Cracking Cause of Neutron and Gamma Ray Shielding Material Composing of High-Density Cement, Crumb Rubber and Lead

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***Abstract*— The cause of the crack under neutron and photon flux of neutron and gamma ray shielding material composing of high-density cement mixed with 5% waste rubber and various percentages of lead powder was investigated. This shielding material was designed and fabricated in our prior work to shield against radiation from synchrotron accelerator. The lead percentages of 2, 4, 6 and 8 wt.% were studied. It was found that with the lead content of at least 4 wt.%, the shielding material cracked under neutron and gamma-ray irradiation. The crystalline phases as well as elemental composition of the shielding material were determined by quantitative X-ray diffraction technique (QXRD). Results revealed the following crystalline phases: barite, portlandite, quartz, calcite, lead and PbO2. Experimental tests were performed to determine the physical bonding of lead power to high-density cement. Results revealed that lead powder exhibited very weak adhesion to the cement, indicating that the poor mechanical interface was the underlying cause of cracking. These data will be helpful in developing this class of radiation shielding material to prevent the cracking problem.**

***Keywords*— Cracking cause, Radiation shielding material, High-Density Cement, Crumb Rubber, Lead**

I. INTRODUCTION

Ionizing radiation is widely used in many fields such as medical, industrial, and others [1]. Nuclear applications can affect human beings from ionizing radiation exposure. In order to control radiation exposure hazard, working areas and public areas have to comply with the maximum permissible dose suggested by ICRP basic radiation safety criteria [2].

The radiation shielding properties depend on material composition [3]. For gamma ray shielding, the important interactions between gamma radiation and matter are photoelectric effect, Compton scattering and pair production. High Z components in material, such as lead and tungsten, contribute to both Compton scattering and photoelectric absorption processes. On the other hand, low Z components in material result in Compton scattering processes [4]. Everything else being equal, the gamma ray attenuation coefficient increases with increasing density of material [5].

For neutron shielding, the most effective shielding material can be obtained by mixing together hydrogenous materials, heavy metal elements and neutron absorbers. For neutron interactions, the inelastic scattering process from heavy elements and elastic scattering process from hydrogen are effective in slowing down fast and intermediate energy neutrons, and the other absorbers can help reduce secondary gamma rays including capturing of thermal neutrons [6].

Tyre rubber wastes represent a significant environmental problem [7]. In this work, crumb rubber, which is a low-cost hydrocarbon polymer, is selected to mix with high-density cement. Crumb rubber is produced from natural rubber. A large number of hydrogen atoms present in the chemical structure makes this hydrocarbon polymer a very suitable choice to shield neutrons [8]. Moreover, different materials such as graphite, polyethylene and cement have suitable fast neutron scattering cross-section that can be used for moderating fast neutrons as well [3].

High-density cement mixed 5 wt.% crumbed rubber was design to be the neutron and gamma ray shielding material [1]. In order to improve the photon attenuation ability of the shielding material, lead powder was added with various percentages from 2% to 8% with 2% increments. Although the addition of lead powder in the high-density cement enhances gamma ray attenuation, increasing amount of lead beyond 2 wt.% resulted in cracking under neutron and gamma-ray irradiation. For the lead content of 0 and 2 wt.%, the cracking problem did not occur.

The aim of this research was to determine the cause of the cracking problem of the shielding material composing of high-density cement mixed with 5% waste rubber and various percentages of lead powder [9] in order to better understand and develop this class of radiation shielding material. The crystalline phases were determined and physical tests were performed to evaluate the adhesion of lead powder to high-density cement.

II. MATERIALS AND METHODS

A. Material Design

Monte Carlo Transport Code is the powerful tool that was used to study the radiation attenuation prior to sample fabrication.

1) Gamma Ray Attenuation

In designing the gamma ray shielding material, the intensity of gamma ray passing through the shielding can be calculated by using Beer-Lambert's law as follows:

$$I = I_0 e^{-\mu x} \quad (1)$$

where I_0 and I are the incident and the attenuated gamma ray intensities, respectively, μ is the photon attenuation coefficient of the material in unit cm^{-1}, and x is the thickness of the material, in unit cm, that is placed in the path of gamma ray beam.

2) Neutron Attenuation

For neutron shielding design, the principle is similar to gamma ray attenuation. Beer-Lambert law can be employed to calculate the neutron attenuation as follows:

$$I = I0 e^{-\Sigma t} \quad (2)$$

where I0 and I are the incident and the attenuated neutron beam intensities, respectively, t is the thickness of the material in unit cm, and Σ is the neutron attenuation coefficient in unit barn (10^{-24} cm^2).

