

Polymer Composites in Present Scenario

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Abstract

Present scenario of growing development demands more sophisticated and engineered materials to facilitate the lifestyle and to meet the challenges in the establishment of colonies in space. The advancements in composite technology meet the growing demands. Composites are the engineered material for future. There are many scopes in composites with various emerging nanotechnology and smart materials. This paper is a brief review of the composite's concept and their types. The differentiation between the electronically conducting polymers and conducting polymer composites are also discussed along with their contemporary applications.

Keywords- Composites, Nanocomposites, Fabrication.

1. Introduction

The discovery of plastic was critical in replacing some of the heavier metal components with lighter components. Earlier, easier to mold plastics had limited use mostly in low-end applications for day to day use. For their high-end applications such as aerospace, marine and automotive; materials must be of high tensile strength at the same time should also be light weight. Search of light-weight high tensile strength hybrid materials, whether conducting or non-conducting of low density host materials opened up the stream of new engineered special materials.

Bone and wood is a natural polymeric composite. The bone is composed of hydroxyapatite reinforced with collagen fibers. The wood lacks dimensional stability because basically it is made up of cellulose, hemicelluloses, and lignin. Among those, lignin, a weaker substance (matrix), holds the long fibers of cellulose (reinforcement). The water adsorption property of wood is due to its abundant hydroxyl functionality. The adsorbed water is held by hydrogen bonding which causes swelling/shrinkage as well as physical degradation and hence mechanical failure (Homan and Jorissen, 2004). In such composites, the water absorption can be tailored by using appropriate reagents (Kumar, 2007; Hon, 2017).

The idea of manmade composites is primitive. An idea of making the dried cake of mud using straws is perhaps the earliest example of a composite in which straw was used to improve the tensile strength of dried mud cake.

Concrete is a good example of a composite in which the gravel of small stones are particulate which provides a good compression strength (Lechtman and Hobbs, 1987) but has poor tensile strength, therefore, steel rods are used as reinforcement. Hence, a good compressive and tensile property is obtained.

2. Composites and Their Classifications

Basically, a composite system is composed of a matrix (host/binder) and one or more reinforcement materials (filler). The reinforcement is dispersed in a matrix (host) which holds the reinforcement material. Generally, reinforcement is used to strengthen either in fibers or particulate form. Hence, a composite material is composed of two or more materials having different properties (Ellyard, 2000; Gibson, 2011;). These different materials provide the overall composite material with enhanced properties that are better than those of the separate constituent parts. In contrast to metallic alloys, composite materials retain its separate physical, chemical, and mechanical properties. It is to mention that in a composite system both the matrix and reinforcement remain distinguishable whereas in an alloy the constituents dissolve (blend) with each other, therefore, are indistinguishable. Generally, composite materials have high strength and stiffness, combined with low density, when compared with bulk materials hence, allowing for a weight reduction in the finished part. Moreover, composites also provide economical/functional benefits to both manufacturers and consumers. On the basis of the form of the structural components, the composites are classified as:

- (i) Whiskers/Fibrous: such composites are composed of fibers/whiskers in a matrix.
- (ii) Laminar: such composites are composed of layers (layer by layer) in a matrix.
- (iii) Particulate: such composites are composed of particles in a matrix.

Figure 1 shows the composites containing fibers, fabric, particulates, and whiskers.

Further, the classification may also be on the basis of different matrixes as

- (i) Polymer matrix based composites
- (ii) Metal matrix based composites
- (iii) Ceramic matrix based composites
- (iv) Carbon-carbon based matrix composites
- (v) Intermetallic/hybrid composites.

Modern composites consist of more advanced materials as most commonly used fiber-reinforced plastics (FRP) (Gibson, 2011). It is a laminated structure consisting of woven fiber fabric reinforcement or layers of unidirectional fiber that is embedded in a polymer matrix material (Bhatnagar et al., 1995; Baillie, 2005; Vassilopoulos et al., 2008; Lopresto et al., 2011).

