The Effect of Fillers on the Tribological Properties of Composites

R. Muraliraja, T. R. Tamilarasan, Sanjith Udayakumar, and C. K. Arvinda Pandian

Abstract Recent advances in the sphere of materials engineering have seen the advent of a new generation of composite materials that have become a replacement for the traditional materials used in different industries over the years. The proven potential of composite technology to deliver in line with critical global trends in the aerospace, automobile, and marine industries has placed it on top of other materials in the market. As composite materials can be lightweight and durable, they score very highly on efficiency measures with desired engineering properties required. A careful and wise combination of matrices, reinforcements, and additives can be tailored for specific applications of the end product. Traditionally, most of the fillers were considered as additives, limiting their contribution to a composite only on reducing their cost. However, the diversity of applications and a broad spectrum of their usage has led to high demand for incorporating fillers in composite technology. In this perspective, the objective of this chapter is to explore the works of literature for providing information about the fillers concerning processing, functions, mechanical and tribological characteristics, environmental impact. Moreover, the recent advances and challenges in employing different types of fillers in different classes of composites have been briefly discussed.

Keywords Fillers · Composites · Wear · Friction

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1 Introduction

"Filler is a solid material capable of changing the physical and chemical properties of materials by surface interaction or its lack thereof and by its own physical characteristics" (Wypych 2016). Fillers are used for diverse industrial applications all over the world based on the nature and needs of the application. These additive components are incorporated in a mixture to elevate the properties and reduce the cost of the composites. The fillers are commercially available in the form of gases, solids, and liquids. Based on the requirements of the material's applications, the fillers are selected among the various available categories. These fillers not only reduce the cost of the composite but also improve the processing and mechanical behaviour of the composite. They serve for a variety of purposes in different industrial applications such as adhesives, agriculture, aerospace, appliances, automotive materials, bottles and containers, building components, business machines, cables and wires, coated fabrics, coatings, paints, face creams and powders and health care medicines, composites used for dental applications, fibres, film, foam, food feed, friction materials, geosynthetics, hoses and pipes, magnetic devices, medical applications, membranes, noise dampening, optical devices, paper, railway transportation, roofing, telecommunication, tires, sealants, sports equipment, waterproofing and windows (Wypych 2016).

Initially, fillers were mainly used to reduce the cost of the material, but it is also subjected to particle size considerations. The effectiveness of a filler addition is influenced by several factors such as purity, surface preparation, shape, and distribution. Fillers can also be used to vary the density of a material. High-density polymer material up to 2 g/cm^3 can be obtained with the help of fillers. Similarly, low-density polymers with lighter materials like foam as a filler can also be obtained (Chiellini and Solaro 1996). The colour of a material can be altered with the help of metallic powder fillers wherein the final polymer gazes like a composite metal. The surface properties of a material can be improved using graphite, PTFE, MOS_2 as fillers to reduce the coefficient of friction, as these types of fillers possess self-lubrication properties (Kano and Akiyama 1996). The shape of a material can be retained in polymer foams with the help of fillers. Besides, the use of hollow spherical particles as a filler provides the best insulation to electric and thermal conduction. The porosity of a material can also be influenced by the right choice of fillers (Srirangan and Paulraj 2016). Fillers are known to significantly affect most of the mechanical properties of materials (Asadi et al. 2011). Fillers contribute towards reducing fire accidents by reducing the auto-ignition temperature, and it decreases the smoke formation during hazardous conditions; also, it increases the char formation. The fillers are also capable of reducing the heat transmission rate and prevent dripping. Some of the critical properties of a material can be improved through the proper and right use of fillers.

2 Fillers in Composites

2.1 Role of Fillers in Composites

Additives, fillers, and reinforcements are mostly used to improve or change the properties of a composite. These days, fillers are used in the composites to improve the properties such as wear, friction, corrosion, heat conductivity, degradation of the material, decrease the thermal expansion, electrical properties, reduce the amount of shrinkage, and swelling. Incorporating fillers such as graphite and SiC particles in the glass-epoxy composite systems significantly improve the friction behaviour and exhibits superior wear-resistant properties (Suresha et al. 2006). For tribological applications, short fibres are used as reinforcement in polymer composites. Fillers in the polyetheretherketone (PEEK) composite have been used to reduce the wear rate, but its level of influence lies on the capability of the composite layer to form transfer films (Bahadur and Gong 1992). Lignin fillers are used in natural rubbers to protect it from thermo-oxidative degradation in the air (Košíková et al. 2007). Fillers that possess high thermal conductivity are added to epoxy composites to increase the breakdown power (Li et al. 2011). The composites used in dental applications also have fillers for several purposes, such as colour, strength, and bonding.

2.2 Classification of Fillers

Fillers are classified mainly by their material, size, and shape. Fibres and particles are commonly used fillers in the polymer matrix and metal matrix composites, respectively. They are either incorporated to serve as reinforcements or fillers. Particulate fillers in the composites are classified based on its size are macrofillers, midifillers, minifillers, microfillers, and nanofillers. The size limits of the particulate fillers are prescribed as 10–100 μm, 1–10 μm, 0.1–1 μm, 0.01–0.1 μm, −0.005 to 0.01 μm, respectively. The characteristics of the fillers are different from one another in the various aspects such as surface properties of the filler, shape, size, distribution of the particles, and impurities. From the literature survey, it was observed that finer particle sized fillers produced composites of better mechanical properties, whereas the coarser particles showed a declining trend of the properties in the composite. Impurities may create serious problems such as unwanted chemical reactions with the matrix materials during elevated temperatures (Nassar et al. 2017; Muraliraja et al. 2018). The toughness of the thermoplastics can be improved by adding rubbery kind of fillers. Layers of fibrous filler materials are implanted in the plastic or polymer material to increase the strength (Raja and Retnam 2019). Fibres are mainly classified into two categories, namely natural and manmade. Natural fibres are extracted from animals, plants, and naturally available minerals. The artificially synt6hesized organic and inorganic are the manmade fibres used in composites (Nassar et al. 2017) Fig. 1.

