Polish Journal of Medical Physics and Engineering 2016;22(2):41-47
doi: 10.1515/pjmpe-2016-0008

Scientific Paper

Experimental investigation on radiation shielding of high performance concrete for nuclear and radiotherapy facilities

Szymon Domański[1,a], Michał A. Gryziński[1], Maciej Maciak[1], Łukasz Murawski[1], Piotr Tulik[1], Katarzyna Tymińska[1]

[1]*National Centre for Nuclear Research, 05-400 Otwock-Świerk, Poland*
[a]*E-mail address: s.domanski@ncbj.gov.pl*

Abstract

This paper presents the set of procedures developed in Radiation Protection Measurements Laboratory at National Centre for Nuclear Research for evaluation of shielding properties of high performance concrete. The purpose of such procedure is to characterize the material behaviour against gamma and neutron radiation. The range of the densities of the concrete specimens was from 2300 to 3900 kg/m^3. The shielding properties against photons were evaluated using ^{137}Cs and ^{60}Co sources. The neutron radiation measurements have been performed by measuring the transmitted radiation from ^{239}PuBe source. Scattered neutron radiation has been evaluated using the shadow cone technique. A set up of ionization chambers was used during all experiments. The gamma dose was measured using C-CO_2 ionization chamber. The neutron dose was evaluated with recombination chamber of REM-2 type with appropriate recombination method applied. The method to distinguish gamma and neutron absorbed dose components in mixed radiation fields using twin detector method was presented. Also, recombination microdosimetric method was applied for the obtained results. Procedures to establish consecutive half value layers and tenth value layers (HVL and TVL) for gamma and neutron radiation were presented. Measured HVL and TVL values were linked with concrete density to highlight well known dependence. Also, influence of specific admixtures to concrete on neutron attenuation properties was studied. The results confirmed the feasibility of approach for the radiation shielding investigations.

Key words: concrete; radiation shields; recombination chambers.

Introduction

Concrete is a cheap, versatile and widely used construction material. It is associated with its efficiency, flexibility, and simple manufacturing process. Among the obvious advantages of concrete are: high strength, good insulation properties, durability and the ability to take any shape. This material is applied as a radiation shield in radiotherapy centers, isotope and accelerator laboratories, and especially in nuclear facilities. In this type of industrial projects besides the regular concrete, a special one is also used containing, besides cement, sand, water and aggregates (stones, slag, etc.), also different types of reinforcements (e.g. steel fibers) and various admixtures (air entraining agents, plasticizers, accelerators, retarders, etc.). A typical concrete mix is prepared from about 25% cement and water, around three quarters of total volume are aggregates [1]. The 80% of the concrete is oxygen and silicon, the rest is calcium, aluminum and small amounts of sodium, potassium and iron [2]. The type and amount of concrete mix ingredients result in a qualitatively different shielding properties for different types of ionizing radiation. The density of the regular concrete is about 2350 kg/m^3, but in practice, due to the diversity of the composition (especially quantity of the grain aggregate), a "regular" is defined as concrete having a density in the range of 2000-2600 kg/m^3.

Radiation shields

Photon radiation (X and gamma) may interact with matter as a result of: the photoelectric effect, Compton scattering, or creation of electron-positron pairs. All of these effects require interaction with the electrons. Therefore the high electron density of heavy materials is essential. A typical shielding material used in this case is lead (Z = 82). Concrete used as a shield against gamma radiation should be composed so that the percentage of "electron-rich" elements is as high as possible. Shielding concrete is prepared on the basis of the heavier aggregate so that, compared with other conventional shielding materials, it has a high effective atomic number (12-13). This translates directly to effectiveness in the fields of gamma radiation and at the same time reduces the space occupied by the shield.

