Graphene: An Effective Lubricant for Tribological Applications

Pranav Dev Srivyas and M. S. Charoo

Abstract The objective of this review paper is to investigate the basic tribological behavior of graphene, the first existing 2D material and to enhance its performance as a self-lubricating material. The significant and prospective impact of this new class of material was first acknowledged in 2004 by Geim and Konstantin Novoselov who were awarded Noble prize for their discovery and development of graphene in 2010. In previous decades, reducing friction coefficient and wear-related failures in mechanical systems has gained serious attention due to friction's adverse impacts on effective life and durability of the mechanical systems. To reduce the friction and wear mechanism in the moving mechanical systems, the research proceeds in the development of novel materials, coatings, and lubricants (both liquid and solid) which have the potential of reducing friction and wear in materials. Despite intense research and development efforts on graphene for numerous existing as well as future applications, its tribological potential as a lubricant is still relatively uncharted. In this review, we provide relevant research of recent tribological studies on graphene especially, its use as a self-lubricating solid or as an additive for lubricating oils. A comprehensive review is provided with the aim to analyze such properties of graphene.

Keywords Graphene · Lubrication · Tribology · Wear · Friction

1 Introduction

In today's world, there is a need of energy resources to meet the growing energy demand, but these energy resources are depleting at a very fast rate which creates tremendous pressure on the engineers, scientist, and the designers. Comprehensive research is on the way to explore the entire possible alternative technologies to

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A. Prasad et al. (eds.), *Advances in Engineering Design*,
Lecture Notes in Mechanical Engineering,
https://doi.org/10.1007/978-981-13-6469-3_22

conserve energy, economic demands, materials, and environment. Therefore, a better tribological property of the materials plays a vital role; is minimizing the energy depletion. Good tribological properties lead to reduce friction, increase the wear resistance of the material which is to be used in various moving mechanical systems and hence also reduce the emission to the environment [1]. Use of Liquid lubricants at the interface of the tribopair is one of the oldest but most effective methods of reducing friction and wear in the mechanical components and systems [2, 3]. According to a study, the worldwide production of the lubricant as well as a coolant during 2010 amounts to approx 170,046.9 metric tons [4]. But most of these lubricants are inherently toxic and of nonbiodegradable nature which affects the environment leads to demand the growth of eco-friendly self-lubricant materials. To enhance the properties of the lubricants and materials used in the composites, various other additives are added to it which enhances the antifriction and antiwear properties. Liquid lubricants reduce the wear and friction of the mechanical component and systems by providing the sliding contact interfaces from metal-to-metal contact by forming a low shear, high durability boundary film on the mating surfaces [5]. Nowadays solid lubricants have emerged as an integral part of materials science and engineering. Solid lubricants can be used in various forms to achieve the set of objectives. However, the use of solid lubricant as coating/filler has greatly expanded the use of solid lubricant materials. Different solid lubricants are used by their operating conditions. Example, MoS_2 is better to be used under a dry condition or in a vacuum; whereas graphite, boric acid shows better properties when used in the humid conditions. Therefore, solid lubricant properties mostly depend on the environment but they are highly durable and easy to deliver to the contact interface [6–9] (Table 1).

Graphene, a 2D carbon material has attained a considerable amount of popularity and scrutiny from the field of science and engineering [10]. Graphene is one such new solid lubricant which is having unusual physical, thermal, nontoxic, eco-friendly, mechanical, and tribological properties. Graphene an allotrope of carbon is a one-atom-thick planar sheet of sp2 bonded carbon atoms densely packed in a honeycomb crystal lattice. Graphene is strongest, chemically as well as thermally stable, gas-impermeable, and atomically thin. It is considered to be green lubricant as it contains C, O, and H instead of heavy metal elements. As in a modern world where the mechanical moving systems attracted a lot of the attention in various diverse applications; this newly emerged solid lubricant has the potential to reduce the friction, wear and increase the life of the system. Graphene nowadays is widely used both in the lubricants as a lubricant additive and as a filler reinforcement/graphene coating in the composites for lubrication means to decrease the friction and wear in tribological applications. It is proved to be equally well for the dry as well as a humid environment which is not in case of other commonly used solid lubricants. Of all the properties, tribological properties along with its applications are still the least explored. This review helps us to highlight these exceptional properties of graphene to reduce friction and wear in the micro- to nanoscale systems. As it is an ultrathin multilayer material, it can also be applied in micro-electromechanical-system (MEMS) and nano-electromechanical-systems

(NEMS) to reduce friction and wear from these systems. Graphene which is an anatomically smooth 2D material with low surface energy can replace the thin solid film is used to reduce adhesion and friction of various tribo surfaces.

In this review, various graphene synthesis techniques are reviewed along with a detailed discussion on the tribological properties of the graphene as a solid lubricant coating/filler in the composite and as a lubricant additive in the mineral/synthetic/biofluids. Also, the literature review is done on the operating conditions at which graphene shows excellent tribological properties. It is expected that this review will prove to be useful to the researchers working in the field of tribology.

