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Importance of atomic composition and moisture content of cement based composites in neutron radiation shielding

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Abstract

Monte Carlo computer simulations confirmed that an increase in density has a minor effect on the weakening of neutron transport and, therefore, the optimum composition of a shielding concrete against gamma radiation is different than the one against neutron radiation. Neutron radiation shielding is a two-step process: slowing down of fast neutrons and absorption of thermal ones. Both result from the atomic composition of the barrier but their dependence on specified atomic compositions and moisture content is different. The aim of the presented research is to develop a high density concrete the composition of which would also assure good efficiency of neutron shielding. Neutron transport through standard cement mortar, PCC mortar, normal-weight concrete and magnetite heavy-weight concrete has been analyzed in the paper. The goal in research was to find an influence of the cement type, polymer addition, density and moisture content on the shielding properties against neutron. The research based on convergent results of MC computer simulations and real experiments confirmed the influence of the cement type on fast neutron attenuation. It was also found that each 1% of moisture content makes 10% increase of fast neutron thermalization effectiveness, what is a little less than it was estimated for cement based mortars. It was also proved that heavy-weight concrete is not proper solution for shielding against fast neutrons, but its efficiency is visible in the case of thermal neutrons absorption, probably due to increase of Fe content at the expense of Si and O in the atomic composition as well as water retained by magnetite aggregate.

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1. Introduction

Previous studies by Piotrowski et al. [1] and Tefelski et al. [2] based on computer simulations using MC method indicate that both the type of concrete (normal and heavy-weight) and its compressive strength have a significant effect on the shielding against neutron. The decrease in the dose is proportional to the strength, apparent increase of cement content and the associated mass of bound water as a result of its hydration. There was presented a true advantage of the heavy-weight concrete over a normal-weight concrete in radiation beam corresponding to the reactor, but it was observed some disturbance in the thickness of approx. 25 cm. The results of the simulations for mono-energetic neutrons with relatively high energies 1 MeV and 10 MeV have shown that a heavy-weight concrete was inferior in the weakening of radiation to a thickness of approx. 30-40 cm, when a change of the trend was found. Additionally, it was concluded that the type of cement did not affect the neutron shielding properties significantly. The next step of these analysis is to perform both computer simulations and experimental studies on influence of specific atomic composition of cement based composites and moisture content on neutron radiation shielding. Results of this analysis are presented in the paper.

Nomenclature	e	
CBC	Cement Based Composites	
MC	Monte Carlo	
XRF	X-Ray Fluorescence	
G-NWC	Granite Normal-Weight Concrete	
M-HWC	Magnetite Heavy-Weight Concrete	

2. CBC for neutron radiation shielding

The most harmful ionizing radiation for human beings is gamma radiation. Recently more often sources emitting neutrons are used as well (e.g. fast neutron therapy, fast neutron rector). It creates the need for concrete shields that are efficient enough for both gamma rays and neutrons. The main problem is a fact that while in case of protection against gamma the increase of density e.g. by the use of heavy-weight CBC, is usually sufficiently effective (but not so economic), for the protection against neutron this solution is insufficient. It is due to the fact that the interaction of the neutrons with the member depends on the energy of neutrons and the probability for given reactions of neutrons with nuclei of atoms in the atomic composition of material. That is why El-Khayat and Akkurt [3] confirmed that the use of barite as a heavy aggregate does not give an improvement of shielding properties of concrete against neutron radiation. In other paper, Akkurt and El-Khayat [4] optimized the barite proportion for simultaneous protection against neutrons and gammas as 53.8%. Moreover, they have 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 In other calculations by Akkurt et al [5] it has been found that marble improved concrete shielding properties against gamma rays but it is not ideal material to improve shielding properties against neutron radiation. Gallego et al. [6] already presented Hormirad TM - a high density concrete composed of magnetite aggregate with boron compounds, which has shown a clearly advantageous behavior, when comparing neutron attenuation against the thickness of the shielding with regard to ordinary concrete. The major importance in the neutron transport through CBC can be attributed to the collisions of neutrons with the nuclei of atoms of the member. A measure of the probability of occurrence of the given reaction (collision) of neutrons with the nucleus is named *cross section*, defined as the effective area in which the neutron must hit to produce a specific phenomenon. As the cross section for absorption of fast (high energy) neutrons is rather small, in order to get the neutron flux weakening by CBC, it should be optimized so that the atoms of which it is composed: first caused precipitation of energy by attenuation in the processes of elastic and inelastic scattering, and then absorption of slow (thermal) neutrons. Protection against neutron radiation is therefore a two-step process (1-attenuation, 2absorption). Attenuation of fast neutrons is mainly due to elastic collisions with the nuclei of light elements (like hydrogen) and inelastic scattering on nuclei of heavy elements, for which the energy of the lowest excited state is

