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Thermal and gamma radiation shielding properties of geopolymer high density concrete

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Abstract: This paper provides a review about the thermal and radiation shielding properties of geopolymer concrete. Geopolymer concrete is a type of concrete that is made by reacting aluminate and silicate bearing materials with a caustic activator. commonly, waste materials such as fly ash or slag from iron and metal production are used which helps to lead a cleaner environment. Radiation-shielding concrete (RSC) is used in nuclear power plants, health care facilities in conducting radiation therapy, nuclear research facilities, and storage/transport casks for radioactive waste. In RSC mixtures, high-density aggregates are used to attenuate gamma rays and light atomic weight aggregates are used to absorb neutrons, so their properties and proportions will affect a mixture's radiation-shielding characteristics. Heavyweight concrete is extensively used as a shield in nuclear plants, radio therapy rooms and for transporting as well as storing radioactive wastes. For this purpose, concrete must have high strength and density. Heavyweight and high strength concrete can be used for shielding purposes. Such concrete with magnetite aggregates can have a density in the range of 3.2–4 t/m3, which is significantly higher than that with normal aggregates.

***Index Terms* - Geopolymer concrete, Compressive strength, Attenuation coefficient, Silica fumes & Fly ash.**

I. INTRODUCTION

Concrete is by far the most widely used material for reactor shielding due to its cheapness and satisfactory mechanical properties. It is usually a mixture of hydrogen and other light nuclei and has a high atomic number. The aggregate of concrete containing many heavy elements plays an important role in improving concrete shielding properties and therefore has good shielding properties for the attenuation of photons and neutrons (Santhoshkumar, et al., 2013). The density of heavyweight concrete is based on the specific gravity of the aggregate and the properties of the other components of concrete

The development of nuclear technology is one of the most significant achievements of the twentieth century. Nuclear technology is currently being used nearly in every field and aspect of human activity - from medicine, manufacturing, agriculture to producing electricity for over 16% of world-wide needs (Wang, et al., 2007). Particle accelerators such as medical cyclotrons accelerate particles to bombard a target to produce radio-nuclides such as fluorine-18. Such bombardment also produces neutrons and gamma rays which are to be shielded. Cyclotron of low-energy (11-12 MeV proton energy) systems come equipped with their own built-in radiation shielding. Others, however, are higher-energy (16-18 MeV proton-energy) accelerators which are intended for installation in a specialized concrete vault for (Alhajali, et al., 2012) radiation protection.

Radiation-shielding concrete (RSC) is used in nuclear power plants, health care facilities in conducting radiation therapy, nuclear research facilities, and storage/transport casks for radioactive waste. In RSC mixtures, high-density aggregates are used to attenuate gamma rays and light atomic weight aggregates are used to absorb neutrons, so their properties and proportions will affect a mixture's radiation-shielding characteristics. The most common naturally occurring aggregates used in RSC are produced from ores of high-density minerals such as hematite, ilmenite, magnetite, and barite (Lee, et al., 2013). Other aggregates used in RSC include aggregates that contain bound water, produced from ores of hydrous iron, serpentine, or bauxite, and aggregates that contain boron, produced from natural borate ores.

The high-density aggregates used in RSC are produced from ore deposits, which contain metallic opaque phases providing the shielding properties to the RSC. Some of the natural minerals used as aggregates in high density concrete are hematite, magnetite, limonite, barite and some of the artificial aggregates include materials like steel punching and iron shot. Bauxite, hydrous iron ore or serpentine, all slightly heavier than normal weight concrete can be used in case of a high fixed water content (Ravindrarajah, et al., 2002). Heavyweight concrete is extensively used as a shield in nuclear plants, radio therapy rooms and for transporting as well as storing radioactive wastes. For this purpose, concrete must have high strength and density. Heavyweight and high strength concrete can be used for shielding purposes. Such concrete with magnetite aggregates can have a density in the range of 3.2–4 t/m^3, which is significantly higher than that with normal aggregates

The standard fire conditions for concrete structures are defined in the range from 20°C to 1200°C. At a temperature of 40°C, free water from capillary pores begins to lose, and to a much lesser degree, physically bound (gel) water also (Chakrabari, et al., 1994). Furthermore, with the long exposure of concrete to a temperature of 100°C, all free capillary water is lost, at 200°C, all physically bound water is lost, while chemically bound water in cement hydrates is lost at 400°C. When the temperature of reinforced concrete reaches 250°C, the yield strength of steel bars is reduced, and at 500°C it is reduced so much that there is a risk of structure collapse this work also indicates that, in addition to other parameters, the applied type of aggregate in concrete can have a significant impact on its resistance under extreme high temperature conditions (Broceta, et al., 2017). The binder material type had a significant influence on the performance of high-strength concrete particularly at temperatures below 800°C. The influence of the binder material type was significantly decreased at temperature of 1000°C. The strengths and stiffness of high-strength concrete were reduced with the increase in temperature without any threshold temperature level (Ravindrarajah, et al., 2002).

The present work proposes to develop and study, the performance of hematite and magnetite incorporated concrete mixes, as a possible application in radiation shielding. In this study, hematite & magnetite in the form of either small size pellets or angular pebbles as a replacement of coarse aggregates are incorporated in the concrete. The choice of these aggregates enhances the shielding property of concrete due to the presence of higher iron content. These build-up materials increase the density within the concrete. Such build-up concrete mixes are then studied for their mechanical properties in fresh and hardened states and gamma radiation characteristics.