In case of neutrons the way they interact with matter varies greatly depending on the kinetic energy of the neutron. Depending on the energy neutrons are divided into two categories: thermal and fast. In experimental practice, the criterion for the division is specific energy - cadmium cutoff

energy (about 0.5 eV), neutrons of higher energies are fast neutrons [3]. Additionally, in the scope of fast neutrons exist the range of intermediate energies from 1 eV to about 0.1 MeV - epithermal (or resonance) neutrons.

In nuclear matter there are 3 main processes the neutron is undergoing. They are two types of scattering: elastic, when the total kinetic energy of the participants is conserved, and inelastic, when a part of the neutron kinetic energy is transferred to the nucleus, causing its excitation. The nucleus then deexcites emitting a photon. Both kinds of scattering occur in full range of energies and their result is thermalization of the neutron. The third process, occurring mostly at low and resonance energies is neutron capture. In the result a compound nucleus is created, usually in unstable excited state. Then it decays emitting alpha, beta and gamma radiation.

The effective neutron radiation shielding materials are the light elements (hydrogen, helium) and the high number of nuclei per volume or high density (water, paraffin or the like HDPE and other plastics). From the point of view of neutron shielding the most important is the amount of hydrogen contained in the concrete. Overall all the hydrogen contained in the concrete is in the form of water, therefore, the higher the water content, the lower amount of concrete is needed for thermalizing and capturing neutrons.

Parameters defining the shielding properties

As a result of interactions that cause ionization of matter the intensity of the primary beam of radiation is weakened by interactions, such as the scattering and absorption of radiation particles. The probability of interaction with matter particles with energy E per unit of path traveled by the particle is described using the linear absorption coefficient $\mu(E)$ in the case of gamma radiation or macroscopic cross section $\Sigma(E)$ in the case of neutron radiation [4].

In the case of photon radiation the intensity of radiation I_0 passing through the uniform shield of thickness of x is the product of two factors: the beam attenuation $e^{-\mu(E)}$ and the buildup parameter $B(E, \mu(E)x)$. Both the parameters are functions of shell thickness, often expressed by the average number of free paths μx travelled by radiation. Generally, this dependence is described by the formula:

$$I(x) = I_0 \cdot B(E, \mu(E)x) \cdot e^{-\mu(E)x} \quad \text{Eq. 1}$$

The build-up factor B $(E,\mu x)$ is an amendment showing the increase in the intensity of secondary and scattered radiation. It depends on:

1. The measured quantity (eg. fluence, dose, detector response, etc.).
2. Geometry
   a. Collimated beam (B = 1)
   b. Wide beam (B > 1)
   c. Uniform point source
   d. other
3. The thickness of the shield; expressed by the average number of free paths μx
   a. Linear attenuation factor μ depends on the photon energy and the type of material (effective atomic number Z).

Half Value Layer – HVL - determines the thickness of the material that suppresses the radiation beam so that the initial radiation dose is reduced by half. In a similar manner defined *Tenth Layer Value* – TVL - is the thickness of the material that suppresses radiation beam tenfold.

The HVL and TVL values are usually expressed in cm. The relation between the linear absorption coefficient, and HVL and TVL is as follows:

$$\text{HVL} = \frac{\ln(2)}{\mu} \cong 0{,}693\, L_\gamma \quad \text{Eq. 2}$$

$$\text{TVL} = \frac{\ln(10)}{\mu} \cong 2{,}3\, L_\gamma \quad \text{Eq. 3}$$

Similar considerations exist for neutron radiation. The reduction of the intensity of the neutron beam in the material is described by formula:

$$I_n(x) = I_0 \cdot B(E, \Sigma x) \cdot e^{-\Sigma x} \quad \text{Eq. 4}$$

For non-collimated radiation beam the build-up factor can be described as:

$$B(E, \Sigma x) = 1 - \beta \cdot \Sigma x \quad \text{Eq. 5}$$

where parameter β is a constant characteristic for the shielding material. Knowing the radiation absorption curve we can determine the macroscopic cross section for neutron absorption by numerical fit for the **equation 4**. A frequently used method is to determine the estimators of the absorption cross section HVL_n TVL_n defined by analogy with the gamma radiation as a shield layer reducing the intensity of the radiation beam to 0.5 and 0.1 of the initial value.

