Study on the Effect of the addition of Synthesized Nano scale Lead Oxide for Concrete Samples used in Gamma-Ray Shielding

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***Abstract*- Our goal in this research is to investigate the effect of concrete incorporated with nanoparticles of lead oxide in powder form by different percentages on Gamma-ray shielding characteristics. The lead oxide nanostructure was synthesized through the reaction of citric acid ($C_6H_7O_8.H_2O$) solution and lead acetate (Pb ($C_2H_3O_2$)) solution as stabilizer and precursor, respectively. In this synthesis, the prepared lead oxide nanostructure was characterized by X-Ray Diffraction (XRD) and Scanning Electron Microscopy (SEM) techniques. The prepared PbO consists of the crystallites about 50 nm. The concrete samples were prepared according to the local standards of building materials and doped with PbO nano powders by different percentages, 0%, 5%and 10% (by weight) by replacing cement and keeping constant w/c ratio. Moreover, commercial PbO bulk powder additive was used to check the effect of particle size on concrete attenuation properties. The γ-rays attenuation coefficients were measured as a function of the additive percentage of nanoparticles of lead oxides and using γ-ray point source,^{126}Ra with different energies in the range (0.295- 1.73) MeV. The results were compared with that for normal concrete incorporated with the same percentage of bulk lead oxide. It was found that the γ-ray attenuation coefficient for concrete doped with nanoparticles of PbO was slightly improved.**

I. Introduction

Concrete has been used in the construction of nuclear facilities because of two primary properties; its structural strength, and its ability to shield radiation. Concrete structures have been known to last for hundreds of years, but they are also known to deteriorate in very short periods of time when exposed to detrimental physical and chemical environmental conditions [1]. The recent research orientation in radiation shielding is to develop denser concrete by adding suitable percentage of additives [5–12]. As an example, minerals such as magnetite, hematite, goethite and limonite were incorporated with concrete and their effects have been evaluated [5–8]. Effects of barite and lead additives in concrete have been separately investigated [9–12]. Similarly, the effect of lime/silica ratio of concrete specimens on gamma absorption and variation of attenuation coefficient for cement specimens have been verified. The use of concrete in nuclear facilities for containment and shielding of radiation and radioactive materials has made its performance crucial for the safe operation of the facility. Spent Nuclear Fuel (SNF) is stored in concrete structures, casks, and vaults for planned storage up to 40 years [2]. Nano particles have attracted increasing attention in recent years and their different types have been used in concrete mixtures in order to improve both the mechanical properties and pore structure of the concrete. The effect of these particles, mostly PbO nanoparticles, has been studied by many researchers.

In the present paper, samples of concrete without and with different percentages by weight (0%, 5%, 10%) of nanoparticles of lead oxide were prepared. Gamma-ray shielding properties of the samples were investigated. Experimental mass attenuation coefficients for these samples were measured. Many previous works compared the experimentally acquired vs. theoretically calculated linear attenuation coefficients for several concrete compositions at different photon energies [16,17].

Photoelectric effect is the major interaction process of γ-rays with the shielding material, while compton scattering and pair production mechanisms contributes only at medium and high energy photons. Linear attenuation coefficient is the simplest absorption coefficient to be measured experimentally using Lamber–Beer law,

$$I = I_o e^{-\mu_m x} \tag{1}$$

where, I_0 and I are the incident and transmitted photon intensities, respectively, x is the thickness of the absorbing material, and μ is the linear attenuation coefficient. Mass attenuation coefficient ($\mu_m = \mu/\rho$) is more convenient to be used as it is related to reaction cross section, σ.

The aim of this work is to study the γ-rays attenuation coefficient of concrete doped by nano-lead oxide at several weight percentages and to determine the nano-size additives that provide maximum improvement of attenuation coefficient compared with that for bulk lead oxide additive of normal size.

II. Experimental Work

A. *Synthesis of PbO nanostructure*

Lead acetate of 5.4055 gm was dissolved in suitable volume of distilled water to obtain 25 ml aqueous solution. Citric acid was prepared as aqueous solution then it was added to the lead acetate. The aqueous solution was then stirred for 0.5 h to mix the solution uniformly until transparent colloidal suspension was obtained. The transparent solution was transformed into dry gel on two steps: evaporating for 20 h at 80°C in a water bath to form wet gelatin, and then drying in oven at 130°C for 8 h. Conditions of formation of gels strongly depended on the nature of the organic acid employed. With citric acid added, white precipitate of lead acetate immediately appeared. Formation of the gel takes place by solvent evaporation during heating. The dry gel was then heated till auto-combustion. Finally, the combustion ash was calcinied at 500°C for 2 h to form PbO nanostructure.