II. Materials and Methods

2.1 Materials

Ordinary Portland cement of 53 grade has been used for all the concrete mixes in this work. Different physical properties are tested and it conforms to IS: 8112 (1989). The fly ash is collected from a local dealer conforming the specifications as laid out for silicious fly ash in IS:3812 (2003) (equivalent to Class F fly ash, ASTM: C618-2012a). Silica fumes are also used as cementitious material. Manufactured sand was used as fine aggregate for concrete. Locally available crushed granite aggregated 20mm down size was used as coarse aggregate. Hematite and magnetite aggregates are collected from iron ore mining sites at hospet, Karnataka, India are shown in Fig.1. Auramix 400 is the chemical admixture collected from Fosroc chemicals (India) Pvt. Ltd which complies with IS:9103-1999(2007). It also complies with ASTM C494 Type G. The basic properties of Coarse aggregate, Hematite & Magnetite aggregates are listed in Table 1.

Fig. 1. Coarse aggregate, Hematite, Magnetite aggregates and sodium silicate

Table 1

Basic test results on aggregates

Test	Coarse agg.	Hematite	Magnetite
Specific gravity	2.73	4.44	3.9
Water absorption, %	1.83	2.67	2.32

Alkali solution preparation

A combination of sodium silicate solution and sodium hydroxide solution/potassium hydroxide solution was chosen as the alkaline liquid. The sodium hydroxide (NaOH) solids were a commercial grade in form of flakes with 97% purity. The sodium hydroxide (NaOH) solution was prepared by dissolving either the flakes or the pellets in water. The mass of NaOH solids in a solution varied depending on the concentration of the solution expressed in terms of molar (M). For instance, NaOH solution with a concentration of 10M consisted of 10x40 = 400 grams of NaOH solids (in flake or pellet form) per litre of the solution, where 40 is the molecular weight of NaOH.

2.2. Mix design

As there are no code provisions for the mix design of geopolymer concrete, the density of geo-polymer concrete is assumed as 2400 Kg/m3. The rest of the calculations are done by considering the density of concrete. The total volume occupied by fine and coarse aggregate is adopted as 77%. The alkaline liquid to fly ash and GGBS ratio is kept as 0.4. The ratio of sodium hydroxide to sodium silicate is kept as 2.5. The conventional method used in the making of normal concrete is adopted to prepare geopolymer concrete.

2.3. Test matrix

Three different concrete mixes were tested herein for their gamma radiation shielding characteristics. One of them was a reference conventional concrete (CC) and the others were hematite and magnetite mixes. 28 cubes of size 100×100×100 mm were cast with each mix to evaluate their compressive strengths at different temperatures. The cube samples were exposed in oven for 2 hours maintained at 150, 300, 450, 600, 750 and 900^{0}C. Also slab specimens with a constant lateral dimension of 100mm×100 mm but with five representative thicknesses (25mm, 50mm, 75mm, 100mm and 125mm respectively) were also cast with each mix to measure the gamma-shielding performances. Thus, a total of eighty-four cube specimens and 15 slab specimens were used to study the strength and gamma radiation shielding performances respectively.

2.4. Gamma attenuation studies

2.4.1. Gamma radiation source

The linear attenuation coefficient (µ), half-value layer (HVL) and tenth-value layer (TVL) of concrete mixes prepared with crushed granular, hematite and magnetite coarse aggregates were measured for a photon energy of 662 KeV for Cs^{137} and two photon energies of 1173 and 1332 KeV for Co^{60}.

2.4.2 Test set up

The attenuation measurements of gamma rays were performed using sodium iodide NaI (Tl) scintillation detector with a Multi-Channel Analyzer (MCA). The arrangements of experimental set up used in the test are shown in Fig. 1. The utilized radiation sources comprised ^{137}Cs and ^{60}Co radioactive elements with photon energies of 662 KeV for Cs-137 and two energy levels of 1173 and 1332 KeV for Co-60 as standard sources with activities in micro curie (5 mCi) for Gamma-rays. After 28 days of ambient curing, specimens were dried at 100^{0}C prior to the test. Test samples with different thicknesses of 25–125 mm were arranged in front of a collimated beam emerged from gamma ray sources as shown in Fig.2. The measurements were conducted for 450 seconds counting time for each sample.

Fig. 2. Gamma Radiation Shielding setup

2.4.3 Radiation flux measurements (Ouda, 2014)

The attenuation coefficient of gamma rays was determined by measuring the fractional radiation intensity N_x passing through the thickness x as compared to the source intensity No. The linear attenuation coefficient (µ) has been obtained from the solution of the exponential Beer–Lambert’s law,

$N_x = N_o e^{-\mu x}$ cm^{-1}

where, ‘µ’ is the linear attenuation coefficient, ‘N_o’ is the intensity of gamma-rays without any sample and ‘N’ is the intensity of the gamma rays, which passes through a sample of thickness ‘X’

Half-value layer (HVL) and tenth-value layer (TVL) are the thicknesses of an absorber that will reduce the gamma radiation to half and to tenth of its intensity, respectively. Those are obtained by using the following equations,

$X_{1/2} = \ln(2/\mu)$

$X_{1/10} = \ln(10/\mu)$

2.5 Compressive strength studies

Compressive strength test was carried out using compression testing machine. A sample size of 100x100x100mm cubes were cast and ambiently cured for a period of 7 and 28 days in an ambient temperature. Test was carried as per IS 516-1959 in a compression testing machine whose load bearing capacity was 2000kN/m.

