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The Influence of the Number of Laser Pulses on the Thickness and Roughness of TiO_2 Thin Films Fabricated Using Pulsed Laser Deposition

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Abstract: In this work Titanium Dioxide thin films were fabricated by pulsed laser deposition technique (PLD) using a Q-switched Nd: YAG laser. Titanium dioxide powder, in the Anatase form, was compressed to form solid disks. Each of these disks was irradiated with different number of laser pulses (5, 10 and 15 pulses) with the same pulse energy (150 mJ) and same Repetition rate (10 Hz). The thickness and topography of each deposited thin films were characterized using atomic force microscopy (AFM). The results showed that the thickness of the film increase exponentially when the number of laser pulses increased. The results showed also that the average roughness (Ra) of the films and the root means square roughness (RMS) increased with increasing the number of pulses exponentially to specific value and then decreased exponentially in a behavior like the Gaussian shape.

Keywords: TiO_2 Thin Films, Pulsed Laser Deposition, Film Thickness, Roughness

1. Introduction

Titanium dioxide (TiO_2) has been proven to be an effective material for applications such as photocatalysis, dye sensitized solar cells, heterogeneous-catalysis, selfcleaning/ antifogging surface coatings [1-4]. etc. Titanium dioxide is a white inorganic solid substance, which is thermally stable, non-flammable, poorly soluble, cheap non-toxic material that has very good semiconducting properties. It is insensitive to visible light, because of its large band gap (3.2 eV) [5], which enables it to absorb only in the near ultraviolet region, it has high transmittance values in the visible region [6]. Titanium dioxide, also known as titanium (IV) oxide or Titania, is the naturally occurring oxide of titanium with the molecular formula TiO_2, and it belongs to the family of transition metal oxides. Due to high refractive index and light scattering, titanium dioxide has been used widely as a white pigment, it can be used also as to generate electricity or to degrade specific organic compounds. Titanium dioxide is found in four different polymorphs in nature: “Anatase”, “Brookite”, “Rutile”, and “TiO_2 (B)”. Additionally, several high pressure forms have been synthesized [7].

Titanium dioxide thin films and coatings are used today commercially to create functional surfaces for e.g. self-cleaning windows and construction parts, low-emission and solar control films on large area glass, anti-fogging mirrors and car windscreens. Furthermore, extensive research in how to apply and benefit from titanium dioxide antibacterial coatings is carried out [8]. It is also widely used in vast range of consumer goods and industrial such as treatment of various surfaces [9]. This generates a significant enthusiasm from scientists for various applications like: photocatalytic, solar cells, gas sensors, anti-reflect coating, and electrochromic systems [10-12]. TiO_2 nanoparticles are presumed to be bound with cotton through ester bonding over the non-homogeneous irregular structure of the cotton making it more self-cleaning, ultraviolet irradiation resistant and antibacterial in nature [13-15]. Several methods for synthesizing and depositing titanium dioxide thin films and coatings are available, the most common being chemical/ physical vapor deposition techniques (CVD/PVD) or sol-gel methods [8], e-beam evaporation, sputtering, spray, laser-deposition, and hydrothermal method [16-19]. In this work pulsed laser deposition (PLD) technique is used to fabricate thin films from TiO_2 material. Laser deposition is one of the physical vapor techniques, which is recognized as one of the simplest and most accurate and efficient methods in terms of quality, simplicity and low cost for the synthesis of thin films materials. PLD also represents a high energy process which provides well adherent thin films with good mechanical rigidity and surfaces with high specific surface area [20, 2]. In addition, PLD also offers advantages such as stoichiometrically transferring material from target to the substrate [20], capability of inert and reactive gas deposition, wide range of operational pressure and temperature, and variety in options for substrate materials. In this work pulsed laser deposition was used to fabricate thin films of pure TiO_2 and the influence of the number of laser pulses on the thickness and roughness of the fabricated films was investigated.