2 Graphene Synthesis

Various synthesis routes are available for the synthesis of graphene. Properties of the graphene synthesized greatly depend on the type and quality of the synthesis root we adopt. Synthesis route adopted to generate the graphene has an effect on the grain size, shape, thickness, density, defect in structure, mechanical, and tribological properties of the graphene generated. The most basic and initially introduced method for the generation of the graphene is scotch tape method which is also called a mechanical exfoliation method. In this method of graphene synthesis, highly ordered pyrolytic graphite (HOPG) is used. Nowadays numerous synthesis process/methods are used for graphene generation. These include dry mechanical or chemical exfoliation; unrolling, and unzipping of carbon nanotubes by using physical, chemical or electrochemical methods; chemical vapor deposition (CVD)/epitaxial growth; arc discharge method; reduction of graphene oxide (GO), and many other organic/synthetic methods [11–20]. CVD is the best synthesis technique to get the best quality of graphene on the catalytic surfaces and it is done in the presence of hydrocarbon gases. Since there are various synthesis routes for the generation of graphene, some of them may be close to perfect but might be costly and some are not so perfect but cheap. It depends a lot that which grade of graphene is required and which synthesis route is adopted.

2.1 Mechanical Exfoliation

Mechanical exfoliation is the primary and the basic synthesis route used for the generation of graphene. It was first developed by Geim and Novoselov in the year 2004 for which they receive the noble prize in the year 2010. It includes isolating monolayer's of graphite. The basic mechanism is repetitive peeling of extremely oriented graphite. Highly ordered pyrolytic graphite (HOPG) is used in this process for the synthesis of graphene. This method is capable of generation of the atomically thin graphene sheet. This includes peeling off one or a few sheets of graphene

Table 1 Properties of graphene [54]

Thermal cond.	Thermal resistance	Specific surface area	Young’s modulus	Fracture strength	Optical transmittance	Sheet resistance	Relaxation length	Phase coherence length	Mobility
$\sim 5000\ \mathrm{Wm^{-1}k^{-1}}$	$\sim 4 \times 10^{-8}\ \mathrm{km^2\ w^{-1}}$	$2300\ \mathrm{m^2g^{-1}}$	~ 1 TPa	130 GPa	$\sim 97.7\%$	1.3×10^4–$5.1 \times 10^4\ \Omega\ \mathrm{sq^{-1}}$	$15{,}000\ \mathrm{cm^2\ v^{-1}\ s^{-1}}$	3–5 μm	$10^8\ \mathrm{A\ cm^{-2}}$

using scotch tape and then depositing it on the substrates. The graphene produced by this synthesis route is of the highest quality with least defects but this process has some constraints, i.e., low productivity [21, 22].

2.2 Chemical Exfoliation

It is another possible synthesis route to obtain graphene which includes wet chemical processing. This processing route includes insertion of the graphite with the reactants which softens the Van-der Walls interactions and helps in the production of graphene sheets. In this synthesis route, the graphite is immersed in the acidic solution generally nitric or sulphuric acid. This method is generally done in two steps: Fist step includes thermal processing and Second includes ultra-sonication to disperse them. This result in the generation of the graphene chemical compounds sheets suspended in the colloidal suspension, which is further deposited on the substrate [23]. In order to achieve the pure quality graphene flakes, the chemical compounds must be removed from the colloidal suspension in the reduced atmosphere using alkaline solutions, by applying hydrogen plasma, by reducing hydrazine vapors or by heat treatments. This chemical exfoliation method has some disadvantages as the graphene flakes obtained are partially oxidized because the reduction processes are not so efficient. Another disadvantage is that the sp2 like graphene bonds are partially degraded to sp2–sp3 structure. The main advantage of this synthesis route is that it permits the correct management of the dimensions of graphene sheets. The dimension of the graphene sheet depends on the time period of sonication. Longer the sonication processes smaller the dimensions of graphene sheet [13]. Another advantage of this route is that high output of the graphene which makes it economically competitive. This route is best to be used for the production of composite materials, coating and for the biomedical applications.

2.3 Epitaxial Growth

It is another synthesis technique which involves the epitaxial growth on the crystalline carbide wafers substrate. In this process, a very thin layer of graphite is used under controlled atmosphere and proper conditions to produce graphene monolayer. A layer of monolayer graphene is produced on the SiC by heating the C–SiC at very high-temperature under the argon/vacuum atmosphere. The silicon which is close to the surface sublimates the carbon atoms but not at high-temperature the graphite reorganizes, and thus graphitization is achieved [24]. High surface roughness (Ra) of graphene is obtained from the Si–face. This synthesis route found is an application in semiconductor industry, semiconductor devices as it is easily deployable. But this synthesis route still required improvement for low-temperature processes as compared to other synthesis processes. The main disadvantage of this method is that the graphene produces by this fabrication route have grain defect and grain boundaries due to which it cannot be perfectly homogenized. The quality as well as quantity produced by this synthesis route is not as much in comparison to the mechanical exfoliation and chemical exfoliation synthesis process, respectively.

2.4 Chemical Vapor Deposition

It is a well-known synthesis process in which the substrate is exposed to gaseous compounds. The graphene growth on the substrate surface is due to thermal decomposition of hydrocarbon gas molecules, i.e., methane/propane/acetylene catalyzed by a metal surface attribute to the segregation or precipitation of the carbon atoms from the metal. Mainly the transitions metals are used as a catalyst for the production of different allotropy of the carbon, so the main focus will be on them for the generation of graphene [25]. In the CVD process, the chemical constituents react in the vapor phase near/on a heated substrate to form a solid deposit. CVD process includes various chemical reactions such as thermal decomposition, reduction, hydrolysis, disproportion, oxidation, carburization, and nitridation. These processes can be used either individually or in combination. The main disadvantage of this processing route is that graphene is to be transferred from the metal to the actual appropriate substrate. It produces high-quality graphene and is mostly used for electronics applications, but the transfer of the graphene from the metal to the actual substrate sometimes leads to the improper alignment which increases the scope of error.