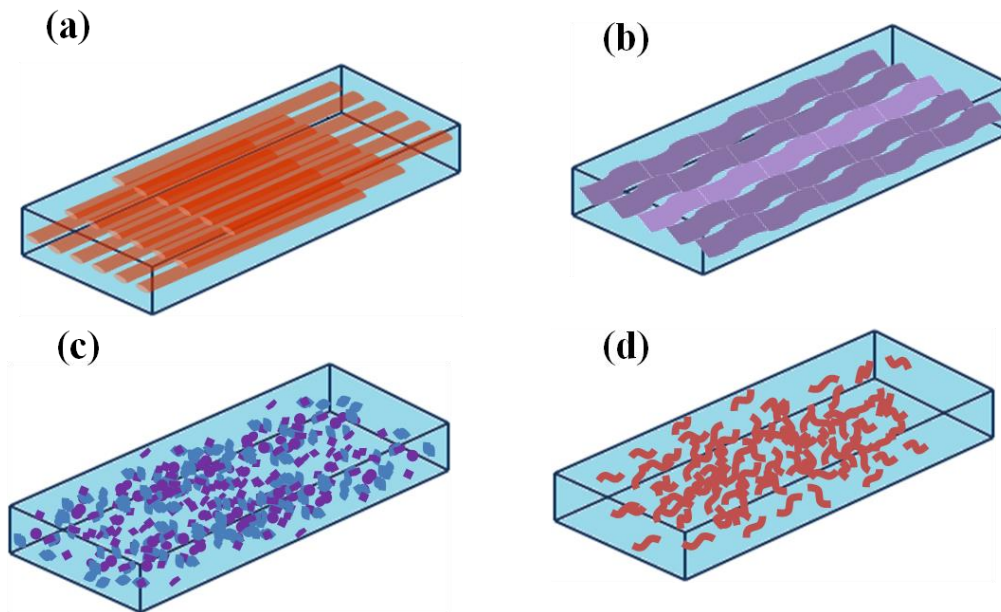


Figure 1. Composite contains (a) fibers (b) fabric (c) particulates (d) whiskers

Advantages of Composite Materials

In place of metals and metal alloys, the composites are custom designed to meet specific application needs as it can provide selective directional properties (Hollaway, 1994). Due to molding property of plastics, the parts formerly assembled out of several smaller metallic components can be fabricated into a larger single part, hence, reducing manufacturing costs. A low cost, low density but stronger/stiffer material with high corrosion resistant property can be fabricated into many complex shapes during fabrication, even providing finished, styled surfaces in the process.

Since our concern is polymer based matrix composites, therefore, we discuss polymer matrix composites and nanocomposites.

The nonlinear conduction property of composites makes these useful for applications that require the resistivity in different ranges (Endo et al., 1976). The origins of terahertz radiation with metamaterials have opened the advancement of several real-world THz applications (Averitt et al., 2007). Therefore, the development of terahertz (THz) devices in advanced technology applications demand the EMI shielding (Dang et al., 2003) composites. Hence, the contemporary research of composites is leading to explore new methods and improvements in existing methods for low percolation threshold to construct a conducting conduit through the polymer.

Often, the heavy and expensive metal fillers result in heavy polymer composites, therefore, a family of low-density carbon, the high performance conducting nanofillers as carbon black

(CB), carbon-nanotube (CNT), carbon-nano-fiber (CNF) and carbon whiskers are preferred. Whereas, CNFs is low in cost and has long fibers (~ 200 μ m) than SWNT (single-wall nanotube) and MWNT (Multi-wallnanotube) hence, the long fibers offer good connectivity around a micrometer-sized polymer particle which may result in good connecting channels for electrical conductions in that polymer matrix.

Conceptually, the conducting filler in a matrix forms connected paths for electronic conduction in that composite system. Generally, chemical or mechanical routes are adopted for the dispersion of fillers in a matrix. If a mechanical mixing process is being adopted then only a good inter-connected network with few agglomerations fillers dispersed in the host (matrix) can lead to an enhanced electrical conductivity of the nanocomposite and this is achieved by increasing the CNF content at percolation concentration (Liu et al., 2014). As a fact, due to the high specific surface area and Van-der Waal interaction, the nanoparticles tend to agglomerate if not dispersed properly in the host polymer, hence, the strength of nanofiller filled polymer composites is reduced by stress concentration effect. Simultaneously, further, it also affects the electric property. Few polymers have high melt viscosities as PTFE (Katepalli et al., 2011), therefore, it is hard to disperse the fillers in such polymer via conventional melt technique. So, mechanical or chemical methods can be employed with or without using sonication. Chemical routes require treatment with co-solvents.

