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THE EFFECT OF GAMMA-RAY IRRADIATION ON MECHANICAL PROPERTIES OF EARLY-AGE CEMENT MORTAR

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ABSTRACT

This study is focused on experimental investigation of the effect of gamma-ray irradiation on early-age cement mortar properties. Several working hypotheses were formulated based on the current research review. The results of the performed experiment in terms of the relative weight and the relative length change are described in detail in this study. Two observations could be made from comparison of the working hypotheses with the obtained experimental results. Firstly, the positive effect of gamma-ray irradiation in terms of the relative weight change was observed within the first 300 hours or within the equivalent absorbed irradiation dose of 450 kGy. Secondly, the shrinkage due to gamma-ray irradiation was smaller for the unsealed samples due to presence of carbon dioxide in the air, which is needed for the carbonation reaction. It is believed that the obtained experimental data themselves provide a platform for validation of related numerical models.

KEYWORDS

Carbonation, Hydration, Microstructure, Mortar, Shrinkage

INTRODUCTION

It can be stated that the influence of gamma-ray irradiation on the hydration reaction of cement-based mortar is poorly understood. According to the current research [1, 2], the early-age mortar samples subjected to gamma-ray irradiation have a significantly reduced amount of micropores and subsequently they show an increase in strength and shielding properties. However, the number and the form of the available results are not enough to comprehend fully the role of gamma-ray irradiation in the hydration and microstructural formation process. Due to the lack of this understanding, the current knowledge of the effect of gamma-ray irradiation on the hardened mortar may be analyzed and extrapolated to the early-age mortar studies.

Regarding the hardened mortar, the peroxide [3], which is formed by the ionization radiation radiolysis, reacts with calcium present in the pore solution while the peroxide octahydrate is produced and decomposed due to its metastability with simultaneous carbonation reaction [4]. From this follows that the gamma-ray irradiation enhances dissolution/precipitation reaction and may heal the cement crystal structure [5]. Furthermore, according to [6], gamma-ray irradiation causes accelerated carbonation and increases the sample carbonate rate, which contradicts [5], where the carbonate rate is not changed, but, in contrast to the normal carbonation, gamma-ray irradiation can provoke formation of a significant amount of vaterite and aragonite instead of calcite, which under normal condition destruct CSH phases. The vaterite carbonation can be one of the key processes in the microstructure porosity decrease and the strength increase of the early-age mortar samples.

With consideration of the observations about the effect of gamma-ray irradiation on hardened cement mortar, five hypotheses related to early-age mortar behaviour were formulated. Then these hypotheses were tried to be confirmed with a new experiment aimed at the investigation of the effect of gamma-ray irradiation on the hydration reaction of the early-age mortar, which is described in detail in this paper.

OBJECTIVE AND WORKING HYPOTHESES

The assessment of the change in the hydration reaction process in terms of the weight and the length change of the early-age cement mortar samples under exposure to gamma-ray irradiation was the objective of the performed experimental investigation.

The following working hypotheses related to early-age mortar were assumed:

(1) The influence of gamma-ray irradiation reduces the amount of micropores [1, 7].

(2) The gamma-ray irradiation causes the peroxide formation and consequently accelerates the vaterite-carbonation reaction [5, 6].

(3) The gamma-ray irradiation induced carbonation and the drop in the microstructure porosity cause an increase of strength [1, 5].

(4) The dose of gamma-ray irradiation absorbed by the samples during the hydration process is too low to induce metamictization of minerals [8, 9].

(5) The alkali-silica reaction is not accelerated by the gamma-ray irradiation dose absorbed during hydration [8, 10].

EXPERIMENT

Mixture composition

The used mortar consisted of Cement type CEM I 42.5R, siliceous aggregate with fraction of 0-4 mm, water-cement ratio of 0.38. The mortar and the cement composition are shown in Tables 1 and 2, respectively. All samples were cured in sealed conditions for eight hours after mixing.