B. Characterization

X-Ray diffraction (XRD) pattern was recorded by powder X-ray diffractometer (Philips, PW 3710, operated at 40 kV, 30 mA) employing Cu K_α radiation (λ = 1.5406 Å). The average crystallite size of the product was calculated from the Scherrer equation using the major diffraction peak of the corresponding PbO (111):

$$t = \frac{k\lambda}{\beta \cos\theta} \qquad (2)$$

where, t is the average crystallite size, k is a constant (0.9), λ is the wavelength of X-ray (Cu Ka), β is the full width at half maxima, and θ is the diffraction angle in degree.

Also, particle morphology and compositional analyses of the prepared PbO nanostructure were carried out using a scanning electron microscopy and an energy dispersive X-Ray analysis (EDAX) set up attached to an SEM (Philips XL-30). The samples were coated with gold to increase their conductivity before scanning.

C. Sample Preparation

Nano lead oxide powder was added to concrete at three concentrations 0% (no additives), 5%, and 10% by weight. For every concentration, disk-shape samples of diameter = 21mm and average thicknesses = 21 mm were casted inside their plastic molds to be used in γ-ray measurements, as presented in Table 1.

Gamma ray intensities behind concrete samples with different percentages of nanoparticles of PbO were measured. A collimated beam of a point isotropic Gamma-Ray sources, ^{226}Ra, with different energy range from (0.29- 1.7) MeV were used. The measurements of the beam intensities after the samples irradiation were carried out using High Purity Germanium (HPGe) detector.

III. Results and Discussion

A. Using XRD Spectra

The X-ray diffraction spectrum of the prepared nanoparticles of PbO sample is shown in Fig. 1. The intense peaks at several diffraction angles indicate the high degree of crystallites of the oxide. The spectrum contains two different types of diffraction patterns, corresponding to, PbO (tetragonal) and PbO (orthorhombic), which matches the JCPDS card number 38-1477. The presence of the low intense peak corresponds to PbO_2 phase that may be generated due to higher oxidation state of lead oxide. Also, the spectrum of the prepared bulk sample of PbO is shown in Fig. 2.

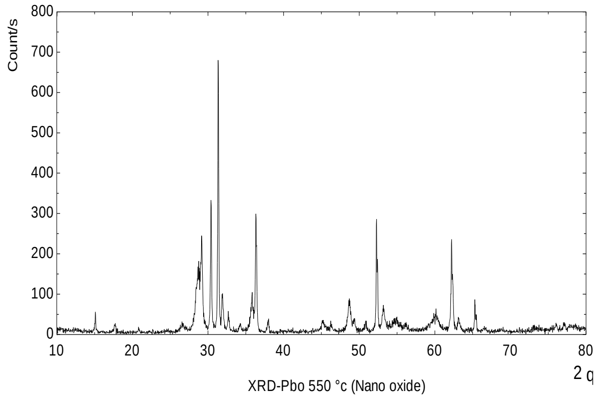

Fig. 1. XRD for nanoparticle sample of PbO

B. Using Scanning Electron Microscopy (SEM)

Figure 3 shows the morphology of pure PbO bulk and PbO nanostructure. SEM investigations of the samples reveal the crystallites nature of the nanoparticles.

TABLE 1
SAMPLES DATA

Sample Number	Composition (Concrete + Additives)	Masses of components (g)				Thickness (mm)	Density (g/cm^3)
		Cement	Sand	Water	Additives		
1	No additives	3.74	14.34	2	Nil	21	2.08
2	5% nano lead oxide	3.55	14.34	2	0.2 g (Nano-Lead oxide)	20	2.188
3	10% nano lead oxide	3.37	14.34	2	0.4 g (Nano-Lead oxide)	20	2.115
4	5% bulk lead oxide	3.55	14.34	2	0.2 g (Bulk Lead oxide)	20	2.10
5	10% bulk lead oxide	3.37	14.34	2	0.4 g (Bulk Lead oxide)	18	2.20

