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Deposition and Characterization of Zinc Assisted Ultrathin Silver Thin Films

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ABSTRACT

The high surface plasmon resonance of ultrathin silver (Ag) films, even for coatings as thin as a few nanometers, has piqued the curiosity of many. This work presents a viable method for improving the performance and stability of ultrathin Ag films by adding a thin layer of zinc (Zn) filler. Glass, quartz, silicon, and polyethylene terephthalate (PET) were used as substrates for the deposition of Ag films with a thickness of less than 15 nm by means of DC magnetron sputtering. Film continuity, adhesion, and suppression of Ag diffusion into the substrate are all much enhanced by this designed interface. In addition, the films that are supported by Zn show better mechanical compatibility and environmental stability, particularly on flexible substrates such as PET. In order to overcome the basic constraints of ultrathin Ag films, the results show that Zn, when used as a filler and interfacial modifier, is beneficial. This method provides an encouraging technique to create transparent conductive electrodes with good performance, which might be used in solar cells, LEDs, and sophisticated plasmonic systems.

Keywords: *Ultrathin films, Silver, Zinc, Plasmonic, Roughness.*

I. INTRODUCTION

Recent years have seen a surge in interest in thin metal films—particularly those of gold, silver, and copper—in the scientific and technological communities for their potential use in a wide variety of applications, such as optical cloaking devices, catalytic coatings, surface plasmon resonance (SPR) biosensors, antibacterial coatings, transparent electrodes for "smart windows" and solar cells, transparent conductive coatings, and many more.

When compared to bulk silver, silver in a thin film (less than 20 nm) has superior optical characteristics and works very well in optical applications. Most plasmonic materials are thin films of silver, which have the unique properties of being both partly transparent and electrically conductive. Among plasmonic materials, silver (Ag) has the reputation of having the best ohmic losses at optical frequencies due to its inherent properties. When it comes to optical devices, silver films outperform other metals due to their superior electrical and optical characteristics. Industrial applications are recommended for silver nanoparticle deposition due to its high reproducibility and similarity in size distribution under same deposition circumstances. However, there are some technological obstacles in the areas of thin film synthesis, nanoscale manufacturing processes, and the shape of metal layers that limit the development of silver-based materials for plasmonic and metamaterial applications.

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A variety of methods are employed for the deposition of thin films of Ag, including glancing angle deposition (GLAD), plasma enhanced atomic layer deposition (PEALD), DC magnetron sputtering, thermal evaporation and low-pressure direct current (DC) plasma sputtering, DC magnetron sputtering, pulsed laser deposition (PLD) RF magnetron sputtering, and electron beam evaporation. The main drawbacks of thermal evaporation technologies are the films' lack of adhesion and stability. For unique patterns, you can utilize GLAD and PEALD. When it comes to regulated and homogenous ultrathin films of Ag, magnetron sputtering is a great option.

The creation, detection, and driving of optically-frequency signals over nanometer-scale metal-dielectric surfaces are known as plasmonics and nano-plasmonics, respectively. Miniaturized improved optical devices, such as energy harvesting cells, metal-dielectric-metal flexible nanostructures, and optical sensors, are the focus of plasmonics, much as they are in photonics. For the purpose of detecting molecule adsorption in biopolymers, Surface Plasmon Resonance (SPR) measurements can be employed. Specifically, a change in the local index of refraction following adsorption on nanoparticles can cause a shift in this resonance, which can be utilized to identify biopolymers.

Although a single ultrathin film deposited using the magnetron sputtering method is straightforward, a comprehensive comprehension of the optical properties of such films integrated into photonic structures (with a thickness of less than 10 nm) remains conceptual and challenging to comprehend in practice. Knowing how to deposit Ag on diverse substrates without a seed layer is crucial for preparing ultra-thin layers for various uses. A number of recent articles have sought to shed light on the issues associated with the deposition and characteristics of ultra-thin Au or Ag films, and they have focused on commercial substrates such as optical glass, oxide coatings, PET plastic, and other polymers.

