Shielding and Radiological Properties Analysis of Pozzolana-Portland Cement (PPC) from Elemental Composition Point of View, Using Neutron Activation Analysis (NAA)

B. J. A. Y. Sogbey[1,2], S. Kwofie[1], E.O. Darko[3], N. Allotey[2]

[1]Department of Materials Engineering, Kwame Nkrumah University of Science and Technology, Kumasi Ghana
[2]National Nuclear Research Institute; Ghana Atomic Energy Commission, P.O. Box LG 80 Legon, Accra, Ghana
[3]Radiation Protection Institute; Ghana Atomic Energy Commission, P.O. Box LG 80 Legon, Accra, Ghana
Email: sobright1knust@yahoo.com

Abstract

The physical properties of the ordinary Portland cement (OPC) are enhanced when the matrix of the OPC is partially replaced by admixtures such as Pozzolana. This research investigated the radiological shielding properties. Neutron activation analysis (NAA) was carried out to determine the elemental compositions of Pozzolana and ordinary Portland cement. The neutron activation analysis results showed that Europium, Hafnium, Copper, Calcium, Iron, Cobalt and Aluminum, all of which have significant macroscopic cross sections (shielding properties) dominating more in Pozzolana cement than in the ordinary Portland cement.

1.0 INTRODUCTION

1.1 Microscopic Cross Sections

The nuclear cross section of a nucleus is used to characterize the probability that a nuclear reaction will occur. The concept of a nuclear cross section can be quantified physically in terms of "characteristic area" where a larger area means a larger probability of interaction. The standard unit for measuring a nuclear cross section (denoted as σ) is the barn, which is equal to 10^{-28} m² or 10^{-24} cm². Cross sections can be measured for all possible interaction processes together, in which case they are called total cross sections, or for specific processes, distinguishing elastic scattering and inelastic scattering; of the latter, amongst neutron cross sections the absorption cross sections are of particular interest. Types of reactions frequently encountered are s: scattering, γ: radiative capture, a: absorption (radiative capture belongs to this type), f: fission, the corresponding notation for cross-sections being: σ_s, σ_γ, σ_a, etc. A special case is the total cross-section σ_t, which gives the probability of a neutron to undergo any sort of reaction ($\sigma_t = \sigma_s + \sigma_\gamma + \sigma_f + ...$).

In nuclear physics, it is conventional to consider the impinging particles as point particles having negligible diameter. Cross sections can be computed for any sort of process, such as capture scattering, production of neutrons, etc. In many cases, the number of particles emitted or scattered in nuclear processes is not measured directly; one merely measures the attenuation produced in a parallel beam of incident particles by the interposition of a known thickness of a particular material. The cross section obtained in this way is called the total cross section and is usually denoted by a σ_t or σ_T.

The typical nuclear radius is of the order of 10^{-12} cm. We might therefore expect the cross sections for nuclear reactions to be of the order of πr^2 or roughly 10^{-24} cm^2 and this unit is given its own name, the barn, and is the unit in which cross sections are usually expressed. Actually the observed cross sections vary enormously. Thus for slow neutrons absorbed by the (n, gamma) reaction the cross section in some cases is as much as 1,000 barns, while the cross sections for transmutations by gamma-ray absorption are in the neighborhood of 0.001 barns (Perkins, 1999).

Nuclear cross sections are used in determining the nuclear reaction rate, and are governed by the reaction rate equation for a particular set of particles (usually viewed as a "beam and target" thought experiment where one particle or nucleus is the "target" [typically at rest] and the other is treated as a "beam" [projectile with a given energy]).

For neutron interactions incident upon a thin sheet of material (ideally made of a single type of isotope), the nuclear reaction rate equation is written as:

$$\mathbf{r_x = \Phi\, \sigma_x\, \rho_A = \Phi \sum} \qquad \textbf{(1)}$$

or

$$\mathbf{\Phi(x) = \Phi_o e^{-\Sigma T X}} \qquad \textbf{(2)}$$

where:

- r_x **:** number of reactions of type x, units: [1/time/volume]
- Φ : neutron beam flux intensity through an absorber of thickness x,[1/area/time]
- Φ_0 = neutron beam flux intensity at zero absorber thickness
- σ_x : microscopic cross section for reaction x, units: [area] (usually barns or cm^2).
- ρ_A : density of atoms in the target in units of [1/volume]
- $\Sigma_T = \sum_{\mathbf{s}} + \sum_a \equiv \sigma_x\, \rho_A =$: total macroscopic cross-section [1/length]
- $\sum_s =$ macroscopic cross-section for scattering
- $\sum_{\mathbf{a}} =$ macroscopic cross-section for absorption
- e = base of the natural logarithm system
- x = thickness of materials

Formally, the equation above defines the macroscopic neutron cross-section (for reaction x) as the proportionality constant between a neutron flux incident on a (thin) piece of material and the number of reactions that occur (per unit volume) in that material. The distinction between macroscopic and microscopic cross-section is that the former is a property of a specific lump of material (with its density), while the latter is an intrinsic property of a type of nuclei. The table below shows some elements with their total macroscopic cross sections.

Table 1: Elements or Compounds & Their Macroscopic Cross Sections for neutrons

Elements or Compound	Density [g/cm^3]	Macroscopic Cross Section, $\sum$ [cm-^{1}]
H_2O	1	3.472
Co	8.9	4.097
Hf	13.3	5.069
Sc	2.5	1.608
Mn	7.2	1.221
Pb	11.35	0.369
Fe	7.9	1.155
Eu	5.22	89.166
Concrete	2.3	**0.240**

Source: (Glasstone and Sesonske, 1967).

The table above showed that concrete (made from OPC) has a shielding property, the macroscopic cross section to be **0.240.**

Table 1b: Total Mass and Linear Attenuation Coefficients [$\frac{\mu}{\rho}$], (cm^2/g) μ_l (cm-^{1}) for Some Materials at 0.1 MeV, gamma energy

	Al	Fe	Ca	K	Na	Pb	H_2O	Concrete
$\frac{\mu}{\rho}$ [cm^2/g]	0.161	0.344	0.238	0.215	0.151	5.29	0.167	0.169
μ_l [cm-^{1}]	0.435	2.718	0.369	0.187	0.147	59.94	0.167	0.389
ρ[g/cm^3]	2.7	7.90	1.55	0.87	0.971	11.35	1	2.3

Source: (Cember, 2009).

Table 2: Chemical Composition of Portland and Pozzolana Cement (%wt)

	Chemical Composition (% wt)	
Compound	**Pozzolana**	**Portland**
SiO	46.25	27.43
Al_2O_3	17.34	5.4
Fe_2O_3	**10.26**	**3.48**
CaO	10.18	53.71
MgO	2.9	1.41
K_2O	1.64	0.92
Na_2O	3.64	0.16
SO_3	0.8	2.59
Cl^-	0.01	0.004

Source: [SBEIDCO, (2009)]

Table 2 showed that Pozzolana has more Fe content than OPC. The significant point here is that Fe contributes to shielding property in concrete.

1.2 Neutron Activation Analysis (NAA)

Neutron activation analysis (NAA) is a physical method of analysis of material for elemental composition. This was first identified by George Hevesy and N. Levy after analysis of dysprosium in rare earth using an isotopic neutron source. (Rezaei et al, 2007, Mesima, 2008). This is the most common form of activation analysis. It is a sensitive analytical technique useful for performing both qualitative and quantitative multi-element analysis of elements in samples from almost every conceivable field of scientific or technical interest (Glascock, 2004). NAA can be grouped into three main categories. These are Instrumental Neutron Activation Analysis (INAA) if no chemical treatment is done to obtain the radionuclide of interest; if chemical separation is done after irradiation to remove interference the technique is called Radiochemical Neutron Activation Analysis (RNAA), and when pre-irradiation chemical separation is employed, the procedure is called Chemical Neutron Activation Analysis (CNAA) (Zimbal, 2007).

1.2.1 Methods of quantification of amount of element in a sample

Two methods of quantifying elements in samples during activation analysis are described.

1.2.2 Absolute method

This method enables the number of atoms of the target nuclide and hence the weight of the elements present to be calculated based on the knowledge or the measurability of t, σ, ϕ, λ and $A_{i.}$ The uncertainties in the nuclear constants (ϕ and σ) and the necessity of measuring absolute disintegration rates make the absolute activation analysis method quite difficult to use (Quarshie, 2008, Adotey, 2003).