Tab. 1 - Composition of mortar

Component	Weight
Cement (CEM I 42.5R Mokrá, CZ)	2 625 g
Water	1 000 g
Siliceous aggregates with fraction of 0-4 mm	4 200 g

Tab. 2 - Composition of cement CEM I 42.5R Mokrá

Component	% by weight
Clinker composition	
C_3S	68.5
C_2S	11.6
C_3A	7.4
C_4AF	11.5
C_{free}	1.0
Cement components	
Clinker	95.0
Gypsum	3.5
Fly ash	1.3
Slag	0.2

Sample geometry

The shape of the sample with dimensions of 10x10x80 mm was selected so that the sample cross-section was small enough to ensure the gamma-heating dissipation.

Experimental sets

The insulated (sealed) and not insulated (unsealed) samples were considered in this experimental investigation so that the effect of drying on the hydration process could be assessed. The insulation was provided by a polyethylene foil. The following experimental sets were investigated:

- *IrIn* – irradiated insulated sample
- *IrNi* – irradiated not insulated sample
- *NrIn* – not irradiated insulated sample (reference sample)
- *NrNi* – not irradiated not insulated sample (reference sample).

Irradiation facility

^{60}Co Irradiation Facility UGU-420 of The Joint Institute for Power and Nuclear Research - Sosny of the National Academy of Sciences of Belarus, Minsk with the following irradiator characteristics: the height of the active part is 50 cm; the generating dose rate is 0.1-10 Gy/s and the total number of sources is 768 with the total activity of $4.4 \cdot 10^{15}$ Bq (120 kCi) was used in order to carry out the presented experimental investigation.

Irradiation

The sample irradiation started at the sample age of eight hours.

Measurements

The effect of gamma-ray irradiation was measured in terms of the weight and the length change of the samples. The insulation was removed from the samples for the dimension and the weight measurement. The environmental temperature and the relative humidity were also recorded. The radiation dose rate for the samples was equal to 3.72 kGy/h and 4.02 kGy/h for the experimental sets *IrIn* and *IrNi*, respectively, with the corresponding total absorbed radiation dose of $2.3 \cdot 10^3$ kGy and $2.6 \cdot 10^3$ kGy at the end of the experiment. The average duration of irradiation per day was 13 hours.

RESULTS

The changes of the relative weight and the relative length of the early-age cement mortar samples over time are shown in Figures 1 and 2, respectively. The changes of the environmental relative humidity and the temperature are shown in Figure 3. These results represent the complete package of information about the gamma-ray irradiation induced volumetric and weight changes and together with the effect of environment they can be used for validation of related numerical models.

