

Graphene and Its Industrial Applications – A Review

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Abstract Graphene has been recently introduced as a promising material for various applications due to its outstanding mechanical, electrical, and thermal properties. It is classified as an allotropic form of carbon with the size of a single layer of graphite. This paper provides an extensive review regarding different critical applications of graphene including three categories namely: energy harvesting, strain sensors technology, and steel industry. The paper highlights what has been traversed in each category and provides an insight for researchers on what still needs to be investigated, which would open new horizons for scientific research and industrial applications. This field of research is expected to yield results that will have a considerable advance specially in 3D strain sensing technology. Also a special attention is given to the application of graphene in steel coating, steel welding, and lubrication.

Keywords Graphene, Energy Harvesting, Strain Sensors, Steel Industry

1. Introduction

Graphene is a 2D transparent nanomaterial that is both durable and flexible. It is classified as an allotropic form of carbon with the size of a single layer of graphite. In 2004, graphene was first introduced to the world when Andre Geim and Kostya Novoselov managed to segregate a single atomic layer of carbon. Since then, this promising material has drawn the attention of many researchers in the domains of materials science and mechanical engineering to manufacture and exploit it. There are two main techniques to fabricate graphene; the first is Top-down technique by breaking down of graphite into graphene using external mechanical or electrical, while the second way is Bottom-up by building up graphene from molecular building blocks or from carbon sources.

Novoselov et al. [1] compared four different synthesis processes of graphene with respect to the obtained quality and price as shown in figure (1). A list of characteristics such as the size of layer and applications were used to differentiate between the syntheses methods [2]. For coating applications, it was found, among other fabrication methods that the optimal fabrication methods of graphene films are chemical vapor deposition (CVD) and liquid phase exfoliation methods because both methods are scalable and show relative higher cost-efficiency than other methods. Graphene can either be used as a surface coating material as a composite or in a pristine form to improve corrosion

resistance [3] & [4]. It is chemically inert and has impermeability to most of gases. These outstanding properties reinforce its chances to be the most suitable material in coating purposes [5,6].

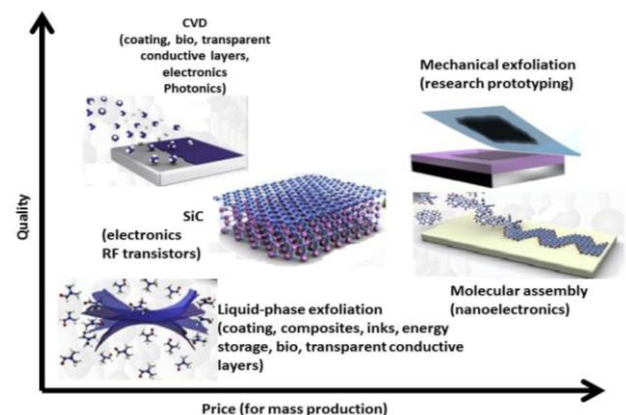


Figure 1. A comparison between various methods for production of graphene regarding to the price and quality [1]

Many research efforts focused on the mechanical characterization of graphene to test its suitability for industrial applications. It was found that a graphene layer possesses a modulus of elasticity around 1 TPa and a tensile strength of almost 130 GPa. Also, it showed a significant thermal conductivity above $3000 \text{ Wm}^{-1}\text{K}^{-1}$ and excellent electron mobility at room temperature with values more than $15000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ [7]. High electric conductivity that graphene possesses [8-10] promoted it to be exploited as a conductor in the field of sensors technology and energy harvesting [11].

For energy storage applications, the corrosion behavior and electrical conductivity are the two main properties to be taken into consideration. Many types of steel require surface

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Published online at <http://journal.sapub.org/ijme>

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modifications to enhance their electric conductivity while maintaining high corrosion resistance for the long-term service. Hence, several coating systems have been considered to improve steel performance in energy storage applications, e.g., polymer-based coatings [12,13], multilayer coatings [14,15] and ceramic based coatings [16,17]. As for polymer coatings, they easily initiate microcracks in mechanical structures and reducing their reliability. This increases the need to find an alternative which can be adopted for long life span applications.

