INVESTIGATION OF THE GAMMA SHIELDING EFFICIENCY REDUCTION BY DEPOSITING PbO AND MnO₂ COMPOSITION ON VARIOUS TYPES OF SUBSTRATES

by

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In this study, a PbO, MnO_2 , PbO + MnO_2 thin film (nano layer) was deposited onto a microscope glass, indium tin oxide glass, and aluminum with thicknesses 0.1 mm and 0.5 mm substrate, samples were prepared by the thermal spray technique. The coating of thin films was carried out onto the substrate at a temperature of 35 °C, furthermore, the prepared shield films were annealed at 100 °C and 250 °C for one hour. The shielding efficiency of each thin film on each substrate at different temperatures was analyzed using radioactive source ⁶⁰Co and NaI(Tl) well-type scintillation detector. The experimental results obtained were summarized: adding PbO or MnO₂ thin film together or separately to the substrate increases the attenuation of gamma-rays depending on the type of samples and annealed temperature. The minimum gamma reduction rate was found 0.3 % for indium tin oxide glass deposited with MnO₂ and annealed at 250 °C while the maximum was found in Al with thickness 0.1 mm (9.6 %) deposited with both PbO or MnO₂ together. This means that shielding efficiency increases by increasing the thickness of the thin film, annealed temperature and substrate type.

Key words: gamma shielding, MnO₂, PbO composition, thermal spray, scintillation detector

INTRODUCTION

The fields of industry, medicine, research centers, and particle accelerators use radiation in their facilities, making it very useful for regular use. The working of many applications depends on radiation. Exposure to an excessive amount of ionizing radiation may lead to permanent tissue damage, cancer and acute radiation syndrome [1, 2]. Nowadays radiation research not only has been of high relevance to human life but also for microelectronic and semiconductor devices. Due to the high demands in the number of manufactured devices and their areas of application and decrease in the geometric dimensions of microcircuits and semiconductor devices, which demand to leave most traditional methods of protection against the negative effects of ionizing radiation, the assessment of microelectronics device degradation under operating conditions when exposed to ionizing radiation and increased background radiation become a significant area of study [2-4].

Shielding materials are useful for protecting humans and their background from the danger of radiation [3, 5]. Shields are used to protect employees or patients from these side effects, decreasing exposure to radiation involved in these procedures. Absorption of radiation by shielding can protect humans, this absorption occurs via the incoming photons through three mechanisms, photoelectric absorption, Compton scattering, and pair production, which have an important role in the process of gamma radiation [6, 7]. These three mechanisms lead to the transfer of the incident photon energy partially or completely and reduce the intensity of radiation to safe levels [8, 9].

There are different types of radiation shields, from metal sheets to glass and concrete. Simple lead is very effective at shielding gamma radiation and has historically been the most common shield due to its high density which makes it highly effective for reducing the energy of photons. However, its use should be limited in medical treatment facilities due to its toxic nature. In this respect, much research has been done throughout the world to study environmentally friendly shields that substitute lead [9, 10]. The most suitable candidate for this is concrete because of its simple manufacturing, low cost and wide range of composition while over time it loses water and is prone to cracking, these properties lead most researchers to look for an alternative radiation shield [11-13]. Through the use of effective shields this protection can be achieved. For that reason researchers require the studying of the characteristics of potential shielding

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materials, such as expected energies, availability of materials, mechanical properties, characteristics of different radiation attenuation, cost, and ease of production.

Radiation shielding glasses have a similar advantage to concretes, except they offer the additional benefit of being transparent. Adding a secondary layer to glass may increase the absorption of radiation and alter the characteristics of the glass to fit its application because thin film materials, in the size from a few nanometers to micrometers, have electrical, magnetic, chemical, optical, thermal, acoustic, mechanical and other properties different from bulk materials due to the quantum confinement effects, large surface to volume ratio and many other interesting effects [14, 15]. In view of this, the main aim of this study is to assess the prospects of using a microscope, indium tin oxide glasses deposited and aluminum sheets with lead (II) oxide (PbO), manganese dioxide (MnO₂), and both (PbO + MnO₂) thin films by depositing samples with PbO, MnO_2 and $PbO + MnO_2$ composition, to evaluate the change in the gamma-ray shielding efficiency and create local protection against the ionizing radiation.