2. The Experimental Part

The schematic diagram of the experimental setup used in this work is shown in figure 1. Q switched Nd YAG laser with second harmonic generation crystal (λ =532 nm) was used for the fabrication of TiO_2 thin films. The target material was pure Titanium dioxide (Anatase) with grain size (10 nm) and purity (99.5%) supplied from (Nanjing Mission Advanced material company - republic of china). It was prepared by sintering pure TiO_2 powder with pressure up to (10000 Kg) with mini press machine supplied from Philips to form disks with diameter (2 cm) and thickness (0.4 cm). The target placed on adjustable stand in order to control the distance from target to substrate. The substrate was glass slides supplied from (Ningbo Greetmed medical instruments Co., Ltd, Hi-Tech zone people's - republic of china) cut in (1x1 cm^2). All the substrates were cleaned in pure acetone solution and triple rinsed with de-ionized water prior to the deposition and then mounted on the substrate holder which joined to rotating motor. The rotation speed was kept at (30 rpm). The incident laser beam maintained at (45°) angle to the target surface. Three series of TiO_2 thin films were grouped (A, B and C) corresponds to number of laser pulses (5, 10 and 15 pulses) with energy of (150 mJ) for each pulse and the same pulse repetition rate (10 Hz) applied for each of the three series. Deposition was carried out at atmosphere and room temperature.

After deposition, the surface topography of TiO_2 thin films was observed using atomic force microscopy (AFM) (SPA-400 microscope with etched silicon cantilever with spring constant of 42 N/m^{-1} and SPI 3800N controller, Seiko Instruments Inc., Japan) with a resolution of (0.01 nm) in the X-Y and Z directions. Images were taken under contact mode with a scan Rate (2.0 Hz). The thin films thickness and surface roughness were determined using software (Nano Navi Station version 5.60 A copy right 2005-2010, SII Nano Technology Inc).

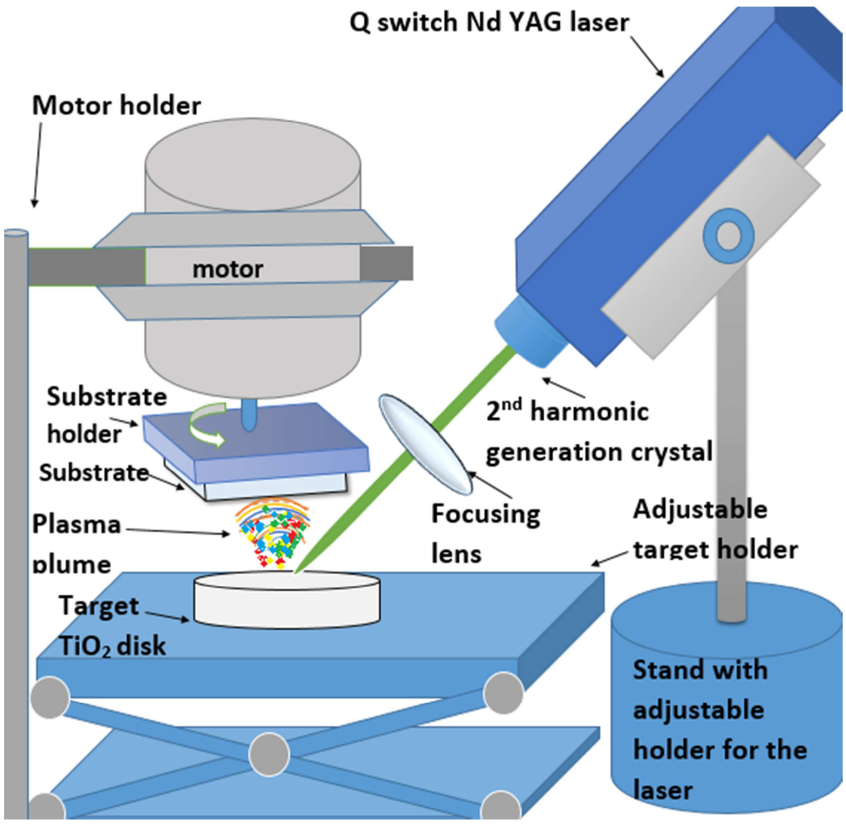

Figure 1. *Schematic diagram of the experimental setup of pulsed laser deposition.*

3. Results and Discussion

Figure 2 - A, B and C shows three dimensional and top view AFM images of three TiO_2 thin films deposited on glass substrates at room temperature and atmospheric pressure using pulsed laser deposition technique (PLD), each target was fired with (5, 10 and 15 pulses) of laser, successively. The pulse energy was (150 mJ) and the repetition rate was (10 Hz).