2.5 Hummer Method

This processing route is used for the synthesis of the graphene oxide. In this process, oxidation of graphite flakes are done to get the desired material. In this synthesis process, graphite flakes and sodium nitride are mixed in concentrated sulfuric acid in a volumetric flask kept at low temperature (0–5 °C) which is continuously stirred for 120 min. After that, the potassium permanganate is added to the suspension. The rate of addition of potassium permanganate is carefully controlled to keep the reaction temperature low (below 15 °C). The mixture is then stirred until it becomes pasty brownish. The paste is then kept stirring for two days after that its solution is diluted with the slow addition of water. The reaction temperature increases and the solution color changes to brown color. The solution is finally treated with hydrogen peroxide to terminate the reaction by the appearance of yellow color. Now, the process of purification takes place in which the mixture is washed by rinsing and centrifugation with hydrochloric acid and then with de-ionized water. The final step is the drying process under vacuum at room temperature, to produce graphene oxide as powder

Table 2 Synthesis methods and mechanisms [13, 21–28]

Synthesis method	Mechanism	Precursors used	Graphene produced/ number and size
Mechanical Exfoliation	Peeling off layers using scotch tape	Graphite	Pristine (SLG, FLG; 10 μm)
Chemical exfoliation	Decomposition of graphite based compounds, reduction, and subsequent exfoliation	Graphene oxide/ graphite flakes	Chemically Modified Graphene (SLG, FLG; > 100 μm)
Epitaxial growth	Thermal decomposition of hydrocarbons on top of SiC crystals	SiC	Pristine (SLG, FLG, > 50 μm)
Chemical vapor deposition	Carbon segregation or precipitation over transition metals	Transition metals, Polycrystalline Ni films, Copper Foils	Pristine (SLG, FLG, > 100 μm)
Liquid-phase exfoliation	Exposing graphite flakes or graphene oxide to solvents and applying sonication	Graphite/GO	Pristine (SLG, FLG, 20 μm)
Unzipped CNT	Longitudinal unzipping of CNT	MWCNT	Chemically Modified Graphene (SLG, FLG, 10 μm)
Solvothermal synthesis	Pyrolysis and filtering of solvothermal product	Solvothermal Products (Na + Ethanol)	Chemically Modified Graphene (SLG, FLG, 20 μm)

[26, 27]. Another improvement in the hummer method to get the high-quality graphene is the modified hummer method which involves both oxidation and exfoliation of graphite sheets due to thermal treatment of solution [28]. The summary of the synthesis methods and mechanisms are given in Table 2.

3 Tribological Behavior of Graphene

Graphene played a vital role in reducing friction and wear in various mechanical and tribological applications. So the detailed literature review of graphene as an antifriction, antiwear reinforcement in composites and as an additive in lubricants for low as well as high-temperature applications are discussed in the sections below.

3.1 Friction and Wear of Graphene

Many researchers worked a lot to reduce the friction and wear behavior at the micro as well as nanoscale by introducing graphene coatings as a solid lubricant in various tribological applications [29, 30]. Graphene produced by using chemical vapor deposition synthesis route was estimated under the normal load up to 70 μN. While examining the frictional behavior of the CVD-graphene, it was found that the Coefficient of Friction (COF) is affected by various parameters as well as the materials of the initial substrate. It was observed that the coatings grown on the nickel exhibit better frictional behavior than that of the coating grown on the copper metal. It was also found that the COF considerably enhanced while the transfer of the graphene from the particular substrate like copper and nickel. The tribological properties of the coatings mostly depend on the adhesion between the substrate and the coatings. It was suggested from this research that few nanometer thick graphene samples proved to be good as solid lubricant at both micro as well as nanoscale [31]. Shin et al. in their investigation studied the COF of graphene produced by Epitaxial Growth and exfoliation method. The graphene with few layers under normal load up to 0.5 μN using an AFM tip radius of one meter was studied. It was observed that COF was not dependent on the number of layers. The COF for one to three-layer graphene was around 0.03. They also studied the defects in the structure of the defect. It was observed that the defects in the crystal structure of the graphene increase the COF by two to three times [32]. Won et al. studied the friction and wear mechanism of CVD produced graphene on the copper substrate under load of 20 μN with chrome steel ball as the counterbody (1 mm diameter). It was reported that the deposition parameters are crucial to get the graphene with minimum defect. It was also reported that the number of the layers varying from one to seven does not affect the COF a lot [33]. Yan et al. in their investigation studied the COF of graphene transferred on to the substrate under the load of up to 40 μN. It was reported that the applied load is a critical parameter which influenced the friction coefficient [34]. Berman et al. reported that the multilayer graphene flakes may be successfully used as a solid lubricant for chrome steel. It was observed that employing a low concentration of graphene flakes reduced the COF by six times [35, 36]. The brief summary of the friction behavior of graphene in micro- and nanoscale is given in Table 3.