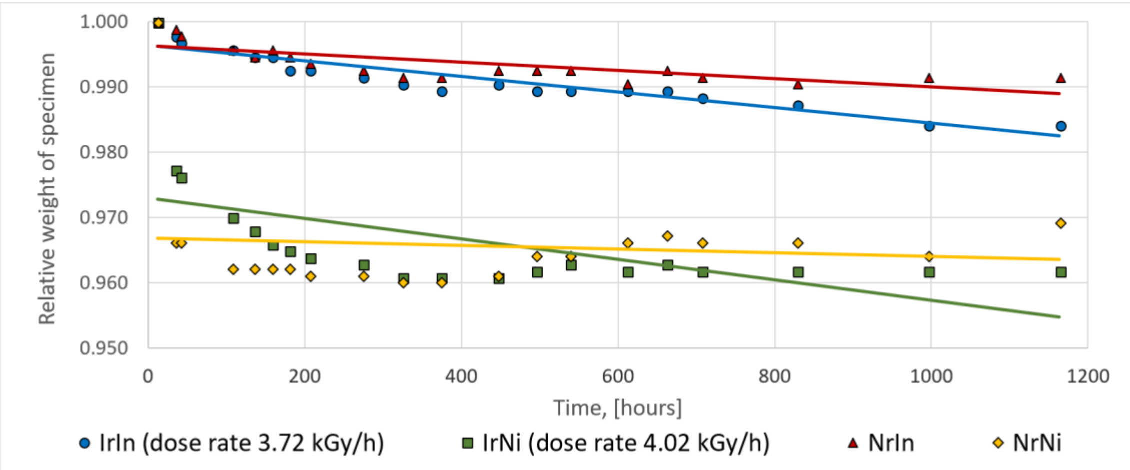

Fig. 1 – Change of relative weight of early-age cement mortar samples over time

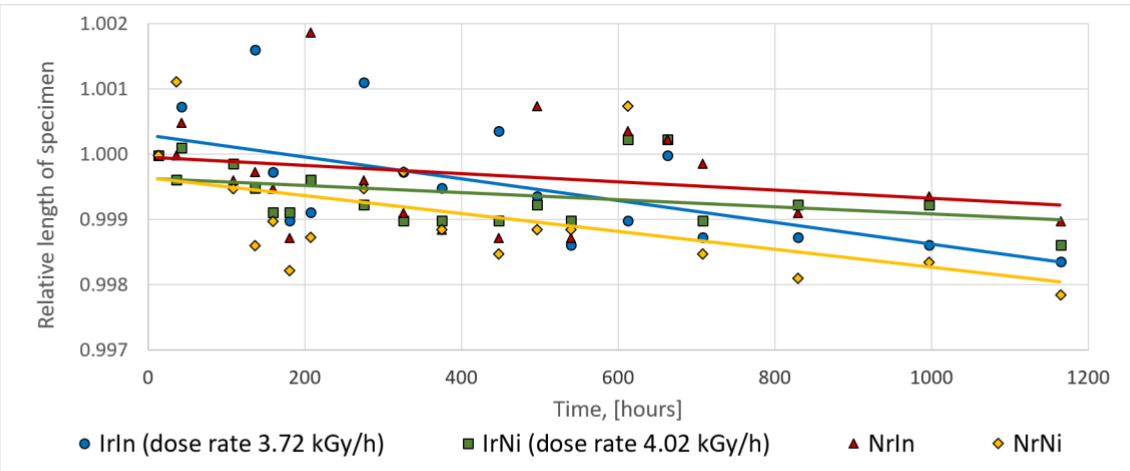

Fig. 2 – Change of relative length of samples over time

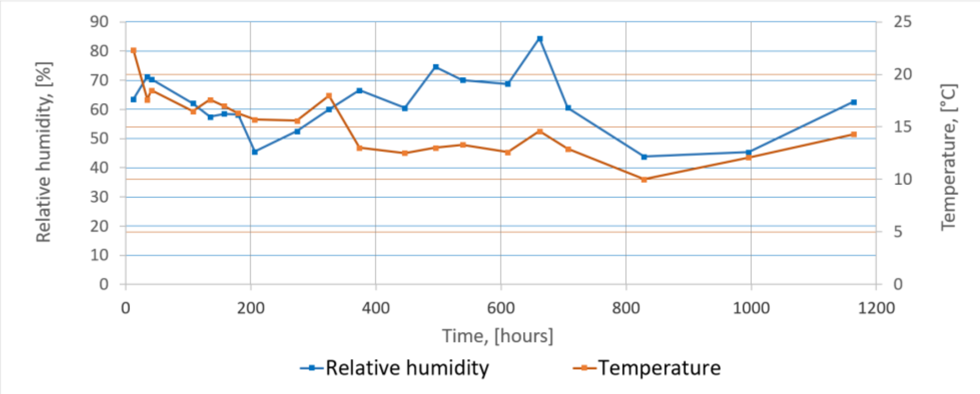

Fig. 3 – Environmental relative humidity and temperature

The weight of *IrNi* and *NrNi* samples decreased rapidly due to the drying, as can be seen in Figure 1. The slight increase of the weight of these samples after 500 and 1000 hours corresponded to the increase of the relative humidity in the laboratory, see Figure 3. The *IrNi* sample absorbed substantially less water than the *NrNi* sample. The decreased ability of the samples to absorb water from the environment is an indirect evidence of the drop in the microstructure porosity (working hypothesis (1)). A similar effect was observed in the relative length change, see Figure 2.

