Pulse Energy Effect on the Optical Properties of Pulse Laser Deposited SiO₂ Thin Films

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ABSTRACT
In this work the effect of laser pulse energy on the optical properties of five samples of SiO₂ thin film deposited using pulse laser deposition technique was studied. Pulse energies of 100, 150, 180, 200 and 250 mj with fixed pulse repetition rate and number of pulses of 2 Hz, and 10 pulses, were used. The target to substrate distance and angle were fixed. The film thickness was measured by FESEM, and the transmission spectrum at certain wavelengths for each film was recorded. SiO₂ thin films transmission data and the measured film thicknesses were used to deduce their optical properties. The results showed that increasing the pulse energy results in an increase of the film thickness and the morphology of the films becomes more dense and non-smooth at higher pulse energy, also the optical properties showed to be affected by the thickness variation and hence by the pulse energy.

Keyword: PLD, Laser energy, SiO₂ thin films, Optical properties

I. INTRODUCTION
The deposition of thin films with adequate morphology and a crystalline structure is a key point in the development of many research fields [1]. To control the properties of the deposit film the deposition parameters needs to be adjusted and optimized. In thin films prepared by pulsed laser deposition (PLD) technique a variety of deposition parameters corresponds to the laser properties such as laser pulse energy, pulse repetition rate, etc., in addition to the rest of the deposition conditions such as target-substrate distance and angle, substrate temperature, ambient gas and pressure are all fundamental deposition parameters that determine the properties of the fabricated film [2]. Pulsed laser deposition has been successfully used to deposit an extraordinarily wide range of materials [3]. The PLD is a widely used technique for the fabrication of thin films because of its numerous advantages, such as its simplicity, etc. [4]. It becomes a promising technique for the production of oxide thin films and new materials with complex stoichiometry and multilayered structures [5]. The PLD enables the deposition of many complex materials over a wide range of background gas compositions and pressures. Extensive studies of the correlation between film structure and deposition parameters have been carried out over the past five decades [6]. Silicon dioxide is one of the most applied low-index materials in optical interference coatings [7]. Thin SiO₂ films deposited by pulsed laser deposition (PLD) are widely studied for application in microelectronics such as gate oxide films in semiconductor devices. In semiconductor industry, very thin continuous films of typically less than 4 nm are required. For that reason, the growth during the early stages of deposition is critical for applications in semiconductor devices. In optical applications conversely, nucleation plays a minor role, since rather thick layers are needed (up to several hundred nm). There is an evident lack of investigation on the optical properties such as the transmittance [8]. Before applying silicon dioxide films in multilayer stacks, fundamental questions on optical film quality have to be investigated.

In this work five samples of SiO₂ thin film were
fabricated using pulsed laser deposition using Q-switched Nd:YAG with the wavelength of 1064 nm, 10 pulses with pulse repetition rate of 2 Hz, the target to substrate distance and angle were fixed to 2 cm and 45°, respectively. Varied pulse energy of 100, 150, 180, 200 and 250 mJ were used. The film thickness was measured by Field Emission scanning Electron Microscope (FESEM) measurement tool, and the transmission spectrum at certain wavelengths for each film was recorded. SiO₂ thin films transmission data and the measured film thicknesses were used to calculate their optical properties.

II. MATERIALS, TOOLS AND METHODS:

A. MATERIALS:
The material used in this work were silicon dioxide (SiO₂) of 99.9% purity and refractive index of 1.46 it was prepared in disc form by mixing 50:50 ratio of the SiO₂ powder and Potassium bromide (IR spectroscopy grade).

B. TOOLS AND EQUIPMENTS:
Different tools and equipments were used to complete this work. They are described below with their specifications and needs:

1. Pressing machine:
   A hand press machine manufactured by Shimadzu (Japan) was used in this work [10]. The machine is used to press the SiO₂ powder in disc form (after mixing with Potassium Bromide) for target ablation.

2. Q-Switched Nd: YAG Laser:
   Q-switched Nd: YAG model OW D1 [11] was used to deposit SiO₂ thin film.

3. Scanning Electron Microscope (FESEM):
   Scanning electron microscopy type TESCAN MIRA 3 was used to measure the films thickness. MIRA3 is a high performance SEM system which gives high resolution and low-noise imaging [12]. MIRA3 offers all the advantages that come with the latest technologies and developments in SEM; delivering faster image acquisition, an ultra-fast scanning system, dynamic and static compensation and built-in scripting for user-defined applications [13].

4. DIFFERENT LIGHT SOURCES:
   Different monochromatic light sources were used to record the transmission spectra for the fabricated films [13-15].

