Effect of lead oxide on concrete density for radiation shielding purposes

Siti Amira Othman*

Department of Physics and Chemistry, Faculty of Applied Sciences and Technology, Universiti Tun Hussein Onn Malaysia
siti.amira@uthm.edu.my*

Abstract. Selection of suitable shielding materials depends on the type, intensity and energy of radiation source, as well as the shielding properties such as mechanical strength, economic preparation and resistance to radiation damage. When compared to ordinary concrete, the addition of some materials with various fractions in the composition of concrete has provided concrete forms more efficient for gamma ray shielding. Concrete has proven to be an tremendous and adaptable shielding material with a well-established linear attenuation coefficient for gamma rays and neutrons. The objectives of this study is to determine the exact composition of concrete constituents and its density. Concrete samples were prepared by mixing cement, water, aggregate and additive with different composition and densities. Density was increases by decreasing the aggregates percent and increasing the additives such as lead oxide. Sample will be compacting to forming a solid material through pelleting and sintering process. Finally, it will undergo for radiation attenuation test. It was found that, substitution of lead oxide by aggregate causes increasing in the specific density and compressive strength of the concrete.

1. Introduction
Concrete is a composite material composed of coarse aggregate bonded together with a fluid cement which hardens over time. Most concretes used are lime-based concretes such as Portland cement concrete or concretes made with other hydraulic cements, such as cement fondu. However, road surfaces are also a type of concrete, asphalt concrete, where the cement material is bitumen, and polymer concretes are sometimes used where the cementing material is a polymer. Increasing the amount of cement in the concrete mixture increases of the yield stress[1]. Good quality aggregate should consist of particles of adequate strength and desirable engineering properties as well as resistance to exposure conditions. The aggregate properties affect the strength, stiffness, and long-term deformation of hardened concrete [2]. For example, some aggregates may react negatively with cement or, in contrast, they may interact beneficially with the cement paste, enhancing the concrete’s strength or stiffness. As a result, concrete properties such as elastic modulus, creep, or shrinkage can vary as much as 100% depending on the aggregate type.
2. Methodology
The materials used for construction of concrete are Portland cement (type I) with a density of 400kg/m³. To increase workability of concrete, about 2.5wt% of cement was used. Distilled water for dilution process. The water to cement ratio is 0.45 by weight. An aggregate with a density 2400kg/cm³ was used in concrete production. Aggregates were graded in accordance to grain size classification ASTM C150, Type I. Lead oxide was used as an additive in forms of powder.

2.1. Sample preparation
Cement, water, aggregate (sand and gravel) and additive will be mix with percentage value of aggregate is vary. Ordinary concrete (without additive) is prepared as a sample reference. By using Ball Mill machine, samples will be grind and blends with speed 250 rpm for 2 hours. Samples will be add with water. By using Hydraulic Press machine, the samples will be press 2 ton about 1 minute. Samples were undergo water curing process for about 7 days by immersed in water at about 25°C. After 7 days of water curing, the samples were expose to heat at temperature of 180°C for about 2 hours. Sample will measure including dose of radiation, intensity of radiation, thickness of concrete and concrete strength.

3. Results and Discussion

3.1 Density Test
In order to produce a high quality concrete, various trial of water to cement ratio of 0.4, 0.8, 1.2, 1.4, 1.6 and 2.0 were prepared. Fig. 1(a) shows the graph of density of concrete and water to cement ratio. The graph shows a decreasing density from 0.4 to 2.0 water to cement ratio. The results obtained were related to the complete hydration in concrete. Each of the compounds in Portland cement will undergoes hydration and contributes to the final concrete product when the water is added. Complete hydration of Portland cement will occur at approximately w/c = 0.4. For the water to cement ratio below than 0.4, the hydration process might be incomplete as the water should fill all the gel pores in order to accomplish the hydration process.

