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NGS-Concrete — New Generation Shielding Concrete against Ionizing Radiation — the Potential Evaluation and Preliminary Investigation

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Concrete is a common material used as a shielding against ionizing radiation due to the relatively low cost and the ability to meet the structural function. State of the art in concrete shielding is that while in case of gamma radiation an increase in density by a change of aggregate is usually efficient enough, protection against neutrons is more complex. It is due to the differences in interactions of neutrons with the matter, depending on their kinetic energy and reaction cross-sections with the component atoms of the cement paste and the aggregate. Last progress in concrete evolution due to use of polymer additives (e.g. superplasticizers) together with reactive additions (e.g. silica fume) allows for a new look at the concrete design for radiation shielding purposes. One of the main advantages of concrete is its composite-type and there is a potential for the optimization of its constituents as well as mixture proportions. The paper presents the preliminary results of the project NGS-Concrete — new-generation shielding concrete against ionizing radiation. The aim of the project is to design the composition of concrete against ionizing radiation, achieved by the use of experiment based multi-criteria optimization of materials supported by the Monte Carlo simulations. The purpose of presented studies was to evaluate neutron shielding properties of ordinary and heavy-weight magnetite concrete modified with epoxy resin and gadolinium oxide. At first the shielding efficiency against neutrons from LWR neutron flux source and Pu–Be was simulated in MCNP code. At the end the comparison of MCNP simulated results and real experiment was presented. It was proved that both methods of modification can improve neutron shielding properties concrete but gadolinium oxide is an efficient additive only for low energy neutron attenuation.

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1. Introduction

The NGS-Concrete (new generation shielding concrete against ionizing radiation) is a research project that aims to design a novel composition of concrete against ionizing radiation by the use of experimental approach based on multi-criteria optimization of materials and supported by the Monte Carlo simulations. Shielding concrete should be designed and fabricated in a particular way in order to meet all the requirements, not only as an effective ionizing radiation shielding but also to maintain its original strength and durability against pressures and tensions. The aim of the project in terms of material optimization was to perform some modifications in the composite at the atomic level including using fillers containing the atoms that are able to scatter and attenuate the neutron radiation. It was assumed that these modifications will improve the neutron shielding properties without compromising other mechanical features of the concrete.

Studies have shown that while in case of shielding against gamma radiation, an increase in density of a concrete by a change of aggregate type causes usually efficient protection, providing shielding against neutrons could be more complex and sophisticated task [1]. It is due to the differences in interactions of free neutrons of different energies with the matter which depends strongly on the reaction cross-sections for different atoms of the cement paste and the aggregate. Nevertheless, some general recommendation have been already stated, such as increase of hydrogen content in concrete (more chemically bounded water) or addition of effective neutron absorbers (boron) [2]. Akkurt and Elkhayat [3] stated that the most effective shielding material for mixed neutron and gamma-rays is obtained by mixed hydrogenous materials, heavy metal elements, and other neutron absorbers. Inelastic scattering by heavy elements and elastic scattering by hydrogen are quite effective to slow down fast and intermediate energy neutrons, and the absorbers can reduce secondary gamma-rays as well as thermal neutrons. Thus the evaluation process for the effectiveness of a shielding material must include, beside other parameters, its ability to attenuate gammas and neutrons in other research.

Preliminary studies of Piotrowski et al. [4] were based on Monte Carlo computer simulations and it was indicated that both the compressive strength and type of concretes (normal and heavy-weight) have a significant effect on the shielding against neutron. It was concluded that the decrease in the neutron dose detected behind the concrete shielding was proportional to the concrete

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strength and an increase in the cement content in the mixture and the associated mass of bound water as a result of its hydration. Concerning the density, the results of the simulations for mono-energetic neutrons with energies 1 MeV and 10 MeV showed that heavy-weight barite concrete was inferior to the weakening of radiation up to a thickness of approximately 30–40 cm, when a change in this relation was found. In the same paper it was found, however, that the type of cement with different chemical composition did not significantly affect the neutron shielding properties.