Moreover, the strain gauges have been in use for many years and are the main sensing element for several types of sensors such as pressure sensors, load cells, torque sensors, and position sensors. They are considered as smart devices which have a wide range of applications where the mechanical deformation or structural changes can be detected, such as damage detection, characterization of structures, fatigue studies of materials and the internal activities in human body [18].

One key parameter in selecting sensing material for a strain gauge is the type of application itself. Recent examples for strain sensors are in artificial intelligent systems, humanoids, and human health monitoring [19,20]. Popular strain-sensitive materials are of two types: semiconductors and metallic materials. Static strain gages are usually made of a copper-nickel alloy and this material enables a minimum variation of resistance with temperature. Dynamic strain gauges, however, are often fabricated from iron-nickel-chrome alloy, which enhances gage factor as compared to static one. Karma (a nickel chrome alloy) is also another alternative material which is used to fabricate strain gauges [21].

The metal-based strain sensors are relying on the geometrical change of conduction path and have gauge factor (GF) ranging from 2-5 [22,23]. They, also, represent a reliable and prevailing technology. Semiconductor strain sensor types take advantage of piezoresistive effect and raise the GF to be greater than 100 [24].

Although the traditional strain gages based on alloys and semiconducting materials possesses an improved sensitivity (nearly 103 of gauge factor), they are not capable for application in specific fields due to their poor stretchability (nearly 5%) [25]. In addition, they are more brittle and only designed for small strains. Along with the continuous efforts which have been exerted to obtain novel types of strain gauges with higher gauge factors (sensitivity), cheaper, and easier use, different materials have been implemented including carbon derivatives, such as low-dimension carbon allotropes [22,26].

Till the moment, various techniques have been suggested to fabricate highly stretchable and sensitive strain sensors using conductive polymer composites containing nanofillers, e.g. silver nanowires [4], carbon nanotubes (CNTs) [27], and graphene (GE) [28]. The next sections summarize the recent research contributions classified into three main categories: strain sensors, energy harvesting, and steel coatings.

2. Graphene Synthesis Methods

There are two different approaches for graphene manufacturing as shown in figure (2). The first one is Top-down approach, in which, graphite is broken down into graphene flakes using mechanical or electrical external forces. The second approach runs in an opposite way, which is achieved by building up graphene from molecular building blocks or from carbon sources [29]. In order to fulfil industrial needs, there are two important factors concerning scalability when focusing on the proficiency of any synthetic route to graphene. First one, a process must produce enhanced quality in the 2D crystal lattice to ensure high mobility. In the second one, the method must provide fine control over crystallite thickness in order to deliver uniform device performance [30].

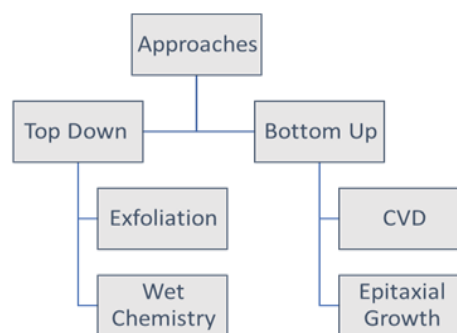


Figure 2. Different routes for graphene production

In this review different synthetic methods are discussed under the two mentioned categories of graphene manufacturing taking into concern that any synthetic route can be judged by four main factors: Quality of the produced graphene, the controllability of process, the complexity of process, and amount of produced graphene.

2.1. Exfoliation Method

This method has two main branches scotch tape and liquid phase exfoliation.

In scotch tape technique, multiple layers of graphene stick to the tape after peeling it off the graphite. By repeated peeling, the number of graphene layers is reduced. Commercial solvents such as acetone are used to remove the tape glue. Finally, the tape is removed by unused peeling process. This method is featured by low complexity, high quality of produced graphene, lack of controllability, and limited amount of produced graphene [31].

Liquid phase Exfoliation (LPT) technique is based on dispersing the graphite in an organic solvent which has nearly the same surface energy as graphite [32]. The solution is sonicated in an ultrasound bath for several hundred hours or by applying a potential difference [33]. After the dispersion, thicker flakes are to be disposed by centrifugation of solution. The quality of obtained graphene is nearly the same as in scotch tape technique, whereas the amount of produced graphene is much higher with lower complexity.