Additionally the value of $L_n=1/\Sigma$ is defined as the mean free path of neutrons in the material.

Ionizing radiation sources

As photon radiation sources one can use X-ray units or sealed radioactive isotope sources. The most commonly used sources are ^{137}Cs isotope emitting gamma rays with an energy of 0.662 MeV or ^{60}Co emitting two energy lines (1.17 and 1.332 MeV) with almost the same intensity and the average energy of 1.25 MeV. Such sources can be found in the form of bars (linear radiation source), or spread evenly on the surface (the plane source), the most common and simplest are point sources of radiation, of which the transversal dimension is much smaller (typically 10 times) compared to the distance to measuring radiation detector.

As sources of neutron radiation one can use the sealed radiation sources of proper design. Heavy isotope nucleus (e.g. ^{235}U, ^{239}Pu, ^{233}U) can undergo a fission, by reaction with neutron of a suitable energy, into two nuclei of comparable weight. During this process fast neutrons are emitted. For certain isotopes (e.g. ^{252}Cf, ^{250}Cf, ^{250}Cm) spontaneous fission, with neutron emission, may occur. The neutrons produced in this way are also sources of gamma rays resulting from the

decay products, inelastic scattering, the neutron capture reaction or activation of stable isotopes from the environment. The most common source of this type is a source containing ^{252}Cf. Its advantage is that the neutrons spectrum is similar to the one of neutrons generated in a fission reactor core.

A very popular neutron source is a nuclear reaction of type (α, n) induced in certain isotopes using alpha particles coming from alpha decay of the other isotopes. Such sources, contain isotopic converters (^{9}Be, ^{10}B, ^{11}B, ^{18}O or ^{19}F) with alpha emitters, mainly actinides (uranium, plutonium, americium). The most common sources of this type are ^{239}PuBe and ^{241}AmBe. The emitted neutrons have average energies higher than the average energy of neutrons produced in the fission reaction. For example, the ^{239}PuBe source emits the neutrons from reaction ^{9}Be(α, n)^{12}C. The neutrons have the energy range of 0.5-11.5 MeV with the average of about 4.4 MeV. Specific for this source are also gammas with an energy of 4.438 MeV, accompanying each neutron emission.

Another type of source used for shielding materials tests are sources of photoneutron from reaction (γ, n). The great advantage of such sources is that they are the source of monoenergetic neutron beam. Unfortunately, production of gamma radiation with sufficiently high energy (typically above 2.5 MeV) requires costly accelerator installation.

Parameters describing the radiation beam or field formed by source are:

Uniformity coefficient h – defined by the ratio of the thickness of the first half value layer to the thickness of the second half value layer. Its change indicates the changes in the radiation spectrum after undergoing successive layers of shielding. When 1.HVL=2.HVL=3.HVL etc. the beam is monoenergetic.

Radiation quality factor, Q – allows to determine the value of the absorbed dose equivalent H or energy deposited by radiation in living matter (tissue, organ), taking into account the biological effects caused by different types of radiation. Its formal unit (often overlooked) is Sv/Gy.

In accordance with the recommendations of the ICRP [5] for gamma radiation Q_γ equals to 1, for neutron Q_n value depends on the neutron energy and is between 5 and 20 Sv/Gy. The factor of neutron radiation depends on the linear energy transfer LET. In the case of the continuous radiation spectrum the sufficient information is the spectrum.

The spectra of isotopic sources are well known, so the determination of the quality factor for such radiation is relatively easy. Unfortunately, the radiation spectrum behind the shield changes (h≠1). In such a case, the numerical modeling of the neutron transport through the shielding or experimental determination of neutron spectra becomes necessary. The alternative is a direct measurement of the radiation LET with recombination chamber or proportional counter filled with tissue equivalent gas.

