Chapter

Synthesis, Dielectric and Electrical Properties of Silver-Polymer Nanocomposites

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Abstract

Metallic nanoparticles and its composites have emerged as valuable asset in all phases of material science and engineering including electronic, optics and electromagnetic domains. Silver nanoparticles (Ag NPs) are one of the most vital and fascinating nanomaterials among several metallic nanoparticles due to its large surface ratio and outstanding properties with diverse field of potential applications. We demonstrated various synthesis techniques of nanocomposites, silver nanoparticles and composite based on these particles have shown great importance because of the remarkable properties (high electrical and thermal conductivity, good chemical stability and catalytic properties) of silver nanoparticles. This chapter provides various synthesis techniques for preparation of silver nanoparticles and their composites with dielectric and electrical properties in a lucid manner. The detail discussions of silver-polymer nanocomposites, emphasizing on each individual synthesis routes and properties have been carried out.

Keywords: Ag nanoparticles, composites, polymer manocomposites, properties

1. Introduction

In recent decade nanotechnology (nano signify very small that denotes to one billionth or 10^{-9} m in size) is a recognized as one of the most emerging fields of contemporary research deals with synthesis, manufacturing, strategy and tailoring of particle size approximately varying from 1 to 100 nm. The nanoparticles have unique magnetic, electronic and optical properties because of their high surface area to volume ratio and wide variety of applications including environmental health, optics, electronics; optoelectronics, catalysis and energy storage devices [1–6]. Nanoparticles possess small size; composition and shape have differences in their physical and chemical behaviors from their parent materials. Moreover, the smaller size of the nanomaterials also helps them to penetrate exact cellular locations and additional surface area facilitates increased absorption and targeted delivery of the substances [6–8]. A large number of nanomaterials have multitude of technological applicability in the field of engineering including electronics, biomedical, drug-gene delivery, environment, catalysis, light emitters, single electron transistors, non-linear optical or photo-electrochemical devices [9, 10]. The synthesis of nanomaterials by using chemical and physical methods is relatively

expensive and potentially hazardous to the environment due to the effect of toxic chemicals and it is responsible for various biological risks [11]. Nanomaterials have rised as appropriate alternatives to overcome the shortcomings of micro-composites and monolithics, while challenges exist related to preparation for controlling elemental composition and stoichiometry in the nanocluster phase. Engineered nanomaterials produced with nanoscale dimension are commonly grouped into four kinds: carbon, metal oxides, dendrimers and composites [12]. However, composites are another powerful tool for the progress of specific materials according to our requirements. Nanocomposites are composites in which at least one of the phases exhibit dimensions in the nanometer range (1 nm = 10^{-9} m) and also shows high performance material with unusual performance and unique design possibilities [13]. The nanoparticles based composite by using various polymeric matrices and fillers such as metal powders (conductive filler) have made immense interest from academia and industry [14, 15]. These nanocomposites show various performances like high strength, stiffness at elevated temperature, corrosion resistance, low weight, low maintenance cost and attractive thermal coefficient [16]. Besides, composites generally divided into two stages such as continuous phase which is commonly uses as matrix and the other phases such as reinforcement which is embedded into the matrix. Moreover, the unique combination of matrix materials including polymer, carbon, metal, ceramics and different types of the reinforcements such as fibers, particles and layered materials have been widely utilized for the fabrication of composites [17]. It has been reported that, there is change in particle properties when size of the particle is less than a particular level called critical size [13]. In addition, as dimensions reach the nanometer level, interactions at phase interfaces become largely improved and this is important to enhance material properties. In view of this, the surface area-volume ratio of reinforced materials employed in the preparation of nanocomposites is the crucial role to the understanding of their structure–property co-relations. Furthermore, the discovery (1991) of carbon material especially carbon nanotube (CNTs) [18] and their subsequent use to fabricate composites with improved mechanical, thermal and electrical properties [13, 19] added a new and attractive dimension in the field of nanotechnology. Currently, nanocomposites offer new technology and business opportunities for all areas of industry making environmental friendly [20].

The synthesis and characterization of composites using organic and inorganic nanomaterials have been extensively investigated area of research with improved functional properties and wide range of potential applications such as coating, packaging materials, sensors, energy storage, etc. [21, 22]. The measurement of various properties of the prepared composite materials mostly depends on the characteristics of the original materials. There is a variety of properties of the matrix materials, for instance it combines the dispersed phase mutually. It protects the dispersed phase from chemical action and maintain in suitable position and its direction [23]. It is revealed that the properties of the polymer composites associated with chemistry extend of polymer chain and thermoset cure can change from the interface between reinforcement and matrix. The silver nanoparticle based polymer composites can be used as biocompatible materials with improved antimicrobial activity as well as good electrical conductivity and catalytic properties [24–26]. The polymer based nanocomposites using nanoparticles have been successfully synthesized by various researchers via free radical thermal polymerization [27] and photo polymerization [28]. There are several attempts to synthesize Ag based polymer composites: For example; Nikfarjam *et al*. [26] have fabricated Ag nanoparticle filled poly(methylmethacrylate) based composites by using in-situ photoiniferter mediated photo polymerization technique with good thermal stability and enhanced mechanical properties.

Urged by scientific interest and potential application for the various green synthetic techniques of silver nanoparticle related polymer based composites research has increased to a surprising scale, opening new challenges and opportunity for the further modulation of properties. This typical chapter is primarily divided into two major sections. The first section covers various synthetic routes of silver (Ag) nanoparticles. The second section covers the synthesis, dielectric and electrical properties of silver-polymer nanocomposites in the field of energy storage devices. We end this chapter with a few words on this new and exciting research area of the Ag nanoparticles followed by summary and perspective.