Table 1: the monochromatic light sources used in this work

| Light Source                        | Wavelength (nm) |
|------------------------------------|-----------------|
| Diode Laser                        | 532             |
| He-Ne Laser                        | 632.8           |
| Diode laser                        | 660             |
| Omega XP Laser (red probe)         | 675             |
| Omega XP Laser (IR 820 nm probe)   | 820             |
| Omega XP Laser (IR 915 nm probe)   | 915             |

5. THE PHOTODETECTOR:
   A silicon pin photodiode was used in this work for detecting the transmitted intensity of each light source after passed through the fabricated SiO₂ films.

C. THE METHOD:
The procedure to fabricate SiO₂ thin films and study the effect of the pulse energy on their properties was done as follows:

- The experimental setup used to produce SiO₂ thin films was arranged as shown in figure (1).
- Different disks of SiO₂ (as targets) were prepared by the press machine.

![Figure 1: Schematic diagram of the experimental setup for fabrication of SiO₂ thin films](image-url)
➢ The distance and the angle between the target and the glass substrate were fixed to 2 cm, and 45°, respectively.
➢ The glass substrates were cut into the dimensions suitable for SEM imaging 2X2 cm, and then washed with distilled water and cleaned with alcohol.
➢ The Q-Switched Nd: YAG laser machine was switched on and 10 pulses with energy of 100 mj and a R.R. of 2 Hz was used to deposit SiO₂ thin film on the glass substrate.
➢ The above step was repeated four times with varied pulse energy of 150, 180, 200 and 250 mj with fixed repetition rate and number of pulses.
➢ The fabricated SiO₂ thin films were examined using FESEM to measure their thicknesses.
➢ The relation between the laser pulse energy and the SiO₂ thin film thickness was plotted.
➢ The transmission spectra of the fabricated SiO₂ films were recorded using different monochromatic light sources.
➢ Thicknesses of the SiO₂ films and the transmission data were used to calculate the optical properties for each film.
➢ The refractive index of each thin film was calculated using the measured reflectivity R and the glass refractive index μ₀ according to [16,17]:

\[ \mu = \left( \frac{\mu_s [1 + \sqrt{R}]}{1 - \sqrt{R}} \right)^{1/2} \]  

(1)

\[ \mu_s = \frac{1}{T_s \left( \frac{1}{T_s^2} - 1 \right)^{1/2}} \]

where T_s represents the transmission of the glass substrate.
➢ The absorption coefficients were deduced from the measured value of reflectivity R, the transmittance T, refractive index μ_s, and thickness t according to [16,17]:

\[ \alpha = \frac{1}{t} \mu \left( \frac{1 - R}{T} \right)^2 \]  

(2)

III. RESULTS AND DISCUSSION:
The results presented here were composed of two parts: a- the influence of the pulse energy deposition parameter of the PLD on the thickness of the fabricated SiO₂ thin films and b- the effect of the pulse energy on the optical properties of the produced SiO₂ thin films.

A. Pulse energy effect on the thickness of the SiO₂ thin films:
The five samples of the SiO₂ thin films using 10 pulses with pulse repetition rate of 2 Hz and varying pulse energies of 100, 150, 180, 200 and 250 mj. Then the five deposited SiO₂ thin film samples were imaged using FESEM machine and their thicknesses were measured and tabulated in table (2) with the corresponding pulse energy used. Figure (2-1a) shows the morphology of the SiO₂ thin films deposited using laser pulse energy of 100 mj and figure (2-1b) shows the FESEM thickness measurement of the produced SiO₂ thin film.

Figure 2-1 a: SiO₂ thin film deposited on glass substrate with laser energy of 100 mj and repetition rate of 2 Hz
Figure 2-1 b: The thickness measurement of the SiO$_2$ thin film deposited on glass substrate with laser energy of 100 mj and repetition rate of 2 Hz

The FESEM image together with the thickness measurement shown in figures (2-1a, and b), respectively, illustrate that the SiO$_2$ thin film has a thickness of 0.39 µm and it’s clear that the fabricated film is dense and has smooth film morphology.

Figure 2-2 a: SiO$_2$ thin film deposited on glass substrate with laser energy of 250 mj and repetition rate of 2 Hz

Figure (2-2 a) shows the SiO$_2$ thin film that was achieved laser pulse energy of 250 mj, while the number of pulses, the pulse repetition rate and other deposition parameters were the same. The film thickness that results when the pulse energy was 250 mj is shown in figure (2-2 b).

