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Study of Mechanical, Physical and Gamma Radiation Attenuation Properties for Concrete Mixes Prepared from Naturally Occurring Ores and Synthetic Aggregates

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Abstract

Ordinary concrete is widely used in many applications that concerned with gamma and neutron radiation shielding. They are widely used in nuclear power plant, nuclear shelters, radiotherapy-mega voltages rooms and for transporting as well as storage of radioactive wastes. In this study four different concrete mixes were prepared using dolomite, barite, goethite and steel slag as coarse aggregates. Fine aggregates for all samples selected to be 50% local sand and 50% limonite with theaddition of 10% silica fume (SF) and 10% fly ash (FA) by replacement of the total cement weight. The cement type used in this study was Portland blast furnace slag cement (PBFSC) with quality CEM/B-S 42.5 N according to EN 197-1standard. Many investigations were performed on physical, mechanical and radiation attenuation properties of these mixes to stand on the best concrete mixes that can be used for radiation shielding applications. A verification using WinXcom program (Version 3.1) was made for radiation attenuation test results. The results revealed that all concrete mixes; goethite-limonite concrete (G.L), Barite-limonite concrete (B.L), steal slag-limonite concrete (S.L) and dolomite concrete (D.C) had good physical and mechanical properties that classified them as high performance concrete. Barite-limonite concrete (B.L) and steal slag-limonite concrete (S.L) were the best for γ-ray attenuation at relatively low and high energy ranges (<1MeV and >1MeV). The results obtained from WinXcom program (Version 3.1) showed good agreement with the experimental results of the γ-ray attenuation tests.

Keywords: High performance concrete, Radiation shielding, Mass attenuation coefficient (σ), Half value layer (HVL) and WinXcom program.

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1. Introduction

Radiation is a general word that was used frequently in the past decades to describe electromagnetic waves but nowadays radiations refers to the all kinds of electromagnetic waves in addition to the atomic and subatomic particles that have been discovered [1]. One of many types of classifications that organizes and classifies the different types of radiation indicates that radiation can be divided into two major categories, the first is the non ionizing radiation and the second is the ionizing radiation.

In shielding issues, we concerned with the ionizing radiation especially the indirect ionizing radiation (γ-rays and neutrons), because the direct ionizing radiation (α, β, P,….)is considered as small external threat as this type includes charged particles which known with their small ranges in the medii through which they pass so, they lose their full energies in quite small ranges [2]. The serious problem always is in the indirect ionizing radiation since they have high penetrating ability and longer ranges. Hence any shield that can attenuate them to the desired level will automatically attenuate the others to negligible value. In attenuating gamma radiation only high (z) elements are preferred thus dense materials are required like steel and lead for example. However, in attenuating neutrons, especially fast neutrons, light elements like hydrogen and oxygen are needed and as a conclusion from the former, the shielding barrier should have high density and in the same time high hydrogen or light elements content so, considering this conflict, the optimum shielding barrier can be used in this case is concrete especially heavy weight concrete which achieves a good compromise between the high density and high hydrogen content that gives heavy weight concrete the priority in attenuating gamma and neutrons in the same time.

Heavy weight concrete is widely used as a shielding barrier in nuclear plants, nuclear shelters, radiotherapy-megavoltage rooms and for transporting as well as storing radioactive wastes [3]. For these purposes heavy weight concrete must have good physical and mechanical characteristics beside having good attenuating properties for both gamma rays and neutrons. To achieve the desired physical, mechanical and radiation attenuation properties, the selection of local suitable aggregates and additives from which the shield is to be made becomes of great importance [4]. Some of the natural minerals used as coarse and fine aggregates in heavy weight concrete are hematite, magnetite, ilmenite, barite, limonite, goethite, serpentine and some of the artificial synthetic aggregates include materials like iron shots, steel punching, iron fibers, heavy slag and boron frits. It is essential that aggregates used in heavy weight concrete must be inert with respect to alkalis and have good mechanical-physical properties to obtain adequate mix [3].

The main aim of this research is to investigate the suitability of some different mixtures with different types of coarse aggregates for yielding high performance heavy weight concrete that could enhance the attenuation properties and thus the shielding efficiency against X-rays and γ-rays.