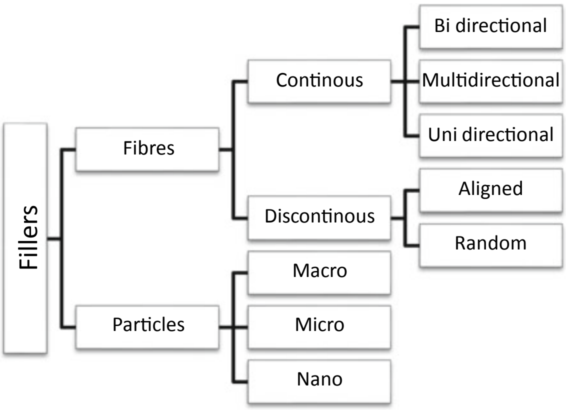

Fig. 1 Classification of fillers (Nassar et al. 2017)

2.3 Fillers in Different Processing Methods *Wypych (2016)*

The preparations composites based on different fillers are mentioned in the table given below. The goal of this section is to identify the different types of processes or methods used to incorporate fillers in newly formulated composite materials Table 1.

Table 1 Different processing methods used to incorporate fillers in composites

No	Fillers	Process/methods	References
1	Glass fibre	Blow moulding	Palutkiewicz et al. (2019)
2	Carbon fibre sheet	Compression moulding	Wulfsberg et al. (2014)
3	Particulate filler (nano-YSZ)	Dip coating	Bakhsheshi-Rad et al. (2016)
4	Particulate fillers	Dispersion	Li et al. (2012)
5	Biomass residues as fibre fillers	Extrusion	Bajwa et al. (2011)
6	Wood fibre	Foaming	Matuana et al. (1998)
7	Cellulose fibre	Injection moulding	Graupner et al. (2016)
8	Glass fibre (treated)	Pultrusion	Chen et al. (2017)
9	Silane treated glass fibre	Reaction injection moulding	Yoo et al. (2017)
10	Natural fibre (Jute)	Resin transfer moulding	Ashworth et al. (2016)
11	Natural fibres (agave, coir, and pine)	Rotational molding	Cisneros-López et al. (2017)
12	Chopped carbon fibre	Sheet molding	Tang et al. (2019)
13	Multi-walled carbon nanotubes	Spinning	Lai et al. (2015)
14	Hemp fibres	Thermoforming	Ciupan et al. (2017)

2.4 Factors Affecting the Functions of Fillers

Fillers generally reduce the weight of the composite without compromising its strength. They also reduce the cost of the matrix material by replacing the specific content of the matrix. The main factors influencing the properties of the composites are the shape and the size of the fillers. Fillers are generally in the shapes of spheres and flakes. Typically, fillers that are sized in between of micrometers and nanometers are preferred, as they have more surface to the contact area. Based on the size and the shape, different fillers tend to exhibit different effects on the properties and behaviour of the composites. In particular, the fillers are selected as per the application for which the composites are developed.

2.5 Effect of Fillers on Mechanical Properties of the Composites

The addition of fillers to a base matrix tends to enhance the mechanical properties of the composite. Factors like, "dispersion, distribution and adhesion of fillers along with the interface between filler and matrix" have an influential role over the mechanical behaviour of the composites. Polymer-based composites showed enhancement of the mechanical properties like ultimate tensile strength, impact strength and hardness. Both natural, as well as artificially prepared fillers, have demonstrated a significant contribution in improving the properties of the base material. Composites with filler provide additional energy-absorbing damage modes, effective stress transfer between the particles and the matrix with combined advantages of the constituent phases.

In an experimental study by Naidu et al. (2019), the influence of graphitic carbon nitride (g-C(g-C_3N_4) nanofillers on the mechanical properties of epoxy-glass fibre composites was investigated. Various proportions of the filler (1, 1.5, 2, 2.5, and 3%) were added to the epoxy matrix and mechanical characterization of the filled composites revelated that for 2 wt% of g-C_3N_4 filling, the tensile and flexural strength was enhanced by 11% and 13% respectively (Naidu et al. 2019). Pandian and Jailani (2019) performed a comparative study on the effect of silica fumes on the mechanical attributes of jute-linen composites. The composites were prepared with and without incorporating silica fumes (1 wt%). The results inferred that the tensile strength of the filler added composite had increased by 7% and the flexural property too had improved by 5.2%, which was more than that of the composite without filler (Pandian and Jailani 2019). The researchers also explored the effect of industrial waste silica fumes as fillers on polymer composites. The dynamic mechanical properties of industrial waste silica fumes (1, 2 and 3 wt%) incorporated natural fibre fabrics reinforced polymer composites were evaluated. The incorporation of a small quantity of silica fumes had significantly increased the composite's dynamic mechanical properties ("storage modulus, loss modulus, and glass transition temperature"). However, for a higher weight fraction (above 2%) of silica fume addition, no further improvement in the mechanical properties of the composite was observed, which could be ascribed to the agglomeration of silica fumes (Arvinda Pandian and Siddhi Jailani 2019).

Nano silicon dioxide of different concentrations (0, 5 and 10%) were incorporated into jute-epoxy composites, and their influence on the fatigue, flexural and tensile attributes were studied. The Young's modulus, peak load, ultimate tensile and flexural attributes were found to be the highest in case of 5% filler loaded composite. Similarly, in the fatigue test, the composite with 5% nano silicon dioxide withstood the highest number of cycles and exhibited good results compared to the other loaded composites (Ashik et al. 2017). The potential of molybdenum disulphide or shungite, organo-modified montmorillonite, graphite nanoplates as fillers in Ultra-high-molecular-weight polyethylene (UHMWPE) was investigated. The deformation and strength characteristics of UHMWPE composites for filler content of up to 0.06 vol% were studied. This study showed that filler addition had a significant effect on the deformation and strength characteristics. Under the filler contents of up to 0.06 vol%, the highest increase in the elastic modulus of composites was provided by the lamellar nanofillers, namely graphite nanoplates and montmorillonite (Grinev et al. 2018). Mohanty et al. (2014) studied the effect of alumina nanoparticles, glass fibre and carbon fibre on the tensile properties of the incorporated epoxy composites. Four different types of composites were prepared with different concentrations of the additives mentioned above and their properties were evaluated. Alumina particles, short glass and carbon fibres were added separately in the range of 1–5 wt% in the first three types of composites, while the fourth type was synthesized by combining both alumina particles (2 wt%) and fibres. Of all the synthesized composites, the combination of alumina particles and fibres in the epoxy composites exhibited excellent tensile strength and modulus.

