p ISSN 1907-0322
e ISSN 2527-6433

Experimental Study of Concrete Composites of Fly Ash and Ferronickel Slag for Gamma-Ray Shielding

Studi Eksperimen Komposit Beton dari Fly Ash dan Ferronickel Slag sebagai Perisai Radiasi Gamma

P. M. Wongso[1]*, S. Dewang[1], B. Y. E. B. Jumpeno[2], O. A. Firmansyah[2], J. Mellawati[2]

[1] Physics Department, Faculty of Math and Science, Hasanuddin University
Jl. Perintis Kemerdekaan, KM. 10, Tamalanrea, Makassar, Indonesia
[2] Center for Technology of Radiation Safety and Metrology (PTKMR), National Nuclear Energy Agency
Jl. Lebak Bulus Raya No. 49, Jakarta 12440, Indonesia
[*] E-mail : patrickjilc@yahoo.co.id

ABSTRACT

This study aim is to determine the effect of partial replacement of cement by high percentages of fly ash and ferronickel slag as gamma radiation shielding properties of concrete. The percentages of replacement were 30% for fly ash and 10%, 20%, and 30% for ferronickel slag by weight of fine aggregate. Several physical parameters (linear attenuation coefficients, mass attenuation coefficients, half value layer, tenth value layer, mean free path, effective atomic number, and effective electron density) of concretes were measured using 600cc Thin Window Ionization Chamber. A broad beam transmission geometry method with ^{137}Cs source was used for the radiation intensity measurements. The elemental compositions of the concretes were analyzed by using an energy dispersive X-ray Fluorescence Spectrometer (EDXRF). In this experiment, the concrete composite sample with composition of 0% fly ash and 30% ferronickel slag shows the most effective result in absorbing low energy gamma rays, therefore it has the potential as a candidate for gamma radiation shielding.

Keywords: shield effectiveness, fly ash, ferronickel slag, XRF

ABSTRAK

Penelitian ini bertujuan untuk mengetahui pengaruh penggantian sebagian semen dengan persentase fly ash dan ferronickel slag pada sifat perisai radiasi gamma dari beton. Persentase penggantian adalah 30% untuk fly ash dan 10%, 20%, dan 30% untuk ferronickel slag berdasarkan berat agregat halus. Beberapa parameter fisika (koefisien atenuasi linier, koefisien atenuasi massa, half value layer, tenth value layer, mean free path, nomor atom efektif, dan densitas elektron efektif) dari beton diukur menggunakan 600cc Thin Window Ionisation Chamber. Metode Broad Beam Geometry dengan sumber ^{137}Cs digunakan untuk pengukuran intensitas radiasi. Komposisi unsur dari beton dianalisis dengan menggunakan Energy Dispersive X-ray Fluorescence Spectrometer (EDXRF). Dalam eksperimen ini, sampel komposit beton dengan komposisi fly ash 0% dan ferronickel slag 30% menunjukkan hasil yang paling efektif dalam menyerap radiasi rendah, sehingga sampel ini berpotensi dijadikan kandidat untuk perisai radiasi gamma.

Kata kunci: efektivitas perisai, fly ash, ferronickel slag, XRF

p ISSN 1907-0322
e ISSN 2527-6433

INTRODUCTION

The use of X-rays and gamma rays in nuclear technology and development have increased in various aspects of life, such as disease diagnosis, industry, agriculture, nuclear diagnostics, radiation biopsy, crystallography, spectrometry, and radiation protection [1],[2]. In connection with the use of radiation, knowledge of the interaction of X-rays and gamma rays with material is important in terms of radiation protection [1]. Protection against radiation is very important in order to minimize the impact that will occur from radiation exposure.

High Z materials such as lead and its derivatives (lead sulfate, lead chloride, lead bromide, lead iodide, etc.) are widely used as gamma radiation shields in different technological applications. Nevertheless, lead and its derivatives have toxic effects that can threaten human health [1],[2]. Therefore, shielding technologists try to develop another type of gamma radiation protection material to replace the use of lead.

