Journal of ELECTRONIC MATERIALS, Vol. 49, No. 3, 2020
https://doi.org/10.1007/s11664-019-07908-x

Thermally Conducting Polymer Composites with EMI Shielding: A review

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Polymer composites have been a material of choice for lightweight and durable applications in sectors ranging from automobile, packaging, structural components, and electronics to energy harvesting. Their versatility and ability to be tailored to application requirements have made them prospective alternatives for metal enclosures used in communication systems, power electronics, electric motors, and generators. The easy processing and high strength-to-weight ratio provide advantages over traditional materials that involve time- and-labor intensive processes. However, high thermal conductivity (TC) and electromagnetic interference (EMI) shielding are factors limiting their penetration into niche markets, and thus the development of alternatives with high TC and EMI shielding efficiency is critical. Thermally conductive polymer composites and EMI shielding effectiveness (SE) is a current issue in different applications including polymers providing light weight, corrosion resistance, and ease of processing as compared with metal. This paper focuses on improvements in the TC and shielding effectiveness of polymers by incorporating various fillers including carbon-based, mineral-based, and hybrid fillers. The paper reviews the current research worldwide regarding the enhancement of the TC and shielding effectiveness of polymer composites.

Key words: Thermal conductivity, EMI shielding, hybrid filler, polymer composite

INTRODUCTION

Polymers and composites are used extensively in electrical and electronic applications as enclosures with electromagnetic interference (EMI) shielding. In the modern electronics industry, thermal management is a critical issue. The power required for some processor modules can approach 250 W, and if the heat generated is not dissipated properly, then the system can break down. In order to resolve this issue, composites with high thermal conductivity and the ability to dissipate heat easily are strongly recommended.[1,2] Further, they provide design freedom for construction parts. With the use of various fillers and filling content, a particular adjustment of the thermal properties may be achieved.

Polymers have many advantages over metals due to their cost-effective, feasible processing methods and high corrosion resistance. Many polymers are electrical insulators, with TC of between 0.1 W/mK and 0.5 W/mK, which is due to the presence of an amorphous state. The three types of carriers found to transfer energy in solids are phonons, electrons, and photons. The quantized modes of lattice vibration in a stable crystal are called phonons, and they are an essential mechanism for the conduction of

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(Received April 17, 2019; accepted December 17, 2019;

heat in the polymer. Normally, a polymer has many defects in its amorphous phase, which contribute to the maximum phonon scattering, and due to this phenomenon, polymers have low TC.[3] Much research has been carried out over the past decade aimed at enhancing the TC of polymer nanocomposites using numerous fillers including boron nitride (BN),[4] carbon nanotubes (CNT),[5] aluminium oxide, and graphene.[6]

Another important requirement for a polymer enclosure is EMI shielding, which prevents interference of undesirable EM waves with the equipment. Many materials such as fabric and polymer-rubber blend composites are processed to inhibit the interaction of the electromagnetic radiation in the system. Fabric acts as protection to inhibit the penetration of radiation.[7–10] Enormous research efforts have been focused on developing cost-effective and lightweight EMI-protective materials which deal with the unfavorable consequences of EMI. When metals are used as EMI shields, their negative features such as heavy weight, physical stress, susceptibility to corrosion, and complex processability hinder their prospective end use. There is thus high demand for lighterweight, flexible, noncorrosive, and processable EMI protective materials.[11–13]

As compared with traditional metals, polymer nanocomposites offer an attractive alternative because of their lightweight, noncorrosive, and facile processability features. Metallic protective materials are only able to protect against EMI emission at the surface, while EMI shielding polymer nanocomposites comprise numerous fillers such as fiber nanoparticles, conductive polymers, and carbonaceous fillers that enhance absorption and dissipation while also limiting the reflection at the surface.[14,15] In the design of the material, polymer nanocomposites offer better flexibility than metals. The incorporation of fillers inside the insulating polymer matrix can provide the necessary mechanical and electric properties to meet various needs. The appropriate choice of geometry, morphology, and volume fraction of fillers can improve the EMI shielding properties of polymer nanocomposites.[3,16] A number of research works have investigated thermoplastic and thermoset polymer composites with regard to their EMI shielding properties. Compared with thermoset materials, thermoplastic composites offer improved recyclability and fracture durability, greater resistance to chemicals, less water-intensive production techniques, weldability, lower energy requirements, and reduced hazardous natural organic compounds and additives, making thermoplastic composites more eco-friendly.

FUNDAMENTALS AND MECHANISM OF THERMAL CONDUCTIVITY (TC)

TC refers to a material's ability to conduct heat (measured in units of W/mK). It can be described as the capacity of a material to transport a specific quantity of heat energy in 1 s via a plate of a specific area (1 m^2) and thickness of (1 m) when its opposite face differs in temperature by 1 K.[17] TC, denoted by λ, is defined as

$$= a \cdot r \cdot c_{\mathrm{p}} \tag{1}$$

where a is known as the thermal diffusivity, ρ is density and C_{p} is the specific heat capacity. The value of specific heat capacity C_{p} is obtained from the properties of a known reference material and the measured temperature differential. If the density of a material is known, then TC can be measured by Eq. 1. There are two ways to measure TC, through-plane and in-plane (Fig. 1).

For application in a heat-dissipating medium, the composite should exhibit through-plane conductivity. This assists in the transfer of heat from inside the enclosure to the outer surface, while in-plane conductivity assists in distributing the heat uniformly throughout the enclosure surface.

Fundamentals and Mechanisms of Heat Transfer and Thermal Conductivity

The modes of heat transfer are conduction, convection, and radiation (Fig. 2). Conduction involves the transport of heat through the direct collision of molecules. It enables the transport of thermal energy from an area of higher kinetic energy to an area of lower kinetic energy. In conduction, higher-speed particles collide with lower-speed particles, thereby increasing the kinetic energy of the lower-speed particles. Thus, conduction takes place through direct physical contact, and it is the most common form of heat transfer. Convection is defined as the transfer of heat through a fluid such as a liquid or gas. When such liquid or gas comes into contact with heat sources, it carries heat along, and this process is known as convection. The transfer of heat occurs due to the bulk movement of molecules inside the fluid. At the molecular level, the molecules expand upon the introduction of thermal energy. As the temperature of the fluid mass increases, the volume of the fluid increases proportionally. This effect causes displacement of the fluid. As the immediately heated air rises, it pushes denser, colder air down. This series of events is how convection currents are formed. The

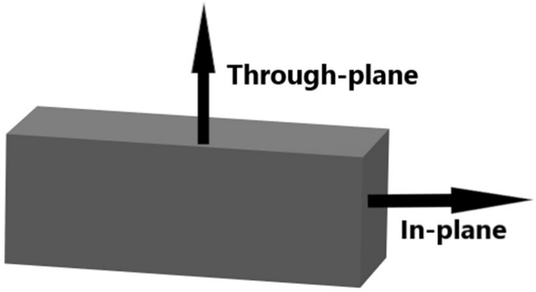

Fig. 1. Through-plane and in-plane thermal conductivity.