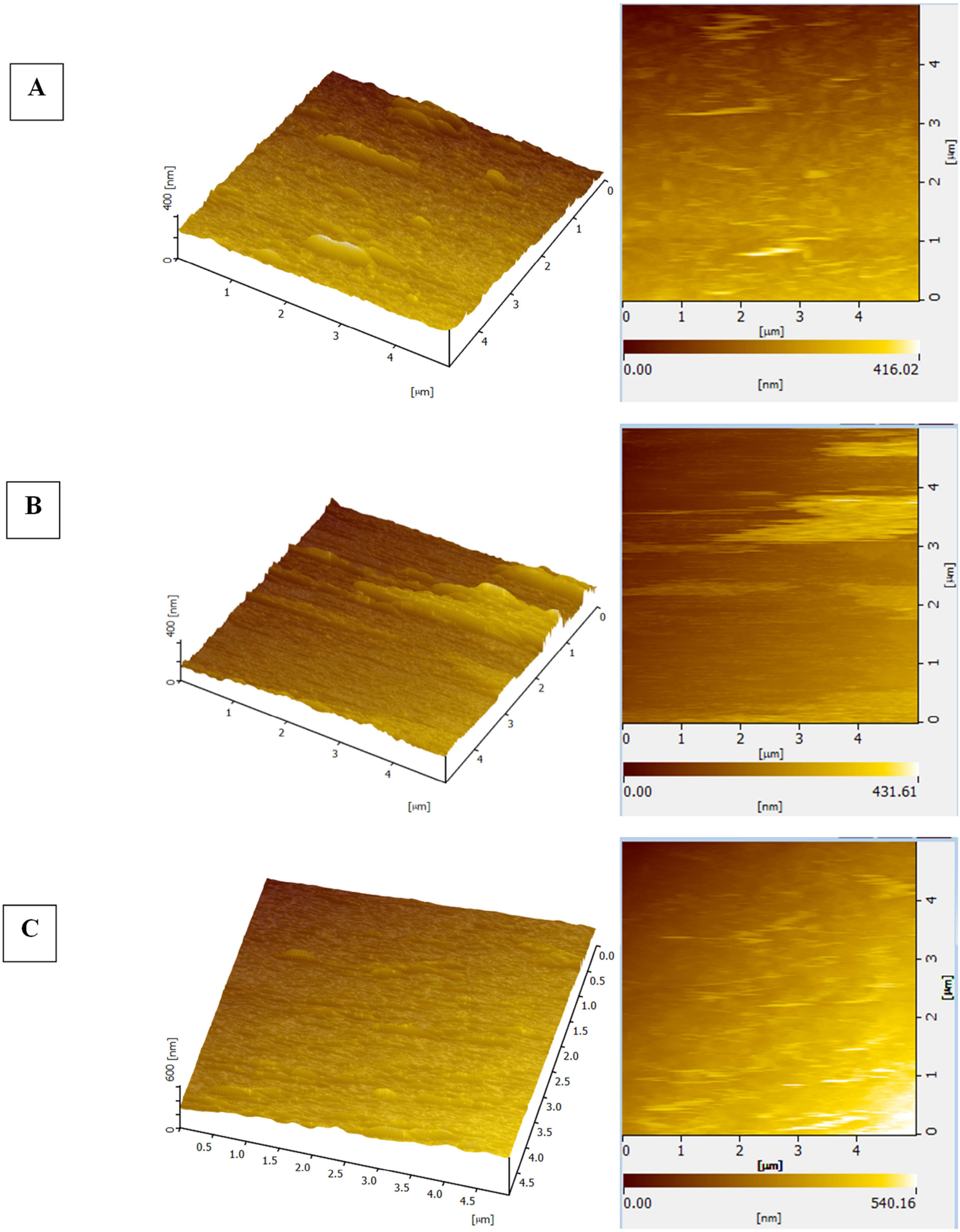

Figure 2. *Three dimensional and Top view AFM images of TiO_2 thin films fabricated using PLD with different number of laser pulses [A]: 5 pulses, [B]: 10 pulses and [C]: 15 pulses. each pulse with energy (150 mJ).*