As for water to cement ratio equal to 0.46, Abram’s Law stated that higher water to cement ratio will lower the strength of concrete [3]. This is because as water was added over and over, it will be evaporated which will leave cavities in concrete and indirectly reduce the density of concrete. In other words, the more water was use to mix the concrete, the weaker the concrete mix. The less water use to mix the concrete, the stronger the concrete mix [4]. A mix with little water and more concrete mix will be dryer and less workable but stronger. Table 1 shows the density results. The samples were polished before conducting the test to remove all entire unwanted particles from the surface of concrete. The next phase in producing concrete was to find the best ratio between fine aggregates and cement. Table 1 shows the results obtained from the mix design of concrete using cement, water and sand.

Besides that, table 1 shows the results obtained from the density test of plain concrete samples. From figure 1(b) and 1(c) shows density of concrete is decrease when weight of fine and coarse aggregate is increase due to bond characteristics [5]. The use of larger maximum size of aggregate affects the concrete in several ways. First, since larger aggregates have less specific surface area and the aggregate–paste bond strength is less, aggregate fails along surfaces of aggregates resulting in reduced compressive strength of concrete. Secondly, for a given volume of concrete, using larger aggregate results in a smaller volume of paste, thereby providing more restraint to volume changes of the paste [6].
### Table 1. Ratio of concrete sample

#### Density of concrete to determine water to cement ratio

| Samples | Water (g) | Cement (g) | Water : Cement | Density (g/cm³) |
|---------|-----------|------------|----------------|----------------|
| A       | 2.0       | 5.0        | 0.40           | 2.5117         |
| B       | 4.0       | 5.0        | 0.80           | 2.3605         |
| C       | 6.0       | 5.0        | 1.20           | 2.1561         |
| D       | 8.0       | 5.0        | 1.60           | 1.7218         |
| E       | 10.0      | 5.0        | 2.00           | 1.5086         |

#### Density of concrete to determine fine aggregate to cement ratio

| Samples | Water (g) | Cement (g) | Fine aggregate (g) | Fine aggregate : Cement | Density (g/cm³) |
|---------|-----------|------------|--------------------|-------------------------|----------------|
| A       | 2.0       | 5.0        | 2.0                | 0.4                     | 2.610          |
| B       | 2.0       | 5.0        | 2.5                | 0.5                     | 2.551          |
| C       | 2.0       | 5.0        | 3.0                | 0.6                     | 2.502          |
| D       | 2.0       | 5.0        | 3.5                | 0.7                     | 2.440          |
| E       | 2.0       | 5.0        | 4.0                | 0.8                     | 2.411          |

#### Density of concrete to determine coarse aggregate to cement ratio

| Samples | Water (g) | Cement (g) | Fine aggregate (g) | Coarse aggregate (g) | Coarse aggregate : Cement | Density (g/cm³) |
|---------|-----------|------------|--------------------|----------------------|--------------------------|----------------|
| A       | 2.0       | 5.0        | 2.0                | 2.0                  | 0.4                      | 2.732          |
| B       | 2.0       | 5.0        | 2.0                | 2.5                  | 0.5                      | 2.705          |
| C       | 2.0       | 5.0        | 2.0                | 3.0                  | 0.6                      | 2.621          |
| D       | 2.0       | 5.0        | 2.0                | 3.5                  | 0.7                      | 2.543          |
| E       | 2.0       | 5.0        | 2.0                | 4.0                  | 0.8                      | 2.487          |

#### Density of High Density Concrete

| Samples | Water (g) | Cement (g) | Fine aggregate (g) | Coarse aggregate (g) | Lead (g) | Density (g/cm³) |
|---------|-----------|------------|--------------------|----------------------|----------|----------------|
| A       | 2.0       | 5.0        | 2.0                | 2.0                  | 2.0      | 2.811          |
| B       | 2.0       | 5.0        | 2.0                | 2.0                  | 4.0      | 2.925          |
| C       | 2.0       | 5.0        | 2.0                | 2.0                  | 6.0      | 3.207          |
| D       | 2.0       | 5.0        | 2.0                | 2.0                  | 8.0      | 3.296          |
| E       | 2.0       | 5.0        | 2.0                | 2.0                  | 10.0     | 3.433          |
Figure 1. (a) Graph density of concrete and water to cement ratio (b) Graph density of concrete and fine aggregate to cement ratio (c) Graph Density of Concrete and coarse aggregate to cement ratio (d) Graph density of concrete and weight of lead in concrete (all in gram unit).