B. Sample Preparation

High-density cement, crumb rubber and lead with the specific portions were mixed in a mixer. The mixture was poured into the mold and kept curing before undergoing evaluations as shown in Fig.1.

(a) (b)

Fig. 1 Sample fabrication (a) Hydration process in the mold (b) Finished sample

All samples were evaluated for gamma ray and neutron attenuations. Elemental compositions were determined by scanning electron microscope (SEM) and electron dispersive microscope (EDS). Morphological compositions were determined by X-ray diffraction (XRD). In addition, lead adhesion test was carried out to determine the physical bonding between lead particles and fabricated high-density cement.

III. RESULTS AND DISCUSSIONS

A. Material Analysis

The elemental composition of the lead powder used in this study was investigated by SEM and EDS.

From the EDS result, the lead powder was determined to compose of carbon, oxygen, tin, antimony and lead with the weight and atomic percentages shown in Table I. The powder consists mainly of lead, which is expected. Although the atomic percentage of carbon is about 2 times that of lead, suggesting the presence of PbC_2, this molecule does not officially exist. Carbon present in the powder must have been carbon powder.

TABLE I : WEIGHT AND ATOMIC PERCENTAGES OF ELEMENTAL COMPOSITION OF THE LEAD POWDER BASED ON EDS ANALYSIS

Element	Weight %	Atomic %
O	7.84	51.26
Sn	2.88	2.52
Sb	3.52	3.02
Pb	85.74	43.18

B. Photon and Neutron Attenuation

Monte Carlo Transport Code (MCNP) was utilized to investigate the neutron and photon transmission of materials. MCNP is widely used for neutron and photon transport simulations, which can be applied for various applications including radiation shielding. For this study, in the geometry specification, a specific point source was arranged in the center line in front of the samples. Mode n p was designated to be transported for neutron and photon transmissions. For gamma ray transmission, mode p was used. Neutron and gamma ray were emitted from the sources. Am/Be-241 was used as the neutron source and Co-60 was used as the gamma ray source. The macrobody capability was used to specify boxes or cube shape with dimensions 5x5x5 inches and combined using Boolean operators. Flux averaged over the surface of the shielding material was identified by tally type F2 in unit particles per cm^2 as shown in Fig. 2.

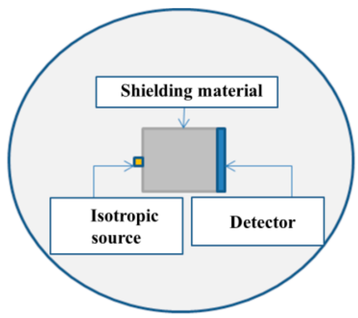


Fig. 2 Simplified 2D geometry in the simulation

The behaviour of gamma ray and neutron shielding materials (high-density cement mixed with 5 wt.% crumbed rubber and various percentages of lead powder) was analysed by the MCNP simulation. Results are presented graphically in Fig. 3 and Fig. 5.

1) Photon Attenuation Simulation

Fig. 3 illustrates the gamma radiation shielding characteristics. The lead powder at 2, 4, 6, and 8 weight percent is denoted as HDCR5L2, HDCR5L4, HDCR5L6 and HDCR5L8, respectively.

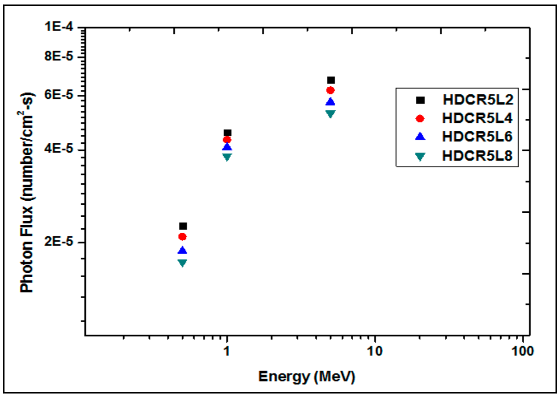


Fig. 3 Transmitted photon flux from high-density cement mixed with 5 wt.% crumbed rubber and various percentages of lead powder

The results show that for all gamma ray energies simulated, the transmitted photon flux decreased with increasing lead content in the high density cement. This result conforms to our previous conclusion that increasing heavy element composition results in higher gamma ray attenuation coefficient [9].

