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Monte Carlo simulations for optimization of neutron shielding concrete

Research Article

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Abstract: Concrete is one of the main materials used for gamma and neutron shielding. While in case of gamma rays an increase in density is usually efficient enough, protection against neutrons is more complex. The aim of this paper is to show the possibility of using the Monte Carlo codes for evaluation and optimization of concrete mix to reach better neutron shielding. Two codes (MCNPX and SPOT - written by authors) were used to simulate neutron transport through a wall made of different concretes. It is showed that concrete of higher compressive strength attenuates neutrons more effectively. The advantage of heavyweight concrete (with barite aggregate), usually used for gamma shielding, over the ordinary concrete was not so clear. Neutron shielding depends on many factors e.g. neutron energy, barrier thickness and atomic composition. All this makes a proper design of concrete as a very important issue for nuclear power plant safety assurance.

Keywords: radiation protection • concrete design and optimization • Monte Carlo

1. Introduction

One of the main radiation shielding materials used in nuclear power plants is a cement concrete. It attenuates both gamma and neutron radiations. One of the main advantages of concrete, in comparison to the others, is that it is a composite-type material and there is a possibility to optimize its constituents and mix proportions for better properties. Optimization has already been widely used in concrete mix design for better mechanical properties but in radiation protection the geometry of barriers is designed not due to the static conditions but due to efficiency of shielding properties of an element. In general, it leads to massive structures which cost are not negligible. Most

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of all, facilities space and weight reduction is desirable.Ordinary concrete of a density $\varrho = 2000 \div 2600\ kg/m^3$, is a mixture of cement, coarse and fine aggregate, water and eventually additives and admixtures that are set by cement hydratation. Usually for shielding barrier a heavyweight concrete ($\varrho > 2600\ kg/m^3$) is used. It is obtained by addition of heavy components (mainly aggregate and fillers) like basalt, magnetite, barite, limonite, iron and metal ash and slag. The report [1] presents the analysis of concrete components for The MARIA nuclear reactor and for The Żarnowiec nuclear power plant with domestic polish sand, gravel and cement. The goal is to achieve better radiation shielding properties with no increase in weight as well as improvement or no loss in other properties e.g. compressive strength and durability. The aim of this paper is to show the possibility of using the Monte Carlo (MC) simulation codes for evaluation and optimization of concrete mix to reach better neutron shielding. The performance of neutron shielding concrete has already been simulated and investigated experimentally by Okuno et al [2]. Gallego et al [3] compared experimental results using ^{241}Am-Be neutron soures to MCNP5 calculation resutls (MCNP5 is the same package as MCNPX [4] for low energy neutrons: less than 20 MeV) and they found some differences with regard to the experiments. In this paper, only theoretical study was presented. Authors are planning an experimental verification of the SPOT results on real samples of concrete of different compositions in a real neutron fluxes. This work is than an entry point for further experimental data validation.

2. Monte Carlo simulations

In Poland SAMSY [5] numerical programs as well as ANISN [6], DOT [7], MORSE [8] and MCNP/MCNPX [9] have been the most commonly used to design the shield against the radiation of nuclear facilities, such as the MARIA nuclear reactor in Świerk and The Żarnowiec nuclear power plant [1], the accelerator in the SINS and sub-critical assembly SAD [10]. These programs allow for the solution of Boltzmann transport equation using remove-diffusion method (SAMSY), discrete ordinates method (ANISN, DOT) or the Monte Carlo (MC) method (MORSE, MCNP). However, the solution of complex shielding problems in a nuclear reactor uses a combination of programs based on the method of discrete ordinates and schemes based on the MC method. In this case, DOT programs are used for calculation of two-dimensional distribution of neutron radiation and gamma rays in a reactor core, while programs based on the MC method in the other parts of the object. It should be mentioned, that simultaneously to the development of computational methods and numerical programs, work on nuclear cross sections is beeing conducted. In this paper, the core of nuclear reactor was calculated with APOLLO 2 [11] and shielding using MC software with ENDF/B nuclear data as the cover for calculations. MC simulation method is one of the most reliable methods for determining the ability of materials to shield against the radiation. Calzada et al [12] already used MC simulations to optimize the composition of new shielding material composed of steel resin, paraffin/polyethylene and a boron compound that radiation attenuation is higher compared to heavy weight concrete. This method is a statistical method based on the pseudo-random number generator, the