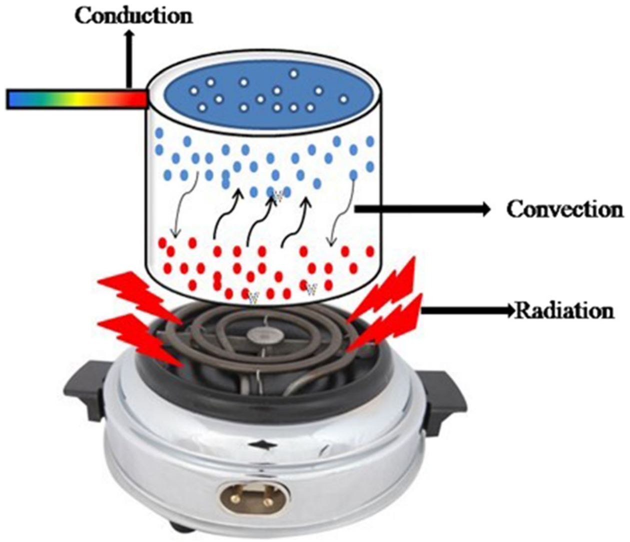

Fig. 2. General mechanisms of heat transfer.

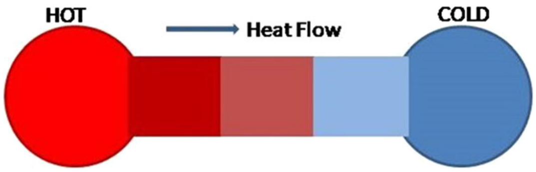

Fig. 3. Mechanism of heat flow in solid.

Table I. Thermal conductivity of various metals[19]

Material	Conductivity values at 25°C (W/mK)
Ag	406
Cu	385
Au	314
Al	20
Ni	158

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mechanism of radiation is different from the other two heat transfer methods, as it involves the emission of electromagnetic waves, which carry the energy from the emitting object. Radiation involves the transfer of heat energy with the aid of electromagnetic waves. Conduction and convection occur only when the object is in direct contact with the heat supply. In radiation, there may be no direct contact with heat energy. Radiation energy travels in a straight line through space and is converted to heat energy when absorbed. The converted heat energy is then transferred into the material with the aid of conduction and convection.

Thermal Conductivity in Metals Versus Polymers

The heat flow in a solid material is caused by phonons and electrons, but in metals, TC occurs because of the free carrier electrons. When the temperature of an object differs from that of its surroundings or any other object, the flow of heat energy from the hot surface to the colder one is also called heat transfer. It takes place in such a manner that the object and the surroundings maintain temperature equilibrium. The free electrons inside the metal can easily move throughout the solid material and transfer the heat energy from one end to the other, making them highly thermally conductive[18] (Fig. 3). The TC of various metals is displayed in Table I.

In the case of insulating materials, the lower heat conduction is due to tightly packed electrons which restrict the mobility of free electrons. Hence, the heat transfer may take place in the form of vibration of atoms. The flow of heat in polymers relies upon various parameters including crystallinity, temperature, and orientation of macromolecules. Phonons are commonly known heat transfer agents in the absence of free electrons.[20] When the surface of the polymer comes in contact with the heat flow, the transfer of heat to the primary atom of the molecular chain takes place in the form of vibration, then to the nearest atom, and then to the next atom. In a polymer, the flow of heat will not take place as a wave; it will diffuse slowly. While the tightly packed lattice of a good conductor facilitates the fast transfer of heat energy from the first to last atom, in poor conductors, the energy may be used for enabling the vibration and rotation of atoms, leading to considerable loss of TC.[21] The mechanism is displayed in Fig. 4. Crystallinity or orderly arrangement of atoms is a prerequisite for high TC. It also depends on the binding energy, hardness of material and stiffness of the structure. Polymers are generally amorphous or semi-crystalline in nature and never truly crystalline (Fig. 5).

In an amorphous structure, the molecules are arranged in random order; when the surface of the monomer collides with the heat source, the heat energy is transferred to the first atom nearest the heat source. The heat is then transferred to the next nearest atom, then to the next, and so on. In amorphous systems, propagation of heat no longer acts like a wave as in the crystal structures; however, the propagation is much slower in the case of a polymer, because of the disordered vibrations and rotations of polymer molecules around the equilibrium positions, scattered to nearby chains.[22] The TC of the different polymers is summarized in Table II. Li[23] studied the TC of polyamide 66(PA66), in which PA66 nanocomposites filled with flake graphite (FG) were prepared by twin-screw extrusion. The effects of filler content, particle size, and particle size mixing on the TC and the mechanical and rheological properties of the composites were studied. The results showed that as the FG content

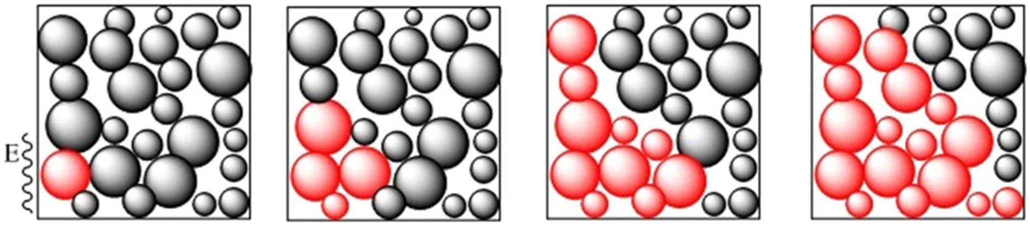

Fig. 4. The mechanism for TC in crystalline polymers. Reprinted with permission of Ref. 142.

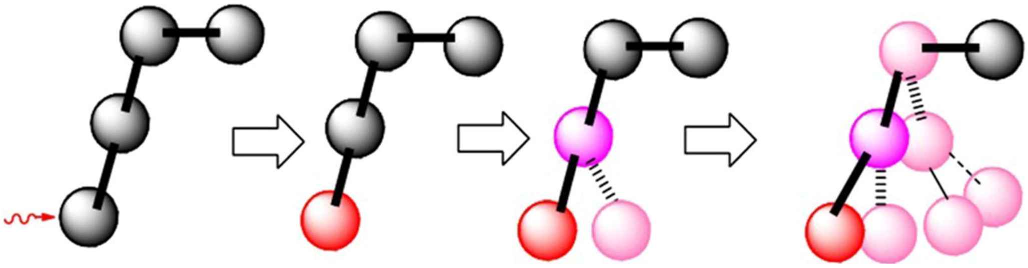

Fig. 5. Phonon transport in amorphous polymers. Reprinted with permission of Ref. 142.

Table II. Thermal conductivity of different polymers[19]

Polymers	Thermal conductivity at 25°C
Low-density polyethylene	0.30
High-density polyethylene	0.44
Polypropylene	0.11
Polystyrene	0.14
Nylon-6	0.25
Nylon-6.6	0.26
Poly(ethylene terephthalate)	0.15
Poly (butylene terephthalate)	0.29
Polycarbonate	0.33
Polyetheretherketone	0.25
Polyphenylene sulfide	0.30
Polysulfone	0.22

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increased from 0 wt.% to 50 wt.%, the TC of the composites filled with 100 μm FG gradually increased, whereas the mechanical properties and rheological properties decreased. At 50 wt.% loading, TC reached 3.07 (W/mK). With the increase in particle size, the TC and rheological properties of the composites improved, but the mechanical properties first increased and then decreased. The composite filled with 100 μm FG had relatively optimal mechanical properties.