much less than for light elements (e.g. Pb - 0.4 MeV, O - 6.0 MeV). From the 60's it is assumed that the concrete containing 8-10% of water (free and chemically bound) with the possible addition of boron silicate (about 70 kg/m³) and thickness necessary to shield the gamma radiation from reactor will also be effective enough for neutron radiation, Pohl [7].

2.1. Calculation of atomic composition of concrete

Portland cement is a hydraulic mineral binder derived from mineral raw materials - marl, limestone, clays and iron ore mixed with gypsum after burning. Marl is a sedimentary rock composed of calcium and magnesium carbonates and clay minerals (minerals with the chemical composition of hydrated aluminum silicates of aluminum, magnesium and iron); limestone is a sedimentary rock composed mainly of calcium carbonate and the clay is a sedimentary rock, mainly composed of clay minerals, quartz and feldspar. The chemical composition of cement can be presented in the oxide form of chemical compounds. To calculate the atomic composition of cement it is necessary to know the atomic mass of each element and to calculate the molecule, how many atoms of a given element contains a molecule and what are the atomic weights of the individual elements? Then, knowing from results of measurements (e.g. XRF) the percentage of each component in cement, e.g. SiO₂ = 18.9% in Lafarge CEM I, and corrected content in order to receive a total 100% (resulting 19.7%), one calculates the percentage of each element (multiplied by the quantity in the molecule) by the atomic mass of the whole molecule and multiply by the percentage of particles in the cement. An example of such calculation to obtain the amount of the oxide expressed in percentage is presented in equation 1.

$$\%O = \frac{2 \cdot 15.99}{2 \cdot 15.99 + 28.08} \cdot 19.7\% = 10.5\%$$
(1)

By an analogy, calculations with other molecules are proceeded. To calculate the total amount of a given element in the cement one should sum up the amount of all the molecules in which it appeared (see Tab.1).

Cement CEM I	XRF Content	Corrected content	Atom	Si	0	Al	Fe	Ca	Mg	S	K	Na	Cl
component	[%]	[%]	Atomic mass [g/mole]	28.1	16.0	27.0	55.8	40.1	24.3	32.1	39.1	23.0	35.5
SiO ₂	18.9	19.7	60.1	9.2	10.5								
Al ₂ O ₃	5.1	5.3	102.0		2.5	2.8							
Fe ₂ O ₃	3.1	3.3	159.6		1.0		2.3						
CaO	63.2	66.0	56.1		18.8			47.2					
MgO	1.3	1.4	40.3		0.6				0.8				
SO_3	3.2	3.4	80.1		2.0					1.4			
K_2O	0.6	0.6	94.2		0.1						0.5		
Na ₂ O	0.2	0.2	62.0		0.1							0.1	
Cl	0.1	0.1	35.5										0.1
Total	95.7	100.0		9.2	35.6	2.8	2.3	47.2	0.8	1.4	0.5	0.1	0.1

Table 1. Example of atomic composition of Portland cement.

Those and other molecules are also presented in the aggregate, additives, admixtures and water (here not all the water from w/c ratio but only 20% of it is assumed – that is the chemically bounded water in cement hydrates). The percentage composition of the individual component of concrete should be determined. Then proceed similarly to the method described above, the amount of the atoms can be calculated by multiplying the percentage of the component of the concrete in which the element is located (see Tab.2).