Utilization of various types of recycled materials, waste, or byproducts shows good prospects economically and/or ecologically as an alternative for radiation shielding materials. By product uses for instance of slag can also be used as an aggregate in cement and concrete mortars. Introducing alternative materials for cement can solve problems related to aggregate mining, waste disposal, and reduce the cost of concrete production for construction [3].

H. Singh, et al utilized fly ash to replace the use of clay in bricks. The results obtained are economical bricks, because fly ash can replace its composition by weight up to 50%, feasible, eco-friendly and can be used as a shield against low energy gamma radiation [4]. A. K. Saha and P. K. Sarker utilized ferronickel slag (FNS) as fine aggregate and fly ash as a substitute for cement with a concentration of 30%, a cement replacement by fly ash was shown to successfully mitigate the deleterious expansion due to alkali-silica reaction of the FNS [5].

The combination of concrete from fly ash and FNS can be a decent alternative for concrete products in terms of its mechanical strength. However, as a shield to X-ray radiation or gamma ray, the properties of the concrete from FNS has never been tested, thus, this paper presents a study on concrete utilizing two industrial by-products by evaluating it's attenuation properties as gamma ray shielding.

THEORETICAL BACKGROUND

The characteristics of gamma ray attenuation from the shielding material can be tested by calculating various physical parameters such as linear attenuation coefficient (μ), half-value layer (HVL), tenth-value layer (TVL), and mean free path (MFP). These are important parameters in the characterization of gamma radiation in a material. These parameters differ depending on the incident radiation energy, mass density and atomic number of the material.

The linear attenuation coefficient can be determined by the Beer-Lambert law [6]:

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

where μ is the linear attenuation coefficient of radiation, x is the thickness of the absorber, I_0 is the intensity of radiation without absorber, and I is the intensity of the radiation transmitted through the absorber with thickness x [7]. For this law to be valid, the photon rays must be a parallel and mono-energetic. If there is more than one different element in material, then the attenuation coefficient can be determined using a mixture rule [1].

Equation (1) is valid only in a narrow beam geometry, when a collimated beam of radiation is used. However, in a broad beam geometry, we must take into account the buildup factor, hence Equation (2) will give us a more realiable value [8].

$$I = B I_0 e^{-\mu x} \quad (2)$$

The mass attenuation coefficient, μ_m, can be calculated by the equation:

$$\mu_m = \frac{\mu}{\rho} = \sum w_i \left(\frac{\mu}{\rho}\right)_i \quad (3)$$

where, w_i, is the mass fraction, and, ρ, is the density [9].

The half value layer (HVL) or tenth value layer (TVL) for a material is the minimum thickness of the absorber to reduce gamma radiation by half and one tenth the incident radiation intensity. This value can be calculated by equation [3]:

$$HVL = \frac{ln2}{\mu} \quad (4)$$

$$TVL = \frac{ln10}{\mu} \quad (5)$$

Mean free path (MFP) is defined as the average distance traveled between two photon interactions, with mathematical equations [1]:

p ISSN 1907-0322
e ISSN 2527-6433

$$MFP = \frac{1}{\mu} \quad (6)$$

The density of the effective atomic number and the effective electron number can be calculated by the equation:

$$Z_{eff} = \frac{\sigma_a}{\sigma_e} \quad (7)$$

$$N_{eff} = \frac{\mu_m}{\sigma_e} = \frac{N_A}{M} Z_{eff} \sum n_i \quad (8)$$

where N_A is Avogadro's number, and σ_a, and σ_e is the cross sections of the total atoms and electrons [10]:

$$\sigma_a = \frac{\mu_m N}{N_A} \quad (9)$$

$$\sigma_e = \frac{1}{N_A} \sum \frac{f_i N_i}{Z_i} (\mu_m)_i = \frac{\sigma_a}{Z_{eff}} \quad (10)$$

MATERIALS AND METHODS

Materials and equipments

In this study, portland cement (OPC) was used as a concrete binder, fly ash as a partial replacement for cement, ferronickel slag with a maximum size of 2 mm as fine aggregate, and destilled water to made concrete samples.

