

Biodegradable Polymer Nanocomposites: A Review of Properties

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Abstract

Biodegradable polymers exhibit shortcomings, including low thermal stability and electrical conductivity. These challenges limit the broad applications of several applications, such as electronic devices. They show suitable dielectric, thermal, and electrical conductivity compared to the biodegradable polymer alone. Several methods can improve biodegradable polymers' dielectric, thermal, and electric conductivity, including co-polymerization, blending, and cross-linking with other polymers. Furthermore, the formation of nanocomposites seems to be the most effective method to improve the properties and performance of biodegradable polymers. This book chapter summarized biodegradable polymers' dielectric, thermal, and electrical conductivity. Biodegradable polymers nanocomposites consisting of polymers blend, inorganic, and other nanomaterials were discussed.

Keywords: Biodegradable polymers; Thermal conductivity; Dielectric; Nanocomposites.

1. Introduction

Polymers are multifunctional materials that can be alternatives to other materials such as metals, glass, and other traditional materials [1–5]. They can be pure organic [6–9] or hybrid materials including coordination polymers or metal-organic frameworks (MOFs) [10,11,20,12–19]. They offered various advantages such as being lightweight, flexible, and low cost. They are widely used for applications including electromagnetic interference shielding, biosensors, transient electronics skin (wearable) [21,22], wound dressings [23,24], tissue engineering [25], bone tissue scaffolds [26], self-healing strain sensor [27], sensors [28–30], optoelectronics [31], biomedicine[32–34], biotechnology [35–47], gene-delivery[48,49], environmental[50–58], and energy [11,16,56,58–71]. Polymers, including organic and coordination, are promising for future applications. In 1980, the term biodegradable polymers were introduced. Biodegradable polymers are widely applied for tissue engineering. Biodegradable polymers can be degraded into carbon dioxide (CO₂), water, biomass, and humus (dark organic matter in soil) using living organisms, photocatalysis, or catalysis. Biodegradation in a natural system is biologically benign using microorganisms or enzymes. The European standard of EN 13432 defined biodegradable materials as materials that can be degraded > 90% of their mass via biological environment for six months.

We introduce biodegradable polymers and the development of polymers nanocomposites. Here, we discuss biodegradable polymer nanocomposites' dielectric, thermal, and electrical conductivity. The synthesis of polymer nanocomposites exhibits high electric and thermal conductivity with good dielectric properties. The properties of polymers can be improved via conjugation with inorganic and organic materials.

2. Biodegradable polymers

The term “biodegradable” ensures products' reliability in the environment without harmful effects. The ASTM (e.g., ASTM D5511-18 and ASTM D5526-18, American Society for Testing and Materials) defined biodegradable materials as materials that exhibited 70% degradation during 30 days under anaerobic conditions[72,73]. Under aerobic conditions, ASTM D6400-19 and ASTM D6868-19 require 90% mineralization degradation of the material into CO₂ within 180 days[74,75]. Biodegradable polymers can be synthesized or extracted from different sources (**Figure 1**). They can be obtained from agro-resource biomass (i.e., agro-polymers). They can be extracted from microorganisms. They can be obtained from the conventional synthesis of bio-derived monomers. Petrochemicals are important sources of

monomers used for the synthesis of degradable polymers. They can be classified as (**Figure 1**):-

(1) Natural-based materials, e.g., polysaccharides (e.g., cellulose, lignin, starch, chitin/chitosan) or proteins (e.g., collagen, Silk fibril (SF)).

(2) Synthetic polymers, fossil oil, or petroleum-derived polymers e.g., poly(ϵ -caprolactone) (PCL) poly(lactic acid) (PLA), poly(butylsuccinate), poly(glycolic acid) (PGA), poly(ϵ -caprolactone) (PCL), Poly(vinyl alcohol) (PVA), Poly(vinylpyrrolidone) (PVP), polybutylene succinate (PBS), and poly(hydroxyl butyrate) (PHB)[76–78].

Natural polymers can be classified as polysaccharides, polyamides, and polynucleotides (**Figure 1**). They can also be listed as plant-based polysaccharides (e.g., cellulose, alginate, or starch) and animal-derived polymers (e.g., collagen, silk fibroin, or chitosan). They exhibit advantages, such as high biocompatibility, low toxicity, low cost, good mechanical properties, and high biodegradability. Several linkages ensure high biodegradability (**Figure 2**).

Cellulose has been considered the most abundant biopolymer in nature (**Figure 2**)[65,79–87]. It is a linear polysaccharide biopolymer consisting of β -1,4-linked D-glucose units. It can be extracted from several sources, such as trees and cotton. The chemical structure of cellulose shows a large number of hydroxyl ($-\text{OH}$) groups[88]. Cellulose can proceed into the nanoscale, such as nanofibers (CNFs) and nanocrystals (CNCs).

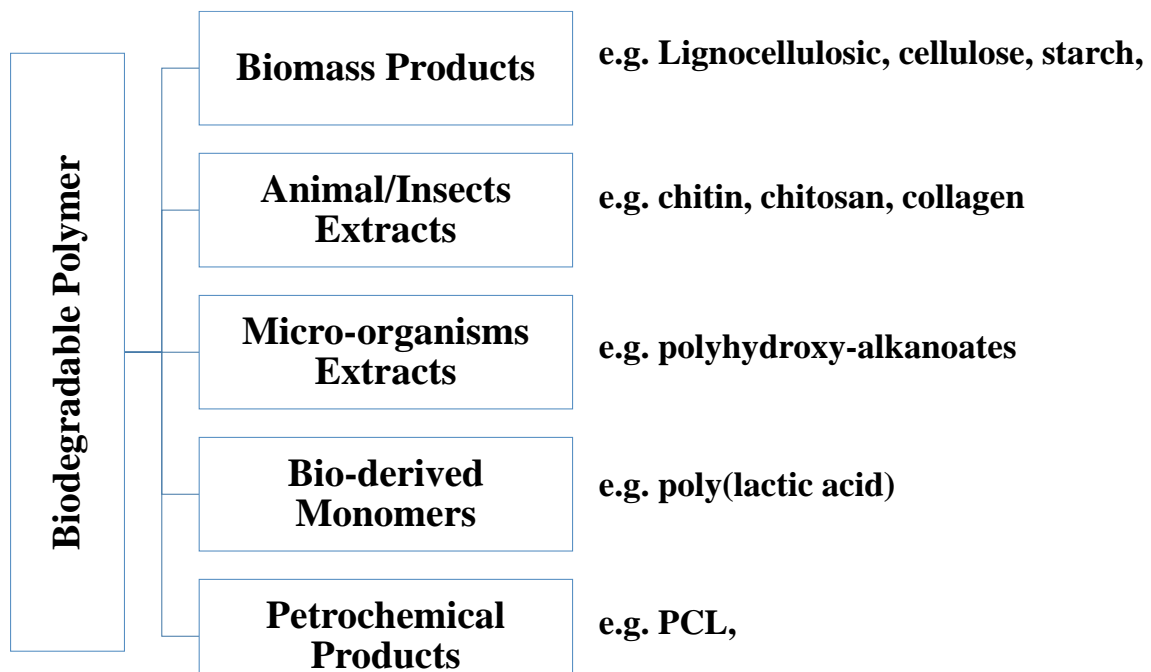


Figure 1. Biodegradable polymer sources.

