

Surface Morphology and Elemental Analysis of Al_2O_3 Thin Films Prepared by DC Reactive Sputtering Technique Using Different Gas Mixtures

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Abstract

In this study, Al_2O_3 thin films were prepared by dc reactive sputtering technique using different gas mixtures of argon and oxygen gases (90:10, 70:30, 50:50, 30:70, and 10:90). These films were characterized to introduce their surface morphology and elemental composition as functions of the oxygen content in the gas mixture. The gas mixing ratio plays a crucial role in controlling the nanoscale morphology of the prepared thin films. The $[\text{Al}]/[\text{O}]$ ratio varies non-linearly with the $\text{Ar}:\text{O}_2$ mixing ratio. Increasing the oxygen content leads to a progressive decrease in surface roughness, resulting in smoother and more uniform films with finer granular features. These results presented herein are useful to optimize the sputtering process to achieve desired surface properties for various applications, as surface roughness profoundly impacts adhesion, optical properties, and device performance.

Keywords: Aluminum oxide; DC sputtering; Surface morphology; Elemental analysis

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1. Introduction

The gas mixing ratio in DC reactive sputtering is a critical parameter that profoundly influences the structural characteristics of nanostructured metal oxide thin films, dictating properties such as crystallinity, stoichiometry, grain size, morphology, and defect density. In DC reactive sputtering, a pure metal target is sputtered in a reactive gas atmosphere, typically a mixture of an inert gas (e.g., Ar) and a reactive gas (e.g., O_2 for oxide films). The ratio of these gases directly impacts the plasma chemistry, the sputtering yield of the target, and the reaction pathways at the target surface and on the growing film.

At low reactive gas flow rates (i.e., high Ar-to-reactive gas ratio), the sputtering process operates in a metallic mode. Here, the target surface remains largely metallic, leading to a high sputtering rate of metal atoms. The reactive gas primarily reacts with the sputtered metal atoms in the plasma phase or on the substrate surface. This often results in films with a high density, good crystallinity, and a relatively stoichiometric composition, as there are ample metal atoms available for reaction. However, insufficient reactive gas can lead to oxygen vacancies or a sub-stoichiometric oxide phase, compromising the desired functional properties. For instance, in the context of transparent conducting oxides, oxygen vacancies can

act as charge carriers, but an excess can lead to reduced transparency or stability.

As the reactive gas flow rate increases, a transition from the "metallic mode" to the "transition mode" and eventually to the "poisoned mode" occurs. In the transition mode, the target surface starts to become partially covered by the reactive gas, forming a thin compound layer (e.g., metal oxide). This surface poisoning reduces the sputtering yield of metal atoms because the compound layer has a lower sputtering rate than the pure metal. The film growth rate decreases, and the plasma characteristics change due to altered secondary electron emission from the poisoned target. This region is often characterized by unstable sputtering conditions, making it challenging to achieve reproducible film properties. However, some researchers deliberately operate in this region to fine-tune specific structural properties, as it offers a balance between metallic and fully oxidized target states.

In the "poisoned mode" (high reactive gas flow rate, low Ar-to-reactive gas ratio), the target surface is fully covered by the reactive compound. The sputtering yield is significantly reduced, leading to much slower deposition rates. While this mode ensures a highly stoichiometric and fully oxidized film, it can also lead to increased porosity, reduced film density, and a decrease in crystallinity due to limited atomic mobility on the growing surface at lower deposition rates.

Excessive reactive gas can also lead to the formation of undesirable phases or increased impurity incorporation. Furthermore, ion bombardment effects from the plasma can be altered; with more reactive gas, the plasma potential might shift, affecting the energy of ions impinging on the substrate, which in turn influences film densification, stress, and crystallographic orientation.

The gas mixing ratio critically influences the mean free path of sputtered species and the reactive gas molecules, affecting collision events in the plasma. This, in turn, impacts the energy and angular distribution of species arriving at the substrate, playing a significant role in determining the film's morphology and texture. For example, a higher reactive gas partial pressure can increase gas-phase scattering, reducing the kinetic energy of arriving species and potentially leading to more porous films. Conversely, optimal kinetic energy can promote surface diffusion and denser film growth. Therefore, precise control over the gas mixing ratio is paramount for tailoring the desired structural characteristics, optimizing the physical and chemical properties, and ensuring the reproducible fabrication of high-performance nanostructured metal oxide thin films for diverse applications in electronics, optics, and sensors.

