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Neutron Shielding Properties of Some Vermiculite-Loaded New Samples

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1. Introduction

Nuclear reactor technology is known as an emerging area of study from past to present. It is an implementation of the nuclear sciences including reactions about atomic nucleus and productions. During the construction of a nuclear reactor, the most important issue is nuclear safety. The term of security can be attributed radiation shielding processes. For nuclear reactors, there are several different materials used to radiation shielding. While determining the most appropriate material to shield, the type and energy of radiation is extremely important.

There are two types of nuclear reactions reveals very large energies. These are the disintegration of atomic nuclei (fission) and merging small atomic nuclei (fusion) reactions. Therefore, nuclear reactors can be divided into two groups according to the type of reaction occurred during as fission reactors and fusion reactors. Currently a nuclear reactor working with fusion reactions is not available. Today, there are the hundreds of nuclear reactors based on the fission reactions. For the realization of nuclear fission, a large fissile atomic nucleus such as ^{235}U can absorb a neutron particle. At the end of nuclear fission event, fission products (two or more light nucleus, kinetic energy, gamma radiation and free neutrons) arise. Fission reactions are controls by using neutron attenuators such as heavy water, cadmium, graphite, beryllium and several hydrocarbons. While designing a reactor shield materials against gamma and neutron radiations should be used.

Vermiculite is a monoclinic-prismatic crystal mineral including Al_2O_3, H_2O, MgO, FeO and SiO_2. It is used in heat applications, as soil conditioner, as loose-fill insulation, as absorber package material and lightweight aggregate for plaster etc. Its chemical formula is known as $(MgFe,Al)_3(Al,Si)_4O_{10}(OH)_2 \cdot 4H_2O$ and physical density of it about 2.5 g.cm^{-3}. Melting point of vermiculite is above 1350^{0}C. This mineral can be used as additive building material in terms of mineral properties.

Vermiculite is a component of the phyllosilicate or sheet silicate group of minerals. It has high-level exfoliation property. So if vermiculite is heated, it expands to many times its original volume. This feature is a striking ability for a mineral. Because it looks like *vermicularis*, its name is vermiculite. They molecular structure consists of two tetrahedral layers as silica and alumina and an octahedral layer including O, Mg, Fe and hydroxyl molecules. Water located between layers is an important member in the vermiculite. If mineral heats suddenly, inter-layer water transforms to steam and exfoliation feature occurs.

Vermiculite is clean to handle, odorless and mould durable. It has a wide range of uses as thermal insulation, fire durability, liquid absorption capability, low density and usefulness etc… The main uses of minerals are listed as follows;

i. Construction Industry (lightweight concretes, vermiculite-loaded plasters, loosefill insulation)
ii. Animal Feedstuff Industry
iii. Industrial Insulation for High Temperatures (up to 1100^{0}C)
iv. Automotive Industry
v. Packaging Materials
vi. Horticulture