II. REVIEW OF LITERATURE

Tran, Thanh et al., (2023) A wide variety of optoelectronic devices and optical coatings utilize silver thin films. However, they tend to form islands by the Volmer-Weber process on surfaces like glass because of how poorly they wette. Using magnetron sputtering and a low-energy ion beam treatment, this research demonstrates how to create continuous silver sheets as thin as 6 nm. A notional 1 nm seed layer is produced using a single-beam ion source, and it considerably improves the wettability of the subsequent silver films. A resistance as low as 11.4 μΩ.cm is produced by this method, which results in a continuous covering of around 6 nm. These films showed significantly improved glass adhesion in the traditional 100-grid tape test compared to silver films sputtered without ion beam treatment, and their transmittance spectra were determined to be comparable to the values derived from models. Images captured using high-resolution scanning electron microscopy during the early stages of development reveal that films subjected to an ion beam treatment have a tendency to form islands, whereas films that are not treated with an ion beam treatment promote nucleation. Based on X-ray diffraction patterns, the soft ion beam treatment inhibits (111) crystallization and promotes the

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growth of large crystals oriented (200). This method has a lot of potential for improving the properties of silver thin films, which might lead to their use in optoelectronics and optical coatings.

Li, Dong et al., (2022) As the metal with the lowest electrical resistance and the best optical properties in the visible and near-IR bands, Ag is ideal for use in transparent conductors made of thin metal sheets. An ultra-thin, see-through electrode made of silver film with aluminum as its seed layer was created in this work utilizing a resistive thermal evaporation technique. Films of silver with a flat surface and good electrical conductivity are produced when a thin layer of aluminum changes the rate of development of thermal evaporation of Ag. Different electrical, optical, and surface morphological methods have also shown that the film's threshold thickness has decreased. Results from examinations of surface morphology, sheet resistance, and transmittance spectra indicated that the ultra-thin silver film was positively affected by the aluminum layer's infiltration. Furthermore, a 10 nm silver film has an average transmittance of 40% and a sheet resistance of 13 Ωsq -1 in the 400-2500 nm range. Research shows that adding an Al seed layer to transparent conductive silver films improves their properties. This was followed by the recommendation of an alternate method for seed layer deposition of 1 nm Al. To do this, the 1 nm layer of aluminum invading material must be divided into two or more layers. The 5 nm silver film has an average transmittance of 60% and a sheet resistance of less than 100 Ω sq-1 in the 400-2500 nm region.

Filip, Ana et al., (2022) Many are interested in ultrathin silver (Ag) films because of its strong surface plasmon resonance, which is achieved even for coatings as thin as a few nanometers. Optical glass and polyethylene terephthalate (PET) are among the substrates that have been deposited ultrathin films with minimal roughness using radio frequency (RF) magnetron sputtering. The films' thicknesses vary between 1 and 9 nanometers. As seen by the UV-Vis spectra, the films have robust surface plasmon resonance capabilities up to a 7 nm thickness. Looking at the films using AFM and SEM, one would think the depositions are smooth and uniform because of the extremely little roughness. The bandgap values are inversely proportional to the film thickness. Visual domain minima in the 400-600 nm wavelength region and values below 1 are observed in film refractive indices as assessed by ellipsometry experiments. Ultrathin Ag films with better surface plasmon resonance and decreased roughness might be made using the results, which could have photonic applications.

Liu, Hong et al., (2010) According to this research, a thin Ni seed layer assisted deposition on Ag films may significantly improve surface plasmon resonance (SPR). A 50 nm thick coating of Ag was deposited on quartz and silicon substrates by means of electron beam evaporation, covering a very thin layer of Ni, measuring around 2 nm. The root-mean-square (rms) surface roughness and correlation length of pure Ag films are >4 nm and 28 nm, respectively. On the other hand, Ag/Ni films have these parameters reduced to around 1.3 nm and 19 nm, respectively. Despite the pure Ag film having a lower complete width at half-maximum, the experimental and theoretical results show that the Ag/Ni films perform better in terms of SPR. Ag films with a Ge seed layer have also been made using the same methods. Since the Ge seed layer undergoes more absorption damping, the SPR

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curve remains unchanged regardless of whether the surface roughness is reduced to below 0.7 nm. It has been shown that Ni acts as a growth layer for the Ag film, smoothing it out and keeping and even increasing the plasmonic properties of the combined structures. This suggests that optical metamaterials and low-loss plasmonic devices might be good applications for it.