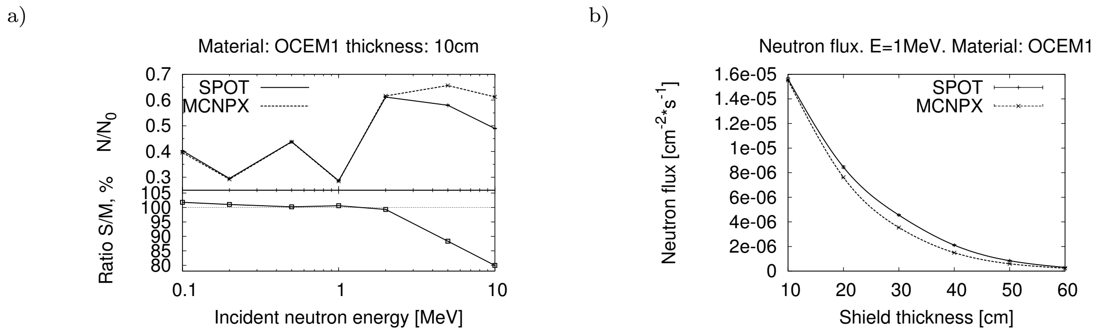

Figure 1. **SPOT and MCNPX comparison: a) neutron transmittance vs. incident neutron energy for 10 cm concrete thickness, b) neutron flux for 1.0 MeV incident neutron energy transmittance vs. concrete thickness**

cross sections for reactions in material already known and the assumed model of particle motion. A complete set of simulation must also include a geometry of the shielding material, an atomic composition of material, parameters of radioactive source and a type and a placement of radiation detectors. The MC simulation method has one important drawback - the real time of simulation significantly increases when more accurate data has to be calculated, especially when high complexity test system is considered. The most advanced MC computational package is MCNPX [4] from Los Alamos National Laboratory. It is very flexible package, which can be used in many applications [13] including shielding analysis [14]. In this paper, data from simulation of neutron transport in concrete shields using MCNPX and also authors' simulation software called SPOT are presented. SPOT, a quite simple software for engineering calculations, was prepared using the assumptions of MCNPX software, but including only the elastic neutron scattering and absorption in the model. The advantage of SPOT is a wide range of possible modifications. A semi-quantitative agreement between data obtained from SPOT and MCNPX was obtained up to 2.0 MeV of neutron initial energy (Fig.1a). It is consistent with the model used, because in this energy range, the elastic scattering process is dominating in neutron collisions with atoms. The comparison of 1.0 MeV neutron flux depending on thickness of concrete shield for SPOT and MCNPX shows that obtained results are in good agreement (Fig.1b).

Calculations in MCNPX were performed for 10^7 neutron histories, whereas in SPOT were performed for 10 batches of 10^5 histories. In MCNPX nuclear data was included from ENDF/B-VI.0, ENDF/B-VI.8, ENDF/B-VII.0 databases. In SPOT, the ENDF/B-VI.8 databases were used (JANIS [15] software was used to extract cross sections). No variance-reduction techniques were applied. The basic steps of MC neutron transport simulation consists of a particle generation (initial energy, position and direction in the domain of θ, ϕ angles tossed from pseudo–random number generator – RNG). Next step is a transport code. For non–charged particle like neutron it is a straight line with a distance acquired from RNG with use of exponential distribution. Distance is easily calculated from equation 1.

$$d = -\frac{1}{\Sigma_t} \ln(x) \tag{1}$$

where x is a number from RNG (linear distribution), Σ_t is macroscopic total cross-section of medium.