The other results were presented by Gallego et al. [5] who analysed magnetite-based concrete and found its advantageous behaviour, when comparing neutron attenuation from ^{241}Am–Be source against the thickness of the shielding with regard to ordinary concrete.

The possibility of improving neutron shielding properties of concrete by polymer addition has already been presented in 70's by Belyakov et al. [6, 7]. New approach in this field was presented by Malkapur et al. [8] in study on the effect of mix parameters and hydrogen loading on the neutron radiation shielding characteristics of latex modified concrete mixes. Here an increasing trend of total neutron cross-section and decreasing trend of dose transmission values with increased hydrogen loading was observed in comparison with control concrete with a few exceptions.

The epoxy-cement concretes are the most common practical example of using of polymer cement concrete (PCC). The aim of modification by epoxy resin is the same as in the case of other types of PCC but degree of improvement is usually higher. Following Czarnecki and Łukowski [9] the chemical resistance of epoxy-cement concrete is better (also to acids), and the compressive strength — in opposition to the other types of PCC — is not worsened. The tensile strength and flexural strength can be twice higher, and abrasion resistance even three times higher than that of unmodified concrete. The other advantages of epoxy-cement concrete is an increase of tensile strength and adhesion, improvement of tightness, decrease of modulus of elasticity, improvement of frost resistance. It was found also by that PCC with 20% epoxy can effectively reduce overall deterioration of concrete especially those exposed to sea water [10]. Regarding shielding properties in a research on cement based mortars it has been demonstrated a significant benefit on neutron shielding properties of epoxy polymer addition as the PCC repair mortars exhibited superior properties in terms of both thermal neutron absorption and fast neutrons attenuation [11].

2. Materials and methods

The basic concrete for analyses was an ordinary concrete ($w/c = 0.4$) composed of CEM I 42.5R and river gravel and sand of a density 2.65×10^3 kg/m^3. To improve gamma shielding properties volumetric change of coarse aggregate type to a magnetite crushed aggregate with a density of 4.8×10^3 kg/m^3 was proposed. The composition of reference concretes are presented in Table I and their atomic compositions in Table II.

TABLE I

Reference concrete composition.

Component	Ordinary concrete (OC)	Magnetite concrete (MC)
cement CEM I 42.5 R	380	380
coarse aggregate	1345	2610
sand (0/2)	576	480
water	152	152
w/c ratio	0.4	0.4

TABLE II

Reference composition of normal concrete (first line) and magnetite concrete (second line).

	Mass	Rel. cont.	Element H	O	Na	Mg	Al	Si	S	Cl	K	Ca	Fe
			M [g/mol]										
	$\left[\frac{kg}{m^3}\right]$	[%]	1.0	16.0	23.0	24.3	27.0	28.1	32.1	35.5	39.1	40.1	55.8
cem.	380	15.49		5.80	0.02	0.14	0.46	1.51	0.22	0.01	0.09	7.69	0.37
	380	10.48		3.73	0.02	0.09	0.29	0.97	0.14	0.01	0.06	4.95	0.24
aggr.	1921	78.31		40.7	1.22	3.98	6.54	27.0			0.66		2.37
	3090	85.31		28.3	0.17		0.18	7.98	0.02		0.13	1.35	49.6
H_2O*	152	6.20	0.14	1.10									
	152	4.20	0.1	0.75									
tot.	2453	100	0.14	47.6	1.25	4.11	7.0	28.5	0.22	0.01	0.75	7.69	2.74
	3622	100	0.1	33.1	0.18	0.09	0.48	8.87	0.17	0.01	0.18	6.54	49.8

*only 20% of water mass was assumed as a water kept (bounded) in concrete.