EXPERIMENTAL METHOD

Shield sample preparation

Synthesis of microscope glass, indium tin oxide (-ITO) glasses, and two pieces of aluminum with different thicknesses of 0.1 mm and 0.5 mm with dimensions of about 2 cm 3 cm were prepared as the samples. Then, the samples were used as a substrate for the spray deposition of manganese dioxide (MnO_2) and lead (II) oxide (PbO) separately and both MnO_2 + PbO together thin films, fig. 1. The substrates were soaked in chromic acid for 24 hours, washed with distilled water, and rinsed in acetone. Then, the substrates were ultrasonically cleaned with ethanol and distilled water, respectively, for 15 minutes to remove the remaining residual and finally left to dry in the desiccator before being used for deposition. The chemical materials used for the preparation of MnO₂ and PbO thin films were bought

from the pro analysis ACS Company without further purification with molarity concentration 0.15. Both MnO_2 and PbO separately were peppered with acetone then the solution was stirred with a magnetic stirrer at room temperature to get a homogenous solution.

The precursor growth MnO_2 and PbO solution flow rate is about 1 mL per second, and the distance between the glass substrates and the spray gun atomizer is around 30 cm as shown in fig. 2. Each thermal spray growth cycle lasts for three seconds, after which there is a 10 second break. The waiting time allows the glass substrate to attain the required temperature for growth before starting the succeeding growth thermal spray cycle. This simple thermal spray growth process required three cycles to produce consistent MnO_2 and PbO throughout the entire substrates at different growth temperatures.

Attenuation of radiation by shielding

Gamma-rays are ionizing radiation and are thus hazardous to life. Due to their high penetration power, they can damage bone marrow and internal organs. The intensity of gamma-rays in materials follows the exponential law [16]

$$I \quad I_{\rm O} {\rm e}^{\ \mu x} \tag{1}$$

where *I* is the intensity of the photons that passed through the absorber, $I_{\rm O}$ – the initial intensity of the photons, x [cm] – the thickness of the absorber, and μ [cm⁻¹] is the linear attenuation coefficient. This coefficient is a constant that describes the fraction of attenuated incident photons in a monoenergetic beam per unit thickness of a material.

Count measurement

Thallium-activated sodium iodide NaI (T1) as a detector and ⁶⁰Co as a radioactive source were used to collect the data. Both detector and radioactive source were placed inside the enclosed geometry of the lead block, ⁶⁰Co at one end of the enclosed shield and the



Figure 1. Shields that were used in the study; (a) ITO glass samples, (b) normal glass samples, and (c) aluminum samples



NaI (TI) detector on the other side. The background radiation was measured before placing the radioactive source in front of the detector. Then the count rate of the main gamma peaks (1173 keV and 1332 keV) was measured after placing ⁶⁰Co in front of the detector. Then the attenuation of radiation in each sheet was obtained by putting the prepared shield between the source and the detector and recording the count rate again. After that, the prepared shield was put in the oven at temperatures of 100 °C and 250 °C. The same procedure for count measurement was recorded for both 100 °C and 250 °C.

Data analyses

The radioactive source spectrum was analyzed by pulse master computerizing software and data measurement was recorded. Due to the decay of radioactive material being a random process and fluctuation in the number of counts emitted by the same isotopes, the counting statistics were conducted by repeating the data of the same sample many times. To obtain the count rate, the measured counts were subtracted from the background radiation and divided by a finite period of time. Furthermore, the Statistical Package for Social Sciences software and Excel software were used for data analyses.

RESULTS AND DISCUSSION

This work investigated the gamma-ray shielding capabilities attenuation by depositing microscope glass, ITO glass, and aluminum with thickness of 0.1 mm and 0.5 mm sheets with deposition of MnO_2 , PbO, and both MnO_2 and PbO together with a thin films composite at three different temperatures 37 °C, 100 °C and 250 °C.

Figures 3-6 show the evaluation results of the efficiency of reducing the intensity of gamma radiation using ITO glass, microscope glass, and Al with two different thickness samples at three different temperatures, depending on the type of thin film deposited on the shield.

Comparing the fig. 3(a)-(d) shield without thin film and shield deposited with thin film composition, mostly the highest reduction rate was found at 250 °C, it is clear that an increase in the processing temperature increases the post-processing grain size which leads to a reduction in the transportation of radiation [17, 18]. Results of adding the PbO layer to all study samples found the highest intensity reduction at 250 °C while the lowest was found in ambient temperature, except the microscope glass shield. In the case of the MnO₂ thin film, fig. 5, the highest reduction in ITO glass and Al with a thickness of 0.1 mm was found at 250 °C, while in both microscope glass and aluminum shield (0.5 mm) it was found at room temperature.

Furthermore, one can demonstrate that fig. 3(d) shielding efficiency increases by increasing the thickness of the thin film as well, adding both PbO and MnO_2 layers together to the shield could change the intensity of the gamma-ray, these results indicate a good agreement with other researchers [19, 20]. Also, one can note that the influence of microscope glass composition was more pronounced than ITO glass. This difference is due to the coating glass substrate with a thin and uniform layer of indium tin oxide, making the glass both highly transparent and low resistance.