2.2. Wet Chemistry

The most common technique here is called "Hummer's Method". The benefits of the hummer's method are its low-cost and massive productivity [30]. This technique is achieved by adding graphite powder to sodium nitrate. Afterwards, a concentrated sulphuric acid is added during stirring followed by adding KMnO_4 progressively to the mixture while maintaining low temperature (less than 20°C) to prohibit overheating and predicted explosion. The mixture is stirred for 12 hrs. while keeping the temperature at 30°C . Then, water is added under potent stirring to dilute the mixture. To ensure the completion of reaction with KMnO_4 , the suspension should be treated with 30% H_2O_2 solution. Finally, graphene oxide sheets are, thus, obtained by using HCl and H_2O to wash the resulting mixture, which is followed by filtration and drying [34].

2.3. Epitaxial Growth

Graphene can be synthesized by heating SiC precious stone at a somewhat high temperature ($\sim 2000\text{ K}$) in ultra-high vacuum. Multilayered graphene structure is yielded after desorption of Si from the top layers of SiC crystalline wafer and the produced structure acts like graphene [35]. Controlling the obtained number of layers occurs by constraining time or temperature of the warmth treatment. The ability to produce large amounts of graphene through epitaxial growth is much lower compared to liquid-phase exfoliation. Also, the intricacy of this technique is comparatively low. Moreover, the quality of produced graphene is lower as compared to the exfoliated one [31].

2.4. Chemical Vapor Deposition (CVD)

Graphene layers are grown in a closed chamber by exposing of a Ni film to a mixture of H_2 , CH_4 , and Argon at about 1000°C (Samples are heated by filament or plasma) [36]. After decomposing the methane into C and H_2 , the hydrogen evaporates followed by the diffusion of carbon atoms into Ni substrate surface. By cooling down in an Ar atmosphere, a layer of graphene begins to grow on the surface. Furthermore, controlling the pattern of the Ni layer is used to control the shape of the produced graphene. It is possible to use a polymer support over which graphene layers could be transferred. After the Ni substrate is etched, the polymer support is peeled off resulting stamped graphene over the substrate. This method can be used to decrease the resistance by stamping of graphene layers on each other [31].

It was also reported that Ni has a good solubility for carbon atoms and this solubility depends on the cooling rate. At low cooling rates, there is no segregation because C atoms have sufficient time to diffuse through Ni substrate. At moderate cooling rates, C atoms begin to segregate and form graphene. At higher cooling rates, C atoms clearly segregate out of Ni, forming a defective graphitic structure with lower crystallinity [37]. Moreover, the replacement of nickel substrate by a copper one had also been reported to result in a single-layer graphene with a few-layer graphene structure

(less than 5%), which didn't grow larger with time [38].

Wang et al. [39] had demonstrated a new method of growing substrate-free few-layered graphene. MgO-supported Co catalysts were used to grow graphene in a ceramic boat (at 1000°C for 30 min) under CH_4 and Ar gas envelope of 1:4 volume ratio and total 375 mL/min flow rate. After the reaction, MgO and Co were washed from the product by using concentrated HCl , followed by rinsing with clean distilled water and drying at 70°C . They confirmed, by microstructure characterization using SEM, HRTEM and Raman spectroscopic, the presence of rippled graphene sheets, containing at least five layers.

3. Recent Trends of Applications

It is becoming a fact that graphene is considered as an exact two-dimensional gapless semiconductor and the most durable material ever measured which can endure up to 25% tensile elastic strain [40]. As mentioned in the previous sections, the exceptional corrosion resistance property makes it the best candidate as a corrosion barrier for steel industry. The new trends of applications are summarized in the upcoming sections.

3.1. Strain Sensors

Recent research efforts focused in utilizing Graphene as an elastic material with weak sheet resistance in the manufacturing of improved strain sensors [41-43]. Moreover, 2D nanoflakes usually offer a piezoresistivity which is more than nanowires by one order of magnitude, as their electrical percolation network is hugely suffers from geometrical discontinuities [41]. The implementation of this idea on a large scale is constrained by economic challenge manifested in high production cost of graphene and CNTs.