2.6 Thermal performance analysis

The cube samples were kept in an oven for 2 hours maintained at 150, 300, 450, 600, 750 and 900^{0}C and are tested for compressive strength. Test was carried as per IS 516-1959 guidelines. For the practical application of concrete walls made up of different materials, the thermal performance of a Containment structure wall is determined. Thermal analysis is carried out in COMSOL software. COMSOL is a general-purpose simulation software for modelling designs, devices and processes in all fields of engineering and scientific research. The heat transfer through the walls is assumed to be one dimensional. The Containment structure wall consists of concrete wall layer of 1.2m thick. The rate of heat flow in hematite and magnetite concrete are compared with conventional concrete.

III. Results and conclusions

3.1 Strength properties

Compressive strength properties of conventional, hematite and magnetite concretes are determined at room temperature for 7 and 28 days curing and are shown in table 2,

Table 2

Compressive strength for 7- and 28-days curing

Curing period	Conventional concrete(mpa)	Magnetite concrete(mpa)	Hematite concrete(mpa)
7 days	51	54	55
28 days	78	80	82

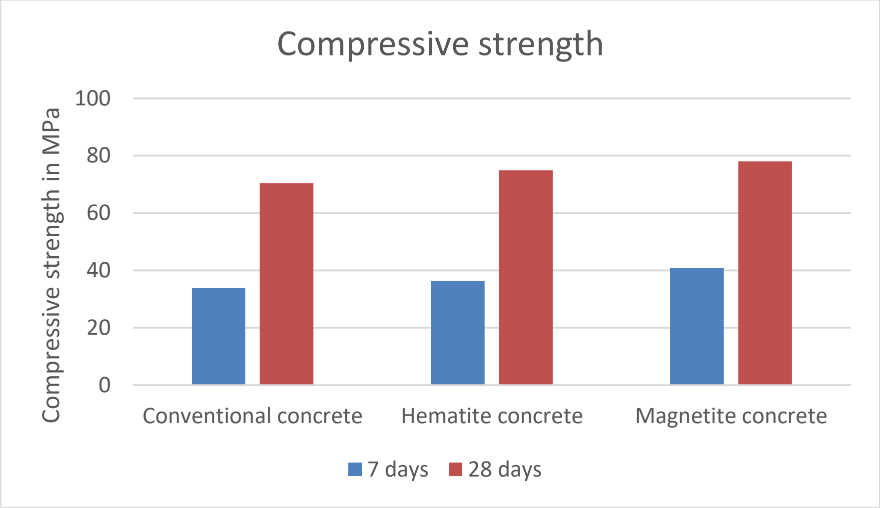

Fig. 3. Compressive strength for 7- and 28-days curing

We can clearly see from figure 3, results of compressive strength that as the density of aggregate material increases, Compressive strength increases. The increase in compressive strength from target strength (M60) is 17.4%, 24.78% and 30% for conventional, hematite and magnetite concretes respectively. Geopolymer Hematite concrete has highest compressive strength among three concretes at room temperature for both 7 and 28 days of curing. From the bar graph above, geopolymer Conventional concrete has lowest Compressive strength compared to Hematite and Magnetite concretes.

3.2. Shielding characteristics of concrete mixes

Significant improvements in the gamma radiation shielding properties have been observed for both the high-density concrete mixes. Table 2, summarizes the gamma shielding properties such as attenuation coefficient, Half value layer, tenth value layer and Mean free path of all the different mixes.

Table 3

Relationship between the attenuation coefficients (l), the half-value layer (HVL) and the tenth-value layer (TVL) of concrete mixes made with the 3 different coarse aggregates.

Type of	ɣ - Source	Photon	Thickness	Attenuation coefficient	Half value layer	Tenth value	Mean free path
concrete		energy (MeV)	(mm)	μ (cm^{-1})	(cm)	layer (cm)	(cm)
Normal concrete	Co^{60}	1.173 &	25	0.59688	1.161	4.96	2.576
		1.332	50	0.82265	0.842	3.01	1.251
			75	1.18201	0.586	1.94	0.846
			100	1.451978	0.477	1.585	0.688
			125	1.751347	0.395	1.314	0.57