Chitosan consists of randomly distributed β -(1-4)-linked D-glucosamine and N-acetyl-d-glucosamine[89–91]. It is the de-acetylated form of chitin offering amino groups. It can be dissolved in an acidic solution forming a cationic polysaccharide. It exhibits good adhesive properties with high antibacterial activity.

Synthetic polymers are usually extracted or synthesized via well-known reactions such as condensation or ring-opening polymerization (ROP). Several synthesis methods were reported for the synthesis of new organic polymers. Most of these methods require the use of a catalyst. The synthesis procedures aim to polymerize monomers with suitable functional groups that tend to polymerization. There are three main steps for polymerization: initiation, propagation, and termination. MOFs are coordination polymer formed via self-assembly of metal ions and organic linkers [64,66,69,92–98]. They can be decomposed into their constituent in harsh condition i.e. acid or base depending on the MOFs materials.

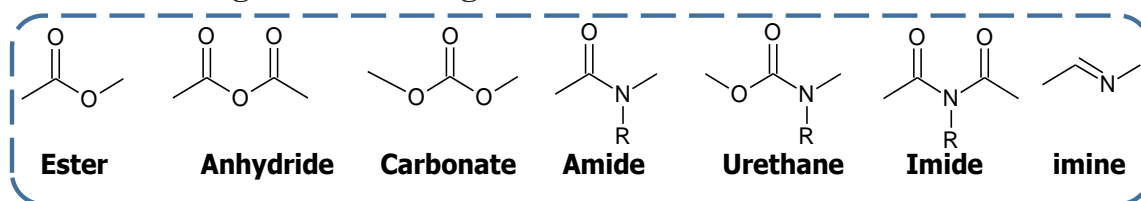
Aliphatic polyester polylactide (PLA) can be synthesized via ROP reaction of lactide using a catalyst such as tin (II) octoate. It can also be prepared via the condensation of lactic acid. It can also be extracted from corn or wheat. It displays good transparency with thermoplastic properties. It can be considered bioplastic. It is an everyday use for three-dimensional printing (3D printing)[78]. It can be degraded via hydrolysis, thermal decomposition, or photodegradation.

Like PLA, PCL can be obtained by ROP of ϵ -caprolactone [26]. United States Food and Drug Administration (US-FDA) approved PCL for biomedical applications such as tissue engineering. PCL exhibits high biocompatibility, high biodegradability, good chemical resistance, and high ductility. It displays a melting point of 65 °C with tunable viscosity enabling processing using several methods. However, it has high Young's modulus and strength.

A water-soluble polymer such as PVA can be synthesized via hydrolysis poly(vinyl acetate)[99]. It exhibits high transparency, high strength, good flexibility, and high biocompatibility. However, it has a high density of –OH groups; it is hard to shape via conventional processes such as melting methods. However, it can be easily blended with other materials via a mixing procedure. PVA-based materials were widely used for resistance random access memory [100–102]. PVP is another water-soluble polymer. It can be synthesized via the radical polymerization of N-vinylpyrrolidone [103]. PVP displays high

chemical resistance, easy processability, high transparency, good biocompatibility, and low cost. PVP was used for several applications, such as wearable electronic devices and gas sensors[104].

Common Biodegradable Linkages



Natural Biodegradable Polymers



Synthetic Biodegradable Polymers

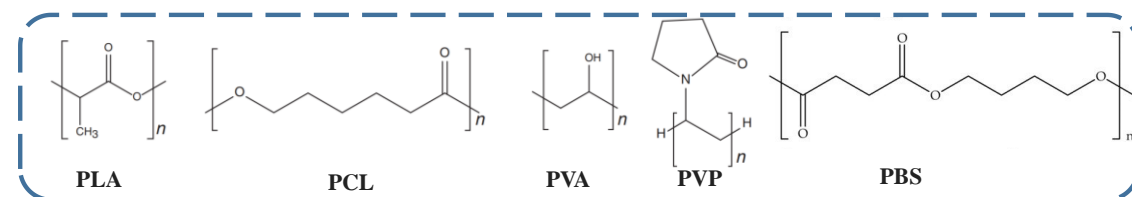


Figure 2 Common biodegradable linkage and chemical structure for common biodegradable polymers.

Natural polymers exhibit good intrinsic biocompatibility and enzymatic degradability, enabling intensive applications for biomedical applications (**Figure 2**). Biodegradable polymers are common clinical polymers, including polyglycolide, polylactides, and polycaprolactone. They can be classified as molecular, microstructural, and macroscopic. The properties of biodegradable polymers can be evaluated using UV–Vis spectroscopy, mass loss profiles, mass spectrometry, nuclear magnetic resonance (NMR), Fourier transform infrared (FT-IR) spectroscopy, Gel permeation chromatography (GPC), scanning electron microscopy (SEM), transmission electron microscopy (TEM), and atomic force microscopy (AFM).

The properties of polymers depend on several parameters, including chemical composition, particle size, morphology, and surface structure. Small particles polymers undergo aggregation into bulk materials, especially without proper stabilization. Nanoscale polymers have high

surface energy pushing the materials to aggregate to achieve high stability. Distributing the polymers uniformly in matrices can reduce or prevent the assembly.

The size of a polymer affects the properties of polymers. The morphology of the polymers can be spherical, rod, or plate, depending on the radius (R) and the thickness (t, **Figure 3**). Therefore, the ratio between volumes of interface material to the volume of the particle ($V_{\text{interface}}/V_{\text{particle}}$) increases with the decrease of particle size. The aspect ratio (the ratio of the diameter ($2r$) to the length (L)) determines the morphology of the polymers. Based on the aspect ratio, the morphology can be a plate, sphere, and rod with an aspect ratio of < 1 , 1 , and >1 , respectively (**Figure 3**).