Density of concrete is determined by the aggregate density as well as soft with porous concrete produce weak concrete with lower wear resistance [7]. Bulk density of aggregates is the mass of aggregates required to fill the container of a unit volume after aggregates are batched based on volume. A new concrete mix design from different weight of lead was done based on the previous density test and the density was determined using the same equipment. The samples were polished before conducting the test as it will remove unwanted particle from the surface of concrete. The result obtained was shown in Table 1. The mix design of concrete actually really important as it was the key to the production of the high quality concrete [8]. In Fig. 1 above, it can be observed that the pattern of density of concrete is increasing with the higher weight of lead. Instead of the suitable mix design, the pattern of increasing in density of concrete was the result from the lead powder which helps a lot in increasing the density. Lead is a high density material. It also displays a marked tendency toward covalent bonding. Because of that, it is kind of suitable to produce a high dense concrete using this admixture. High density concrete is mainly used for the purpose of radiation shielding, for counterweights and other uses where high density is required. The high density concrete has a better shielding property, so that it can protect harmful radiation [9].
According to the recent study made by previous researchers [4][10], the highest density of concrete that they can produce using lead is 3.542 g/cm³ which is higher compared to the concrete made in this study which the highest density is 3.433 g/cm³. This might be due to the difference in the period of water curing which water curing process in this research was made for 7 days. As mentioned earlier, water curing plays an important role in concrete making as the process will improve the workability and the structure thus effect the density of concrete.

3.2 Analysis for radiation attenuation test
Radiation attenuation test was done using NaI Scintillation Detector with Cs-137 as the source of radiation. The count time was set as 150 seconds. Table 2 below shows the results obtained from the radiation test. The background reading was repeated three times to reduce errors and the average counts rate was calculated.

| Background reading (Counts/s) | Average Counts rate (Counts/s) | Corrected count rate (Counts/s) |
|-----------------------------|-----------------------------|-------------------------------|
| 28476                       | 30020                       | 30020/150 = 200.13            |
| 30118                       |                            |                               |
| 31467                       |                            |                               |

Table 2. Result of radiation count rate

| Sample | Thickness of sample (g/cm²) | Counts rate (Counts/s) | Average Counts rate (Counts/s) | Corrected Counts rate (Counts/s) |
|--------|----------------------------|------------------------|-------------------------------|---------------------------------|
| A      | 2.4160                     | 41864                  | 42917                         | 78.10                           |
| B      | 2.5891                     | 41929                  | 41511                         | 77.81                           |
| C      | 2.7161                     | 41507                  | 41473                         | 76.07                           |
| D      | 2.8350                     | 41316                  | 41604                         | 75.79                           |
| E      | 2.9726                     | 41379                  | 41198                         | 75.79                           |

From the graph plotted in Fig. 2, it can be observed that the pattern of the counts rate is decreasing as the density of the concrete increase. The decline of the count rate shows that γ-ray had been absorbed by the concrete. Gamma ray will be interact with atomic particles of concrete and make them lose the energy. The particles in the concrete samples were packed as the density is increase which will contribute to the loss of gamma energy as many interactions occurred between concrete particles [11]. In conclusion, gamma-ray absorption will increase as the density of materials increased. In this radiation attenuation test, the count rate represented the intensity that passes through the concrete samples. It can be said that the intensity of the gamma ray decreased as the density of samples increases. The thickness relate to the density proportionally [12].
3.3 Analysis for linear attenuation coefficient and half value layer