3.2 *Tribological Behavior of Graphene as Reinforcement in Composites*

Several investigations have been done to study the tribological behavior of graphene as reinforcement in various composites materials. Tai et al. [37] studied the tribological behavior of Ultra-High Relative Molecular Mass Polythene (UHMWPE)/Graphene Oxide (GO) composite. The composite samples were fabricated using toluene-assisted mixing followed by hot-pressing technique. The tribological and mechanical properties of base composite and the GO/UHMWPE composites were examined. It was reported that GO nanosheets content up to 1 wt %, increases the wear resistance and hardness of the composites very significantly, while the friction coefficient will increase rapidly with the increase in the

Table 3 Tribological behavior of graphene on micro/nanoscale [29, 36]

Graphene in Microscale tribology					
Substrate	Deposition method	Ra (nm)	Counterbody	Contact pressure, load/operating conditions	COF
SiO_2	CVD	1	Fused silica lens	5–10 μm (Air, Room Temperature)	0.15
SiC	CVD	1	Diamond tip	0.05 μm (Air, Room Temperature)	0.07
Ni	CVD	–	Si_3N_4 ball	2 N (Air, 24 °C) (45% RH)	0.06
Cu	Solution	–	Steel ball	220 Mpa, 20 μN (Air, Room Temperature)	0.2
Steel C440	solution	15	Steel ball	1–5 N (Air, Room Temperature)	0.15
PET	Solution	5	SiO_2	20, 40 μN (Air, Room Temperature)	0.08
Ni	Exfol.	1	Fused silica lens	5–10 μm (Air, Room Temperature)	0.05
Si	Exfol.	1	Diamond tip	0.05 μm (Air, Room Temperature)	0.07
Si	Sputtering	–	Steel ball	100–400 μN (Air, Room Temperature)	0.12
Graphene in Nanoscale tribology					
Si	Exfol.	0.5	SiN	0.01–0.5 Nn (Air, Room Temperature)	0.025
SiO_2	Exfol.	0.5	Si	1 nN (Air, Room Temperature)	0.6–0.15
SiO_2	Exfol.	0.5	Si	1 nN (Air, Room Temperature)	–
Si	Exfol.	0.5	Si	1 nN (Air, Room Temperature)	–
SiO_2	Exfol.	0.5	DLC	5 nN (Air, Room Temperature)	0.4–1.0
Si	Exfol.	0.5	Si	3–30 nN (Air, Room Temperature)	0.02
Si	Solution	0.5	Si	1 nN (Air, Room Temperature)	0.10
SiO_2 (GNP Coated)	Solution	–	SiO_2 Ball (DLC coated)	1 nN (Dry Nitrogen)	0.004
SiO_2 (GNP Coated)	Solution	–	SiO_2 Ball	1 nN (Dry Nitrogen)	0.04
SiC	Thermal decomp	–	Si	40 nN (Air, Room Temperature)	0.2