2. Methodology of research

2.1 Materials

The aggregates selected were chosen regarding the radiation shielding point of view and in the same time satisfy the requirements of construction applications. Consequently, four different types of course aggregates were used , dolomite [$CaMg(CO_3)_2$] from Helwan, Cairo, Egypt, goethite [α-FeO(OH)] and barite [$BaSO_4$] both from El-Bahariya Oasis, Western Desert, Egypt, steel slag (by product from iron industry) obtained from Iron and Steel Factory, Eltebin, Helwan, Egypt. The fine aggregates used are local sand, Helwan, Egypt and limonite [$Fe_2O_3 \cdot nH_2O$] from El-Bahariya Oasis, Western Desert, Egypt. The materials used in this study are Portland blast furnace slag cement CEM/B-S 42.5 N which is compatible with ASTM C-150 [5], from El-Aamryah Cement Company, Egypt. The additives used are fly ash (FA) class F, silica fume (SF) and super plasticizer Sikament-NN (type G) all from Sika Company, El-Obour, Egypt. Coarse and fine aggregates were sieved in order to get coarse aggregates in the range 5-20 mm and fine aggregates with particle size <5 mm. Some important physic-mechanical properties of aggregates are presented in Table 1 and evaluated according to the limits stated by [6, 7].Chemical analysis was performed for aggregates, cement, and additives using XRF Spectrometer as shown in Table 2.

Table 1: Some important physical and mechanical properties of coarse and fine aggregates.

Property	Barite	Goethite	Steel slag	Dolomite	Limonite	Sand
Specific gravity	**4.4**	**4.04**	**4.46**	**2.68**	**2.22**	**2.65**
Water absorption, %	**1.5**	**13.5**	**0.52**	**0.7**	**30.8**	**0.4**
Crushing value, %	**63.3**	**20**	**16.83**	**-**	**-**	**-**

2.2 Mix proportions

Four different concrete mixes were prepared using goethite, barite, dolomite and steel slag as coarse aggregates. Fine aggregates used in all mixes were (50% local sand – 50% limonite) in addition to 10% silica fume (SF) and 10% fly ash (FA) as a partial replacement from the total cement content. All concrete mixes were prepared according to the American Concrete Institute method (ACI) of absolute volumes [8]. The ACI method is generally considered to be more convenient and suitable for heavy weight concrete. The mix proportions per 1m^3 for all concrete mixes are shown in Table 3.All aggregates used in this study were used in saturated surface dry form to eliminate the effect of water absorption during mixing in order to evaluate the real effect of aggregates on concrete mixes properties [3]. The water to cement ratio for all mixes were set to be 0.43.

Table 2: Chemical analysis for cement, additives and aggregates

Oxides	PBFSC	SF	FA	Coarse aggregates				Fine aggregates	
				Goethite	Barite	Steel Slag	Dolomite	Limonite	Sand
SiO_2	23.33	96.81	61.13	11.2	1.16	8.13	2.24	16.3	94.84
Al_2O_3	5.91	0.25	27.68	3.39	0.64	2.01	0.95	2.97	2.12
Fe_2O_3	3.29	0.45	4.15	67.0	20.84	37.0	0.61	68.1	0.82
CaO	57.07	0.16	1.32	6.49	1.59	44.4	37.9	4.16	0.52
MgO	3.10	0.26	0.44	0.992	1.63	1.15	15.03	0.643	0.1
SO_3^{--}	2.9	0.14	0.28	1.9	4.42	0.864	0.39	2.9	0.11
Cl^-	0.03	0.03	0.07	0.923	0.41	0.15	0.13	0.62	0.06
Na_2O	0.24	0.14	0.15	1.46	-	0.973	0.25	0.985	0.27
K_2O	0.25	0.28	0.85	1.8	0.34	0.358	0.07	0.74	0.69
TiO_2	0.08	-	2.07	1.49	-	1.08	0.13	1.28	0.12
BaO	-	-	0.04	-	66.77	-	-	-	-
P_2O_5	-	0.03	0.61	0.91	0.28	1.74	0.03	0.83	0.05
V_2O_5	-	-	-	-	-	0.104	-	-	-
Cr_2O_3	-	-	-	0.416	0.14	0.167	-	-	-
MnO	-	0.05	-	0.292	1.1	1.82	-	-	-
CeO_2	-	-	-	0.278	-	-	-	-	-
Sm_2O_5	-	-	-	0.314	-	-	-	0.168	0.06
L.O.I	2.97	0.95	0.85	0.3	0.2	-	42.25	0.15	0.1
Total	99.17	99.55	99.64	99.15	99.52	99.94	99.94	99.84	99.86