Yliniemi, Kirsi et al., (2008) Stainless steel and glass surfaces may be treated with ultra-thin coatings containing Ag nanoparticles using a simple three-step procedure. Silver ions and the two amino groups of DIAMO create a complex when the sample is soaked in silver nitrate, leading to the formation of massive surface clusters. The first step is to immerse the sample in N-(2-aminoethyl)-3-aminopropyl-trimethoxysilane (DIAMO), which will attach a monolayer of the compound to the surface of the sample. In the annealing process, these silver clusters scatter and link to the surface to generate silver nanoparticles. With the use of FE-SEM, UV/Vis, and FE-AES, the film formation was characterized. Additionally, the effect of nanoparticle attachment and surface SERS activity on antibacterial characteristics was investigated.

III. MATERIALS AND METHODS

This experiment made use of glass, quartz, and silicon substrates. The substrates were prepared for deposition by washing them with distilled water, rinsing them with acetone and ethanol, and finally drying them with 99.999% pure N2 gas. A DC magnetron reactive sputtering device called the ATS Hind High Vacuum 500 was used to carry out the deposition procedures. In order to determine the thin films' purity, the sputtering chamber was evacuated to a pressure of 5×10^{-6} mbar prior to deposition. To make sure the target surface wasn't contaminated, pre-sputtering was done for 10 minutes at 8.5×10^{-3} mbar and 30 W DC power. From a 2 inch diameter and 0.25 inch thick, 99.99 percent pure target, silver was sputtered. The carrier gas for sputtering was argon, which has a purity level of 99.999%. Ultrathin Zn-assisted Ag films were deposited by optimizing three deposition process parameters: sputtering power, working pressure, and deposition rate. These values are detailed in Table 1.

Table 1: Process Optimization Parameters for Ag and Zn-Based Thin Films

Process parameter	Target Material (Ag)	Target Material (Zn)
Sputtering power (W)	5	10
Working pressure (mbar) or Flow rate (sccm)	5×10^{-3} (23)	8.5×10^{-3} (31)
Deposition rate (nm/min)	1.5	2

Three distinct conditions were used for the deposition: first, as Ag deposition; second, as Zn and Ag deposition; and third, substrate preheating at 100 °C for 30 minutes, Zn deposition, and holding at 100 °C for 1 hour; cooling to room temperature (35 °C), and finally, Ag deposition, and annealing at 100 °C inside the chamber for 1 hour. Condition 1, Condition 2, and Condition 3 will be used to describe these deposition situations going forward.

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The optical profilometer was used to determine the film thickness (t), while the atomic force microscope (AFM) (BRUKER, Dimension icon) was used to assess the surface roughness (Ra) of the deposited Zn-assisted Ag films with scan areas of 3 μm × 3 μm and 10 μm × 10 μm for each sample. Scanning electron microscopy (SEM) was used to examine the surface morphology of very thin Ag sheets (Apreo S). A four-point probe (JANDAL, RM3000) equipped with the proper electronics for detecting voltage and current is used to determine the sheet resistance (Rs). Perkin Emler LAMDA 950 UV-VIS spectroscopy was used to get the transmittance data. For every example, we averaged the values and determined the standard deviation.

IV. RESULTS AND DISCUSSION

Surface morphology

The SEM was used to examine the surface morphology of the ultrathin films made of Ag. The surface of the 6 nm Ag film shows discontinuities in Figure 1, which are caused by the energy mismatch between the substrate and the Ag, which results in the growth of an island of Ag film on top of the quartz substrate, also known as an open circuit. On the other hand, the surface of the film shows continuity, which is the result of the 3D assisted growth of the film, with a relative surface area (RS) of 60 Ω /sq. c) The longer the annealing process, the more consistent the surface of the Ag film appears, with a resistance of 12 Ω /Sq.