Fig. 4 shows the relationship between lead percentage from 2 to 8% and density of the shielding material. As expected, the addition of lead to high-density cement mixed with 5 wt.% crumb rubber resulted in an increase in the density of the shielding material. As the gamma radiation attenuation depends on atomic number and density of elements in the shielding material (as well as the energy of the incident photon) [4], the shielding efficiency increased with increasing lead content.

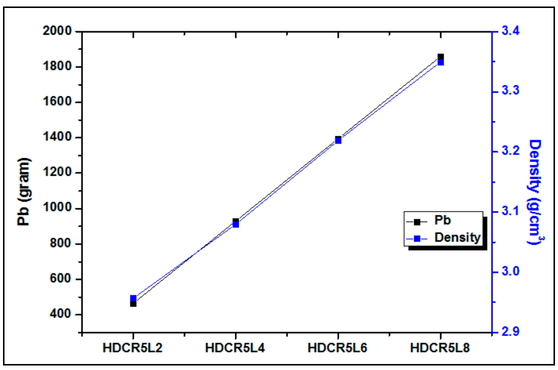


Fig. 4 Relationship between lead percentage, lead content and density of the shielding material

2) Neutron Attenuation Simulation

The simulation result of neutron attenuation shown in Fig. 5 indicates that the neutron attenuations under different lead percentages were very similar.

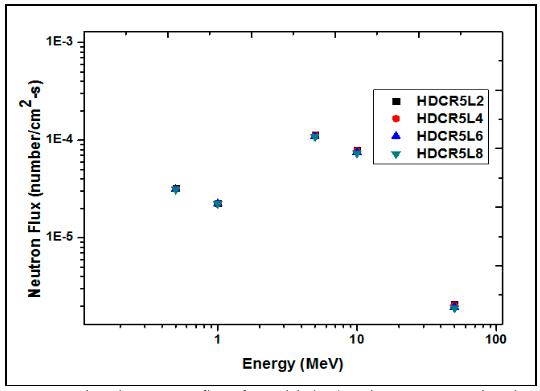


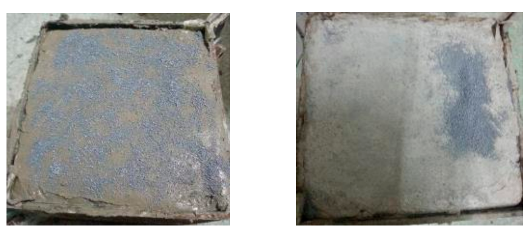
Fig. 5 Transmitted neutron flux from high-density cement mixed with 5 wt.% crumbed rubber and various percentages of lead powder

This is because the number of heavy elements in barite and the number of hydrogen atoms in crumb rubber were fixed and because lead is quite ineffective in attenuating neutrons. Thus, it can be concluded that increasing lead content from 2 to 8 wt.% was ineffective for neutron attenuation under incident neutron energies studied.

3) Lead Adhesion Testing

Cement is mainly used to bind aggregates together in concrete. Since the surface of lead powder particle is smooth unlike sand, it may affect cohesion of lead particles with high-density cement mixed 5 wt.% crumb rubber. Although the addition of lead powder to HDCR5 increased the gamma ray attenuation, increasing amount of lead beyond 2 wt.% resulted in cracking under neutron and gamma-ray irradiation. The cause of the cracking problem was determined.

The lead adhesion testing was conducted to investigate the physical bonding between lead particles and high-density cement mixed 5 wt.% crumb rubber. Lead powder was spread over the surface of the wet shielding as shown in Fig. 6(a). Moderate force was applied to the powder so that some of it was submerged in the wet shielding. After the shielding became dried, most of the lead particles on the surface were easily removed by simply wiping fingers over with little force. Fig. 6(b) shows the surface of the dry shielding after most of lead particles were removed. The agglomeration of lead on the right side is from removed lead powder.



(a) (b)

Fig. 6 (a) Lead powder on surface of wet shielding, (b) After removal of lead powder from surface of dried shielding

Thus, from this experiment, it can be concluded that lead particles do not physically adhere to high-density cement mixed 5 wt.% crumb rubber. The poor mechanical interface was the underlying mechanism for cracking. Therefore, lead particles present in the shielding actually behaves similar to “air pockets.” Under neutron and gamma-ray irradiation, stresses are generated in the shielding material and these “air pockets” represent weak areas of the material, resulting in generation and propagation of cracks. To prevent the cracking problem with the presence of lead powder, the entire surface of each lead particle must be treated to make it to bond strongly with the shielding material. Treatment can include coating with an appropriate material to be determined in future studies.

