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Development and Production of High Heat Resistant Heavy Concrete Shielding Materials for Neutron and Gamma Radiation

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Abstract

Design and production of new heavy concrete is needed to radiation absorption character at high teperature. For fast neutron and gamma radiation, absorption cross sections of minerals such as hematite (Fe_2O_3), titanium oxide (TiO_2), aluminum oxide (Al_2O_3), limonite (FeO (OH) nH_2O), siderite ($FeCO_3$), silicon dioxide (SiO_2), datolite ($CaBSiO_4$), barite ($BaSO_4$), galena (Pbs) were detected by using a Monte Carlo simulation program, called as GEANT4 code. The mass percentages of the materials used in the production calculated in accord with the found simulation results. New heavy concrete with resistantance to high temperature by using aluminous cement in the production was designed and produced. The behavior of new heavy concrete under high temperature was determined by "high temperature experiments". Than experimental neutron transmission measurement and mechanical resistance tests were carried out. The results were compared with normal concretes and it was seen that the performance of gamma and fast neutron radiation absorption of the new heavy concrete was higher. According to the results obtained in the present work, we suggest that the new heavy concretes can be used in nuclear reactors, in radiation therapy rooms, in storage of nuclear waste, in nuclear research laboratory and in nuclear shelters.

Keywords: Heavy concrete, Geant4, Gamma and neutron shielding

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Introduction

The neutron particles can easily penetrate into the materials without being affected by the Coulomb force of the atoms and they are very dangerous for causing high ionization such as gamma rays (METTLER et al. 1990).

Such radiation may cause serious harm to the environment and people unless good shielding is not provided.

The results of this work show that the new heavy concretes can be used in many areas such as nuclear reactors, radiation therapy rooms, storage of nuclear waste, nuclear research laboratory and nuclear shelters. Exposure to radiation at high doses can lead to health problems such as cancer, genetic disorders and tumor formation (KİRSTEN et al. 2002). Neutrons can be stopped by elements such as hydrogen whose atomic weight is small, as well as some elements with large atomic weight. Additionally, gamma rays can be stopped by high density materials such as lead. On the other hand, heavy concrete is generally used to prevent radiation leaks in radiation practice areas. Also, it provides shelter against the possibility of nuclear warfare and it is used as a shield in X-ray film practiced room's walls. (JOSEPH et al. 2014). The absorption ability of heavy concrete for both neutron and gamma radiation can be increased by adding some minerals into. The water absorption of Portland cement by adding alkali metals such as calcium (Ca), strontium (Sr), barium (Ba), radium (Ra) magnesium (Mg) have been increased. By increasing the hydrogen content in concrete, heavy concrete was produced and patented with neutron and gamma radiation shielding properties. (TANİUCHİ et al. 2007). Boron and its compounds may be sufficient to stop neutrons, as well as light, atomic materials such as hydrogen and oxygen in heavier concrete, by more than 50% by weight. When producing heavy concrete, commonly preferred compounds are iron ore such as Barite ($BaSO_4$) and Limonite ($2Fe_2O_3.H_2O$), Magnetite (Fe_3O_4).