An interesting study by Swain and Biswas (2017) on the influence of moisture on different mechanical attributes of jute/epoxy with Al_2O_3 fillers at wet and dry conditions was conducted. The maximum impact and flexural attributes were 1.902 J and 72.94 MPa, respectively. The observations from the experimental results indicated that the mechanical properties began to decrease on absorbing water (Kane et al. 2016). A similar study by Sideridis et al. (2017) to examine the effect of water absorption on the flexural attributes of low-content iron particle–epoxy composites was performed. The flexural strength and strain witnessed a decrease with an increase in the filler content in the presence of water (Sideridis et al. 2017).

Aveen et al. (2019) developed the glass epoxy composite with aluminium powder, mother of pearl and fly ash powder as fillers. The study was conducted to determine the mechanical properties of fabricated composites by conducting flexural and tensile tests. In the total volume, the filler material was varied in percentage composition by 3, 6 and 9%. From the tests, it was found that the composite with aluminium filler material exhibited better tensile property while the fly ash filler composite exhibited better flexural property (Aveen et al. 2019). Sudheer et al. (2014) investigated the mechanical characteristics of potassium titanate whisker (PTW) incorporated epoxy composites. PTW inclusions showed a positive effect on hardness, density and stiffness properties of the composites (Sudheer et al. 2014). Investigation on the effects of polymer–filler and filler–filler interactions on mechanical as well as dynamic rheological properties of High-density polyethylene (HDPE)–wood composites were conducted. The results indicated that enhanced filler–filler interaction increased the complex viscosity and storage modulus of composites and decreased the mechanical properties. Enhanced polymer–filler interaction increased the complex viscosity and storage modulus and also improved the mechanical properties (Yang et al. 2010).

2.6 Environmental Benefits and Health Hazards

In modern days, there are hundreds of sources of industrial waste polluting the environment, consequently having damaging effects on the earth and its inhabitants. Also, owing to "ecological necessities and strict regulations", incorporating natural fibres in the place of synthetic fibres has become inevitable "for the manufacturers to accomplish new composite materials originated from renewable sources" (Arvinda Pandian et al. 2017). Hence the "whole gamut of engineering sector" has started shifting towards natural fibre-oriented materials from synthetic materials (Arvinda Pandian and Siddhi Jailani 2018).

Several natural fillers such as coconut coir, rice husk and wheat husk fillers were incorporated as fillers, and the mechanical attributes of filled glass/epoxy composites were characterized. Amongst the different composites, the composite with coconut coir had exhibited excellent mechanical attributes (Dhawan et al. 2013). In another study, ground walnut shells, organic waste fillers were used to modify the properties of epoxy composites. Composites with 20, 30, 40, and 50 wt% of walnut shell were prepared and tested. The composites containing the natural filler exhibited a considerable degree of improvement in hardness and stiffness, yet witnessed a decrease in the impact, and tensile properties (Salasinska et al. 2018). Bamboo, E-glass, and coconut shells were also used in polyester composites, and a significant improvement in the fatigue life and tensile strength of the composite was observed with the addition of coconut shell powder as a filler (Raja and Retnam 2019).

3 Effect of Fillers on Tribological Properties of Composites

3.1 Tribology

The understanding of the design, friction, wear, and lubrication of interacting surfaces in relative motion are described in the scope of tribological science. Almost every industrial part or component is subjected to different types of direct and indirect tribological loadings, namely adhesive and abrasive, during their service. Therefore, the tribological behaviour of materials becomes an essential criterion to be considered in the design of any mechanical part. Wear is the dominant reason behind material wastage and loss of mechanical, thermal, or electrical performance of the related application. Friction is the primary reason behind wear and energy loss because it affects the period of complete utilisation of the material. The two influential tribological properties of a mechanical system, i.e., friction and wear, are neither mutually exclusive nor completely inclusive. It is hardly discerned that tribology affects the life of people to a much greater extent than what is actually realized. Any reduction in wear or improvement friction control in an application can result in a considerable amount of savings. Several measures have been developed to reduce the coefficient of friction as much as possible by either eliminating or at least controlling the factors such as surface finish, temperature, operational load, relative speed, nature of relative motion between the surfaces, and lubrication characteristics. Friction in a mechanical system can generally be reduced by the use of sacrificial bearing surfaces made of wear-resistant or low shear materials, modifying the surface of the moving or stationary component by coating the surface, replace sliding friction by rolling friction, or improve the lubricity between the sliding surfaces by changing the viscosity, use of improved additives or suitable lubricants (Kumar and Srivastava 2016; Vinayagamoorthy 2018).

The huge amount of direct and indirect costs incurred by tribological deficiencies and failures dramatically affects the economy of a country. The expenditure mostly incurs due to the simultaneous loss of material and energy on every mechanical component in operation. Among the two characteristic properties in the tribological behaviour of a system, wear is a more critical factor compared to friction as excessive wear of a component in a machine may result in catastrophic failures and operational breakdowns, which can adversely impact productivity and hence, cost (Holmberg and Erdemir 2017; Tzanakis et al. 2012). There is a need to emphasize the importance of sustainable tribology in our era to showing that tribology is frequently the primary cause and, simultaneously, the solution for most of the mechanical maintenance problems. The development and deployment of modern tribological solutions for engineering systems can provide measurable beneficial, financial, and environmental outcomes to society and the industry.

Studies have revealed that 90% of failures in mechanical parts and components occur as a result of tribological loadings (Jost 2006). Therefore, a substantial sum of expenditure involved in repair costs can be lowered if a proper understanding of tribological principles and its applications are made. In tribology, the friction and wear of a material depend on some of the essential parameters such as surface roughness, relative motion, velocity, type of material, load, temperature, stick-slip, relative humidity, lubrication and vibration (Blanchet 2012). Nonetheless, effective modern methodologies for good tribological design can be very costly, yet efficient. An understanding of the nature of wear and friction in the system is essential to formulate the right mechanism to control their tribological behaviour. The criteria by which the wear life or frictional behaviour of a product is regulated may vary strongly across different segments of application. In some applications, the function is far more critical than manufacturing costs. One effective way of reducing wear and controlling friction is by the use of lubricants, which comprises of dry and wet lubricants. A wide range of liquids and soft solids have been studied to exhibit effective lubricant properties and have also proven to be cost-effective solutions. The apparent advantage of solid lubrication over oil lubricants is their superior cleanliness.