Particle mixing has been found to improve TC, with maximum values achieved for 20 μm and 100 μm particles in a mass ratio of 1:2. Zhang et al.[24] fabricated carbon materials (carbon black [CB], multi-walled carbon nanotubes [MWCNT],exfoliated graphite [EG]) filled with polycarbonate (PC) matrix and obtained nanocomposites with high TC by including 10 weight fraction of EG filler to PC matrix; the TC value of the composite reached 1.06 W/mK, which was five times that of the PC. When the ratio of EG:MWCNT was 9:1, the TC reached the highest value of 1.19 W/mK, showing that the mixture of EG and MWCNT provided a synergistic effect. The TC of the EG/MWCNT/PC composite was improved by 26-fold over the natural PC resin at a loading of 40 mass%, from 0.21 to 5.76 W m^{-1} K^{-1}, which may also provide greater opportunity for its further use in electronics, aerospace, and LED lamps. Jia et al.[25] studied the enhancement of the TC of PA6 using low-melt-temperature (LMTM) tin (Sn) following a melt processing method (extrusion and injection molding). The authors found that the incorporation of Sn into the PA6/Sn matrix became constant due to the agglomeration of Sn. But when 20 wt.% (5.4 vol.%) of Sn was incorporated into PA6 containing 50 wt.% (33.3 vol.%) of graphite, the composite yielded a TC value of 5.364 versus 1.852 W/mK, which illustrates that the addition of graphite and Sn had an enormous synergistic effect in improving the TC of nylon6.

FUNDAMENTALS AND MECHANISM OF ELECTROMAGNETIC INTERFERENCE (EMI) SHIELDING

The use of electronic devices has accelerated in recent years, leading to greater demand for material used for EMI shielding. Devices used in present-day technology in the fields of defense, aerospace, telecommunications, electronics, and automobile manufacturing may also result in an excessive amount of EMI radiation, in turn leading to the disturbance or destruction of other devices. This affords a major opportunity for the further development of EMI shielding materials. Asia-Pacific and North America are the largest regions for the EMI shielding materials market and are predicted to retain a strong position in the future as well.

The basic principle of EMI shielding is to reduce or prevent the effects of EMI generated from electrical equipment in the working of another similar appliance. The effect of using a conducting material, creating current and magnetic polarization within the conducting material which is opposite the source of the electromagnetic field, and thereby reducing the effect of the radiation source, represents the shielding effectiveness (SE).[26] SE is defined as the ratio of incident electromagnetic waves to the reflected or transmitted electromagnetic waves at the same location, i.e., the attenuation value of the electromagnetic signal, in units o dB.

According to the Schelkunoff electromagnetic shielding theory, the shielding effect of a metal material is described as the combination of the effect of reflection loss of electromagnetic waves, the loss of absorption of electromagnetic wave and the loss of electromagnetic wave in the process of reflection within the shielded material.[27,28] Silver, copper, aluminum are excellent electrical conductors; their relative conductivity is high and the electromagnetic effect is mainly reflection loss. Iron and iron-nickel alloy are highly permeable materials; the relative permeability is very high and the electromagnetic shield is mainly based on absorption loss.[27,29]

The effectiveness of EMI shielding plays a vital role in the selection of shielding material, possessing a stealth effect. This can be calculated as the ratio of infringing energy to the residual energy. The infringing energy includes the portion of a penetrating electromagnetic wave into shielding material that is either absorbed or reflected, whereas residual energy is constituted of the energy that is neither reflected nor absorbed by the shielding material. Nevertheless, that part of the energy escapes from the shield. All the electromagnetic waves are composed of two major components, the magnetic field (H) and electric field (E).[30] These two fields predominantly intersect at a right angle with each other; later the interaction paves a directional pathway for the propagation of electromagnetic radiation represented in Fig. 6.

The total shielding effect after passing electromagnetic radiation through a shielding material can be calculated by the sum of absorption, reflection, and multiple reflection values, and is represented as

$$\mathrm{SE} = A + R + B \tag{2}$$

where SE is the electromagnetic shielding effect; R is the surface single reflection, A is the absorption, B is the internal multiple reflection attenuation (Fig. 7).

Reflection Reflection takes place when the shielding material has a high conductive capacity. The reflection of the incident wave can be dependent upon the wave frequency, ion charges and magnetic properties of the material. In the case of composite materials, the filler must contain free electrons.[31]

Multiple Reflections Multiple reflections occur because of the presence of various phases inside the shielding material. Materials having large specific surface areas such as foams and composites show multiple reflections of EM waves.[32]

Absorption For absorption, the shielding material must comprise electric and magnetic dipoles. Material with a high dielectric constant can provide an electric dipole, and material with high magnetic permeability provides a magnetic dipole for the absorption of EM waves. Electric dipoles present

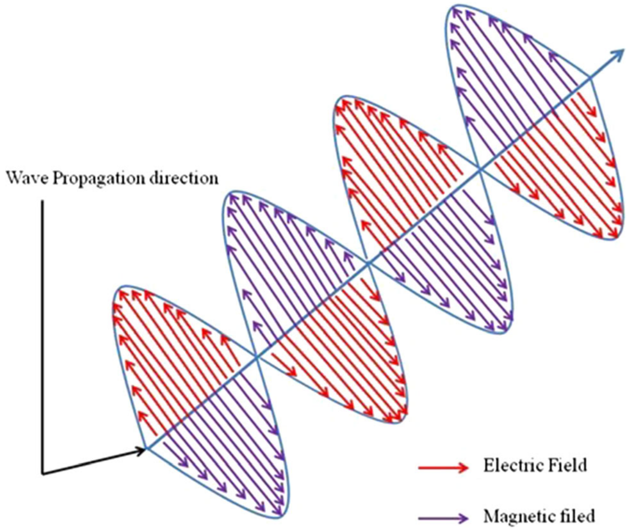

Fig. 6. Fundamentals of electromagnetic radiation.

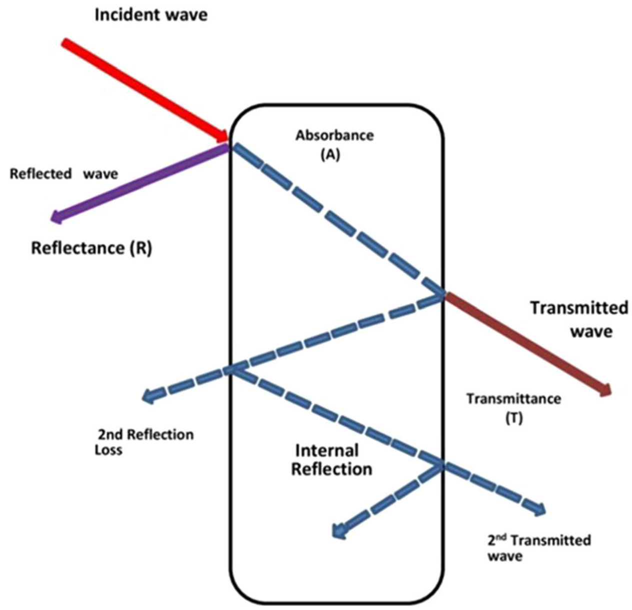

Fig. 7. Mechanism of EMI shielding.

within the shielding material can destroy the electric field of EM waves by converting it to heat energy. A material with good shielding properties obtains better absorption loss (insertion loss), lower volume, and surface resistivity properties.[16]

Transmission When electromagnetic waves penetrate the shielding material without any attenuation, the transmission of a high number of EM waves is observed. Many nonconductive materials such as glass and polymers provide small or zero transmission loss to the EM waves.