2. Synthesis of Ag nanoparticles

The new synthesis strategy for the material fabrications are of essential importance of nanostructure material in the field of nanotechnology. The applications of nanomaterials are possible only when nano-structured materials are made available with required size, morphology, crystal, chemical composition and their unique properties in the field of various technological applications [29–31]. Silver nanoparticles (Ag NPs) have outstanding microbial resistant ability. These nanoparticles have variety of applications in our daily life including clothes, household and personal care products and mostly owing to their antimicrobial properties. Moreover, the silver based nanostructured materials with specific physical, chemical and optical properties, especially altering sizes and shapes have been widely used in the field of electronic devices, paints, coatings, soaps, detergents, etc. [32]. In the above discussion, the following details of Ag based nanomaterials are significant to consider in their synthesis methods such as surface property, particle composition, size distribution, morphology and different types of reducing and capping agents used. Generally, the methods used for the preparation of metallic Ag Nps are classified into two categories, namely top-down or bottom-up approach [33]. The top down approach involves bulk materials and decrease them into nano-sized particles by using physical/chemical and mechanical processes [34]. The top-down approach is also used for the fabrication of many materials including semiconductor industry [35], in this approach metal oxide semiconductor field effect transistor (MOSFET) are imprinted onto a silica wafer by lithography based technique [36]. On the other hand, the preparation in bottom-up method requires single atoms and molecules into larger nanostructures to achieve nano-size particles [34]. Currently, the synthetic methods are divided into physical, chemical and biological green syntheses. In this respect, the physical and chemical synthesis method tend to more serious and hazardous as compared to the biological synthesis of Ag nanoparticles. This shows outstanding properties including high yield, solubility and stability [37]. The following segment discuss different synthesis methods in detail of Ag nanoparticle and their mechanisms, explaining how shape and size controlled Ag Nps can be achieved by proper selection of precursor chemicals, reducing and capping agent as well as concentration and molar ratio of chemicals.

2.1 Physical methods

The physical synthesis of Ag nanoparticles involves various processes such as evaporation-condensation process and the laser ablation technique [38, 39]. This technique is to synthesize large amount of Ag nanoparticles with high purity without use of chemicals that release toxic substances and expose human health and environment. However, there is a great challenge of agglomeration of nanoparticles because it is not used in the capping agents. Besides, both methods consume more

power and require relatively longer duration of synthesis and difficult equipment, all of which increase their operating cost. The evaporation-condensation method commonly uses a gas phase technique that utilizes a tube furnace to synthesize nanospheres at atmospheric pressure. A variety of nanospheres using several materials including Au, Ag and PbS have been prepared by this method [40]. The centre of the tube furnace comprises a vessel carrying a base metal source which is evaporated into the carrier gas, permitting the final of nanoparticles. The shape, size and yield of the nanoparticles can be controlled by changing the plan of reaction facilities. Further, the synthesis of Ag nanoparticles by evaporation-condensation technique through tube furnace has various drawbacks. In this technique, the tube furnace occupies a huge space, consumes high energy elevating the surrounding temperature of the metal source and gives a longer period to maintain its thermal stability. To overcome these difficulties, Jung *et al.* [41] have synthesized Ag nanoparticles with higher concentration using ceramic heater and also utilizes in the efficiency.

Furthermore, another crucial approach for physical synthesis of Ag nanoparticle using laser ablation technique. The preparation of Ag nanoparticles through laser ablation of a bulk metal source placed in a liquid environment. Once irradiating with a pulse laser, the liquid environment may contain Ag nanoparticle of the base metal source, cleared from other compounds, ions or reducing agents [42]. However, there are various factors like laser power, duration of irradiation, type of base metal source and property of liquid media manipulate the features of the metal nanoparticles produced. Consequently, different chemical synthesis, the preparation of nanoparticles via laser ablation technique is pure and uncontaminated and in this method it uses mild surfactants in the solvent without using any other chemical reagents [43].

2.2 Chemical method

The synthesis of silver (Ag) nanoparticles by chemical method is the most commonly used technique and it is stable, colloidal dispersions in water or other appropriate organic solvents. The chemical process for synthesis of Ag nanoparticles in solution comprises the three major behaviors: (i) metal precursor, (ii) reducing agent and (iii) stabilizing or capping agent. The Ag nanoparticles are mainly chemically synthesized through Brust-Schiffrin synthesis (BSS) or the Turkevich method [43–46]. In order to achieve specific shape, size and various optical properties of the metal nanoparticles, it is essential to control the reducing agents and stabilizers are also taken into consideration. The uses of stabilizing agent during the preparation of metal nanoparticles are typically for avoiding aggregation [47]. So therefore, the following factors are needed to be considered for the safety and effectiveness of this technique, which includes the choice of appropriate solvent, use of environment friendly reducing agent and selection of non-toxic substances. There are various reducing agents used for the preparation of nanoparticles such as $NaBH_4$, N_2H_4, tri-sodium citrate (TSC), sodium citrate and N,N-dimethylformamide (DMF). Besides, in order to avoid aggregation between Ag nanoparticles, surfactant can be used such as sodium dodecyl sulphate (SDS), oleylamine and some polymeric materials like polyvinylpyrrolidone (PVP), polymethacrylic acid, polyethylene glycol (PEG) and polymethylmethacrylate have been reported to be the efficient protecting agents to stabilize as capping agents for stabilization [48–50]. This stabilization or capping agents can be accomplishing either through electrostatic or steric repulsion. For example, electrostatic stabilization is generally reached through anionic species like citrate, halide, carboxylates that adsorb or interact with Ag nanoparticles to impart a negative charge on the Ag nanoparticle surfaces. Thus,

the surface charge of Ag nanoparticles can be controlled by coating with citrate ions to give a strong negative charge. Further, the polyethylene glycol (PEG) coated nanoparticles show good stability in highly concentrated salt solution, whereas lipoic acid coated particles with carboxylic group can also be used for bio conjugation. Meanwhile, the morphology of nanoparticles is strongly manipulated by the temperature variant were implemented during the preparation. The Ag nanoparticles exhibit much deviation in shape ranging from spherical to trigonal/hexagonal. It is also reported that ascorbic acid used as reducing agent for the formation of room temperature flower like silver nano architecture with average particle size is about 20 nm. Consequently, Ag nanoparticles have been synthesized by the polyol process with the support of supercritical carbon dioxide from nitrate salt of silver as the base material, polyvinylpyrrolidone (PVP) as the stabilizer for the silver clusters and ethylene glycol act as the reducing agent and solvent. However, polyvinylpyrrolidone (PVP) not only protected the nanosized silver particles from aggregation but also help nucleation phenomenon [51, 52].