Figure 3 depicts the influence of the number of laser pulses on the thickness of the films. The thickness growth exponentially proportional to the number of laser pulses. The growth of the films, as shown in the figure, at lower number of pulses increase rapidly as pulses increased till reaches specific thickness then the growth behavior become slow, which is due to that each laser pulse with specific energy directed onto the surface of a target, depositing energy by a combination of photothermal and photochemical processes. Give a sufficient intensity of incident laser light, a combination of thermal vaporization and electronic bond-breaking takes place, must leading to removal of specific (nm) thickness layers of material per a laser pulse and deposit this removed volume vapor on the substrate as a thin film, but after few first pulses plume of species created and the deposition rate drops and the relation between the deposited thin films and the number laser pulses will no longer remains linear, it behaves in a logarithmic manner that is due to all of the ablated material will not reach the substrate a portion will be reflected back onto the target surface due to collisions within the plume and between the plume and the shock-wave generated in the background gas where its pressure increase with the number of laser pulses and hence the deposition rate decrease with increasing number of pulses [22, 23]. Also due to the formation of vapor above the target after the first pulse, this vapor absorbs some of the next laser pulse energy and decrease the deposited thickness.

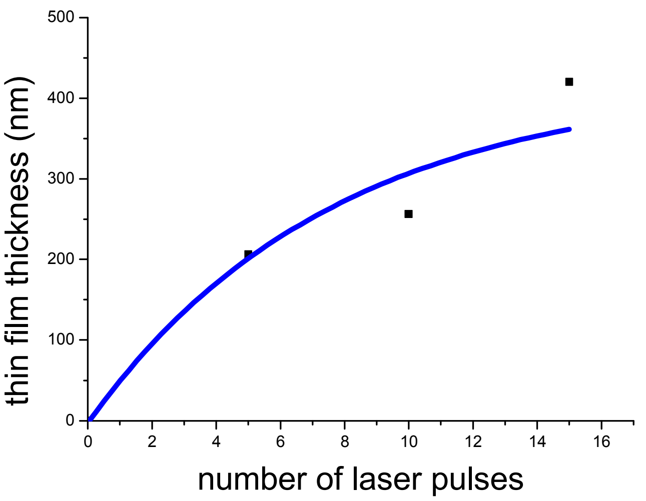

Figure 3. *The Thickness of TiO_2 thin films as a function of the number of laser pulses (5, 10 and 15 pulses) with the same pulse energy (150 mJ) and the same pulse repetition rate (10 Hz).*

Figure 4 illustrates the influence of the number of laser pulses on the average roughness (Ra) of the films. It is clear from the figure that the average roughness of the TiO_2 thin films strongly dependent on the number of laser pulses and the roughens increase exponentially till reaches the maximum value and then fall in exponential decay.

In the beginning of the deposition process the increase in roughness is due to the following possible reasons:

Target Erosion that due to the repeated ablation at the same target site leads to the formation of surface ripples and cones [24], that their formation may possibly due to the inhomogeneous etching resulted from the interference of the incident beam with waves scattered from defects. Once cones and ripples are formed, thermal and/or mechanical shock may eject droplets and/or solid particles from the sharp edges of the structure.

Clustering when vapor plume expands into a background oxygen gas, collisions may cause atoms and ions in the plume to coalesce into nm-scale clusters [25]. On the other hand, the roughness starts to decrease after reaching the maximum value, as increasing the number of laser pulses thermal energy of the vapor above the target will increase and hence the plume expands and compresses the background gas, creating a shock-wave which slows the plume expansion [23, 26]. As in this work where the target-to-substrate distance is too close, the expanding vapor reaches the substrate with typical energies of 0.1-100 eV per atom [27], depositing sub-monolayer thicknesses of material per pulse. The kinetic energy of the condensing vapor is transferred to the target surface, supplying thermal and kinetic energy to the deposition process. This energy encourages the formation of smooth, high-quality films by enhancing the mobility of the atoms on the surface of the film that give the adatoms more chance to grow together in ordered manner leading to formation of thin films with large crystallites and discouraging the growth of islands on the film surface [28]. Another factor reducing roughness is the low speed rotating substrate, as the number of pulses increase the substrate will rotate many times and thus the plume imping the substrate randomly at various positions that enhance filling of the empty channels between the islands even if the uncovered surface area of the channels is large, secondary nuclei are formed in the channels with the progress of pulses and substrate rotation and they grow coalescence and fill in as before and the film becomes continuous and smoother than before.