Intensity was measured using 600cc Thin Window Ionization Chamber Type 2575C SER Cap B detector, PTW UNIDOS Webline Electrometer reader, and ^{137}Cs OB.6 gamma radiation source with photon energy of 662 keV and activity of 74 GBq made by Buchler GmbH. Element mass fraction was measured using XRF Analyzers instrument.

Preparation of samples

The compositions of concrete samples are seen in Table 1. Ferronickel slag was used as much as 10%, 20%, and 30% by weight in the overall mass of concrete before mixing. 0% fly ash was used in the first three mixes and 30% was used in the other three mixes. The water-cement ratio in the sample is made constant for all mixes, with a ratio of 0.42 [11],12]. The materials were homogenized and casted with sample dimensions of 1 cm in thickness, 8 cm in length, and 7 cm in width. Each sample was demolded one day after casting at room temperature, then the concrete sample was cured with water for 28 days.

Table 1. Concrete samples composition

Sample Labels	Material Composition (%)		
	Cement	Fly ash	Ferronickel slag
S1 (PC-FNS10)	90	0	10
S2 (PC-FNS20)	80	0	20
S3 (PC-FNS30)	70	0	30
S4 (FA-FNS10)	60	30	10
S5 (FA-FNS20)	50	30	20
S6 (FA-FNS30)	40	30	30

Samples testing with ^{137}Cs gamma source

Broad beam geometry method was used in the experiment. To compensate with the scattering radiation, the sample was designed like a thin slab and the attenuator-to-detector distance was set as in the Figure 1. Background radiation was measured, then gamma radiation without the sample was measured, and finally, gamma radiation was measured from each sample three times with six variations of composition (S1, S2, S3, S4, S5, S6).

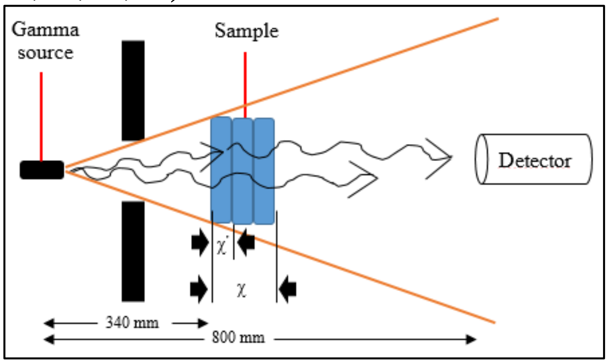

Figure 1. Sample testing scheme with ^{137}Cs gamma source

Sampels testing with X-ray fluorescence (XRF)

Concrete samples of S1, S3, S4, and S6 were grinded to pass the 200-mesh filter. The samples were put into cuvettes with a diameter of 1,5 cm and a height of 2,5 cm, and then tested with the XRF Analyzer. Table 2. Shows the percentage of mass of concrete sample elements used in this study. These four samples already represent the data needed to find out what the differences in the composition of the material elements are, if the material in the sample is reduced or added.

RESULTS AND DISCUSSION

X-ray fluorescence (XRF) test results

X-ray Fluorescence (XRF) testing on concrete samples S1, S3, S4, and S6 were carried out to identify the weight percentage of major and minor constituents which was then processed using WinXCom [13], the mass fraction of each element in the samples are presented in Table 2.