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Compone nt	in m ³	Relative content	Atom	Н	С	0	Na	Mg	Al	Si	S	Cl	K	Ca	Fe
	[kg]	[%]	[g/mole]	1.0	12.0	16.0	23.0	24.3	27.0	28.1	32.1	35.5	39.1	40.1	55.8
CEM I	380	15.5				5.80	0.02	0.14	0.46	1.51	0.22	0.01	0.09	7.69	0.37
	380	10.5				3.73	0.02	0.09	0.29	0.97	0.14	0.01	0.06	4.95	0.24
Aggregate	1921	78.3				40.7	1.22	3.98	6.54	27.0			0.66		2.37
	3090	85.3				28.3	0.17		0.18	7.98	0.02		0.13	1.35	49.6
Water	152	6.2		0.14		1.10									
H_2O^*	152	4.2		0.1		0.75									
Total	2453	100,0		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		0.1	_	33.1	0.18	0.09	0.48	8.87	0.17	0.01	0.18	6.54	49.8

Table 2. Atomic composition of G-NWC (0.4) - black color, and M-HWC (0.4) - grey color.

*Only 20% (by mass) of added water was assumed as a water bounded in concrete.

The research was made on granite normal-weight concrete (G-NWC) with different w/c ratios (0.4, 0.5 and 0.6) and magnetite heavy-weight concrete (M-HWC) with w/c =0.4 and the same volume composition of aggregate as G-NWC. The particle size distribution of granite aggregate with sand was optimized to obtain minimum cavity. The composition was 30% of sand, 28% of granite 2/8 and 42% of granite 8/16. The composition of magnetite aggregate and sand was fixed to obtain the same level of sieve curve (30.1% - fraction 0/2; 27.9% fraction 2/8; 41.8% fraction 8/16). This solution eliminates the influence of granulometry on the results for both mechanical properties and radiation protection. For comparison, the results for a sample of standard cement mortar CEM I according to EN 196-1 and PCC mortar from Piotrowski and Skubalski [8] are presented. The compressive strength of concrete was evaluated and concrete from C20/25 to C40/50 was obtained. Interesting is that only M-HWC fulfills the requirements for structural concrete (min C40/50) presented in ETC-C edition 2010[†] [9]. The characteristics of CBC samples is presented in Table 3.

Table 3. Characteristic of CBC sample	les.
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Symbol	aggregate	Cement CEM I	w/c	Water [kg/m ³]	Density in dry-air state [kg/m ³]	f _{ci,min} [MPa]	f _{cm} [MPa]	Compressive strength class
G-NWC (0.4)	crushed granite ($\delta = 2.65 \text{ kg/m}^3$)	380	0.4	152	2380	46.5	49.9	C 35/45
G-NWC (0.5)	$(\delta = 2.65 \text{ kg/m}^3)$ + river sand $(\delta = 2.65 \text{ kg/m}^3)$	380	0.5	190	2352	38.5	40.3	C 25/30
G-NWC (0.6)	1921 kg/m ³	380	0.6	228	2354	34.4	37.2	C 25/30
M-HWC (0.4)	crushed magnetite ($\delta = 4.80 \text{ kg/m}^3$) 2610 kg/m ³ + river sand ($\delta = 2.65 \text{ kg/m}^3$) 480 kg/m ³	380	0.4	152	3608	55.1	56.9	C40/50
Mortar CEM I	CEN Standard sand acc. to EN 196-1 1350 g*	450 g*	0.5	225 g*	not measured	33.6	36.2	Cement class 32.5
PCC Mortar	CEN Standard sand acc. to EN 196-1 1350 g*	450 g* +10% polymer	0.5	225 g*	not measured	40.0	42.1	PCC Repair Mortar R4

* for preparation of the standard mortar according to EN 196-1

⁺ In ETC-C edition 2012 and RCC-CW 2015 this requirement has been changed for durability requirements in accordance to exposure class

