Buildup factor and mechanical properties of high-density cement mixed with crumb rubber and prompt gamma ray study

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Abstract. High-density cement mixed with crumb rubber has been studied to be a gamma ray and neutron shielding material, especially for photonuclear reactions that may occur from accelerators where both types of radiation exist. The Buildup factors from gamma ray scattering, prompt and secondary gamma ray emissions from neutron capture and mechanical properties were evaluated. For buildup factor studies, two different geometries were used: narrow beam and broad beam. Prompt Gamma Neutron Activation Analysis (PGNAA) was carried out to determine the prompt and secondary gamma ray emissions. The compressive strength of samples was evaluated by using compression testing machine which was central point loading crushing test. The results revealed that addition of crumb rubber increased the buildup factor. Gamma ray spectra following PGNAA revealed no prompt or secondary gamma ray emission. Mechanical testing indicated that the compressive strength of the shielding material decreased with increasing volume percentage of crumb rubber.

1. Introduction
Radiation shielding is based on the principle of radiation attenuation, which is the ability of a shielding material to effectively reduce the radiation’s intensity by blocking particles or electromagnetic energy. Some radiation sources emit both neutron and gamma ray from absorption of high-energy gamma ray by atomic nuclei and ejection of protons and neutrons from the nuclei. Therefore, under this situation, neutrons and gamma ray are the main types of nuclear radiation that have to be considered in shielding design [1].
Gamma radiation interacts with the shielding material and become attenuated by photoelectric effect, Compton scattering, and pair production processes. The effectiveness of gamma ray shielding depends on the atomic composition (Z) of the shielding material and the photon energy. Certain gamma ray interaction processes can also result in emission of secondary photons [2].
The transmission of gamma radiation through materials depends on two factors: the attenuation factor and the buildup factor [3]. The attenuation factor accounts for the relaxation length which is the reduction of uncollided photons. The buildup factor accounts for the increased radiation intensity from secondary or scattered radiation and also the number of relaxation lengths [3], which can be computed from the multiplication of linear attenuation coefficient and shielding thickness or μx.
The buildup factor is considerably important for multi-energy gamma-rays with poor geometry for the reason that attenuation coefficients and cross sections of various mediums are not accurate. The
buildup factor is described as the ratio of total value of specified radiation quantity at any point of contribution to the value from radiation transmission at the point without undergoing a collision [4]. Moreover, prompt gamma ray may occur from a neutron interaction process. When a neutron interacts with the target nucleus via a non-elastic collision, the compound nucleus forms in an excited state due to the binding energy of the neutron with the nucleus. The compound nucleus will instantaneously de-excite into a more stable configuration through emission of one or more characteristic prompt gamma rays and secondary gamma rays [5]. This interaction is called the neutron capture or (n, gamma) reaction. Prompt Gamma Neutron Activation Analysis (PGNAA) is a non-destructive method that is useful for simultaneous determination of a trace amount of many elements in samples. This technique measures the prompt gamma rays and secondary gamma ray emission during the neutron interaction. This work focused on the buildup factor, which is the effect of the scattered and secondary radiation and is a function of the shielding material, the thickness and the energy of the radiation. The shielding material evaluated in the present study was high-density cement mixed with various percentages of crumb rubber [6]. Moreover, the prompt and secondary gamma rays will be considered in this study by the PGNAA method. These two pieces of information will help to completely determine the safety of the shielding material before it can be put into real applications. Moreover, the mechanical properties of the shielding material were evaluated as well.

2. Materials and methods

2.1. Materials and shielding preparation

Natural rubbers are often used in many industries including tyre industries, and are the potential products for neutron shielding. The tyre wastes have been increasing because of the rising demands for automobiles. Crumb rubbers are obtained from tyre wastes by recycling via mechanical process. Crumb rubbers have the special properties to effectively attenuate neutron radiation because they contain a high density of hydrogen and carbon atoms. The shielding material in this work was designed to simultaneously shield against the neutron radiation and gamma radiation, thus, mixing hydrogenous materials, heavy elements and other neutron absorbers help increase the effectiveness of shielding materials [7]. The shielding material in this research composed of Portland cement type I, barite and various percentages of crumb rubber from 5 to 25 vol%, which 5 vol% step increment. All compositions were mixed together and fabricated into a cube shape with dimensions 5 x 5 inches [6].