p ISSN 1907-0322
e ISSN 2527-6433

Table 2. Mass fraction values of each element in concrete samples

Elements	Mass Fraction			
	S1	S3	S4	S6
Oxygen	0.336951	0.359230	0.364091	0.379747
Aluminum	-	0.010427	0.015718	0.014712
Silicon	0.097562	0.101206	0.137096	0.142092
Sulfur	-	0.020747	-	0.017020
Potassium	0.005562	0.005147	0.007222	0.006890
Calcium	0.510976	0.447283	0.411296	0.373761
Titanium	0.001756	0.001607	0.004364	0.003884
Chromium	0.000342	0.001430	0.001033	0.000958
Iron	0.043438	0.050502	0.056023	0.058049
Zinc	0.000550	0.000359	0.000366	0.000360
Strontium	0.002148	0.001683	0.001826	0.001683
Zirconium	-	-	0.000444	0.000444
Niobium	0.000236	0.000174	0.000204	0.000145
Molybdenum	0.000177	0.000109	0.000131	0.000079
Ruthenium	0.000059	-	0.000004	-
Rhodium	0.000051	-	-	-
Indium	0.000063	0.000051	0.000063	0.000052
Tin	0.000065	-	0.000050	0.000060
Antimony	0.000063	0.000044	0.000068	0.000064

Gamma-ray attenuation properties

Concrete with cement binder has a greater density than concrete with a mixture of fly ash, this is because fly ash has a relatively low density, this value can be used to calculate the mass attenuation coefficient of concretes. The intensity of radiation without a shield and with a shield has been measured experimentally to obtain the values of μ, μ_m, HVL, TVL, and MFP from each concrete sample from fly ash and ferronickel slag which can be seen in Table 3.

Table 3. Calculation values of ρ, μ, μ_m, HVL, TVL, and MFP

Samples	ρ (g/cm³)	μ (1/cm)	μ_m (cm²/g)	HVL (cm)	TVL (cm)	MFP (cm)
S1	1.831	0.1436	0.078	4.83	16.0	6.963
S2	1.922	0.1467	0.076	4.72	15.7	6.817
S3	2.096	0.1580	0.075	4.39	14.6	6.328
S4	1.734	0.1310	0.076	5.29	17.6	7.631
S5	1.808	0.1395	0.077	4.97	16.5	7.171
S6	1.948	0.1457	0.075	4.76	15.8	6.862

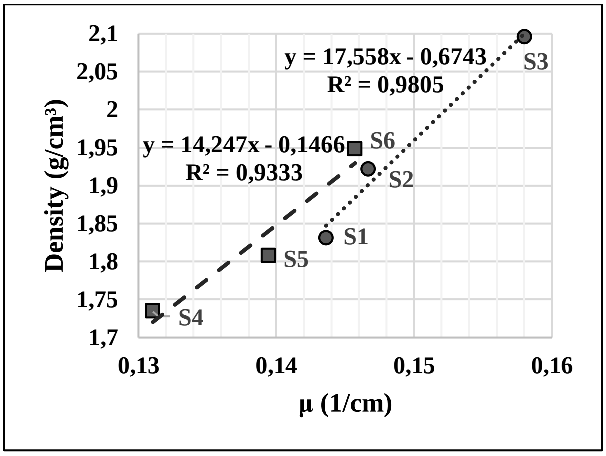

Figure 2. Relationship graph of density and linear attenuation coefficient

The value of the linear attenuation coefficient increased with the addition of ferronickel slag concentration, and decreased when fly ash was added. This happens because the addition of ferronickel slag increases the density of the concrete, while the addition of fly ash decreases the density of the concrete. The relationship between density and linear attenuation coefficients can be seen in Figure 2.

Based on Table 3, it is clear that the mass attenuation coefficient of each concrete sample shows a constant value. Although the concrete density increases with the addition of ferronickel slag and decreases with the addition of fly ash, the attenuation coefficient does not change significantly because the attenuation coefficient of the mass does not affect the density of the material, but depends on the chemical composition of the material.