Recombination Quality Factor of radiation (RQF or Q_4) is used as a measurable indicator of numerical values similar to the radiation quality factor [6]. To determine the RQF recombination chambers are used. Measurement of RQF involves the determination of changes in the efficiency of ions collection f(U) at two suitably selected voltage polarizing, the reference voltage, U_S, and a voltage recombination U_R in relation to the same difference in the reference radiation, i.e. one for which Q = 1, according to the formula:

$$\mathrm{RQF} = \frac{\mathrm{f}(U_S)-\mathrm{f}(U_R)}{\mathrm{f_{ref}}(U_S)-\mathrm{f_{ref}}(U_R)} \qquad \text{Eq. 6}$$

As a reference gamma radiation is meant radiation of ^{137}Cs isotope.

Radiation properties after shield passage

The best accuracy in gamma and X beams provides spectrometric measurement (e.g. a NaI crystal with a photomultiplier cooperating with multichannel analyzer). As a result we observe the reduction of intensity of gamma lines of different radiation energies. However, in the case of monoenergetic gamma sources displayed in a collimated beam geometry, giving the at measurements of HVL and TVL uniformity coefficient beam h≈1, we can simplify the measurement using less demanding radiation detectors such as Geiger-Muller or ionization chambers.

The main problem that we encounter performing the measurements in the neutron fields dose is to separate the dose component derived from gamma radiation. The classic solution is to two-detector method of determining the dose components. This method uses two detectors with different relative neutron and gamma sensitivity - detector of neutron sensitivity similar to the gamma sensitivity, denoted T, and detector "insensitive to neutrons", that is low relative sensitivity of neutron, denoted U. The absorbed dose rate in the mixed radiation is obtained by solving the simultaneous equations:

$$R_T = A_T\left(h_T D_\gamma + k_T D_T\right) \qquad \text{Eq. 7}$$

$$R_U = A_U\left(h_U D_\gamma + k_U D_T\right) \qquad \text{Eq. 8}$$

where R is the detector response (depending on the type of detector it may be a current, charge, quantity or pulse rate); h_T H_U - the relative sensitivity of detectors T and U to gamma radiation in the investigated field, ie. the ratio of detectors response (sensitivity) to the absorbed dose rate (or more precisely the tissue kerma rate) of gamma radiation in the field to the response in calibration field (^{137}Cs); k_T, k_U - relative neutron sensitivity of detectors T and U, i.e. the ratio of response to the neutron absorbed dose rate in the test field to the response to the absorbed dose rate of gamma radiation $D^*(10)$ in the calibration field. The next possible step is to calculate the dose equivalent $H^*(10)$. For this purpose we use the formula:

$$H^*(10) = D_\gamma^*(10)\cdot Q_\gamma + D_n^*(10)\cdot Q_n \qquad \text{Eq. 9}$$

where D with respective index means absorbed dose rate components determined by us, and the Q coefficients correspond to the quality coefficients of gamma and neutron radiation. For the measurement of the neutron dose one can use

dosimeters of the Leake or Brown type [3], in which the detector is proportional counter with a working gas containing ^{3}He or BF_3 with a natural content of ^{10}B. The distinctive feature of these detectors is a thick layer of polyethylene moderator (in the shape of a sphere for Leake or cylinder for the Brown type) surrounding the counter. These devices are designed to be absolutely insensitive to gamma radiation. For gamma measurements a G-M counter, not sensitive to neutron radiation, can be used.

Microdosimetric recombination method

Method RMM consists of examining the current-voltage characteristic shape of the recombination chamber [7]. This analysis will provide information on energy loss distribution in the gas filling recombination chamber and thus the determination of quality factors for the individual components of the radiation dose in a volume of the detector.