Carbon nanotubes and graphene have shown outstanding mechanical, electrical, piezoresistive and other physical properties which make them suitable for several applications in recent years. However, compared to CNTs, graphene have attracted enormous attention due to an ideal 2D structure which has its own merits in scalable devices fabrication through top-down approaches [40,44]. Moreover, graphene is a lower priced material with higher availability in a large-scale industrial production compared to CNTs.

There are different fabrication procedures and structures in graphene-based strain sensors providing various properties suitable for different applications (0D fullerene, 1D fiber, 2D film, 3D porous structure) [45]. The 3D graphene-based structures are very important part in the manufacturing of strain sensors because they can be massively produced in large quantities. Also, these structures can be stretched in all Cartesian axes and detects tiny or large force with ultrahigh sensitivity [45].

Fu et al. fabricated a strain sensor using CVD-grown graphene with higher sensitivity (GF~151) [46]. Also, J. Zhao et al. developed a graphene-based strain sensor with a sensitivity factor of more than 300 [47]. They illustrated the

piezoresistive behavior of the graphene films by using a charge tunneling model. Bao *et al.* had fabricated a more advanced strain sensor with a sensitivity factor of around 150 through using a sprayed solution technique [22]. The properties of the graphene films were explained through a percolation networks model. In addition, Li *et al.* created graphene woven fabrics with a gauge factor of 103 under 2-6% strains and 106 under higher strains [48]. Wang and collaborators [49] fabricated a composite of graphene Nanoplatelets with PDMS through simple sonication and molding processes. This device reached gauge factors of approximately 230 at a GNPs concentration of around 8 vol %, and within a strain of 2%.

Yong Lin *et al.* [50] fabricated a highly stretchable strain sensor with a high gauge factor from graphene elastomer composites by subsection functionalized graphene into the mixture of NR latex (NRL) and SBR (SBRL) latex creating electrostatic interactions between functionalized graphene (f-GE, positively charged) and NR latex particles (negatively charged) and they found that the fabricated strain sensors manifested outstanding characteristics such as good stretchability (up to 120%), high sensitivity (GF~82.5) and acceptable fatigue life (about 300 cycles) even when strained up to 100%, which makes able to detect and measure human motion under large strain. Moreover, the double-interconnected GE network enables the strain sensors with the excellent performances.

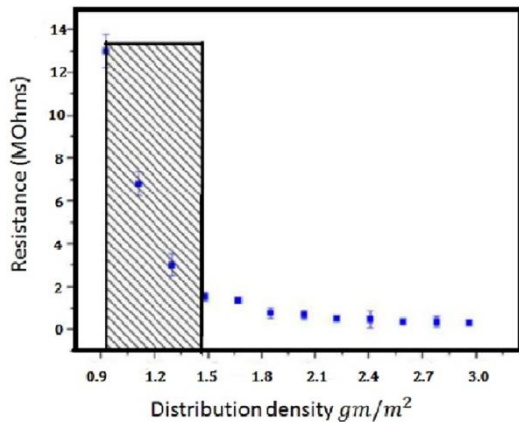


Figure 3. a) The film resistance as a function of the distribution density of graphene, b) Corresponding changes in resistance under different strains. (Inset) Reliability measurement of gauge factor over 100 strain cycles [18]

Dong Zhang, *et al.* [18] had synthesized novel type of strain sensors by using a spray coating technique to fabricate a sandwich-structured composite through sandwiching polymer layers (Styrene-Acrylate emulsion) between graphene layers and the substrate (stretchable rubber) and this composite was tested by digital multimeter to measure electrical resistance during tensile test. They discovered that the manufactured strain sensors which could endure a large tensile strain (25% strain) demonstrated high sensitivity to mechanical strain with GF of 6-35. Also, the resistance of the samples decreased with the increasing of the distribution density from 0.93g/m² to 1.48 g/m², and

kept almost unchanged at the distribution density from 2.59 g/m² to 2.96 g/m² as shown in figure 3. In Addition, the results demonstrated that these graphene composites were more sensitive compared to those without the polymer layers. Thus, the internal crack size in the composite films progress with time as a result of increasing strain. This results in a decrease of conductive paths and an increase of the sensitivity factor.