In literature, there are several studies about vermiculite and its usage. A comparative study about the effects of grinding and ultrasonic treatment on vermiculite was done. The effect of mechanical treatment and cation type on the clay micro porosity of the Santa Olalla vermiculite untreated and mechanically treated (sonicated and ground) and saturated with different captions was investigated. An experimental study was performed about thermal conductivity of expanded vermiculite based samples. In this paper, measurements were carried out on samples in the temperature range of 300-1100 K. Because of the high heat insulation properties of vermiculite mineral, we frequently encounter studies on thermal properties of it. In another study, researchers produced new materials including vermiculite that can withstand up to 1150^{0}C. The cement-vermiculite composition was used to produce new materials to leach ^{54}Mn and ^{89}Sr radionuclide. Researchers have received the best results when using 95% Portland cement and 5% vermiculite composition. The effect on ultrasounds on natural macroscopic vermiculite flakes has been studied and effects of ultrasound treatment on the several parameters (particle sizes, crystal structure, surface area, etc...) were investigated. Finally, micron and submicron-sized vermiculites were prepared. High surface area silica was obtained by selectively leaching vermiculite. In this study, also the characteristics of the porous silica obtained from vermiculite are compared with those from other clay minerals. Studies are performed on the material composition and typical characteristics of micaceous minerals of the vermiculite series in the Tebinbulak deposit. Thermal treatments of nano-layered vermiculite samples were studied up to 900^{0}C. In another thermal effect study was achieved for 15-800^{0}C temperature range vermiculite originated by Tanzania region. Thermal properties of polypropylene-vermiculite composites were investigated using differential scanning calorimetry (DSC) and thermogravimetry (TG) techniques. At the end of this study, new composites with high thermal stability were produced. The effect of sodium ion exchange on the properties of vermiculite was studied by several methodology (scanning electron microscopy, X-ray fluorescence spectroscopy, inductively coupled plasma mass spectroscopy, X-ray diffraction and thermo mechanical analysis) and sodium exchange lowered the exfoliation onset temperature to below 300 °C. A comparative study about oil affinity of expanded and hydrophobized vermiculite was done. According to the results of this study, the expanded vermiculite had a greater affinity for oil than hydrophobized vermiculite. XRD characteristics of Poland vermiculites were studied and crystal structure of it was determined. A general study about typical properties and some parameters of different vermiculites was performed. In this study, especially heat conduction coefficients were commented and an evaluation was done about vermiculite based building materials. In another study, flyash-based fibre-reinforced hybrid phenolic composites filled with vermiculite were fabricated and characterized for their physical, thermal, mechanical and tribological performance.

Neutron shielding studies have a wide range of literature. For example, colemanite and epoxy resin mixtures have been prepared for neutron shielding applications by Okuno, 2005. Agosteo et al. have investigated double differential neutron distribution and neutron attenuation in concrete using 100-250 MeV proton accelerator. In another study, neutron transmission measurements were studied through pyrolytic graphite crystals by Adib et al. Neutron attenuation properties of zirconium borohydride and zirconium hydride were determined by Hayashi et al., 2009. Sato et al. designed a new material evaluation method by using a pulsed neutron transmission with pixel type detectors.

In this paper; we investigated usability of vermiculite loaded samples for nuclear reactor shielding processes, because of excellent thermal insulation properties of it. This mineral was doped in cement and new samples including different vermiculite percents were produced. 4.5 MeV neutron dose transmission values were determined. Also 4.5 MeV neutron attenuation lengths were calculated for each sample.

2. Experiments

2.1 Sample preparation

Before the mixing procedure, a part of mixing water at the percentage of water absorption capacity of expanded vermiculite aggregate by weight was added to vermiculite to make it fully saturated with water. Fig.1 shows the expanded vermiculite particles saturated with water.

Fig. 1. Expanded vermiculite fine aggregate saturated with water.

Then, the rest of the mixing water cement and silica fume or steel fiber were mixed together for 1 minute in a mixer, and finally, expanded vermiculite aggregate saturated with water was added to cement slurry and mixed for 3 minutes again, to get a homogenous structure. Fig.2 shows the fresh state of the mixture of lightweight mortar.

Fig. 2. Fresh state of lightweight mortar prepared with expanded vermiculite aggregate.

The prepared fresh mortar were cast in standard cube (with an edge of 150 mm) molds, in two layers, each layer being compacted by self-weight on the shaker for 10 s. All the specimens were kept in moulds for 24 h at room temperature of about 20°C, and then demoulded, and after demoulding all specimens were cured in water at 23 ±2 °C for 27-days. After 28 days curing, three plate specimens with a dimension of 150x100x20mm for neutron dose transmission measurements were obtained by cutting the cube specimens using a stone saw. Plate specimens obtained by the way was illustrated in Fig.3. We obtained 12 different samples. Codes and contents of samples were shown in Table.1.