Normal concrete	Cs^{137}	0.662	25	0.475702	1.277	4.2438	1.8435
			50	0.957589	0.7236	2.4039	1.044289
			75	1.40713	0.4924	1.6359	0.7106
			100	1.783761	0.388	1.2905	0.5606
			125	2.246561	0.308	1.0246	0.4451
Hematite concrete	Co^{60}	1.173 &	25	0.473126	1.456	4.8655	2.11
		1.332	50	0.971133	0.7507	2.37	1.029
			75	1.360061	0.3909	1.69	0.735
			100	1.688699	0.2969	1.36	0.592
			125	2.362628	0.2608	0.974	0.423
Hematite concrete	Cs^{137}	0.662	25	0.537835	1.456	4.28	1.85
			50	1.214227	0.7507	1.895	0.8235
			75	1.772634	0.3909	1.2986	0.5641
			100	2.334091	0.2969	0.986	0.4284
			125	2.657187	0.2608	0.866	0.37633
Magnetite concrete	Co^{60}	1.173 &	25	0.489794	1.414	4.69	2.0416
		1.332	50	0.790861	0.876	2.91	1.26
			75	1.291051	0.536	1.78	0.77
			100	1.718174	0.4033	1.33	0.58
			125	2.621522	0.264	0.878	0.38
Magnetite concrete	Cs^{137}	0.662	25	0.475702	1.288	4.8391	2.102155
			50	0.959608	0.722	2.3988	1.04
			75	1.616746	0.4286	1.4238	0.61
			100	2.367112	0.2927	0.9724	0.422
			125	2.742458	0.2526	0.8393	0.364

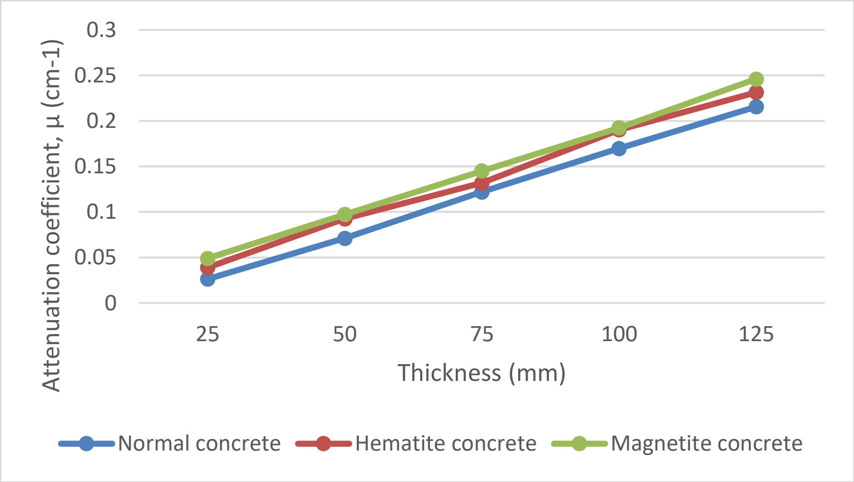

Fig. 4. Variation of linear attenuation coefficients with concrete thickness made for Co-60 with photon energies of 1.173 and 1.333 MeV.

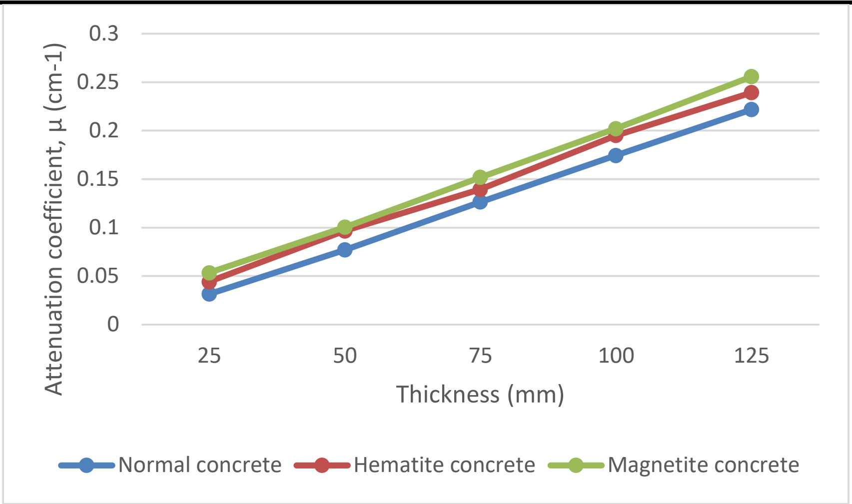

Fig. 5. Variation of linear attenuation coefficient with concrete thickness for Cs^{137} with photon energies of 0.662

Fig. 4. Shows variation of linear attenuation coefficients for concrete thickness made for Co-60 with photon energies of 1.173 and 1.333 MeV. For 25mm thickness, the increase in attenuation coefficient was found to be 48.47% & 87.02% for hematite and magnetite concretes with respect to conventional concrete respectively. For 50mm thickness, attenuation coefficient of hematite and magnetite concrete increases to 29.95% & 36.70% respectively in comparison with conventional concrete. There is increase in attenuation coefficient of 75mm thickness sample by 8.11% & 18.85% for hematite and magnetite concrete with reference concrete mix. For 100mm thickness, the increase in attenuation coefficient was found to be 12.13% & 13.43% for hematite and magnetite concretes with respect to conventional concrete respectively. For 125mm thickness, attenuation coefficient of hematite and magnetite concrete increased by 7.2% & 14.04% respectively in comparison with conventional concrete.

