Oxygen Content-Dependent Surface Roughness of Nanostructured Silicon Dioxide Thin Films

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Abstract

In this work, nanostructured SiO_2 thin films were deposited on glass substrates using DC reactive magnetron sputtering technique. Gas mixtures of argon and oxygen at different mixing ratios were used to synthesize SiO_2 nanoparticles. Smoother surfaces were produced at intermediate gas ratios, with the lowest RMS roughness observed at an Ar:O_2 ratio of 70:30. The film deposited at a 50:50 ratio exhibited the highest absorbance in the visible range (400–600 nm), making it promising for optoelectronic applications. The results confirm that the Ar:O_2 gas ratio plays a critical role in tuning the structural and optical properties of SiO_2 nanostructured thin films.

Keywords: Silicon dioxide; Surface roughness; DC sputtering; Nanoparticles

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1. Introduction

Silicon dioxide (SiO_2), commonly known as silica, is a common oxide compound made up of silicon and oxygen atoms bonded together. Within the silica molecules, every silicon atom connects with four oxygen atoms while every oxygen atom connects with two silicon atoms by Si-O bonds [1-3]. This compound is prevalent in various crystalline forms, notably quartz [4,5], and is also found in amorphous forms such as glass [6,7]. The basic structural unit of the silicon dioxide is a tetrahedron formed by a silicon atom and four oxygen atoms. The silicon atom sits in the center of the structure and is chemically bonded to the oxygen atoms in the four corners of the tetrahedron [8]. The oxygen atom has two valence electrons, and thus it has a possibility to form a bond to the silicon atom of the neighboring tetrahedron. Since the oxide is amorphous, all of the oxygen atoms do not form bonds between the adjacent structural units. Consequently, depending on the bonding state of the oxygen atoms, they are referred to as bridging or non-bridging oxygen [9,10]. The amorphous structure forms a more open network and thus the density of the oxide is less than the crystalline form (i.e., the quartz) [11]. SiO_2 is characterized by a substantial band gap, this large band gap classifies SiO_2 as an insulator, making it an excellent electrical insulator in various applications [12-14]. SiO_2 nanoparticles are used in various fields, including chemical, biological, and medical applications, due to their chemical stability, optical transparency, low toxicity [15], with low refractive index [16]. It serves as a fundamental component in microelectronics [17], solar cells [18], and protective coatings [19]. SiO_2 films are widely used as low-refractive-index layers in multilayer optical devices [20,21], as passivation and protective coatings for silicon devices [22], and as scratch-resistant coatings for plastic ophthalmic lenses [23], and so on. The physical and optical properties of SiO_2 layer depend on the method of deposition [24-28]. In reactive sputtering used to prepare nanostructured SiO_2 thin films, compound thin films are deposited in the presence of a reactive gas [29,30]. The reactive gas reacts with the sputtered material and forms a compound. This process makes it possible to deposit a wide variety of compounds (oxides, nitrides, carbides, etc.) with a wide range of properties [31,32]. The ability to deposit high-quality SiO_2 thin films with controlled thickness and structural properties is crucial for enhancing the performance of advanced technologies [33]. The key parameters influencing the film quality include oxygen partial pressure, DC power, working gas pressure, and substrate temperature, which must be optimized to achieve the desired structural, optical, and electrical properties [34-36].

The aim of this research is to introduce the effect of oxygen gas in the gas mixture of a dc reactive sputtering technique on the surface roughness of silicon dioxide thin films.