Then particle interactions are considered in a manner of RNG with use of correct cross-sections. In case of SPOT only absorption and elastic scattering was considered whereas in MCNPX a lot of interactions are possible, like production of neutrons $(n, 2n), (n, 3n), (n, xn)$, gamma rays, electrons, particles $(d, t, \alpha, ...)$, ions and heavy-ions. In such cases another particle tracks are generated. Also photons and other particles like light ions may be generated. For example, if elastic scattering is chosen, a nuclide should be tossed with use of macroscopic cross-sections. MCNPX uses many advanced modelling techniques like thermal treatment based on the free gas aproximation, chemical binding, crystal structure influcence, and cross-section data are corrected accordingly to the simulation conditions. In case of elastic scattering the particle energy loss is generally obtained from momentum and energy conservation principles in a center of mass frame. Then a new "motion" is started with angle generated by RNG using differential angular cross-sections. Theses steps are repeated until an absorption is chosen or particle leaves the material. In this case particle counter is increased, flux is updated and for SPOT equivalent dose is updated. In case of MCNPX histogram of selected energy bins is created and on this basis equivalent dose is calculated in external program. In MCNPX there are many physics models codes like inner nucleus cascades which helps obtaining correct solution within high energy ranges. For inelastic scattering, there are many laws incorporated. There are photo effect, pair production, Compton effect, electron interactions like Bremstrahlung, Auger transitions and so on. MCNPX can be used in wide areas of nuclear physics simulation. The SPOT program is a very thin subset of this, but quite useful for our simulation conditions (energy up to 2.0 MeV, only neutron transport). SPOT is also not suited for complex medium geometry but suited for calculation of neutron transport in inhomogeneous medium. The proper improvement is under preparation stage.

3. Shielding concrete optimization

Lately a big progress in concrete evolution has been made. It is mostly due to use of polymer additives (e.g. superplasticizers) together with reactive additions (e.g. silica fume), that allows for decrease of w/c ratio, increase of tightness and mechanical properties without loss in workability of fresh concrete mix. Also an intensive development of Polymer-Cement Concrete (PCC) in the last decade has been observed [16]. This progress allows for a new look at the concrete optimization for better radiation shielding properties. Already some calculations and investigations on shielding (mainly against gamma rays) has been made. Akkurt et al. [17] proved that the type of the aggregate is more important than the amount of aggregate for gamma ray shielding and the barite-loaded concretes would be preferred for this purpose. Sato et al. [18] proposed for a radioactive waste management a multilayered concrete structures with boron-doped low activation concrete wall. Here a flexible neutron shielding resin, recently developed by Sukegawa et al. [19], could be used. This solution could be problematic in a reactor building construction due to the particularity of concrete works technology and problems that can arise at the interface like it happens in multilayer repair systems [20]. Bashter [21], apart from gamma shielding analyses, showed that calculated effective macroscopic neutron removal cross-sections for six different concretes can be increased up to 50% with relation to ordinary one by a change of an aggregate type. Measured values differs from the calculated ones within the range $13.5 \div 33.7\%$. It shows 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 free neutrons with the matter, depending on their kinetic energy and cross-sections for different reactions of the component atoms of the cement paste and the aggregate. Some general recommendations can be presented here e.g. increase of hydrogen content in concrete (more chemically bounded water) or addition of neutron absorbers (boron). It is worth to mention that shielding process and its intensity against different radiation depends not only on the chemical composition but the structure of the barrier (e.g. heterogeneites) [22] and local defects (e.g. cracks) as well [23].

4. Dose calculation

In radiation protection doses refer mostly to absorbed dose and relate to the X-rays and gamma rays. Formally, absorbed dose D, is defined by equation (2) as a measure of the energy deposited in a medium by ionizing radiation:

$$D = \frac{\Delta E}{\Delta m} \quad \left[\frac{J}{kg} = Gy\ (gray)\right] \tag{2}$$

where: ΔE – the mean energy imparted by ionizing radiation to matter in a volume element, Δm – the mass of matter in the volume element.