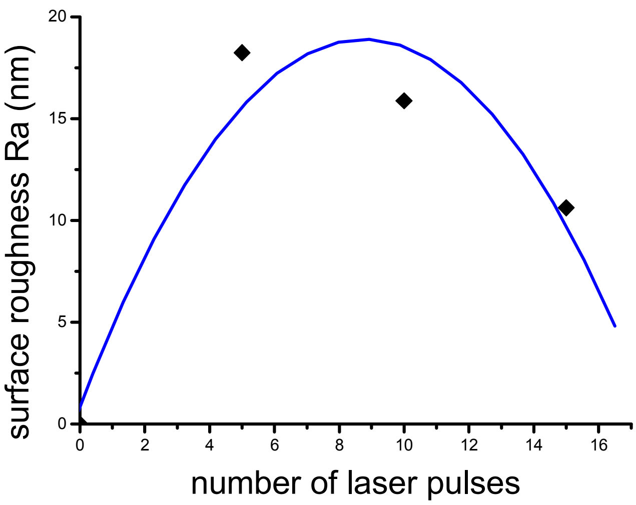

Figure 4. *Surface average roughness (Ra) of the TiO_2 thin films fabricated using different number of laser pulses (5, 10 and 15 pulses) with the same pulse energy (150 mJ) and the same pulse repetition rate (10 Hz).*

Figure 5 depicts the influence of the number of laser pulses on the root mean square roughness (RMS) of the films. The behavior of the curve is just like that in figure 4. The difference between (Ra) and (RMS) is that each of them is calculated differently, (Ra) is the average roughness of the surface measuring the microscopic peaks and valleys while (RMS) is the Root Mean Squire measuring the microscopic peaks and valleys.

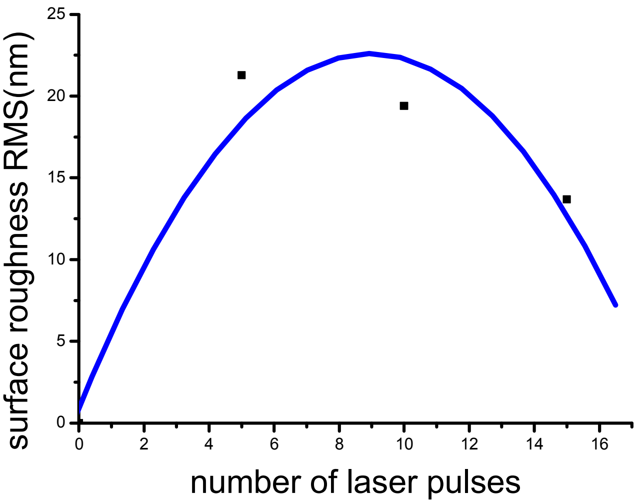

Figure 5. *The root mean square roughness (RMS) of the TiO_2 thin films fabricated using different number of laser pulses (5, 10 and 15 pulses) with the same pulse energy (150 mJ) and the same pulse repetition rate (10 Hz).*

4. Conclusions

This work demonstrated the possibility of fabricating Nano-structured TiO_2 thin films on glass substrates by pulsed laser deposition (PLD) at room temperature and atmospheric pressure. The fabricated films were characterized using atomic force microscopy (AFM) and the relations between the number laser pulses and film thickness and morphology were deduced. The results showed that the number of laser pulses has a significant influence on the thickness and the roughness of the fabricated films, three samples were deposited at different number of laser pulses while all other experimental conditions including pulse energy, pressure, time, and temperature were maintained constant and found that the thickness increase exponentially with the number of laser pulses starting with (5) pulses the film was about (200) nm and the experimental data were fitted in order to estimate the rate of deposition per pulse to fabricate films with specific thickness depending on the number of pulses used in applications like optical coatings, gas sensors, etc. on the other hand the average roughness and the root means square roughness (RMS) increased with increasing the number of pulses exponentially and reaches maxima and then decreased exponentially in a behavior like the Gaussian, as smoothness is an important factor in many application we recommend that using higher number of pulses to obtain high quality smooth films, in spite of some applications need thinner films that means low number of pulses needed, higher number of pulses used but with relative low energy for each pulse.

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