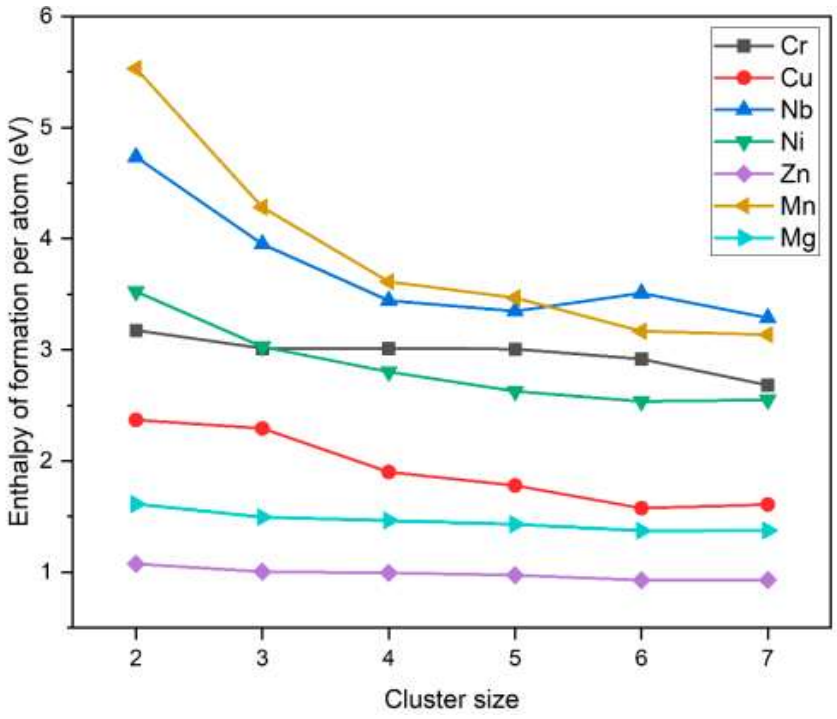

Figure 1: Variation of Formation Enthalpy of Neutral Clusters with Cluster Size

Good continuity (low sheet resistance) was observed in the Zn-assisted Ag film formed at condition 3 (Figure 2). The reason behind this is the formation of a 3-dimensional enhanced atomically graded

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contact by the Zn filler metal, the substrate, and the ultrathin Ag. Depositing and studying the optoelectronic characteristics of ultrathin Ag films was done under the same environment 3.

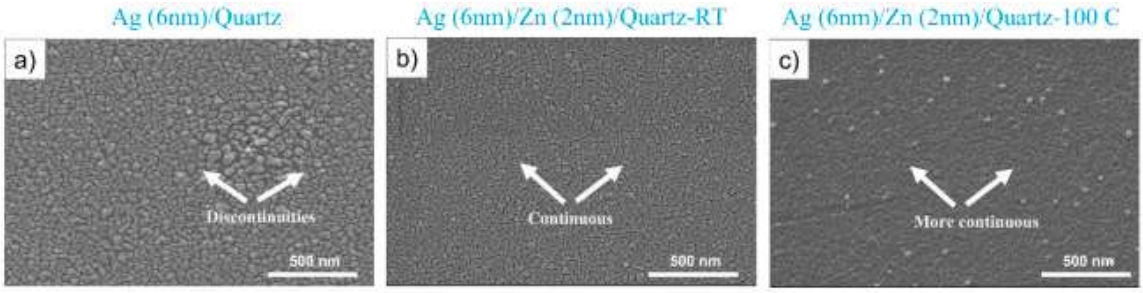

Figure 2: SEM Micrographs of Ag Thin Films Deposited on Quartz Substrate

Figure 2 shows scanning electron micrographs of Ag thin films that have been deposited on a quartz substrate. The films are arranged as follows: (a) as deposited Ag (6 nm) film; (b) as deposited Zn (2 nm) assisted Ag (6 nm) film; (c) as a Zn assisted Ag film (substrates are heated to 100 °C for 30 minutes, then Zn (2 nm) is deposited and held at 100 °C for 1 hour. After cooling to room temperature (35 °C), Ag (6 nm) is deposited and annealed at 100 °C for 1 hour.) or as condition 3, Zn (2 nm) assisted Ag (6 nm) film.

Surface roughness

Zn filer metal 3D-assisted Ag ultrathin films had their surface roughness evaluated using AFM. At condition 3, which involves preheating the substrates at 100 °C for 30 minutes, deposition of Zn 2 nm, holding at 100 °C for 1 hour, cooling to room temperature (35 °C), and finally, deposition of Ag 8 nm, followed by annealing at 100 °C for 1 hour, are shown in two- and three-dimensional AFM images in Figures 3 and 5. These films had an average surface roughness of 2.57 nm, 2.09 nm, and 2.02 nm, respectively.