However, the biological effects do not depend only on the absorbed dose but on the sensitivity of particular organs or tissues and the radiation type as well. Radiation type is described by the ionization density (numbers of ions produced in the individual path of radiation). Therefore the two doses are defined, which take into consideration all these aspects. The equivalent dose H_T takes into consideration the radiation type (by multiplication factor w_R) and the effective dose D_E takes into consideration the sensitivity of individual organs or tissues to radiation (by multiplication factor w_T). The equivalent dose is defined by equation (3):

$$H_T = w_R \cdot D_T \tag{3}$$

where: D_T - the absorbed dose delivered by radiation type R averaged over a tissue or organ T, w_R - the radiation weighting factor for radiation type R.

The effective dose (4) is a summation of tissue equivalent doses multiplied by the appropriate tissue weighting factor:

$$D_E = \sum w_T \cdot H_T \tag{4}$$

where: H_T - the equivalent dose in tissue T, w_T - the tissue weighting factor for tissue T.

In the Polish legal system, the binding values of radiation weighting factors (Tab.1) and tissue weighting factors (Tab.2) are given in the Regulation of the Council of Ministers [24] in accordance with the recommendations of

Table 1. **Radiation weighting factors w_R [24]**

Radiation type and energy	Radiation weighting factor, w_R
Photons, all energies	1
Electrons, myons, all energies	1
Neutrons below 10 keV	5
from 10 keV to 100 keV	10
from 100 keV to 2 MeV	20
from 2 MeV to 20 MeV	10
over 20 MeV	5
Protons over 2 MeV	5
Alpha particles, fission fragments, heavy nuclei	20

Table 2. **Tissue weighting factor w_T [24]**

Tissue (organ), T	Tissue (organ) weighting factor,w_T
Gonads	0.20
Bone marrow (red)	0.12
Colon	0.12
Lung	0.12
Stomach	0.12
Bladder	0.05
Chest	0.05
Liver	0.05
Oesophagus	0.05
Thyroid gland	0.05
Skin	0.01
Bone surface	0.01
Adrenals, brain, small intestine, kidney muscle, pancreas,spleen, thymus, uterus (the weighting factor 0.05 is applied to the average dose of these organs)	0.05

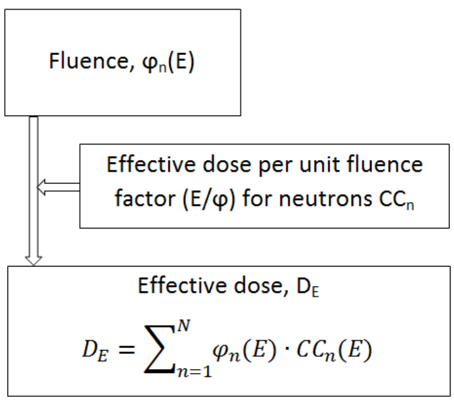

Figure 2. The model of effective dose calculation used in this paper. N - number of neutron energy groups, D_E - effective dose, E - neutron energy

Table 3. **The conversion coefficients CC_n as the effective dose per unit fluence**

Energy $[MeV]$	CC_n $[pSv \cdot cm^2]$	Energy $[MeV]$	CC_n $[pSv \cdot cm^2]$	Energy $[MeV]$	CC_n $[pSv \cdot cm^2]$
1.0E-09	2.40	1.0E-03	6.04	1.2	130
1.0E-08	2.89	2.0E-03	6.05	2.0	178
2.5E-08	3.30	5.0E-03	6.52	3.0	222
1.0E-07	4.13	0.01	7.70	4.0	250
2.0E-07	4.59	0.02	10.2	5.0	272
5.0E-07	5.20	0.03	12.7	6.0	282
1.0E-06	5.63	0.05	17.3	7.0	290
2.0E-06	5.96	0.07	21.5	8.0	297
5.0E-06	6.28	0.10	27.2	9.0	303
1.0E-05	6.44	0.15	35.2	10	309
2.0E-05	6.51	0.20	42.4	12	322
5.0E-05	6.51	0.30	54.7	14	333
1.0E-04	6.45	0.50	75.0	15	338
2.0E-04	6.32	0.70	92.8	16	342
5.0E-04	6.14	0.90	108	18	345
		1.00	116	20	343

ICRP [25]. When effective dose for all body is calculated, it corresponds to the equivalent dose because a sum of all tissue weighting factors is equal to 1 ($\sum w_T = 1$). In this paper the effective doses were determined using the neutron flux and the conversion coefficients CC_n as the effective dose per unit fluence (Fig.2). The conversion coefficients (Tab.3) depend on neutron energy and were taken from the ICRU Report [26].