The higher weight of the *IrNi* sample during the first 300 hours (up to approximately 450 kGy in terms of the absorbed gamma-ray dose), see Figure 1, could be caused by the accelerated carbonation (working hypothesis (2)). After the first 300 hours, the *IrNi* sample had a higher weight loss than the *NrNi* sample, which means that the radiation-induced water decomposition with the subsequent gas evaporation became more important in terms of the weight loss. Presumably, the rate of carbonation reaction under the exposure to gamma-ray irradiation is much higher than the usual carbonation rate, but the reaction slows down after the absorption of a specific dose of radiation by the sample, which means that the difference in the carbonate rate of the irradiated and not irradiated samples decreases with time. This observation supplements the data presented in [5] and [6] without controversy.

The *IrNi* sample shrank less than the *NrNi* sample, see Figure 2, possibly due to the carbonation reaction, which reduced the shrinkage. However, the comparison of the *IrIn* and the *NrIn* samples showed the opposite trend. The irradiated samples (*IrIn* and *IrNi*) absorbed a different irradiation dose over time. However, the comparison of the relative length of these samples with dependence on the absorbed irradiation dose shown in Figure 4 had a similar trend with that shown in Figure 2. It means that the sample insulation affected negatively the irradiated early-age mortar due to the lack of carbon dioxide available for the carbonation reaction, which not only reduced the volumetric changes, but also could increase the strength of the samples (working hypothesis (3)).

The weight of the *IrIn* and the *NrIn* samples decreased much slower than that of the *IrNi* and the *NrNi* samples, as can be seen in Figure 1, however, the weight loss of the *NrIn* sample is rather high. This loss was caused by an instant drop in the relative humidity from 100\% to the ambient humidity due to the insulation removal, which was necessary in order to perform the measurements. Such a drop in the relative humidity caused the rapid free water evaporation.

The relative weight change of the *IrIn* sample in the first 300 hours was similar to that of the *NrIn* sample, see Figure 1. After the first 300 hours, the relative weight of the *IrIn* sample became smaller than that of the *NrIn* sample. This effect was similar to those in the *IrNi* and *NrNi* samples, but it was less pronounced. That confirmed both the accelerated carbonation during the first 300 hours (working hypothesis (2)) and the effect of the lack of carbon dioxide for the carbonation reaction due to the insulation.

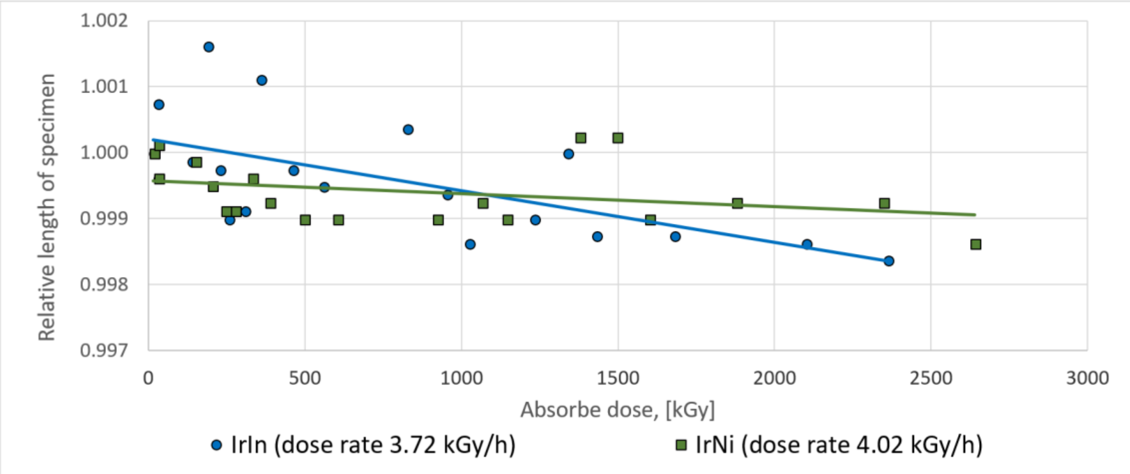

Fig. 4 – Change of relative length of samples with absorbed irradiation dose

The relative weight and the relative length of the *IrIn* sample reduced faster, see Figures 1 and 2, due to the summation the insulation removal effect and the radiation-induced water decomposition.