Carmela Bonavolontà *et al.* [51] had presented a novel route to synthesis a graphene-based film on a PMMA substrate fabricated by spreading graphene layers on PMMA substrate. They studied the piezoelectric response through applying bending stress to the composite sample and they used an infrared camera (Flir ThermoCam, SC6000) to monitor the outer shell of the PMMA/graphene material in order explain thermal changes.

The same authors in [51] demonstrated a gauge factor of 50 from the relationship between the electrical resistance and the applied stress. Also, they investigated the influence of bending on the electrical properties of the composite material by measuring the Voltage-Current (V-I) utilizing constructed electrical connection and they found that the increase of bending force yields decreases in current as shown in figure (4).

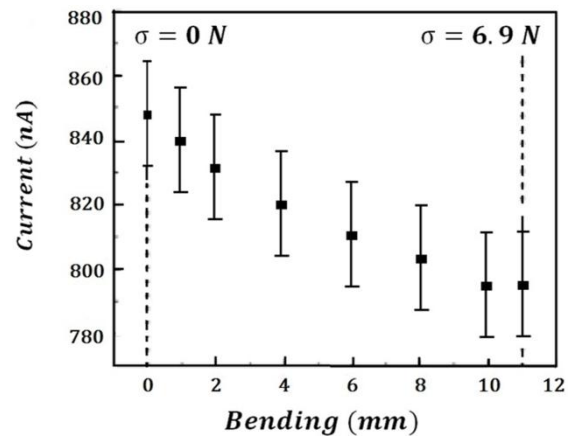


Figure 4. Electrical current variation due to the mechanical stress as measured on a PMMA/graphene sample. A constant voltage bias $V = 5$ V is applied to the sample while the applied force varies cyclically between unload ($F = 0$ N) and load ($F = 6.9$ N) [51]

Jung Jin Park *et al.* [52] used a layer by layer assembly technique to fabricate new types of graphene strain gauges based on stretchable yarns. They chose this fabrication method due to its simplicity, cost effectiveness, and scalability and they observed that these sensors showed high stretchability (up to 150%) and versatility. They also found that this type of sensors can be used to detect human motions in different scales. In Addition, they compared three types of yarns; rubber yarns (RY), nylon covered rubber (NCRY), and wool yarns (WY) and they showed that RY become elongated when stretched and this stretching resulted in the generation of cracks in the GNP coating on the surface of the fibers, which led to the breakage of electrical paths, i.e. an increase in electrical

resistance takes place. As for NCRY, piezoresistive property of the NCRY sensor depends mainly on the deformation of the outer nylon fibers. Under strain, the inner rubber fibers were stretched in the direction of the external force, while the nylon fibers were not directly stretched but deformed along the stretching direction. This dual structure minimized the elongation of the nylon fibers and suppressed the formation of prominent cracks in the GNP coating, which resulted in a smaller increase in the resistance of the NCRY sensor, compared to that of the RY sensor for the same strain. For WY, if it was stretched, the curled wool fibers became straightened and closer to each other. Furthermore, the subsequent contact between the GNP-coated wool fibers increased electrical paths and, hence, a decrease in the electrical resistance.

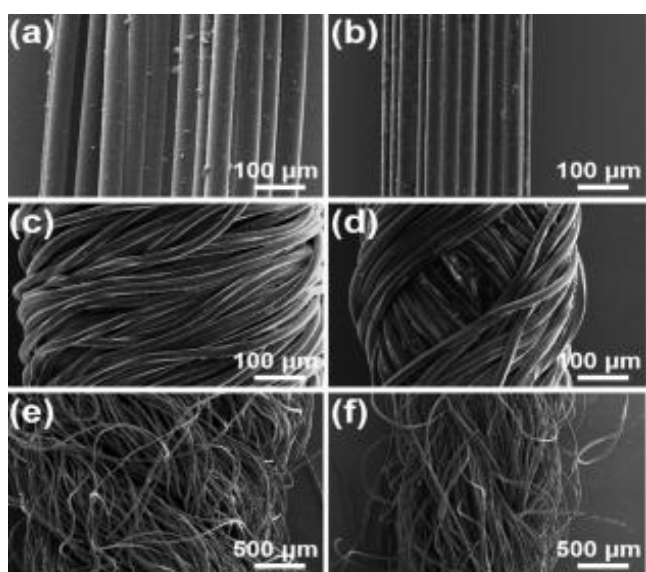


Figure 5. Different routes for graphene production images of the graphene strain sensors based on RY, NCRY, and WY, (a, c, e) without and (b, d, f) with strain (100%, 100%, and 50%, respectively) [52]