4) Samples Analysis

Two samples were prepared for material analysis. The first one consists of the following: Portland cement type A, barite ($BaSO_4$) and 5 wt.% crumb rubber. The other one consists of the above with added lead powder at 2, 4, 6 and 8 weight percent. The samples were prepared with dimensions 5x5x5 inches. Two techniques were used for material analysis as follows.

4.1) Scanning Electron Microscopy and Electron Dispersive Microscopy

The microstructure and elemental composition of the samples were investigated under SEM and EDS, respectively, using the same machine.

In the SEM micrographs shown in Fig. 7, increasing lead composition from 2 to 8% did not change the observed morphological characteristics.

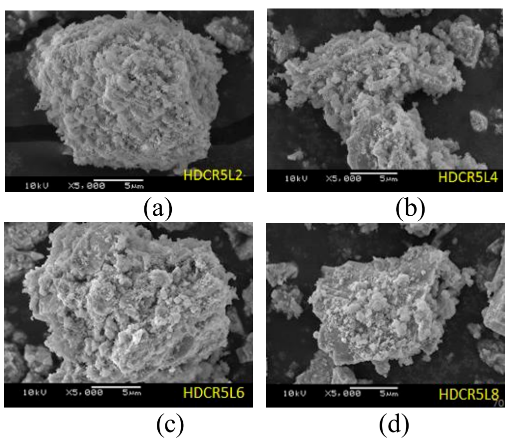


Fig. 7 SEM micrographs of high-density concrete mixed with 5 wt.% crumb rubber and various lead compositions (5,000x magnification): (a) HDCR5L2, (b) HDCR5L4, (c) HDCR5L6, (d) HDCR5L8

The elemental analysis of HDCR5L8 using EDS technique revealed that the sample consisted of barium, oxygen, sulphur, carbon, nitrogen, lead, calcium, silicon and aluminium with the following weight percentages: 35.69, 34.08, 9.27, 6.7, 5.41, 4.31, 2.75, 1.33 and 0.45%, respectively. This EDS result confirms the presence of the added 5 wt% lead. The slightly lower analysed lead quantity of 4.31% may indicate the inherent uncertainly in the analysis or it may be a result of minor inhomogeneity of lead in the small piece of the sample obtained from the large piece of fabricated shielding. The EDS spectrum of this sample is shown in Fig. 8.

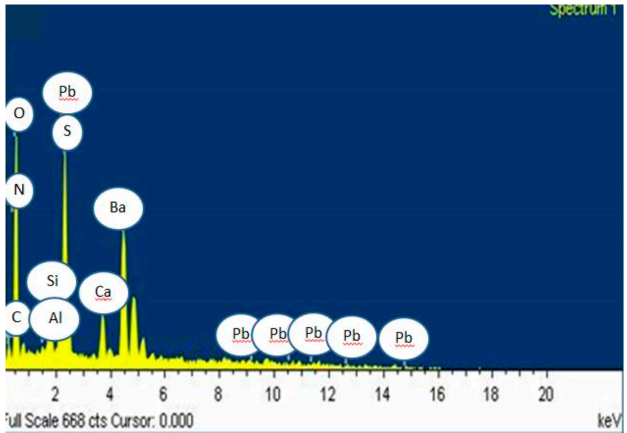


Fig. 8 EDS spectrum of HDCR5L8

4.2) X-ray Diffraction Technique

X-ray diffraction (XRD) is a technique commonly used for the characterization and identification of crystalline phases. This pattern is unique for each particular phase, and it can be used to identify the substance.

The X-ray source operated at 40 kV and 40 mA. Scans were typically collected from 5 degree 2-theta to 80 degree 2-theta using step size of 0.017 degree at a scan time of 2 hours. Samples of HDCR5, HDCR5L2, HDCR5L4, HDCR5L6 and HDCR5L8 were investigated and the crystalline phases were identified.