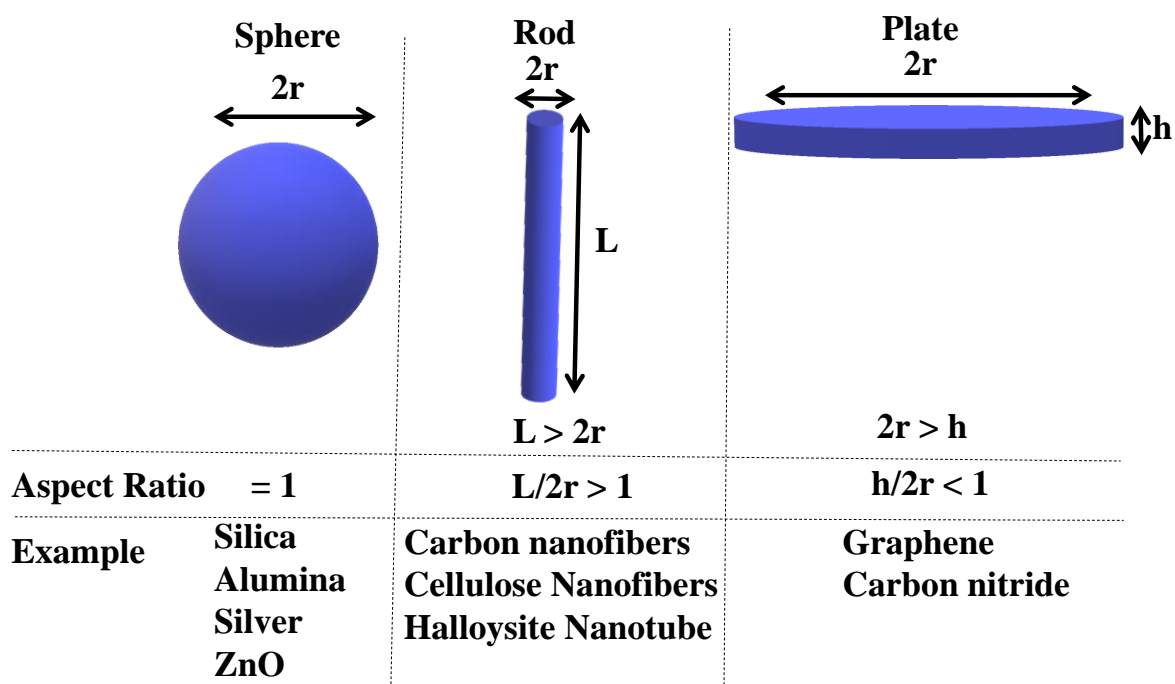


Figure 3. The morphology of polymers particles and their aspect ratio. R is the radius, L is length, and h is the height.

3. Polymers nanocomposites

The properties of the polymer can be significantly improved via several methods, including the addition of nanoparticles, i.e., the formation of nanocomposites. Polymers can be conjugated or blended with other materials to form composites. They can be conjugated with nanomaterials to form polymer nanocomposite (**Figure 4**). Polymer nanocomposites can generally be achieved using several methods such as:-

- (a) Solution method: this method depends on the dissolution in a suitable solvent in the presence of nanoparticles followed by evaporation of solvent or via precipitation.
- (b) Melt mixing: this method depends on the direct melt-mixed of polymer with the nanoparticle.
- (c) In-situ polymerization: this method involves the polymerization of monomers without nanoparticles.
- (d) Template synthesis: this method relies on using a template to synthesize nanoparticles.

Polymer nanocomposites (PNCs) are multiphase materials made of two or more materials containing a polymer and other materials, e.g., metal, ceramic, inorganic, non-metallic, and ionic liquids[105]. PNCs exhibit good physicochemical properties compared to individual components (**Figure 4**). Polymer nanocomposites are prepared via physical blending, dipping coating, casting, template molding, melt blending, mixing (via ultrasonication, shear, three-roll milling, ball milling), double-screw extrusion, and in-situ synthesis. Electro-hydrodynamic (EHD) method [106].

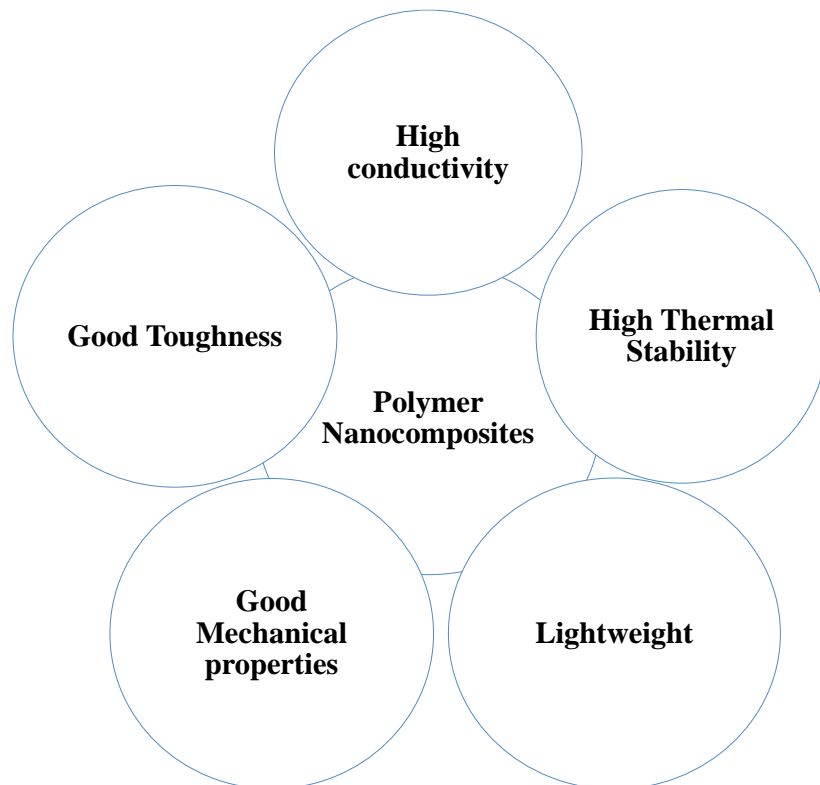


Figure 4. Advantages of polymer nanocomposite.