Fig. 3. View of the samples

Properties of samples including fiber steel (20^{0}C)		
Code of Sample	Vermiculite /Cement Ratio	Fiber Volume Fraction
4F0	4	0
4F15	4	1.5 %
6F0	6	0
6F15	6	1.5 %
8F0	8	0
8F15	8	1.5 %
Properties of samples including silica fume (20^{0}C)		
Code of Sample	Vermiculite /Cement Ratio	Silica Fume Contents
4S0	4	0
4S15	4	5 %
6S0	6	0
6S15	6	5 %
8S0	8	0
8S15	8	5 %

Table 1. Codes and Properties of Samples

2.2 Experimental design

For neutron transmission measurement, we used a ^{241}Am/Be neutron source and a Canberra portable neutron detector equipments. ^{241}Am/Be source emits 4.5 MeV neutron particles. Physical form of ^{241}Am/Be neutron source is compacted mixture of americium oxide with beryllium metal. Fast neutrons are produced by following nuclear reaction,

$$^{9}_{4}Be(\propto, n)^{12}_{6}C$$

5.486 keV maximum energy alpha particles emitting from ^{241}Am. Neutron energy value produced by this nuclear reaction is 4.5 MeV. Radiation characteristics of ^{241}Am/Be neutron source are shown in Table.2 (Dose rate values have been obtained from The Health Physics and Radiological Health Handbook, Scintra _Inc., Revised Edition, 1992.).

The NP-100B detector provides us to detect slow and fast neutrons. Tissue equivalent dose rates of the neutron field can be measured by it. The detectors contain a proportional counter which produces pulses resulting from neutron interactions within it. The probes contain components to moderate and attenuate neutrons. So that the net incident flux at the proportional counter is a thermal and low epithermal flux representative of the tissue equivalent dose rate and the neutron field. Because of neutrons have no charge; they can only be detected indirectly through nuclear reactions that create charged particles. The NP100B detector uses ^{10}B as the conversion target. The charged particle – alpha or proton (respectively) created in the nuclear reaction ionizes the gas. Typical detector properties are shown in Table.3. Equivalent dose rate measurement results have read on RADACS program in system PC. Experimental design is shown in Fig.4.

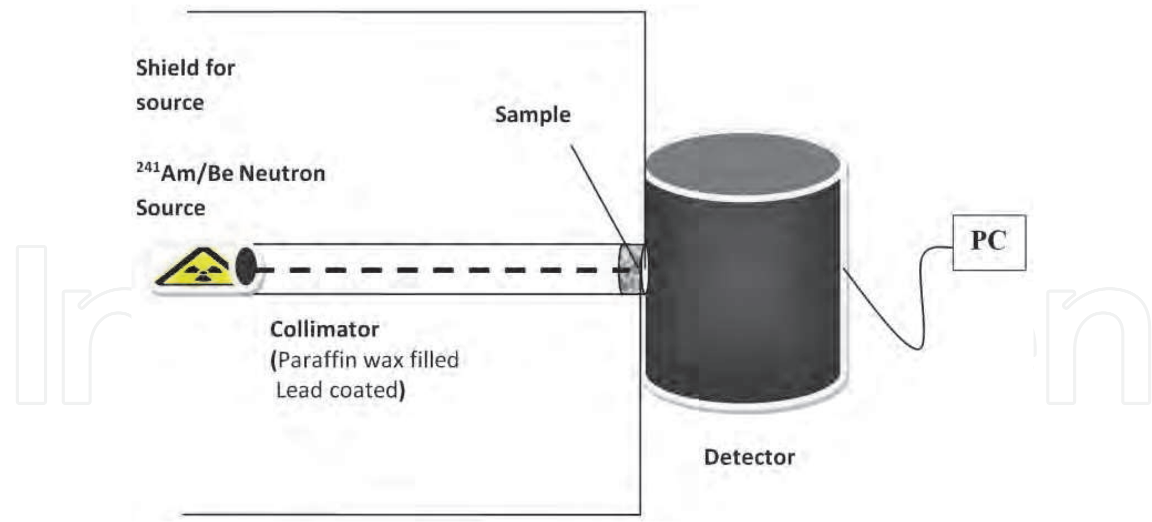

Fig. 4. Experimental Setup

Physical Half-Life: 432.2 years		Specific Activity: 127 GBq/g	
Principle Emissions	**E_{max} (keV)**	**E_{eff}**	**Dose Rate (μSv/h/GBq at 1m)**
Gamma/X-Rays	13.9 (42.7%) 59.5 (35.9%)	-	-
Alpha	5.443 (12.8%) 5.486 (85.2%)	-	85
Neutron	-	4.5 MeV	2