2.3 Photochemical method

The Ag nanoparticle was successfully synthesized by using photo irradiation technique. In this technique, photo assisted synthesis of Ag used for the preparation of stable Ag nanoparticles by irradiation of a reaction mixture with a light source including laser or lamp in presence of photo reducing agents without introducing stabilizers or surfactants [53–55]. For instance, the laser irradiation of an aqueous solution of Ag salt and surfactant can made stable homogeneous dispersion of Ag nanoparticles with good distribution of shape and size of the particles. However, the syntheses of silver nanoparticles with narrow size distribution through ethylene glycol-water solvent system without use of a stabilizer. Further, it is also observed that Ag nanoparticles are synthesized by using per chlorate salt via pulse radiography technique. In this technique, there is a reduction of Ag^{+} to Ag^{0} was accomplished successfully using UV light as a substitute of chemical materials in a rubber matrix using photo reduction of film cast from natural rubber latex (NRL) comprising silver salt with an average size about 10 nm. It is also reported that other synthesis route such as microwave irradiation have also been utilized and this route is of much faster rate than that of the conventional heating via conduction and convection. Their size and preparation time of the nanoparticles is directly proportional to the irradiation power of the source of light [56]. In addition, photochemical processes also suggest a reasonable potential synthesis of controlled shape and size of Ag nanoparticles while multiple preparation steps might be required.

3. Synthesis of polymer-silver nanocomposites

3.1 In-situ polymerization

In-situ polymerization is a very efficient technique for the carbon based conductive fillers to be dispersed homogeneously in the polymer matrix, so it gives a strong interaction between the matrix and the filler particles. However, in-situ polymerization technique normally involves the addition of nanoparticle in a pristine monomer or a solution of monomer through polymerization in the presence of nanomaterials [57]. Many studies have been made to synthesize nanocomposites using in-situ polymerization techniques and also showed that the covalent linkages between the matrix and nanomaterials. Besides, the fabrication of silver nanoparticle is relatively simple, efficient process by using in-situ technique. It is a one step

process for manufacture of nanoparticles which uses the corresponding precursors for synthesis and these nanoparticles can be directly grown using this technique. The most significant benefit of this technique is that it avoids particle agglomeration and maintains homogeneous distribution of the particles in the polymer matrix at the same time but, the main shortcoming of this technique is the slight possibility of left un-reacted educts in path of the reaction. These results may be control on the properties of the final product. For instance: Li *et al.* [58] have prepared polyvinylpyrrolidone (PVP) encapsulated stable Ag nanofluids with various concentrations of Ag via microwave synthesis technique. It is observed that the stable nanofluid comprising Ag nanoparticles of 30 to 60 nm and it is suitable for large scale production of nanofluids. Zhao *et al*. [59] have fabricated Ag deposited cellulose nanocomposites by using microwave assisted technique. It is low cost production and large scale production of prepared cellulose based nanocomposites. Singh *et al.* [60] have synthesized cellulose-Ag nanocomposites by using microwave assisted method and these nanocomposites have superb antimicrobial properties.

3.2 Ex-situ polymerization

Another important technique for preparation of nanoparticles based polymer composites by using ex-situ polymerization. This technique is more appropriate for large scale industrial applications. The main challenges of this method are preparation of nanoparticles with good homogeneity into the polymer matrix and have good thermal stability against aggregation [61]. In order to solve these difficulties sonication techniques were used to disperse the nanoparticles in the polymer matrix. For instance: Dhibar *et al.* [62] synthesized silver deposited polypyrrrole-graphene (Ag-PPy-Gr) nanocomposites used as an excellent candidate in the field of super capacitor applications [62]. In this work, it is observed that the graphene sheets are well uniformly coated in the polypyrrrole surface in the presence of silver nanoparticles. However, the specific capacitance of prepared Ag-PPy-Gr nanocomposites is 472 F/g at a current density 0.5 A/g. The development of these nanocomposites has enhanced electrochemical properties with the presence of both graphene and silver nanoparticles. In the ex-situ polymerization technique the uniform dispersion of nanoparticles in the polymer matrix becomes one important factor for preparation of transparent hybrid nanocomposites.

4. Different properties of polymer-silver nanocomposites

4.1 Dielectric properties

The dielectric properties of poly (vinylidene fluoride) (PVDF) based composites filled with nanosilver (nAg) deposited nickelate (Mg doped $La_{1.9}Sr_{0.1}NiO_4$) particles, which was reported by Thongbai *et al.* [63]. The composites (**Figure 1 (a,b)**) possessed high dielectric permittivity (ε') 62 and simultaneously achieved low dielectric loss (tan δ) of 0.027 at particular frequency region. Thus, these Ag-MLSNO/PVDF composites of greater dielectric properties with good mechanical flexibility and used in high technology future applications [63]. The nAg-MLNSO-PVDF composite were more thermally stable than that of the neat PVDF and it results these polymer composite systems has brilliant flexibility with lower concentration of fillers. These flexible dielectric polymer composites might be used in this modern period of electronic applications.