Aggregates of mineral origin and sizes around 100 mm can be added to these concretes at a rate of approximately 75%. A multi-level concrete has been developed to store radioactive waste in the fuel reactors. Low-activated concrete at the first level, boron-added low-activity concrete at the second-level and ordinary concrete at the third level were made. (HUASİ et al. 2011). The increase in shield concrete temperature may increase the friction by affecting the resistance against the pressure in the opposite direction. The evaporation of water from the warming concrete can reduce the absorption and decay of the neutrons by approximately 30%. It is desirable that the tension caused by the pressure is very low in shield concrete. Because of this effect, there may be radiation leakage from cracks in shield concrete, which can lead to negative results. Heavy concretes used in nuclear power plants are heated when exposed to neutron and gamma radiation and as a result of this warming, the water in the concrete evaporates and cracks form on the surface of the evaporated water, causing radiation leakage in these cracks (SHAYER et al. 2010). Over time, deterioration occurs due to thermal effects, radiation effect, neutralization, freezing and thawing, mechanical vibrations, chemical corrosion, alkali aggregate reaction in shield concrete used in nuclear power plants, treatment rooms and nuclear waste deposits. Good selection of the materials to be used in the production of heavy concrete can eliminate the negative conditions. The concrete can be further strengthened by adjusting the water/cement ratio and adding in various fibers. Considering this issue, heavy concrete was designed and patented for the construction of underground shelters resistant to fire, neutron and gamma radiation, fire and chemicals Boron carbide (B_4C), natural boron (^{10}B) and (^{11}B) compounds, calcium, silicon have been added to the produced heavy concrete to increase the neutron absorption capacities. In order to give mechanical strength to the concrete, iron and steel parts are added to them (HONDORP et al. 1984). In applications where radiation is used for shielding constitutes about 40% of the cost, especially nuclear power plants. For this reason, the efforts to research and develop the material used for shield are becoming more productive and cheaper. 15,000 tons of heavy concrete are produced annually for use in radiation shield units in nuclear power plants. Fe-Ba-Serpentine and cement stone (IBSCS) were added to the structure of this heavy concrete. This heavy concrete (IBSCS) is mixed with iron powder, serpentine, cement and barium water and annealed (PAVLENKO et al. 2008). In another work, zinc-lead silver was used instead of sand in the production of heavy concrete. Gamma radiation measurements of the produced concrete were made and compared with the conventional concrete (MOHAMED, 2017). By adding some metal oxides into the concrete, the radiation holding capacities can be increased. MCNPX and XCOM codes were used to produce heavy concrete with added tungsten oxide (WO_3) whose mass attenuation coefficients were determined. Different types of samples were produced by adding nano and tungsten oxide (WO_3) in concrete. The tungsten oxide (WO_3) ratio in the concrete was increased to determine the shielding properties (TEKİN et al. 2017). In addition to these works, heavy concrete with 4 different contents with high Gamma and neutron radiation absorption were measured and heavy concretes with optimum values were determined (TÜRCK et al. 2011). The high heat resistance of the produced heavy concrete was tested. Fast neutron and radiation absorption experiments were carried out.

Materials and Methods

Monte Carlo Simulation Technique GEANT4 Code:

Simulation is a technique used in high energy physics and radiation shielding studies to predict possible pre- and post-experiment situations. Simulation technique is a time-saving tool for system design and analysis. Simulation helps engineers and planners make timely and intelligent decisions about the design and operation of the system. Simulation can not only solve problems, but it clearly defines the problem and quantitatively evaluates alternative solutions. A technique that can perform state analysis, simulation can perform numerical measurement and analysis for complex problems, and helps to find the best alternative solution in a short time. Experimental research initiated by the discovery of radiation has caused the human body to be exposed to radiation. Radiation taken during experiments gives various damages to the body, according to dose amount. For this reason, the maximum dose limits that the human body can receive per year are reported for various conditions. In radiation experiments, hand dosimeters were started to be used and radiation dose amounts of the environment were recorded. Simulations have been started to simulate Monte Carlo simulation programs to predict situations that might occur before radiation use. Today, most radiation experiments are not carried out, but instead, proven radiation transfer codes are used. The most common usage of Monte Carlo nuclear interactions is the code Geant4 which simulation is made very well. Geant4 simulation code is used in particle physics, nuclear physics, nuclear medicine and it is a kit that simulates the interaction of particles or radiation with matter in science. An input file is needed to run the program. This file contains the information about the radiation applied, the materials used, the geometry of the experiment and the desired result parameters as a result of the simulation. After the simulation is done, the results are read out in the output files. Different software is available for data analysis and graphical drawing that each program is compatible. In this study, neutron and gamma irradiation materials used in concrete design were tested with Geant4 Monte Carlo simulation program written and used by CERN before the experimental procedures.

Sample preparation

The new heavy weight concrete was produced of heat resistant alumina cement and hematite (Fe_2O_3), titanium oxide (TiO_2), aluminum oxide (Al_2O_3), limonite (Fe_2O_3 + $1/2H_2O$), siderite ($FeCO_3$), silicon dioxide (SiO_2), datolite ($CaBSiO_4$), borkarpide (B_4C), barite ($BaSO_4$), Galena (PbS), sand materials are used. In the production, 2mm-3mm diameter sand was used in cubic shape. The percentages of the materials used and the mixture are shown in Table 1. The normal concrete production technique was used in the production. At the rates determined by Monte Carlo simulations, the ingredients were mixed for 25 minutes until homogeneous in dry condition. Water was added to the mixing ratios determined and than a homogeneous mortar was obtained. The prepared mortar was poured into a preformed mold thickness of 10 cm. In order to keep the air inside the molds, the molds were made to vibrate in certain periods. The mortar in the molds was frozen at a temperature of 24 ^{0}C. In order to test the water resistance of the concrete obtained, it was placed in a limy water bath at a temperature of 21-25 ^{0}C for 28 days. Fast neutron dosing experiments of new heavy concrete with the desired properties were carried out. Experimental design is shown in Figure1. After making the best concrete specimens that absorb the radiation, the press and high temperature resistance tests were done. Each sample was placed between the source and the detector as in Figure 1 and measurements were taken for 30 minutes. The dose amount D0 (μSv/h) from the source was measured when there was no sample between the source and the detector. Then the dose amount DN (μSv/h) was measured by placing a sample between the source and the detector. The amount of dose absorbed by the sample was calculated using the equation DA = D0-DN (μSv / h).