Fig. 5. Shows variation of linear attenuation coefficients for concrete thickness made for Cs-137 with photon energies of 0.662 MeV. For 25mm thickness, the increase in attenuation coefficient was found to be 40.31% & 69.84% for hematite and magnetite concretes with respect to conventional concrete respectively. For 50mm thickness, attenuation coefficient of hematite and magnetite concrete increases to 25.42% & 30.35% respectively in comparison with conventional concrete. There is increase in attenuation coefficient of 75mm thickness sample by 10.27% & 20% for hematite and magnetite concrete with reference concrete mix. For 100mm thickness, the increase in attenuation coefficient was found to be 11.80% & 15.81% for hematite and magnetite concretes with respect to conventional concrete respectively. For 125mm thickness, attenuation coefficient of hematite and magnetite concrete increased by 7.89% & 15.33% respectively in comparison with conventional concrete.

It was found that, the linear attenuation coefficients for both Cs-137 and Co-60 increase with concrete thickness. Also, linear attenuation coefficients for the concrete mixes decreases with the increase in gamma ray energy as found in fig.5. and fig. 6. It is clearly seen that, the linear attenuation coefficients depend on the photon energy and the density of the shielding material; accordingly, the concrete samples containing magnetite aggregates are remarkably effective for shielding of gamma rays. There is increase in attenuation coefficient of Cs^{137} over Co^{60} for 25mm thick slab are 20%, 13.62% & 9.1% for conventional, hematite and magnetite concretes respectively. Thus, it can be concluded that the linear attenuation coefficient depends on the photon energy as well.

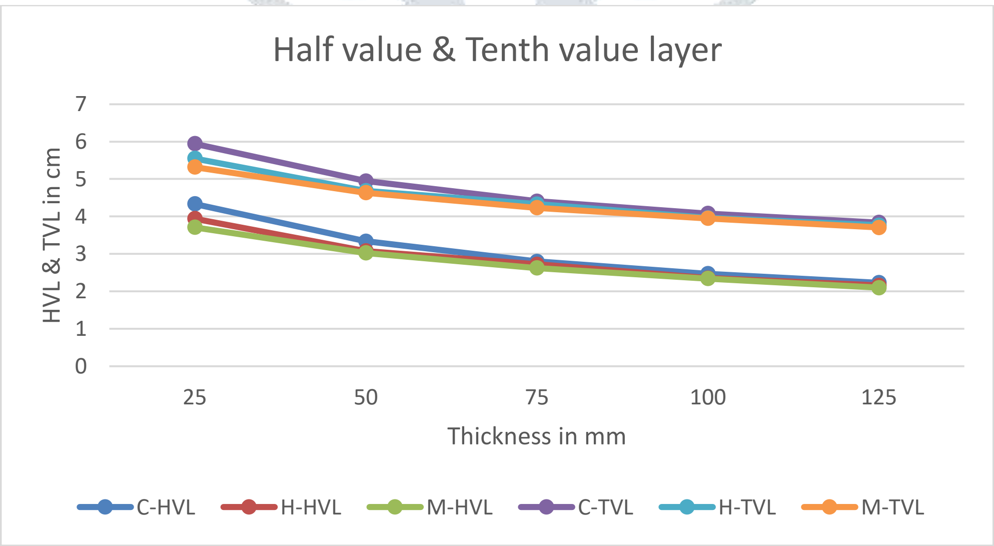

Fig. 6. Half-value layer and tenth-value layer for concrete thickness using ^{60}Co source

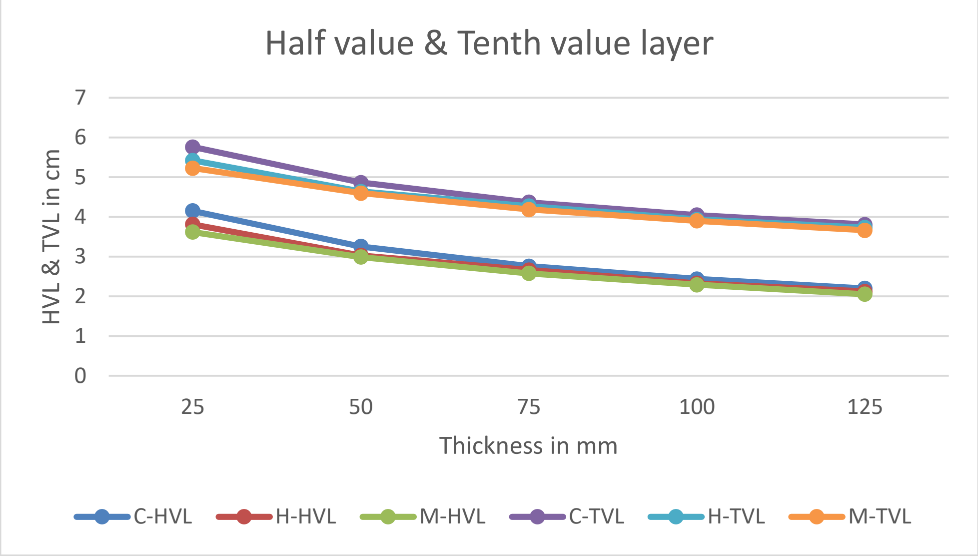

Fig. 7. Half-value layer and tenth-value layer for concrete thickness using ^{137}Cs source

Figs. 6 and 7 shows the HVL & TVL values of concrete mixes for gamma energy emitted by ^{60}Co and ^{137}Cs sources for different concrete thickness. It is seen that, the HVL and TVL values of mixes decrease with the concrete thickness.