Zeng *et al.* [21] have reported silver nanoparticle-modified alumina microsphere hybrid composites for enhanced energy density and thermal conductivity. This

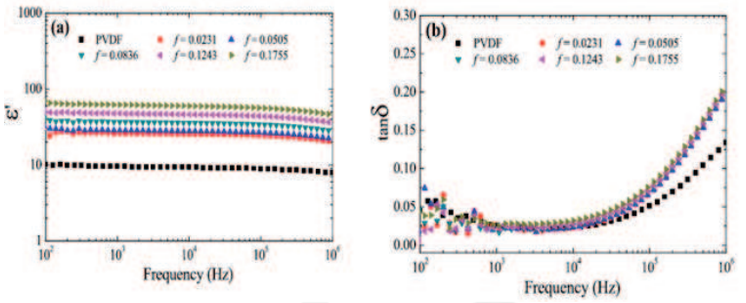

Figure 1.
Frequency dependence of (a) dielectric permittivity (ε') and (b) dielectric loss (tan δ) of nAg-MLSNO-PVDF composites. Reprinted with permission from ref. [63].

report tells us about the dielectric polymer composites which consisted of epoxy resins and silver nanoparticles decorated Al_2O_3 microsphere (Al_2O_3-AgNPs) with improved energy storage density and thermal conductivity. The dielectric loss of pure epoxy resins and Al_2O_3 -epoxy composites increases with increase in frequency. Moreover, the silver nanoparticle which is helpful to obtain Al_2O_3-AgNPs-epoxy resin composites with enhances the interfacial polarization and also leads to the high dielectric constant. For getting low dielectric loss and high breakdown strengths of the composite systems, the silver nanoparticles can be prevented from connecting into conductive pathways and is helpful for obtaining the polymer composites with high dielectric loss and high breakdown strengths [21]. It is observed that dielectric constant increases with Al_2O_3 AgNPs. With the increase in Al_2O_3 microsphere content, the motion of epoxy chain segments would be reserved and confined in the near vicinity, and the steric hindrance effect reduces the corresponding loss.

The enhancement of the thermal conductivity for Ag deposited alumina sphere through interfacial thermal resistance has been reported by Sun *et al.* [64]. In this work, the epoxy resin and commercial Al_2O_3 sphere was chosen as thermally conductive filler. The influence of silver deposition on the out-of-plane thermal conductivity of the composites was investigated. In this study, it is found that the composite with out-of-plane conductivity has increased with the increase in the Ag-deposited Al_2O_3 filler contents. The thermal conductivities of Al_2O_3-AgNPs-epoxy composites (**Figure 2(a)**) continuously improved [1.304 Wm^{-1} K^{-1}] until Ag NP content of 1.96 wt% and then decrease with increase in the filler content. This increase may be due to the bridge of Ag between the filler particles facilitate the heat flow through interfacial boundary, which in turn moderates the interfacial

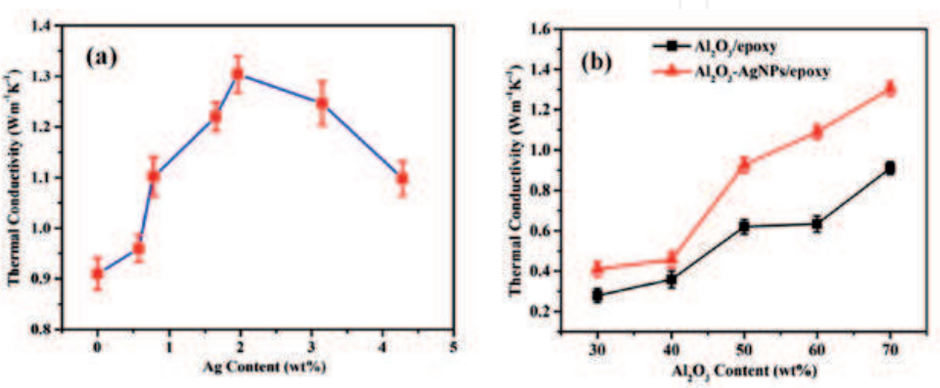

Figure 2.
Thermal conductivity of (a) Al_2O_3-Ag NPs-epoxy composites as a function of Ag contents, (b) variation of thermal conductivity of Al_2O_3-epoxy and Al2O3-AgNP-epoxy composites. Reprinted with permission from ref. [64].

thermal resistance and also help to improve thermal conductivity (**Figure 2(b)**) of the resultant composites [64, 65]. This method is helpful in the designing of thermally conductive polymer composites and can be used in the future generation electronic packaging applications.

Wu *et al.* [22] have reported about dielectric properties and thermal conductivity of the quantum-sized Ag decorated core or polydopamine (PDA)-epoxy based composites. The dopamine molecules are coated and attached to the surface of organic or inorganic matter which forms into supra-molecular polymer chain through the interaction of covalent bond and non-covalent bond [22]. However, the PDA is considerably improved the interfacial interaction between filler particles and polymer matrices, resulting in higher dielectric permittivity and breakdown strength [66, 67]. In this work they have found that, the composite has a low dielectric loss, high resistance and breakdown strength due to the Coulomb barrier formed by the Ag nanoparticles in the polymer. The novel core shell gray powder (AO*) sphere was prepared by the surface treatment of raw AO powder using dopamine. The polydopamine (PDA) shell helps to improve the interaction between AO fillers and epoxy matrix, which shows increase in dielectric permittivity and breakdown strength of the composites. Moreover, the strawberry like core shells AO* in Ag nanoparticles were prepared by hydrolysing silver nitrate and reducing the silver ion. The amalgamation of strawberry based core shell particles showed improved thermo mechanical, thermo stability and storage modulus. The AO* in Ag nanoparticles or epoxy composites indicates higher thermal conductivity values than that of the AO* or epoxy. It is accredited to the exceptional intrinsic thermal conductivity of Ag, mainly contributed from electronic transfer and phonon lattice vibration. Further, the enhanced dielectric, electromechanical and hydrophobic behaviors of core-shell Ag nano-wires (Ag NWs) in SiO_2 filler into polydimethylsiloxane (PDMS) based composites were prepared by sol gel technique and it is reported by Zhang *et al*. [68] and his co-workers. The polydimethylsiloxane (PDMS) is act as a polymer resin which is used in the field of insulators, coatings, medical devices, actuators and adhesives. Because it is due to the unique properties such as high electric insulation, good mechanical flexibility, mechanical stability under different ranges of temperature and high response speed [69].