4. Dielectric Properties

The electrical insulator materials that can be polarized under an applied electric field are called dielectric materials. This material exhibits high electrical polarizability due to the accumulation of the electric charges on the dielectric materials under an electric field. The materials are insulators; thus, no flow of the electric charges can be performed through the material. The dielectric materials are not loosely bound or free, causing dielectric polarization that offers positive charges in the field's direction and negative charges that shift opposite the electric field. The material performance determines by a dielectric constant (k) that defines the material's storage capacity for the electrical charge. Materials with high- k values can be used as gate dielectrics in metal–oxide–silicon (MOS) transistors, memory cells, and supercapacitors. On the other hand, materials with low- k values are essential as electrical insulators.

The dielectric properties of polymers enable their applications for electronic skins (e-skins), capacitors, and dielectric resonator. A parallel plate capacitor exhibits a capacitance (C), defined as the ability to store a charge. The capacitance can be determined via the equation of $C = \epsilon A/d$, where ϵ , A , and d are the dielectric constant, the area, and the space between the two electrodes, respectively. A dielectric layer of polymer is sandwiched between the two electrodes. The dielectric properties depend on several parameters such as composition, size, porosity, thin-film thickness, defects, operating frequency, doping concentration, and atomic number[31].

Ion-gel of cellulose can be used as dielectric materials [107]. Cellulose such as CNC was reported as a flexible field-effect transistor (FET) [108]. CNC can serve as substrate and dielectric materials. However, cellulose-based devices need a large operating voltage. The performance of cellulose can be improved via several methods, including mixing cellulose derivatives such as cyanoethyl cellulose-containing barium strontium titanate (BST, $\text{Ba}_x\text{Sr}_{1-x}\text{TiO}_3$) nanoparticles[109]. The materials exhibit high dielectric properties, making them suitable for preparing ultralow-power electronics with a high on/off ratio and low operating voltage.

The thin film consisting of chitosan and Y_2O_3 nanoparticles exhibits a low current leakage current compared to pure chitosan. The electric and dielectric performance of chitosan can be improved via the impregnation of a high- K material (i.e., high dielectric constant)[40]. Similarly, silver nanoparticles (Ag NPs)/chitosan composite was used as a dielectric gate for

transparent resistive switching memory [110]. The composite offered high biocompatibility and required low-power operation conditions [110].

PVA has a dielectric constant of 5–8 in the 10–106 Hz range. It exhibits a higher dielectric constant than other biodegradable polymers [111,112]. An aqueous solution of PVA can be mixed with inorganic fillers materials such as Au NPs/graphene oxide (GO) [113], GO [114], and MoS₂ [115]. Inorganic nanoparticles improved the dielectric constant and strength of PVA films. A composite film of biodegradable poly(butylene succinate) and graphite exhibited a K-value of 113, which was 28 times higher than the pristine polymer matrix [116].

Various dielectric materials were mixed with biodegradable polymers such as PLA for electronic devices. PLA/TiO₂ was reported for the humidity sensor [117]. It can be used for the detection of humidity in the range of 20-90% [117]. PLA/barium titanate (BaTiO₃) nanowires (NWs) produce printable energy harvesters that can proceed using the 3D printing method [118]. BaTiO₃ nanowires can be well aligned, offering 273% higher power generation capacity than conventional cast nanocomposites with randomly oriented nanowires. Al₂O₃ improved the dielectric properties of cellulose acetate [119]. 3D printing was also reported for PLA/NiTi nanowires [120]. PLA/NiTi nanowires were reported as a sensor for measuring temperature and strain simultaneously [120]. Electrospinning of PLA/boron dye was reported to detect oxygen in real-time for tissue scaffold applications [121]. PLA/boron dye sensor can determine the low level of dissolved oxygen (< 15 ppm) in the scaffold [121]. Other materials, including cellulose nanocrystals (CNC)[122], clays [123], ZnO [124], SnO₂ [125], cobalt ferrites [126], nitride nanosheets /copper calcium titanate [127], gold nanoparticles [128], or PbS [129] were also reported to improve the dielectric properties of polymers. There are several mechanism for the dielectric improvement using these nanoparticles including the decrease in energy band gap [125].

Carbon nanomaterials such as carbon nanotubes improved the dielectric properties of commercially available polymers, Ecoflex[®] [130]. It was found that there is an increase in the dielectric value with the rise of the carbon nanotube's contents. Reduced graphene oxide (rGO) improved the dielectric properties of polyaniline (PANI)/calcium copper titanate (CaCu₃Ti₄O₁₂) [131]. It can be also improved PVP-PVA [132], and starch [133].

5. Thermal Properties

The thermal properties of polymers can be determined using several analytical techniques such as thermogravimetric analysis (TGA), differential thermal analysis (DTA), and differential

scanning calorimetry (DSC). TGA can be connected with other analytical instruments to characterize degradable production, such as TGA\FT-IR spectroscopy.

A composite of silica/carbon exhibited the thermal properties of biodegradable and commercial polymer Ecoflex (a blend of L,D-poly(lactide (L,D-PLA) and PCL) [134]. DSC analysis indicated an improvement in the composite's glass transition temperature (T_g) and crystallization temperature. There is an enhancement of decomposition onset and maximum temperatures by 15 °C and 16 °C, respectively. The thermal improvement is due to increased cross-linking caused due to the inorganic nanoparticles [134]. Carbon nanomaterials such as CNT enhanced the T_m of PLA [135]. CNC improved the thermal stability of polyaniline (PANI) [136].

The coordination of Cd^{2+} ions with chitosan improved the thermal stability of chitosan [137–139]. The presence of CdS quantum dots enhanced the thermal stability of chitosan. The thermal decomposition of CdS/chitosan was increased by 60 °C compared to pure chitosan [137,138,140–142]. Co-doped ZnO improved thermal stability of carboxymethyl cellulose (CMC)[143].

TGA and TGA-FTIR were used to characterize PCL and PLA's thermal stability and degradation [144]. Data analysis showed that PCL exhibited higher thermal stability than PLA. However, PLA has higher activation energy showing higher degradation kinetics. This observation indicates that the degradation rate depends on temperature. A blend of PLA and PCL showed lower thermal stabilities than both polymers. However, the thermal stability can be improved by adding TiO_2 nanoparticles[144]. TiO_2 NPs also improved the thermal stability of PLA/poly(hydroxybutyrate-co-valerate) (PHBV) [145].