1.2.3 Comparative method

The comparative activation analysis method involves irradiation and counting of a sample and a standard(s) containing a known amount of the element(s) of interest under identical conditions (same neutron flux and same counting geometry). Under such conditions the following equations apply (Quarshie, 2008).

$$A_S = N_S \sigma\phi\, (1-\ell^{-\lambda t}) \qquad (3)$$

$$A_R = N_R \sigma\phi\, (1-\ell^{-\lambda t}) \qquad (4)$$

where:
A_S = activity (disintegrations/sec) of the nuclide in the sample at the end of irradiation.
A_R = activity (disintegrations/sec) of the nuclide in the standard at the end of irradiation.
Hence;

$$\frac{A_S}{A_R} = \frac{N_S}{N_R} = \frac{W_S}{W_R} = \frac{C_S}{C_R} \qquad (5)$$

where;
WS = weight of the element in the sample in μg
WR = weight of the element in the standard in μg
CS = measured activity of nuclide in the sample
CR = measured activity of the nuclide in the standard
Therefore,

$$\textbf{Concentration of element (μg/g)} = \frac{C_S W_R}{C_R W} \qquad (6)$$

where W = weight of sample in grams

2.0 Materials and Methods

The Pozzolana was obtained from BRRI-CSIR, Kumasi. **ASTM type-1 OPC used** as reference was taken from the cement factory (GHACEM). The chemical composition of Pozzolana and OPC used in the present work are as shown in Table 1.

3.0 Sample Preparation and Analysis

Each samples weighing about 0.1g was placed on a polyethylene of 1mm wall thickness, wrapped and then heat sealed. The wrapped samples were then concealed in rabbit capsules of height 5 mm and external diameter of 1.4 mm, this was then stuffed with cotton and then heat sealed. For long and medium-lived irradiation, all the samples were concealed in one capsule, stuffed with cotton wool and then heat sealed. For short-lived irradiation, each sample was placed in one individual capsule.
The samples were sent into the inner irradiation sites of the reactor for thermal irradiation by means of pneumatic transfer system operating at 65 psi. Three types of

irradiations schemes were used: one to determine short-lived radionuclides (10sec) and other medium (1hr) or long-lived radionuclides (above 1hr). The samples for medium-lived were sent into the reactor for irradiation time of one hour and left overnight to decay before counting . The samples for long-lived were sent into the reactor for irradiation time of more than one hour and left for two weeks to decay before counting .
In the same manner, the samples for short-lived radionuclides were irradiated one after the other in their individual capsules for ten seconds and the counting done immediately.
SL-7 (IAEA Standard Comparator) was used for the Pozzolana composition while GBW-7 (Chinese Standard Comparator) was used for the OPC compositional analysis.

4.0 RESULTS AND DISCUSSION

Neutron Activation Analysis of OPC and Pozzolana Results
Table 3: Compositional Percentage of OPC

(Neutron Activation Analysis: Ghana Atomic Energy Commission)

Elements	**Concentration in mg/kg**	**Composition (%wt)**
AI	**77,463.5 ± 3,063.2**	**11**
Ca	**609,355 ± 30,100**	**87**
Cu	**44.42 ± 2.21**	**0.006**
Fe	**32,217 ± 1,411**	**4.6**
Mg	**45,195 ± 2,117**	**6.5**
Mn	**410.88 ± 20.52**	**0.06**
Ti	**2,737.32 ± 135.57**	**0.4**

Figure 1 below shows the elemental composition in Pozzolana cement, with Fe, followed by Al, being in significant quantity. Figure 2 then shows the respective densities of these elements in Pozzolana and OPC. In Figure 3, common elements found in both Pozzolana and ordinary Portland cements are shown with Fe being the highest with significant shielding properties in Pozzolana. In Figure 4, linear attenuation coefficients of these elements are shown, namely, Fe, Cu, Al, Ca, K and Na with Cu being the highest followed by Fe. Referenced to the cumulative shielding effect of elements, Pozzolana stands to be a better shielding material in comparison with OPC. Figure 5, in another development, shows elements of significant macroscopic cross sections found in Pozzolana cement. These elements are Eu, Hf and Co with Eu having the highest macroscopic cross section putting Pozzolana cement as a better neutron shielding material compared with OPC.