5. Simulation results

Simulations presented in this paper assumed no cracks and the homogeneity of concrete (no structural heterogeneity due to aggregates arrangement) - the atoms were homogeneously spread in the atomic structure. Obviously it is not always true, for example in case of poor quality of concrete works. Next assumption concerns water demand of 25% of mass of cement used for hydratation. A preliminary study was a SPOT simulation performed for two separate groups of concretes of different class of compressive strength; from C30/37 to C45/55 (Group A) and from C25/30 to C50/60 (Group B). Change in compressive strength class was a result of different concrete composition (Tab.4) that leads to differences in atomic compositions (Tab.5). Calculations has been made for a 25 cm thick wall.

SPOT simulations source was based on a neutron flux from a Light Water Reactor (LWR) core calculated using APOLLO 2. Calculation has been made for 10^5 histories. Results present a clear decrease in effective dose behind barrier with an increase in compressive strength of concrete both for group A and B: 31% and 44% respectively (Fig.3). The next investigation was made for two types of concrete: ordinary concrete and the heavyweight concrete with barite, that is commonly used for gamma shielding (Tab.6). In these concretes, two different cement classes (CEM I 42.5 R and CEM II/B-V 32.5R) were considered. Differences in mix lead to

Table 4. Concrete compositions of different compressive strength class for SPOT simulations

GROUP A samples		C30A	C40A	C45A	GROUP B samples		C25B	C35B	C50B
Compressive strength class of concrete		C30/37	C40/50	C45/55	Compressive strength class of concrete		C25/30	C35/45	C50/60
CEM I 52.5 N	$[kg/m^3]$	275	325	375	CEM I 32.5 R	$[kg/m^3]$	350	407	488
Crushed Sand 0/2	$[kg/m^3]$	765	729	676	Sand 0/2	$[kg/m^3]$	610	611	604
Crushed limestone 2/8	$[kg/m^3]$	255	230	206	River Gravel 2/8	$[kg/m^3]$	666	667	658
Crushed limestone 8/14	$[kg/m^3]$	569	576	601	River Gravel 8/16	$[kg/m^3]$	574	575	567
Crushed limestone 14/20	$[kg/m^3]$	390	401	412	Aggregate total	$[kg/m^3]$	1850	1853	1829
Aggregate total	$[kg/m^3]$	1979	1936	1895	Super-plasticizer	$[l/m^3]$	-	1.63	7.32
Water	$[l/m^3]$	197	192	188	Water	$[l/m^3]$	189	170	152
Hydrated water	$[l/m^3]$	68.75	81,25	93.75	Hydrated water	$[l/m^3]$	87.50	101.75	122.00
w/c ratio		0.72	0.59	0.50	w/c ratio		0.54	0.42	0.31
$f_{ck,cyl,28days}$	[MPa]	34.96	41.34	48.77	$f_{ck,cube,28days}$	[MPa]	31.47	45.67	62.10

Table 5. Concrete atomic compositions of different compressive strength class for SPOT simulations

	Sample	C30A	C40A	C45A	C25B	C35B	C50B
	Compressive strength class of concrete	C30/37	C40/50	C45/55	C25/30	C35/45	C50/60
Atomic composition	H	0.33%	0.39%	0.44%	0.43%	0.48%	0.56%
	O	52.28%	52.10%	51.93%	51.98%	51.81%	51.56%
	Na	0.02%	0.02%	0.02%	0.02%	0.03%	0.03%
	Mg	0.10%	0.12%	0.13%	0.13%	0.14%	0.17%
	Al	0.33%	0.39%	0.44%	0.43%	0.48%	0.56%
	Si	40.85%	39.85%	38.88%	39.15%	38.20%	36.84%
	S	0.16%	0.19%	0.21%	0.21%	0.23%	0.27%
	Cl	0.01%	0.01%	0.01%	0.01%	0.01%	0.01%
	K	0.06%	0.07%	0.08%	0.08%	0.09%	0.11%
	Ca	5.59%	6.55%	7.49%	7.22%	8.13%	9.44%
	Fe	0.27%	0.32%	0.36%	0.35%	0.40%	0.46%

