

Chapter 2

Silver Concentration Effects in PEO–Ag Nanocomposite Thin Films and their Applications

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1. Introduction

Polymer nanocomposites containing metallic nanoparticles have emerged as an important class of multifunctional materials because they combine the flexibility, low weight, and processing advantages of polymers with the electrical, optical, and thermal characteristics of metallic nanostructures. Among metallic nanofillers, silver (Ag) nanoparticles are particularly attractive because of their high electrical conductivity, high surface-to-volume ratio, chemical stability, and distinctive nanoscale electronic properties. These characteristics make Ag nanoparticles useful for tailoring the dielectric, conductive, optical, and sensing properties of polymer-based materials (Rycenga et al., 2011; Moharana et al., 2021).

Polyethylene oxide (PEO) is a particularly useful polymer matrix because of its semicrystalline structure, flexible backbone, good film-forming ability, and ether oxygen groups. The oxygen atoms along the PEO chain can interact with metal species and influence nanoparticle dispersion, polymer-chain organization, and charge-carrier mobility. Pure PEO has relatively low electrical conductivity because of its limited concentration of mobile charge carriers. Incorporating conductive Ag nanoparticles can modify this behavior by introducing highly polarizable interfaces and additional pathways for charge migration (Armand, 1983; Wright, 1975).

Silver concentration is one of the principal variables determining the functional behavior of PEO–Ag nanocomposite films. At low Ag concentrations, the nanoparticles tend to remain isolated within the polymer matrix. Their contribution to electrical behavior is therefore mainly associated with localized polarization and charge accumulation at the Ag/PEO interfaces. Increasing Ag concentration decreases the average separation between neighboring particles, facilitating hopping and tunneling of charge carriers. At still higher concentrations, the particles may approach a percolation threshold at which interconnected conducting regions are established and electrical conductivity increases substantially (Dang et al., 2012; Lewis, 2004).

The concentration dependence of these properties means that increasing the Ag content does not necessarily improve every characteristic of the composite. Well-dispersed nanoparticles increase the number of metal–polymer interfaces and can enhance dielectric polarization, whereas excessive Ag loading can promote aggregation and conductive leakage, leading to higher dielectric losses. An optimum concentration is therefore expected in which beneficial interfacial polarization and charge transport are enhanced without converting the material into an excessively lossy conductive system (Barber et al., 2009; Sahu et al., 2020).

Physical vapor deposition (PVD) is particularly attractive for investigating these concentration-dependent phenomena because it offers control over film composition, thickness, morphology, and interface formation. PVD-based fabrication under controlled vacuum

conditions can produce high-purity thin films and permits systematic variation of metallic content. Consequently, PVD provides an appropriate platform for correlating Ag concentration with structural evolution, dielectric behavior, relaxation dynamics, and charge transport in PEO–Ag nanocomposites (Nur-E-Alam et al., 2021).

This chapter examines how variation in silver concentration modifies the structure and electrical behavior of PEO–Ag nanocomposite thin films fabricated by PVD. Particular attention is given to the progression from isolated Ag nanoparticles to interacting clusters and eventually conductive networks, and to how this evolution affects crystallinity, interfacial polarization, dielectric relaxation, and charge-transport mechanisms.

2. PVD Fabrication

Physical vapor deposition is a vacuum-based thin-film fabrication process in which a source material is converted into the vapor phase and subsequently condensed onto a substrate. PVD techniques are widely employed for metallic, ceramic, polymeric, and hybrid coatings because they provide effective control over film thickness, composition, purity, and microstructure (Ohring, 2002). These advantages are particularly relevant to polymer–metal nanocomposites, where small variations in nanoparticle concentration and spatial distribution can substantially alter electrical behavior.

PEO–Ag nanocomposite thin films can be produced using PVD-related approaches such as thermal evaporation, magnetron sputtering, sequential deposition, or co-deposition. During Ag deposition, silver atoms reaching the polymer surface undergo nucleation and growth. Depending on deposition conditions, these atoms may remain as relatively isolated nanoscale domains, combine into larger clusters, or develop interconnected metallic regions. Ag concentration, deposition rate, substrate temperature, vacuum conditions, and film thickness therefore collectively determine the final morphology.

At low silver concentrations, the relatively small population of deposited Ag atoms favors the formation of separated nanoparticles surrounded by PEO. Interactions between the Ag surface and oxygen-containing groups of the polymer may influence nucleation and stabilization of these domains. Because the metallic particles remain physically separated, direct electron conduction between them is restricted.