2.2. Importance and calculation of moisture content

Taking into account the specific parameters influencing neutron radiation shielding, hydrogen content is one of the main factor influencing thermalization of fast neutrons. Of course, moisture content is one of the main source of hydrogen in a shielding CBC, but the other aspects for keeping the hydrogen in the atomic composition should be analyzed as well. The water can be retained both in aggregate and cement matrix. In the previous studies presented by Piotrowski and Skubalski [8] it was found that cement type, despite the similar chemical atomic composition, is influencing results of fast neutron attenuation. Experimentally slowing down of fast neutrons by cement mortars with CEM I was by 70% more effective than by mortars with CEM II and CEM IV, probably due to difference in chemically bound water in the hydrates. It is also shown that each 1% increase of moisture makes around 15% increase of effectiveness of fast neutron thermalization. The next source of hydrogen in CBC could be aggregate. It is recommended to use aggregates with high percentage of bound water like serpentine, Kharita et al [10], or peridotite, Okuno et al. [11]. The problem could be due to the fact that gamma and neutron radiation lead to decrease of technical properties (e.g. compressive strength) and loss of water as well. This effect, presented in detail by Brandt [12] and Brandt and Jóźwiak-Niedźwiedzka [13], makes concrete less usable and less effective in shielding with time. ANSI/ANS-6.4-2006 [14] addresses the loss of free water from concrete to heating and its effects on shielding. It is stated that the effectiveness of concrete neutron shields can be easily calculated using modern radiation transport codes only if the degree of concrete dehydration can be estimated.

The moisture content in specimens was measured according to the procedure for water absorption measurement described in Polish Standard PN-B-06250:1988. Moisture content is calculated as a mass of water in the sample in relation to a mass of totally dry sample that was fixed by putting it into the 105°C until its mass is constant (difference in 24h interval less than 0.2%).

3. MC simulations

For neutron shielding problems, the Boltzmann transport equation for neutrons should be resolved, but it cannot be solved analytically unless a lot of simplifying assumptions are made. One of the simplest method for solving it is a use of the computer MC methods developed by Metropolis and Ulam [15] during their work in Los Alamos laboratory. It is a numerical method of multiple sampling of a neutron history and the final result is obtained statistically. A simplified algorithm of the MC neutron transport simulation was presented by Tefelski et al [2]. The MC simulation in this research was performed using MCNP code on a geometry and Pu-Be neutron source as it was used in experiment.

4. Experiment description

As the neutron shielding by CBC is related to attenuation of fast neutrons and absorption of the thermalized ones, two experimental programs were performed. In both cases, the helium counter was used and neutrons were emitted by a Pu-Be source. Specific cylindrical samples (height -35 cm, diameter -10cm) were prepared for the experiments. This shape allows to place a cylindrical detector (length -31 cm, diameter -2 cm) inside the sample in a special hole. As a result, the CBC cover for helium counter detector was constant, equal to 4 cm.

In the first experiment, the system was built in order to stop all the thermal neutrons (Fig.1a). It was obtained by using a filter sheet made of cadmium. As only fast neutrons emitted by the source pass into the measuring channel, with CBC sample, a detector placed inside the sample records only thermal neutrons that are a result of slowing fast neutrons within material. This is mainly an effect of collisions with the hydrogen and high cross section nuclei of atoms contained in the sample, so the influence of atomic composition and moisture (hydrogen) content in CBC can be easily interpreted. In the second study, in order to eliminate all fast neutrons, the neutron flux was attenuated by the moderator - paraffin surrounding the Pu-Be source in a special paraffin block (Fig.1b).

Interpretation of both experimental results is as follow: better CBC is that which slows down the larger number of fast neutrons, measured indirectly by an increase in the number of registered thermal neutrons in the first study and at the same time the one that absorbs more thermal neutrons in the second study.

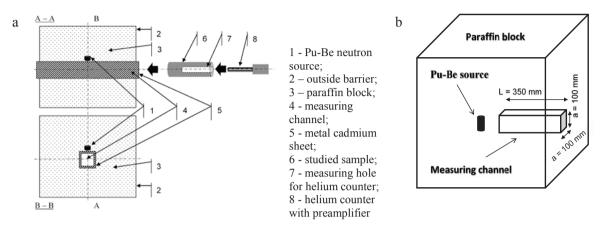


Fig. 1. (a) The experimental system for fast neutron attenuation measurement with cadmium sheet channel; (b) Paraffin block used in thermal neutron absorption measurement.

5. Results

The results of experiment could be presented in relative number of thermal neutrons (Wcp) registered by the helium counter in relation to a fixed initial value - background (cps_0), according to equation 2:

$$Wcp = \frac{\Delta[cps_0; cps_t]}{cps_0} \cdot 100\%$$
⁽²⁾

where Wcp - the relative number of registered thermal neutrons; cps_t - the number of registered thermal neutrons emerging from a sample of concrete; cps_0 - the number of registered thermal neutrons without concrete samples in the test channel.