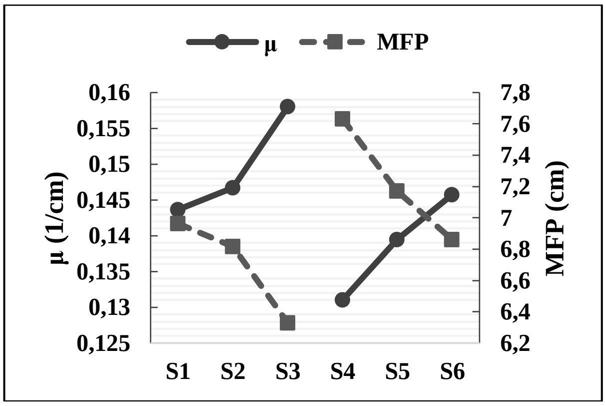

Figure 3. Relationship graph of linear attenuation coefficient and MFP of concrete

p ISSN 1907-0322
e ISSN 2527-6433

Mean free path (MFP) represents the average distance traveled between two successive photon interactions. The shorter MFP indicates more interaction of photons to material, and hence the ability of the shield to attenuate radiation is better. The MFP value is the opposite of the measurement of the linear attenuation coefficient. In Figure 3, based on the sample with no addition of fly ash and with the addition of fly ash. The smallest MFP values are obtained in sample S3 and S6, with the values of 6,328 cm and 6,862 cm respectively, which indicate that the composition of sample S3 and S6 have good effectiveness.

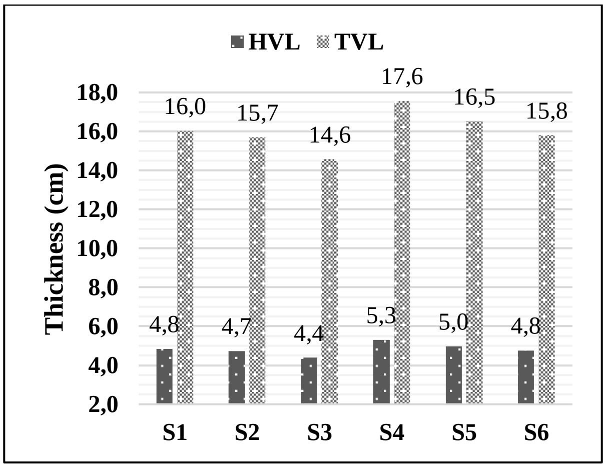

Figure 4. HVL and TVL values

The effectiveness of the shield of concrete samples affects the value of HVL and TVL, the lower the value of HVL and TVL, the better the material in attenuating radiation. The addition of ferronickel slag concentration showed a decrease in the value of HVL and TVL, which means the effectiveness of the shield is getting better. The HVL and TVL values are inversely proportional with the density, for that, S3 with high density showed lower values of HVL and TVL than that of S4 and consequently, S3 is remarkably effective for gamma radiation shielding.

Table 4. Calculation values of μ_m, σ_a, σ_e, Z_{eff} and N_{eff}

Samples	μ_m (cm²/g)	σ_a (barn/mol)	σ_e (barn/mol)	Z_{eff}	N_{eff}
S1	0.0774	0.3356	0.0258	13.025	3.0031
S3	0.0773	0.3259	0.0258	12.655	3.0008
S4	0.0772	0.3220	0.0258	12.503	2.9980
S6	0.0772	0.3162	0.0258	12.278	2.9979

The value of the mass attenuation coefficient theoretically is calculated from the Phy-X/PSD software [14], the calculation results show values that are almost similar to the values of the mass attenuation coefficient experimentally, the values of σ_a and σ_e are obtained from calculations with equations 9 and 10. In Table 4, as expected, the effective atomic number of all concretes were obtained between the atomic number of O (Z=8), Si (Z=14) and Ca (Z=20). The highest value of Z_{eff} is observed for S1, while the lowest value is observed for S6, the cause for these observations is undoubtedly because the elemental abundance of Ca and O elements. The associate parameter to the effective atomic number, used for energy deposition by photons in biological shielding is effective electron density. The effective electron density is practically constant for all concrete samples in the range of $2.9979\text{-}3.0031\times10^{23}$ electrons per gram.