reinforcement. Min et al. [38] investigated the Graphene Oxide (GO)/Polyimide (PI) Nanocomposite fabricated using situ polymerization. They studied the tribological behaviors of the composite under dry friction, seawater lubrication, and pure water lubrication conditions. The GO/PI composite exhibited better results beneath seawater-lubricated condition than other conditions because of the excellent lubricating effect of seawater. GO as reinforcement greatly improved the thermal stability of the composites. The tensile modulus and tensile stress of the nanocomposite improved significantly by adding graphene oxide (GO). The incorporation of GO under seawater lubrication can greatly improve the wear resistance of Polyimide. Best results were obtained with 0.5 wt% GO reinforced to PI composite. Zhu et al. [38] studied the dry sliding tribological behavior of Ni_3Al matrix composites (NMCs). It was reported that Ni_3Al matrix composites with 0.5 wt% graphene nanoplatelets (GNPs) sliding against different counter face balls with an applied load of 10 N and a sliding velocity of 0.234 ms^{-1}. When the composite sliding against GCr15 steel, a consistent and thick friction layer is formed, leading to a lower COF, but when the composite sliding against Si_3N_4 and Al_2O_3, the formation and stability of the friction layers are restricted within the severe wear regime, and thus composite exhibit higher friction coefficients and wear rates. Yao et al. [40] studied the combined effect of lubrication of WS2 and multilayer graphene (MLG). The prepared sample of NiAl–5 wt% WS2 (NB)–1.5 wt% MLG exhibited excellent tribological properties. The MLG play the role of reinforcement particles and improved loading carrying ability. The addition of a combination of MLG and WS2 offered to possessed superior antifriction and the wear resistance. Gonzalez et al. [41] studied the dry sliding behavior of a graphene/alumina composite against alumina under dry conditions. The testing was done on the reciprocating tribometer with an applied load of 20 N, a sliding distance of up to 10 km and a sliding speed of 0.06 ms^{-1}. The composite showed a 10% lower friction coefficient and half the wear rate than the monolithic alumina. It has been also found that this behavior is related to the presence of graphene nanoplatelets (GNPs). GNPs form a self-lubricating layer that provides enough lubrication so as to decrease both friction coefficient and wear rate. These GNPs act as a self-lubricating layer on the contact surface between the composite and the Al_2O_3 ball that acts as counterpart material. Yazdani et al. [42] studied the tribological performance of the hot-pressed pure alumina and its composites containing numerous hybrid contents of GNPs and carbon nanotubes (CNTs) under different loading conditions. The composite reinforced with 0.5wt% graphene nanoplatelets reduced the COF up to 23% and enhance the wear rate by 70%. The hybrid reinforcement consisting of 0.3 wt% GNPs and 1 wt% CNTs shows even better performance, with an 86% reduction in the wear rate. GNPs played a vital role in the formation of a tribo-film on the worn surface by exfoliation. Llorente et al. [43] studied the friction and wear behavior of graphene/silicon carbide (SiC) composites under the dry sliding conditions and using silicon nitride balls as countersurface. GNPs composites showed an improvement in the wear resistance as compared to monolithic silicon carbide, with enhancements of 70 vol. % for the material containing up to 20 vol. % of GNPs. Under dry sliding conditions, the wear resistance of SiC ceramics considerably enhances with the addition of GNPs. 20 vol. % GNPs composite clearly showing the best wear resistant performance which is 70% more as compared to the monolithic SiC, whereas 5 vol. % reduced graphene oxides composites exhibit excellent fracture toughness. Kalin et al. [44] studied the effect of the morphology solid lubricant nanoparticles on Poly-Ether-Ether-Ketone (PEEK) composites on the mechanical and tribological characteristics. The results obtained under dry sliding tribological conditions show that the materials have an important effect on the friction coefficient and the wear, primarily by affecting their hardness. The carbon-based particles deteriorated the wear and tear behavior by 20 wt% (CNT) and the maximum amount of three times in the case of the GNP. Tabandeh et al. [45] studied the tribological behavior of Al matrix composites reinforced by GNPs, on a pin-on-disk tribometer. The results showed that the wear of Al-1 wt% GNP is enhanced with an increase in the normal loads. However, the friction coefficient of the Al-1 wt% GNP reduced with increasing normal loads. It has been found that the GNPs reinforced nanocomposites showed excellent tribological properties. Belmonte et al. [46] studied the tribological properties of GNPs/Si_3N_4 composites in a reciprocating ball-on-plate tribometer under iso-octane lubrication. GNPs are excellent nanofillers which improve the tribological performance of ceramics. Under the high contact pressures, GNPs are able to decrease the friction and enhance the wear resistance up to 56% due to the exfoliation of the GNPs that creates a protective tribo-film. The exfoliation of the nanoplatelets (NPs) generates graphene flakes, which effectively limits wear volume by protective tribo-film. Xu et al. [47] studied the self-lubrication characteristics of multilayer graphene and high-temperature tribological properties of graphene titanium aluminum matrix composite from the temperature ranges from 100–700 °C using a rotating ball-on-disk tribometer at a load of 10 N and speed of 0.2 ms^{-1}. During the temperature range from 100 to 550 °C, MLG presents good lubricating properties. Above 600 °C, MLG lost their self-lubrication characteristics due to the formation of the oxide layer which improves the oxidation resistance of GTMC by restricting the grain boundaries and inhibiting the inflow of oxygen through grain boundaries. The brief summary of the test parameters of graphene as reinforcement composite in different materials are given in Table 4.

3.3 Tribological Behavior of Graphene as Lubricant Additives

Lin et al. [48] investigated the chemically modified the graphene platelets with oleic and stearic acids in a reflux reaction. They then investigated the tribological properties of modified graphene platelets based lubricant using a four-ball tester. The results show that the lubricants containing 0.75 wt% of the MGNP additive is efficient to reduce the wear of the materials and there is also an increase in the load carrying capacity of the lubricant. Senatore et al. [49] investigate the tribological behavior of graphene oxide nanosheets in mineral oil under boundary, mixed, and elastohydrodynamic lubrication regimes. The GO nanosheets were synthesized by using modified hummer method. Experimental studies were done on the pin-on-disc configuration tribometer. The studies concluded that the lubricant-containing GO additives are more efficient to reduce the friction behavior by forming a protective layer between two tribo surfaces which prevent direct metal-to-metal contact. Zhe et al. [50], in their study, compared the performance of lubricating oils with different additives. It was observed that most of the additives are toxic and produces poisonous gases when burnt in the environment; but the lubricant congaing GO as an additive is eco-friendly and green lubricant as GO consist of C, H, and O. It was also reported that the GO-based lubricating oil is efficient to reduce the wear and friction in the material even at high temperature. These GO-based lubricant also proved to be good for high sliding speed conditions. 0.5 wt% GO as an additive proved to be best in all the composition variations. Dou et al. [51] in their

Table 4 Tribological properties of graphene as reinforcement in composites [37–47]