The decrease in HVL for Co-60 source for 25, 50, 75, 100 and 125mm thickness are 9.2%, 7.6%, 2.86%, 4.47% & 3.15% for hematite concrete and 14.54%, 9.3%, 6.09%, 4.87% & 5.85% for magnetite concrete respectively with respect to convention concrete. For Cs-137, the HVL reduces by 8.19%, 7.07%, 3.62%, 4.52%, 3.19% for hematite concrete and 12.77%, 8%, 6.88%, 5.76% & 6.39% for magnetite concrete of 25, 50, 75, 100 and 125mm thickness respectively.

The decrease in TVL for Co-60 source for 25, 50, 75, 100 and 125mm thickness are 6.73%, 5.26%, 1.81%, 2.7% & 1.82% for hematite concrete and 10.60%, 6.27%, 3.86%, 2.9% & 3.39% for magnetite concrete respectively with respect to convention concrete. For Cs-137, the TVL reduces by 5.9%, 4.73%, 2.28%, 2.72%, 1.84% for hematite concrete and 9.2%, 5.3%, 4.34%, 3.46% & 3.6% for magnetite concrete of 25, 50, 75, 100 and 125mm thickness respectively.

The lower the values of HVL and TVL, the better are the radiation shielding materials in terms of the thickness requirements. At a photon energy of 0.662 MeV for Cs137 source, the values of HVL and TVL for magnetite aggregate are lower as compared to the other mixes at the same energy. The results indicated also that, the values of HVL and TVL are inversely proportional to the concrete density.

3.3 Thermal performance analysis

It is observed from the results depicted in Fig. 8.. that, with reference to the control concrete mix, both hematite and magnetite concrete mixes achieved reasonable higher compressive strengths, as expected. The reference control concrete mix attained a higher strength of 70.44 MPa and among the high-density mixes, hematite and magnetite concretes attained maximum compressive strengths of 74.87 and 78.045 MPa respectively at 28-days of testing. The density of conventional, hematite and magnetite concrete was found to be 2490, 3402 and 3556 kg/m^3 respectively. It can be observed that all the concrete mixes have attained compressive strengths greater than 60 MPa at 28 days of curing and hence satisfy the range of strengths required for a structural grade concrete

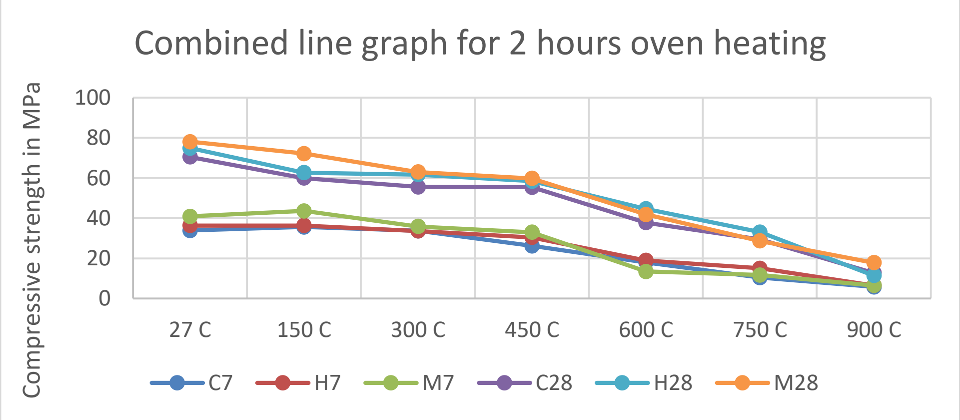

Fig. 8. Combined graph for 7 and 28 days curing for 2 hours oven heating

Concrete mix with magnetite aggregate showed higher physico-mechanical properties than that containing hematite and normal aggregates. The increase in compressive strength values at room temperature was found to be 17.4%, 24.78% and 30.07% for conventional, hematite and Magnetite concrete respectively. It is observed from the results of compressive strength that as the density of aggregate material increases, Compressive strength increases. Density being 3556kg/ m^3 for magnetite aggregate has highest compressive strength. Initially compressive strength of all concretes increases from room temperature to 150°C. Further, the strength values of all three concrete decreases with the increase in temperature. From 450°C to 600°C there is sudden fall of strength by 31.85%, 23.65% and 30.03% for conventional, hematite & magnetite concrete respectively