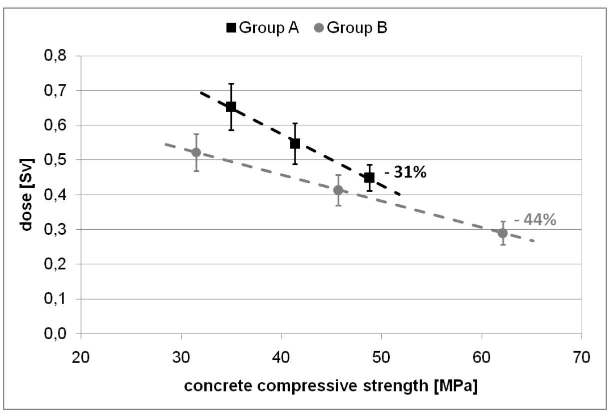

Figure 3. Effective dose behind 25 cm thick concrete of different compressive strength (SPOT simulation)

Table 6. **Volume composition of ordinary and heavyweight concrete for MCNPX and SPOT simulations**

	Concrete	Ordinary	Heavyweight
Volume composition	Microfiller	Silica fume 5%	Barite powder 1%
	Aggregate	SiO_2 76%	$BaSO_4$ 82%
	Water	6%	5%
	Admixture	0.19%	0.80%
	Cement	12%	11%

Table 7. **Atomic composition of ordinary and heavyweight concrete for MCNPX and SPOT simulations**

	Concrete	Ordinary		Heavyweight	
	Cement	CEM I	CEM II	CEM I	CEM II
Atomic composition	H	0.79%	0.79%	0.62%	0.62%
	C	0.10%	0.10%	0.04%	0.04%
	O	52.98%	53.38%	31.27%	31.65%
	Na	0.07%	0.09%	0.02%	0.03%
	Mg	0.18%	0.21%	0.09%	0.12%
	Al	1.16%	1.56%	0.32%	0.69%
	Si	37.99%	38.56%	2.41%	2.95%
	S	0.17%	0.13%	10.67%	10.63%
	Cl	0.01%	0.01%	0.01%	0.01%
	K	0.17%	0.24%	0.06%	0.13%
	Ca	5.82%	4.27%	5.36%	3.89%
	Fe	0.57%	0.67%	4.01%	4.11%
	Ba	0.00%	0.00%	45.12%	45.12%

specific atomic concrete compositions (Tab.7). SPOT simulations source was again a real neutron flux of 10^5 histories and ordinary and heavyweight concretes with CEM I and CEM II of thickness 10 to 40 cm (step 5 cm) were investigated. Results are presented in relative dose to ordinary concrete with CEM I. The advantage of heavyweight concrete is quite clear but a little disturbance at 25 cm appeared (Fig.4).

MCNPX simulation was performed using a monoenergetic point source of 10^7 histories for ordinary and heavyweight concrete with CEM I and CEM II and thickness 10 to 60 cm (step 10 cm). Results for 0.1 MeV are quite similar to the SPOT ones but the disturbance at 20-30 cm is much smaller (Fig.5). The disturbance in SPOT simulations for real neutron flux can be explained by the MCNPX results for 1.0 MeV (Fig.6) and less for 10.0 MeV (Fig.7) point source that are quite different than for 0.1 MeV. The figures show that heavyweight concrete is not the best solution for 1.0 MeV neutron radiation shielding up to thickness of 30-40 cm, where a change in the trend line is observed.