Thermal analysis of pure PCL and PCL: lithium thiocyanate (LiSCN) salt complexes using different compositions were reported (**Figure 5**) [146]. DSC thermogram for pure PCL showed a relatively sharp endothermic peak at 64 °C. This observation indicates the high purity of the PCL. However, DSC thermograms of the PCL\LiSCN composite showed a shifting in the melting temperature (T_m) toward lower temperatures with increased LiSCN contents. The drop of T_m in the presence of the salt addition is due to the decrease in size and free energy (**Figure 5**).

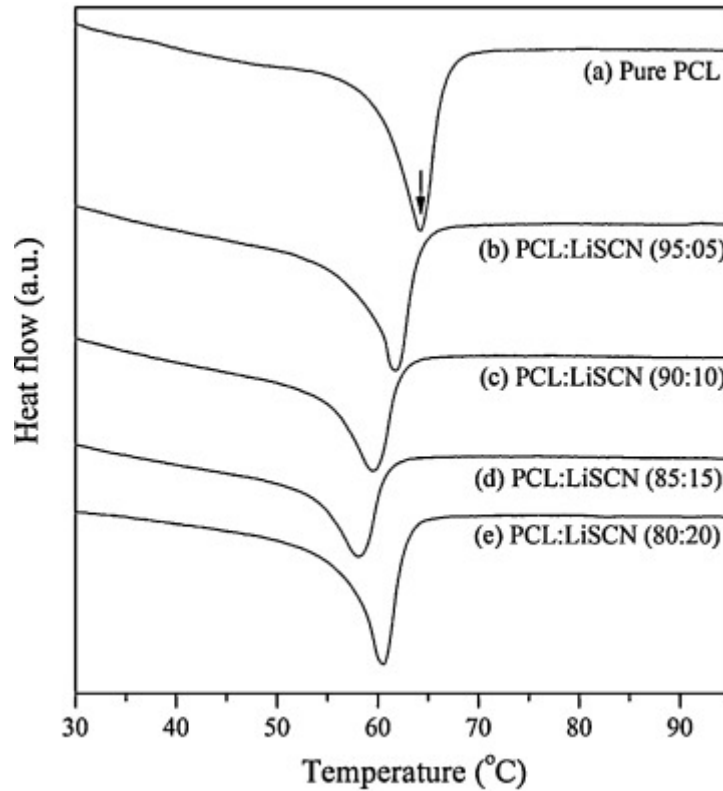


Figure 5 DSC analysis of PCL:LiSCN films embedded different concentrations of LiSCN [146]. Copyright 2015. Reproduced with permission from Elsevier.

6. Thermal Conductivity for Biodegradable Polymer Nanocomposites

Thermal conductivity is the ability to conduct heat. Thermal conductivity is measured in watts per meter kelvin ($\text{W}^{-1}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$) according to the International System of Units (SI). Organic polymers are heat insulators with low thermal conductivity (TC, $0.2 \text{ W}^{-1}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$). The low TC values of polymers hinder their adoption in electronic applications. The TC of polymers can be improved via several methods, including synthesizing polymers nanocomposites with organic and inorganic materials.

A solvent-free spherical cellulose nanocrystals fluids (CNCfs) embedded PLA membrane was fabricated using electrospinning [147]. PLA/CNCfs fibrous membrane exhibited high thermal conductivity of $0.27 \text{ W}^{-1}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ [147]. A melt extrusion-stretching method was reported to process a biodegradable composite of PLA/poly(butylene adipate-co-butylene terephthalate)/carbon nanofiber (PLA/PBAT/CNF) [148]. PLA/PBAT/CNF containing CNF of 10 wt.% exhibited in-plane TC of $1.53 \text{ W}^{-1}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$. CNF offered a 31.9% increment in the thermal conductivity of the biocomposite[148].

Hydroxyl-functionalized hexagonal boron nitride (OH-h-BN) nanoplatelets improved thermal conductivity for chitosan microspheres (CSM)[149]. OH-h-BN/CSM nanocomposites exhibit thermal conductivity of $5.66 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$. It showed 502% and 1914% enhancement compared to OH-h-BN/CS nanocomposites and pure chitosan, respectively [149]. A simple and environmentally friendly strategy was reported to fabricate epoxy/BN composites[150]. The synthesis method improved the through-plane TC of the prepared composite. The effect of filler sizes of BN on thermal conductivity was also investigated. The data analysis showed that the high content of BN in the epoxy composite ensured a higher through-plane TC of $2.01 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ [150].

Metallic nanoparticles exhibit high thermal conductivity ensuring significant improvement for polymers nanocomposites. Metallic nanoparticles such as silver nanowires (Ag NWs) improved the through-plane thermal conductivity of cigarette filters (CF, non-biodegradable cellulose acetate), offering TC of $1.71 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ at the loading of 2.8 vol% Ag NWs¹¹⁵. A cellulose nanofibers/MXene@Ag film showed TC of $22.43 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ due to the synergistic effect of Ag NPs¹¹⁶.

Carbon nanomaterials such as graphene improve the thermal conductivity of polymers. They have high TC at ambient temperature. Graphene exhibits a high aspect ratio with high TC ($\geq 5000 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$)[151]. The TC value of epoxy (BE)/4,4'-diaminodiphenyl methane/GNP (graphite nanoplatelets) and BE/DDMGNP-10 nanocomposites was enhanced to $2.21 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ which is ten times higher than conventional EP [152]. Cellulose/multiwall carbon nanotube (MWCNT) exhibited in-plane and through-plane TC of 1.98 and $0.34 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, respectively [153]. The composite was prepared via several steps, including 1) a simple paper-making step, 2) a layer-by-layer assembly step, and 3) *in-situ* welding (**Figure 6A**). There is a significant increase in the in-plane TC with the addition of CNT content without a substantial increase in the through-plant conductivity (**Figure 6B**)[153]. MWCNT and GNP improved thermal and electrical conductivity for composites of polyoxymethylene (POM)/ PLA/MWCNT and POM/graphene nanoplate (GNP) (denoted as PMCNT and PMGNP, respectively)[154]. PMCNT and PMGNP exhibited electrical and through-plane thermal conductivities of $3484 \text{ S}\cdot\text{m}^{-1}$ and $1.95 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, respectively, for PMCNT40, and $2695 \text{ S}\cdot\text{m}^{-1}$ and $4.24 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$, for the corresponding values for PMGNP48 [154]. MWCNT improved the thermal and electrical conductivity compared to GNPs. GNPs (loading of 40 wt.%) improved the thermal and electrical conductivity for the PBAT/PLA blend (ratio of PBAT:PLA was 75:25), offering thermal conductivity and electrical conductivity of 3.15

$\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ and $338 \text{ S}\cdot\text{m}^{-1}$, respectively[155]. The polymer's properties are enhanced with the GNPs content (**Figure 7**). The presence of PLA ensures a confined composite of PBAT and GNPs according to TEM images (**Figure 7**).