2.2. Measurement system

2.2.1. Buildup factor measurement and electronic system.

To determine the buildup factor, two geometric arrangements were required, which were narrow beam (good-geometry) and broad beam (poor-geometry). Transmissions of the incident photons were measured, allowing calculation of the attenuation coefficient. For the narrow beam geometry, the gamma radiation was collimated using a cylindrical lead collimator. The thickness of the lead collimator for the source holder was 5 cm with the central hole of 1 cm in diameter and 4.5 cm in height. The diameter of the lead collimator on the detector was 16 cm with the central hole of 3 cm in diameter. The distance between the isotropic source and the detector was 17 cm. The distance between the isotropic source and the shielding material was 12 cm. The broad beam geometry was obtained by simply excluding both collimators. Figures 1 and 2 show the narrow beam and broad beam arrangements, respectively.

The electronic system for radiation detection employed in this study consisted of the following units: NaI(Tl) detector (CANBERRA Model 802-2x2, crystal size 2x2 inches), pre-amplifier and multi-channel analyzer (MCA). Co-60 gamma isotopic source was used to measure 1.1732 MeV and 1.3325 MeV gamma rays emission.
The buildup factor can be calculated by using the total gamma intensity ratio with existence of collimator (good-geometry) or \((I/I_0)b\) divided by the total gamma intensity ratio without existence of collimator (poor-geometry) or \((I/I_0)g\) as follows [8]:

\[
B = \frac{(I/I_0)b}{(I/I_0)g}
\]  

(1)

The mean free path \((\lambda)\), which describes the average distance between two successive photon interactions with a unit of cm, can be calculated from the follow equation [8]:

\[
\lambda = \frac{1}{\mu}
\]

(2)

Where \(\mu\) is linear attenuation coefficient in unit cm\(^{-1}\).

2.2.2. Prompt gamma neutron activation analysis.

PGNAA is a measurement technique for non-destructive elemental analysis. A high-purity germanium (HpGe) detector model GC3021 with 30% relative efficiency at 1.1732 and 1.3325 MeV and Am/Be-241 source were used to measure prompt gamma ray emitted from the irradiated specimens. The distance from the neutron source to the specimen was 12 cm. The shielding material was placed 3 cm in front of the detector. The HpGe detector was covered with polyethylene in order to minimize unwanted neutron radiation from getting into the detector and causing damages. This measurement operated at a high voltage of 2,900 V. The system is shown in Figure 3. Each specimen was irradiated for 1 hour using the Am/Be-241 neutron source and the gamma ray spectrum was recorded from 20 to 5,300 keV.

2.3. Compressive strength testing

The compressive strength is the most significant mechanical engineering property of hardening concrete [9]. The compressive strength testing normally requires specimens with a cubical shape [10].
thus, being the reason that specimens in this work were molded into a cubical shape. The compressive strength of samples was evaluated by using a compression testing machine (CTM), which was a central point loading crushing test. Compressive strength testing was conducted after 28 days of curing time.

3. Results and discussions

3.1. Results of mean free path, linear attenuation coefficient and buildup factor

The intensity of broad beam is denoted by \( I_b \) and the intensity of narrow beam is denoted by \( I_g \). The results of gamma ray transmissions of narrow beam (good-geometry) and broad beam (poor-geometry) revealed that the \( \ln(I/I_0)b \) and \( \ln(I/I_0)g \) values decreased when the amount of crumble rubbers increased as shown in Table 1. HDC denotes high-density cement; HDCRx denotes high-density cement mixed with \( x \) volume percent of crumble rubber.

