Gamma and neutron attenuation properties of barite-cement mixture

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Abstract. For the neutron radiography facility renovation plan at Thai Research Reactor, mixed barite-concrete blocks of different compositions were tested for their photon and neutron radiation attenuation properties. ^{60}Co and ^{137}Cs isotopes were used as the gamma sources; ^{241}Am-Be was used as the neutron source. For detection, a scintillation counter and a BF_3 tube were used. The intensities at various energies were measured and attenuation coefficients were calculated. Samples of barite mixture were analyzed with X-ray. The results involving the effects of barite are reported and discussed.

1. Introduction

Nuclear radiation can be generated from many sources, natural and man-made, such as radioisotopes, reactors, accelerators. Such radiation is widely used in many fields such as nuclear medicine, medical diagnosis, industrial tracing and radiography. Besides its beneficial effects, radiation can also cause harm. Adequate protection is thus the first priority when handling radiation sources.

Neutron radiography (NR) experiment at Thai Research Reactor 1/Modification 1 (TRR-1/M1) is one of several beam experiment facilities at Thailand's only research reactor. NR has been in operation for more than 20 years. Its current wall setup is shown in figure 1. Due to dislocation of concrete blocks, there has been an initiation to renovate the facility with several improvements including new concrete walls.

When the beam shutter is opened, neutrons from the reactor core travel along the neutron beam tube (left side of the figure) inside a wall. The beam is then incident on a sample before being absorbed into the beam stopper embedded in the concrete wall. For the purpose of increasing its potential to conduct future research, the facility is currently undergoing renovation stages, which involve replacing the beam shutter, the shielding walls, and the imaging system. Based on recent literature [1-3], barite concrete blocks were tested for their radiation shielding properties. Another available aggregate, copper slag, which seemed to possess desirable characteristics in terms of strength and gamma shielding [4-5], was also used for comparison.

In this work, the process using these concrete samples as shielding material for gamma and neutron radiation is discussed. The content is organized in the following manner. In section 2, the methods of study are described. Section 3 goes through the results on concrete properties. Finally, the summary and conclusion are given in section 4.

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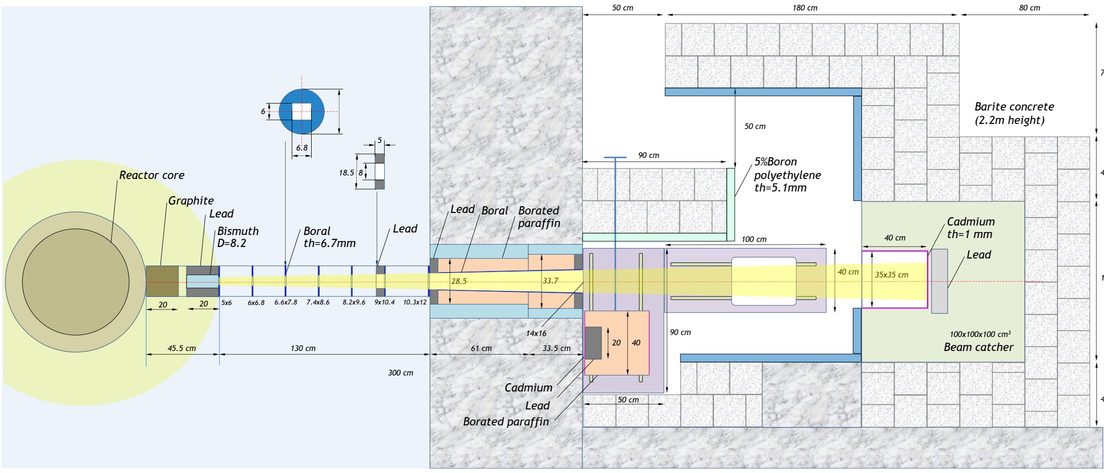

Figure 1. Top view of the current neutron radiography facility at TRR-1/M1. From the left side to the right side is the reactor core inside a water pool, the beam tube consisting of graphite, lead, bismuth, and boral, the reactor wall with a beam tube inside, the beam shutter, the sample staging area, beam stopper, and the concrete shielding walls.