Material	Reinforcement	Fabrication route	Test motion/counterbody/ normal load (N)/temperature	Contact velocity/ sliding distance	COF	Wear rate (mm^3/Nm)/ Weight loss (µg)
UHMWPE	0.1 wt% GO	Hot pressing	Reciprocating/ZrO_2 ball/5 N	9 cm/s	0.115	$1.35*10^{-5}$
UHMWPE	0.3 wt% GO				0.116	$1.53*10^{-5}$
UHMWPE	0.7 wt% GO				0.116	$1.1*10^{-5}$
UHMWPE	1.0 wt% GO				0.120	$1.1*10^{-5}$
UHMWPE	2.0 wt% GO				0.115	$1.05*10^{-5}$
UHMWPE	3.0 wt% GO				0.115	$1.03*10^{-5}$
Polyimide	0.1 wt% GO	Situ polymerization	Reciprocating/steel ball/3 N	0.156 m/s	0.41	$6*10^{-6}$
Polyimide	0.3 wt% GO				0.46	$13.5*10^{-6}$
Polyimide	0.5 wt% GO				0.54	$8*10^{-6}$
Polyimide	0.7 wt% GO				0.49	$7.9*10^{-6}$
Polyimide	1.0 wt% GO				0.55	$6.5*10^{-6}$
NiAl	1.5 wt% MLG	Spark plasma sintering	Rotary/Ni_3N_4 ball/2, 6, 12, 16 N	0.2 m/s	(0.6–0.8), (0.5–0.6), 0.5, 0.42	$2.82*10^{-5}$, $3.57*10^{-5}$, $4.02*10^{-5}$, $5.24*10^{-5}$
NiAl	1.5 wt% MLG + 5 wt% WS_2		Rotary/Al_2O_3 ball/10 N		0.35	$0.40*10^{-5}$
NiAl			Rotary/Ni_3N_4 ball/10 N		0.21	$0.17*10^{-5}$
NiAl			Rotary/Al_2O_3 ball/10 N		0.25	$0.11*10^{-5}$
Al_2O_3	0.22 wt% GNP	Spark plasma sintering	Reciprocating/Al_2O_3/20 N	0.06 m/s/10 km	0.67	$3.5*10^{-9}$
Al_2O_3	0.5 wt% GNP		Reciprocating/Si_3N_4 Ball/5, 15, 25, 35 N	10 mm/s	0.40, 0.48, 0.58, 0.54	−, 2.3 µg, 6 µg, 5 µg
Al_2O_3	2 wt% GNP			10 mm/s	0.49, 0.55, 0.54, 0.53	−, 7 µg, 9 µg, 40 µg
Al_2O_3	5 wt% GNP			10 mm/s	0.45, 0.54, 0.51, 0.45	−, 62 µg, 80 µg, 160 µg

(continued)

Table 4 (continued)

Material	Reinforcement	Fabrication route	Test motion/counterbody/normal load (N)/temperature	Contact velocity/sliding distance	COF	Wear rate (mm^3/Nm)/Weight loss (μg)
SiC	5 wt% GO	Spark plasma sintering	Reciprocating/Si_3N_4 Ball/5 N	0.1 m/s/360 m	0.71	$1.4*10^{-3}$
SiC	5 wt% GO				0.75	$1.3*10^{-3}$
SiC	20 wt% GO				0.96	$0.5*10^{-3}$
Al	0.1 wt% GNP	Spark plasma sintering	Reciprocating/440C steel disc/5 N, 10 N, 15 N	100 RPM/1.13 km	0.36, 0.34, 0.25	0.008 gms, 0.001gms, 0.016 grms
Al	1 wt% GNP		Reciprocating/440C steel disc/5, 10, 15 N	50 RPM/1.13 km	0.339, 0.36, 0.258	0.011gms, 0.0180 gms, 0.022 gms
Al	1 wt% GNP		Reciprocating/440C steel disc/5, 10, 15 N	100 RPM/1.13 km	0.33, 0.33, 0.239	0.0051 gms, 0.015 gms, 0.02 gms
Al	1 wt% GNP		Reciprocating/440C steel disc/5, 10, 15 N	150 RPM/1.13 km	0.33, 0.33, 0.239	0.0120 gms, 0.0155 gms, 0.027 gms
PEEK	2 wt% GNP	Spark plasma sintering	Reciprocating/100Cr6 PIN/1 MPa	0.05 m/s	0.62	$8.8*10^{-6}$
Si_3N_4	3 wt% GNP	Spark plasma sintering	Reciprocating/Si_3N_4 Ball/5, 100, 150, 200 N	0.1 m/s/360 m	0.225, 0.185, 0.169, 0.16	$1.0*10^{-3}$, $1.1*10^{-3}$, $1.3*10^{-3}$, $1.5*10^{-3}$
TiAl	3.5 wt% MLG	Spark plasma sintering	Rotary/Ni_3N_4 Ball/10 N/100 °C	0.2 m/s	0.360	$0.91*10^{-4}$
			Rotary/Ni_3N_4 Ball/10 N/200 °C		0.350	$0.95*10^{-4}$
			Rotary/Ni_3N_4 Ball/10 N/300 °C		0.375	$1.00*10^{-4}$
			Rotary/Ni_3N_4 Ball/10 N/400 °C		0.375	$1.01*10^{-4}$
			Rotary/Ni_3N_4 Ball/10 N/500 °C		0.370	$1.40*10^{-4}$
			Rotary/Ni_3N_4 Ball/10 N/600 °C		0.510	$2.30*10^{-4}$
			Rotary/Ni_3N_4 Ball/10 N/700 °C		0.525	$2.20*10^{-4}$