EMI Shielding in Metals Versus Polymers

In electromagnetic shielding applications, metal-based materials are often used due to their excellent electrical conductivity. The presence of free electrons and shallow skin depth in metal shielding of the electromagnetic wave occurs through surface reflection. Conductive metals such as aluminium, copper, chromium, and nickel have certain limitations, e.g. aluminum-based materials have low impact resistance and steel has higher density.[33] The disadvantages of using metals as EMI shielding materials are that they are heavy, costly, rigid, and provide poor corrosion resistance.[34] These limitations have driven research focusing on metal-coated materials; the coating on the surface of the metal can be achieved using different metallization methods. The common metals showing improved electromagnetic shielding include pre-tin-plated steel and copper alloys, and aluminium nickel, silver, and copper.

Pre-Tin-Plated Steel

Pre-tin-plated steel provides EMI SE in a wide frequency range from kilohertz to gigahertz. It is an ideal low-cost solution that works better in the low-frequency region. The lower hundred range of carbon provides lower-frequency shielding properties that are missing in copper alloy77 or aluminum. One of the major advantages in using pre-tin-plated steel is the corrosion resistance of steel to prevent rusting. Pre-tin-plated steel also provides good soldering properties, enabling easy assembly.

Copper

Copper is the most common material used for EMI shielding because it is incredibly effective in reducing magnetic and electric waves, such as reduction in EMI between a magnetic resonance imaging (MRI) system and a computer. The versatility of this metal makes it easy to couple with brass, phosphorous, bronze, and beryllium to form desired alloys. Copper is expensive relative to other shielding materials, although it provides better conductivity.

Aluminum

Aluminum provides good mechanical properties, a high strength-to-weight ratio, and high TC values, in addition to EMI shielding. When aluminium comes in contact with the atmosphere, a thin invisible oxide skin forms immediately, which protects the metal from further oxidation. This self-protection characteristic renders aluminum highly resistant to corrosion even in industrial atmospheres that often corrode other metals.

Metal enclosures are one of the most important parts for use in an EMI shielding composite, as they provide high TC along with better reflection properties. Metal enclosures with high-density matrices lead to increased weight of the entire assembly. This makes it difficult for military personnel to carry electronic appliances during mission-critical operations or high-altitude transport. When metal enclosures are used in wet and humid environments, oxidation may occur, and subsequently corrosion. This allows the growth of microbes on the surface, leading to further property deterioration. Metal enclosures must be machined via multi-step processes to obtain the desired surface textures, shapes, and sizes. Further, post-fabrication processes are required, which are expensive and time-consuming. Intricate shapes and designs are very difficult to incorporate into a metallic substrate, which in turn affects the visual appearance of the system. Modern sophisticated design flexibility is much less feasible in the case of metals as compared with their nonmetallic counterparts (Fig. 8).

In the quest for lighter construction materials, many researchers have chosen thermoplastics, which are simpler and faster to manufacture. With metals, the enclosure's shape must be stamped, a costly and time-consuming process that is not always easily adaptable to intricate shapes. Thermoplastics, however, may be easily molded into

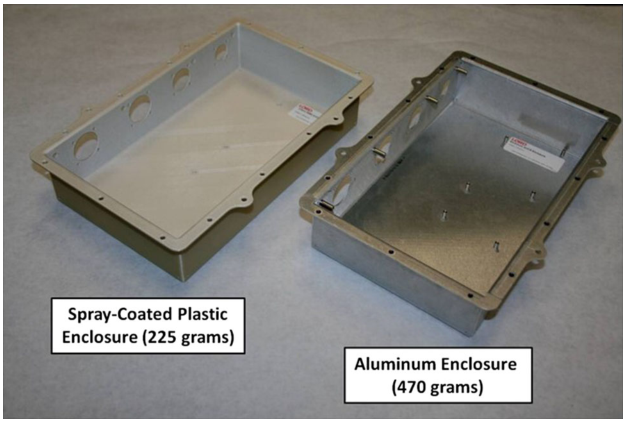

Fig. 8. Comparison between metal and polymer housing.

complex shapes by injection molding. This combination of lightweight design and manufacturing speed makes plastic enclosures more attractive than metallic enclosures. However, plastics are inherently electrically insulating, thereby providing negligible EMI protection. To overcome this problem, manufacturers often resort to the use of an electroless plating process or coating the plastics with heavily packed conductive coatings, which transforms the plastic into an EMI shield.

EMI Shielding in Polymers

As discussed in the previous section, EMI shielding in metals is due to reflection rather than absorption. They are commonly used because of the presence of electrons at the surface and low skin depths. Since the EMI shielding methodology (reflection or absorption) can be tailored by controlling electric conductivity and dielectric constant values, conducting polymers may have a variety of benefits over metal shielding, which is heavy, corrodible and physically rigid.[36] The skin depth of the polymer composite can be improved by reducing the size of filler.[37] The polymer composite with conducting filler is broadly used for the shielding application (Table III).

Because of the skin effect, a composite material having a large filler size is less effective than the composite with a small filler. The typically used metal sheet as a shield tends to leak the radiation and decrease the effectiveness of the shield. The addition of polymer matrix composite is attractive for shielding material. The effect of filled epoxy composites with organic as well as inorganic fillers has been studied by many researchers.[38,39] With the addition of 15–20% of CNT in epoxy and polyurethane, Li et al. and Liu et al. recorded an EMI shielding value of 15-20 dB. However, higher loading of fillers leads to difficulty in processing and a reduction in mechanical strength that must be addressed for developing a commercially viable product.