2. Experimental work

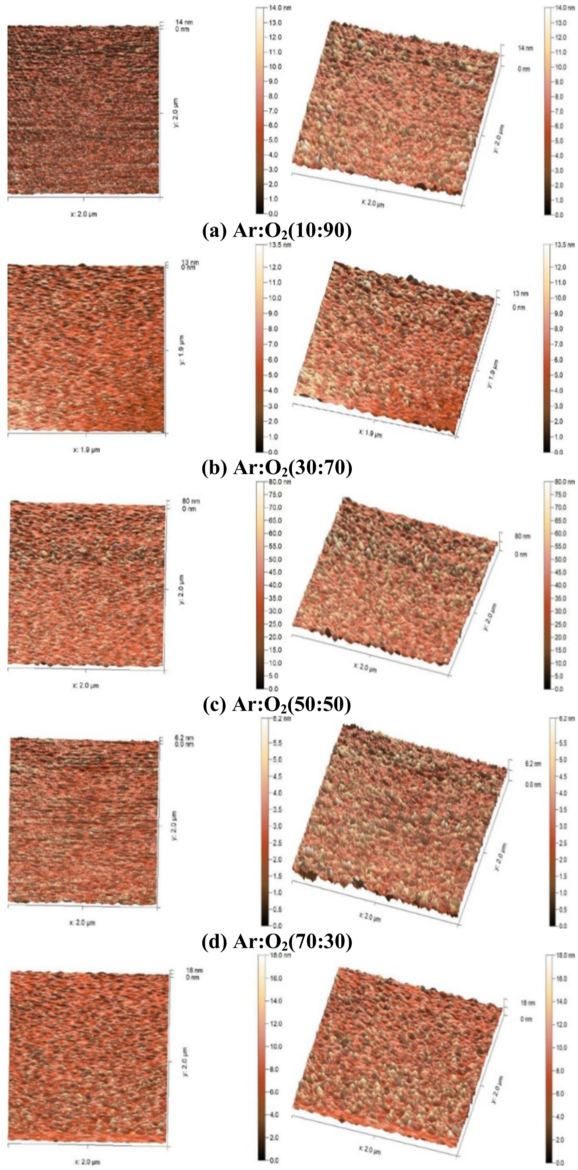
A reactive magnetron sputtering system was utilized to deposit SiO_2 thin films onto glass substrates. Initially, a base pressure of approximately 0.15 mbar was attained using a two-stage Leybold-Heraeuos rotary pump (24 m^3/h). Prior to the deposition process, both the targets and the glass substrates were thoroughly cleaned and dried. The 99.9% pure silicon target was meticulously secured on the cathode. Plasma essential for sputtering was generated via the electrical discharge of argon, powered by a high-voltage DC supply. The discharge current was kept constant at 35 mA, an inter-electrode distance of 4 cm, and deposition time of 60 min. The Ar:O_2 gas mixing ratios were (30:70), (70:30), (50:50), (10:90), and (90:10).

3. Results and discussion

Figure (1) shows the 2D and 3D atomic force microscopy (AFM) images for the nanostructured SiO_2 thin films prepared using different mixing ratios of Ar:O_2 gases after deposition time of one hour, while table (1) shows the RMS roughness of nanostructured SiO_2 thin films prepared at different Ar:O_2 gas mixing ratios. The AFM images offer a detailed visual representation of the surface morphology of prepared SiO_2 thin films. A comprehensive analysis of these images, coupled with the RMS roughness data, reveals how the Ar:O_2 gas mixing ratio critically influences the nanoscale features, uniformity, and overall smoothness of the deposited SiO_2 films. Clearly, all images display a granular or textured surface, characteristic of thin films grown by sputtering. However, significant differences in grain size, uniformity, and peak-to-valley variations are evident. The accompanying color scales (representing height) are crucial for interpreting the vertical topography. For example, images (a) and (e) have relatively narrow height scales (around 13.5 nm and 6.2 nm, respectively), indicating smoother surfaces with smaller height variations. In contrast, image (c) displays a much larger height scale (up to 80 nm), immediately suggesting a significantly rougher surface.

The sample prepared using 10:90 mixture corresponds to an oxygen-rich environment. The surface appears relatively uniform with small, densely packed grains. The height scale is 13.5 nm, indicating modest vertical variations. According to table (1), the RMS roughness for this sample is 1.31 nm. This low roughness value confirms the visual impression of a smooth and uniform surface, consistent with the manuscript's claim that films prepared with higher oxygen content exhibit low surface roughness. The enhanced crystallinity observed at higher oxygen ratios in XRD analysis might contribute to the formation of more organized, albeit small, grains, leading to a smoother film.