The determination of linear attenuation coefficient can present the performance of concrete towards radiation shielding [13]. In simple explanation, the higher the attenuation of gamma ray, the performance of concrete will be good. In this part, concrete samples were radiated with Cs-137 with the energy of 0.662 MeV. The value of linear attenuation coefficient for every samples were calculated using Eq. 1. 

$$\mu = \frac{1}{x} \ln \left( \frac{I}{I_o} \right)$$  

(1)

where $\mu$ is the linear attenuation coefficient in cm$^{-1}$ and $x$ is the thickness of the samples in cm. In this study, the thicknesses, $x$ of the samples were fixed as 1 cm. $I_0$ is the gamma ray counts without an absorber and $I$ is the gamma ray count with absorber. Half-value layer was defined as the thickness of the material that reduces the intensity of the beam into half of its original value [14]. It was related to the linear attenuation coefficient by Eq. 2 below.

$$HVL = \ln 2 / \mu$$  

(2)

where $\mu$ (cm$^{-1}$) is the linear attenuation coefficient.

Table 3. Half value layer for concrete samples

| Sample | Thickness (g/cm$^2$) | Density (g/cm$^3$) | $I$  | $I_o$ | $\mu$ (cm$^{-1}$) | HVL (cm) |
|--------|---------------------|-------------------|-----|-------|------------------|---------|
| A      | 2.4160              | 2.811             | 84.89 | 124.42 | 0.382            | 1.814   |
| B      | 2.5891              | 2.925             | 78.10 | 124.42 | 0.465            | 1.491   |
| C      | 2.7161              | 3.207             | 77.81 | 124.42 | 0.469            | 1.477   |
| D      | 2.8350              | 3.296             | 76.07 | 124.42 | 0.492            | 1.408   |
| E      | 2.9726              | 3.433             | 75.79 | 124.42 | 0.495            | 1.400   |
From table 3, it can be conclude that HVL was decreased as the thickness is increases. HVL is an indirect measure of photon energy or beam hardness. HVL is an important quality control test as it is used to measure whether or not there is sufficient filtration in the gamma ray beam to remove energy radiation, which can be damaging [15]. It also helps to determine the type and thickness of shielding required. When a gamma photon passes through a concrete, it does not deposit gradually its energy as an alpha particle on its way: either it interacts and disappears as such, or passes all the way through without interacting. In the case of a large number of gamma, a beam or a number of gamma emerge unscathed. The beam is attenuated. The stronger the ‘probability of interaction’, the faster the attenuation.

![Graph thickness and HVL for concrete](image)

### 3.4 Analysis for Hardness Test

In this research, hardness test was done using Vickers Hardness Method. This test was done in order to relate the hardness and the structure of concrete. The table 4 below shows the results obtained from the hardness test. The value of test force that has been used in this hardness test is quite high which is 4.903 N. In Vickers Hardness test, the value of test force is quite important as it can indicate the degree of hardness of material.

| Lead (g) | Hardness (GPa) | Average hardness (GPa) |
|----------|----------------|------------------------|
| 2        | 2.275          | 2.325                  | 2.312                  | 2.304                  |
| 4        | 4.374          | 4.549                  | 4.599                  | 4.507                  |
| 6        | 5.884          | 5.355                  | 5.433                  | 5.557                  |
| 8        | 7.257          | 7.591                  | 7.217                  | 7.355                  |
| 10       | 9.414          | 9.581                  | 9.752                  | 9.582                  |
Fig 4. shows a graph of hardness and weight of lead. Hardness was increase as weight of lead increase. Many factors influence the rate at which the hardness of concrete after mixing. Strength of concrete is directly related to the structure of the hydrated cement paste. Air in concrete produces voids [16]. Excess of water in concrete evaporate leave the voids in the concrete. Consequently, as the w/c ratio increases, the porosity of the cement paste in the concrete also increases. As the porosity increases, the compressive strength of the concrete decreases. Air also is created during improper mixing, consolidating and placement of the concrete. Air pockets, or irregularly sized air voids, are spread throughout the concrete and have negative effects on product appearance, strength and durability.