Song et al.[40] studied the effect of grapheme (GR) and carbon black (CB) on the conductive composite of PBT/GR/CB prepared by melt blending. The rheological properties, morphology, mechanical strength, electrical resistivity and EMI SE of the conductive composite were studied. The authors found that as the loading of CB increased, the electrical resistivity of the composite decreased, with values of 3.5 Ω at 35% graphite and 15% CB loading, and the highest EMI SE of 40–60 dB was found within a frequency range of 30–3000 MHz. The EMI shielding effect of paint-like nanocomposite layers containing graphene nanoplatelets and different concentrations of PANI/HCl and PEDOT/PSS was studied by Drakakisa et al.[41] As shown, optimized paint content was necessary to achieve uniform and homogeneous nanocomposite layers providing effective electromagnetic shielding. The electromagnetic shielding was studied within a frequency range of 4–20 GHz. Pardo et al.[42] investigated the effect of CNT on 5% loading, and PC composites gave at least initially appropriate conductivity values (> 1022 s/cm) for the application of EMI shielding. In this compression-molded nanocomposite, an SE value of 40 dB was found, which is suitable for use in electronics housings. Al-Saleh et al.[43] investigated conductive polymer nanocomposites with an extremely low electrical percolation threshold and found that extremely high EMI SE was created by simple wet-mixing. The electrical percolation threshold occurred at 0.054 vol.% CNT. The nanocomposites confirmed improvement within the EMI SE with the increase in CNT content, and the EMI SE provided independence on the frequency within the X-band range. For example, at 10 wt.% CNT, the nanocomposites showed SE of 50 dB. Wanga et al.[44] studied RGO/Mn_3O_4 nanocomposites synthesized using a one-step hydrothermal approach, which demonstrated greater dielectric loss properties attributable to Debye dipolar relaxation, interfacial polarization relaxation, and the unique conductivity of RGO. A much improved overall dielectric loss performance and EMI SE were achieved in comparison with pure Mn_3O_4 nanoparticles. These nanocomposites may be suitable for application in microwave technology and can also be extended to other areas including supercapacitors and lithium-ion batteries. Liu et al.[45] prepared polymer nanocomposites containing epoxy as matrix and packed with nano-sized MWCNT, Fe3O4, and Fe as filler for application in EMI shielding. The nanocomposite specimens contained different filler systems with different weight fractions, and their absorption value was characterized using a vector network analyzer. The experimental data revealed better absorption for tri-layer laminated nanocomposites, with values up to 40 dB over a frequency range of 13–40 GHz in the high-frequency region.

CHALLENGES AND ASSOCIATED FACTORS

Plastics are inherently electrically insulating, and thus impart negligible EMI protection and very low TC. In order to enhance the TC and SE, researchers are now focusing on the addition of conducting filler. The addition of a higher percentage of filler can improve both the TC and SE value, but it creates processing challenges. For industry, the effective processing of a material is more critical than in experimental research. Defects at the atomic scale can have serious implications for TC, and macroscopic defects also lead to more severe consequences. Porosity is a macroscopic defect that arises due to high filler loading in the composites and causes decreases in TC. The microscopic voids in the polymer matrix lead to phonon scattering. Burger et al.[46] showed that a pressed sample showed higher TC than the molded sample only above a certain loading of filler in the polymer matrix. At this particular filler loading, the composite provided relatively high viscosity, which was attributed to better filler dispersion. When the viscosity of a sample increases, the porosity of the cured sample will increase. This problem can be solved using a press-molding process, and the pressed sample achieves better TC.[47] A major processing challenge reported in the literature involves the mechanical approach. In order to enhance the mechanical properties of the composite, it is necessary to achieve the proper dispersion state of the filler in the matrix, using methods such as roll milling,[48,49] press molding,[50] and mixing.[51] Chakraborty et al.[52] showed that roll mill mixing increased the dispersion of CNT into an epoxy matrix, but re-agglomeration of the CNT occurred during the curing process. Ming et al.[53] reported that the conductivity of the composite increased by three to four times within the direction of pressing for a metal–organic framework loaded with graphite. A current challenge in the processing of composites is the enhancement of both the thermal and shielding properties by the addition of high filler loading to the composite.

FACTORS AFFECTING THERMAL CONDUCTIVITY AND EMI SHIELDING IN POLYMERS

Types and Classes of Fillers

Since most polymers have very low TC, researchers have sought methods to improve both TC and EMI shielding. However, the outcome is dependent on the shape and type of fillers in addition to their properties. A number of fillers have been studied for their effect on TC and EMI shielding, including carbon-based fillers,[54] graphite,[55] carbon nanotubes,[56] carbon black,[57] graphene,[58] metallic fillers,[59] silver, copper,[60] aluminium,[61] TiO_2,[62] ceramic fillers,[63] boron nitride,[64] and silicon.[65] The majority of these studies have chosen carbon nanotubes as organic fillers and boron nitride in the inorganic class.[66] High TC depends upon various factors, including filler particle size, purity, and loading. Materials including fibers are highly anisotropic and often provide much better conductivity along the principal plane. Excessive filler loading above 30% is needed to attain an appropriate TC value for polymer composites, which creates a challenge in processing the composites.[67] Moreover, the use of high inorganic filler loading significantly affects the polymeric composite mechanical behavior and bulk density.

Carbon-Based Fillers

Carbon-based fillers such as graphite, carbon fiber, and carbon black possess high TC and are lightweight. Graphite, a common and easily available form of carbon, is widely used as conductive filler because of its high TC, cost-effectiveness and uniform dispersion within the polymer application (Table IV). A polymer/vapor-grown carbon fiber (VGCF) composite was recently studied by Chen et al.,[37] as VGCF mat-reinforced epoxy composites showed an unprecedented high TC of 695 W/mK with a reinforcement of 56%, by volume, of heat-treated VGCF. Carbon black is also used as conducting filler in the polymer matrix, and it demonstrates superior electrical conductivity as well as TC properties.[70–72] Alexander et al.[60] reported that a single graphene sheet showed high thermal conductivity of about 800 W/mK. Although graphite is less expensive than all other carbon-based fillers such as single-walled nanotubes (SWNTs) and VGCFs, using graphite in higher loading leads to increased viscosity and difficulty in processing. The use of exfoliated graphite flakes has helped in opening a wide variety of applications, where high TC, electromagnetic shielding gas barrier resistance is necessary.

Compared with metals, carbon-based composites are largely used in shielding material due to their light weight, flexibility and anti-corrosion properties. A different form of carbon filler, such as graphite, carbon black and carbon fiber, is used as fillers for the EMI shielding composite. It was reported that carbon fiber has good electrical conductivity properties and mechanical strength. Carbon fibers are used extensively as conducting fillers in shielding materials to enhance SE.[73] Jana et al.[74] investigated various aspect ratios of carbon fiber for the shielding efficiency in a frequency range of 8–12 GHz. They found that a highly conducting network was formed within the composite with a higher fiber aspect ratio, helping to enhance EMI properties.

Metallic Fillers

The addition of metallic fillers in the polymer increases the electrical as well as TC in the

Table III. Shielding effectiveness of polymer composites/nanocomposites with the varying volume fraction of filler loading

Composite system	Volume %	EMI shielding effectiveness (dB)
PBT + graphite + carbon black	35/15	64
Reduced graphene oxide (rGO) + carbon fiber (CF)	0.75	37.8
Carbon nanofiber (CNF)	3–8	30–50
Nickel + polyaniline (PANI)	5	60
Carbon fiber/PES	29.5	65
UHMWPE/CNT	10	50
Nickel-coated carbon fiber/PC/ABS composites	22.3	47
Graphite nanosheets	–	38
PAN/PVC/PMMA polyester composites	–	50–60
High-density polyethylene/vanadium trioxide (HDPE/V_2O_3)	≈ 30	54
PANI + PVC	–	55.2
PPY + polyethylene	–	40–50
Polycaprolactone/CNT nanocomposites	0.249	60
Polyethersulfone + nickel coating	7	87
Al flakes/PES	20	4.3
Ni fiber/PES	7	2.7
Stainless steel fiber/PC (polycarbonate)	1.1	1.7
MWCNT/PAK (polyacrylate)	8.1	1.9
CNT BP (buckypaper)/PE	38	1.9

Only the data from Przemyslaw et al.[143] are used.