The *IrIn* and the *NrIn* samples swelled during the first several hours up to the several days, see Figure 2. This effect is common for concrete. The absence of the swelling of the irradiated samples after 28 days of the experiment is an indirect evidence that no metamictization or accelerated alkali-silica reaction started (working hypotheses (4) and (5), respectively).

CONCLUSION

The working hypotheses were confirmed indirectly, however the direct confirmations are still needed. Therefore, the experimental investigation of the sample microstructure, the chemical composition and the strength should be performed.

Two important observations have been made based on comparison of the working hypotheses and the experimental results. Firstly, the shrinkage due to gamma-ray irradiation was smaller for the unsealed samples. It is believed that this effect is associated with the unlimited source of carbon dioxide in the air, which is needed for the carbonation reaction. Secondly, the positive effect of the gamma-ray irradiation in terms of the reduced relative weight loss was observed within the first 300 hours, or within the equivalent absorbed radiation dose of 450 kGy.

It is believed that the obtained results along with the above discussion can provide a platform for validation of the numerical models which describe the effect of gamma-ray irradiation on the early-age cement properties.

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REFERENCES

[1] Rezaei-Ochbelagh D, Mosavinejad H G, Molaei M, Khodadoost M., 2010. Effect of low-dose gamma-radiation on concrete during solidification. International Journal of Physical Sciences, vol. 5: 1496-1500

[2] Burnham S., Huang L., Jevremovic T., 2016. Examining the Effect of Gamma Radiation Exposure in Early Stage of Concrete Curing on its Strength and Long-Term Durability. In: Proceedings of the 24th International Conference on Nuclear Engineering, edited by American Society of Mechanical Engineers, V005T15A068-V005T15A068, doi: 10.1115/ICONE24-60924

[3] Le Caër S., 2011. Water radiolysis: influence of oxide surfaces on H_2 production under ionizing radiation. Water, vol. 3: 235-253, doi: 10.3390/w3010235

[4] Bouniol P, Aspart A., 1998. Disappearance of oxygen in concrete under irradiation: the role of peroxides in radiolysis. Cement and concrete research, vol. 28: 1669-1681, doi: 10.1016/S0008-8846(98)00138-0

[5] Maruyama I, Kontani O, Takizawa M, Sawada S, Ishikawao S, Yasukouchi J, Sato O, Etoh J, Igari T., 2017. Development of soundness assessment procedure for concrete members affected by neutron and gamma-ray irradiation. Journal of Advanced Concrete Technology, vol. 15: 440-523, doi: 10.3151/jact.15.440

[6] Vodák F, Trtík K, Sopko V, Kapičková O, Demo, P., 2005. Effect of γ-irradiation on strength of concrete for nuclear-safety structures. Cement and concrete research, vol. 35: 1447-1451, doi: 10.1016/j.cemconres.2004.10.016

[7] Vodák F, Vydra V, Trtík K, Kapičková O., 2011. Effect of gamma irradiation on properties of hardened cement paste. Materials and structures, vol. 44: 101-107, doi: 10.1617/s11527-010-9612-x

[8] Ichikawa T, Kimura T., 2007. Effect of nuclear radiation on alkali-silica reaction of concrete. Journal of Nuclear Science and Technology, vol. 44: 1281-1284, doi: 10.1080/18811248.2007.9711372

[9] Maruyama I, Muto S., 2016. Change in relative density of natural rock minerals due to electron irradiation. Journal of Advanced Concrete Technology, vol. 14: 706-716, doi: 10.3151/jact.14.706

[10] Pomaro B., 2016. A review on radiation damage in concrete for nuclear facilities: from experiments to modeling. Modelling and Simulation in Engineering, vol. 2016, doi: 10.1155/2016/4165746