investigation used the crumpled graphene ball as an additive in the Poly Alpha Olefin (PAO) lubricating oil. It is proved to be high-performance additive. 0.01–0.1 wt% of the crumpled graphene ball proved to be best to reduce the friction and wear of materials. Azman et al. [52] studied the effect of GNP as an additive in the palm oil trimethylolpropane ester blended Poly Alpha Olefin. It was observed that there is a decrease in the COF and wear rate of the material by using the blended lubricating oil with 0.05 wt% GNP as an additive. Meng et al. [53] in their investigation studied the tribological behavior of engine oil with 0.06–0.10 wt% Sc–Ag/GN as an additive. The tests were conducted in the four-ball tester. It was reported that the oil with additive proved to behave better tribological properties than the base engine oil. As the nano–Ag and GN particles form a protective film between the two materials and prevent metal to metal contact, hence reduce the friction and wear of the tribopair. Rasheed et al. [54] investigated the tribological performance of graphene-based nanolubricant in a four-stroke IC engine test rig. The lubricating oil SN/CF API 20W50 was used and 0.01 wt% additive was added to the lubricating oil. It was observed that there is a reduction in the COF by 21%, increase in thermal conductivity at 80 °C by 23% and enhancement in the heat transfer rate by 70% is achieved. Wei al. [55] used liquid-phase exfoliated modified graphene by oleic acid as an additive in the lubricating oil. It was found that there was an increase in the wear and friction of the material by 14% and 17%, respectively. It was reported that modified graphene as an additive also increases the load carrying capacity of the lubricant. Kinoshita et al. [56] investigated the behavior of graphene oxide nanoparticles in a water-based coolant to reduce friction. It was reported that the COF is reduced to 0.05 and no surface wear was reported for over 60,000 cycles. Eswaraiah et al. [57] in their investigation reported that there is an increase in the frictional characteristics, wear resistance, and extreme pressure properties by 80, 33, and 40% by the use of 0.025 mg/ml of graphene as an additive in the engine oil. The brief summary of the test parameters of graphene as additive in different lubricants are given in Table 5 (Table 6).

Table 5 Tribological properties of graphene as an additive in lubricants [48–57]

Lubricant	Additive	Material	Counterbody	Test motion/ temperature/ normal load (N)	Frequency (Hz)/Speed (rpm)	Stoke (mm)	COF
HC Base oli	0.5 wt% GO	ALSI 52100 Steel	ALSI 52100 Steel ball	Recipro/50 °C/ 50 N Recipro/50 °C/ 100 N Recipro/50 °C/ 150 N	50 Hz	2	0.13, 0.11, 0.105
HC Base oli	0.5 wt% GO	ALSI 52100 Steel	ALSI 52100 Steel ball	Recipro/100 °C/ 50 N Recipro/100 °C/ 100 N Recipro/100 °C/ 150 N	50 Hz	2	0.17, 0.15, 0.14
HC Base oli	0.5 wt% GO	ALSI 52100 Steel	ALSI 52100 Steel ball	Recipro/150 °C/ 50 N Recipro/150 °C/ 100 N Recipro/150 °C/ 150 N	50 Hz	2	0.18, 0.15, 0.14

(continued)

Table 5 (continued)

Lubricant	Additive	Material	Counterbody	Test motion/ temperature/ normal load (N)	Frequency (Hz)/Speed (rpm)	Stoke (mm)	COF
HC Base oli	0.5 wt% GO	ALSI 52100 Steel	ALSI 52100 Steel ball	Recipro/50 °C/ 100 N	10 Hz, 20 Hz, 30 Hz, 40 Hz, 50 Hz	2	0.122, 0.12, 0.119, 0.18, 0.117
PAO40	0.05 wt% GO	ALSI 52100 Steel	ALSI 52100 Steel ball	Reciprocating/ 100 N Reciprocating/ 200 N Reciprocating/ 300 N Reciprocating/ 400 N Reciprocating/ 500 N	30 Hz	2	0.121, 0.119, 0.125, 0.126, 0.130
PAO40	0.5 wt% GO	ALSI 52100 Steel	ALSI 52100 Steel ball	Reciprocating/ 100 N Reciprocating/ 200 N Reciprocating/ 300 N Reciprocating/ 400 N Reciprocating/ 500 N	30 Hz	2	0.118, 0.118, 0.116, 0.118, 0.124
PAO40	0.1 wt% GO	ALSI 52100 Steel	ALSI 52100 Steel ball	Reciprocating/ 100 N Reciprocating/ 200 N Reciprocating/ 300 N Reciprocating/ 400 N Reciprocating/ 500 N	30 Hz	2	0.117, 0.114, 0.112, 0.113, 0.116
PAO40	1 wt% GO	ALSI 52100 Steel	ALSI 52100 Steel ball	Reciprocating/ 300 N	10 Hz, 20 Hz, 30 Hz, 40 Hz, 50 Hz	2	0.09, 0.16, 0.112, 0.113, 0.114
PAO4	0.01 wt% τ–GO	ALSI52100 Steel disc	M50 steel ball	Rotary/10 N	10 mm/s	–	0.132
PAO4	0.1 wt% τ–GO	ALSI52100 Steel disc	M50 steel ball	Rotary/10 N	10 mm/s	–	0.131
PAO 10	0.01 wt% GNP	ALSI 52100 Steel	ALSI 52100 Steel	Rotary/392 N	1200 rpm	–	0.08
PAO 10	0.03wt% GNP	ALSI 52100 Steel	ALSI 52100 Steel	Rotary/392 N	1200 rpm	–	0.081
PAO 10	0.05wt% GNP	ALSI 52100 Steel	ALSI 52100 Steel	Rotary/392 N	1200 rpm	–	0.073
PAO 10	0.1wt% GNP	ALSI 52100 Steel	ALSI 52100 Steel	Rotary/392 N	1200 rpm	–	0.087
PAO 10	0.2wt% GNP	ALSI 52100 Steel	ALSI 52100 Steel	Rotary/392 N	1200 rpm	–	0.081
PAO 10	0.5wt% GNP	ALSI 52100 Steel	ALSI 52100 Steel	Rotary/392 N	1200 rpm	–	0.080
PAO 10	1wt% GNP	ALSI 52100 Steel	ALSI 52100 Steel	Rotary/392 N	1200 rpm	–	0.086
PAO 10	3wt% GNP	ALSI 52100 Steel	ALSI 52100 Steel	Rotary/392 N	1200 rpm	–	0.089
SN 350	0.075wt% MGP	ALSI 52100 Steel	ALSI 52100 Steel ball	Rotary/75°C/ 147 N	1200 rpm	–	0.121
SN 150	0.1 wt% GO	X155CrVMo12–1 steel disc	X45Cr13 steel ball	Rotary/25 °C/ 60 N	1 m/s, 1.5 m/s, 2 m/s	–	0.134, 0.141, 0.152
SN 150	0.1 wt% GO	X155CrVMo12–1 steel disc	X45Cr13 steel ball	Rotary/50 °C/ 60 N	1 m/s, 1.5 m/s, 2 m/s	–	0.131, 0.141, 0.153