Composite materials emerged as a potential alternative for several classes of other materials in engineering technology, wherein two or more natural or artificial elements are combined by a series of chemical or physical processes to obtain a final product with added strength, efficiency or durability. The technical advantage of employing composites for manufacturing mechanical components or devices is that they can be prepared with the desired engineering properties by a careful selection of matrix and a compatible reinforcement. In the scope of tribology, composite materials are expected to provide a viable support structure to support the load and frictional heat. The inherent nature of the composite system exhibits mechanical stability and also fashions the attractions of means of dissipating frictional work. The continuous progress and innovations in modern technology are laying out newer rheological demands on lubricants, which are not often met by conventional lubricants (Dorri Moghadam et al. 2015; Omrani et al. 2016). On the other hand, self-lubricating composites have gained importance, as they can be tailored by using a base matrix, reinforcements, solid lubricants, or additives that may be required for the typical application. Advancements in the field of tribo-engineering have offered attractive solutions as self-lubricating composites, which are mostly compositions of metallic, ceramic or polymeric matrices added with functional fillers providing the desired tribological functionality (Erdemir 2005).

In general, the role of a filler in a composite material is versatile, and the nature of role purely is influenced by the composite matrix and the filler material itself. Some of the unique advantages of using fillers are to strengthen the matrix (load carrying capacity), improve the sub-surface crack arresting ability, enhancement of the thermal or electrical conductivity of the material, and providing a lubricating effect at the interface by decreasing the shear stress. Although fillers demonstrate superior behaviour in terms of the mechanical and tribological properties, the use of a specific type of fillers (e.g., particulate type) may somewhat affect the other properties; therefore, proper optimization of the mechanical and tribological properties of the composite has to be carried to avoid this disadvantageous compromise of the behaviour. Occasionally, in some cases, fillers are also used as cheap additives, as they reduce the material costs in polymer applications besides enhancing the tribological behaviour of the material (Bobby and Samad 2017; Saba et al. 2014; Senbet 2008).

3.2 Common Testing Methods for Tribology Tests

There is an endless number of methods that can be employed to characterize the tribological behaviour of a particular material. In order to make meaningful interpretations of the tribological outcomes, the most suitable test for the particular purpose needs to be selected. The nature of results obtained from a tribo test does not merely relate to the behavioural characteristics and properties of the composite but represents the mechanical system as well. Proper testing of composites using standardised methods can reveal information on the likely product life of the system. In the tribological characterisation of composites, the experimental attributes such as the matrix, type and structure of reinforcement, filler-matrix interface, and internal lubricants usually influence the measurement of wear and friction. The testing of wear and friction differ for different tribological systems, and therefore, composites need to be tested under standardised procedures before application of the actual results. Standard testing methods and devices have been elucidated in several references (Nirmal et al. 2011). Most of the methods have been exclusively published by the Society of Automotive Engineers (SAE) and American Society of Standards and Materials (ASTM) as standardised procedures for testing and evaluating material properties. Following the standard guidelines will enable the use of identical devices to obtain very nearly identical results facilitating comparability and correlatability.

Some of the commonly used tribological testing machines are dry sand rubber wheel (based on ASTM G65), pin on drum (based on ASTM A514), pin on disc (ASTM G99), Linear tribo-machine, Block on ring (ASTM G77, G137-95), Block on disc (ASTM G99), and 4-ball test (ASTM D2266) (Bhushan 2000; Singh et al. 2016). One of the most widely used methods for evaluating the tribological characteristics is the Pin-on-disc wear and friction testing that follows the methods as per the ASTM G99 standard. The specimen (pin) is held vertically or horizontally under loading against a rotating counterface (disc). The counterface exhibits a constant area of contact throughout the test as a result of which the method is more suitable for application involving sliding wear. The distinct operating parameters include applied load, sliding distance, wet or dry sliding condition, sliding velocity while the specimen contact area is maintained constant with res0pect to sliding time. The Pin on drum wear test rig is built based on ASTM A514 standard. The specimen travels horizontally (linearly) against a rotating drum that rotates at the desired speed using a drive chain. While the wear test can be conducted for both abrasive or adhesive conditions, wear simulates the applications of goods on rotating rollers or conveyor belts.

The Block on ring wear testing, which is based on ASTM G77 standard has a working principle very similar to that of Pin on disc testing method. The specimen is held against a rotating wheel or ring at 90° to the wheel or ring axis of rotation. The characteristic operating parameters include applied load, sliding velocity, temperature, sliding distance, wet or dry sliding condition but the specimen contact area is varied with respect to sliding time. This testing method simulates applications such as pulleys and camshafts. The Dry sand rubber wheel wear testing is based on the ASTM G65 standard. The specimen is held against a rubber wheel, while sand is introduced to the rubber interface to simulate an abrasive testing condition. The testing method can also be used for adhesive testing in the absence of sand. This kind of wear test typically simulates the applications such as tyres, bushes, bearings and rollers.

Although the standard methods of testing can produce adequate results to predict the failure mode or lifetime of a tribological system, the standard tests may not simulate the experience of the composite in the practical system. Although simple bench model tests can easily, quickly, and cheaply evaluate an extensive range of materials under well-controlled and simulated test conditions, they do carry some limitations. Generally, the practical representation and realism of the data and features of surface damage, and the prospects of making reliable inferences about the performance or usability from the tests to an application is somewhat decreased as we move from a field test to a simpler model test. The primary criterion for confirming whether a chosen model test is applicable or not is based on to what extent the wear mechanisms appearing in the application during the actual service conditions are being reproduced. Moreover, it is critical to comprehend the actual mechanism of wear for accurately characterizing the tribological behaviour of the tested materials.