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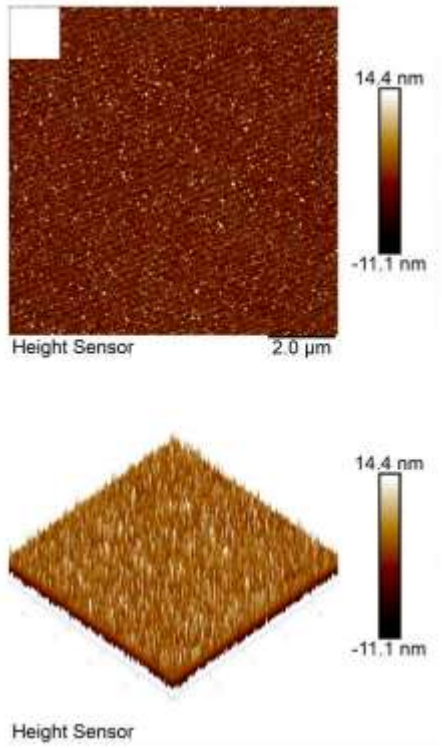

Figure 3: 2D and 3D AFM Analysis of Zn (2 nm)/Ag (6 nm) Films Deposited on Glass Substrate (10 µm × 10 µm)

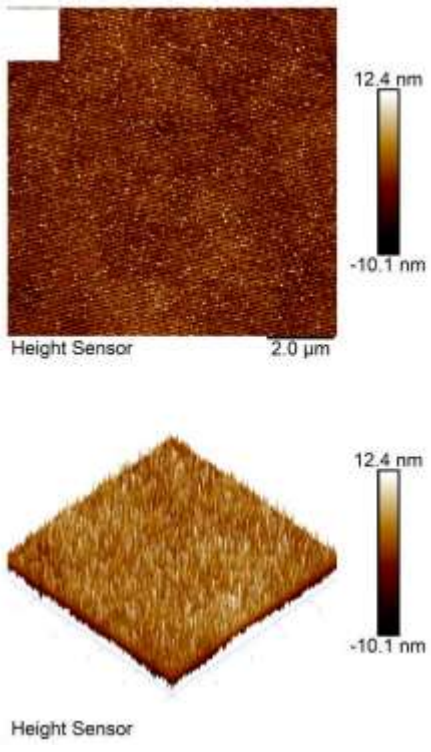

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Figure 4: 2D and 3D AFM Analysis of Zn (2 nm)/Ag (6 nm) Thin Films Deposited on Quartz (10 µm × 10 µm)

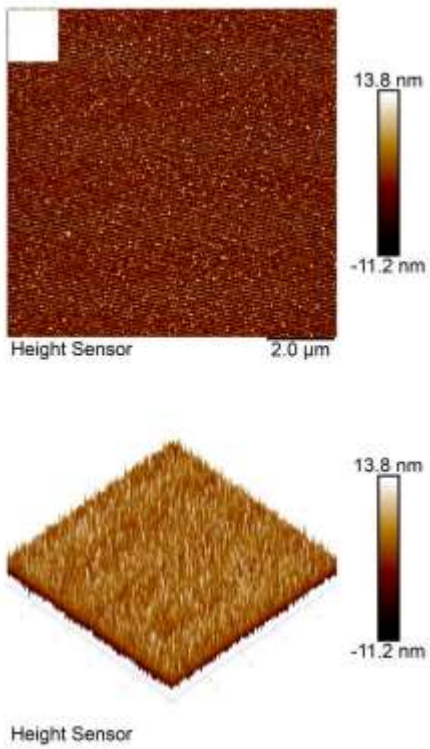

Figure 5: 2D and 3D AFM Analysis of Zn (2 nm)/Ag (6 nm) Thin Films Deposited on Quartz–Silicon (10 µm × 10 µm)

Optoelectronic properties

A transparent and conductive film was achieved by depositing ultrathin Zn 3D aided Ag films on glass, quartz, and PET substrates. You can see the results of the measurements for the Ag films' optical transmittance and sheet resistance in Table 2. In Table 2 you can see the numbers. The sheet resistance and scattering are both affected by the increasing thickness of Ag, since the bulk nature of Ag causes a decrease in transmittance along with an increase in scattering. Films of Ag (8 nm) and Zn (2 nm) placed on quartz have a greater FoM.