Zang *et al.* [70] have reported that excellent energy storage density and efficiency in blend polymer poly (vinylidene fluoride) (PVDF) based composites by design of core-shell structured inorganic fibers and sandwich structured films. The dielectric material having great storage density is estimated to be obtained to minimize the size and weight of the dielectric capacitors, which is helpful in the pulsed power system and electric vehicles [70, 71]. Here they have taken into account the energy storage density and the energy storage efficiency for the polymer based composites using silver as nanofillers. These composites exhibited high polarization, low breakdown strength and high remnant polarization, which limits its energy storage density and efficiency [71, 72]. The silver (Ag) nanoparticle encapsulated 0.5Ba $(Zr_{0.2}Ti_{0.8})$ O_3-0.5 $(Ba_{0.7}Ca_{0.3})TiO_3$ (BCZT)- PVDF composites with improved dielectric constant. Further, the Al_2O_3 fibers outer layer not only discharge the dielectric difference between the BCTZ and polymer matrix, which will weaken the distortion of electric field but also confines the mobility of electronics by using Ag particles and the interfacial charges. In this study both energy storage density and efficiency have been improved with the sandwich-structured PVDF films. Luo *et al.* [73] have been synthesized an Ag deposited $BaTiO_3$ hybrid and fabricated BT-Ag-PVDF composites. The effect of Ag content on the dielectric properties of the composites were analyzed based on the diffused electrical double layer theory. The experimental results revealed that, the dielectric constant of BT-Ag-PVDF composites significantly enhanced

with the increase in the filler content about 613 (61 wt%) and reduced the dielectric loss of about 0.29 at 1 KHz. Furthermore, It was revealed that significantly enhancement of dielectric constant appeared when the inter particle distance decreased to a critical value of about 20 nm [73]. The facile method for the synthesis of Ag nanoparticles was successfully reinforced into the epoxy matrix *via* in-situ photochemical method. The Al-epoxy and Al-Ag-epoxy composites were prepared by mixing methods. The result showed that the dielectric constant of the Al-Ag-epoxy composite increases as compared to the Al-epoxy composites with simultaneously achieved negligible dielectric loss. Thus, these metal-polymer nanocomposites by in-situ photochemical method can be applied for the development of high-K polymer matrix to various fillers of host [74]. However, it is reported that the composites with high dielectric constant of core shell Ag@TiO_2 fillers successfully embedded into the polytetrafluoroethylene (PTFE) matrix, which provides the thickness of cell about 8–10 nm. It is observed that the composite of Ag@TiO_2 with rutile shells containing high dielectric constant than the anatase shells of Ag@TiO_2. The resultant dielectric constant maximum up to 200 at 100 Hz, which was 100 times higher than the pristine PTFE matrix with well suppressed dielectric loss of 0.005. Thus, the composites have potential applications in embedded devices [75].

Kuang *et al.* have been reported core shell Ag@C nanoparticles with diameter of 100–120 nm silver shell and 60–80 nm of carbon shell by hydrothermal method. The composite of Ag@C-PVDF composites was synthesized by solution casting method. The result of the composite illustrated that remarkably higher dielectric constant with low loss of 0.08 at 1000 Hz as compared to neat PVDF matrix due to the effect of the interfacial polarization. Furthermore, the permittivity and the tan δ of the composites increased with the increase of temperature [76]. The facile method for the synthesis of nanocomposites films of PVA reinforced into the silver nanoparticles and graphene oxide (GO) via solvent casting technique. It is observed that the composites have decrease in the real and imaginary part of impedance along with bulk resistivity on increasing GO content. Thus, on account of these above results of the GO-Ag-PVA nanocomposites can be employed as better promising candidate in the charge storage devices applications [77]. Xu et al. [78] have been demonstrated a new approach of ternary P(VDF-HFP) composites of Ag nanoparticles was decorated on TiO_2 nanowires with various aspects ratio *via* hydrothermal process. The result showed that the composites (**Figure 3(a,b)**) containing high aspect ratio of TiO_2 nanowires (i.e., pa-TiO_2) exhibits high dielectric constant (33.4) with simultaneously well suppressed dielectric loss of 0.023 at 100 Hz, which was 42.1% higher than that of Ag/pr-TiO_2/P(VDF-HFP) composites and also 227% greater than the pristine P (VDF-HFP) matrix. Hence, this method

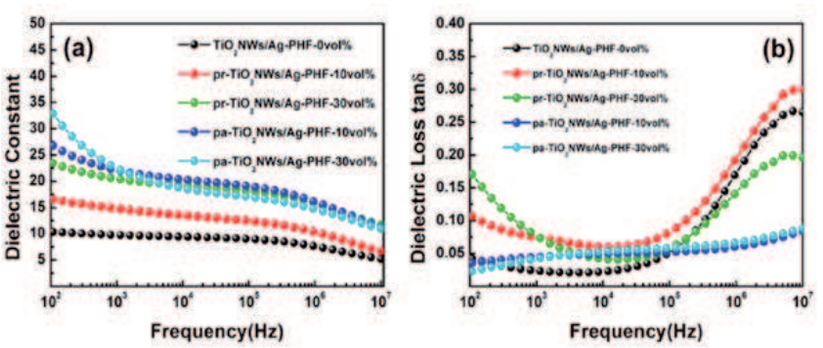

Figure 3.
Frequency dependence of (a) dielectric constant and (b) dielectric loss (tan δ) of Ag-TiO_2-P(VDF –HFP) ternary composites. Reprinted with permission from ref. [78].

could be provide a green route for the fabrication of high performance polymer composites for electric and electronic storage devices applications [78].