3.3 Wear and Frictional Behaviour of Filled Composites

The addition of filler materials to composites is mainly targeted towards improving the thermal, mechanical and tribological properties. In the scope of improving the tribological properties, the ability of the fillers lies mainly in influencing the development of transfer film and counterface adhesion. Composites are classified based on the type of the primary matrix, or reinforcement or the nature of the interface while the fillers used in composites are generally categorised into metallic fillers, ceramic fillers, polymeric fillers, and mineral fillers. Most of the time, micro- and nano-sized inorganic fillers are used for modifying the tribological behaviour of composites. The systemic effect of a filler on the tribological behaviour of a composite only becomes noticeable when the filler is at the surface. The ability of fillers to reduce friction depends on the degree of fineness of the filler particles, as they need to be fine enough to effectively increase the deflection temperature of the matrix near the surface because frictional heating and matrix softening are two factors responsible for high friction. In terms of the contribution of fillers towards wear resistance, the proportion of the filler in the matrix will determine the property. In general, for composites, the best abrasion resistance and low coefficient of friction will be obtained with those fillers providing the highest packing fraction (P_f) or at concentrations approaching P_f. With the development of mature technology, industries have begun using fillers in diverse applications as multifunctional additives rather than just cost-reducing expedients. Powdered Teflon, graphite, micro-size CuO, silica, $CaCO_3$, PTFE and molybdenum sulphide (MoS2) and so on, are some of the prominent fillers used in composites for improving the tribological behaviour. A considerable amount of work on the evaluation of the tribological behaviour of composites has been carried out previously. The scope of this review is limited to briefly examining the various kinds of fillers used in composites for improving their tribological behaviour.

3.3.1 Ceramic Fillers

Ceramics, such as metallic oxides, carbides, borides, nitrides, silicides, etc., are known to retain their mechanical properties at higher temperatures. This inherent property of ceramics has been exploited in ceramic fillers which are primarily compounds of the aforementioned inorganic groups. The properties of the filler and their functionality comes from the inherent nature of the base ceramic itself. The inherent advantages of ceramics in terms of their high hardness, strength, melting point, and abrasion resistance make them a much-preferred variety of fillers in composites operating in high-temperature applications. While the most usual type of bonding observed in ceramics is a combination of ionic and covalent, Van-der-Waals forces or a metallic component may also be present depending on the type of ceramic. Some of the commonly used ceramic fillers in composites are AlN, Al_2O_3, SiC, Si_3N_4, $Sr_2Ce_2Ti_5O_{16}$, zirconium silicate ($ZrSiO_4$), wollastonite ($CaSiO_3$), silicon dioxide (SiO_2), beryllium oxide (BeO), CeO_2, boron nitride (BN), and ZnO.

Addition of ceramic particulate, whisker or microfibre fillers to composites or nanocomposites have revealed a dramatic improvement in the wear resistance of composites as much as three times the magnitude of the unfilled composites. Nanometer-sized particles have been extensively used as fillers for obtaining superior tribological characteristics. Hong-Bin Qiao et al. (2007) investigated the wear and friction properties of the Al_2O_3 particles (5%) filled polyetheretherketone (PEEK) and polytetrafluoroethylene (10%) PEEK against medium carbon steel (AISI 1045 steel) ring under dry sliding conditions. The PEEK composites exhibited a notable decrease in the wear rate with the addition of the nanometer and micron-sized Al_2O_3 (in the absence of PTFE) but had less effect on the friction coefficient. In contrast, the wear rate, as well as friction coefficient, had significantly lowered for the pure PEEK composite filled with 10 mass % of PTFE. But, when 10 mass % PTFE was filled into the Al_2O_3/PEEK composites, the behaviour contradicted the expectation wherein the coefficient of friction decreased while the wear rate increased (Qiao et al. 2007).

A comparative evaluation of using SiC and Al_2O_3 on the wear behaviour of jute/epoxy composite was conducted by varying the weight percent of fillers with respect to the resin. In the absence of fillers, the jute–epoxy composites were easily subjected to wear under higher normal loads and sliding velocities whereas the addition of ceramic fillers displayed considerably lowered the wear rate of jute epoxy composites. The lowest coefficient of friction was observed in 15 wt% filled jute–epoxy composites which complemented its higher wear resistance. Furthermore, Al_2O_3 filled jute epoxy composites exhibited a lower coefficient of friction and wear loss compared to SiC filled composites for all compositions. The wear mechanism of the in-filled was characterized by microcracking, pit and debris formation in contrast to the unfilled composite, which was dominated by fibre breakage and plastic deformation (Sabeel Ahmed et al. 2012).

In another study, the effect of different loads and abrasion distance was investigated for Polyamide 66/Polypropylene (PA66/PP) blend, nano clay filled PA66/PP and short carbon fiber reinforced PA66/PP nanocomposites based on the three-body abrasive wear mechanism. The addition of nano clay/short carbon fiber and PA66/PP had exhibited a sound effect on the wear rate for different abrading distance and loads. On comparison of the effects of their addition, nano clay/short carbon fibre composites demonstrated a lower wear rate than short carbon fiber filled PA66/PP composites (Ravi Kumar et al. 2009). The possibility of using Rice bran ceramics (RBC) as fillers were explored, as the hard-porous carbon material, made from rice bran provide low friction and high wear resistance characteristics (Shibata et al. 2014). The tribological properties of the thermoplastic resin-based RBC composites were experimentally determined for polyamide 66 (PA66), polyamide 11, polyoxymethylene, polybutylene terephthalate, and polypropylene matrix resins. Higher wear resistance and lower friction levels were observed for thermoplastic-based RBC compared to their pure resins. Also, the RBC fillers had the upper hand over Glass fibre (GF) fillers, as a substantial improvement in the strength of the composites was noticed whereas the friction coefficient and wear did not witness any effect. The RBC particles, in addition to improving the fracture toughness of the composite, also contributed to a decrease in the friction coefficient, resulting in mild wear. Thus, the RBC based composited indicated their great potential as an anti-wear hard particulate filler in the industry (Shibata et al. 2014, 2012). So far, four different forms of glasses, namely plain GFs, hollow glass beads (GBs), solid GBs, and glass flakes have been incorporated in glass-filled thermoplastic composites. Of all the forms, the lowest wear rate was observed for GF-filled composites and solid GB-filled composites, whereas the hollow GB-filled composite showed the highest wear (Klaas et al. 2005).