This method can be used in radiation fields of unknown components and spectrum in a relatively wide range of dose rates. Measurements are based on determination of current-voltage characteristics of the chamber in the studied radiation fields and in a standard field of gamma radiation of ^{137}Cs isotope. An illustrative series of characteristics for a set of concrete slabs is shown in the **figure 1**.

The result is graphs **(figure 2)** showing the contribution of individual fractions of radiation with different LET created by ionizing radiation in the gas of recombination chamber. Here we can observe the change of individual components of the dose with a change in the thickness of the concrete shielding. The first interval corresponds to the percentage part of the gamma component. The next three columns can be interpreted as the interaction of neutrons with the molecules of the gas filling the recombination chamber. Observing the change in the composition of the radiation after passing through each layer, you can see the growth of scattered gamma radiation coming from non-elastic collisions of neutrons with the gas filling the chamber. We can also observe a decreasing number and energy of recoil protons, indicating the gradual slowing of neutrons down to intermediate energy. Passing through the last three layers, the proportions of the bars practically do not change, which means a total domination of the radiation composition by the scattered radiation.

The undoubted disadvantage of measurements performed using this method is the relatively long time it takes to make one complete characterization of the chamber (at least 5h). In return, however, we get full information about the quality of the measured radiation field. Among the advantages one can include no need for a second radiation detector measuring gamma radiation only.

Instrumentation

Tests were performed on 40 cm × 40 cm × 5 cm slabs with density approx. 3000 kg/m^3. Units were exposed to gamma radiation (^{60}Co, ^{137}Cs) and neutron radiation ($^{239}PuBe$) under controlled conditions.

Gamma sources were exposed with Tema Synergie irradiator. The device produces collimated beam directed through a bench for precise positioning of radiation detectors. Plutonium-Berylium source was exposed in free field geometry, directly from the specially designed container for storing high activity isotopic sources. During the exposition source were placed in the axis of calibration bench, 100 cm over the floor. Additionally, the laboratory is equipped with two kinds of neutron sources: $^{241}AmBe$ and ^{252}Cf, recommended by international standards [8] as reference standards for neutron dosimeter calibrations.

To estimate parameters of neutron radiation passing through concrete, two recombination chambers REM-2 and GW2 were used alternatively. REM-2 № 8 chamber [9] is a high pressure recombination chamber, filled with tissue equivalent gas containing 11% of hydrogen. It has a similar sensitivity for neutron and gamma radiation. Second chamber, GW2 has a similar construction [10], but it is filled with CO_2. Hydrogen free filling assures weak sensitivity for neutron radiation.

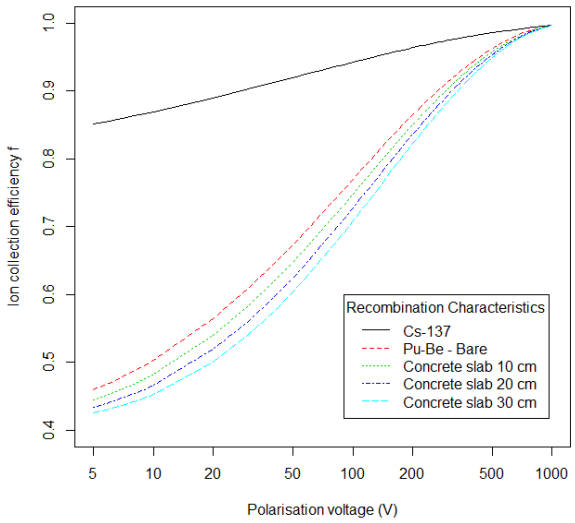

Figure 1. Series of ion collection efficiency curves measured in reference field of ^{137}Cs and $^{239}PuBe$ ionizing radiation and behind concrete slabs.