Memoon et al. [53] had fabricated a novel, highly flexible and piezoresistive strain sensor with a special 3D conductive network by 3D printing (FDM) technique. They used a filament extruder setup to synthesis a composite of conductive graphene pellets and flexible thermoplastic polyurethane (TPU) pellets. The size of the fabricated strain sensor was 2 x 1.5 cm (length x width) and the layer had a depth of 200 mm. They observed that the 3D printed strain sensor had a desirable combination of high sensitivity (GF of 11 with a strain of 10%, 80 with a strain of 100%) and good stretchability. Also, it showed a good durability and stability (stretch/release test of 6000 cycles) and a rapid response speed.

Chen et al. [54] fabricated a 0D graphene-based strain sensor through screen printing technique by combining 1D silver nanowire in a 2D layered graphene structure with 0D fullerene as a lubricant between layers. Two main interesting outputs were obtained from the work. The first observation was that the obtained 0D structure strain sensor shows insignificant hysteresis error at 0.8 mm s⁻¹ strain rate. The

second interesting result is that the sensitivity factor of the developed sensor reaches a gauge factor of 2392.9 corresponding to strains up to 62%.

Meshari Alsharari et al. [55] manufactured a 3D printed strain sensor by adding thermoplastic polyurethane (TPU) with different weight percentage to graphene based polylactic acid (PLA) to make different composites. The stretchability was enhanced by up to four times, while maintaining stable sensitivity of the strain sensors. Also, the cyclic loading and unloading of the printed samples showed good sensing stability and recoverability.

Xinda Li et al. [56] have proposed a fabrication of strain sensor based on FDM 3D printing of a commercial rubber/poly (vinyl alcohol) (rubber/ PVA) blend filament and RGO-GNF as a coating. The sensitivity factor values reached 13 at 40% strain. This value is higher than that of the metal strain sensor (GF=2).

McGhee et al. [57] compared between carbon dispersed polylactic acid (PLA), carbon dispersed acrylonitrile butadiene styrene (ABS), and graphene dispersed PLA. They found that increasing the thickness of the sensor leads to a decrease in reliability of measurements for carbon dispersed plastics. All three plastics give stable measurements up to 0.8mm layer thickness.

Zhihui Zeng et al. [58] used unidirectional freeze drying method as a novel technique to fabricate a nanocomposite strain sensor based on embedding RGO foam in polydimethylsiloxane (PDMS) followed by simple mechanical compression. They observed that the transverse directional sensing strain can reach as high as 122% producing a gauge factor of about 7.2. However, the longitudinal direction possessed a gauge factor from 12.2 to 27.0 at strain up to 57%.

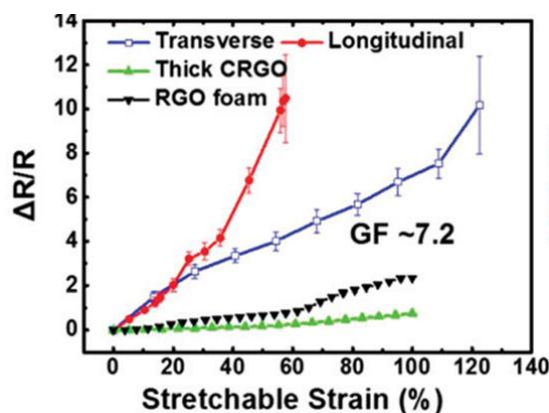


Figure 6. The relative resistance change ratio versus strain curves of various strain sensors [58]

T. Maturos et al. [59] prepared a composite material from 3D CVD-grown graphene foam (GF) and PDMS using dip coating technique. They concluded that the flexibility of the obtained composite is related proportionally with curing agent ratio. Also, they found that the optimum PDMS: curing ratio is 10:1 which produced the highest gauge factor (3.7).