Phromviyo *et al.* [79] has been provided Ag@Ale–NPs composites were successfully fabricated on PVDF matrix via hydrothermal process using *Aloe vera* plant extracted solution as surface stabilizer and reducing agent. The composite of 0.18 volume fraction Ag@Ale-NP content exhibited high dielectric permittivity of 92.5 with low loss tangent of 0.049 at 1 KHz at room temperature. However, with the increase in filler content up to 0.22 the dielectric permittivity dramatically enhanced of ≈ 257.2 with very low loss of 0.26. Thus, the Ag@Ale-NPs-PVDF nanocomposites have greatly improved dielectric permittivity, due to the interfacial polarization effect and also the formation of micro-capacitor in the PVDF matrix [79]. Ghosh *et al.* have been prepared a CCTO ceramic fillers were prepared by sol–gel method and they successfully decorated by Ag nanoparticles embedded into the PVDF matrix to formed P/C-Ag: 30 composites via modified seeding method. These composites have high dielectric constant and negligible dielectric loss, which was 20% higher than that of the unmodified P/C: 30 composites. Thus, these types of work could be provided a better promising candidate of high performance polymer composites for energy storage applications [80].

The enhancement of both dielectric constant and breakdown strength of the modified Ag-OMMT nanoparticles incorporated in to the P(VDF-HFP) matrix to form P(VDF-HFP)-Ag-OMMT composites. However, the dielectric constant and breakdown strength of the P(VDF-HFP)-Ag-OMMT composites significantly increased with 4 vol% of Ag-OMMT contents and showed an energy density of 10.51 J cm^{-3} at 400 MV m^{-1}, which is ~2.25 times higher than that of pristine P(VDF-HFP) film. Thus, it is a simple and facile method to increase the energy density of polymer dielectric films by the addition of very low loading hybrid fillers [81]. Zou *et al.* [82] have been demonstrated silver (Ag) nanoparticles were dispersed homogeneously in P(VDF-TrFE) matrix to fabricate the composites by solvent casting method. The results showed that the enhancement of ferroelectric and dielectric properties of the composites with an increase in 38% of remnant polarization and 47% of dielectric constant than that of the pure P(VDF-TrFE) films [82]. Wang *et al.* have synthesized a PVDF based nanocomposites of Ag particles embedded in to the $BaTiO_3$ nanofillers via coaxial electro spinning technology followed by calcinations. The result indicated that, the composite of PVDF-Ag@BT exhibits high dielectric constant of 38.87 with well suppressed dielectric loss of 0.02 at 1 KHz. This remarkably enhancement of the dielectric constant may be due to the uniform distribution of the Ag nanoparticles on to the BT surface and the interfacial polarization effect. Furthermore, the Ag nanoparticles on the $BaTiO_3$ nanofibers exhibit the Coulomb blockade effect in the composites, which inhibits the electron migration and suppress the dielectric loss for better electronics applications [83].

The new strategy of core–shell graphene@polydopamine-Ag nanoplatelets and corresponding thermoplastic polyurethane (TPU)/Gns@PDA-Ag nanocomposites was reported by Zhu *et al.* [84]. This core shell nanocomposites were demonstrated by using dopamine oxidation polymerization and chemical reduction of silver nitride. The finding results indicates that, the dielectric constant for the 4 wt% of the TPU-Gns@PDA-Ag nanocomposites was 117.81, which was about 14 times greater than that of the pristine TPU matrix. Meanwhile, PDA shell not only promoted the homogeneous dispersion of Gns in the TPU matrix, but also enhanced the interface bonding between the nanoplatelets and TPU matrix. However, the Ag nanoparticles were uniformly embedded on the PDA shell, and the Coulomb blocking effect was successfully introduced into the composite. As a result, TPU-Gns@PDA-Ag nanocomposites exhibits low dielectric loss and high breakdown field strength compares with TPU-Gns nanocomposites. Thus, the obtained TPU-Gns@

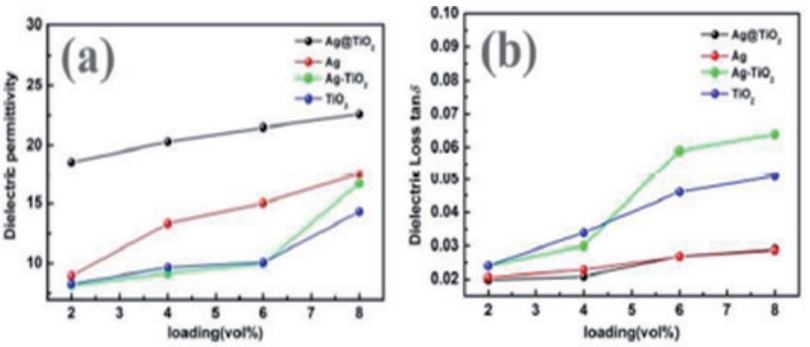

Figure 4.
Comparison of (a) dielectric permittivity and (b) dielectric loss of P(VDF-HFP)-Ag-TiO2 composites. Reprinted with permission from ref. [85].