Increasing Ag concentration increases the density of metallic particles and decreases their mean separation. This change is important because the probability of charge transfer between neighboring particles increases rapidly as the insulating polymer barrier becomes thinner. Continued Ag incorporation can therefore produce a morphological progression from isolated nanoparticles to clusters and eventually interconnected metallic pathways. This concentration-driven evolution is closely related to percolation behavior, in which a critical concentration

produces a pronounced change in macroscopic electrical conductivity (Stauffer & Aharony, 1994).

Excessive Ag incorporation, however, may promote uncontrolled aggregation and structural heterogeneity. Large clusters reduce the effective interfacial area relative to uniformly dispersed nanoparticles and can generate leakage pathways. The ability of PVD to regulate Ag loading is therefore important for identifying a concentration range that provides sufficient nanoparticle interaction for enhanced electrical properties while preserving desirable film uniformity.

3. Structural Changes

PEO is a semicrystalline polymer containing ordered crystalline domains together with less ordered amorphous regions. These two phases have different implications for electrical behavior. Crystalline domains constrain polymer-chain motion, whereas amorphous regions provide greater segmental mobility. Consequently, structural changes induced by Ag incorporation can indirectly affect dielectric relaxation and charge transport.

X-ray diffraction (XRD) can be used to follow these concentration-dependent changes. Incorporation of Ag nanoparticles can disturb regular PEO chain packing and alter the relative contributions of crystalline and amorphous regions. Nanoparticles positioned between polymer chains may act as structural obstacles that inhibit efficient chain organization. A reduction in PEO crystallinity can consequently increase the amorphous fraction, which generally provides greater polymer segmental mobility and can facilitate charge migration (Arya & Sharma, 2018).

At sufficiently high Ag loading, crystalline metallic domains may become increasingly important. The appearance or strengthening of diffraction features associated with metallic Ag would indicate increased formation or growth of crystalline silver domains. Thus, concentration-dependent XRD behavior can provide evidence for simultaneous modification of the polymer structure and evolution of the metallic phase.

Fourier-transform infrared spectroscopy (FTIR) provides complementary information about interactions between PEO and Ag. The ether groups of PEO constitute potential interaction sites for metallic species. Changes in the position, intensity, or shape of characteristic PEO vibrational bands after Ag incorporation can therefore indicate modification of the local chemical environment. Such interactions are important because they can influence both chain arrangement and local charge distribution.

Surface and microstructural techniques such as scanning electron microscopy (SEM), transmission electron microscopy (TEM), and atomic force microscopy (AFM) can provide direct information about particle distribution and film morphology. At relatively low Ag concentration, separated metallic domains are expected to dominate. Increasing concentration promotes particle growth and clustering, leading to greater surface heterogeneity and potentially

increased roughness. At sufficiently high concentrations, neighboring metallic domains can approach or contact one another, establishing the morphological conditions required for percolative conduction.

The structural response to increasing Ag concentration is therefore central to understanding the electrical behavior of PEO–Ag films. The important variable is not simply the total amount of silver but how that silver is distributed within the polymer. Uniformly dispersed particles can maximize interfacial area, whereas aggregated or interconnected particles increasingly favor conductive transport. The resulting balance determines whether increasing Ag loading primarily enhances dielectric polarization or produces substantial electrical leakage.

4. Dielectric Behavior

The dielectric response of PEO–Ag nanocomposite thin films originates from polarization processes within the polymer and at the interfaces between phases. Introducing conductive Ag nanoparticles into comparatively insulating PEO creates a heterogeneous electrical structure characterized by strong contrasts in conductivity and permittivity. These interfaces can therefore make a substantial contribution to the effective dielectric response.

An important mechanism is Maxwell–Wagner–Sillars interfacial polarization. When an alternating electric field is applied to a heterogeneous system containing phases of different conductivity, mobile charges can accumulate at their boundaries. In PEO–Ag films, the large conductivity contrast between metallic Ag and polymeric PEO favors charge accumulation at Ag/PEO interfaces, increasing the effective polarization of the nanocomposite (Lewis, 2004).

At low Ag concentration, the particles are relatively isolated, so polarization occurs predominantly around individual metallic domains. Long-range charge transport remains restricted by the surrounding insulating polymer. As the Ag concentration increases, the number and density of Ag/PEO interfaces increase, creating more sites for charge accumulation. This can produce an increase in effective dielectric permittivity, particularly at lower frequencies where charge carriers have sufficient time to respond to the external field (Sahu et al., 2020).