Table IV. Thermal conductivity of carbon-based filler[19]

Filler type	Thermal conductivity at 25°C (W/mK)
Carbon black	6–174
Graphite	100–400
Diamond	2000
CNT	2000–6000
Carbon fiber (PAN-based)	8–70
Carbon fiber (Pitch-based)	530–1100

Material from Dehaghani et al.[19] is from open access sources available under a Creative Commons license.

Table V. Shielding effectiveness of different metals. Reprinted with permission of Ref. 35

Filler system	Volume %	EMI shielding effectiveness (dB)
Ni fiber (2 mm dia × 2 mm)	7	58
Steel fiber	20	42
Carbon filament	7	32
Al flakes	20	26
Ni particles	9.4	23
Carbon fiber	20	19
Ni fiber (20 mm dia × 1 mm)	19	5

composites obtained. However, the density of the polymer composite is also increased with the incorporation of metallic fillers. Metallic fillers used to enhance TC include aluminum, copper, silver, and nickel, which have high thermal and electrical conductivity. Polymers such as polyethylene,[75] polypropylene,[76] polyamide,[77] polyvinylchloride and epoxy resins[78,79] reinforced with metallic fillers have been studied. The properties of TC may depend upon the TC of metallic fillers, their particle sizes, the volume percentage and the proper mixing of fillers in the polymer composites. Metals, owing to their high electrical conductivity, are widely used in EMI shielding applications.

Conductive solid fillers such as metallic powders, metal flakes, metal-coated fibers, and metal nanowires are mixed with polymer matrices for developing conductive composites (Table V). Polymer nanocomposites with high EMI shielding properties can be prepared as bulk, foam and layered structures. Metals can absorb, reflect and transmit electromagnetic radiation easily due to the high conductivity. Utilizing metallic filler renders thermal and electrical conductivity to inherently insulating plastics and rubbers. This helps to dissipate the static charge and heat developed in electrical equipment. The most common metallic fillers used for shielding material are aluminium flakes, copper fibers, steel fibers, etc. Baker et al.[80] studied the EMI SE of stainless steel fiber in an acrylonitrile butadiene styrene (ABS) polymer matrix. The incorporation of conducting steel fiber inside the polymer matrix created a conducting mesh, which may be used for EMI shielding purposes. The SE obtained was found to be 11 dB in the X-band region. They also found that the shielding value was doubled by doubling the thickness of the sample. The SE of the composite material was achieved with the aid of the alignment of the fibers in the material, incident electric field, opening size and properties of the material. This composite material is used for shielding purposes and for the construction of electromagnetic absorbent walls.

Ceramic Fillers

To enhance electrical conductivity, metal and ceramic particle-reinforced polymer composites have been widely used as electronic materials. Ceramic materials such as BN, aluminum nitride, silicon carbide, and beryllium oxide have gained increasing attention as thermally conductive fillers due to their high TC and electrical resistivity properties.[81,82] Various factors can affect the TC of a ceramic–polymer composite, including filler packing density,[83] particle size and size distribution,[81] surface treatment,[84] and mixing method.[85] Among the different ceramic substances, Al_2O_3 has been shown to provide excellent chemical stability and low cost compared with other ceramic materials, while Al_2O_3 has exhibited much lower TC of between 30 W/mK and 42 W/mK, which is very low compared with other ceramic materials. SiC has a wide variety of applications in high-temperature electronic devices because of its high TC and stability. However, SiC has a very high dielectric constant (40-1 MHz), which presents limitations with regard to highly integrated electronic devices. Si_3N_4 has provided much lower TC than AIN or BN, but it has advantages over AIN and BN in terms of good chemical stability, cost-effectiveness, and higher erosion resistance. This has been confirmed by various industrial applications.[86,87] Because of the presence of a honeycomb molecular structure in BN, it shows an anisotropic heat flow. The TC value will be 20 times higher (600 W/mK) when BN nanosheets are aligned perpendicular to the C-axis. Therefore, BN nanocomposites show higher TC by following a specific procedure.[88]

Yuchang et al.[89] studied the electromagnetic shielding properties of graphene nanosheets and Al_2O_3 ceramic composite in a frequency range of 8.2–12.4 GHz. They found that the GN/Al_2O_3 ceramic showed high mechanical strength, tunable electromagnetic properties, and good microwave absorption and overall EMI shielding performance, demonstrating a remarkable capacity for implementation in microwave applications. Zhang et al.[90] prepared a multilayer film via a casting layer with the aid of the layer. The unique structure of the film provided a high electrically and thermally conductive network in the in-plane direction, and the ordered multilayer film exhibited excellent EMI SE of 37.92 dB.

Shape and Size of Fillers

The shape and size of the filler are a key factor determining TC and mechanical strength.[91,92] Zhou et al.[93] used Al_2O_3 particles of different shapes and sizes to fill silicon rubber and noted that the nanosized Al_2O_3 composite had greater thermal and mechanical strength than the micro-sized Al_2O_3 composite. The particle size and content of the composite determine the average inter-particle distance,[94,95] which is related to the TC and mechanical properties in the composite. A small particle size results in lower particle distance and increased chance to form the TC pathway. Hence, the optimization of particle size and content may be a very facile and feasible way to prepare composites with good synthetic properties (Fig. 9).

The morphology of the filler is also a key factor for a shielding composite material. Various important

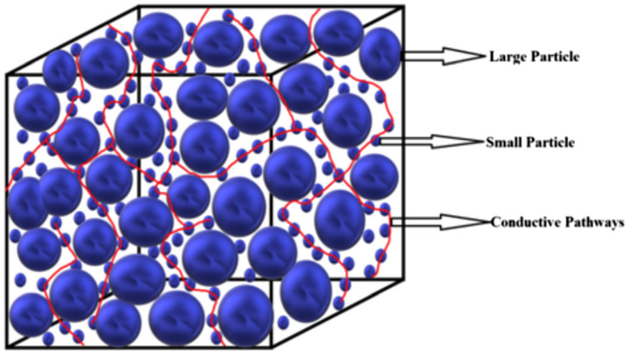

Fig. 9. Schematic diagram showing the distribution of a hybrid of large- and small-sized particles.

parameters will affect the properties of a composite material, including the shape, dispersion, and particle size of the filler. The filler size inside the polymeric composite also affects the tribological behavior, dielectric loss, and overall electrochemical performance, thermal stability, conductivity, and mechanical properties.[96–102] Koops studied the effect of the particle size of the filler on the shielding performance of different phases of a composite[103] and found that the overall shielding properties increased as the filler size decreased. Dong et al.[104] investigated the effect of WO_3 particle size on epoxy-based composite shields using low-energy gamma rays. They found that the linear attenuation coefficients of the composites increased because the filler particle length decreased. Azman and colleagues considered the effect of filler particle size on tungsten oxide-epoxy composites. They reported that the nano-sized WO_3 yielded higher attenuation values than the micro-sized particles in lower tube voltages (25–35 kV); however, at higher x-ray tube voltages (40–120 kV), the effect was negligible.