At the same time, changing the composition of microscope glasses, ITO glass, and Al by adding PbO, MnO_2 or both composition lead to an increase in shielding efficiency, depending on the thin film tem-







■ 37 °C

Figure 3. Count rate of different shields deposited with; (a) no deposition, (b) Pb thin-film, (c) MnO₂ thin film, and (d) both PbO and MnO₂ thin-film at three different temperature degrees



Figure 4. Americium spectrum analyses attenuated using the ITO glass shield compared to; (a) ITO glass added with PbO thin film and (b) ITO glass added with MnO2 thin film

perature production. This effect was due to the absorption capabilities of MnO_2 and PbO, as well as a change in the grain size of the substrate and thin film at a dif-

ferent temperature, which had a direct effect on the shielding efficiency. Table 1 demonstrates the reduction rate percentage of a gamma-ray by adding thin film, and indicates an improvement in the shielding enhancement of both used types of glasses and aluminum sheet samples.

The general view of the presented trends in the reduction rate indicates that a change in the composition of the studied substrate led to an increase in the gamma radiation absorption efficiency, as well as a decrease in its efficiency if the shield did not contain MnO_2 and PbO. Moreover, results indicate that the reduction rate of the substrate containing MnO_2 is more than PbO while the substrate that contains both PbO and MnO_2 has the highest reduction rate. Furthermore, the paired sample *T*-test was applied to the data to examine whether there were significant differences between the shield before and after adding the thin film. The significant (p < 0.05) correlation was found between the radiation shielding before and after adding PbO, MnO₂ and both compositions.

On the other hand, behavioral changes were seen in each shield different from the other one, namely low, medium, and high reduction rates, as a consequence of the interaction between the substrate and thin film. Our results indicated that the minimum attenuation of radiation of 0.3 % found in the ITO glass shield deposited with MnO_2 thin film at 250 °C while, the maximum reduction rate (9.6 %) of radiation noted

| | Reduction rate [%] | | | | | | | | |
|--------------|--------------------|--------|--------|------------------|--------|--------|---------------|--------|--------|
| Shield type | РЬО | | | MnO ₂ | | | $PbO + MnO_2$ | | |
| | 37 °C | 100 °C | 250 °C | 37 °C | 100 °C | 250 °C | 37 °C | 100 °C | 250 °C |
| ITO glass | 4.2 | 5.5 | 0.6 | 4.4 | 5.8 | 0.3 | 5 | 7.1 | 1.6 |
| Normal glass | 2.2 | 0.5 | 0.4 | 8 | 0.6 | 0.9 | 3.7 | 0.8 | 0.9 |
| Al | 2.9 | 3.2 | 2.2 | 8.4 | 2.6 | 1 | 5.3 | 4.2 | 2.1 |
| Al (0.1 mm) | 7.3 | 3.9 | 5.5 | 8 | 4.2 | 4.8 | 7.4 | 2.1 | 9.6 |
| Minimum | 2.2 | 0.5 | 0.4 | 4.4 | 0.6 | 0.3 | 3.7 | 0.8 | 0.9 |
| Maximum | 7.3 | 5.5 | 5.5 | 8.4 | 5.8 | 4.8 | 7.4 | 7.1 | 9.6 |

Table 1. The percentage of the reduction rate of gamma radiation intensity of the shield containing PbO, MnO₂, or both composition thin film



Figure 5. Results of linear attenuation coefficients (cm⁻¹) of (a) PbO to the glass shield, (b) MnO₂ to the glass shield (c) PbO and MnO₂ to the glass shield, (d) PbO to the aluminum shield, (e) MnO₂ to the aluminum shield, and (f) PbO and MnO₂ to the aluminum shield, as a function of temperature

in the substrate aluminum with (0.1 mm) thickness deposited with both PbO and MnO_2 composition together. Figure 4(a) and 4(b) depicts the comparison of behavioral changes of attenuation of the americium spectrum using the ITO glass substrate added by PbO

and MnO₂ composition, respectively. This effect was most pronounced for gamma rays with low energy.

Figure 5(a)-5(c) shows the dependences of the change in the linear attenuation coefficient of gamma radiation with a different substrate for the different

temperatures deposited by PbO, MnO₂, and both compositions (PbO and MnO₂), respectively. The general view of the presented trends in the change in the coefficients linear attenuation indicates that a change in the thin film deposited on substrate composition or preparing thin films at different temperatures led to variation in gamma-ray intensity. A different trend was observed about the linear attenuation coefficient between microscope glass and ITO glass, especially at 100 °C. With increasing temperature, ITO glass deposited with PbO, MnO₂, and both compositions recorded the highest µ at 100 °C while microscope glass recorded the lowest one at this temperature. By increasing temperature, especially at ~200 °C, amorphous thin films of ITO undergo a phase transition and develop polycrystalline phases with increased optical gap energies [21].