*http://www.stuarthunt.com/pdfs/Americium_241Beryllium.pdf

Table 2. Radiation characteristics of ^{241}Am-Be neutron source*

Specifications of Canberra NP100B Neutron Detector	
Detector Type	BF3 Proportional Counter
Detector Sensitivities	0–100 mSv/h (0–10 Rem/h)
Energy Range	0.025 eV – 15 MeV
Operating Temperature Range	–10 °C to +50 °C (+14 °F to +122 °F)
Size (mm.)	244 x 292 mm
(Dia. x inch)	(9.6 x 11.5 in.)
Weight kg (lb)	10 kg (22 lb)
Housing	Moisture Proof Aluminum
Operating Humidity	0–100% non-condensing
Detector Linearity	±5%
Accuracy	±10%
High Voltage Supply (internally generated)	1750–1950 V

*http://www.canberra.com/pdf/Products/RMS_pdf/NPSeries.pdf

Table 3. Typical Properties of Detector*

We determined dose transmission values of vermiculite loaded samples. Firstly, we counted equivalent dose rate by fast neutrons while there is no sample between source and detector system. And then we measured for each sample neutron equivalent dose rate while there is our sample between ^{241}Am-Be source box and detector probe. The ratio of two values is called dose transmission.

3. Results and discussion

Nowadays concrete is often used in radiation shielding process. In several studies, some additive materials were added in concrete to increase its radiation shielding capacity. In this study, as an additive material, we have used vermiculite mineral with a good heat insulation material. Produced samples have three different vermiculite and cement ratio values. 4.5 MeV neutron dose transmission values (Fig.5) and attenuation lengths of samples (Table.4) were obtained. Attenuation length is just equal to the average distance a particle travels before being scattered or absorbed. It is a useful parameter for shielding calculations. Also we calculated experimental 4.5 MeV neutron total macroscopic cross sections (μ) using by dose transmission values. The various types of interactions of neutrons with matter are combined into a total cross-section value:

$$\Sigma_{total} = \Sigma_{scatter} + \Sigma_{capture} + \Sigma_{fission} + ... \quad (1)$$

The attenuation relation in the case of neutrons is thus:

$$I_X = I_0 e^{-\Sigma_{total} x} \quad (2)$$

$$\Sigma_{total} = \frac{\ln\left(\frac{I_0}{I}\right)}{x} \quad (3)$$

where I_0 is known as beam intensity value, at a material thickness of $x = 0$. Equivalent dose rate has been used instead of beam intensity because of our equivalent dose rate measurements. Experimental 4.5 MeV neutron total macroscopic cross sections were shown in Table.5.