Table 3: Mix proportions of concrete mixes

Mixes	Concrete ingredients, kg/m^3									
	PBFSC	Fine aggregates		Coarse aggregates				Additives		S.P
		Sand	Limonite	Goethite	Barite	Steel slag	Dolomite	SF	FA	
G.L	400	270.77	226.83	1651.3	-	-	-	50	50	12.5
B.L	400	270.77	226.83	-	1798.3	-	-	50	50	12.5
S.L	400	270.77	226.83	-	-	1822.86	-	50	50	12.5
D.C	500	554.8	-	-	-	-	1126.3	-	-	-

2.3 Mixing, curing and testing specimens

The mixing method used for the concrete mixes in this study is similar to that used for conventional concrete. The materials were mixed using a mixer with a capacity of 56 dm^3. The mixing sequence for each mix was as follows; dry mixing of coarse aggregates and fine aggregates, followed by cement mixed with mineral cementing materials for 2 min, adding of 80% of the mixing water with the dry mixture for 1.5 min, the rest 20% of the mixing water was added to the rotating mixer in a gradual matter. All mixes were mixed for 5 min as a total time. In order to prevent fresh concrete from segregation especially that we used heavy aggregates. After the mixing was finished, slump test was performed on the fresh concrete to evaluate the workability according to ASTM C143 [9]. All concrete specimens were cast in three layers in 100x100x100 mm cubic molds and cylindrical molds [200 mm in length – 100 mm in diameter].After casting the specimens in their molds, the specimens were consolidated using a vibrating table Fig 1, then the specimens were covered to avoid water evaporation and kept for 24 hours. After removing specimens from their molds, they were cured by immersing them in curing tanks until the date of testing Fig 2.

Bulk density

For concrete, expressing the density in kilograms per cubic meter is widely common when aggregates are to be actually batched by volume and here the density to be calculated is called bulk density (ρ). The bulk density for hardened concrete mixes was performed according to ECCS 203-2001 [10].

Compressive strength

The compressive strength of concrete is usually determined by applying a uniformly distributed increasing compression load on a cubic specimen using suitable testing equipment until failure. The testing equipment used for this test is 2000 KN universal machine Fig 3. The test was performed using a set of three cubic specimens [100×100×100 mm^3] for each concrete mix at curing age 28 days and the compressive strength at 90 days was estimated using the following general equation:

$$(f'_c)_t = \frac{t}{a+bt} \times (f'_c)_{28} \qquad \text{..............................} \qquad (1)$$

According to ACI 209/71 standard, this equation was obtained as a result of a study of concrete strength versus time with different types of concrete [11]. The constants a (days) and b both are related to the characteristics of the used mix design and curing conditions and they could be determined by solving two simultaneous equations at two different ages using previous experimental work which was suitable and agreeable with the concrete mixes used in this study [3, 12].

Ultrasonic pulse velocity test

This test considered as a good and easy technique for assessment of concrete quality by measuring the velocity of the ultrasonic pulse that passing through a known thickness of the concrete specimen according to ASTM C597 [13]. The ultrasonic pulse velocity test is used mainly to measure the concrete quality however; it can be used to confirm the compressive strength test results. The device used for this test is shown in Fig 6.