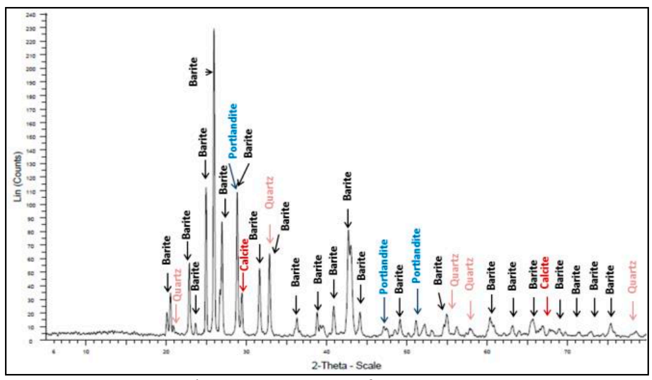


Fig. 9 XRD spectra of HDCR5

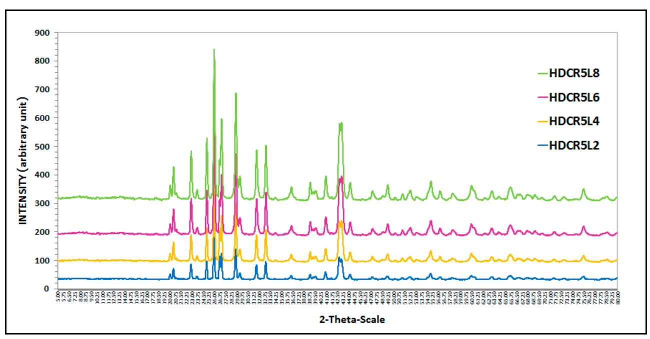


Fig. 10 XRD spectra of HDCR5L2, HDCR5L4, HDCR5L6 and HDCR5L8

The generated XRD spectra are displayed in Fig. 9 and Fig. 10. The peak locations and intensities for HDCR5L2, HDCR5L4, HDCR5L6 and HDCR5L8 are similar, but are distinctly different from those of HDCR5 because the material compositions are not same. There is no lead compound in HDCR5.

For HDCR5, the crystalline phases were determined to consist of barite ($BaSO_4$), portlandite [$Ca(OH)_2$], quartz (SiO_2) and calcite ($CaCO_3$), which has the crystal structures of orthorhombic, hexagonal, cubic and rhombohedral, respectively. The analyzed quantity of each phase is listed in Table II.

The analyzed phase composition of HDCR5L8 is listed in Table II. The crystalline phases of HDCR5L8 consisted of barite, portlandite, quartz, calcite, PbO_2 and Pb. The presence of PbO_2 and Pb was expected because of the added lead powder.

TABLE II : QUANTITATIVE ESTIMATE OF WEIGHT PERCENTAGES OF THE MINERAL PHASES IN HDCR5 AND HDCR5L8 SAMPLES BASED ON XRD ANALYSIS

Phase	**Chemical Formula**	**Quantity (wt.%)**	
		HDCR5	**HDCR5L8**
Barite	$BaSO_4$	75.029	72.769
Portlandite	$Ca(OH)_2$	0.146	1.198
Quartz	SiO_2	8.343	5.240
Calcite	$CaCO_3$	16.482	20.558
Lead oxide	PbO_2	-	0.108
Lead	Pb	-	0.128

Although the analysis for HDCR5L8 revealed only ~ 0.24 wt.% of Pb and PbO_2, their presence confirms the added lead powder. The discrepancy between the analysed result and the expected result (5 wt.%) may be attributable to the inherent uncertainly in the analysis because other phases vary quite significantly as well, especially for portlandite which was analysed to be 10 times more abundant in HDCR5L8 than in HDCR5. Also, it may be because lead powder the fine power obtained from the large piece of fabricated shielding remained at the bottom during analysis because of its higher density, so other phases present in the high-density concrete effectively shielded lead powder.

Although discrepancy between the analysed EDS and XRD results and the expected result occurred, both analytical techniques confirmed the presence of the added lead powder and the expected phases.

IV. CONCLUSIONS

The cracking cause of neutron and gamma ray shielding material composing of high-density cement mixed with 5% crumb rubber and various percentages of lead powder was evaluated. It was found that with the lead content of at least 4 wt.%, the shielding material cracked under neutron and gamma-ray irradiation. Furthermore, experimental tests revealed that lead particles present in the shielding behaves similar to “air pockets.” Under neutron and gamma-ray irradiation, stresses are generated in the shielding material and these “air pockets” represent weak areas of the material, resulting in generation and propagation of cracks. To prevent the cracking problem with the presence of lead powder, the entire surface of each lead particle must be treated to make it to bond strongly with the shielding material. These data will be helpful in developing this class of radiation shielding material to prevent the cracking problem.

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