Conclusion

It was found that, use lead oxide as additives in concrete causes increasing in specific density and compressive strength of the concrete. Besides that, level of radiation intensity towards concrete composition can be identified. As a result enhances the shielding material efficiency against gamma ray. Based on the result and the discussion obtained from the research, the following conclusion can be drawn:

1. After a few trials, water to cement ratio was decided as 0.40 according to the density test.
2. The highest density of the concrete produced in this research was 3.433 g/cm³.
3. The intensity of gamma ray was reduced with the increasing in the density of concrete.
4. The hardness of concrete increase as the composition of lead increase.

References

[1] Khaled Marar, Ozgur Eren. Effect of cement content and water/cement ratio on fresh concrete properties without admixtures, International Journal of the Physical Sciences 24 (2011) 5752-5765.
[2] Ihab Adam, Kenji Sakata, Toshiki Ayano. Influence of coarse aggregate on the shrinkage of normal and high-strength concretes, J. Faculty of Environmental Science and Technology 6 (2001) 41-45.
[3] Sandor Popovics, Janos Ujhelyi. Contribution to the concrete strength versus water-cement ratio relationship, J. Materials in Civil Engineering 7 (2008) 459.
[4] Saleh H. Alsayed, Mohammad A. Amjad. Strength, water absorption and porosity of concrete incorporating natural and crushed aggregate, J. King Saud University- Engineering Sciences 1 (1996) 109-119.
[5] Shohana Iffat. Relation between density and compressive strength of hardened concrete, Concrete Research Letters 4 (2015) 182-189.
[6] S. Kolias, C. Georgiou. The effect of paste volume and of water content on the strength and water absorption of concrete, Cement and Concrete Composites 2 (2005) 211-216.

[7] Nwokoye S.O, Ezeagu C.A. Optimization of the strength of concrete made from Nigerian processed aggregate, Internatiol Refereed J. of Engineering and Science 4 (2016) 45-61.

[8] Mohd El Khatieb. Trial concrete mix design comparison using local materials under local environment, International Journal of Science and Technology 10 (2017) 1-11.

[9] Suleyman Ozen, Cengiz Sengul, Teoman Erenoglu, Unercolak, Iskander Atilla Reyhancan, Mehmet Ali Tasdemir. Properties of heavyweight concrete for structural and radiation shielding purposes, Arabian J. for Science and Engineering 4 (2016) 1573-1584.

[10] El-Sayed A. Waly, Mohamed A. Bourham. Comparative study of different concrete composition as gamma-ray shielding material. Annals of nuclear energy. 85(2015) 306-310.

[11] Emna Bouali, Abdelhak Ayadi, El- Hadj Kadri, Abdelhak Kaci, Hamza Soualhi, Amjad Kallel. Rheological and mechanical properties of heavy density concrete including barite powder, Research Article- Civil Engineering 45 (2020) 3999-4011.

[12] O. Agar, M.I.Sayyed, F. Akman, H.O. Tekin, M.R. Kacal. An extensive investigation on gamma ray shielding features of Pd/Ag- based alloys, Nuclear Engineering and Technology 3 (2019) 853-859.

[13] Almahdi Mousa, Kus Kusminarto and Gede Bayu Suparta. A new simple method to measure the X-ray linear attenuation coefficients of materials using Micro- Digital Radiography Machine, International Journal of Applied Engineering Research 12 (2017) 10589-10594.

[14] Akkas. Determination of the tenth and half-value layer thickness of concretes with different densities, Acta Physica Polonica A. 129 (2016) 770-72.

[15] M. Begum, A.S. Mollah, M.A. Zaman, A.K.M.M. Rahman. Quality control tests in some diagnostic X-Ray Units in Bangladesh, Bangladesh Journal of Medical Physics 4 (2011) 59-66.

[16] P. Shawnim, F. Mohammad. Porosity, permeability and microstructure of foamed concrete through SEM images, J. Civil Engineering, Science and Technology 10 (2019) 22-23.