| Shielding type | \( I_b \)  | \( I_g \)  | \( \ln(I/I_0)b \) | \( \ln(I/I_0)g \) |
|---------------|-----------|-----------|-----------------|-----------------|
| HDC           | 20358.6   | 21390.7   | -1.805          | -1.769          |
| HDCR5         | 22074.6   | 21427.0   | -1.757          | -1.768          |
| HDCR10        | 24161.2   | 23575.7   | -1.635          | -1.657          |
| HDCR15        | 24251.0   | 23977.0   | -1.630          | -1.656          |
| HDCR20        | 22644.0   | 24508.7   | -1.614          | -1.634          |
| HDCR25        | 27386.6   | 26374.7   | -1.509          | -1.560          |

The reason why the \( \ln(I/I_0)b \) and \( \ln(I/I_0)g \) values decreased with increasing crumble rubber content is because the reduction of barite quantity in the shielding material as a result of addition of crumb rubber. Since barite has the elemental composition known as \( \text{BaSO}_4 \), Ba presents a heavy element with the characteristic properties to attenuate gamma radiation. The high-density cement in this work contained 45.45 w% of barite [6] to function as an effective gamma ray absorber.

| Shielding type | Mean free path | Buildup factor | Standard Deviation |
|---------------|----------------|----------------|--------------------|
| HDC           | 7.175          | 0.965          | 1.023              |
| HDCR5         | 7.182          | 1.044          | 1.091              |
| HDCR10        | 7.592          | 1.031          | 1.158              |
| HDCR15        | 7.670          | 1.025          | 1.497              |
| HDCR20        | 7.773          | 1.019          | 1.159              |
| HDCR25        | 8.139          | 1.053          | 1.218              |

The mean free path parameter describes the average distance travelled by photons between any consecutive interaction (scattering or absorption). The results revealed that the mean free paths for the case of high density cement (HDC) and high density cement mixed 5 vol% crumb rubber (HDCR5) are lower than the others. Adding more crumb rubber increased the mean free path quite substantially. This behavior can be explained that as the atomic density of rubber is lower than that of HDC and as heavy elements in HDC exhibit large scattering cross-sections for gamma rays compared to H and C, adding more crumb rubber means that gamma rays will be able to travel further before undergoing any interaction. This effectively increased the mean free path of gamma ray observed in the experimental results shown in Figure 4. The effect of crumb rubber addition on linear attenuation coefficient is shown in Figure 5.
The linear attenuation coefficient indicates how effective a shielding material is in attenuating radiation, and is used to calculate the penetration and the energy deposition by photons in shielding materials. The linear attenuation coefficients of high density cement mixed with 5, 10, 15, 20 and 25 vol% crumb rubber were evaluated to be 0.1392, 0.1304, 0.1303, 0.1286 and 0.1228 cm\(^{-1}\), respectively, indicating that the value decreased with increasing crumb rubber addition. This result is expected because as the crumb rubber percentage increased, it effectively lowers the overall Z value and the density of the shielding, resulting in less gamma ray attenuation. In fact, the linear attenuation coefficient must have the opposite trend compared to the mean free path with increasing crumb rubber percentage, and this is clearly observed in results presented Figures 4 and 5.

The results of the buildup factor experiment revealed that the buildup factors of high density cement mixed with 5, 10, 15, 20 and 25 vol% crumb rubber were 1.044, 1.030, 1.025, 1.019 and 1.053, respectively. Normally, the value of the buildup factor should be unity for the case of a collimated beam with monoenergetic photon and should be greater than 1 for other cases [11], for the reason that some scattered gamma ray occurred from shielding materials. The reduction of the buildup factor with increasing crumb rubber content up to 20 vol% is a highly desirable feature because it indicates that addition of crumb rubber helped reduce secondary and scattered gamma ray emissions. Moreover, all of the evaluated buildup factor values were very close to unity, indicating that secondary or scattered gamma radiation were effectively not present.