Calculation of elastic modulus

The simple definition of modulus of elasticity is that value which measures the concrete’s resistance to being deformed elastically when a load is applied to it, also it can be defined as the slope of the stress strain curve for concrete in the elastic deformation region [14]. By increasing the modulus of elasticity, the concrete becomes stiffer, harder and resistant to deformation and excessive loads. In this study elastic modulus for concrete mixes were calculated using the following practical equation:

$$E = k_1 \times k_2 \times 3.35 \times 10^4 \times \left(\frac{\gamma}{2.4}\right)^2 \times \left(\frac{\sigma}{60}\right)^{1/3} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (2)$$

Where: σ is the compressive strength after 28 days, γ is the concrete density, k_1 the correction factor for coarse aggregates ranges from 0.95 to 1.2, k_2 the correction factor for mineral additions ranges from 0.95 to 1[15].

Gamma rays attenuation measurements

In this study, cylindrical samples of dimensions 200 mm in length and 100 mm in diameter were prepared for all concrete mixes and then cut to different thicknesses as shown in Fig 5. The gamma sources used in these tests are Ba-133, Cs-137 and Co-60 and the detector used in detection and measurement is NaI(Tl), 3’’x3’’scintillation detector with multichannel analyzer using software (UCS-30) version 1.1.06 USB, Spectrum Technique 2010. In order to achieve the good geometry condition, the gamma sources were fixed inside 30 mm lead holder (source collimator) with an aperture 3 mm in diameter, while the scintillation detector had been surrounded by lead slabs at the sides and covered with 5 mm lead sheet (detector collimator) in order to prevent scattered gamma rays from reaching the detector. The source beam, the samples and the detector were placed in the same horizontal plane and the distance between the source and the face of the detector was 310 mm fixed for all measurements Fig 6. First, three counts were taken without any samples and the average was taken to be the initial intensity (I_o). Then three counts were taken with each sample thickness and the average of these readings was taken to represent the transmitted intensity at certain thickness (I_x). The live time was adjusted to be 10 min for each reading [16].

After the readings were taken, a relation between ln $\left(\frac{I_x}{I_o}\right)$ and density thickness (ρx) for each concrete mix was plotted and the slope value (without the negative sign) for each relation was taken to be the mass attenuation coefficient (σ) for the mix. Then the linear attenuation coefficient (μ) was calculated from the following equation:

$$\mu = \sigma \times \rho \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (3)$$

The Half value layer (HVL) and Tenth value layer (TVL) for each concrete mix were obtained from the following equations [3].

$$HVL = \frac{\ln 2}{\mu} \quad , \; TVL = \frac{\ln 10}{\mu} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (4)$$

After recording all these results that obtained experimentally, the mass attenuation coefficients for the different concrete mixes were calculated theoretically as well using WinXcom program (version 3.1), then calculated coefficients were compared with those obtained experimentally.

3. Results and Discussion

3.1 Slump test results

The results showed that the slump values of the investigated concrete mixes ranged from 6 cm to 10 cm as shown in Table 4, which means that all mixes were accepted from the workability point of view. The highest slump values obtained with D.C and B.L mixes respectively and the lowest value obtained with G.L mix. These results can be attributed mainly to the differences in the water absorption ratios of the different aggregates in addition to the high specific gravity values for the used coarse aggregates except for dolomite used in D.C mix (see Table 2).

Table 4: Slump values for the concrete mixes.

Concrete mix	G.L	B.L	S.L	D.C
Slump value, cm	6.0	8.0	7.5	10.0

3.2 Bulk density

The bulk densities for different concrete mixes are shown in Table 5. The results obtained were logic as the bulk density of the concrete prepared is directly proportional to the specific gravity of the aggregates used in the mix design (see Table 2); therefore, S.L mix were found to have the highest value of bulk density. S.L, B.L and G.L all can be classified as heavy density concrete because all of them were found to have bulk density values more than 2600 kg/m^3 [17]. On the other hand D.C couldn't meet the requirements of the dense concrete as its density was 2570 kg/m^3, so it can be classified as normal weight concrete.