Table 4: Elemental Composition for Pozzolana

Element	Concentration in mg/kg	Composition (%wt)
AI	79570.17±244.73	42
Ca	8803.52±782.4	4.6
Ce	922.01±82.31	0.48
Co	101.90±4.53	0.05
Cr	316.53±30.49	0.17
Cs	286.61±25.96	0.15
Cu	69.56±5.14	0.04
Eu	14.31±2.19	0.008
Fe	88403.64±2375.93	46
Hf	161.40±9.77	0.08
K	8027.281±74.71	4.21
La	17.93±2.44	0.009
Mn	969.04±19.13	0.51
Na	2844.501±17 .54	1.50
Sc	53.43±0.40	0.03
V	83.37±4.15	0.04

(Neutron Activation Analysis: Ghana Atomic Energy Commission)

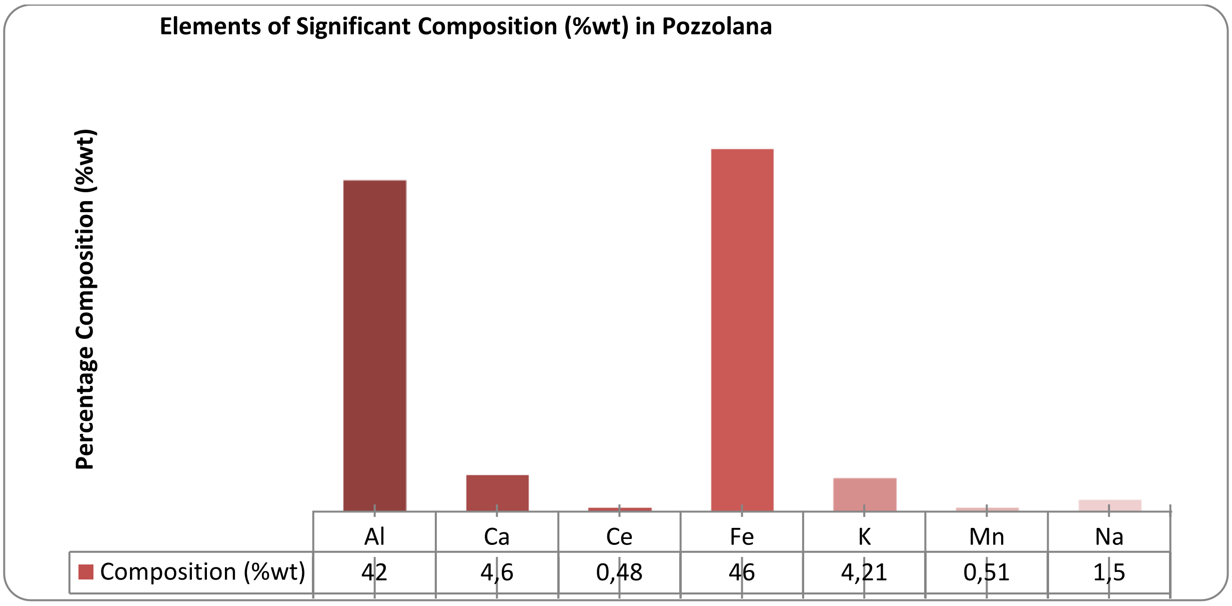

Figure 1: Percentage Composition of Some Significant Elements in Pozzolana

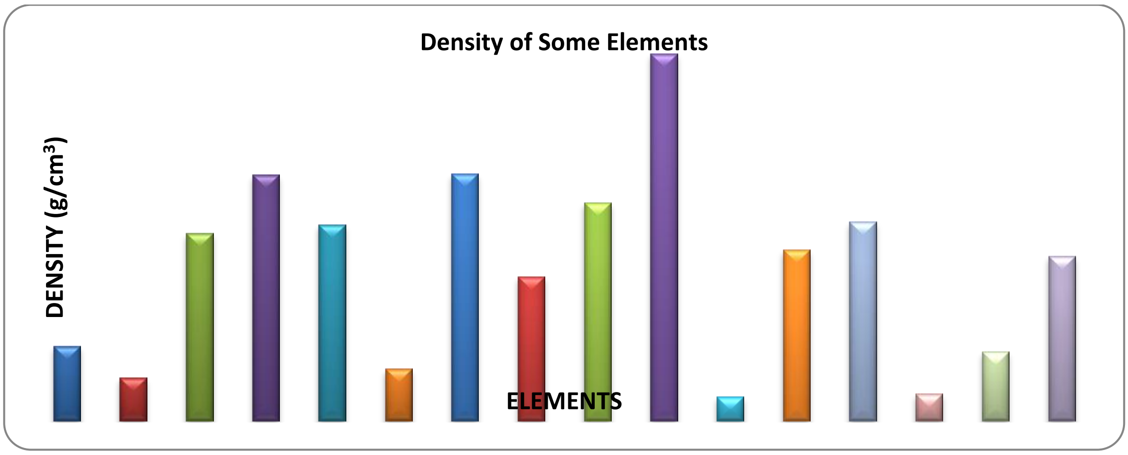

Figure 2: Density of Some Elements in Pozzolana or OPC

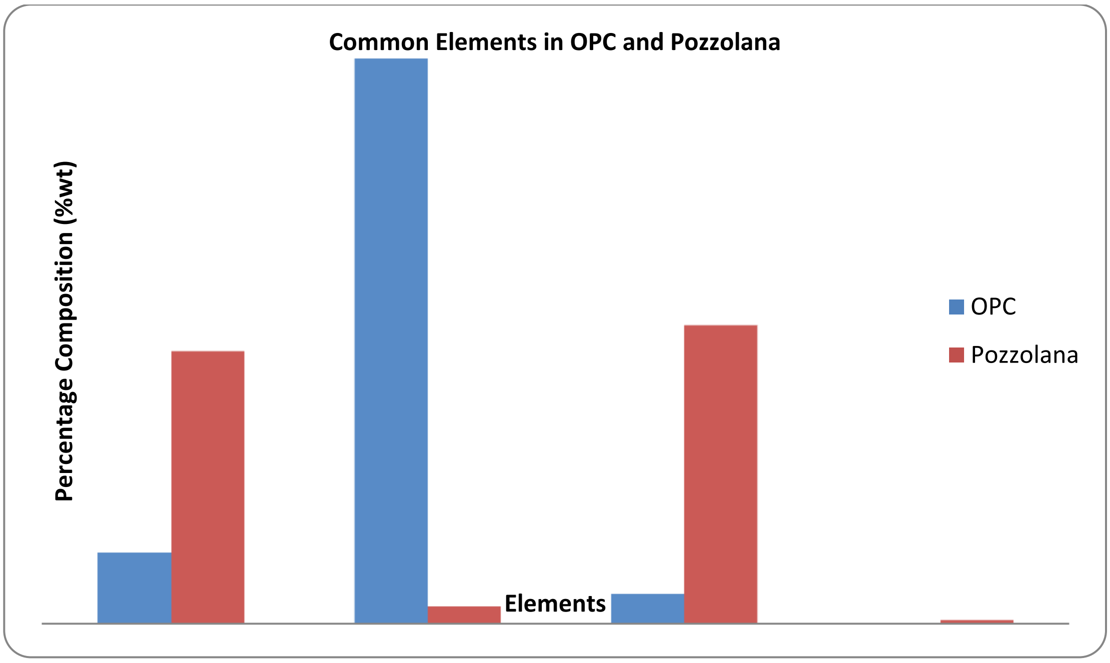

Figure 3: Elements in OPC and Pozzolana of Significant Shielding Characteristics

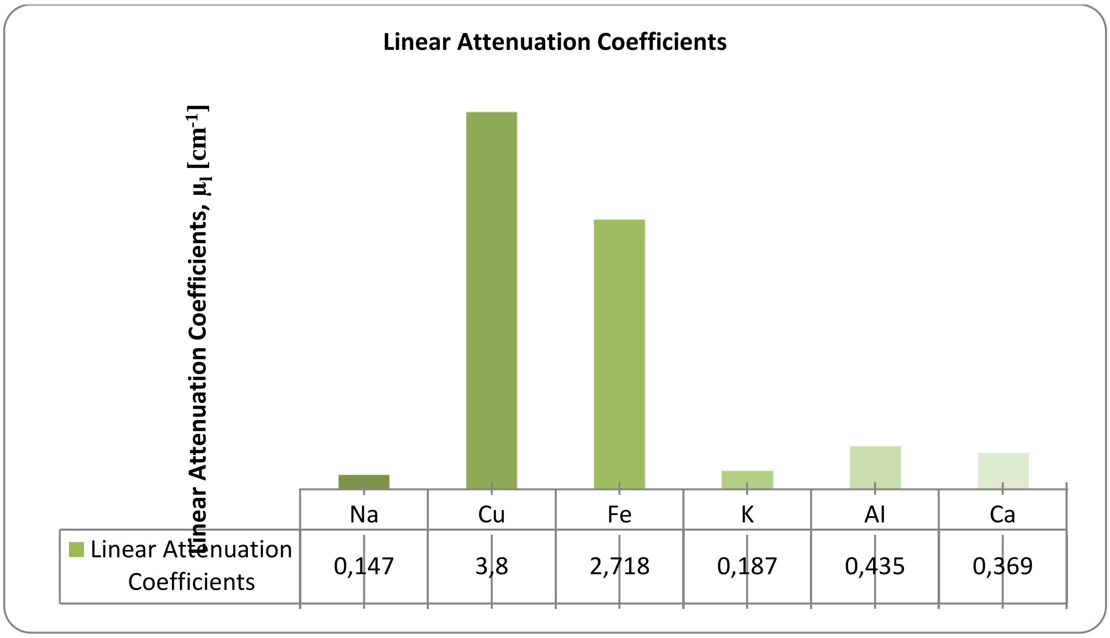

Figure 4: Linear Attenuation Coefficients of Some Elements in Pozzolana or OPC

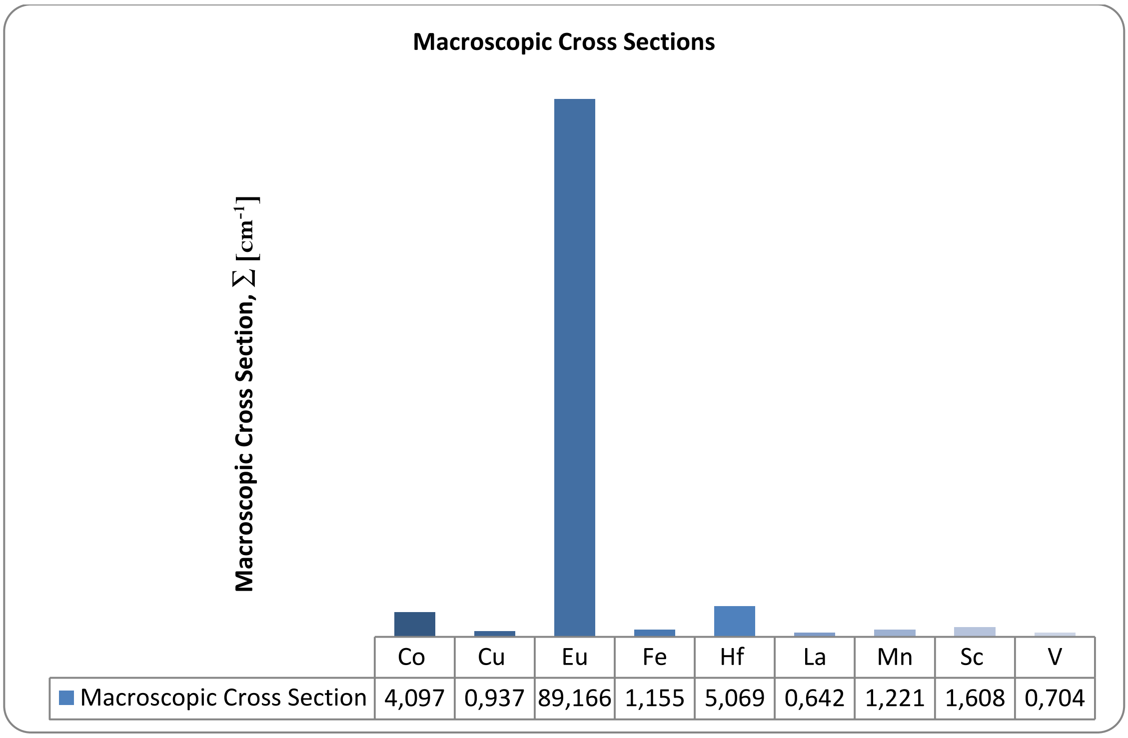

Figure 5: Macroscopic Cross Section of Some Elements in Pozzolana or OPC