Metallic compounds such as ZnO, TiO_2, CuO have been exploited as filler owing to their desirable tribological properties. Addition of ceramic fillers in polymers had improved the performance during abrasive wear. The effectiveness of the filler to function as a wear-resisting material is determined by factors such as the content of filler added, the interaction of filler matrix and the type of the matrix used (Suresha et al. 2010). Nanosized CuO filled and short carbon (CF) and aramid (Kevlar) fibres-reinforced polyphenylene sulfide (PPS) composites prepared by compression moulding were investigated for their tribological behaviour using a pin-on-disc apparatus. In case of the filled composite, a steady-state wear rate was observed while it was reduced to half of that in the case of filled CF reinforcement composite. However, the addition of CuO filler had not contributed to improvement in the wear resistance of the fabric-reinforced composites owing to the poorly developed transfer film in the presence of fibres and as a result, the composites became fragile (Bahadur and Polineni 1996). Li et al. (2002) reported the use of nano ZnO as filling material to PTFE to reduce the wear rate of the polymer. The wear rate obtained for the composite containing 15 vol% nano ZnO was determined as the optimum level of nano ZnO to be incorporated, however a higher coefficient of friction was observed for the nanocomposite than that of the unfilled PTFE (Li et al. 2001). Short carbon fibre (SCF), graphite flakes, and microparticles of TiO_2 and ZnS were used as fillers in thermoplastic composites, such as polyetheretherketone (PEEK) and polyetherimide (PEI). The tribological characteristics of the two types of high-temperature-resistant filled thermoplastic composites were evaluated under dry sliding conditions against steel counterparts. The addition of fillers like SCF and graphite flakes demonstrated considerable improvement in the wear resistance and the load-carrying capacity of the base polymers.

Nonetheless, with the addition of microparticles of TiO_2 and ZnS, the coefficient of friction and wear rate of the composites had further reduced especially at high temperatures (Chang et al. 2007). TiO_2 and ZrO_2 were incorporated as ceramic fillers in the reinforcement of bamboo-glass hybrid polymer composites. These compounds have identical morphology and were incorporated as fillers with particular emphasis on exploring their effect on the tribological behaviour of reinforced thermosets (including micro-particle). Among the two inorganic fillers, the comparison of the wear characteristics of bamboo-glass-epoxy hybrid composites revealed that ZrO_2 fillers gave better strength and wear-resistant properties when compared to the TiO_2 filled composite (Latha and Rao 2018).

The potential of Silicon carbide (SiC) or boron carbide (B_4C) ceramic fillers was investigated by introducing them into three-dimensional needled carbon fibre to prepare ceramic modified carbon/carbon (C/C) composites. On observing their morphology, the pore size distribution was found to be uniform on the addition of the ceramic fillers in the C/C composites. Further, the ceramic modification considerably lowered the COF fade in seawater conditions for C/C composites. C/C-SiC_f displayed superiority over C/SiC for its excellent stable friction behaviour without any fade in seawater conditions. Also, a cumulative effect of lubrication from water film and SiO_2 film seemed to increase the COF fade for C/SiC compared to that of C/B_4C (Cai et al. 2013). A combination of nanostructured fillers based on BN and SiO_2 micro powders was used to reinforce $AK_{12}M_2MgN$ alloy. The structure of the fillers led to a uniform dispersion of the structural components of the alloy and resulted in a considerable increase in its wear resistance and a decrease in the coefficient of friction (Komarov et al. 2013). A synergistic effect was observed for epoxy based composited filled with varying concentrations of short carbon fibre (SCF) and solid lubricants, i.e. PTFE and graphite. These composited were also supplemented with the addition of varying amounts of sub-micron sized TiO_2 (300 nm). In the investigation by Zhang et al. (2004), it was inferred that the synergistic effect was subjected to tribological characteristics of the composites in comparison with monolithic systems (Zhang et al. 2004).

Nanoparticles of SiO_2, SiC, Si_3N_4 and ZrO_2 used as fillers have demonstrated to be very effective in lowering the coefficient of friction and specific wear rate of PEEK composites when sliding against the steel counter surfaces (Wang et al. 2000, 1996). Apart from these, nano laminated Ti_3SiC_2, granite dust and powder, porcelain waste, nanodiamond and Ti_3AlC_2TiC particles and fly ash have also been as fillers in metal matrix and polymer matrix composites.

3.3.2 Carbon and Organic Fillers

Carbon black and carbon fibres have been used as additives in a wide range of thermoplastic, and thermoset resins. The layered structure of graphite, which is characterized by a weak Van-Der Waals force between the layers possess a high electrical conductivity. The nature of the graphite structure also helps in reducing friction and wear by forming a lubricant film between the mating surfaces, which is utilized in its application as a filler. Carbon black, on being incorporated as fillers in composites showed attractive tribological characteristics. The addition of carbon black fillers (at 1–5% filling) along with PTFE composites exhibited a tremendous increase in the wear resistance of up to 700 times. The excellent dispersion of fillers in the PTFE matrix enhanced the interaction effect of carbon black in the dry conditions of testing against smooth metal surfaces.

Moreover, the wear rate of a PTFE composite mainly depends on the properties of an ultra-disperse filler, such as its specific surface area. With an increase in the specific surface area of the added filler, the wear rate witnesses a proportional increase due to the increase in the area of interphase interaction of the components per unit mass of the filler and high activity at its surface centres (Aderikha and Shapovalov 2010). Carbon black derived from wood apple shell, obtained by pyrolysis at 400 °C was also used as a filler in an epoxy resin. Carbon black particulates composite showed minimum wear on comparison with raw particulate composite (Ojha et al. 2014). Hybrid fillers containing multi-walled CNTs and carbon black in natural rubber were also investigated for dry friction and wear behaviour. It was inferred that a considerable increase in the wear resistance and unexpected reduction in the friction coefficient resulted from the increase in the applied load for every fixed sliding speed (Ojha et al. 2014). The investigations on the mechanism of wear in the low-filled PTFE-CB composites revealed a delamination mode of wear wherein the changes in the wear resistance was relatable with the structural transformations of the composite.