The preparation of Al_2O_3 thin films can be achieved through a diverse array of deposition techniques, each offering distinct advantages in terms of film quality, thickness control, stoichiometry, and deposition rate. Common physical vapor deposition (PVD) methods include sputtering (DC, RF, reactive sputtering) and pulsed-laser deposition (PLD), which involve bombarding an Al target with energetic ions in an oxygen atmosphere or ablating an Al_2O_3 target, respectively. Chemical vapor deposition (CVD) techniques, such as atomic layer deposition (ALD) and plasma-enhanced CVD (PECVD), utilize gaseous precursors that react on the substrate surface to form the film. ALD, in particular, is highly favored for its ability to produce ultrathin, highly conformal, and pinhole-free films with atomic-level thickness control, owing to its self-limiting growth mechanism. Solution-based methods like sol-gel deposition also offer a low-cost alternative for preparing Al_2O_3 films, although often with less precise thickness control and potentially higher impurity levels. The choice of deposition technique is often dictated by the desired film properties, application requirements, and economic considerations.

The aim of this work is to introduce the effects of partial gas content of oxygen in the gas mixture on the structural characteristics of nanostructured Al_2O_3 thin films by dc reactive sputtering technique.

2. Experimental Part

A homemade DC reactive magnetron sputtering system was used to prepare Al_2O_3 thin films. A high-

purity (99.99%) aluminum sheet served as the sputtering target, used in an oxygen atmosphere to deposit Al_2O_3 films onto glass substrates. Prior to deposition, both the targets and glass substrates underwent thorough cleaning and drying. The target was meticulously positioned on the cathode, and sputtering plasma was generated by an electrical discharge of argon, powered by a high-voltage DC supply (up to 5kV). The technique involved gas mixtures of argon and oxygen with various mixing ratios (10:90, 30:70, 50:50, 70:30, 90:10). The deposition chamber was initially evacuated to 0.001 Torr, and the process was conducted at room temperature with a discharge voltage of 650-700 V and a discharge current of 25 mA. Deposition times for all samples were fixed at 30, 45, 60, 90, and 120 minutes, with an inter-electrode distance of 4 cm. The gas mixture pressure was maintained at approximately 0.15 mbar. Gas mixture flow rate was precisely controllable, and chamber temperature was measured by a precision thermometer. Argon and oxygen were supplied from gas cylinders to the mixer, with ratios controlled by fine needle valves. Both electrodes incorporated a water-cooling system via internal channels. Further specifics on this sputtering system are available in references [24-27]. The prepared samples were characterized by the field-emission scanning electron microscopy (FE-SEM) and energy-dispersive X-ray spectroscopy (EDS).

3. Results and Discussion

Figure (1) shows the FE-SEM images offering a visual insight into the surface morphology and nanoparticle characteristics of Al_2O_3 thin films prepared in this work. These images reveal how the gas mixing ratio critically influences the film's nanostructure, including particle size, distribution, and overall surface homogeneity. Using gas mixing ratio of 90:10, which represents an argon-rich and oxygen-deficient environment, the surface is characterized by relatively large, irregularly shaped agglomerates of particles. The film appears less dense, with visible voids and a somewhat heterogeneous distribution of material. This morphology is consistent with incomplete oxidation or a lower sticking coefficient of aluminum species due to insufficient reactive gas, leading to the formation of larger, loosely packed structures. The lack of sufficient oxygen during deposition can hinder the formation of a uniform, fine-grained Al_2O_3 film. As the oxygen content in the gas mixture is increased (70:30), the film's morphology begins to show an improvement in terms of particle size and distribution. The image reveals smaller, more uniformly distributed nanoparticles compared to the 90:10 ratio. While some agglomeration is still present, the overall surface appears denser and more homogeneous. This indicates that a moderate increase in oxygen supply promotes better oxidation of the