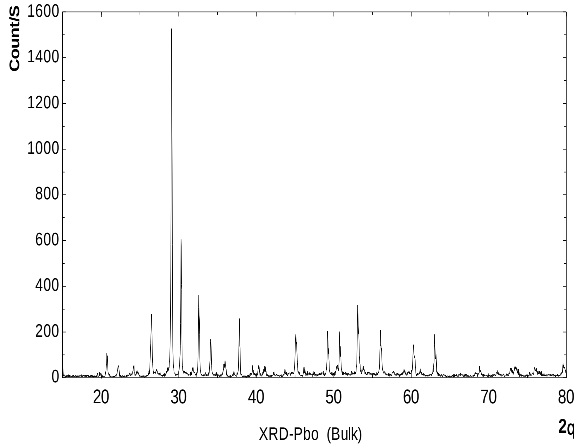

Fig .2. XRD Spectrum for Bulk sample of PbO

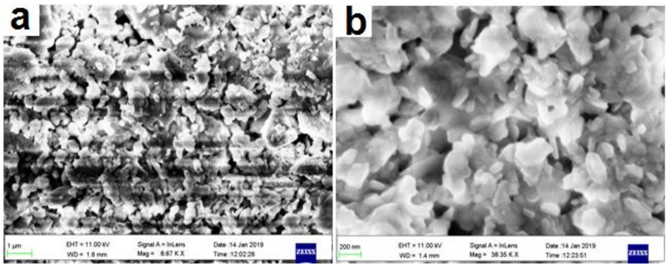

Fig. 3. SEM for lead oxide, a) for bulk particle, (b for nano particles

Linear attenuation coefficient is the simplest absorption coefficient to measure experimentally, but it is not usually tabulated because it depends on density of the absorbing material. Gamma rays interact primarily with atomic electrons; therefore, the attenuation coefficient is proportional to the electron density which is proportional to the bulk density of the absorbing material.

The ratio of the linear attenuation coefficient to the density (μ/ρ) is called the mass attenuation coefficient, μ_m and has the dimensions of area per unit mass (cm^2/g). The units of this coefficient hint that one may think of it as the effective cross section of electrons per unit mass of absorber.

Table 2 shows the variation of γ-ray counts per second –as the peak area- and the mass attenuation coefficient (μ/ρ) in (cm^2/g) with the γ-ray energies, for samples of concrete without and with bulk-lead oxide of 5% ratio and nanoparticles of lead oxide of 5% ratio. Figures (5-7) show γ-ray mass attenuation coefficients (μ/ρ) vs. γ-ray energies in keV, for these samples of concrete.

Gamma ray intensities behind samples was measured using a collimated gamma beam from a point isotropic ^{226}Ra source, with range of energy 185.7 keV to 1764 keV using High Pure Germanium (HpGe) detector. It was found that the mass attenuation coefficient (μ/ρ) decrease with increasing γ-ray energies in keV. Below 1MeV γ-ray energy, there was no improvement in shielding properties using nanoparticle size at 5%. Between 1-1.8 MeV γ-ray energies, attenuation coefficient was significantly higher for the 5% PbO nanoparticle doped concrete.

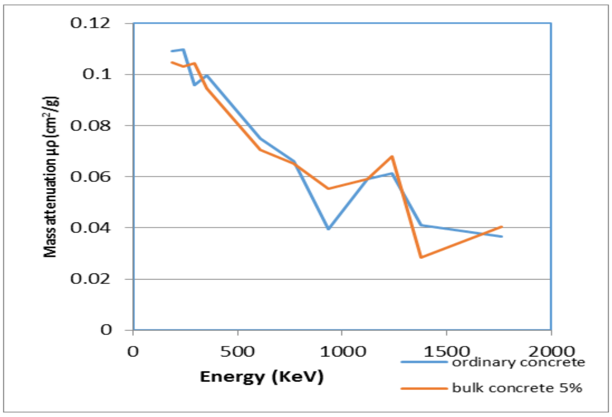

Fig. 5. Mass attenuation coefficient of ordinary concrete and concrete 5% bulk PbO