Table 5: Bulk densities of the concrete mixes

Concrete mix	G.L	B.L	S.L	D.C
Bulk density, kg/m^3	**2906**	**2963**	**2994**	**2570**

3.3 Compressive strength

After curing for 28 days, the concrete mixes gained the most of their strength because of the formation of the hydration products and the domination of hydrated calcium silicate (C-S-H gel) among these hydration products. The compressive strength of all concrete mixes in this study was higher than that of traditional concrete. Using Portland blast furnace slag cement with high content was one of the reasons that lead to the concrete mixes high strength, also the addition of fly ash and silica fume (see Table 3) participated in the development of the strength of the mixes due to their good filling effect and the pozzolanic activity. The compressive strength of the different concrete mixes at 28 days and 90 days (estimated) is plotted in Fig 7. The results obtained revealed that D.C and S.L mixes were significantly higher than the other two mixes G.L and B.L mixes and the differences could be attributed to the physic-mechanical properties of the coarse and fine aggregates used (see Table 2). The use of limonite as aportion of the fine aggregates was due to its good shielding properties but it had a bad effect on the strength of the mixes because its high water absorption value (30.8%). The relative small crushing value and high specific gravity (4.04) had a good effect on gaining strength in G.L mix but on the other hand its high water absorption value (13.5%) minimize and curtail this good effect. The high crushing value of barite (63.3%) had a significant bad effect on the B.L strength even if its low water absorption value (1.5%) and high specific gravity (4.4). the high specific gravity of steel slag (4.46) and its good physic-mechanical properties in addition to the porous structure all that enhanced the strength of the S.L mixes in spite of using limonite as a portion of the fine aggregates in the mix. Using sand only as fine aggregate beside dolomite as coarse aggregate in D.C mix had a good effect on the strength due to their low water absorption values (0.4%, 0.7%) respectively, and the convergence in the specific gravity values of both of them which had a great effect on strengthening the physical bonds between them and also with the binder.

3.4 Ultrasonic pulse velocity test

As known, this test performed to assess the quality of concrete represented in uniformity, homogeneity, presence or absence of cracks and durability. The results obtained from this test also confirm those obtained from the compressive strength test. The results plotted in Fig 8 showed that all concrete mixes in this study classified as excellent in quality because all of them have ultrasonic pulse velocity higher than 4500 m/sec [18]. The results also showed high agreement with the compressive strength test results.

3.5 Modulus of elasticity

As shown in Fig 9, all mixes in this study had modulus of elasticity greater than that for normal concrete that means greater than 30 Gpa [19] so all concrete mixes are stiffer, harder and resistant to deformation and excessive loads.

3.6 Gamma rays attenuation measurements

The aim of this part is to investigate the attenuation properties for the concrete mixes used in this study and determine the best mix among them. Fig 10 indicates the relation between ln

(I_x/I_o) versus barrier density thickness (ρx) for the four concrete mixes. Moreover, graphs of (σ) against $E\gamma$ were plotted together with the calculated values of (σ) that obtained using WinXcom program (version 3.1) for the four concrete mixes as shown in Fig 11. The obtained values of mass attenuation coefficient (σ), linear attenuation coefficient (μ), half value layer (HVL) and tenth value layer (TVL) for all mixes are shown in Table 6. From the results obtained we found that B.L mix has the greatest linear attenuation coefficient against gamma rays of energies 356 and 662 keV and thus the corresponding minimum required HVL for these energies. The reason could be due to the high atomic number of barium (56) which is the effective element in barite that was used as coarse aggregate in B.L mix. The photo electric effect has a significant contribution in the attenuation process with these energies and as well known the microscopic attenuation cross section due to the photo electric effect is directly proportional to (Z^n) where n varies from 4-5, so with the high (Z) value of barium (56) which is greater than the double of that for iron (Z=26), that explains the priority of B.L mix on S.L mix with the gamma energy lines; 356 and 662 keV. On the other hand at the gamma energies 1173 and 1332 keV, S.L mix regain its priority on B.L. This is due to its higher bulk density and also because the contribution of the photo electric effect in this energy range is very small and the dominant mechanism is the Compton scattering which is linearly proportional to (Z).