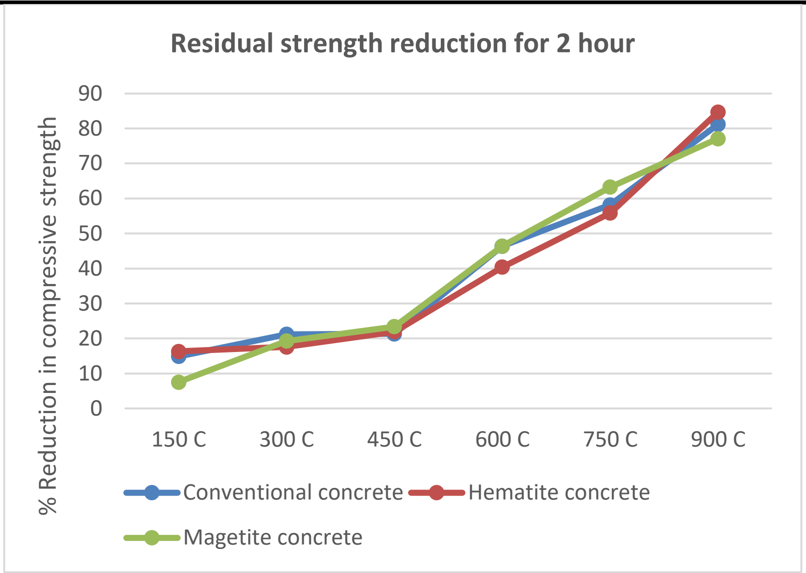

Fig. 10. Strength reduction for 1 and 2 hours oven drying

Residual strength reduction from room temperature to 450ºC for 2 hours lies within 20% of strength at room temperature. Beyond 450ºC, there is great reduction in compressive strength up to 900ºC. Between 750ºC- 900ºC, the strength lies in the range from 50-80%. The graphical representation of strength reduction for 1 and 2 hours oven drying is represented in Fig.10.

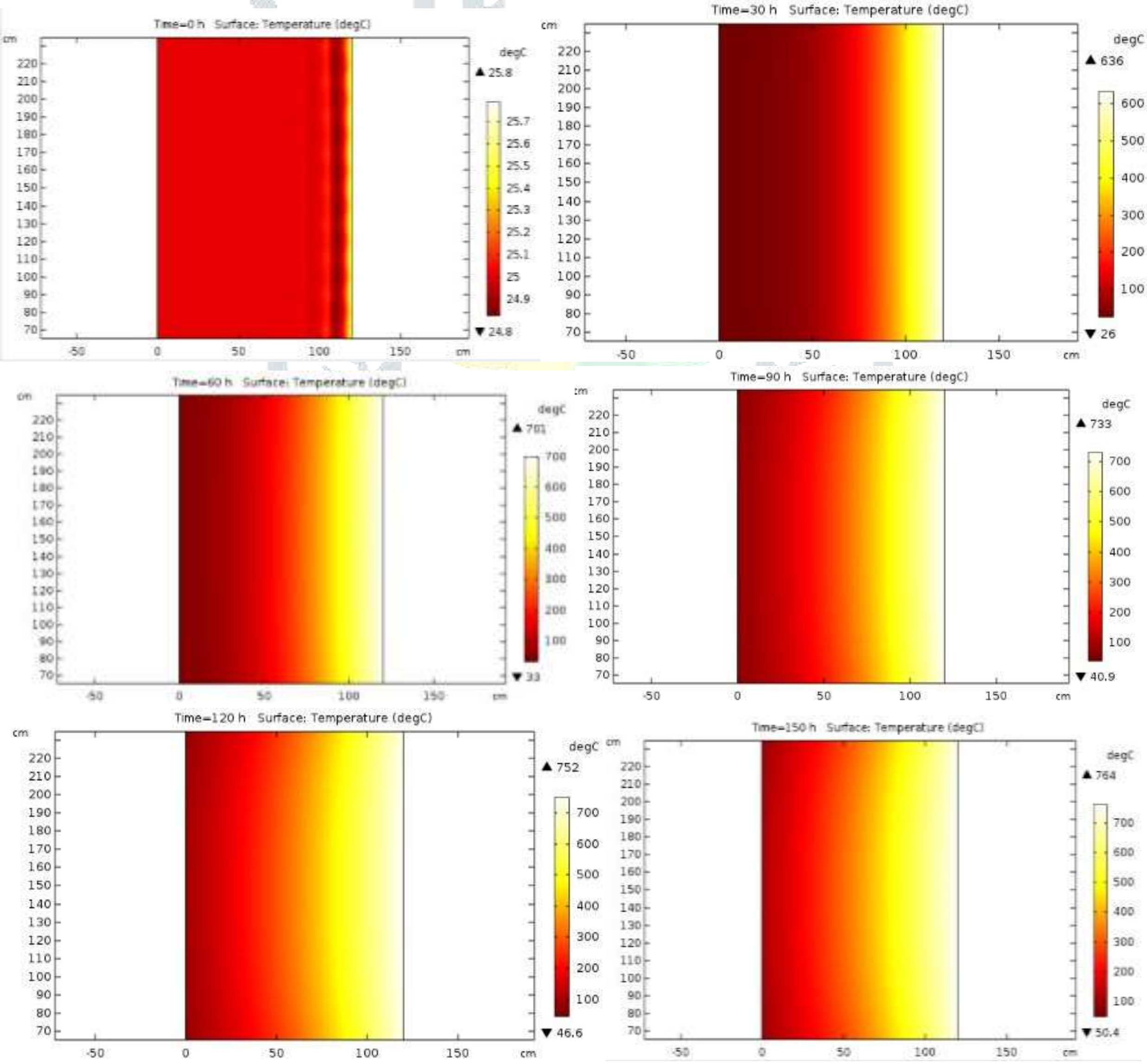

Figure 11. Variation of surface temperature from 0 hour to 150 hours for magnetite concrete wall (900^0C)

From figure 11, it is observed that change in temperature after 150 hours is negligible. Hence wall made up of Magnetite

concrete has time lag of 150 hours for temperature of 900^0C. Thus, steady state of heat flow is achieved at 150 hours. At the inner surface of the wall, the temperature has decreased to 764°C over the duration of 150 hours.