3. Fabrication Methods

The general fabrication methods of polymer composites are classified into five approaches

- (i) Intercalation method.
- (ii) *in situ* polymerization.
- (iii) Sol-gel method.
- (iv) Direct mechanical mixing of a polymer and nanofillers.
- (v) Simple melt compounding method -no surface modification of filler: It is an industrial research by using a simple melt compounding method (no surface modification of filler is required).

Generally, a good mechanical strength (filler must bind well with matrix) in composites is the need for their practical use. Therefore, prior surface treatments of fillers are preferred for a good adhesion of filler-matrix.

Intercalation Method

In this method, an inorganic filler i.e. *layered silicates* (as montmorillonite and mica) is downsized to nano-dimension by intercalation process. An organic compound is intercalated into inter-layer spacing of the silicate which results in an inhomogeneous distribution of plate-like nanofillers. Since the silicate is hydrophilic and the organic compound is hydrophobic, therefore it must be treated with some organic surfactants. The preferable organic compounds should have quaternary cation functionality such as phosphonium salts, amino acid,

imidazolium and alkylammonium. The exfoliation of silicate is generally performed via a mechanical or chemical method (*in situ* polymerization or *in situ* intercalation polymerization process) (Usuki et al., 1993 Fudala et al., 1999 Lin et al., 2001 Zhang et al., 2014). It is also revealed that similar intercalation also occurs in the interlayer spacing of graphite. Hence, the fabrication of graphite/polymer nanocomposites by the *in situ* intercalation polymerization is possible. Further, in the mechanical method, the polymer is directly intercalated into layered silicates in a suitable solvent (Shioyama, 1997; Ogata et al., 1997; Shioyama et al., 1998; Fudala et al., 1999; Lin et al., 2001; Ray and Okamoto, 2003; Beall et al., 2011). This method can also be employed for the fabrication of nanocomposites even when polymeric systems are unsuitable for *in situ* polymerization.

In Situ Polymerization

In *in situ* polymerization, the inorganic (metal) /organic nano particles are dispersed in a monomer solution (Ratna et al., 2003). The wettability of nanoparticles with the monomer is improved by using a metal precursor (organically modified particle surface). The metal/polymer nanocomposites synthesized *via* this method can be found in references (Vaia et al., 1993; Vaia and Giannelis, 1997). The synthesis of many precious organometallic/polymer nanocomposites (as platinum, silver, palladium, and gold) involves reduction of metallic ions in complexes or polymerization (Vaia and Giannelis, 1997).

Sol-Gel Method

Sol-gel is a kind of bottom-up technique to carry out the synthesis of organic/inorganic molecular hybrid materials along with *in situ* formation of a nanofillers and *in situ* polymerization. The feature of this technology makes able to disperse inorganic fillers which are shorter in dimension than the molecular chain length of the matrix. Since the alcohol- and water-soluble polymers form hydrogen bonds with hydroxyl groups on inorganic fillers surfaces, therefore, their use as hybrid materials are limited in sol-gel technique.

The metal alkoxides reactions (Mayer, 1988; Chujo and Saegusa, 1992; Nakao, 1993 Reynaud et al., 2001) are the basis of this method. A silica/polymer molecular hybrid materials reaction is given below



Above reaction includes hydrolysis and polycondensation reactions of tetraalkoxysilane. The main drawback of this method is that reactions employed have a dramatic effect. Hence, at the molecular level, it is not possible to control the arrangement and size (angstroms to several nanometers) of inorganic domains in a hybrid material.

To solve the difficulty of synthesis of hybrid materials, a site-selective molecular hybrid method (a segmented-polymer and silica) using a silane-modified polymer have been

developed. In this, the alkoxy silane oligomers are introduced selectively at appropriate sites in each polymer. This approach has been used to improve the properties of polyurethane (Chujo and Tamaki, 2001; Goda and Frank, 2001). The advantage of this approach is that microstructure can be controlled in the hybrid material. Hence, no interaction with metal alkoxides takes place and this approach is suitable for the conventional sol-gel technology. Attention is required during various complex reactions taking place in the synthesis processes.

4. Direct Mixing of Polymer and Nanofillers

It is a top-down approach based on the breakdown of aggregated fillers to nm -scale during the mixing process in a polymer. Hence, it is a suitable method of fabricating polymer-based composites containing nm and μm sized fillers. There are two general ways of mixing the polymer and fillers.