The exceptional properties of the single and multi-walled carbon nanotubes (CNTs) favour them as a recognized filler for epoxy-based composites and coatings. Various boating and automotive applications utilised a wide range of reinforced polymer–matrix composites because of the capacity of these composites to withstand longer durations of sliding contact-based wear conditions. The tribological response of an epoxy matrix of polyamide filled with graphite and/or carbon nanotubes was highlighted by the enhanced wear resistance which was owing to the carbon fillers dispersed in the epoxy matrix. Of all the carbon-based fillers, the most desirable result was obtained for the composited filled with TCNTs. The TCNTs were well dispersed in the epoxy matrix due to the presence of the NH_2 groups. Despite the enhanced wear resistance exhibited by EpCNTs and Ep-Graphite, Ep-Hybrid demonstrated relatively much inferior tribological properties. Subsequently, even the combination of graphite and CNT fillers did not witness any synergistic effect (Sakka et al. 2017). Sam-Daliri et al. (2019) found a novel method to improve the tribological behaviour of an unsaturated polyester matrix wherein a relatively small amount (0.2 wt%) of well-dispersed MWCNTs within the wood flour polyester composite was used a filler in the polyester matrix. The enhanced tribological behaviour was attributed to the transfer of a soft layer of wood flour on the worn-out area during the wear process, which acted as a self-lubricating material (Nabinejad et al. 2019). Composites were developed with a novel combination of multiwalled carbon nanotubes (MWCNTs) and short carbon fibres (SCF) as fillers for an automotive brake system. All the combinations of the composite with the carbon fiber had shown superior properties of lower wear rate and coefficient of friction (Gbadeyan et al. 2018).

Researchers have extensively used carbon fibres for their desirable mechanical properties. Surface modified carbon nanofibres (CNF) filled PTFE composites were subjected to tribological characterization under dry sliding conditions. The CNF was treated with HNO_3 followed by coupling agent treatment before incorporation to the PTFE matrix. The optimum content of CNF in the PTFE resulted in reduction of the wear rate, almost 30% lower than that of untreated CNF filled PTFE for an applied load of 200 N (Shi et al. 2008). Suresha et al. (2010) compared the wear characteristics of carbon–epoxy and graphite filled carbon–epoxy composites at different loads in abrasive condition using different grades of SiC abrasive paper (150 and 320 grit size). Improved abrasion resistance was witnessed for graphite filled carbon-epoxy composites investigated for different loads and distances of abrasion. The improvement in the properties corresponds to the filler to filler interaction and uniformity in the distribution of the added fillers in the carbon-epoxy matrix (Suresha et al. 2010). The abrasion and attrition wear of experimental composites related to dental application with samples of different resin viscosities was investigated. Although raising the resin viscosity lowered the wear resistance, it had minimal influence on composites holding nonbonded nanofiller. However, an increase in resin viscosity increased abrasion and attrition in composites containing silanated nanofiller, with equivocal effects in composites containing unsilanated nanofiller (Musanje et al. 2006).

Any discussion on carbon-based fillers is incomplete without highlighting the use of graphene/graphene oxide fillers. The tribological properties of metal matrix composites containing graphene were reported in several papers earlier. The functional advantage of using graphene fillers is that multilayer graphene and reduced graphene oxide can not only enhance the strength drastically but also reduce the friction coefficient and wear rate of composites (Li et al. 2017a, b; Llorente et al. 2019). Carbon fibre composites have gained much importance in marine, sports, construction industries besides aerospace applications.

3.3.3 Metallic Fillers

Mild steel substrates coated with metallic fillers such as micro-nickel, aluminium, silver and zinc powders have demonstrated remarkable tribological properties and surface energy characteristics. The attractive results exhibited by the metallic filler-based composites has encouraged several other metallic powders to be explored as a viable possibility. Three types of metallic fillers, namely steel fibers, brass fibers and copper powder, were used in the preparation of non-asbestos organic (NAO) composites. The tribological properties of the composites were evaluated for different loads and speeds. Although the addition of metallic fillers led to enhancement in friction performance of the composites, for every subsequent increase in the amount of the metallic filler, the wear resistance exhibited an increase. The higher wear resistance due to increasing metallic contents resulted in an increase in the thermal conductivity

(TC). Especially, copper filler (10%) based composites showed significant performance from both wear and friction properties followed brass, while iron powder based composite showed moderate behaviour (Kumar and Bijwe 2010).

Researchers attempted to investigate the influence of various metallic fillers (Cu, steel, or Al) on the friction and wear performance of brake pad composites. The experiments were conducted using a small-scale friction tester against two counter disks (grey cast iron and aluminium metal matrix composite (Al-MMC)) at ambient and elevated temperature ranges. The ambient temperature tests against grey cast iron disc revealed that the composites with Cu fibers showed a noticeable negative μ–ν (friction coefficient versus sliding velocity) inferring that stick-slip may occur at low speeds. At higher temperatures, wear tests showed that the Cu-fiber composites exhibited better wear resistance than the other composites. The tests with steel fibers revealed that they were not well-matched with Al-MMC disks due to substantial material loss and irregular friction behaviour during sliding at elevated temperatures (Jang et al. 2004). The use of copper and its alloys to increase the thermal diffusivity at the friction interface has become a common practice. The natural tendency of copper to endure the high temperatures attained at the interface for higher levels of the friction coefficient is taken as an advantage. During high temperatures, the copper oxide formed at the interfacial layer dissipates the frictional heat effectively. Hence, copper and its alloys are added as fillers to regulate the friction level while avoiding the fierceness against the counterpart. Likewise, aluminium fibers are also added to the composites in applications where aluminium metal matrix composite (Al-MMC) brake rotors are used (Wilson and Alpas 1996; Urquhart 1991).