The aim of material modification was to use the additives that could improve the neutron shielding properties — gadolinium and epoxy resin. Gadolinium has the highest thermal neutron capture cross-section among the known elements. Additionally, metallic gadolinium is relatively stable in dry air, but it tarnishes quickly in moist air, forming a loosely adhering gadolinium(III) oxide: $4Gd + 3O_2 \rightarrow 2Gd_2O_3$. In the current study a fine white powder (Fig. 1) similar to a hydrated lime was used. The dosage of fine gadolinium (99.5% Gd_2O_3) powder was varied from 1% up to 5% as it is in case of other metallic oxide, TiO_2, used for photocatalytic purposes.

The epoxy resin is a typical polymer composed of carbon (76%), oxide (17%), and hydrogen (7%) atoms. The addition of epoxy resin makes a significant increase of hydrogen content in atomic composition which is favourable from neutron shielding point of view. The addition of epoxy resin was 10% in relation to cement as it is assumed that bigger dosage can decrease a hydration of a cement and in a result decrease of a compressive strength of concrete.

The Monte Carlo neutron transport simulation was performed with Monte Carlo N-Particle Transport Code System, MCNP 6.0. In case of MCNP, the standard library, also used in this work, is ENDF/B VI. A random number generator sample provided experimentally evaluated cross-sections for different reactions. Standard procedures called *tallies f1* — current integrated over a surface and *tallies f2* — flux averaged over a surface [12] were

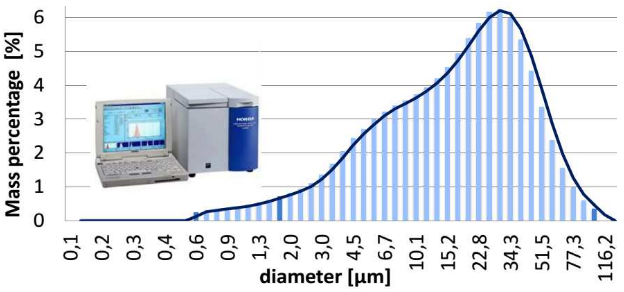

Fig. 1. Laser granulometry result for a gadolinium powder used in research.

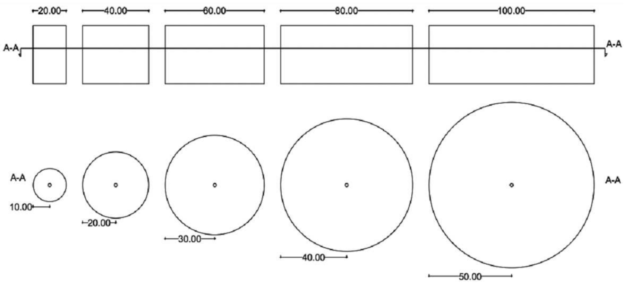

Fig. 2. Laser granulometry result for a gadolinium powder used in research.

used. Every simulation was performed for 1×10^7 neutron histories to get fair statistical properties. Finally the statistical result were available and relative error was less then 10^{-4}. All tallies passed the 10 statistical checks for the tally fluctuation chart bin result. The simulated geometry was the cylinders of thickness 10, 30, and 50 cm in radius dimension with a hole inside (Fig. 2). The source was simulated inside the sample and the outer surface was treated as a detector.

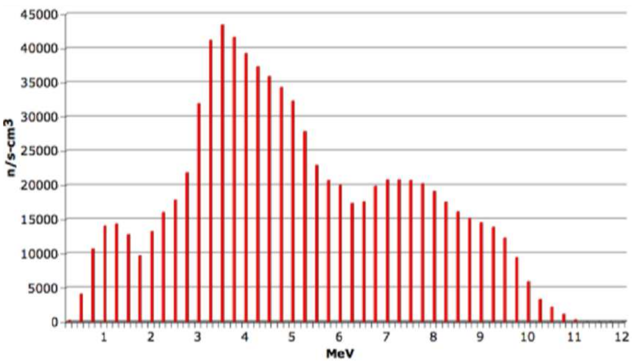

Fig. 3. Energy histogram of a Pu–Be source used in MCNP calculations.