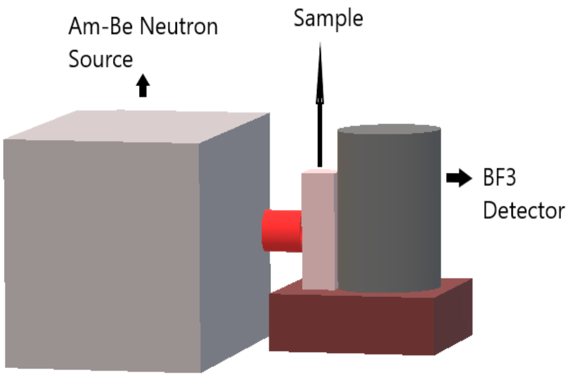


Figure1. Neutron dose measurement system

Equivalent dose measurements were performed using a ^{241}Am/Be neutron source with a mean energy of 4.5 MeV and a Canberra BF_3 gaseous neutron detector.

Results and Discussion

Neutron radiation measurement results

Elastic scattering, inelastic scattering, trapping, or escape can occur when neutron particles collide with atoms of the target. The likelihood that these interactions will occur over the distance that neutron particles take on at the material is expressed in terms of the total macroscopic cross section. These interactions depend on both the atomic density of the target material and neutron energy. The higher the total macroscopic cross section value of a material, the higher the probability that the materials atoms interact with the neutrons. These values for materials used for neutron shield must be high. The total macroscopic cross sections of the four new heavy concrete produced were calculated using the Monte Carlo Simulation Geant4 code. The theoretical results show that neutron radiation is compared to paraffin and conventional concrete used in shield Experimentally equivalent dose measurements of neutron radiation were performed. The results were compared with paraffin and conventional concrete. The results are shown in Table 2 and Figure2.

Table 1 Percentage of New Weight Concrete (%)

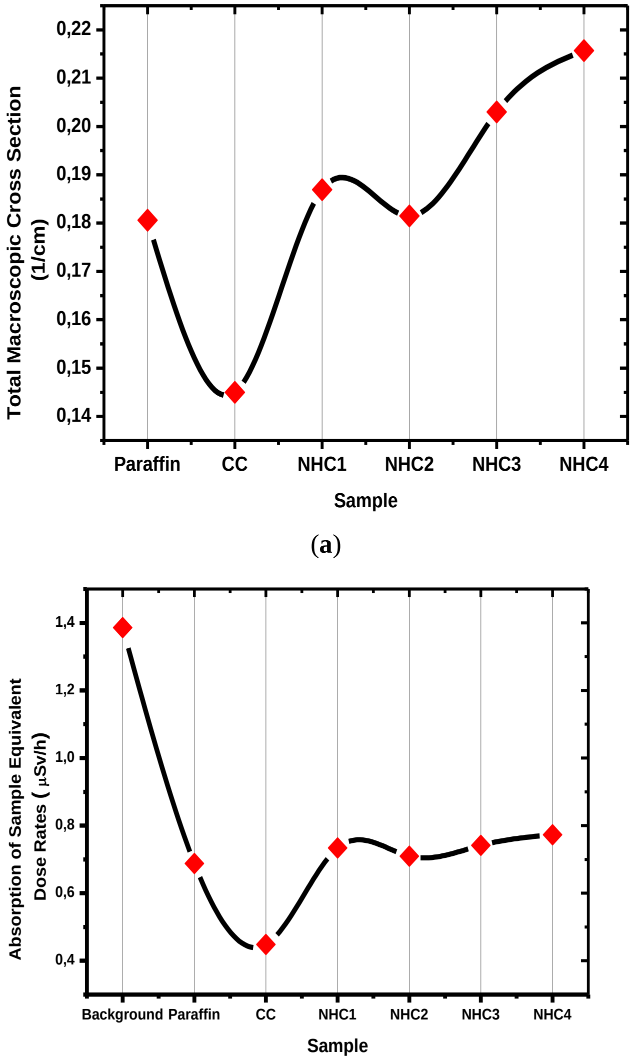
New Heavy Concrete	**Aluminate Cement**	**Water (H2O)**	**Sand**	**Aluminum Oxide (Al_2O_3)**	**Hematite (Fe_2O_3)**	**Barium sulfate ($BaSO_4$)**	**Titanium Oxide (TiO)**	**Silicone Dioxide (SiO_2)**	**Limonite (Fe_2O_3+1/2H_2O)**	**Siderite ($FeCO_3$)**	**Boron Carbide (B_4C)**
NHC1	10	15.5	48.3	6.6	13	-	6.6	-	-		
NHC2	10	15	45.04	5	5	8.3	10	-	-	-	1.66
NHC3	10	15	20	5	10	-	5	5	10	10	10
NHC4	10	15	20	5	15	-	5	-	20	5	5