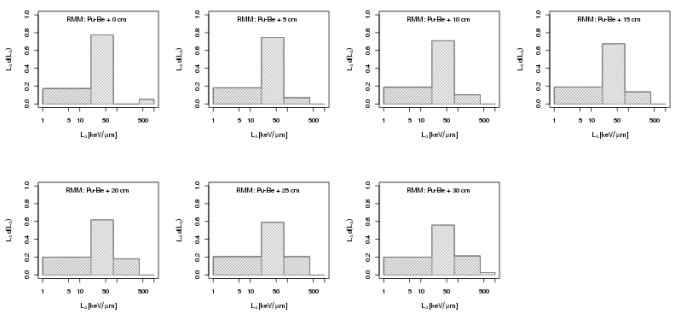

Figure 2. RMM graphs: a) histogram for bare $^{239}PuBe$ source. b) - g) histograms for consecutive thickness of 5 cm, 10 cm, 15 cm, 20 cm, 25 cm, 30 cm of portland concrete slabs.

Recombination chamber REM-2 operates with the recombination voltage $U_R = 60$ V and saturation voltage $U_S = 1000$ V. Ionization chamber GW2 operates with the voltage $U_S = \pm 30$ V. The setup of twin chambers allows for determination of particular dose components $D_n^*(10)$ and $D_\gamma^*(10)$. If additional recombination techniques are applied it is possible to estimate the ambient dose equivalent $H^*(10)$.

The measurements were conducted for various thickness of each concrete. The slabs were set on a steel frame, strong enough to carry at least ten concrete plates with a total mass up to 300 kg. The distance between the source and the detector was set to 132.8 cm. The scattered fraction of neutrons was measured using shadow cone and estimated as $D_n^*(10)_{\text{dispersed}}$ =8.73±8.7 μGy/h. The value was used as a shift correction of neutron dose rate $D_n^*(10)_{\text{total}}$ according to the formula:

$$D_n^*(10) = D_n^*(10)_{\text{total}} - D_n^*(10)_{\text{dispersed}} \qquad \text{Eq. 10}$$

Using obtained measurement results, one can plot the functions describing beam attenuation versus increasing shield layer thickness. Exemplary plot is shown on the **figure 3**.

To determine the absorption of radiation from gamma sources only GW2 chamber was employed. One can easily read out HVL and TVL values or determine linear attenuation coefficients using **equations 1** or **4**.

To determine the RMM histogram for selected types of concrete chamber REM-2 № 8 was used. The current voltage characteristics, which later appointed curves of ions collection effectiveness, was measured in the voltage range from ±5 to ±1000 V. Deconvolution of ions collection effectiveness function to the form of histograms-LET spectrum estimation was performed according to the algorithm RMM [7] implemented for this purpose in R language [11], and using the non-negative least squares NNLS model [12].

Measurement data analysis

The final result of the study for each concrete, are the values of HVL and TVL determined on the basis of attenuation curves. The resulting values are presented on the **figure 5**. We can clearly state that the density of the shield is an important factor in attenuation. Hence reduction of high specific gravity enhance the attenuation performance of concretes.

In opposition to the previous figure is an attempt to search a similar dependence for neutron radiation. As one can see in the graph in **figure 6**, for this particular problem, it is hard to find a simple relation between the applied aggregate and shielding parameters studied concrete mixtures. The accuracy of the designated half value layers due to the uncertainty of the source position, the measurement technique and the data extrapolation is at the level of 5.5% for gamma radiation and 16% for neutron radiation. Measured results do not show any correlation to concrete density.

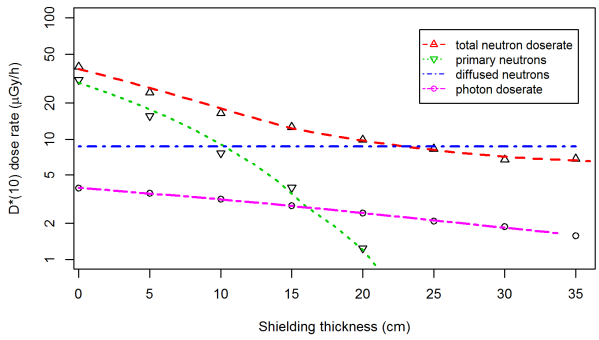

Figure 3. Gamma and neutron dose rate from ^{239}PuBe source measured behind portland concrete slabs. Absorbed dose from direct and in scattered neutrons determined with shadow cone technique.