First MCNP simulations were performed for a neutron flux from a light water reactor (LWR) core with total flux of 4.64×10^9 n/(cm^2 s) calculated using APOLLO 2 [4]. By the use of this neutron source first the influence of gadolinium oxide content in 1, 2 and 5% in relation to cement was analysed by the comparison of neutron flux on the different levels of energy counted in the detector. Then there was analysed an effect of introduction of 3% of gadolinium oxide and 10% epoxy resin on the effective doses calculated for a human body behind a shielding. The synergy effect was also taken into account. As in the experiment the neutron source was a Pu–Be and in final MCNP calculations for linear attenuation coefficient estimation the same source and the real geometry of experiment [11] were taken as a input data. The energy histogram used in MCNP simulations was taken from Harvey [13] as presented in Fig. 3.

To validate the MCNP simulation results, an experiment was conducted to measure the degree of absorption of thermal neutrons in the concrete of a certain thickness. The details of experimental test setup was described in studies of Piotrowski et al. [11]. The comparison was made on the base of linear attenuation coefficient [14, 15] μ (cm^{-1}) obtained by

$$\mu = \frac{1}{x} \ln \frac{I_0}{I}, \tag{1}$$

where x is the material thickness and I and I_0 are the background subtracted number of counts recorded in detector with and without concrete material between detector and source, respectively.

3. Results and discussions

The first MCNP simulations were performed for a neutron flux from a light water reactor (LWR). Figure 4 presents the results as a neutron flux behind concrete shields of different thickness composed of an ordinary concrete and the one modified by 1, 2, and 5% of gadolinium oxide. It is clearly visible that gadolinium is very effective in small thickness (10 cm) in almost all thermal neutron range (up to 1×10^{-1} MeV) but for bigger thickness the energy border of effectiveness is moved to smaller energies (for 30 cm thick — 1×10^{-4} MeV and for 50 cm thick — 2×10^{-7} MeV). Another observation is that increasing gadolinium content from 1 to 5% in relation to cement does not make an important progress in neutron shielding efficiency.

Next MCNP simulations performed for the same neutron flux shows effective and relative doses calculated for a human body behind concretes modified by epoxy resin and gadolinium oxide. The results of effective dose in Fig. 4 show that both modifications are effective. In a relative (to non-modified magnetite concrete) dose graph presented in Fig. 5 there is visible a slight advantage of 10% of epoxy over 3% of gadolinium oxide addition and the synergy effect of both modifications is not observed.

The last results are the thermal neutron linear attenuation coefficients calculated using MCNP and obtained in real experiment when a Pu–Be neutron source was used.

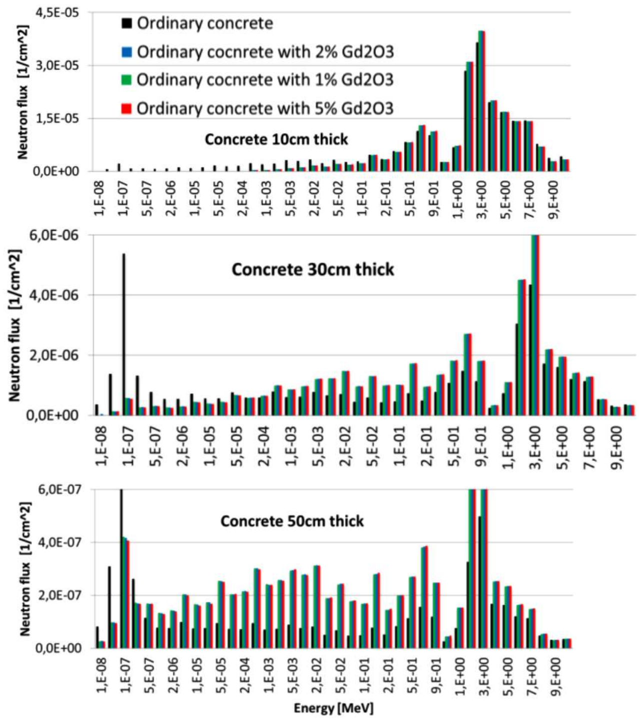

Fig. 4. PuBe neutron flux behind ordinary concrete shields of different thickness modified by the gadolinium powder (Gd_2O_3).