Following are the steps in this process

Melt-Compounding Method

In the absence of any solvents, the mixing of nanofillers into a polymer above its softening temperature is known as melt-compounding method (Takadama et al., 2004; Kalaitzidou Fukuda et al., 2005; Kalaitzidou et al., 2007). In this method, the hydrodynamic force (shear stress hydrodynamic force) induced in the polymer during melt is used for the breakdown of aggregated fillers. It mainly depends on the internal shear stresses induced by viscous drag on the fillers during melt-compounding.

The induced shear stresses can work either along the cross section in the agglomerate (rupture model (Jankong and Srikulkit, 2008)) or remove a primary particle or a group of primary particles from the surface of the larger agglomerate (onion peeling model (Manas-Zloczower et al., 1982)). In either case, the dispersive action occurs only when the shear stress becomes larger than a critical threshold value for breaking down of filler's agglomerates inside the kneaded polymer melt (Palmgren, 1975; Shiga and Furuta, 1985; Hashimoto et al., 2006).

Solution Mixing Process

In this method, the nanofillers are pre-dispersed in a polymer using a solvent. Then evaporation of solvent from solution ensures finely dispersion in the polymer matrix (Carotenuto et al., 1996; Cho and Paul, 2001 Andrews et al., 2002; Preghenella et al., 2005; Yang and Nelson, 2006; Kalaitzidou et al., 2007). Generally, surface modification of nanofillers or ultrasonic waves is used to disperse the nanofillers. Ultrasonic waves provide an external force to overcome the less value of shear force during the mixing process in the absence of a solvent. Few polymers such as polyethylene are not solvable in solvents having a low boiling point; therefore, a solution-mixing method is limited for few polymer matrixes.

In a typical example of the dispersion state of micron-sized silica additives in the PFA-matrix composite exhibited isolate silica particles. The addition of nm-sized silica formed the micro-sized agglomerated cluster. Hence, in kneaded PFA melt, the μm -sized silica agglomerates (additives) gets apart only when the hydrodynamic force exceeds the cohesive force acting among the agglomerated silica particles. Otherwise, it results in isolated dispersion of large silica particles in the PFA matrix when the cohesive force exceeds hydrodynamic force. Hence, to overcome the difficulty of dispersion of nanoparticles, they are chemically treated using surface modifiers as silane (a coupling agent) before dispersion.

Since silica has a low chemical affinity (hydrophilic surfaces or poor wettability) therefore it is not easy to disperse it completely into hydrophobic perfluoropolymer even after surface modification. Hence, it is difficult to fabricate silica/perfluoropolymer composite. The dispersion can be facile by combining the melt-compounding method with ultrasonic mixing (Agarwal et al., 2007). It has been possible to disperse (not uniform) titanium oxide nanoparticles into kneaded polymer melts by utilizing ultrasonic waves.

Simple Melt Compounding Method -No Surface Modification of Filler

(Suetsugu and Sato, 2006)

This research is an extension of melt-compounding approach which does not require any surface modification (use of organic surfactants) of inorganic particulate fillers (nm). Hence, it is a simple and versatile method without the use of any organic solvents as required in *in situ* polymerizations, sol-gel processor in solution mixing process for the fabrication of filler/polymer nanocomposites.

Moreover, it is compatible with extrusion and injection molding and hence finds its suitability in current industrial processes for large-scale production of nanocomposites even for the polymers which are not suitable for above-mentioned methods.

This concept concentrates on fracture strength required for the agglomerated nanoparticles. Hence, reducing the fracture strength before a melt-compounding stage is the key strategy of dispersion in this approach (Watanabe et al., 2006; Tanahashi et al., 2007; Tanahashi, 2010).

The main factors of dispersion state in this method is

- Shear stress – induced in a kneaded polymer melt.
- The threshold value needed for the breakdown of agglomerates.

The melt compounding process allows the breaking of agglomerates and ensures the uniform dispersion of nanofillers when the induced shear stress is higher than the agglomerate strength.

The authors have published a work to disperse the nm-sized spherical silica particles in various filler/polymer nanocomposites (Watanabe et al., 2006; Tanahashi et al., 2009; Tanahashi, 2010). In this work, to ensure a bulk porosity of silica particles (the open packaging), they have used Derjaguin-Landau-Verwey-Overbeek (DLVO) theory (Deraguin and Landau, 1941) for the stability of colloidal dispersion in the system.