Furthermore, the μ variations with increasing temperature are noticeable at different substrate thicknesses. Figure 5(d)-5(f) shows how shielding efficiency depends on the linear attenuation coefficient, depending on the thickness of the protective aluminum. As can be noted from the presented data, the shielding efficiency increases by increasing the thickness of Al, which can be explained quite simply: by reducing thickness of the protective shield, the minimum quantity of ionizing radiation was absorbed into it.

Figures 6-8 show the changing trend of half-value layer (cm) values of investigated glass and aluminum shield deposited with studied thin film as a function of temperature. In the figures, the vertical primary axis represents the comparison between the half-value layer between ITO and microscope glass, while the secondary vertical axis presents the difference between two aluminum shields with different thicknesses (0.5 mm and 0.1 mm). The microscope glass sample deposited with all study thin film has the greatest value of the half-value layer across most temperature degrees when compared to the ITO studied glasses except adding the MnO₂ thin film at 250 °C.



Figure 6. Variation of the half-value layer (cm) of investigated substrate added with PbO thin film as a function of temperature



Figure 7. Variation of the halfvalue layer (cm) of investigated substrate added with MnO₂ thin film as a function of temperature



Figure 8. Variation of the halfvalue layer (cm) of investigated substrate added with both PbO and MnO₂ thin films as a function of temperature

At the same time, the shield thickness in combination with the thin film was the most effective parameter in shielding properties [20, 21]. The efficiency of the shield increased by adding the thin film to the thin substrate with minimum thickness. This means that the effectiveness of shielding by adding a thin film on aluminum with a thickness of 0.1 mm was more than Al with 0.5 mm. In terms of the half-value layer, results showed that the half-value layer of Al with a thickness of 0.5 mm samples greater than Al with 0.1 mm. All obtained observations confirm that the addition of PbO and MnO₂ separately and together in thin films to the study samples can be considered promising materials for nuclear radiation attenuation.

CONCLUSION

This research summarizes the positive effect of adding thin film to nuclear shielding. To achieve that PbO and MnO_2 separately and together as thin film composition were added to four different shields (microscope glass, ITO glass, Al with a thickness of 0.1 mm, and Al with a thickness of 0.5 mm) at three different temperatures. During the experimental study, it was found that the addition of PbO and MnO_2 sepa-

rately or together to the composition of glasses or aluminum led to an increase in the efficiency of shielding gamma radiation. According to the temperature of the shield preparation, the difference between the substrate deposited with PbO and MnO_2 in terms of attenuation was different. However, when PbO and MnO_2 were used together as glass and Al deposits, the screening efficiency increased more compared to the cases of depositing with only one of the PbO and MnO_2 . Therefore, one can conclude that the shielding efficiency had not only dependence on the type of thin film composition but also on their thickness, layers, and temperature as well, the variation of which played a very important role in determining the absorbing and shielding characteristics.

AUTHORS' CONTRIBUTION

The corresponding author W. A. Alhamdi measured the attenuation of gamma-rays, analyzed the data and wrote the paper while author H. Khalil prepared the shield samples.

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ИСТРАЖИВАЊЕ ЕФИКАСНОСТИ ГАМА ЗАШТИТЕ ДЕПОНОВАЊЕМ РЬО И MnO₂ САСТАВА НА РАЗЛИЧИТЕ ВРСТЕ ПОДЛОГА

У овом раду, танак филм PbO, MnO₂, PbO + MnO $_2$ (нано слој) нанесен је на микроскопско стакло подлоге од индијум калајоксида и алуминијума дебљине 0,1 mm и 0,5 mm и узорци су припремљени техником термичког спреја. Облагање танких филмова на подлогу вршено је на температури од 35 °C, а затим су припремљени филмови жарени на 100 °C и 250 °C, током једног сата. Ефикасност заштите сваког танког филма на свакој подлози на различитим температурама анализирана је коришћењем радиоактивног извора⁶⁰Co и NaI(Tl) сцинтилационог детектора облика јаме. Добијени експериментални резултати сумирани су на следећи начин: додавањем танког филма PbO или MnO₂, заједно или одвојено на подлогу, повећава се слабљење гама зрачења у зависности од врсте узорака и температуре жарења. Минимална стопа гама редукције износила је 0.3 % за стакло од индијум калајоксида депоновано са MnO ₂и жарено на 250 °C, док је максимално слабљење од 9.6 % пронађено у алуминијуму дебљине 0,1 mm депонованим са PbO, или MnO ₂заједно. Утврђено је да се ефикасност заштите повећава са повећањем дебљине танког филма, температуре жарења и да зависи од врсте подлоге.

Кључне речи: зашишийа од гама зрачења, MnO₂, PbO сасийав, шермички сиреј, сциншилациони дешекшор