CONCLUSION

Radiation shielding parameters in concrete composites from ferronickel slag and fly ash were investigated to obtain an effective shield. The composition of S3 concrete without fly ash and S6 with fly ash showed good effectiveness as gamma radiation shields, but the composition of S3 concrete had more potential as a gamma radiation shield. This study shows that the use of concrete from fly ash and ferronickel slag can be beneficial to address the issues of radiation shield, cost effectiveness, waste management and the disposal of byproducts in a useful manner.

ACKNOWLEDGEMENTS

The authors would like to acknowledge Center for Technology of Radiation Safety and Metrology (PTKMR) National Nuclear Energy Agency of Indonesia (BATAN) for facilities provided and assistance received to conduct this study.

REFERENCES

[1]. F. Akman *et al.*, “Study of gamma radiation attenuation properties of some selected ternary alloys,” *J. Alloys Compd.*, vol. 782, pp. 315–322, 2019.

[2] M.I. Sayyed *et al.*, Radiation Protective

p ISSN 1907-0322
e ISSN 2527-6433

qualities of some selected lead and bismuth salts in the wide gamma energy region," *Nucl. Eng. Technol.*, vol. 51, no. 3, pp. 860–866, 2019.

[3] A.S. Ouda and H.A. Abdel-Gawwad, "The effect of replacing sand by iron slag on physical, mechanical and radiological properties of cement mortar," *HBRC J.*, vol. 13, no. 3, pp. 255–261, 2017.

[4] H.S. Mann *et al.*, "Experimental investigation of clay fly ash bricks for gamma-ray shielding," *Nucl. Eng. Technol.*, vol. 48, no. 5, pp. 1230–1236, 2016.

[5] A.K. Saha and P.K. Sarker, "Sustainable use of ferronickel slag fine aggregate and fly ash in structural concrete: mechanical properties and leaching study," *J. Clean. Prod.*, vol. 162, pp. 438–448, 2017.

[6] M.R. Kaçal, F. Akman, and M.I. Sayyed, "Evaluation of gamma-ray and neutron attenuation properties of some polymers," *Nucl. Eng. Technol.*, vol. 51, no. 3, pp. 818–824, 2019.

[7] D.R. Mcalister, "Gamma ray attenuation properties of common shielding materials," *PG Research Foundation, University Lane Lisle USA*, 2012.

[8] D.N. Hopkins, M. Maqbool, and M.S. Islam, "Linear attenuation coefficient and buildup factor of MCPS-96 alloy for dose accuracy, beam collimation, and radiation protection," *Radiol. Phys. Technol.*, vol. 5, no. 2, pp. 229-236, 2012.

[9] H.S. Mann, G.S. Brar, and G.S. Mudahar, "Gamma-ray shielding effectiveness of novel light-weight clay-flyash bricks," *Radiat. Phys. Chem.*, vol. 127, pp. 97–101, 2016.

[10] N. Abbasova *et al.*, "Investigation of gamma-ray attenuation parameters of some materials used in dental applications," *Results Phys.*, vol. 12, no. November 2018, pp. 2202–2205, 2019.

[11] P.C. Aïtcin, "Science and Technology of concrete admixture (chapter 1): The importance of the water-cement and water-binder ratios," Elsevier Ltd, pp. 483-487, 2016.

[12] Badan Standarisasi Nasional, "Tata cara pemilihan campuran untuk beton normal, beton berat dan beton massa SNI 7576-2012."

[13] L. Gerward *et al.*, "WinXCom - A program for calculating X-ray attenuation coefficients," vol. 71, pp. 653-654, 2004.

[14] E. Sakar *et al.*, "Phy-X / PSD: development of a user friendly online software for calculation of parameters relevant to radiation shielding and dosimetry," *Radiat. Phys. Chem.*, p. 108496, 2019.