PDA-Ag composite shows a better material for the potential applications in the field of energy storage [84]. Xiao *et al.* [85] have prepared the filler of Ag@TiO_2 core shell nanoparticles by vapor-thermal method and successfully embedded into the P(VDF-HFP) matrix to formed the P(VDF-HFP)-Ag@TiO_2 composites. The results revealed that the composite of P(VDF-HFP)-Ag@TiO_2 (**Figure 4(a,b)**) exhibits high dielectric constant (≈36) at 13 vol% of filler loading which was 4 times larger than that of the pristine P(VDF-HFP) matrix and well suppressed dielectric loss 0.03 at 1 KHz due to the effect of duplex polarization. Thus, this facile method of composites fabrication could be used as a smart functional material for embedded devices in the field of electronic industries [85].

Yang *et al.* reported the ternary composites system of polyimide (PI) embedded $CaCu_3Ti_4O_{12}$-Ag nanoparticles (CCTO@Ag) to fabricate PI-CCTO@Ag composites. The results showed that the Ag nanoparticle functionalized CCTO-PI composites having high dielectric constant value about 103 at 100 Hz at 3 vol% of filler content. This high dielectric constant based composite materials have well agreed with the percolation theory, space charge polarization and Maxwell-Wagner-Sillars effect. Furthermore, the dielectric loss value was reduced about 0.018 at 100 Hz due to blockage of charge transfer by insulating polyimide chains. Thus, the easy process ability, good flexibility and excellent dielectric properties of the composite have drawn potential applications in the field of charge-storage capacitors and embedded devices in electronic industry [86]. Chen *et al*. have been synthesized Ag-TiO_2-core shell nanoparticles (NPs) modified by two surfactants, i.e. octyl phosphonic acid (OPA) and pentafluorobenzyl phosphonic acid (PFBPA) reinforced polytetrafluorooethylene (PTFE) based composites. Thus, the results showed that the nanocomposites by PFBPA exhibits higher permittivity than the OPA because of its uniform dispersion stability of inorganic fillers into the PTFE polymer matrix and results increase the interfacial polarization effect. Therefore, nanocomposites could be used as better promising materials for applications of embedded devices and high frequency fields in electronics [87].

Zhu *et al.* [88] have prepared a flexible polymer composite by using solution casting method with core-shell nanostructure (Ag cores coated by SiO_2 shell) and incorporating P(VDF-TrFE) matrix. The Ag@SiO_2 nanoparticles were prepared by a simple Stober method. The resultant composite showed increased in dielectric constant with lower dielectric loss along with improved breakdown field of 10 wt% and thickness of the SiO_2 shell about 10 and 20 nm. The enhancement of the dielectric performance suggested a promising way to fabricate composites for potential applications in electronic devices [66, 67, 88]. Fang *et al.* [89] have been reported BT-Ag hybrid particles with variation of Ag mass fractions of 0.31, 0.49, and 0.61 were synthesized by chemically depositing nano Ag on the surface of 100 nm BT

particles. The results showed that the enhancement of the dielectric permittivity of the BT-Ag-PVDF (0.31, 0.49, 0.61 wt% of filler contents) composites exhibits 283, 350, and 783, respectively. On the other hand, the dielectric loss about <1 at 100 Hz, which is approximately three times higher than that of the untreated BT nanoparticles and it may be attributed to the low conductivity of the composites. Thus this method provided a way to fabricate BT-Ag hybrid particles into the polymer matrix for desired dielectric performance. Furthermore, the synthesized composites have attractive for potential applications in the high density packing technology and electronic systems [83, 84, 89]. Similarly, Wageh *et al*. [90] have successfully demonstrated a nano-silver decorated on the surface of the reduced graphene oxide (Ag-RGO) and incorporated into the polar poly (vinylidene fluoride) (PVDF) matrix. The results of these composites (**Figure 5(a,b)**) have lower electrical conductivity as well as increase in dielectric constant of 97 at 1 kHz of 1.5 vol% of Ag-RGO contents, while at the same time maintain the low loss. This enhancement in dielectric constant can be attributed to interfacial polarization or Maxwell-Wagner-Sillars (MWS) effect [79]. Thus, these results of the Ag–RGO–PVDF composite systems have drawn great promise for use as dielectric material for embedded capacitor applications [90].

4.2 Electrical properties

The simple method for the synthesis of electrically conductive composites comprising silver nanoparticles decorated on the carbon nanotubes (CNT) (Ag@CNT) incorporated into the epoxy resins by using wet chemistry reaction. The obtained result of the composites showed that higher electrical conductivity 0.10 wt% of Ag@CNTs, which was four times higher than the pure and functionalized CNTs. Thus, this enhancement of electrical conductivity of the Ag@CNTs-epoxy based composites was used as a potential application in the field of electronic package industries [91]. Sahu *et al*. [92] have been prepared polyaniline (PANI) coated functionalized multiwalled carbon nanotubes (f-CNTs) and silver nanoparticles (AgNPs) (PANI@f-CNTs and PANI@AgNPs hybrid nanofillers) based polymethylmethacrylate (PMMA) composites by in-situ polymerization technique. It is observed that the AC electrical conductivity (σ_{ac}), dielectric permittivity and dielectric loss (tan δ) of the resultant nanocomposites have significantly enhanced. Although the real impedance decreases by the incorporation of the hybrid nanofillers into the PMMA matrix, which concludes that the synthesized polymeric materials have most promising dielectric application [92]. Yusof *et al*. [93] have been synthesized a MWNT-Ag hybrids via reducing aqueous silver salt with the