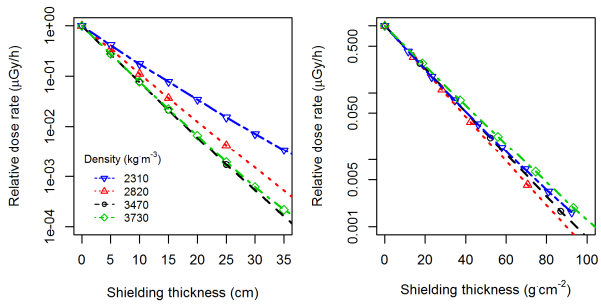

Figure 4. Transmission of gamma radiation from ^{137}Cs source thru concrete slabs of different density.

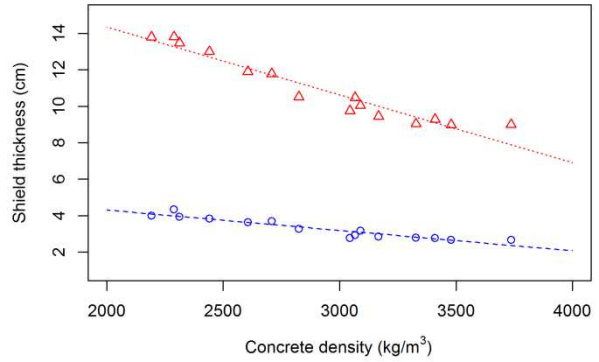

Figure 5. Variation of HVL and TVL values, gamma attenuation estimators, as a function of the density of different concretes for ^{137}Cs source.

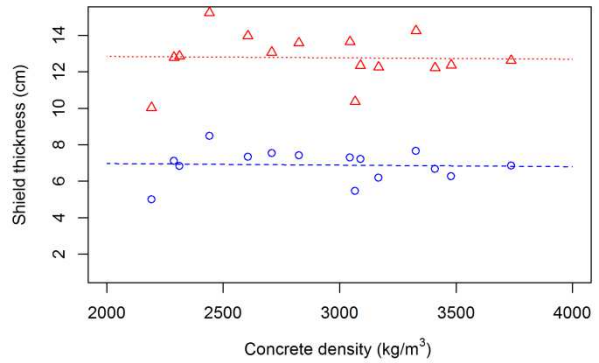

Figure 6. Measured values of 1.HVL$_n$ (circles) and 1.HVL$_n$+2.HVL$_n$ (triangles) for various types of high performance concretes.

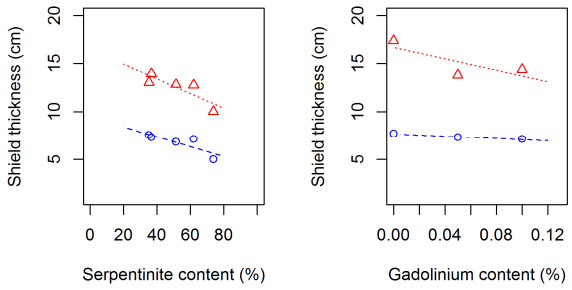

Figure 7. Measured enhancement of neutron radiation attenuation (HVL_n decrease) related to Hydrogen (left) and Gadolinium (right graph) contents. Circles represent 1.HVL_n values and triangles represent sum of 1.HVL_n and 2.HVL_n.