6. Conclusions

The aim of this paper was to show the possibility of using the the Monte Carlo simulation codes for evaluation and optimization of concrete for better neutron shielding properties. It was proved that neutron radiation shielding

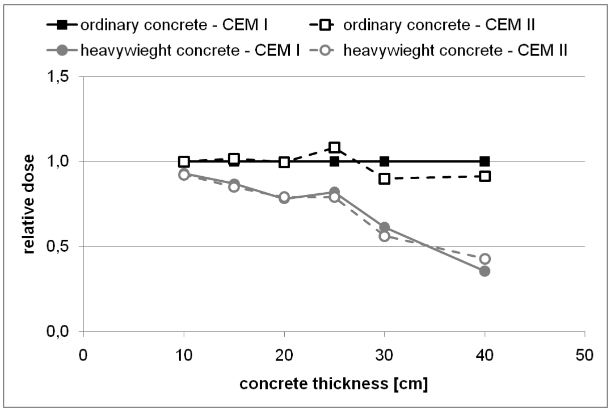

Figure 4. **SPOT simulations results for LWR neutron flux**

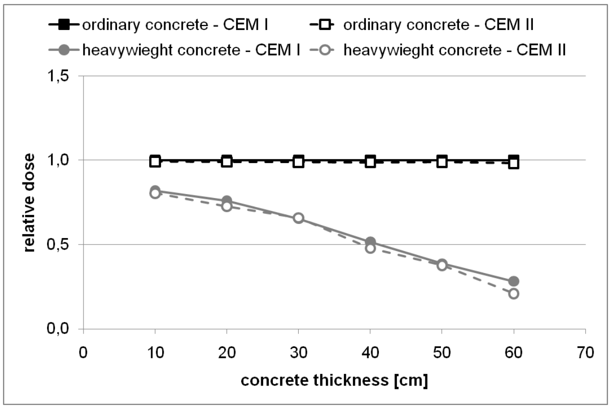

Figure 5. **MCNPX simulations results for 0.1 MeV point source**

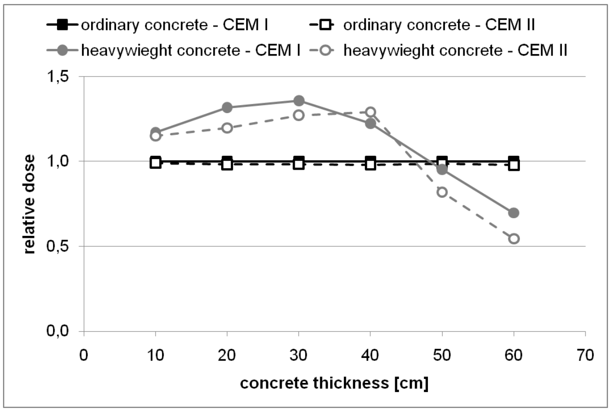

Figure 6. **MCNPX simulations results for 1.0 MeV point source**

is much more complex phenomena than protection against gamma radiation. The investigation showed that increasing density of material has a marginal influence on neutron shielding efficiency. Neutron shielding should

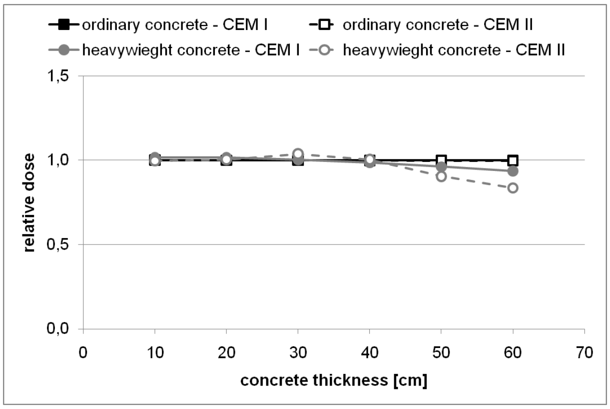

Figure 7. **MCNPX simulations results for 10.0 MeV point source**

be solved taking in mind two aspects slowing down neutrons, which is best done with light nuclides (hydrogen) and neutron absorption with nuclides with high cross section for neutron absorption like boron and gadolinium. In fact, neutron shielding for the given barrier depends on many factors e.g. neutron energy, barrier thickness and atomic composition. All this makes a very important issue for nuclear power plant and other facility (eg. accelerator) safety assurance to design concrete properly for protection against radiation not only from photons (gamma) but neutrons as well.

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