The relationship is not expected to remain beneficial indefinitely. As particles become closely spaced, tunneling and direct conduction become increasingly important. Close to the percolation region, very large apparent dielectric responses can be accompanied by substantial dielectric loss. Once continuous conductive pathways develop, leakage conduction may dominate over charge-storage behavior. Consequently, Ag concentration must be optimized rather than simply maximized.

Frequency also strongly affects dielectric response. At low frequencies, interfacial and space charges have sufficient time to migrate and accumulate, resulting in stronger polarization. As frequency increases, slower polarization mechanisms can no longer follow rapid reversals of

the electric field. The dielectric constant therefore generally decreases with increasing frequency, while the loss response can reveal characteristic relaxation processes.

Silver concentration consequently affects both the magnitude and frequency dependence of dielectric properties. Low concentrations predominantly modify localized interfacial polarization, intermediate concentrations can maximize interfacial effects and charge accumulation, and high concentrations increasingly favor conductive losses. This progression establishes a direct relationship between Ag-induced structural evolution and dielectric performance.

5. Relaxation Behavior

Dielectric relaxation provides information about the time-dependent response of polymer chains, dipoles, trapped charges, and mobile carriers to an alternating electric field. In PEO–Ag films, relaxation behavior is particularly important because silver concentration simultaneously modifies polymer-chain dynamics, interfacial density, carrier concentration, and nanoparticle connectivity.

In PEO, relaxation processes are associated partly with motion of polymer segments in amorphous regions. Cooperative segmental motion is commonly associated with α -relaxation and depends strongly on temperature, molecular flexibility, and crystallinity. The polar ether groups of PEO also contribute to the dielectric response. Changes in crystallinity or local chain organization produced by Ag incorporation can therefore influence the characteristic relaxation behavior of the polymer (Wright, 1975).

At low Ag concentration, isolated particles create localized Ag/PEO interfaces. Charges can accumulate at these interfaces because of the conductivity difference between the two phases, producing interfacial relaxation. Polymer chains immediately surrounding the nanoparticle may also experience an environment different from that of the bulk polymer. Consequently, even relatively small amounts of Ag can modify relaxation strength and characteristic relaxation time (Lewis, 2004).

As Ag concentration increases, the number of interfaces initially increases and the mean interparticle distance decreases. The additional interfaces provide more charge-trapping and polarization sites, while shorter particle separation facilitates carrier transfer. Relaxation can therefore become faster as charge carriers encounter lower effective barriers between neighboring conducting domains. Near the percolation threshold, the dominant electrical response may progressively change from localized interfacial polarization to collective carrier movement through interacting or interconnected metallic regions.

Electric modulus analysis is useful for examining these processes because it can reduce the dominance of electrode polarization.

$$M^* = M' + jM''$$

$$\tau = \frac{1}{2\pi f_{max}}$$

Related metal-containing polymer nanocomposites have demonstrated concentration-dependent hopping conductivity and dielectric relaxation behavior (Kudryashov et al., 2021).

Temperature-dependent measurements can further distinguish the underlying mechanisms. Thermal energy promotes segmental motion and helps carriers overcome local potential barriers. If Ag incorporation creates more favorable charge-transfer pathways, the apparent activation energy associated with relaxation or conduction may decrease. Relaxation analysis therefore provides an important connection between microscopic Ag distribution and macroscopic electrical response.

6. Charge Transport

Charge transport in PEO–Ag nanocomposite films is determined primarily by Ag concentration, interparticle separation, particle connectivity, and the properties of the intervening polymer. Pure PEO is comparatively insulating, whereas metallic Ag possesses very high electrical conductivity. Combining the two phases produces a concentration-dependent transition from polymer-dominated transport toward nanoparticle-assisted and eventually network-dominated conduction.

At low Ag concentrations, metallic particles remain separated by comparatively thick polymer barriers. Continuous metallic conduction is therefore unavailable. Charge carriers must instead move through localized processes involving hopping between available states or tunneling between sufficiently close particles. Because the probability of tunneling decreases strongly with increasing barrier width, large interparticle separation produces relatively low conductivity.