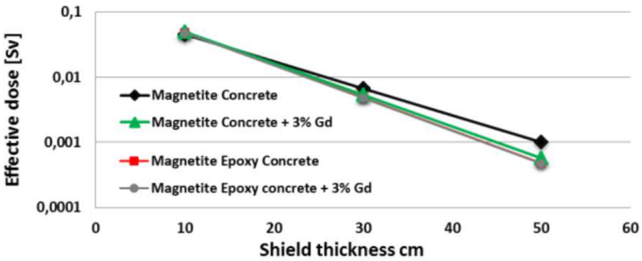

Fig. 5. Effective dose behind magnetite concrete and magnetite concrete modified by epoxy and gadolinium.

They were obtained for standard cement mortar based on CEM I 42.5 R, ordinary concrete, ordinary concrete modified by 5% of gadolinium oxide and magnetite concrete as well (for results see Fig. 6). A good agreement of MCNP simulation with experiments is clear (Fig. 7). The advantage of magnetite concrete was also observed, however unexpectedly the effectiveness of gadolinium additive in neutron shielding was not detected by MCNP results.

4. Conclusion

Based on the presented results the two basic conclusions can be drawn:

- concerning the gadolinium: it is an efficient additive for low energy neutron attenuation but it is useless from the fast neutron shielding point of view — no decrease of flux was observed for all thickness;

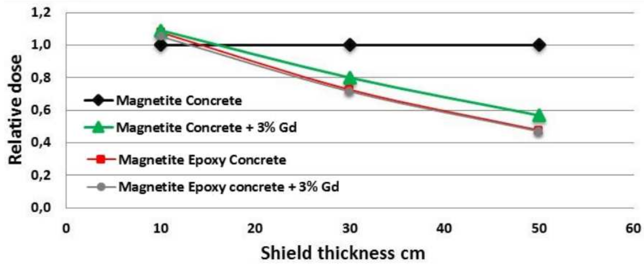

Fig. 6. Relative dose behind magnetite concretes modified by epoxy and gadolinium.

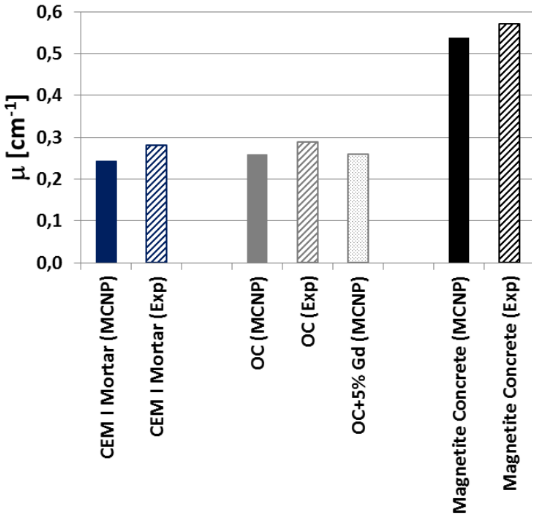

Fig. 7. Thermal neutron linear attenuation coefficient calculated from MCNP simulations and resulting from experiments for different types of shielding concretes.

- concerning the epoxy resin: it improves shielding properties of mortars and concrete against neutron radiation, addition of such polymer into heavy-weight concrete (used against gamma radiation) can improve its usability properties and neutron shielding efficiency as well.

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