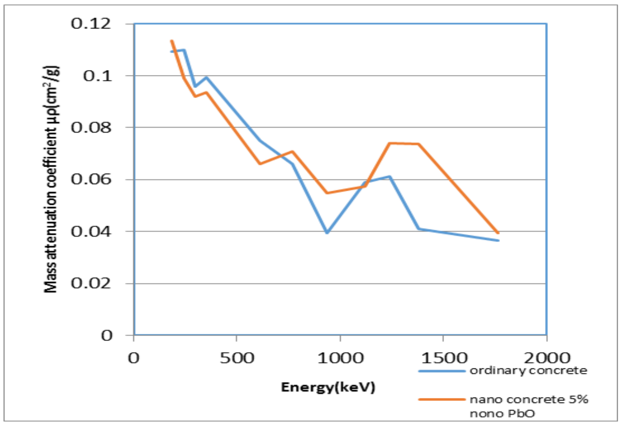

Fig. 6. Mass attenuation coefficient of ordinary concrete and concrete with 5% nano PbO

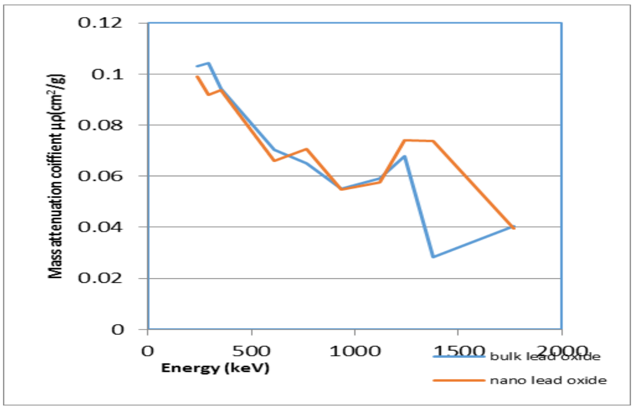

Fig.7. Mass attenuation coefficient of concretewith 5% bulk PbO and with 5% nano PbO

In case of increasing the ratio of nanoparticles of lead oxide up to 10%, results were improved as shown in Table 3 and in Figures (8 - 10).

IV. CONCLUSIONS

Nano lead oxide powder was added to concrete at three concentrations 0% (no additives), 5%, and 10% by weight.

For every concentration, disk-shape samples of diameter = 21mm and average thicknesses = 21 mm were casted inside their plastic molds to be used in γ-ray measurements**.**

TABLE 2

GAMMA INTENSITY AND MASS ATTENUATION OF DIFFERENT NANO COMPOSITIONS (IN CASE OF 5% RATIO)

γ-ray Energy (MeV)	**Intensity %**	**Io(γ-ray intensity without sample)**	**I1 (γ-ray intensity of ordinary concrete)**	**I_2 (γ-ray intensity of concrete with 5% bulk PbO)**	**I_3 (γ-ray intensity of concrete with 5% nano PbO**	**μρ 1(mass attenuation coefficient of ordinary concrete)**	**μρ 2(mass attenuation coefficient concrete with 5% bulk PbO)**	**μρ 3(mass attenuation coefficient concrete with 5% nano PbO)**
185.7	3.59	3770	2340	2430	2330	0.109186	0.104568	0.113599
241.9	7.43	6720	4160	4360	4420	0.109792	0.103004	0.098902
295.2	19.3	15500	10200	10000	10500	0.0958	0.104346	0.091942
352	37.6	27800	18000	18700	18700	0.099511	0.094408	0.093605
609	46.1	25400	18300	18900	19200	0.075057	0.070378	0.066062
768	4.9	2470	1850	1880	1830	0.06617	0.064987	0.070798
934	3.13	1450	1220	1150	1150	0.03954	0.055191	0.054722
1120	15.1	6150	4750	4800	4820	0.059136	0.059009	0.057526
1238	6	2340	1790	1760	1710	0.061341	0.067818	0.074046
1378	4	1340	1120	1190	980	0.041058	0.028266	0.07386
1764	15.4	4410	3760	3720	3730	0.036505	0.040512	0.039534

TABLE 3.

GAMMA INTENSITY AND MASS ATTENUATION OF DIFFERENT NANO COMPOSITIONS (IN CASE OF 10% RATIO)