2. Materials and Methods

The reactor neutron radiography system consists of six main parts: 1) the neutron source, 2) the beam shutter, 3) the collimator, 4) the sample stage, 5) the image capture system, and 6) the shielding walls. In the overall renovation plan, several improvements are for each of these components. The neutron source in this setup comes is the fission reaction of uranium-235. The beam shutter, when in the "Closed" position, is used as the first line of defense against neutron beam. The collimator is used for focusing the beam, and can be used as secondary safety equipment. It is one of the parts not currently being used but planned for the future. The sample stage is also planned to be improved so that it can be remotely controlled. For the image capture system, there are several options: X-ray films, digital imaging plates, or a camera. Finally, shielding walls are planned to be built with a known composition to replace the current walls. To design and evaluate the composition for the new shielding walls, Monte Carlo simulation and actual measurements were conducted. For the shielding wall blocks, barite powder and copper slag was mixed with cement at different ratios. The blocks were tested with gamma and neutron sources.

For characterization, samples were also tested using X-ray sources via fluorescence and diffraction techniques.

2.1. Concrete composition

Barite ($BaSO_4$) powder was formed into cement mortar. For the first batch, four different barite concrete (BaCon) formulae with different barite-to-cement ratios were used. The measured densities are between 2.7-2.9 g cm^{-3}. The compositions are shown in table 1.

Table 1. Weight compositions of barite concrete (BaCon) samples.

formula	cement	fine barite	water	density (g/cm^3)
BaCon 1	280	3090	435	2.91
BaCon 2	310	3050	430	2.72
BaCon 3	350	3000	420	2.91
BaCon 4	400	2940	420	2.78

Another batch of concrete was mixed using copper slag as aggregate, along with a smaller fraction of barite powder. Compositions of copper slag concrete (CuslCon) are shown in table 2.

Table 2. Weight compositions of copper slag concrete samples.

formula	cement	fine barite	agg Cu slag	water	density (g/cm^3)
CuslCon 1	400	400	2050	200	2.68
CuslCon 2	400	800	1700	200	3.04
CuslCon 3	400	1200	1350	200	3.06
CuslCon 4	400	1600	1000	200	2.92

2.1.1. X-ray analysis

Concrete samples were analyzed with X-ray fluorescence (XRF) and X-ray diffraction (XRD) in powder forms. XRF was done using PANalytical Epsilon 5 spectrometer with energy range 25-100 keV (atuo adjust). XRD was done using a Bruker device, operated at 30 mA and 30 kV. From XRD spectra (figure 2), $BaSO_4$ orthorhombic peaks were well identified (a:b:c = 8.792:5.430:7.110, α=90°, β=90°, γ=90°). XRF results are listed in table 3. Composition of copper slag aggregate is shown in table 4.