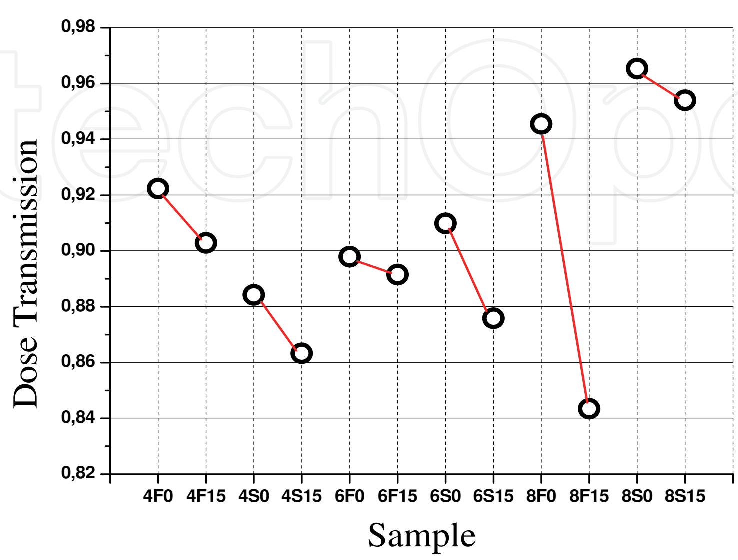

Fig. 5. 4.5 MeV neutron dose transmissions for each samples

Code of Sample	Attenuation Length (cm)
4F0	31.92848
4F15	25.25253
6F0	23.96932
6F15	22.47191
8F0	46.04052
8F15	15.15611
4S0	20.96876
4S15	17.55618
6S0	27.31494
6S15	19.46283
8S0	73.20644
8S15	54.79452

Table 4. 4.5 MeV neutrons attenuation lengths

Code of Sample	$\mu(cm^{-1})$
4F0	0.0313
4F15	0.0396
6F0	0.0417
6F15	0.0445
8F0	0.0217
8F15	0.0650
4S0	0.0477
4S15	0.0570
6S0	0.0366
6S15	0.0514
8S0	0.0137
8S15	0.0183

Table 5. 4.5 MeV neutron total macroscopic cross sections

As can be seen from Fig.5 and Table.4, dose transmission values and attenuation lengths decrease with increasing fiber steel and silica fume contents. This result indicates that neutron shielding capacity of samples is increased by silica and steel amount. According to the results, there is not a consistent relationship between vermiculite content and neutron shielding capacity of samples except of F15-samples. The sample named 8F15 is the best neutron attenuator in all specimens. The reason of this that, this sample has higher vermiculite and fiber steel content than others. The worst sample is 8S0 which has higher vermiculite but lower silica fume content. As a result, to increase neutron shielding capacity of sample, expanded vermiculite and fiber steel may be added in the mortar.

4. Conclussions

At the end of this experimental study, we reached the following outcomes;

1. Vermiculite mineral has high-level thermal insulation capacity. Concrete isn't decomposing with vermiculite addition. This mineral can be used as an additive for radiation shielding process.
2. According to the experimental results, neutron shielding property of concrete increase with increasing fiber steel and silica fume content.
3. To produce good materials which have high radiation shielding capacity and thermal insulation property, vermiculite and fiber steel may be doped in mortar. These materials can be used for neutronic and thermal applications.

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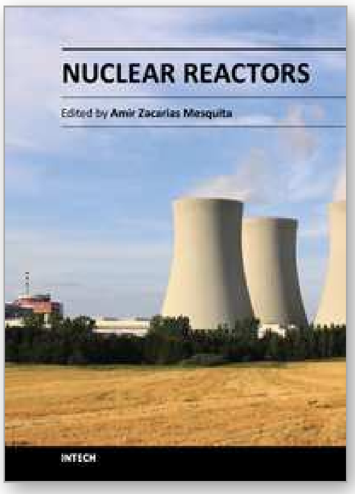

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This book presents a comprehensive review of studies in nuclear reactors technology from authors across the globe. Topics discussed in this compilation include: thermal hydraulic investigation of TRIGA type research reactor, materials testing reactor and high temperature gas-cooled reactor; the use of radiogenic lead recovered from ores as a coolant for fast reactors; decay heat in reactors and spent-fuel pools; present status of two-phase flow studies in reactor components; thermal aspects of conventional and alternative fuels in supercritical water?cooled reactor; two-phase flow coolant behavior in boiling water reactors under earthquake condition; simulation of nuclear reactors core; fuel life control in light-water reactors; methods for monitoring and controlling power in nuclear reactors; structural materials modeling for the next generation of nuclear reactors; application of the results of finite group theory in reactor physics; and the usability of vermiculite as a shield for nuclear reactor.