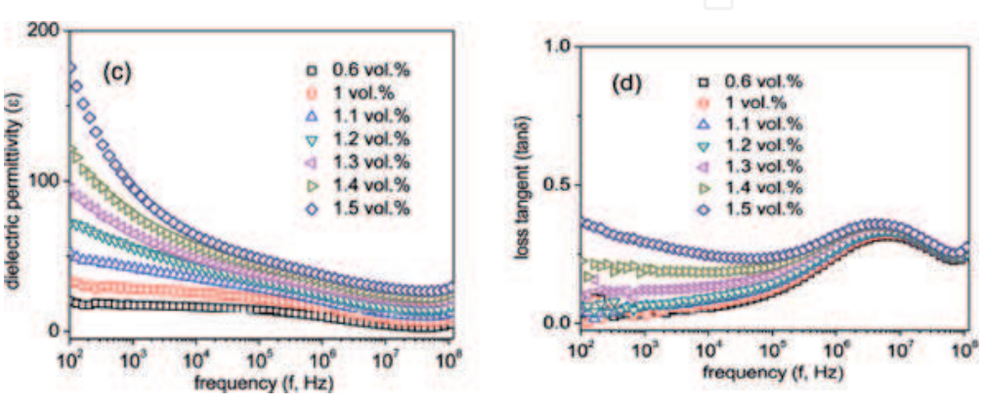

Figure 5.
Frequency dependence of (a) dielectric constant and (b) dielectric loss of Ag-RGO-PVDF composites with various percentage of filler contents. Reprinted with permission from ref. [90].

presence of SDS as stabilizing agent to enhance the deposition of the Ag NP on to the MWNT surface. The result obtained that MWNT-AgNP hybrids remarkably improved the structural, electrical and thermal properties, which could be a better material for the enhancement of electrical properties in the field-effect transistor (FET) applications at lower threshold voltage [93]. A facile and effective method of silver nanoparticles decorated on reduced graphene oxide (R-GO) was incorporated in to the PVDF matrix to formed Ag-RGO-PVDF composites via solution mixing method. The results indicated that the electrical conductivity of the composites increases at above the percolation threshold (0.17 vol %) due to the homogeneous dispersion of the Ag-RGO into the polymer matrix. Thus, this enhancement of electrical properties have drawn an attractive materials for antistatic, electrostatic dissipative and electromagnetic/radio frequency interference shielding applications. Furthermore, the resistivity of the composite system increased with increasing temperature, generating a pronounced positive temperature coefficient effect of resistivity [94]. Hussain *et al.* [95] have synthesized conducting PANI-Ag-MWCNTs composites comprising pure multi-walled carbon nanotubes (MWCNTs) and silver anchored with MWCNTs (Ag-MWCNTs) by using in situ polymerization approach. The electrical conductivity of the Ag-MWCNT (act as filler) is more strong interactions between PANI and Ag-MWCNTs and results improved electrical conductivity of the PANI. Thus, the PANI-Ag-MWCNTs composites were found to be the most stable composites [95]. Hsiao *et al*. [96] have prepared a silver nanoparticle deposited reduced graphene oxide-polyurethane (PU) composites (AgNP@RGO-PU) by electro spinning technique. The experimental result showed that the GO sheets can be attached to the PU surface through hydrogen bonding between GO and PU fibers. They also reported that the AgNPs deposited RGO-PU composites, which were modified by thiophenol through π-π interaction between the functional groups of AgNPs and RGO by dip-coating method. The 2 wt% of AgNP@RGO-PU composite (**Figure 6**) exhibited remarkably enhanced electrical properties (‘10 Ω/sq) [96]. Similarly, Nayak *et al.* [97] have reported expanded graphite (EG) decorated Ag nanoparticles using solid-state decomposition by a simple ‘mix and heat’ technique and prepared nanoparticles were deposited into the epoxy via stir casting method. It is revealed that the EG-Ag-epoxy composites with improved electrical conductivity. The EG-Ag composites exhibits better dispersion and lower porosity in the epoxy matrix which corresponding to high performance of thermal and electrical properties. Therefore, this method provided a green route to fabricate high

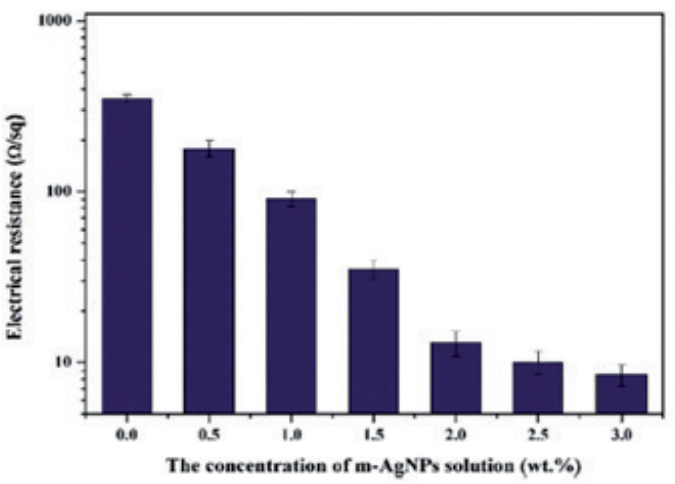

Figure 6.
Electrical resistance of AgNP@RGO-PU composites with various concentrations of filler content. Reprinted with permission from ref. [96].

efficiency and multifunctional epoxy based composite for advanced electronics and optoelectronics applications [97].

5. Conclusion

In summary, the syntheses of Ag nanoparticles have significant aspects of nanotechnology and it is used as nanofillers for fabricating nanocomposites. The Ag nanoparticle is a highly efficient, reliable high yielding and low cost technique. These nanoparticles have gained immense interest due to their unique physical and chemical properties as well as confirmed applicability in diverse fields such as electronics, catalysis, biotechnology and medicine. However, the shape and size distribution of silver nanoparticles can be controlled by adjusting reaction conditions such as reducing agent, stabilizing agent or using various synthesis techniques. The use of silver in the polymer based nanocomposites shows the enhancement in various properties such as dielectric and electrical in the field of energy storage devices. The Ag based nanocomposites have an enormous research interest in recent few times and potentially applicable in various fields especially in embedded high energy storage devices.

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Conflicts of interest

The authors declare no conflict of interest.

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