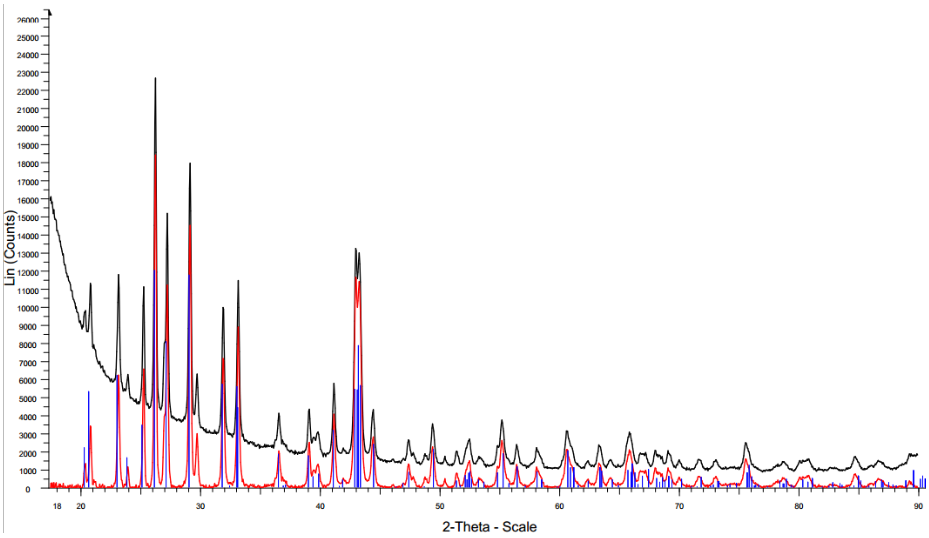

Figure 2. XRD spectrum of one of the BaCon samples (formula 1). Top spectrum is raw measurement. Lower spectrum has background subtraction applied. Vertical lines are barite peaks.

Table 3. Molecular concentration of barite concrete (BaCon) from X-ray fluorescence analysis. The values are normalized (total 100%).

formula	BaO (%)	SO_3 (%)	Sum (%)	Statistical error (%)
1	50.8	23.5	74.2	0.4
2	50.0	23.0	73.0	0.4
3	49.1	22.2	71.4	0.4
4	48.0	21.5	69.5	0.4

Table 4. Major molecular constituents of copper slag aggregate from X-ray florescence analysis. The values are normalized (total 100%).

compound	Fe_2O_3	SiO_2	Al_2O_3	ZnO	CuO	CaO
concentration (%)	74.752	13.396	2.644	2.275	1.609	1.324

2.2. Shielding tests

Monte Carlo simulation was performed along with actual measurements. In the measurements, analyses were carried out using two gamma sources providing know energies. The sources used were Cs-137 and Co-60. After energy spectra were obtained, attenuation coefficients (μ) at three energies were calculated using the available 0.661, 1.173, and 1.333 MeV photopeaks.

2.2.1. Monte Carlo simulation. Simple simulation setup was studied using PHITS (Particle and Heavy Ion Transport code System), a general purpose Monte Carlo particle transport code created by Japan Atomic Energy Agency (JAEA). The setup is shown in figure 3. Two types of sources were used to study the shielding capability of the materials. First one is a 4-MeV gamma source and the second is a 2-MeV neutron source. These energies are assessed to be the maximum energies in the actual radiography setup. Source cross-section area is 10x10 cm^2. Density between barite concrete and copper slag concrete are kept the same in the simulation (3.35 g/cm^3). In actual experiment, the densities are likely different. For the weight composition, copper-slag concrete consists of 45% copper slag (about 60% of it is Fe_2O_3), 40% barite, 10% cement, and 5% water, whereas barite concrete consists of approximately 46% Ba and 5% Fe.

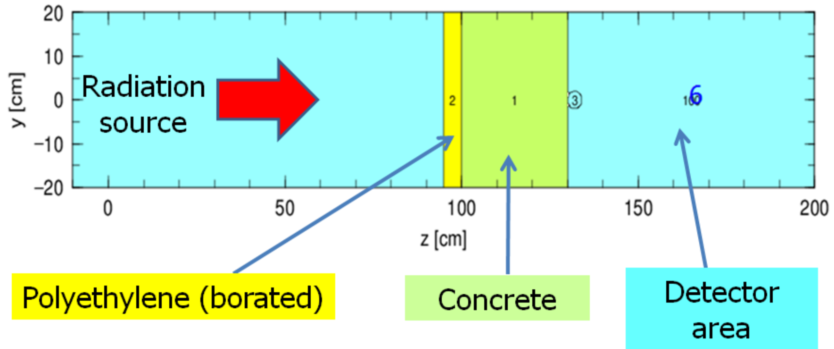

Figure 3. Geometry and location of radiation source, materials, and detector used in Monte Carlo simulation. The concrete volume is changed between different concrete compositions.

2.2.2. Gamma measurement. Lanthanum bromide scintillation detector (Canberra LABR-1.5x1.5) was used to measure gamma signals. A simple linear setup with the source and the detector separated by 40 cm was used, as shown in figure 2. Partial collimation using lead blocks was applied. The counting time was set at 5 minutes per sample. The measured count rates and percent transmission between different concrete compositions are shown in figure 5. It can be seen that the gamma shielding properties of the new concrete samples are comparable to the old one.