Steel fibres have also been presented as a potential replacement for other fillers that lack the mechanical properties as that of steel. Qu et al. (2004) examined the wear and frictional characteristics of composites filled with continuous steel fibres. The fibre orientations are hosted concerning the sliding direction, namely parallel (P) normal (N) and antiparallel (AP) along the fibre direction. The wear rates were found to increase with changing sliding directions and exhibited a dependency on the stability of the film. The friction coefficient (range 0.49 to 0.54) also remained dependent on the fibre alignment directions. At higher temperatures, the iron-rich transfer film formed on the specimen promotes adhesive interaction, and steady-state friction is observed (Qu et al. 2004). The wear and friction characteristics of the friction materials reinforced by brass fibres against grey cast iron demonstrated superior wear resistance of the composite. The interacting elements showed fatigue wear mechanism which was confirmed based on the following inferences: (i) formation of a copper transfer film on the friction surface of the grey cast iron counterpart (ii) the worn surface of the counterpart revealed fatigue cracks. The wear loss and coefficient of friction increased slightly when the mass fraction of brass fibres was over 19% (Xian and Xiaomei 2004). Compression moulding technique to add the carbon fibres and nanofillers like Al and Zn as reinforcements to epoxy hybrid composites proved to be effective in reducing the specific wear rate up to 0.5 wt. % of Al/Zn filler loading (Divya and Suresha 2018).

Apart from the above mentioned, steel wool, nickel, silver, brass fibres, copper powder, sodium are the other types of metallic fillers used in the composites. Besides, mineral silicates (Vasilev et al. 2019), talc (Zhao et al. 2012), feldspar (Cai et al. 2015), nano clays (Bobby and Samad 2017), calcium carbonate are also being used as fillers for many polymer applications. Polyvinyl chloride, polyolefins, phenolics, polyesters, and epoxies are all resins compatible with $CaCO_3$ as fillers, which is not only attributed to economic but performance considerations as well (Palanikumar et al. 2019).

3.3.4 Polymeric Fillers

Polymers have become more of a commodity and relatively less expensive these days. Addition of fillers to reduce cost has become a less significant factor because by judiciously combining the filler with the resin, one can achieve a spectrum of materials with properties intermediate between those of the two ingredients. Polymeric fillers are generally classified into natural and synthetic varieties, wherein natural polymers consist of cellulose fibres, graphite fibres, wood flour, flax, cotton and starch. Synthetic polymers are not available in the natural environment, so these are tailored polymer materials catering to different applications, namely polyamide, polyethene, polytetrafluoroethylene, polyester, aramid, and polyvinyl alcohol fibres. Polymeric fillers usually limit their usage to controlling permeability, instilling softness to the matrix, damping control, and imparting the desired tribological behaviour to the composite system.

Polymer-based fillers such as fluorinated polyether ketone (aryl ether ketone) (FPEK) and polytetrafluoroethylene (PTFE) have also been used to modify the properties of epoxy-based composites. The nature of FPEK tends to render the composite extremely resistant to scratch and as a result, its presence in the corresponding matrix, the composite resists even higher degree of abrasion (Saba et al. 2014). The friction coefficient of the composite is considerably lowered with the addition of PTFE fillers to the composite. PTFE is a commercial filler that could reduce the frictional coefficient, and, due to this fact, sometimes also the wear rate of polymeric composites is reduced. The unique molecular and morphological structure of PTFE renders the polymer its extraordinary tribological properties. A third-body transfer film is formed when the polymer slides against the steel counterparts, which is the mechanism behind the lowering of the frictional coefficient (Zhang et al. 2004; Şahin 2018).

4 Recent Advances, Challenges and Future Trends

Composite materials have become a sustainable alternative to conventional materials used in automotive, aerospace and other industries in the last few years. The speciality of this technology is that the end product material can be engineered with desired specific properties by a careful combination of matrix and additives. Although the composites offer many attractive properties, it is unquestionable that the manufacturing process involved in commercializing these materials present new challenges day by day. On the other hand, the technological capabilities and diversity of applications of different reinforced composites have steadily increased. The broadening spectrum of applications of composites is driven by the need to replace an existing expensive material, usually a metal. While most of the metals are facing pressing demand and acute shortage in metal processing industries, traditional composites have been facing a need for up-gradation to fulfil the demand for superior properties and high performance. In this aspect, nanocomposites are evolving as a promising solution to cater to the needs and requirements of modern industries. The advantages of the newer generation of composites are their ability to improve two or more desired properties simultaneously. The tribological research on the nanocomposites, particularly hybrid nanocomposites and polymer nanocomposites is still at an early stage and has great potential to cater to many domestic and industrial applications.

Traditionally, most fillers were considered as additives, considering their contribution to a composite only focused on reducing their cost. However, the diversity of applications and a broad spectrum of their usage has led to high demand for fillers or reinforcing fillers in composite technology. Although the diversity of fillers has crossed countable limits, the challenge is to select the most suitable and appropriate filler by understanding the performance criteria of the composite being developed. Several commercial polymer composites survived the rigorous market demands, because of the right blending of ingredients to maximize the magnitude of the desired property and simultaneously mitigating the loss of others. Apart from these, filler manufacturers have to necessarily take environmental concerns, such as recycling, sustainability and life cycle impact into consideration. Natural, preferably biological and organic fillers are gaining more dominance over synthetic owing to their environmentally friendly properties. There is no doubt that more emphasis will be imposed to improve the durability and viability of the fillers used in the future. Many reasons such as lack of familiarity, limited availability have so far prevented large scale penetration of natural fillers. However, tremendous growth is still anticipated with large companies entering the market and technological advances being made.

Thus, a stable and healthy advancement in tribological research of nanocomposites is anticipated to take over the current scenario as the trend is moving towards developing novel, green measures, chemical or physical alterations of improving the tribological behaviour of polymer composites (pristine, hybrid and nanocomposites).

5 Conclusion

In this chapter, the different fillers used in the composite have been reviewed broadly for the first time. The most important contribution of the current work is identifying various fillers used in the composite materials and its influence on the tribological behaviour of the composite. It is expected that the review would be beneficial to the researchers seeking guidance on different types of fillers for the preparation of newly formulated composite materials. The roles, classifications, processing methods and the influences of different categories of fillers in the composites are discussed intricately. The challenges faced during the fabrication of composited are highlighted. The recent advancements in the processes of incorporating the fillers in composites and the future possibilities of the research have also been briefly reviewed.

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