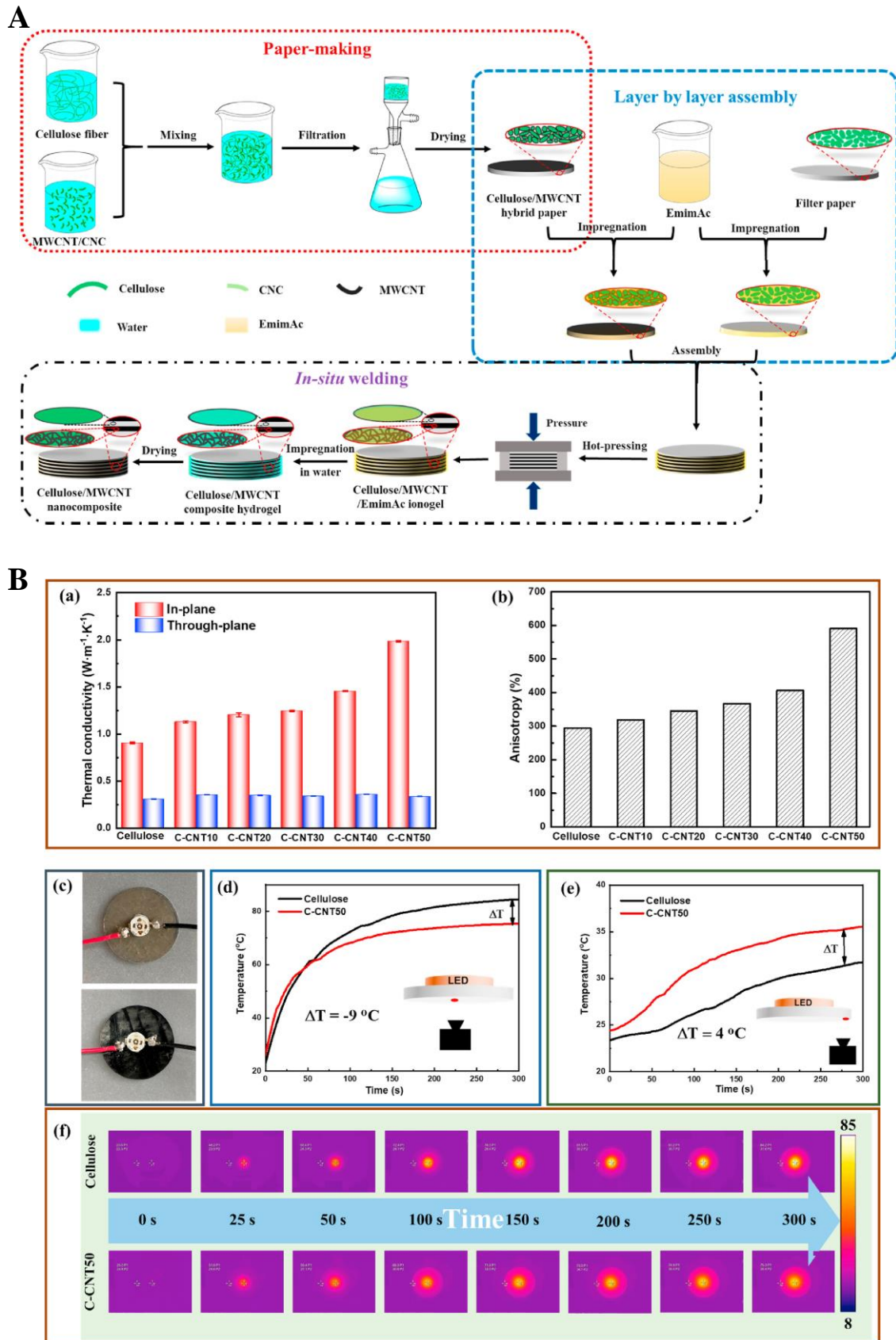


Figure 6 A) Schematic of the fabrication process of the cellulose/MWCNT nanocomposites, and B) a) TC and (b) TC anisotropy of cellulose/MWCNT nanocomposites, (c) Photographs

of cellulose (upper) and C-CNT50 (lower), (d) and (e) Surface temperature. (f) Infrared thermal images [153]. *Copyright 2021. Reproduced with permission from Elsevier.*