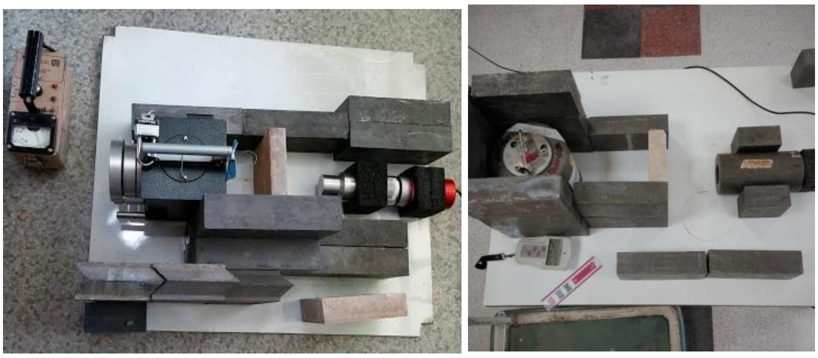

Figure 4. Radiation shielding test setup for Cs-137 source (left) and Co-60 source (right) and LABR setup.

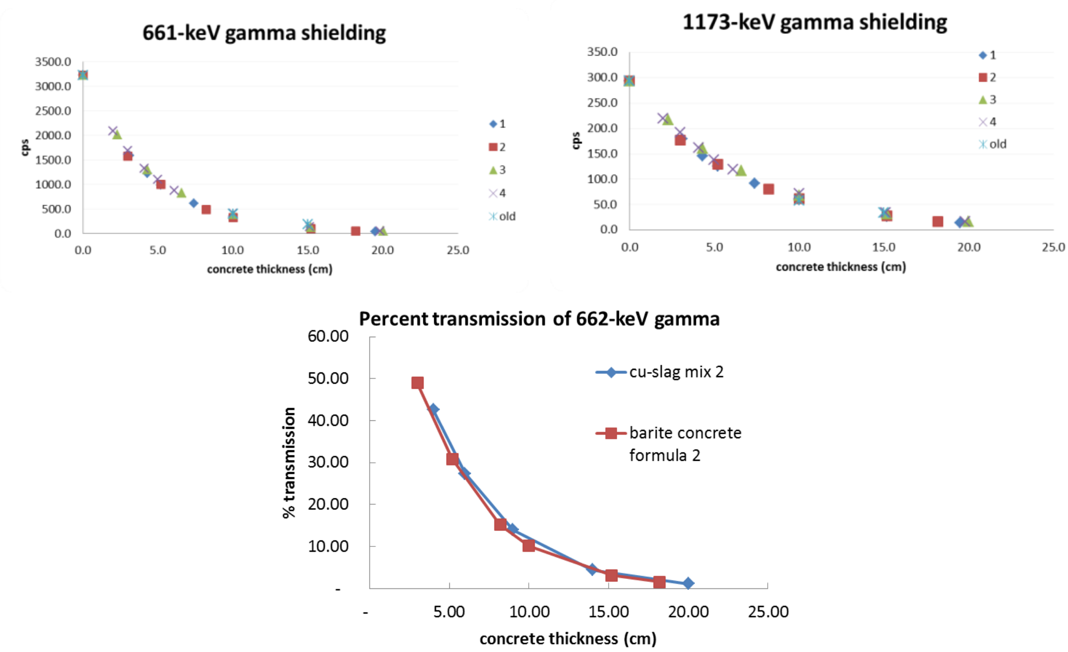

Figure 5. (Top row) Gamma count rates for different BaCon compositions at different values of concrete thickness, and (Bottom row) percent transmission of gamma ray through BaCon formula 2 and CuslCon formula 2.

The gamma intensity is related to the material thickness according to Beer-Lambert law:

$$I(x) = I(0)e^{-\mu x} \quad (1)$$

where I is the intensity as a function of thickness x and μ is the energy-dependent attenuation coefficient.