Table 6: Values of σ, μ, HVL and TVL for the four concrete mixes

Property	*μ, cm^{-1}*				*σ, cm^2/g*				*HVL, cm*				*TVL, cm*			
γ ray energy, keV	*356*	*662*	*1173*	*1332*	*356*	*662*	*1173*	*1332*	*356*	*662*	*1173*	*1332*	*356*	*662*	*1173*	*1332*
G.L mix	***0.253***	***0.218***	***0.166***	***0.160***	***0.087***	***0.075***	***0.057***	***0.055***	***2.74***	***3.18***	***4.17***	***4.33***	***9.10***	***10.56***	***13.87***	***14.39***
B.L mix	***0.329***	***0.258***	***0.172***	***0.166***	***0.111***	***0.087***	***0.058***	***0.056***	***2.11***	***2.68***	***4.02***	***4.17***	***6.99***	***8.92***	***13.38***	***13.87***
S.L mix	***0.272***	***0.225***	***0.180***	***0.177***	***0.091***	***0.075***	***0.060***	***0.059***	***2.55***	***3.08***	***3.85***	***3.92***	***8.46***	***10.23***	***12.79***	***13.01***
D.C mix	***0.247***	***0.216***	***0.154***	***0.146***	***0.096***	***0.084***	***0.060***	***0.057***	***2.81***	***3.21***	***4.50***	***4.75***	***9.32***	***10.66***	***14.95***	***15.77***

4. Conclusion

Low water absorption values of dolomite and sand and the convergence in the specific gravity values for both of them as well as the high specific gravity, acceptable crushing factor and low water absorption value of steel slag, drive D.C and S.L mixes to have the best physical and mechanical properties among all the studied concrete mixes. On the other hand the low crushing value of barite and the high water absorption value of goethite had drawbacks on the physical and mechanical properties for both B.L and G.L mixes inspite of the high specific gravity values of barite and goethite, also the use of limonite as portion of the used fine aggregates in G.L, B.L and S.L mixes had a bad effect on the physic-mechanical properties of these concrete mixes because of its high water absorption value but the use of goethite and limonite enhance the attenuating properties of the mixes they used in. The high specific gravity values of steel slag, barite and goethite and the presence of effective elements that have high Z number like both iron and barium give the priority of S.L, B.L and G.L mixes on D.C mix in attenuating gamma rays. B.L mix was the best concrete mix in attenuating gamma rays at low and intermediate energies however S.L mix was the best one in attenuating gamma rays at higher energies.

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Figure 1 : The cuing tanks

Figure 2: The vibrating table

Figure 3: The universal machine

Figure 4: Ultrasonic instrument PROCEQ, Pundit Lab Company, Model: PL02-0020282BO, Switzerland

Figure 5: The cylindrical samples used for gamma rays attenuation measurements.

Figure 6: The experimental arrangement of the test.

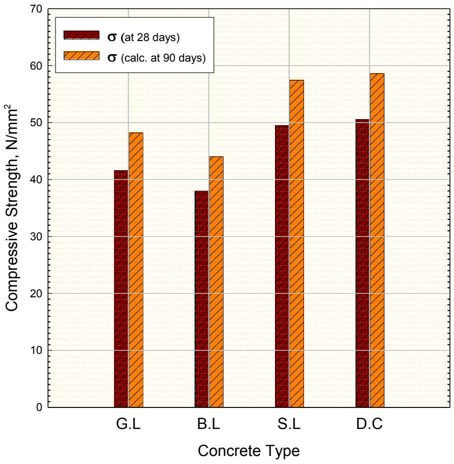


Figure 8: Compressive strength results

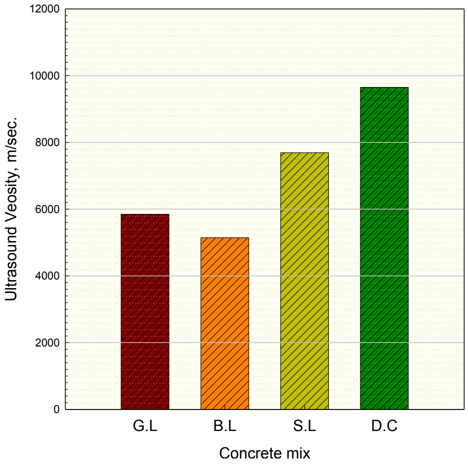


Figure 7: Ultrasonic pulse velocity results.