Hybridization of Fillers

Hybrid reinforced polymer composites offer greater advantages because the properties obtained by the hybrid composite are better than those of the mono-filler-reinforced composite.[105] Two or more different fillers can be used, as in the case of a three-component compound, in which the properties of the different components interact synergistically.[106] The hybridization of two-filler systems in the same matrix provides another dimension to the potential versatility of the parent system. The super microstructure produced in the nanofiller leads to enhanced properties.[107] In polymer nanocomposites, diffusion and transport are dependent on the characteristics of the filler, the degree of adhesion, and their compatibility with the polymer matrix. If the filler is compatible with the polymer matrix, it will take up the free volume in the matrix and produce a tortuous pathway for the permeating molecule. If the filler system is not compatible with the matrix, a void occurs at the interface, which increases the free volume inside the matrix and thereby enhances its permeability. Organic/inorganic hybrid fillers can produce ultrafine dispersion of the filler as well as local interaction between the matrix and fillers, leading to improved properties. According to research, high-efficiency TC can be obtained by using two or more fillers in the same composite. Teng et al.[108] investigated the impact of a hybrid composite between BN and MWCNTs. They found that the TC of the epoxy-based composite was increased by 740% (1.9 W/mK) with the use of a hybrid filler of 30 volume fraction of BN and one volume fraction of MWCNTs. Yang and Gu[109] studied the effects on TC of epoxy-based nanocomposites in a hybrid filler system using CNT and SiC modified with silane. They found that the hybrid filler system offered a superior result and also reduced the cost.

The hybrid filler system also produces a synergistic effect on the shielding composite. Researchers are now developing a plastic enclosure for electronic devices, due to their flexible design and low cost compared with metal. Plastics are electrically insulating, so they show transparent to electromagnetic radiation. Because of the insulating properties of plastics, scientists have investigated EMI-shielding conductive composites using conductive fillers with various conductive fillers including CNT, graphite, carbon fiber, nickel-coated carbon fiber, and carbon black. An EMI shielding value of 50 dB was able to achieve 99.7% shielding efficiency for EMI radiation. Many types of conductive fillers can be used for EMI shielding, but the use of a hybrid filler system inside the polymer matrix can achieve an optimal EMI shielding level. To achieve 99% shielding efficiency, a single filler system of more than 50% is necessary to overcome the problem of high filler loading; scientists have used hybrid conductive fillers including CNT with carbon black,[110–113] CNT with carbon fiber,[114,115] CNT with graphite,[116] carbon fiber with carbon black,[117] and carbon fiber with graphite.[118] In a study of a PP/NCCF composite hybridized with carbon black (3 wt.%),[119] EMI shielding of 44.5 dB was reported, which was much higher than that of a PP composite with NCCF at a frequency of 1.0 GHz.

Filler Loading

Electrically conductive polymer nanocomposites are the focus of a great deal of attention in electronic devices and have been studied in a large number of research areas. Conductive polymeric composites can be utilized in various electronic devices including light-emitting diodes, semiconductors, batteries, EMI shielding, antistatic anti-corrosion coatings, and other functional applications.[120–122] To achieve high TC properties in polymer composites, high filler loading is typically needed, which creates a significant processing challenge. Moreover, the use of a high volume percentage of inorganic filler loading in the polymer can significantly affect the polymer mechanical properties and density. For these reasons, currently, achieving polymer composites with TC higher than 4 W/mK and easy processability is very challenging. The addition of a compatibilizer processing aid and hybridization of fillers can facilitate the processing of polymer nanocomposites to achieve high TC.

EMI efficiency in a range of 8.2–12.4 GHz is necessary for all electronic enclosures. Typically, carbon nanotubes are used as conductive filler for the fabrication of EMI composites. To achieve 90% shielding, a conductive filler of more than 30% is necessary, which creates a processing challenge. At higher filler loading, the mechanical strength of the composite system decreases because of the lower filler–matrix interaction.[123] The incorporation of carbon black into the polymer matrix as conductive filler presents major disadvantages, as the higher amount of carbon black needed to achieve 30– 40% conductivity results in deterioration of mechanical strength.[124] A major advantage in using CNT as conductive filler is that the conducting network can be easily formed by a lower amount of filler loading, due to the percolation thresholds of CNT. For suitable EMI shielding systems, lightweight and mechanically strong materials are more effective. Much research work has been reported on EMI shielding based on CNT polymer composites. Yang et al.[125] reported an EMI shielding value for CNT/polystyrene (PS) composites of about 20 dB at 7 wt.% filler loading. The authors found that the composites were more reflective than absorptive of electromagnetic radiation. Yang et al.[126] reported the effects of carbon nanofiber and CNT inside the PS matrix and reported that the addition of 1 wt.% CNT to a 10 wt.% carbon fiber/PS composite obtained a shielding value of 20.3 dB. Kim et al.[126] investigated the effect of MWCNT on a polymethyl methacrylate (PMMA) film matrix and found a shielding value of 27 dB within a range of 50 MHz to 13.5 GHz. Yuen et al.[127] investigated the processing effect and EMI efficiency of an MWCNT/PMMA composite. The composite was prepared by *in situ* polymerization and *ex situ* polymerization. The authors found that the composite prepared by *in situ* polymerization showed higher shielding than the composite prepared by *ex situ* polymerization. Liu et al.[128] reported the effect of 20 wt.% SWCNT in a PU/SWCNT composite, in which the SE reached 17 dB in a range of 8.2–12.4 GHz.

MOLDABILITY OF CONDUCTING COMPOSITE

According to a recent study, polymer-based nanocomposite materials have great advantages in electronic devices and making of enclosures. Researchers are now focusing on exchanging metal parts with polymer nanocomposites for applications in various sectors, as polymers have several advantages including low weight, anti-corrosion resistance, and easy processability. The addition of a fiber-reinforced composite can yield mechanical properties similar to those of metals, although metal has superior thermal properties compared with polymers. Replacing metal with plastic in various sectors requires high TC. It is known that polymers are good insulators, so they offer negligible thermal properties compared with metals. The insulating properties of the polymer cause transparency to EMI radiation which is necessary for making enclosures for electronic devices. Adding nanofillers such as carbon-based, metallic, or ceramic fillers to plastics can significantly increase thermal and shielding behavior of polymers. To achieve sufficient TC and shielding, more than 30% filler loading is required, which causes unavoidable processing challenges, as the viscosity of the compounds will increase significantly. Thus, extrusion and injection molding restrict the number of fillers and the maximum TC. Excessive loading also dramatically affects mechanical behavior and density.[129]

The most commonly used process, especially in automotive and electronic parts, is injection molding. Many different properties are directly influenced by the process parameters. An optimization technique for the application of product design to mold design and selection of material can be achieved following the Taguchi method. Villmow et al.[130] explored how the parameters of injection molding could affect a composite prepared with polypropylene and CNT using a four-factor design evaluating the effects of holding pressure, injection velocity, mold temperature, and melt temperature of the composite. They found that better dispersion took place when the composite had a lower melt temperature and higher injection velocity compared with injection molding at low velocity and high melt temperature. Chandra et al.[131] reported that to achieve high TC on a polycarbonate matrix, better dispersion of CNT was necessary, and CNT-based nanocomposites should be processed at high melting temperature and low injection speed to achieve proper TC.