Table 2: Effect of Ag Thickness on Haacke FoM of Zn-Assisted Ag Ultrathin Films on Various Substrates at Condition 3

Film	Sheet resistance Ω/sq.	Transmittance (%)	FOM (m Ω -1)
A6/Z2/G	12	62.85	0.80
A6/Z2/Q	12	62.64	0.77
A8/Z2/G	5	59.56	1.12

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A8/Z2/Q	5	60.21	1.26
A8/Z2/PET	15	46.91	0.04

Figures 6 and 7 demonstrate that ultrathin Ag films have high visible-light transmittance. The surface resistance, transmittance, and Hackee figure of merit (FOM) of the 8 nm Ag films coated on quartz substrate were 5 Ohm/Sq., 60.21 percent, and 1.26 mOhm-1, respectively. In comparison to the ultrathin Ag films displayed in Table 2, the Zn-assisted Ag (8 nm) films on PET substrate exhibited comparatively low sheet resistance (15 Ohm/Sq.), transmittance (46.71%), and FOM (0.04 (mOhm-1) values. The graph shown displays the sheet resistance with time, thus you can see that these films were more stable.

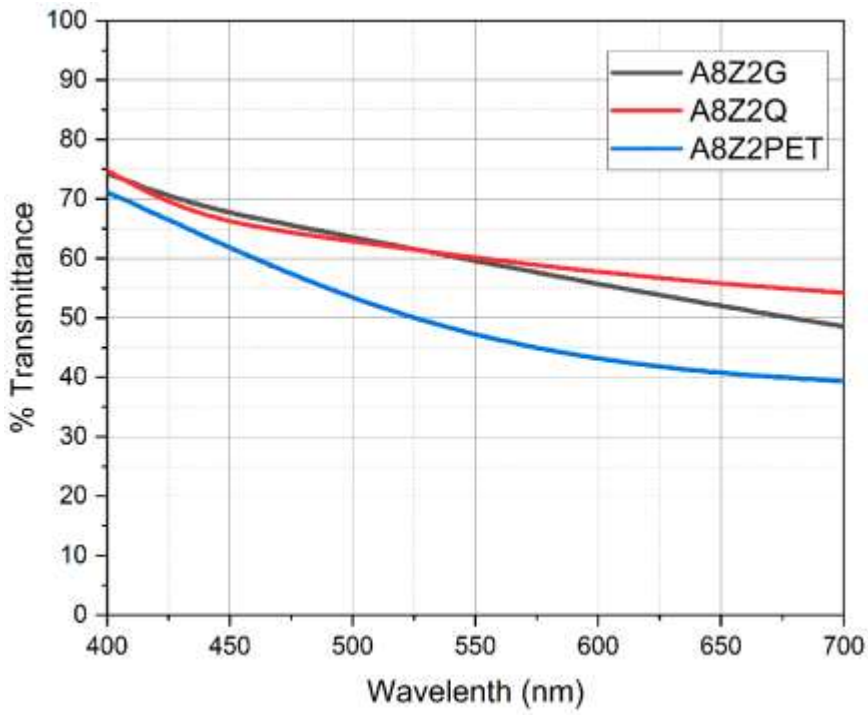

Figure 6: Substrate-Dependent Transmittance of 8 nm Ag Films with 2 nm Zn Filler Layer at Condition 3

