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Comparison between the optical properties of TiO₂ and ZnO thin films deposited using Dc plasma sputtering and pulsed laser deposition

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Abstract:

 TiO_2 and ZnO thin films were deposited using two different techniques, DC magnetron plasma sputtering and pulsed laser deposition, on glass substrates at different powers. The optical properties concerning the absorption, reflection and transmission spectra were studied for the deposited thin films. Noticeable enhancement for all optical properties was noticed for PLD thin films. The surface morphology of the deposits materials has been studied using atomic force microscope (AFM). The average surface roughness increasing from 3 by sputtering to 7 by PLD for TiO₂ thin films and from 1 by sputtering to 9 by PLD for ZnO thin films.

Key words: optical films, Zinc oxide, Titanium Oxide, plasma deposition. Laser deposition Received; 9 Feb. 2018, Revised form; 7 Mar. 2018, Accepted; 7 Mar. 2018, Available online 1 Apr. 2018

1. Introduction

Oxide thin films play a significant role in various optoelectronic devices such as solar cells, displays, touch screens, light emitting diodes, and etc [1-3]. Because of their high conductivity and high transmission in the visible region. Normally very high surface area of the metal oxides improves the efficiency of the optical devices. However, the search for new semiconducting materials to act as efficient light absorbers, has led to alternative materials being considered [4-6]. Now days TiO₂ and ZnO has been the most common materials for this job. Selecting of materials with the correct energetic and the morphology of the metal oxide particles plays an important role in these devices. The ability to manipulate the shape, size, and surface to volume ratio of these oxides is critical in influencing the materials. Also compared to TiO₂, ZnO is supposed to have higher efficiency in the photocatalytic performance in many cases due to its higher quantum efficiency [7,8]. Considerable attempts have been applied to extend the photoresponse of ZnO and TiO_2 to visible light region [8,9].

Many methods to produce oxides particles and thin films with different morphologies have been introduced. Such as atomic layer deposition (ALD) techniques [10], spray pyrolysis [11]. DC and RF magnetron sputtering and pulsed laser deposition [12-14]. Sputtering and deposition processing can be used for controlled tuning of the desired structure that may exhibit interesting properties with strong potential applications [15]. The aim of this work is the preparation and characterization of anti-reflection coating from oxides thin films. We are aiming to improve the transmission of visible light through TiO_2 and ZnOthin films using two different deposition techniques, by controlling the preparation conditions, such as time and power

2. Experimental:

Titanium Dioxide (TiO₂) and Zinc Oxide (ZnO) were purchased from FLUKA Chemical company. The TiO₂ particle size is 20 nm with 99% purity, 4.26 gm/cm³ density and molar weight 79.87 gm/mole. The ZnO particle size is 5µm, 98% purity, 5.6 gm/cm³ density and molar weight 81.3 gm/mole. The powders of Titanium dioxide (TiO₂) and Zinc Oxide (ZnO) are pressed under 10 MB for ZnO powder and 5MB for TiO₂ powder in the form of disk respectively. The dimensions of the target were 5 cm diameter and 0.5 cm thickness fore sputtering system and 3 cm diameter. The films were deposited in a DC magnetron sputtering system of 40 watt and $5x10^{-2}$ m torr vacuum pressure for different deposition times, 10, 30 and 40 mints. Neodymium: YAG laser model (Huafei Tongda Technology-DIAMOND-288 pattern EPLS) with 1064 nm wavelength, 10 ns pulse width and repetition frequency 1-6 Hz, was used for deposition of the thin films at 50, 75 and 100 mJ laser power. For observing the roughness and topography of deposited thin films, Atomic Force Microscopy (AFM) micrographs are taken with a (Digital Instruments, CSPM-AA3000). Typical data has been taken from AFM height images including roughness and average grain size of all thin films. A double -beam

UV/VIS Spectrophotometer (CECIL 2700) is used to measure the optical properties of thin films in the range (190-1100) nm. The background correction is taken for each scan.

3. Results and discussion

The used glass substrate was firstly characterized by spectrophotometer and the variation of transmittance with wavelength was represented in figure 1. At low wavelength in the UV region, the transmittance increases sharply with increasing wavelength up to 400 nm. Then reaching to saturation values, about 0.84 for all visible light spectrum [16, 17]. Figure 2 shows the transmittance and reflectance spectrum for TiO_2 thin films deposited on glass substrate by sputtering method at different sputtering times. The spectrum reveals that the films show more absorption in ultraviolet region and the transmission is gradually **increase** with increasing wave length, reaching the saturation values in the visible region. On the other hand, the transmission decreases by increasing sputtering time. And the optimum values of the transmission were recorded for 10 min sputtering time is about 0.9, which is larger than the substrate transmittance by 0.06 %.



Fig (1): Transmittance spectrum of glass substrate.



Fig (2): A, transmittance B, reflectance of TiO₂ thin films by sputtering deposition at different times

Figure 3 shows the optical properties of ZnO thin films deposited on glass substrate by sputtering method at different times. The optical characteristics, transmission, absorption and reflection are similar to the substrate behaviors. In the visible region, all the optical parameters are constant with increasing sputtering times. The maximum transmittance was 0.88. Which is larger than the substrate transmittance by 0.04. the low values of optical parameters for films prepared by DC plasma sputtering could be attributed to the low of deposition power of the current system [18,19].