5. Nanocomposites

Nanocomposites are the combination of multiphase materials in which one of the constituents has one dimension in the nanometer range [1-100nm] (Uppal and Shiakolas, 2008). A nanostructured material has considerably different or enhanced properties as compared with a larger-dimensional material (μm) of the same composition. Since the surface area per unit volume is inversely proportional to the diameter of a nanomaterial, therefore, the surface area per unit volume increases as the diameter of the nanostructure decreases (Bertsch et al., 1998). Since the chemical and physical interactions are governed by surfaces and surface properties, it's the molecules on the surface that are responsible for any property. Therefore; at the nanoscale, a large surface area for a given volume (Bertsch et al., 1998) enhances the number of molecules and so their physical properties (Uppal and Shiakolas, 2008). Hence, a transition from microparticles (μm) to nanoparticles (nm) yields dramatic changes in the physical properties of the composite.

Application of nanomaterial ranges from different sensors, actuators, smart materials electro-magnetic-optical devices, heat transfer enhancement fluids or interfaces, drug synthesis/design/delivery to energy harvesting devices, memory chips, electronic devices, catalysis, functional polymer composites and coatings and in so many other areas invading various branches of science and technology.

In a nanocomposite, the multi-functionality is attributed to the combination of the constituent materials. The choice of size of the nano constituents is based on the required application. The nm-sized constituent can be

- (i) One dimensional (such as nanofiber and nanowires),
- (ii) Two dimensional (layered mineral like nanoclay) or
- (iii) Three-dimensional (spherical particles in nanoscale range).

Nanomaterial produced via numerous versatile techniques, impart their chemical and physical properties to the composites. The focus of contemporary nanocomposites research is in the areas of manufacturing techniques and combination of material for the manufacturing. As discussed above, the properties of nanocomposite materials depend on the morphology and interfacial characteristics of constituents. Hence; the nanocomposites research is progressing with the use of a combination of characterization (AFM, SEM, TEM STEM etc.) and detailed modeling (Uppal and Shiakolas, 2008).

6. Polymer Matrix Composites

Basically, polymers are considered as insulators. However, they have emerged as potential materials in the 20th century. The useful exploitation of polymers, either active (in electronic, optical, energy storage and mechanical) or passive (containers or coatings) has turned our future in a comfortable zone. The basic electrical conduction mechanism in polymers is generally due to loosely bound ions which is considered undesirable. (Freund et al., 2007; Inzelt, 2008).

Polymeric-based composites were first introduced in 1960. Since then, there were many improvements in consumer goods, sports goods, automotive components, aerospace parts and marine and oil industries. Keeping in mind, the sustainability and performance of any product, the composite material is the option of replacing the use of heavy materials in many applications such as heavy steel and aluminum with carbon-based derivatives. Therefore, it saves 20 to 50% weight by substituting 60 to 80% and aluminum parts in component weight of steel-based applications.

The matrix material used in polymer-based composites can either be thermoset (epoxies, phenolics) or thermoplastic resins (low-density polyethylene (LDPE), high-density polyethylene, polypropylene (HDPE), nylon, acrylics etc.). The choice of filler or reinforcing agent depends upon desired or required properties. The characteristics (electrical, thermal or mechanical) of a polymer composite depend upon the orientation as well as the concentration of fibers and properties of the polymer matrix.

The conductive polymer or electronically conducting polymers are a relatively new class of materials. Their traditional applications are corrosion inhibitors, compact capacitors, antistatic coating, and 'smart' windows while new applications include transistors, organic light-emitting diodes (OLEDs). Since then conductive polymers have been the subject of both theoretical and experimental studies.

On the other hand, the conducting polymer composites are such composites of polymer in which conductive fillers whether metallic or carbonaceous are mixed and hence, the electric characteristics of conducting polymer composites is tunable and versatile. The carbonaceous fillers are advantageous because as carbon black, carbon nanotubes or carbon nanofibers, graphene etc. do not get oxidized (Zois et al., 2003). The other advantage of such conductive composite is that they are flexible, lightweight and have low production costs while possessing electrical property. However, their mechanical characteristics and processing are typical of plastics. They are used as antistatic materials, switching devices, cables, transducers and electrostatic discharge (Gul, 1996; Huang, 2002 ; Ma et al., 2010; Villmow et al., 2011; Antunes et al., 2011; Dang et al., 2012) UV shielding (Sun et al., 2007; Guo et al., 2007; Li et al., 2008) and absorption of microwave radiation (Bregar, 2004; Guo et al., 2007).