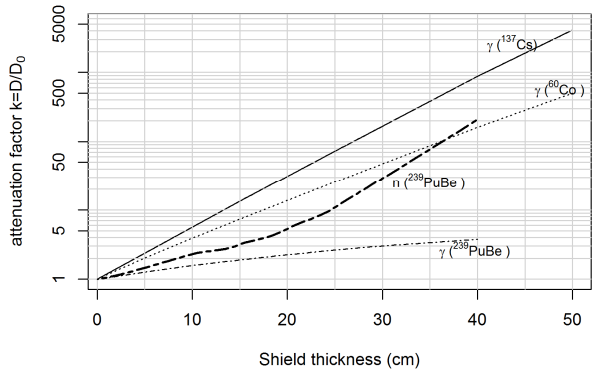

Figure 8. Attenuation factor for ionizing radiation for portland concrete of density $\rho = 2400\ kg/m^3$.

In the case of neutron radiation the type of designed concrete shielding should be considered individually, carefully choosing its elemental composition. In fact, already rough comparison of the HVL_n changes to the content of serpentine aggregate ($3MgO \cdot 2SiO_2 \cdot 2H_2O$) used in the mixture, so tied hydrogen content in the material, can be seen to improve the properties of the shielding (**figure 7**).

Obtained from measurement data attenuation curves shown previously in **figure 3** and **4** can be converted to the nomograms allowing for the determination of the attenuation coefficients in the radiation field. Example nomograms presents **figure 8**.

Summary

During the measurements of neutron fields parameters created by neutron sources carried out so far, we stated that at the calibration hall one can determine the exact value of ambient dose equivalent after passing through the concrete shielding smaller of thickness than about 30 cm. For thicker layers, high contribution of scattered radiation makes it impossible to perform measurements without the use of sophisticated measurement techniques. Moreover the authors skip details of the procedure for radiation fields with high fluxes of neutrons in which gamma generation in n-g reactions become significant. Regarding the results of gamma, one can measure the thickness of the shielding material of more than 140 g/cm^2. Determination of HVL and TVL parameters (shielding properties of various materials, various concretes mixtures) in such fields is much more complicated and also will be presented in another publication. In spite of all described above disadvantages presented technique is good enough to make rough quantitative and qualitative comparison of different materials e.g. samples of different concrete mixtures.

References

[1] Neville AM. Concrete properties. Krakow: Polish Cement Association; 2000.

[2] Kaplan MF. Concrete radiation shielding. New York: John Willey & Sons; 1989.

[3] Knoll GF. Radiation Detection and Measurement. New York: John Wiley & Sons; 2000.

[4] Shultis J, Faw R. Handbook of Nuclear Engineering. in: Radiation Shielding and Radiological Protection. Springer, p. 1313-1448. 2010

[5] ICRP. 1990 Recommendations of the International Commission on Radiological Protection. ICRP Publication 60. Ann ICRP. 1991;21(1-3):1-201.

[6] Zielczyński M, Golnik N, Makarewicz M, Sullivan AH. Definition of radiation quality by initial recombination of ions. In: Proc 7th Symp Microdosimetry; 1980 Sep 8-12; Oxford, UK. p. 853-862.

[7] Golnik N. Microdosimetry Using A Recombination Chamber: Method and Applications. Radiat Prot Dosim. 1995;61(1-3):125-128.

[8] Reference neutron radiations – Part 1: Characteristics and methods of production. ISO 8529-1:2001. Geneva, Switzerland: International Organization for Standardization; 2001.

[9] Zielczyński M, Golnik N. Rekombinacyjne komory jonizacyjne. Seria monografie IEA, Otwock-Świerk. 2000.

[10] Golnik N, Pliszczynski T, Wysocka A, Zielczynski M. Determination of Dose Components in Mixed Radiation Fields by means of Recombination Chambers. 5th Symp Neutr Dosim; 1985 Sep 17-21; Munich; Germany. Luxembourg: Commission of the European Communities, p. 717-725.

[11] R Core Team. R: A language and environment for statistical computing. R Foundation for Statistical Computing, Vienna, Austria: 2015.

[12] Lawson CL, Hanson RJ. Solving Least Squares Problems. Classics in Applied Mathematics. Philadelphia: SIAM. 1995.