Figure 2-2 b: The thickness measurement of SiO$_2$ thin film deposited on glass substrate with laser energy of 250 mj and repetition rate of 2 Hz

Also figure (2-2 a) proved this film is dense and with non-smooth morphology compared to the first film in figure (2-1 a) and this is due to the increasing the pulse energy.
Table 2: thicknesses of the five fabricated SiO$_2$ thin films versus laser pulse energy:

| Pulse energy in (mj) with R.R = 2 Hz | Samples codes | SiO$_2$ thin film thickness in (µm) |
|-------------------------------------|---------------|----------------------------------|
| 100                                 | $S_1$         | 0.39                             |
| 150                                 | $S_2$         | 0.52                             |
| 180                                 | $S_3$         | 0.58                             |
| 200                                 | $S_4$         | 0.62                             |
| 250                                 | $S_5$         | 0.71                             |

The relation between the SiO$_2$ thin film thickness and the pulse energy is plotted in figure 3 and the equation that best describe the deposited thin film thickness energy relation was obtained.

\[
\text{Thickness of SiO}_2\text{ thin films} = A + B_1E_P + B_2E_P^2
\]

Where $E_P$ is the energy of the pulse while $A$, $B_1$ and $B_2$ being constants and their values were shown in figure (3). Also it is clear that increasing the pulse energy results in an increment of the film thickness.

From figure 3 it was found that the thickness of the deposited SiO$_2$ thin films is dependent on the pulse energy used and the equation relate the film thickness and the laser pulse energy is:

\[
\text{Thickness} = AE_P^2 + BE_P + C \quad (3)
\]

B. THE OPTICAL PROPERTIES OF THE FABRICATED SiO$_2$ FILMS:

Optical measurement constitutes the most important means of determining the band structure of the materials [18]. Optical constants of thin films provide us with information concerning microscopic characteristics of the material and its determination is very important for using it in any of such devices [19]. The transmission spectra, refractive indices and absorption coefficients were obtained for each SiO$_2$ thin film. The transmission intensities of different monochromatic light sources were detected before and after deposition for the five SiO$_2$ thin films and the results of sample $S_1$ are tabulated in table (3):

Table 3: Intensities before and after the deposition of sample $S_1$ (of thickness =0.39 µm):

| Wavelength (nm) | Intensity before deposition $I_0 \pm 0.001$ (V) | Intensity after deposition $I \pm 0.001$ (V) |
|-----------------|---------------------------------------------|---------------------------------------------|
| 532             | 340                                         | 319                                         |
| 632.8           | 401                                         | 361                                         |
| 660             | 442                                         | 404                                         |
| 675             | 472                                         | 436                                         |
| 820             | 526                                         | 490                                         |
| 915             | 385                                         | 343                                         |
The data in table (3) above was used to calculate the T% \((T = I/I_0)\) at the given wavelengths for sample \(S_1\) and the same was done with the other four samples. The calculated values are plotted for the five samples \(S_1, S_2, S_3, S_4\) and \(S_5\) in figure (4) as a function of wavelength.

![Figure 4: transmission spectra of the five SiO\(_2\) thin film samples](image)

Figure 4 show that the thickness of the thin film affected its transmission, the large thickness of the thin film gives the lower transmission of the film. Regarding the thickness difference shown in figure 4, it can be said that the SiO\(_2\) thin films have a high transparency varied from (0.89-0.97) % in the investigated spectral range.

The calculated refractive indices for the five samples using equation (1) are plotted as a function of wavelengths as shown in figure (5).

![Figure 5: The refractive index of the five samples of SiO\(_2\) thin films versus wavelengths](image)
Figure 5 shows that sample S₁ which is of smallest thickness among the other has highest refractive indices from 532 to 660 nm. The refractive index of any material in thin film profile is usually deviates from that of the bulk of the same material. This is due to the void fraction typical of the thin film microstructure [20].

The absorption coefficients calculated for the deposited SiO₂ film samples using equation (2) are plotted versus wavelengths in figure (6).

![Absorption coefficients versus wavelengths for five samples of SiO₂](image)

Figures (5) and (6) support the idea of using such film as an optical filter or as a reflector in specific wavelengths deduced from the transmission spectrum. The refractive indices and absorption coefficients of the pulsed laser deposited SiO₂ thin films varies with thickness and for each thickness the transmission is unique, therefore these two optical properties are functions of the film thickness and therefore depend on the pulse energy used for the deposition of such films.

**IV. CONCLUSION:**

From the obtained results the followings can be concluded:

- SiO₂ thin films of different thicknesses and optical properties can be produced using pulsed laser deposition technique.
- Pulse energy of the laser source is an important parameter in fabrication of films prepared using PLD method.
- The thickness of the SiO₂ thin film can be controlled by controlling the pulse energy used in the deposition process.
- The SiO₂ can be used to produce optical components in the range from 532 nm to 915 nm by controlling its thickness.

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