Increasing silver concentration decreases the average interparticle distance and increases the number of possible transport paths. Electron tunneling between adjacent metallic domains consequently becomes more probable. This regime may produce substantial enhancement of conductivity even before physical contact between all neighboring particles occurs. Continued Ag incorporation ultimately produces clusters or connected networks that allow longer-range carrier transport.

The transition can be described using percolation theory (Stauffer & Aharony, 1994).

$$\sigma = \sigma_0(\phi - \phi_c)^t$$

Frequency-dependent conductivity can also provide information about carrier dynamics (Jonscher, 1977). The DC component is associated with long-range carrier migration, whereas the frequency-dependent contribution reflects localized or hopping processes.

$$\sigma(\omega) = \sigma_{dc} + A\omega^s$$

At low Ag concentration, the frequency-dependent contribution is expected to be relatively important because carriers remain localized and transport occurs through separated sites. At intermediate loading, the reduced distance between particles facilitates hopping and tunneling. At high Ag concentrations, the increasing contribution of connected metallic domains strengthens the DC conductivity component and reduces the relative dominance of localized transport.

Temperature-dependent conductivity is frequently analyzed using an Arrhenius-type relationship:

$$\sigma = \sigma_0 \exp(-E_a/kT)$$

Silver concentration can therefore shift PEO–Ag films through distinct transport regimes. Low concentrations retain predominantly dielectric and localized transport behavior; intermediate concentrations enhance hopping, tunneling, and interfacial polarization; and sufficiently high concentrations can produce percolative or network-dominated conduction. The optimum concentration consequently depends on whether the intended application requires high dielectric polarization, controlled semiconducting behavior, or high conductivity.

7. Applications

The concentration-dependent properties of PEO–Ag nanocomposite films make them potentially useful for several classes of functional devices. Their principal advantage is the possibility of combining polymer flexibility with tunable electrical behavior. By controlling Ag concentration, a film can potentially be designed to favor dielectric polarization, moderate conductivity, or more extensive charge transport.

Flexible and wearable electronics represent one relevant application area. PEO provides film-forming characteristics and mechanical flexibility, while Ag provides highly conductive domains. Control of nanoparticle distribution is essential because electrical performance must be achieved without losing the advantages associated with the polymer matrix. The extensive use

of Ag nanostructures in electronic and plasmonic applications demonstrates the technological significance of controlling their nanoscale morphology and connectivity (Rycenga et al., 2011).

Electromagnetic interference shielding represents another possible application. Conductive metallic networks can reflect and attenuate electromagnetic radiation, while polymer matrices provide low density and processability. Ag-containing polymer systems can therefore offer an alternative to conventional heavy metallic shielding materials, particularly when sufficient particle connectivity is established (Geetha et al., 2009).

The enhanced interfacial polarization obtainable at appropriate Ag concentrations also creates opportunities in dielectric and energy-storage applications. Polymer nanocomposites with increased permittivity are of interest for capacitor technologies, although dielectric loss and electrical breakdown remain critical considerations. The desired condition is therefore not necessarily the maximum Ag concentration but a composition below or around the region where interfacial polarization is enhanced without excessive leakage conduction (Barber et al., 2009; Dang et al., 2012).

PEO–Ag films may also be relevant to sensing technologies because conductivity and dielectric response are sensitive to changes in nanoparticle separation, polymer structure, and the surrounding environment. A system operating near a concentration-dependent transport transition can show substantial electrical changes in response to relatively small structural perturbations, providing a potential basis for responsive sensing materials.

Future studies should therefore focus not only on nominal Ag concentration but also on nanoparticle size, distribution, interface chemistry, film thickness, and deposition conditions. A particularly important objective is to experimentally identify the concentration at which the system changes from isolated-particle behavior to tunneling-dominated transport and finally to percolative conduction. Correlating XRD, FTIR, SEM/AFM, dielectric spectroscopy, electric-modulus analysis, and conductivity measurements across the same concentration series would provide a more complete understanding of this transition.

Overall, silver concentration provides a powerful means of tuning PEO–Ag nanocomposite thin films. Increasing Ag content progressively modifies polymer structure, interfacial polarization, dielectric relaxation, and charge-carrier pathways. The most technologically useful composition depends on the intended function: moderate Ag loading may be preferable for dielectric applications, whereas concentrations close to or above the percolation threshold may be required for conductive applications. Careful control of Ag concentration during PVD is therefore central to tailoring PEO–Ag films for future electronic, dielectric, sensing, and energy-related technologies.

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