The electrically conducting polymers are extended systems of conjugated carbon-carbon double bonds (Figure 2). A doping process which involves reduction or oxidation reactions is responsible for the synthesis of conductive polymers provides significant conductivity to polymers (up to 105 S/m) whereas the conducting polymers composed of conductive fillers, form an individual conductive connected (or coherent) phase within a polymer matrix. No reaction or surface interaction is established between the filler and polymer matrix. Fillers are just dispersed and bound in a host polymer matrix. (Petrovic et al., 2005).

The description of the synthesis of first work of conducting polymers was published in 1862 by Henry Letheby (College of London Hospital). The Letheby (1862) prepared poly-aniline by anodic oxidation of aniline in sulfuric acid (called aniline black); it was partly conductive in nature. However, its electronic properties were not studied. Later on, (Natta et al., 1958) synthesized a highly crystalline semiconducting poly-acetylene (a black powder) with a regular structure having conductivity 10^{-11} to 10^{-3} S/cm (Pino and Moretti, 1987) which was curiously explored until 1977. Although, in the early 1970s, well-defined films of poly-acetylene were made successfully. It has conjugated double bond, not equal bonds. Except, a class of conducting polymers, the polymers were made, by addition of metal or carbon powder (discussed above).

Electronically conducting polymers: (Shirakawa et al., 1977) received the prestigious Nobel prize (Chemistry-year 2000) for the development of conductive polymers (Shirakawa et al., 1977). The properties of such polymers depend upon their electrochemical potential. Few conductive polymers are listed in Table 1 chronically along with their band gap and conductivities.

Table 1. Contains the list of conductive polymers

Conductive Polymer and their use	Band gap (eV)	Conductivity (S/cm)
Polyacetylene (1977)	1.5	$10^3 - 1.7 \times 10^5$
Polypyrrole (1979) (microwave absorber)	3.1	$10^2 - 7.5 \times 10^3$
Polyphenylene and analogues Poly(paraphenylene) (1979) (Electroluminescent display as mobile, telephone displays)	3.0	$10^2 - 10^3$
Poly (p-phenylene vinylene) (1979)	2.5	$3 - 5 \times 10^3$
Polyaniline (1980) (EMI shielding)	3.2	30- 200
Polythiophene (1981) (FET)	2.0	$10 - 10^3$

Electronic industry's demand for electrostatic protection and electromagnetic interference. (EMI) shielding requires electrically conducting polymers which are made by mixing of conductive fillers as Ag, Cu, and graphite. It increases the weight as 70% by weight with the polymer matrix. Such fillers cost high and deteriorate properties of the polymer.

Polypyrrole (1979) polyaniline (1980) and polythiophene (1981) are examples of self-conducting polymers. Chemically prepared poly-heterocyclic polymers as polypyrrole and polythiophene tend to be insoluble and infusible and having poor quality and low electro-conductivity as compared to electrochemically prepared poly-heterocyclic polymers. The advantage of the chemical oxidative polymerization is that it follows simple preparation methods and has short reaction times. Hence it is suitable for mass production (Shenoy et al., 2001). As an example, polypyrrole has a range of potential applications in electronic and electrochromic devices, drug delivery, lightweight rechargeable battery, anhydrous electrorheological fluids, membrane separation, sensors, chromatographic stationary phase, microwave shielding and corrosion protection (Ansari, 2006; Liu et al., 2006). Similarly, polyaniline has an electrochromic property. It opened the possibility for smart windows.

Since the conjugated systems have Monomer HOMO (VB)/LUMO (CB) states therefore, there is a band interval energy which depends on the molecular structure of the repeating unit and so optical and electronic properties can be controlled at the molecular level (Figure 2). Since the 1.7eV - 3.1eV is within visible spectrum, hence the conductive polymers are tunable for PV/LED applications. The original thought was to replace copper in printed circuits and transmission lines with light-weight and easy-to-process polymer materials. As discussed above that their traditional applications are an antistatic coating, smart windows, corrosion inhibitors and compact capacitors whereas the new applications include transistors, photovoltaics (OPVs), organic light-emitting diodes (OLEDs), and thermoelectric materials. Conductive polymers are easy to process at low cost.