3.2. Prompt gamma neutron activation analysis results

Figure 6 illustrates the gamma ray spectra obtained from the PGNAA studies. Because the spectra of all shielding designs were very similar, only those for HDCR10 and HDCR20 were shown in the Figure for simplicity. The spectra were also not different when compared with Co-60 background spectrum.

It can be concluded that there was no detectable prompt or secondary gamma ray emission, which is highly desirable. This was a highly desirable feature. However, this does not mean that there was no neutron capture interaction occurring in the shielding material. It means that all the heavy and light elements in the shielding effectively shielded all of the generated prompt and secondary gamma ray. Heavy elemental composites in specimens, mainly being barite, help increase the inelastic scattering interaction, while light elemental composites such as hydrogen and carbon atoms enhance the elastic scattering interaction.
3.3. Compressive strength results

Compressive strength testing was conducted to determine the mechanical property of specimens. The results in Table 3 clearly indicated that more crumb rubber addition resulted in less compressive strength. This result is expected because crumb rubber is mechanically much weaker and less dense than the HDC matrix.

Table 3. The 28 days Compressive Strength on different type of shielding materials.

| Shielding type | Crushing load (kN) | Compressive strength (N/mm²) | Avg Compressive strength (N/mm²) |
|---------------|---------------------|-------------------------------|-----------------------------------|
|               | 1                   | 2                            | 3                                 | 1                   | 2                          | 3                                 |
| HDC           | 199.8               | 297.9                        | 245.6                             | 12.063              | 18.312                     | 14.725                           | 15.03                            |
| HOCR5         | 139.8               | 151.6                        | 158.1                             | 9.9770              | 9.7430                     | 9.6790                           | 9.80                             |
| HOCR10        | 142.5               | 169.7                        | 174.9                             | 8.3610              | 10.087                     | 10.610                           | 9.69                             |
| HOCR15        | 183.0               | 124.6                        | 166.7                             | 11.022              | 7.4120                     | 9.9710                           | 9.47                             |
| HOCR20        | 129.4               | 165.5                        | 144.0                             | 7.8980              | 10.001                     | 8.6930                           | 8.86                             |
| HOCR25        | 104.5               | 114.7                        | 111.0                             | 6.4330              | 6.9730                     | 6.7910                           | 6.73                             |

Factors that can contribute to increase the compressive strength of concrete are mix ratio, size and texture of aggregate, method of compaction and curing periods [10]. Thus, the compressive strength of the shielding with addition of crumb rubber could be enhanced by appropriately adjusting the above-mentioned factors. However, although the shielding becomes mechanically weaker with addition of crumb rubber, the shielding should not be designed and used to physically withstand any load other than the weight of the shielding itself. Several methods can be easily employed to mechanically support the shielding material such as encasement in a stronger material.

4. Conclusions

In this work, the buildup factors as well as scattered and secondary gamma radiation of high-density cement mixed with various volume percentages of crumb rubber were evaluated. The results found that the buildup factors for HOCR5 and HOCR25 were 1.044 and 1.053, respectively, which were very close to unity, indicating that secondary or scattered gamma radiation were effectively not present. Gamma ray spectra from PGNAA analysis revealed no prompt or secondary gamma ray emission from neutron interactions with shielding materials. This was a highly desirable feature. The results of compressive strength testing indicated that the compressive strength of the shielding
materials decreased with increasing volume percentage of crumb rubber, with the compressive strengths of HDCR5 and HDCR25 of 9.800 and 6.733 N/mm², respectively. Several methods can be easily employed to mechanically support the shielding material such as encasement in a stronger material. In conclusion, the safety of the high-density cement mixed with various percentages of crumb rubber was confirmed because the material exhibited several highly desirable features to shield against neutron and gamma radiation simultaneously. This shielding material can be utilized in real applications to ensure radiation safety of personnel. In order to have a complete assessment of the performance of the shielding materials, the usable lifetime of the shielding with respect to gamma ray dose and neutron fluence should be evaluated as well.

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