Table 2. Monte Carlo simulation (GEANT4) and Equivalent Dose Rates by Experiments Results

Sample	**4.5 MeV Neutron Total Macroscopic Cross Sections (cm^{-1}) (Geant4 Theoric)**	**Detector Equivalent Dose Rates 4.5 MeV Neutron (µSv/h) D_N**	**Absorption of Sample Equivalent Dose Rates (µSv/h) $D_A = D_0 - D_N$**	**Absorbed dose percentage of samples (%)**
Background **(D0)**	-	1.3855	-	-
Paraffin	0.1806	0.6973	0.6882	49.1
CC	0.1450	0.9371	0.4484	32.3
NHC1	0.1869	0.6516	0.7339	52.9
NHC2	0.1815	0.6761	0.7094	51.2
NHC3	0.2030	0.6436	0.7419	53.5
NHC4	0.2157	0.6123	0.7732	55.8

Sample Code: CC (Conventional Concrete), **NHC** (New Heavy Concrete)

From the results in Table 2, it can be seen that the total macroscopic cross sections of the four new heavy concrete specimens produced for 4.5 MeV energy neutron are considerably higher than paraffin and conventional concrete. Considering the equivalent dose rates of new heavy concrete and paraffin selected as a reference sample and absorbed by conventional concrete, heavier doses of heavy concrete are seen. The new heavy concrete showed excellent performance by absorbing over 50% of the 1.3855 (µSv/h) dose from the source. The theoretically calculated total macroscopic cross-sections of new heavy concretes are experimentally compatible with the dose rates they absorbed. Total macroscopic cross sections and equivalent dose rates (µSv/h) absorptions of new heavy concretes and reference samples paraffin and conventional concrete are compared in Figure 2.



(a)

(b)

Figure 2 a; Theoretical 4.5 MeV Neutron Total Macroscopic Cross Sections **b**; Experimental Equivalent Dose Rates Absorbed by Sample

As shown in Figure 2, both the total macroscopic cross sections of the new heavy concrete produced and the neutron radiation doses absorbed by them are considerably higher than those of paraffin and conventional concrete.

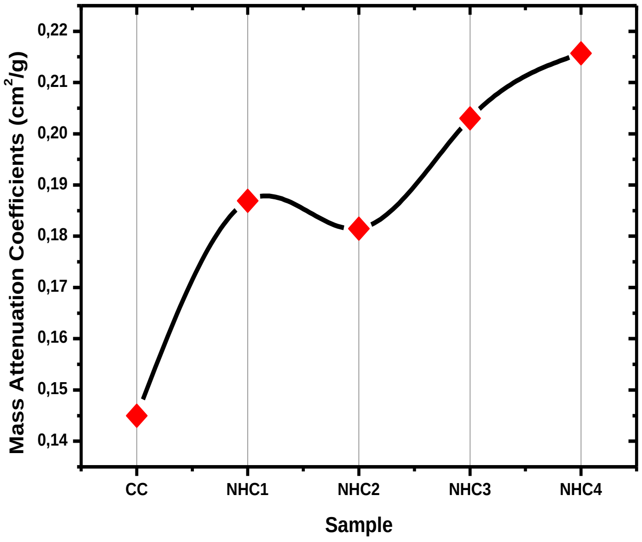
Gamma Radiation Monte Carlo (Geant4) Simulation Accounts