sputtered aluminum, leading to the formation of finer Al_2O_3 nanoparticles and a more compact film structure. The FE-SEM image for the sample prepared using gas mixing ratio of 50:50 displays a surface composed of relatively fine and densely packed nanoparticles. The agglomerates are smaller and less pronounced compared to the lower oxygen ratios, suggesting a more uniform and compact film. This intermediate ratio seems to strike a good balance, allowing for efficient oxidation and particle nucleation, which contributes to a smoother and more homogeneous film. The particles appear to be well-defined and closely spaced, indicating improved film quality. Using mixing ratio of 30:70, where oxygen becomes the dominant gas, the film exhibits a surface with very fine, uniformly distributed nanoparticles. The image shows a highly dense and homogeneous film, with minimal visible voids or large agglomerates. The nanoparticles appear to be very small and tightly packed, forming a continuous layer. This morphology suggests that an oxygen-rich environment facilitates complete oxidation and promotes the formation of very small, well-crystallized Al_2O_3 particles, leading to a highly uniform and smooth film surface. Finally, at the highest oxygen content in the mixture (10:90), the FE-SEM image reveals a surface that is still composed of fine nanoparticles, but with some evidence of increased porosity or a slightly less uniform distribution compared to the 30:70 ratio. While the particles are generally small, there are regions where they appear to form slightly larger clusters or where the packing density might be marginally reduced. This could be due to excessive oxygen leading to gas phase reactions or a change in the sputtering mechanism that affects particle aggregation on the substrate. However, overall, it still represents a relatively fine-grained and dense film compared to the argon-rich conditions.

The FE-SEM images clearly demonstrate that the $\text{Ar}:\text{O}_2$ gas mixing ratio plays a crucial role in controlling the nanoscale morphology of Al_2O_3 thin films prepared by DC reactive sputtering. As the oxygen content increases from 90:10 to 30:70, there is a clear trend towards smaller, more uniformly distributed, and densely packed nanoparticles, leading to smoother and more homogeneous film surfaces. The 30:70 ratio appears to yield the most uniform and fine-grained films, while very high argon content (90:10) results in larger, irregular agglomerates and higher porosity. Very high oxygen content (10:90) maintains fine particles but might introduce slight non-uniformities. These morphological changes are critical for tuning the optical, electrical, and mechanical properties of the Al_2O_3 thin films for various applications.