Stan's group[132] investigated the effect of processing parameters on mechanical strength in a PP/CNT composite. The degree of crystalline morphology of the molded polymers was affected by the injection molding parameters, which affected the mechanical properties of the injection-molded parts. Additionally, the use of a compatibilizer led to changes in the optical parameters for nanocomposite materials processed by injection molding. Compatibilizers enhance the interfacial interaction among the different polymers which are immiscible in nature. They normally block copolymers. These molecules generally tend to recognize the interfaces and stabilize them, thus permitting finer dispersion of collectively incompatible pairs. Coupling agents enhance the interfacial properties of fillers and polymers. Typically, a coupling agent reacts with modified filler surface. However, it exhibits one side group for the interaction of polymeric material.[133] The incorporation of a compatibilizer in the polymer matrix can achieve miscibility, easy processing, and melt flow properties.

PROSPECTIVE APPLICATION OF EMI SHIELDING: THERMAL CONDUCTIVITY

Soldiers and marines are highly dependent upon electronic devices for communication, navigation, and situational awareness. Portable electronic devices (PEDs) are electronic devices such as phones, handheld radios, computers, and tablets that can be easily carried. Such electronic devices are widely used by soldiers and military personnel for various communication purposes. In communication systems, the EMI and thermal dissipation properties of a material play a vital role. The basic requirements for EMI shielding in military applications can be described in two steps. The first step involves averting the ambient EMI sources penetrating the sensitive surroundings containing electronics equipment. The second step involves averting the electromagnetic radiation produced by the equipment being transmitted or conducted through the EMI shielding material. EMI shielding arises when unwanted electromagnetic radiation passes through electronic devices. The phenomenon of EMI shielding depends upon the absorption and reflection of EMI radiation by a material that acts as a barrier. The rapid growth in the field of electronics and radiation sources will increase the demand for EMI shielding. Controlling EMI radiation through the use of a shielding material plays a vital role in military applications. The internal design of the shielding material helps to control EMI radiation in industrial electronics. Much research work has been dedicated to improving the EMI shielding properties of thermoplastic polymers. Singh et al.[134] fabricated an MWCNT/PC composite using a micro-twin extruder with different mass fractions of MWCNT. The composite with 10 wt.% loading of MWCNT showed SE of −27 dB, which was considered for use as a high-strength EMI shielding material. Arjmand et al.[135] studied the EMI SE of a compressed 5 wt.% MWCNT/PC composite and found that the SE value reached 25 dB in the X-band. Pande et al.[136] fabricated an MWCNT/PC composite with high-pressure and low-pressure compression composites. They found that the EMI shielding value of the high-pressure 10 wt.% MWCNT reached 21 dB, whereas the low-pressure MWCNT/PC composite reached 35 dB in the X-band. Gupta et al.[137] studied the EMI SE value of a poly(trimethylene terephthalate) [PTT]/MWCNT composite with different weight fractions of MWCNT loading. The composite showed an SE value of 36–42 dB in a frequency range of 12.4–18 GHz (Ku-band) at 10 wt.% loading of MWCNT.

Thermal management is essential for the dissipation of the heat generated from electronic devices and circuitry during operation. It enhances device reliability and reduces the risk of premature failure. The amount of heat generated is proportional to the power input, provided no other source of energy is involved. Therefore, heat removal and thermal management open an important avenue for research and development of new material for easy dissipation of heat from electronic components. The greatest challenge with thermal management in military applications is the varying boundary conditions that are encountered in a short span of service life. During the course of mission operations, electronic devices are subjected to extreme conditions, which may affect the performance and life of the device. Military defense requires a wide range of computing assessment under various harsh environmental conditions. Inside the circuit board, applications facilitated by higher processing power are necessary. This leads to difficulties in thermal management with higher levels of processing power needed for high-end applications, which can be achieved by replacement of the cooling system for effective heat dissipation. With the increase in multiple slots/internal equipment, heat dissipation in an enclosure increases, resulting in damage to the instruments. Operating altitude is another important consideration, as the density of air decreases at higher altitudes. The result is a reduced cooling capability of air at the same rate. Specifically, at 50% air density reduction (approximately 20,000 ft altitude), the cooling efficiency is decreased by 50%. A number of studies have been reported in relation to the design and development of thermally efficient materials. Zhou et al.[138] prepared a polyamide 6/polycarbonate (PA6/PC) immiscible blend at a ratio of 7:3 using a twin-screw extruder. After the addition of 40 wt.% flake graphite, a TC value of 2.716 W/mK was reached, which was 30.3% higher than that of the mono-PA6/graphite composite. Zhang et al.[139] fabricated a composite using CB, MWCNT, and exfoliated graphite (EG) as filler, and found that the incorporation of EG resulted in higher conductivity. To further improve the TC of the PC matrix, they incorporated a hybrid of EG/MWCNT with a ratio of 9:1, and a TC value of 1.19 W/mK was reached. They also found that the TC value of the PC matrix increased to 5 W/mK when the hybrid filler of EG/MWCNT was increased to 40 mass%, which broadens its field of application in electronic devices and aerospace. Yu et al.[140] formulated a PC/ABS blend composite containing graphite nanoplatelets (GNPs) that were prepared using melt blending. The addition of GNPs to the PC/ABS blend was found to have a synergistic effect, increasing the TC to 3.11 W/mK at 70% weight fraction of the filler. Feng et al.[141] formulated a composite of polypropylene (PP)/flake graphite at a loading of 21.2 vol.% and achieved a TC of 5.4 W/mK.

FUTURE SCOPE AND CONCLUSION

Because of the rapid growth in electronics and communication systems, heat management and shielding of electromagnetic radiation is a critical issue for device design and application. Generally, metals are used for heat dissipation and radiation shielding, but many attempts are being made to replace metal with high-thermal-conductivity polymer-based composites, due to their light weight, chemical stability, easy processability, and low production cost. A high-thermal-conductivity composite can be prepared by incorporating conducting fillers into the polymer matrix. In this review, we have shown the effect of various factors including filler loading, filler shape, hybridization of fillers, and filler type and class on the EMI shielding and TC of polymer composites. While the incorporation of nano- and micro-sized fillers increases the TC, it was found that the hybridization of fillers is more effective. A combination of fillers with different shapes facilitates the formation of continuous networking, thereby effectively transferring heat from one surface to the other. It was also observed that the incorporation of carbonaceous fillers positively influences the effectiveness of EMI shielding while also enhancing the TC. Similarly, properties such as the coefficient of thermal expansion and the dielectric properties of the composite need to be considered in order to develop commercially feasible components. From the literature survey, it was observed that more in-depth analysis is needed to evaluate the moldability of polymer composites with high loading of conductive fillers.

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