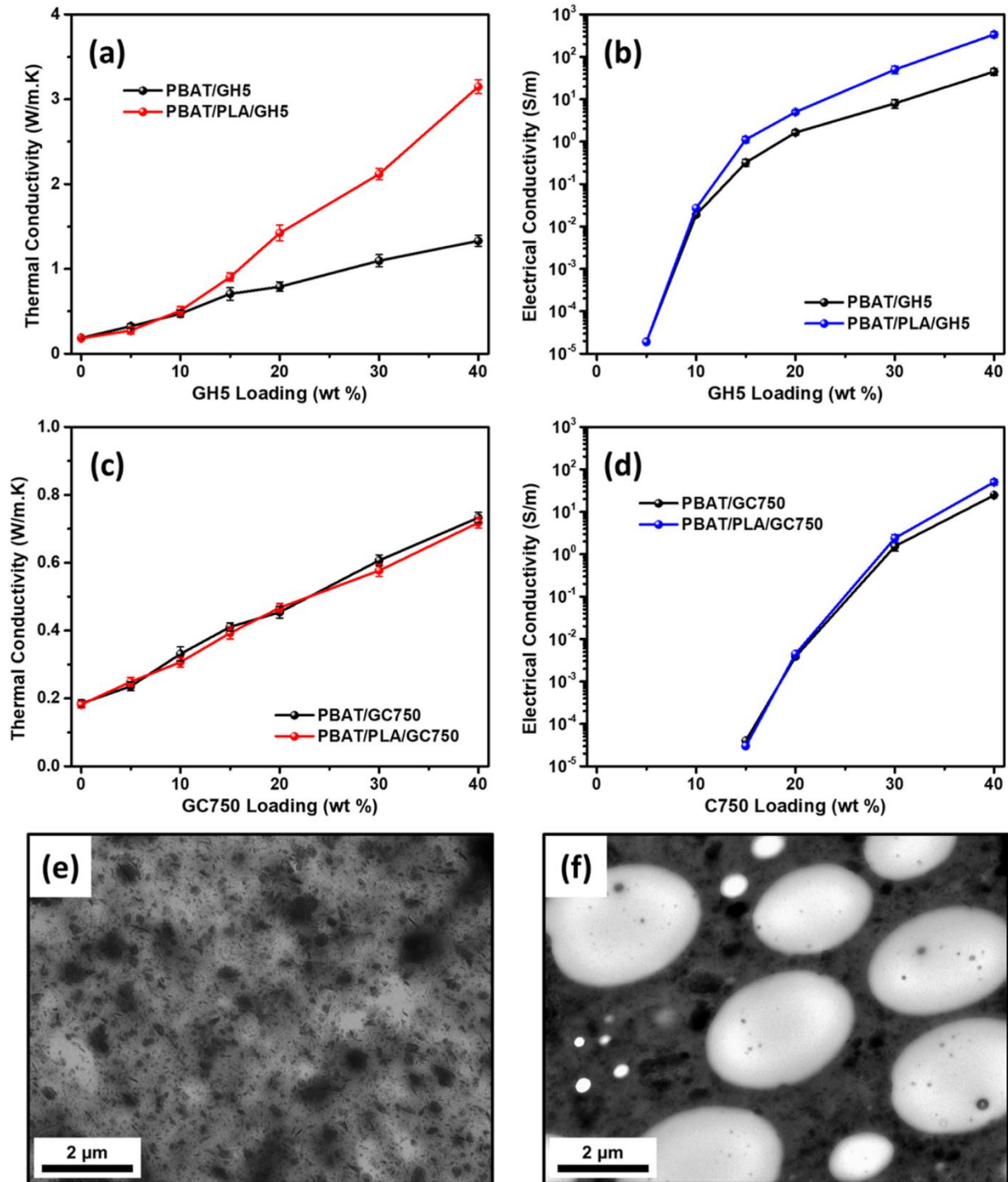


Figure 7 a, c) thermal conductivity, b, d) electrical conductivity, of a-b) PBAT/GNPs and c-d) PBAT/PLA/GNPs, e-f) TEM images of e) PBAT/GNPs, and f) PBAT/PLA/GNPs [155]. *Copyright 2020. Reproduced with permission from Elsevier.*

7. Electrical Conductivity

Electronic devices are usually used in traditional polymers such as polypropylene (PP), polystyrene (PS), polyimide (PI), and poly(ethylene terephthalate) (PET). These polymers are mainly non-degradable, causing environmental concerns. In contrast, biodegradable polymers are environmentally friendly with high biocompatibility and metabolization properties. Thus, they can be used for implantable chips and wearable devices.

Most reported polymers lack high conductivity except for some conducting polymers (CPs). The low conductivity of polymers is due to the covalent bonding. A strong bond, i.e., covalent bonds, prevents electron channels or ion migration. Compared with traditional conductive materials, CPs offer advantages, including high flexibility, lightweight, and low cost. CPs can be classified into two categories: Structural (intrinsic) structural CPs (SCPs): e.g., polyaniline (PANI), polypyrrole (PPy), polyphenyl acetylene (PPA), and their derivatives, and CPs composites (CPCs): incorporating the polymers with conductive fillers, e.g., carbon black (CB), carbon nanotubes (CNTs), and graphene (G).

Several critical parameters affect the polymer's conductivity (**Figure 8**). The size of polymers determines the polymer conductivity. When the volume fraction of polymers increases to a specific critical value, there is a significant increase in the conductivity of the polymer, i.e., conversion of insulator materials to the conductor. The conductivity of biodegradable polymers can be improved by adding conductive nanofillers. It can be improved by adding a small volume fraction of high conductivity materials. The conjugated polymers exhibit electrical conductivity in the range of 10^{-10} to $10^{-5} \text{ S}\cdot\text{cm}^{-1}$. The electrical conductivity can be improved via several methods, including doping with molecules such as salt ions that can improve the polymer's electrical conductivity to $10^4 \text{ S}\cdot\text{cm}^{-1}$. As a rule of thumb, the critical value (defined as the percolation threshold) of the conductive material in the composites offers a significant increase of several orders of magnitude in conductivity. The critical value of the conductivity materials in polymer composite converts an insulator to a conductor. Above this threshold, the relation between the concentration and the conductivity (σ) or electrical resistivity (ρ) can be described by a scaling law[156]:

$$\sigma = \sigma_0[(\varphi - \varphi_c)]^t$$

$$\rho = \rho_0[(\varphi - \varphi_c)]^t$$

where σ_0 , ρ_0 , t , φ , and φ_c represent the intrinsic conductivity of the fillers, the resistivity coefficient, the exponent, the volume fraction of conductive particles, and percolation transition, respectively. The exponent t depends on the dimensionality of the polymers, which is called universal percolation behavior. Based on the theoretical calculation, it equals 1–1.3 and 2 for two-dimensional (2D) networks and a three-dimensional (3D) network.

Nanofiller materials exhibit higher electrical properties than micro-sized fillers. Conductive nanofillers with a large aspect ratio (e.g., CNTs, graphene) exhibited an excellent connected conductive network providing a low percolation threshold. The use of carbon nanomaterials such as graphene improved the thermal conductivity of the biodegradable polymers [157]. Chitosan composite film containing GO exhibits good proton conductivity. PBAT composite containing 40 wt.% of graphene nanoplatelets (GNPs) exhibited electrical and thermal conductivity of $3.15 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ and $338 \text{ S}\cdot\text{m}^{-1}$, respectively [155]. GNPs improved the electrical conductivity for PLA/chitosan (75/25 wt./wt.) [158], and polyhedral oligomeric silsesquioxane/PCL (POSS-PCL) [159]. The use of plasticizers such as glycerol improved the conductivity for poly(vinyl alcohol) (PVA) films and PVA/starch blend [160]. Data analysis reveals that there is an increase of the conductivity for PVA with the increase in the content of glycerol and starch [160]. The same observation was reported for Chitosan/AgNO₃/Al₂O₃ system using glycerol as a plasticizer [161].