Photon energy (keV)	Intensity	Io(γ-ray intensity without sample)	I1 (γ-ray intensity of ordinary concrete)	I_2 (γ-ray intensity of concrete with 10% bulk PbO)	I_3 (γ-ray intensity of concrete with 10% nano PbO	μρ, 1(mass attenuation coefficient of ordinary concrete)	μρ, 2(mass attenuation coefficient of concrete with 10% bulk PbO)	μρ, 3(mass attenuation coefficient of concrete with 10% nano PbO)
185.7	3.59	3770	2340	2300	2070	0.109186	0.124789	0.141732
241.9	7.43	6720	4160	4530	4130	0.109792	0.099587	0.115085
295.2	19.3	15500	10200	10700	10100	0.0958	0.093585	0.101254
352	37.6	27800	18000	19800	18000	0.099511	0.085695	0.102758
609	46.1	25400	18300	20200	18700	0.075057	0.057845	0.072394
768	4.9	2470	1850	2050	1940	0.06617	0.047065	0.057099
934	3.13	1450	1220	1160	1090	0.03954	0.056349	0.067467
1120	15.1	6150	4750	5150	4960	0.059136	0.044812	0.050838
1238	6	2340	1790	1870	1770	0.061341	0.056619	0.065998
1764	15.4	4410	3760	2920	3690	0.036505	0.104114	0.042139

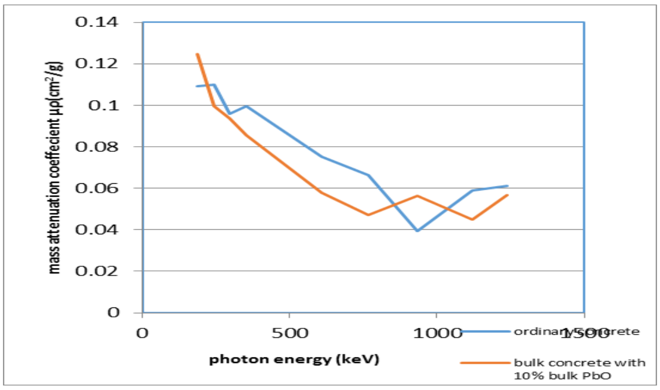

Fig. 8. Mass attenuation coefficient of ordinary concrete and concrete with 10% bulk PbO

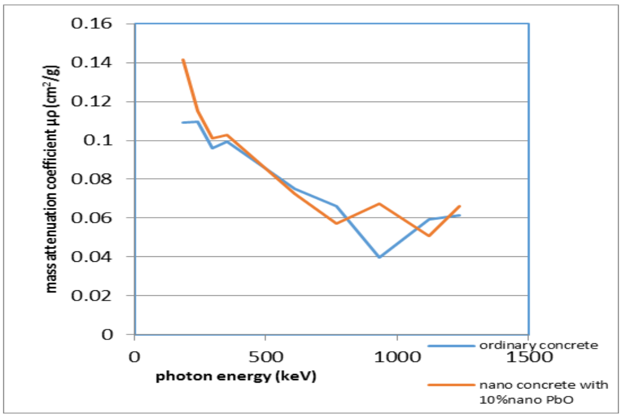

Fig. 9. Mass attenuation coefficient of ordinaryconcrete and concrete with 10% nano PbO

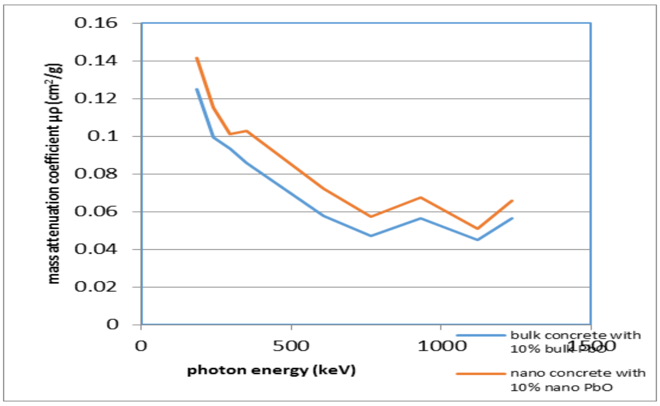

Fig. 10. Mass attenuation coefficient of concrete with 10% bulk PbO, and with 10% nano PbO

The morphological structure and γ-ray shielding properties of the nanoparticles of PbO doped in concretes were studied using High Pure Germanium (HpGe) detector. The prepared lead oxide nanostructure was characterized by X-Ray Diffraction (XRD) and Scanning Electron Microscopy (SEM) techniques. XRD and SEM measurements confirmed the phase purity and particle sizes of the prepared nanoparticles of PbO.

The comparative study performed for the measured values of the mass attenuation coefficients for the concrete doped by nanoparticles of PbO with different percentages didn't reveal any improvement up to 5%, but when using 10% of nanoparticles of lead oxide doping, slightly improvements were attained specially at Gamma- ray energies in the range, 185.7 - 1764 keV.

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