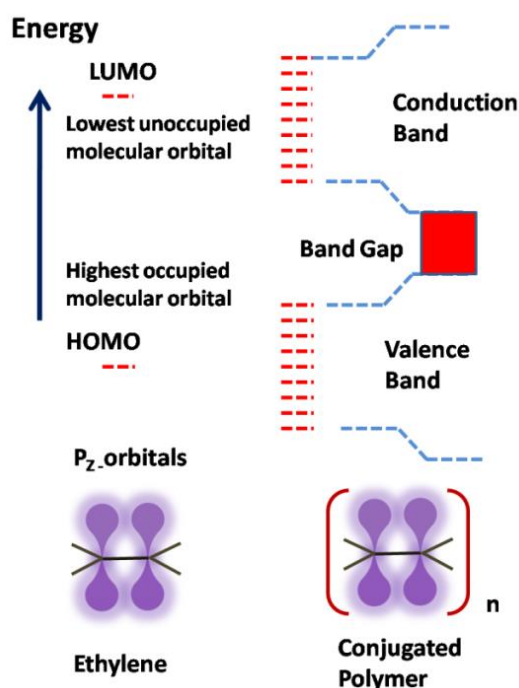


Figure 2. Bandgap in the conjugated polymer

As discussed above, the doping process is used to transform insulating polymers (e.g., semiconducting polyacetylene) in the category of excellent conductors. Addition of electron donors such as Na or K (n-doping, reduction) or electron acceptors such as I₂, FeCl₃ or AsF₅ (p-doping, oxidation) forms charge-transfer complexes. As a result of this, a doped polymer backbone forms the charged ions (Na⁺, I³⁻, K⁺, I⁵⁻, FeCl₄⁻, AsF₆⁻) hence, a negatively or positively charged are formed with the dopant. When an electric potential is applied across it, the motion of counter ions in and out enables to toggle polymer via conducting and insulating state (Figure 3) (Petrovic et al., 2005).

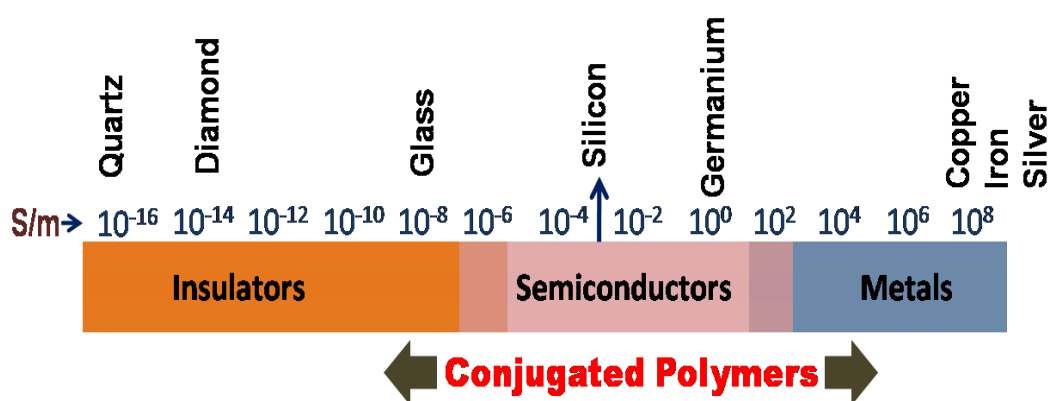


Figure3. Conductivity of conjugated polymers

7. Conclusions

The contemporary applications of the electronically conducted polymer are the transistors, photovoltaic, organic light-emitting diodes and thermoelectric materials. The large conductive polymers are used in printing electronic circuits as actuators, supercapacitors, electrochromism and chemical/biosensors. The processing with nanomaterial provides the higher surface area and better dispersibility and hence, better electrical and physical properties are obtained at low cost. The processability of polymer salts requires the oxidative doping because they are soluble in organic solvents and water. This makes the synthesis crucial and complicated. Still, in recent trends, the OLEDs and polymer solar cells are being promoted. Further, the composites have an enormous scope with emerging nano and smart materials. This helps in innovations to facilitate the lifestyle.

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