(continued)

Table 5 (continued)

Lubricant	Additive	Material	Counterbody	Test motion/ temperature/ normal load (N)	Frequency (Hz)/Speed (rpm)	Stoke (mm)	COF
SN 150	0.1 wt% GO	X155CrVMo12–1 steel disc	X45Cr13 steel ball	Rotary/80 °C/ 60 N	1 m/s, 1.5 m/s, 2 m/s	–	0.141, 0.15, 0.16
SN/ CF20W50	Additive + G60	Steel Ball	Steel Ball	Rotary/40 N	1200 rpm	–	0.011
SN/ CF20W50	G60	Steel Ball	Steel Ball	Rotary/40 N	1200 rpm	–	0.014
SN/ CF20W50	G12	Steel Ball	Steel Ball	Rotary/40 N	1200 rpm	–	0.013
SN/ CF20W50	G8	Steel Ball	Steel Ball	Rotary/40 N	1200 rpm	–	0.014
SJ/ CF20W50	Additive + G60	Steel Ball	Steel Ball	Rotary/40 N	1200 rpm	–	0.017
SJ/ CF20W50	G60	Steel Ball	Steel Ball	Rotary/40 N	1200 rpm	–	0.018
SJ/ CF20W50	G12	Steel Ball	Steel Ball	Rotary/40 N	1200 rpm	–	0.017
SJ/ CF20W50	G8	Steel Ball	Steel Ball	Rotary/40 N	1200 rpm	–	0.017
10W40	0.1wt% GO	GCr15 steel ball	GCr15 steel ball	Rotary/343 N	1200 rpm	–	0.09
10W40	0.1wt% Ag/GN	GCr15 steel ball	GCr15 steel ball	Rotary/343 N	1200 rpm	–	0.09
10W40	0.1wt% Sc-Ag/ GN	GCr15 steel ball	GCr15 steel ball	Rotary/343 N	1200 rpm	–	0.078

Table 6 Overview of tribological properties of widely used solid lubricant [58, 59]

Solid lubricant coating	Deposition method	Coating thickness (μm)	COF	Friction/Wear mechanism
Graphite	Evaporation	0.2–5	Dry: 0.5–0.6 Humid: 0.1-0.2	Interlayer shear Water intercalation
DLC	Sputtering, Ion-Beam	1–3	Dry: 0.001–0.05 Humid: 0.2–0.3	High chemical inertness. The repulsive force due to hydrogen termination
Tetrahedral amorphous carbon	Ion-Beam, Cathode Arc, Pulsed Laser	0.01–1	Dry: 0.7 Humid: 0.1	Tribochemical induces a surface reaction Termination of a top carbon atom
Ultra Nano-Crystalline Diamond	MPCVD, HFCVD	0/5–1.5	Dry: 0.05–0.13 Humid: 0.007–0.1	Tribochemical induced reaction with H, O or OH
MoS_2&WS_2	Sputteing, Thermal Evaporation, CVD, ALD	0.2–2	Dry: 0.02–0.06 Humid: 0.15–0.25	Interlayer shear Transfer film formation
Graphene/Graphene Oxide (GO)	CVD, Mechanical and Chemical Exfoliation	0.001–0.002	Dry: 0.15–0.2 Humid: 0.15–0.2	Interlayer shear Prevention of tribo-corrosion

4 Conclusions

Graphene is proved as a unique and attractive material having promising mechanical, thermal, and tribological properties which found its application in the field of mechanical systems, electronic systems, and also in the field of biomedical. Several investigations make it clear that the graphene and its allotropes enhance the tribological properties of the composites as well as lubricants when used as reinforcement filler/coating or as additives, respectively. This review highlights the recent growth and development of graphene as a lubricant for micro as well as nanoscale applications in tribology. Despite its ultrathin nature it is proved to be effective for high temperature, high load dry sliding conditions. Graphene nanoparticles contributed directly to the latter film formation which plays an effective role in reducing the friction and wear of the tribopairs. Overall in this review graphene is proved to be a very effective material for reducing friction and wear. Growth in the synthesis process of graphene leads to explore some more applications in the field of tribology. The employment of graphene in tribological applications is expected to grow continuously shortly.

Acknowledgements I gratefully acknowledge all the researchers who have worked in the field of tribology, without their significant contribution. This review literature would have been difficult to summarize. I would also want to acknowledge my institute and supervisor for their wholehearted support.

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