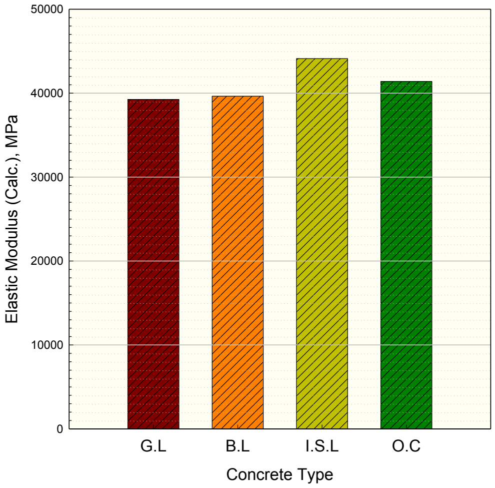
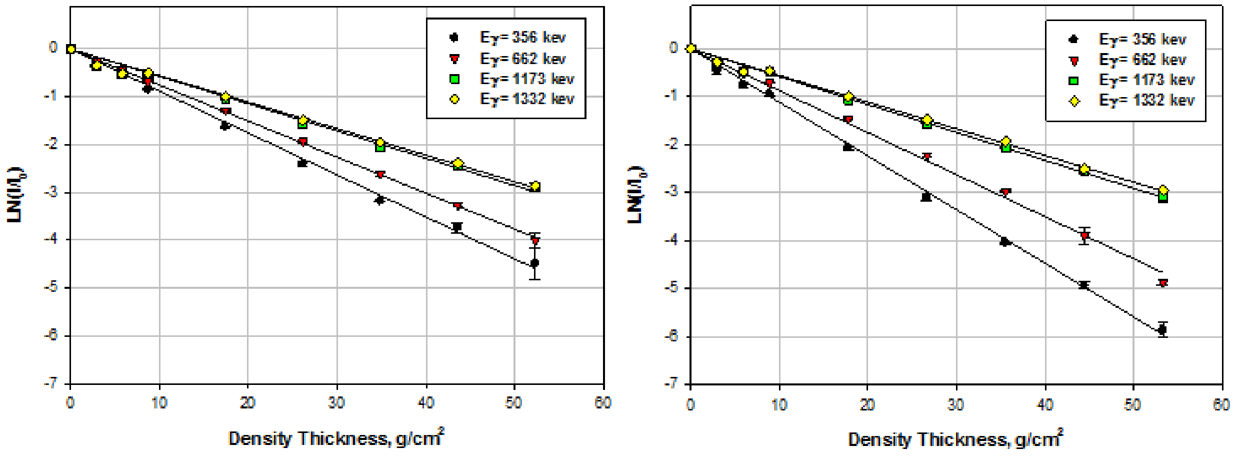
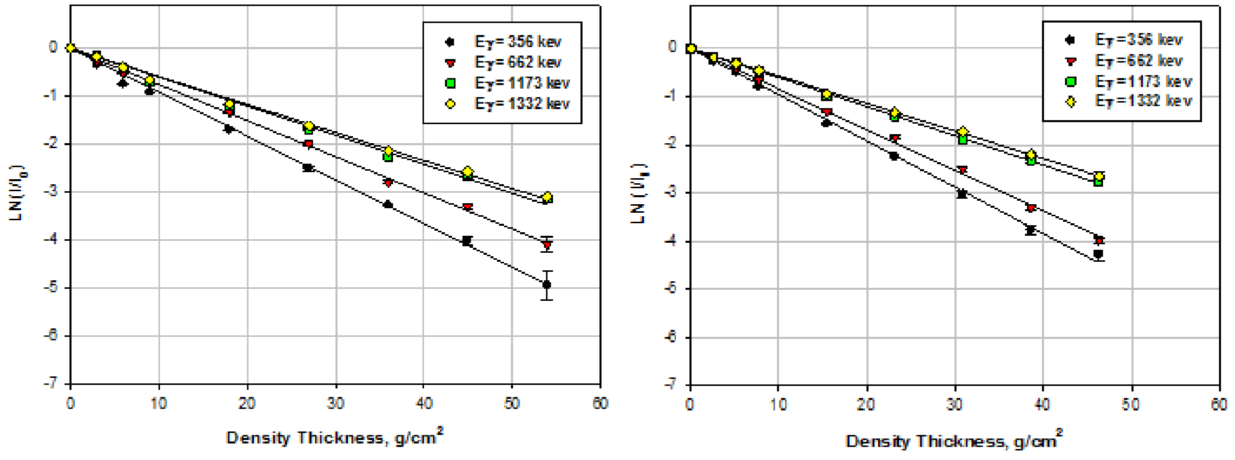