2.2.3. Neutron measurement. Boron trifluoride (BF_3) detector operated with a high voltage supply of 1,800 V was used to detect neutrons. A 50-Ci Am-241/Be neutron source with a collimator was used as the neutron source, with the setup shown in figure 6. This newly arranged facility at TINT, Nakorn Nayok, is aimed for nuclear analytical techniques and irradiation applications. Source-to-detector distance was set at 60 cm. The counting time was 1 minute. Beer-Lambert law is also applied to the data to find out the macroscopic cross section Σ.

Figure 6. Radiation shielding test setup using Am-241/Be source and BF_3 detector.

3. Results and Discussions

Simulation results using the Monte Carlo method are shown in table 5. Two different concrete thicknesses and two radiation sources were used. Compared to the barite concrete, the Cu-slag concrete is predicted to yield similar results in gamma shielding and much better results in neutron shielding.

Table 5. Fluence ratio of Cu-slag concrete to barite concrete from simulation.

concrete thickness	radiation source	fluence ratio of Cu-slag concrete to barite concrete, computed in air space behind concrete	
		neutron	gamma
30 cm	4-MeV gamma	-	1.107
40 cm	4-MeV gamma	-	1.117
30 cm	2-MeV neutron	0.533	1.220
40 cm	2-MeV neutron	0.383	0.951

From the shielding tests, gamma ray attenuation coefficients decrease as the gamma energy increases. For both gamma and neutron shielding properties, there is not a large variation in the coefficient values across different concrete formulae. The coefficients are summarized in table 6. The calculated energy-specific gamma attenuation coefficients are similar across all concrete formulae. With the current neutron setup, the measurements are inconsistent with the simulation predictions. A future experiment setup incorporating high-density polyethylene sleeves for neutron thermalization is planned.

Table 6. Radiation attenuation coefficients of different concrete formulae. Subscript of μ indicates the gamma energy.

formula	*gamma*			*neutron*
	$\mu_{0.661}$ (cm^{-1})	$\mu_{1.173}$ (cm^{-1})	$\mu_{1.333}$ (cm^{-1})	Σ (cm^{-1})
BaCon 1	0.223±0.001	0.157±0.001	0.146±0.001	0.113±0.031
BaCon 2	0.229±0.001	0.156±0.001	0.147±0.001	0.112±0.032
BaCon 3	0.212±0.001	0.145±0.001	0.136±0.001	0.101±0.024
BaCon 4	0.209±0.001	0.144±0.001	0.140±0.002	0.113±0.034
current	0.200±0.011	0.150±0.001	0.141±0.001	0.100±0.006
CuslCon 1	0.185±0.002			0.025±0.002
CuslCon 2	0.222±0.001			0.024±0.002
CuslCon 3	0.212±0.002			0.025±0.002
CuslCon 4	0.206±0.001			0.023±0.002

4. Conclusions

For the purpose of shielding neutron and gamma rays in the neutron radiography area at research reactor TRR-1/M1, concrete blocks, formed by mixing in barite and copper slag were formulated and constructed. Characterization was done using XRF and XRD techniques. The results from the newly formulated concrete (except BaCon 3 and 4) are comparable to that of the current concrete in terms of gamma shielding. Similar gamma shielding properties are observed for the two concrete types. Results agree qualitatively with the prediction from Monte Carlo simulation. Copper slag concrete is theoretically more effective than barite concrete in terms of neutron shielding. However, using the existing experimental setup, the neutron measurements yield the opposite results. New measurements using high-density polyethylene sleeves, which were shown to thermalize neutrons well, are planned for the future study. For the shielding of gamma rays, barite concrete is an effective material. However, the brittleness of the aggregate-free formulae keeps them from practical applications. Experiments with other compositions and larger aggregate size will be conducted at the next stage. Besides using material with good shielding properties, it is equally essential to have good safety equipment such as dose monitoring detectors clearly displaying the dose rate in the area.

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