Table 5: Macroscopic Cross Section Interpretation for Pozzolana

Elements	**Composition (%wt)**	**Macroscopic Cross Section, ∑ [cm-[1]]**	**(%wt) x (∑)**
AI	42	0.099	0.042
Ca	4.60	0.080	0.004
Ce	0.48	0.284	0.0014
Co	0.05	4.097	0.002
Cr	0.17	0.502	0.0009
Cs	0.15	0.408	0.0006
Cu	0.04	0.937	0.0004
Eu	0.008	89.166	0.00713
Fe	46	1.155	0.5313
Hf	0.08	5.069	0.0041
K	4.21	0.048	0.002
La	0.009	0.642	0.00006
Mn	0.51	1.221	0.006
Na	1.50	0.115	0.0017
Sc	0.03	1.608	0.00048
V	0.04	0.704	0.0003
Total $\sum_T$ [cm-[1]]			**0.6044 cm^{-1}**

Table 6: Macroscopic Cross Section Interpretation for OPC

Elements	**Composition (%wt)**	**Macroscopic Cross Section, ∑ [cm-[1]]**	**(%wt) x (∑)**
Al	11	0.099	0.011
Ca	87	0.080	0.07
Cu	0.006	0.937	0.00006
Fe	4.6	1.155	0.053
Mg	6.5	0.158	0.0103
Mn	0.06	1.221	0.0007
Ti	0.4	0.608	0.0024
Total $\sum_T$[cm-[1]]			**0.15 cm^{-1}**

In calculations, it was determined whether or not Pozzolanic materials have a better shielding property than the ordinary Portland cement (OPC).