As a result of the fission reaction in the nuclear reactors, gamma radiation of 7 MeV energy is emitted (FREIDBERG, 2007). Absorbing gamma radiation of a material depends on the mass attenuation coefficient (cm^2/g) and the linear absorption coefficient (cm^{-1}). These values are taken into consideration for gamma radiation shield. Mass attenuation factors and linear absorption coefficient for 7 MeV energy were theoretically calculated so that new heavy concrete could be used in nuclear reactors. In the calculations, the thickness of the samples was taken 1cm and the Geant4 code was used. The greater the mass attenuation coefficient and the linear absorption coefficient of a material, the larger the gamma radiation absorption characteristic. The material has a good feature for gamma radiation shielding as shown in Table 3 and Figure 3. The mass attenuation coefficients and the linear absorption coefficients of a selected reference sample of all new heavy concrete produced are higher than of the conventional concrete.

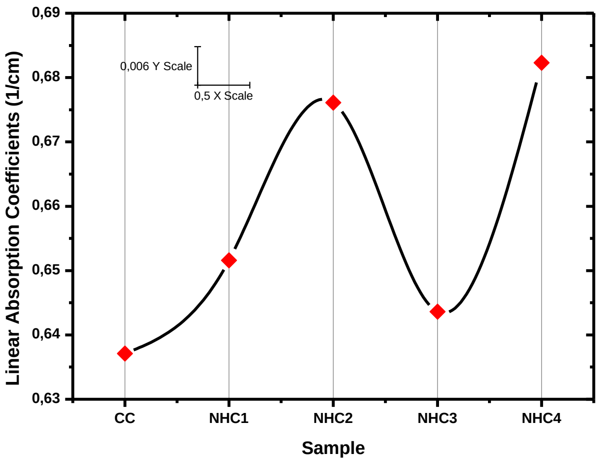
Table 3. GEANT4 Mass Attenuation Coefficient (cm^2/g) and Linear Absorption Coefficient (cm^{-1}) for 7MeV Gamma

Sample	Theoric Mass Attenuation Coefficients (cm^2/g)	Theoric Linear Absorption Coefficients (cm^{-1})
CC	0.1450	0.6371
NHC1	0.1869	0.6516
NHC2	0.1815	0.6761
NHC3	0.2030	0.6436
NHC4	0.2157	0.6823

Sample Code: CC (Conventional Concrete), **NHC** (New Heavy Concrete)



(a)



(**b**)

Figure 3. a; Geant4 Theoric Mass Attenuation Coefficient (cm^2/g) and **b**; Linear Absorption Coefficient (cm^{-1})

The mass attenuation coefficient and the linear absorption coefficient were theoretically calculated for 7 MeV Gamma radiation by using the Geant4 code. It is clear that both mass attenuation coefficient and the linear absorption coefficient of the new heavy concretes produced are larger than those of the conventional concrete selected as the reference sample from Figure 3. According to these results, it was determined that the new heavy concrete absorbed 7MeV gamma radiation well.

Conclusion

In this study, four new heavy concrete was designed and produced. The total macroscopic cross-section values for neutron radiation 4.5 MeV energy shielding were calculated by the Monte Carlo simulation program's Geant4 code. The absorption experiments were carried out for fast neutron of 4.5 MeV. Calculated results were compared with commonly used materials for neutron radiation shielding such as paraffin and traditional concrete. New heavy concrete was found to absorb higher neutron radiation than these materials. The values of the mass attenutaion coefficient and linear absorption coefficients, which are gamma radiation shielding parameters, were calculated theoretically. These results were compared with traditional concrete. It was seen that NHC1, NHC2, NHC3 and NHC4 concretes radiation absorption power much better than traditional concrete. In particular, the NHC4 concrete has proved to be very good at absorption ability gamma from traditional concrete. Alumina cement was used for these concrete to withstand high temperatures. It was determined that new heavy concrete has the ability to shield both gamma and neutron radiation leaks in the treatment of radiation, in nuclear power plants, in the transport and storage of radioactive waste, in space and laboratory research.

It is expected that the new heavy concretes produced in this study has a high absorption ability for fast neutrons and gamma rays and they will contribute to the radiation shielding technology.

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