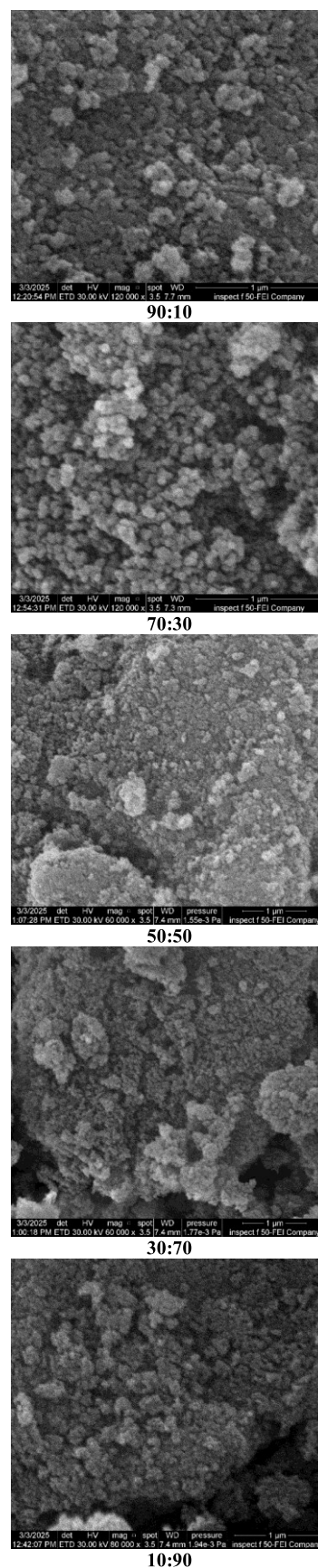


Fig. (1) FE-SEM images of Al_2O_3 thin films prepared in this work

Figure (2) illustrates the variation of the minimum particle size of Al_2O_3 thin films as a function of the Ar:O₂ gas mixing ratio, derived from FE-SEM images. This figure reveals a non-monotonic relationship, highlighting the complex influence of gas composition on nanoparticle growth during DC reactive sputtering. Using mixing ratio of 90:10 (high argon, low oxygen), the minimum particle size is approximately 41 nm. As the oxygen content is increased to 30%, the minimum particle size significantly decreases to about 28 nm. This trend continues as the oxygen content is further increased to 50%, where the minimum particle size reaches its lowest value of approximately 21 nm. This reduction in particle size with increasing oxygen content is generally expected. A higher oxygen partial pressure ensures more complete oxidation of the sputtered aluminum atoms, which can lead to rapid nucleation of Al_2O_3 nanocrystals and inhibit extensive grain growth, resulting in smaller, more uniform particles. However, a critical change occurs beyond the 50% content of oxygen as the oxygen content is increased to 70%, the minimum particle size begins to increase again, reaching approximately 27 nm. This increase continues for the Ar:O₂ ratio of 10:90, where the minimum particle size rises to around 32 nm. This observed increase in particle size at very high oxygen concentrations (low argon) is a noteworthy phenomenon. It might be attributed to several factors, such as increased gas-phase scattering of sputtered species at very high reactive gas pressures, which can reduce the kinetic energy of the arriving particles on the substrate and promote agglomeration. Alternatively, excessive oxygen might lead to target poisoning more rapidly, altering the sputtering yield and affecting the growth kinetics of the Al_2O_3 nanoparticles. This non-linear behavior suggests an optimal Ar:O₂ ratio around 50:50 for achieving the smallest Al_2O_3 nanoparticles under these specific reactive sputtering conditions.

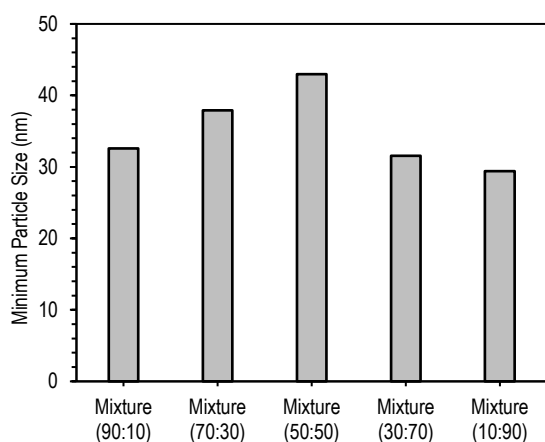


Fig. (2) Variation of minimum particle size with mixing ratio used for preparing Al_2O_3 thin films

The EDX spectra in Fig. (3) illustrate the elemental composition of Al_2O_3 thin films prepared in this work.

Each spectrum displays characteristic peaks corresponding to the constituent elements, primarily aluminum (Al) and oxygen (O), alongside a peak for carbon (C), which is likely due to adventitious carbon contamination on the sample surface or from the sample mounting process inside the FE-SEM instrument, and potentially a Si peak if a silicon substrate was used and the film was thin enough for the electron beam to penetrate. The intensity and relative ratios of the Al and O peaks provide crucial insights into the stoichiometry and oxidation efficiency of the deposited films as a function of the gas mixture. Using the mixing ratio of 90:10 (argon-rich and oxygen-deficient mixture), the spectrum shows a dominant Al peak, while the O peak is significantly smaller. This indicates that the film formed under these conditions is highly oxygen-deficient, suggesting the formation of sub-stoichiometric aluminum oxide (AlO_x , where $x < 1.5$) or even metallic aluminum inclusions within the film. The low oxygen supply limits the complete oxidation of the sputtered aluminum atoms. As the oxygen content is increased to 30%, the relative intensity of the O peak shows a noticeable increase compared to the Al peak. While the film might still be slightly oxygen-deficient, it is clear that a greater amount of oxygen has been incorporated into the film's structure. This signifies improved oxidation efficiency with increased oxygen partial pressure, leading to a composition closer to stoichiometric Al_2O_3 . Increasing the oxygen content to 50%, the relative intensity of the O peak becomes even more prominent and appears to be in a more balanced proportion with the Al peak. This suggests that at this intermediate gas mixing ratio, the film's composition is closer to the ideal stoichiometric Al_2O_3 . The optimized balance between the sputtering rate of aluminum and the reactive oxidation process likely contributes to the formation of a more chemically accurate Al_2O_3 compound. The peak labeled "Al-A" in the spectrum likely refers to a specific X-ray emission line of aluminum (e.g., $\text{Al K}\alpha$ or $\text{Al K}\beta$), confirming its presence. When the oxygen content is further increased to 70%, the relative intensity of the O peak continues to be strong, possibly even slightly surpassing the Al peak in relative magnitude depending on the specific quantification. This indicates that sufficient or even surplus oxygen is available for the complete oxidation of aluminum. The film formed is expected to be well-oxidized and close to, or even slightly oxygen-rich compared to, perfect stoichiometry if any excess oxygen is incorporated into the lattice or as adsorbed species. Finally, at the highest oxygen content (90%), the EDX spectrum shows a very strong O peak relative to the Al peak. This confirms that the film is fully oxidized and likely very close to, or at, the stoichiometric composition of Al_2O_3 . The abundance of oxygen in the sputtering environment ensures that nearly all sputtered aluminum atoms are completely oxidized upon deposition.