Figure 9: Elastic modulus values for the concrete mixes.



a) G.L mix

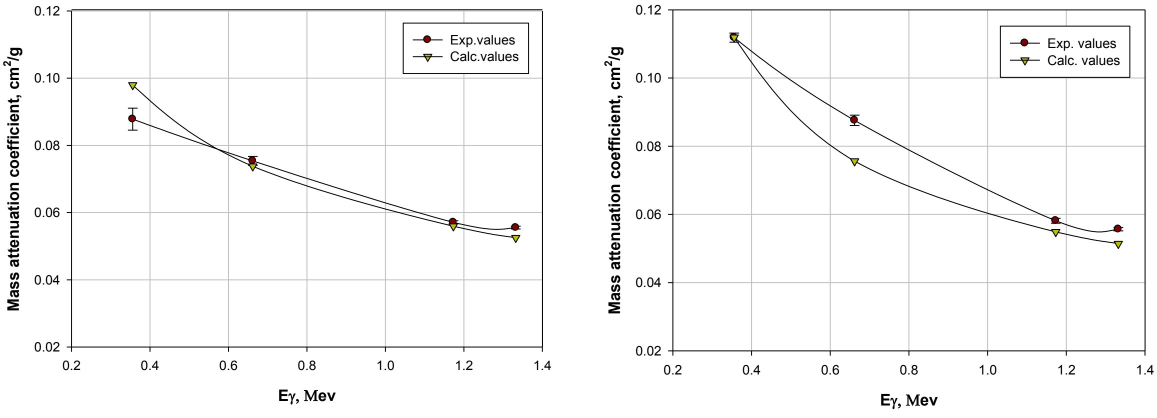
b) B.Lmix



c) S.L mix

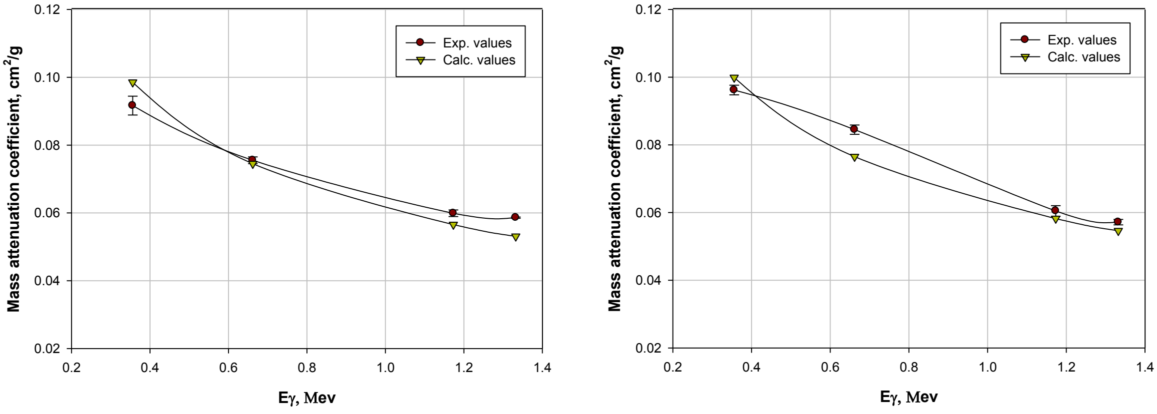
d) D.C mix

Figure 10: Relation between ln (Ix/Io) versus barrier density thickness (ρx) for the four concrete mixes.



a) G.L mix

b) B.L mix



c) S.L mix

d) D.C mix

Figure 11: Relation between mass attenuation coefficients versus gamma energy for the concrete mixes