The sample prepared using 30:70 mixture also represents an oxygen-rich condition, though with slightly less oxygen than the 10:90 ratio. The visual appearance is still quite smooth, with a similar granular texture to the 10:90 sample. The height scale for this image is 14 nm, slightly wider than the 10:90 ratio. Table 1 reports an RMS roughness of 1.76 nm for this sample. While slightly higher than the 10:90 ratio, it still falls within the low roughness range, further supporting the idea that higher oxygen content promotes smooth and uniform surfaces. The slight increase in roughness compared to 10:90 might suggest a subtle decrease in the completeness of oxidation or a slight increase in grain size.



(a) Ar:O_2(10:90)

(b) Ar:O_2(30:70)

(c) Ar:O_2(50:50)

(d) Ar:O_2(70:30)

(e) Ar:O_2(90:100)

Fig. (1) 2D and 3D AFM images of SiO_2 thin films prepared at different Ar:O_2 mixing ratios

Table (1) The RMS roughness of nanostructured SiO_2 thin films prepared at different Ar:O_2 gas mixing ratios.

Ar:O_2 gas mixing ratios	RMS roughness (nm)
10:90	1.31
30:70	1.76
50:50	8.32
70:30	0.71
90:10	2.11

The sample prepared using 50:50 mixture corresponds to an intermediate gas mixing ratio. Visually, this image stands out dramatically from the previous two. The surface appears much rougher, with larger and more distinct granular features. The height scale for this image is a substantial 80 nm, indicating very significant peak-to-valley variations. This visual observation is quantitatively supported by table (1), which shows the highest RMS roughness of 8.32 nm for this sample. This high roughness may be attributed to more advanced grain growth.

The sample prepared using 70:30 mixture represents another argon-rich environment, but with a higher oxygen content than the 90:10 ratio. Visually, this image appears to be the smoothest among all samples. The surface is remarkably uniform with very fine, closely packed features. The height scale is significantly compressed at only 6.2 nm, which is the smallest range among all images. Quantitatively, Table (1) confirms this observation, reporting an RMS roughness of 0.71 nm, which is the lowest among all samples. The manuscript states that this film "showed the smoothest surface... making it ideal for high-quality applications". This finding is consistent across the AFM image and the quantitative data, indicating that a particular balance between argon and oxygen (even in an argon-dominant environment, but with sufficient oxygen) can lead to the smoothest film deposition, possibly due to optimized sputtering and oxidation rates allowing for dense and uniform film growth.

The sample prepared using 90:10 mixture represents an argon-rich environment with very low oxygen content. The surface shows a moderately rough appearance, with discernible grains that are larger than those in the oxygen-rich samples. The height scale for this image is 18 nm. Table (1) confirms this, with an RMS roughness of 2.11 nm. This is consistent with the increasing argon ratio that led to higher surface roughness due to reduced oxidation and less dense film growth. The low oxygen content would likely result in incomplete oxidation of the silicon target, leading to silicon-rich oxide films or even unoxidized silicon, which can contribute to rougher and less uniform film growth.

The AFM images, when analyzed in conjunction with the RMS roughness data, strongly demonstrate the critical influence of the Ar:O_2 gas mixing ratio on the surface morphology of SiO_2 thin films. High oxygen content (10:90, 30:70) generally promotes smoother surfaces, consistent with enhanced oxidation and more uniform film growth. However, there appears to be an optimal ratio, as the 70:30 mixture surprisingly yields the absolute smoothest surface, even smoother than the higher oxygen content ratios. The 50:50 ratio results in the roughest surface based on AFM, indicating significant grain growth or surface irregularity. Argon-rich environments (90:10) tend to produce rougher films due to less efficient oxidation. These findings underscore the complex interplay between plasma chemistry, deposition kinetics, and surface adatom mobility, all of which are modulated by the Ar:O_2 ratio, ultimately dictating the nanoscale topography of the resulting SiO_2 thin films. The detailed AFM analysis provides crucial insights into optimizing deposition parameters for specific application requirements where surface smoothness is paramount.

4. Conclusions

DC reactive magnetron sputtering technique was used as a reliable and flexible technique to prepare SiO_2 thin films. The results indicate that the structural and morphological quality of SiO_2 thin films is highly sensitive to the reactive gas environment during reactive sputtering. Controlled oxygen incorporation enhances film crystallinity, reduces surface roughness, and improves chemical stoichiometry, thereby enabling the fabrication of high-purity, homogeneous SiO_2 nanostructures suitable for advanced optical and electronic applications.

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