The EDX analysis consistently demonstrates that the oxygen content in the Ar:O₂ sputtering gas mixture critically dictates the elemental composition and stoichiometry of the deposited Al₂O₃ thin films. A transition from oxygen-deficient (sub-stoichiometric) films at low oxygen partial pressures to well-oxidized (stoichiometric) films at higher oxygen partial pressures is clearly observed. The 50:50, 30:70, and 10:90 ratios appear to yield films closer to the desired Al₂O₃ stoichiometry, which is crucial for achieving optimal electrical, optical, and structural properties for various applications.

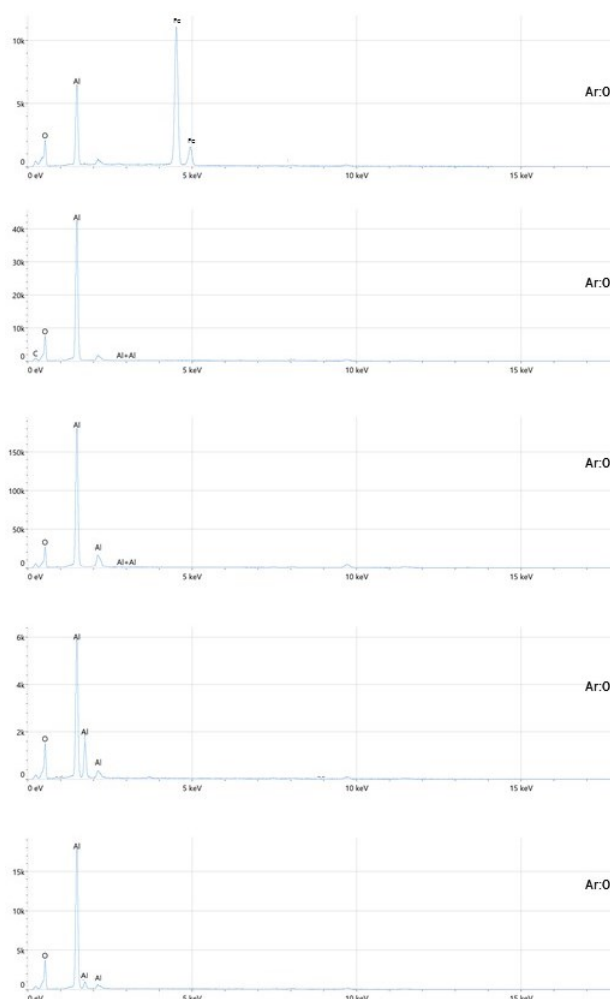


Fig. (3) EDX spectra of Al₂O₃ thin films prepared in this work

4. Conclusions

The gas mixing ratio plays a crucial role in controlling the nanoscale morphology of the prepared thin films. As the oxygen content is increased, there is a clear trend towards smaller, more uniformly distributed, and densely packed nanoparticles, leading to smoother and more homogeneous film surfaces, while very high argon content results in larger, irregular agglomerates and higher porosity. Very high oxygen content maintains fine particles but might introduce slight non-uniformities. The [Al]/[O] ratio varies non-

linearly with the Ar:O₂ mixing ratio. While increasing oxygen generally brings the film closer to stoichiometry, an intermediate mixture presents an anomaly with the highest oxygen deficiency. These results are useful to optimize the sputtering process to produce desired samples for various applications, as surface roughness profoundly impacts adhesion, optical properties, and device performance.

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