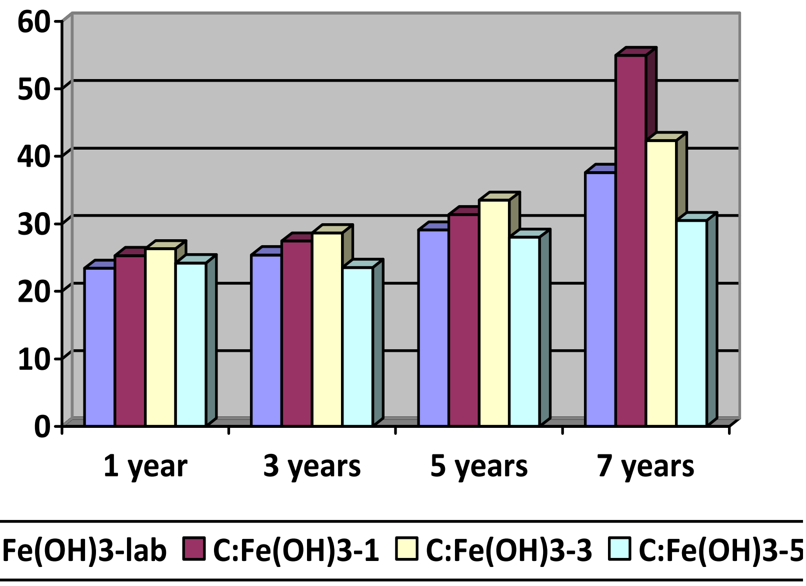


Fig. 2 – Behaviour in time of the compressive strength
for cement-iron hydroxide precipitate matrices.

In Table 4 are presented values obtained for the compressive test on the cement: iron phosphate precipitate matrices after 1, 3, 5 and 7 years of disposal in real and laboratory conditions.

*Table 4*

Compressive strength for cement-iron phosphate precipitates matrices

| No. | Sample | $S_{comp.}$ ($N/mm^2$) | | | |
|---|---|---|---|---|---|
| | | 1 year | 3 years | 5 years | 7 years |
| 1 | (C:FePO4-lab), Cement-Iron phosphate precipitate, laboratory conditions | 25.2 | 38 | 42.3 | 42.81 |
| 2 | (C:FePO4-1) Cement-Iron phosphate precipitate real conditions, position 1 | 29.3 | 39.5 | 45.37 | 52.64 |
| 3 | (C:FePO4-3) Cement-Iron phosphate precipitate real conditions, position 3 | 31.2 | 40.8 | 46.75 | 52.45 |
| 4 | (C:FePO4-5) Cement-Iron phosphate precipitate real conditions, position 5 | 35.7 | 43 | 48 | 54.2 |

The behavior in time of the compressive strength for cement-iron phosphate precipitate matrices is presented in Fig. 3.

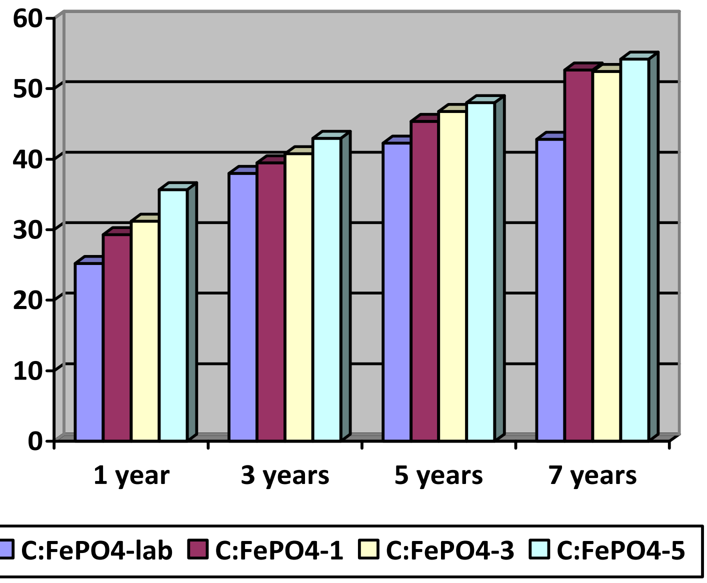


Fig. 3 – Behavior in time of the compressive strength for cement-iron phosphate precipitate matrices.

For presented cases we can observe, as was expected, the increasing of all resistance values for all examined samples. All resistance values are small different depending on disposal conditions.

Comparing the obtained results for the samples kept in real and simulated conditions, we can see that the mechanical resistance of the samples kept in real conditions is similar to those kept in laboratory conditions.

## 4. CONCLUSIONS

The compressive strength obtained for the samples which contain cement - iron hydroxide and cement-iron phosphate are smaller than the reference samples which contain cement-water but they are included in the accepted limits for the embedding matrix with cement (above $5N/mm^2$).

The compressive strength for system with iron precipitates increase in time and they are not influenced on the disposal conditions.

The results obtained by mechanical tests will be correlated with the structural modifications (after three, five and seven years of disposal).

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