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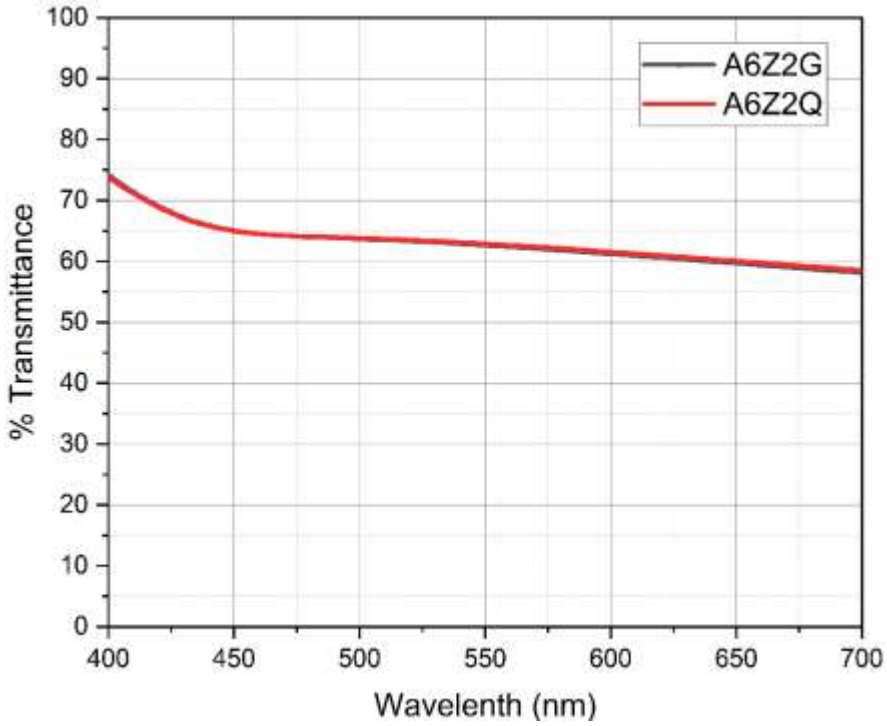

Figure 7: Substrate-Dependent Transmittance of 6 nm Ag Films with Zn Filler Layer at Condition 3

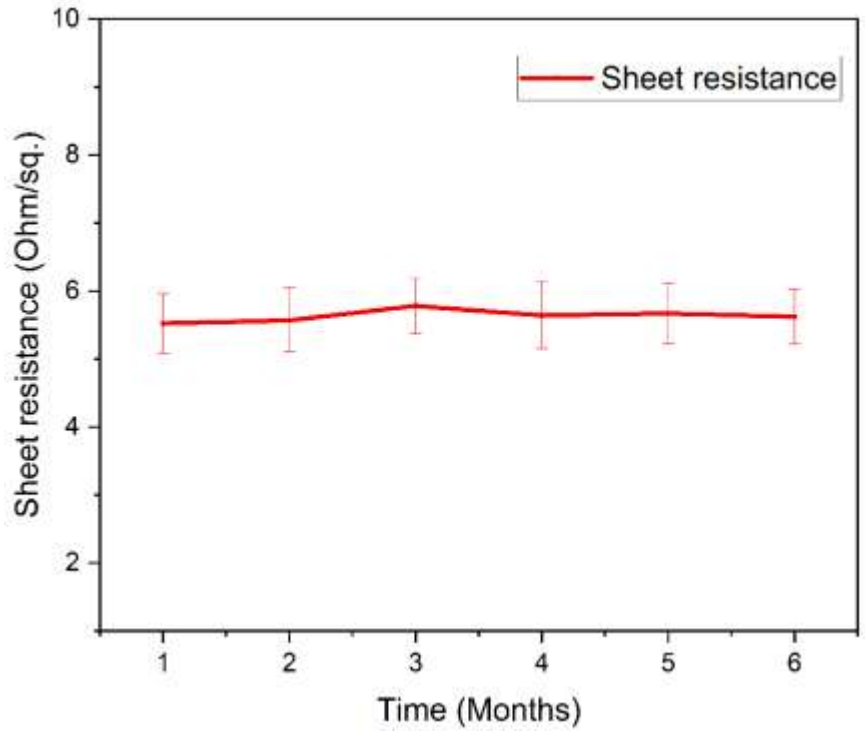

Figure 8: Variation of Sheet Resistance with Time for Zn (2 nm)/Ag (8 nm) Ultrathin Films at

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Condition 3

V. CONCLUSION

Zinc allows for the creation of a chemically graded interface in three dimensions, which improves film continuity and adhesion on various substrates such as PET, silicon, glass, and quartz. The Zn-assisted films solve the problems of island formation and poor wettability that are common with ultrathin Ag layers by smoothing out the surface and decreasing discontinuities. Reduced sheet resistance, increased optical transparency (from moderate to high), and enhanced environmental stability are all desirable properties of the optimized films. Because of improved interface formation and structural regularity, films deposited under regulated temperature settings outperform the other samples investigated. These findings provide credence to the idea that film quality is heavily dependent on substrate preparation and deposition circumstances.

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