This behavior can be explained as flows: Practically, ZnO and TiO₂ thin films have a wide optical band gap about 3.3 eV and 3.05 eV respectively, which is compatible with short wavelength and UV band [20]. So, at short wave length, interaction between incident photon and material will occur, and films will absorb photons with band gab energy. When the wavelength increases, the incident photon does not have enough energy to interact with atoms, thus the photon will transmitted. The increase in the absorbance with increasing deposition times could be attributed to the increasing of thin film thickness and surface roughness.



Fig (3): A, transmittance B, reflectance of Zno thin films by sputtering deposition at different times

AFM images of TiO₂ and ZnO films are shown in Figures 4 and 5 respectively. The rms roughness and average grain size are extracted from AFM data and listed in Table 1 for both of TiO₂ and ZnO films. All films surfaces are inhomogeneous and very thick. It can be clearly seen from the figures 4 and 5, that grains size finally, increases with increasing deposition time. This attributed to the two types of formation, the first one is the

increases of supporting beam ablation with large grains and the second, may attributed to possibility some of small grains agglomerated to form greater grains [17]. This will lead to increase in the surface roughness and grain size diameter, which in turn leads to decrease in reflectance. Surface roughness is important factor and it inversely proportional to reflectance of the surface. Glass substrate have 0.48 roughness, then high reflectance [21, 22].

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Sputtering							
	ZnO		TiO_2				
Pulse time Min.	Average grain size [nm]	RMS roughness [nm]	Average grain size [nm]	RMS roughness [nm]			
10	1.2	0.585	3.65	2.36			
20	1.5	0.8	5.73	2.79			
40	3	1.6	6.14	3.77			

Table (1): average grain size and rms roughness for ZnO and TiO₂ thin films by sputtering deposition



Fig (4): AFM images for TiO₂ thin films by sputtering deposition at 10 and 40 min



Fig (5): AFM images for ZnO thin films by sputtering deposition at 10 and 40 min

According to AFM results, ZnO thin film recorded very low particle size and rms roughness comparing by TiO_2 parameters, as shown in table (4-1). The rms roughness of ZnO ranging from 0.58 to 1.6 at different deposition time. These values are close to the glass substrate roughness. In addition to the small particle size, which will expect to reduce the film thickness.

On the other hand, pulsed laser deposition technique was used to re prepare the same oxides thin films at 50, 75 and 100 mJ power.



Fig (6): A, transmittance B, reflectance of TiO_2 thin films by PLD at different laser powers

Figure 6 shows the transmittance and reflectance spectrum for TiO₂ thin at different deposition pulsed energies. The absorbance and reflectivity are softly decrease by increasing wavelength, reaching the saturation near to the infrared region. However, the films show more absorption in ultraviolet region. The transmission is gradually increased with increasing wavelength, but, decreases by increasing PLD powers. The optimum values of the transmission were recorded for 50 mJ was about 0.985. Comparing by TiO_2 thin films prepared by sputtering method, the PLD thin films recorded desirable optical characteristics. For examples at 750 nm wavelength, which is the threshold of saturation in the optical parameters. PLD TiO₂ thin films recorded 0.985 transmittance and 0.011 reflectance at 50 mJ power. But sputtering TiO₂ thin films recorded 0.9 transmittance and 0.087 reflectance at the same laser power.

These values reflect improvements in the film transmission by about 0.16% higher than the glass

substrate and 0.094% higher than the plasma thin film. The improvement in the transmittance could be attributed to the increasing in the film's surface roughness, leading to the reduction in the reflectance.

Figure 7 shows the optical properties of ZnO thin films at different laser power energy. By using PLD technique, ZnO thin films are completely different from plasma sputtering thin films in values and behaviors. The absorbance is gradually decreased with increasing wavelength as a result of thick thin film formation. The transmittance is improved by about 0.16 % higher than the sputtering one.

The change in the optical properties with deposition power could be explained according to the AFM characterization for the two films as shown in figures 8 and 9 for TiO₂ and ZnO respectively . The rms roughness and average grain size are extracted from AFM data and listed in Table 2 for both of TiO₂ and ZnO films.



Fig (7): A, transmittance B, reflectance of Zno thin films by PLD at different laser powers

PLD							
	TiO ₂		ZnO				
Pulse energy [mJ]	Average grain size [nm]	RMS roughness [nm]	Average grain size [nm]	RMS roughness [nm]			
50	6.52	5.3	9.5	5			
75	8	7.6	11	6.28			
100	31.5	8.69	24.3	13			

Table (2): average grain size and rms roughness for ZnO and TiO₂ thin films by PLD

It can be clearly seen from the figures that grains size increases with increasing laser power and the films surface becomes more homogeneous at high laser power. The average grain size and rms roughness are higher than the values which recorded by sputtering for the two thin films. These values clearly explain the enhancement of the transmittance of PLD films as a result of a decreasing in the reflectivity parameters of the thin films. Which attributed to the increasing in average surface roughness from 3 by sputtering to 7 by PLD for TiO_2 thin films and from 1 sputtering to 9 by PLD for ZnO thin films.



Fig (8): AFM images for TiO₂ thin films by PLD at 50 and 100 mJ power



Fig (9): AFM images for ZnO thin films by PLD at 50 and 100 mJ power

4. Conclusion

We have successfully deposited TiO_2 and ZnO thin films by DC magnetron sputtering and pulsed laser deposition. The result was clearly that the film's color, surface morphology and optical properties could be directly controlled by deposition times and laser pulse power. The surface roughness was increased supplement with larger grain sizes of the films due to energy of atom enhancement. The UV-VIS spectroscopy showed that the PLD thin films were highly transparent. The transmittance is improved by about 0.16 % higher than the sputtering deposition one.

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