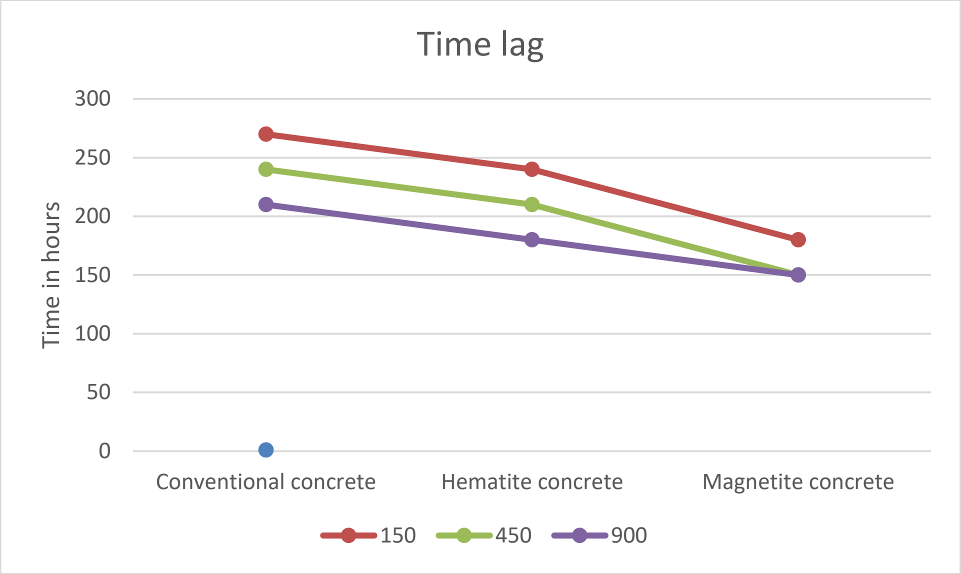

Figure 9. Time lag graph for 3 different temperatures

The time lag for 150^{0}C, 450^{0}C and 900^{0}C are 270, 240 and 180 hours respectively for Conventional concrete; 240, 210 and 150 hours respectively for Hematite concrete and 210, 180 and 150 hours respectively for Magnetite concrete as shown in figure 9. Heat flow always takes place from higher temperature end to the lower level. From above table, it is observed that as the temperature increases, time lag decreases. Also, as the density of concrete wall increases, Time lag decreases. Denser the wall, more is the heat flow.

IV. Conclusions

Concrete mix with hematite aggregate showed higher physico-mechanical properties than that containing hematite and normal aggregates. The geopolymer hematite mix, geopolymer magnetite mix and geopolymer normal concrete mix gained 78%, 77% and 73% of target strength in first 7 days and gained the required strength after 28 days ambient curing.

From the experimental results, it was observed that initially compressive strength of all geopolymer concretes remains constant from room temperature to 150^{o}C. Further, the strength values of all three geopolymer concrete decreases with the increase in temperature. From 450^{o}C to 600^{o}C there is sudden fall of strength by 54%, 56% and 37% for geopolymer normal concrete, geopolymer magnetite concrete and geopolymer hematite concrete respectively.

It was observed from the experimental results that, among all slabs, 125mm thick concrete slab showed better shielding performance for hematite and magnetite concrete than conventional concrete. The increase in attenuation coefficient were 7.2% and 14.02% for Co-60 and 7.8% and 15.33% for Cs-137 source respectively.

HVL decreases with the increase in concrete thickness. 25mm thick concrete slab has maximum HVL of 4.33, 3.93 & 3.70 cm for Co-60 and 4.15, 3.81 & 3.62cm for Cs-137 source respectively. The HVL of 125mm is least with 2.22, 2.15 & 2.09cm for Co-60 and 2.19, 2.12 & 2.05cm for Cs-137 source respectively. Maximum reduction in HVL for hematite and magnetite concrete are found in 25mm thick slab as 9.2% and 14.54% for Co-60 and 8.19% and 12.77% for Cs-137 respectively.

It was found that, the linear attenuation coefficients for both Cs^{137} and Co^{60} increase with shield concrete thickness. On an average, for every increase in thickness of concrete by 25mm, attenuation coefficient increases by 2.5%. There is increase in attenuation coefficient of Cs^{137} over Co^{60} for 25mm thick slab are 20%, 13.62% & 9.1% for conventional, hematite and magnetite concretes respectively. Thus, it can be concluded that the linear attenuation coefficient depends on the photon energy as well.

TVL decreases with the increase in concrete thickness. 25mm thick concrete slab has maximum TVL of 5.94, 5.54 & 5.31cm for Co-60 and 5.76, 5.42 & 5.23cm for Cs-137 source respectively. The TVL of 125mm is least with 2.22, 2.15 & 2.09cm for Co-60 and 2.19, 2.12 & 2.05cm for Cs-137 source respectively. Maximum reduction in TVL for hematite and magnetite concrete are also found in 25mm thick slab as 6.73% and 10.60% for Co-60 and 5.90% and 9.20% for Cs-137 respectively.

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