For gamma-ray and X-ray shielding analysis, linear attenuation coefficients μ_l, [cm^{-1}] was considered, the higher the μ_l, the better the material for shielding. This relation is given by:

$$\frac{I}{I_O} = \mathbf{B}e^{-\mu x} = \mathbf{B_T} = \frac{60IPd^2}{YWT} \quad \mathbf{(7)}$$

Whereas, for neutron shielding, total macroscopic cross section $\sum_T$, [cm^{-1}] was considered, the higher the $\sum_T$, the better the material for neutron shielding. The relation is given by:

$$\Phi(x) = \Phi_o e^{-\Sigma T X} \quad (8)$$

5.0 CONCLUSION

It is shown in Tables 5 and 6 above, that total macroscopic cross section ($\sum_T$) of **Pozzolana** is greater than that of **OPC** hence Pozzolana is a better shielding material than OPC.

ACKNOWLEDGMENTS

I acknowledge the contribution from Ghana Atomic Energy Commission (GAEC), Messrs I. Lawson and R. E. Quagraine, M. A. Addo, J. K. Gbadago, S. Akligo, N. S. Opata, I. K. Baidoo and F. Anyang all of National Nuclear Research Institute, GAEC.

Mr Nikolas Sosu, Head, Engineering Section, Ghana Standards Board (GSB), Mr. William Nimako, Head, Civil Engineering Laboratory, GSB, Messrs Vincent Agbodemegbe and Said Ibrahim Said all of Department of Nuclear Engineering, University of Ghana: Francis Mudy and Mawuli Tsaku of Civil Engineering Laboratory, GSB. W.O Armah: Dept. of Physics, University of Ghana, Legon and Appropriate Technology Support Division; Science and Technology Advocacy Renaissance Foundation for Africa (STARFA), Ghana.

REFERENCES

1. Cember Herman, Introdution to Health Physics, Fourth Edition, 2009.
2. Farnell CNS, Manor Way, Borehamwood, Herts: PUNDIT plus, Operating manual, 1991.
3. Gladstone Samuel and Alexander Sesonske: Nuclear Reactor Engineering, 1967
4. Glascock M. D., 2004. An overview of Neutron Activation Analysis, University of Missouri Research Reactor.
5. Turner James E.: Atoms, Radiation and Radiation Protection, 3rd Edition, 2007.
6. Zimbal, A. 2007. Measurement of the spectral fluence rate ofreference neutron sources with a liquid scintillation detector, Physikalisch-Technische Bundesanstalt, D-38116 Braunschweig, Germany. Radiation protection dosimetry Advance Access.