The conductivity of PLA can be improved by incorporating gold nanoparticles (Au NPs) [162]. The PLA/Au NPs/indium tin oxide (ITO) electrode was used as an electrochemical biosensor for the selective identification of leukemia cancer cells [162]. Silver nanowires improved the conductivity of PLA [163]. PLA/Ag NPs were reported as electrodes for the organic light-emitting diodes (OLEDs) [163]. It offered high transparency with good electrical conductivity, even after 10 000 cycles [163]. AgNPs-Ni₂O₃ improved the electrical conductivity of cellulose nanowhiskers/polypyrrole [164]. Ag NPs can be also used to improve the electrical conductivity for chitosan [165].



Figure 8. Key parameters are affecting polymer's conductivity.

The perfect interconnection between polymers and conductive nanofillers is critical for constructing a conductive nanocomposite. The homogenous distribution of nanofiller in the polymer nanocomposite improved the electrical conductivity of the composite. Naturally, the nanofillers tend to distribute randomly in the polymer matrix. It was reported that some fillers tend to spread to the polymer's surface at higher concentrations.

A study showed a correlation between the dispersion state of MWCNTs in PP and electrical conductivity [166]. The composite was synthesized using two different procedures; 1) chemical modification of MWCNTs and 2) incorporation of a master batch [polypropylene-grafted-maleic anhydride (PP-g-MA)] as a compatibilizer followed by a simple melt blending. Based on the optical microscopic images of MWCNTs/PP composite, the addition of a compatibilizer enhanced the uniform dispersion of MWCNTs in the PP matrix. Although it reduced the conductivity. However, the post-heat treatment improved the interconnection between MWCNTs and PP, leading to higher electrical conductivity. The uniform distribution and construction of a compelling relationship between the composite are crucial. Amphiphilic polymers prevent the aggregation in nanocomposites non-covalently attachment.

The addition of CNT improved the conductivity of several polymers, such as PCL[167,168], polypropylene (PP)[168], PLA[169], and chitosan-PCL[170]. The dispersion of conductive carbon nanoparticles (e.g., CB, CNTs) in PLA can be improved via the addition of ramie fiber (RF)/PLA, resulting in different sensing mechanisms [169]. PCL/CNTs nanocomposites were synthesized via in-situ grafting and polymerization of PCL and CNTs [171]. It exhibits good conductivity enabling quantitative sensing of organic vapors[171]. The melt-blown method synthesized PCL/PP/CNTs [168]. A 3 wt.% CNTs offered a significant high electrical resistance of the polymer nanocomposites, enabling the detection of various solvents[172]. PCL/CNTs/PLA blends containing four wt.% CNTs were synthesized via the melt-spun procedure as fibers[168]. The fibers exhibited good electrical properties for application as a textile sensor. CNTs/PCL composite can be used to monitor the temperature in a range of 20-80 °C [168]. MWCNT exhibited more remarkable POM/PLA composite improvement than GNPs (**Figure 9**)[154]. The conductivity depends on the loading of both materials (**Figure 9**)[154].

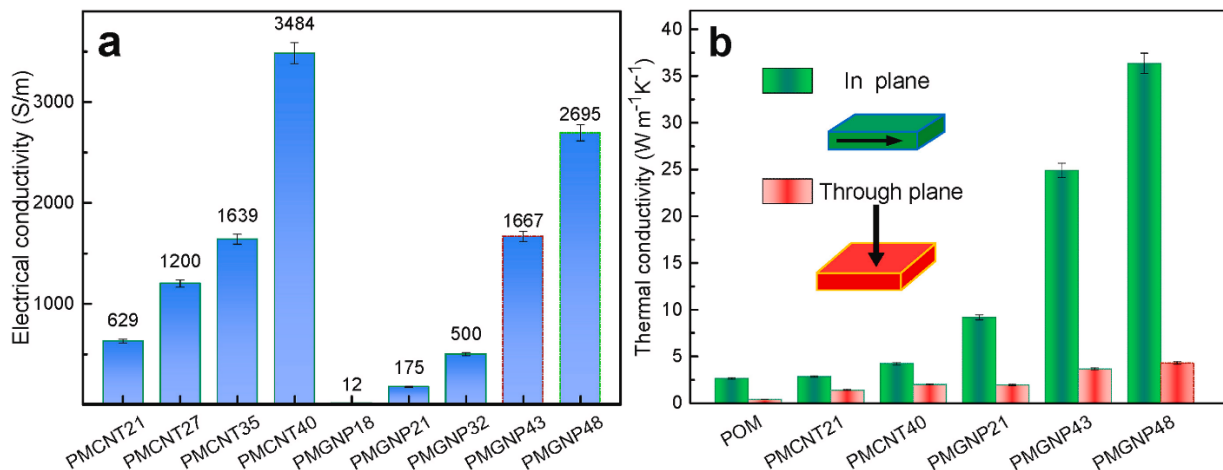


Figure 9. (a) Electrical and (d) thermal conductivity.[154] *Copyright 2021. Reproduced with permission from Elsevier.*

The nanofiller materials such as metallic nanoparticles or metal oxides with high conductivity ensure high conductivity. The electrical properties of PVA can be improved by adding In₂O₃/Cr₂O₃ nanoparticles [128] and GO [129]. 3D printing PVA/GO devices can be operated in a small gate voltage range of -0.5 to 2.5 V for electronic devices and circuits[129]. Graphite/diamond particles [173] and G [174] enhanced the electroconductive of PLA nanocomposites. Nano-clay improved the electrical conductivity of bacterial cellulose (BC)/PANI to 0.49 S•cm⁻¹ using only 5 wt.% of the clay [175]. It exhibited 16 folds improvement in the electrical conductivity for the polymer blend [175]. The improvement in the conductivity of biodegradable polymers enable the fabrication of energy storage devices with high biodegradability [176].

8. Conclusions

This chapter discusses the dielectric, thermal, and electrical conductivity properties of biodegradable polymers nanocomposites. The fabrication process of biodegradable polymer nanocomposite can significantly influence the electrical properties. There are several methods for the synthesis and fabrication of polymer nanocomposites. However, preparing nanocomposite with high conductivity, i.e., low percolation threshold, remains challenging. There is a lack of clear procedures to characterize biodegradable polymers' dielectric, thermal, and conductivity. Potentially, there is high progress in evaluating the material's properties to ensure high benefits.

9. References

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