Result of the first experiment (Tab.4) proved that heavy weight-concrete is not good solution for shielding against fast neutrons, as the number of thermal neutrons registered by a helium counter for M-HWC (0.4) increased only by 14%, that is less than 1/3 of that observed for G-NWC (0.4). The increase of compressive strength is not correlated to effectiveness of shielding. The other trend is visible when comparing only the normal-weight concretes. If the w/c decreases (so compressive strength increases) there is an increase of fast neutron thermalization as well but the differences are rather small and the firm statement should be avoided. It is also shown that taking the linear correlation from previous research, each 1% of moisture in concrete makes around 10% increase of effectiveness of fast neutron thermalization, a little less than it is for mortars -15% (Fig.2a).

Results of second experiment are different. In relation to the initial number of thermal neutrons in the channel, the 4 cm thick G-NWC absorbs around 70%, regardless of the w/c ratio and related technical properties (e.g. compressive strength). Heavy weight-concrete proved to be better in thermal neutron shielding – the result for M-HWC (90%) was 1/3 higher than for NWCs.

The experimental results are in accordance to MCNP simulations (Fig.2b). It is proved by similarity of results of MCNP computer simulations and experiments expressed as a linear attenuation coefficient - μ , calculated according to equation 3 already presented by Akkurt et al. [5]:

$$\mu = \frac{1}{x} \cdot \ln \frac{I_0}{I} [cm^{-1}]$$
(3)

where x is the material thickness in cm 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.

Symbol	Fast neutron attenuation	Moisture content	Fast neutron attenuation	Thermal neutron absorption
	Dry state	Air dry state	Air dry state	Dry state
	[%]	[%]	[%]	[%]
G-NWC (0.4)	44	2.3	67	70
G-NWC (0.5)	42	3.1	72	69
G-NWC (0.6)	36	3.5	73	69
M-HWC (0.4)	14	1.9	33	90
Mortar CEM I	46	2.4	77	63
PCC Mortar	46	5.2	134	66

Table 4. Results of shielding properties, Wcp.

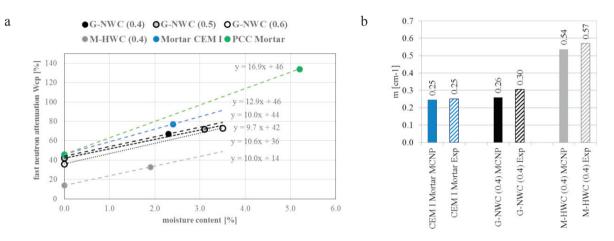


Fig. 2. (a) The relation of fast neutron attenuation vs. moisture content; (b) linear attenuation coefficient for thermal neutrons calculated by MC simulations (MCNP) and resulted from experiments (Exp).

6. Conclusions

The aim of the paper was to present an analysis of neutrons transport through standard cement mortar, PCC mortar, normal-weight concrete and magnetite heavy-weight concrete. The goal was to find an influence of the cement type, polymer addition, density and moisture content on the shielding properties of a composite against neutron. First experimental studies confirmed an influence of the cement type on fast neutron attenuation. Taking into account the previously stated linear correlation in the measurements of fast neutrons attenuation with different moisture content it was shown that for normal-weight and heavy-weight concrete each 1% of moisture makes around 10% increase of effectiveness of fast neutron thermalization (a little less than it was estimated for cement based mortars). The investigation confirmed that increasing density of material has a marginal influence on fast neutron shielding efficiency, so performance-based design of such concrete should take into account other conditions like atomic composition and moisture (hydrogen) content. It is proved that heavy-weight concrete is not proper solution for shielding against fast neutrons but its efficiency is visible in the case of thermal neutrons absorption, probably due to increase of Fe content at the expense of Si and O in the atomic composition. The influence of water retained by magnetite concrete could also have an influence. The further investigations are in

progress. The results of experiment are in accordance to MCNP calculations so it confirmed the statement of NESCC Final Report of the Concrete Task Group [16] that the effectiveness of concrete neutron shields is easily calculated using modern radiation if the degree of concrete dehydration can be estimated transport codes like MCNP.

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