Rujing Zhang *et al.* [60] used a novel technique based on bubble templating and fixation by freezing to fabricate 3D bubble-derived graphene foams (BGFs). They added BGFs to PDMS and by vacuum filtration they obtained a BGFs/PDMS composite. The GF was calculated as about 1.6 within 10% and ≈ 60 within 30 – 50%.

3.2. Energy Harvesting Applications

Recently, ferroelectric materials have attracted enormous attention because of their application in energy harvesting owing to its piezoelectric nature. In the decade before this, energy harvesting from piezoelectric materials had been largely based on bulk materials and have usually been of cantilever type, which requires special designs to operate at lower frequencies (in a frequency range < 100 Hz) [61,62].

In the same track of research, Wang and Song [63] introduced a nanogenerator, to represent the energy harvesting devices which convert mechanical energy into electrical energy from Nano-piezoelectric materials utilizing low frequency mechanical vibrations available in ambient atmosphere and human body motions. In general, the inorganic materials such as lead zirconium titanate, barium titanate, and so on have higher piezoelectric coefficients but are brittle, whereas the polymer ferroelectrics, poly(vinylidene fluoride-trifluoroethylene) [PVDF-TrFE], have lower piezoelectric coefficients but are flexible and can endure higher strains compared to inorganic types [64,65].

Venkateswarlu Bhavanasi *et al.* [66] investigated energy harvesting behavior of flexible PVDF-TrFE bilayer films with graphene oxide synthesized using drop casting technique followed by vacuum drying. They had chosen PVDF-TrFE due to its high piezoelectric coefficients among polymer ferroelectrics and graphene oxide as GO layers acquire negative charges owing to the existence of functional groups. Utilizing the charge on GO, an electrostatic nanogenerator has been fabricated to retain and convert the energy taken from mechanical vibrations. They observed that the adhesion of the GO films with the non-poled PVDF-TrFE films is poor due to the hydrophobicity and chemical inertness of PVDF-TrFE. In Addition, they observed that the obtained film shows outstanding energy harvesting leverage with power output of about $4.41 \mu\text{Wcm}^{-2}$ and a voltage output of about 4 V compared to poled PVDF-TrFE films alone (power output of $1.77 \mu\text{Wcm}^{-2}$ and voltage output of 1.9 V) owing to the improved elastic modulus of the bilayer films, the effect of electrostatic contribution from GO layers, and residual tensile stress.

Sanghoon Park *et al.* [67] fabricated a PVDF film based piezoelectric generator with graphene electrodes on both sides of the PVDF film. The film was placed vertically between graphene sheets from both sides after dipping it into the water. After that they compared energy harvesting performance of the film with PVDF film with metal electrodes by fixing the film in a bar frame which was weighed about 70 g and had additional hung weights to apply longitudinal tensile stress. The samples were

acoustically excited using a loudspeaker by tuning the excitation frequency between 100 Hz to 300 Hz and the excitation signals amplitude remained constant at 5 V. A decibel meter was used to measure loudness (105 to 110 dB) at 10 cm distance. They observed that devices based on metallic electrodes produced a higher output power as compared to graphene electrodes. Also, they concluded that the thin piezo-film was more effective in converting acoustic waves into electrical charge because free standing thin film is more flexible and more sensitive to the acoustic waves.

Manoj Kandpal *et al.* [68] reported a robust and a low-cost spin coating approach for fabrication of large area flexible piezoelectric nanocomposite generator device for harvesting bio-mechanical motions based on additions of ZnO and BaTiO₃ fillers on SU-8 matrix. They investigated the effect of adding graphene on these piezoelectric nanocomposites and they found that the average signal magnitude over the cycles was ~ 182 mV, ~ 592 mV without graphene for ZnO and BaTiO₃ respectively. Whereas, with the addition of graphene it spiked up significantly to ~ 385 mV, ~ 1.5 V for ZnO and BaTiO₃ films.

Liangke Wu *et al.* [69] fabricated piezoelectric nanocomposite films by adding reduced graphene oxide to poly(vinylidene fluoride) and they found that Compared to pristine poly(vinylidene fluoride) films, the open-circuit voltage, the density of harvested power of alternating current, and direct current of the poly(vinylidene fluoride)/reduced graphene oxide nanocomposite films improved by 105%, 153%, and 233%, respectively, indicating the suitability of this nanocomposite films for a broad range of applications.

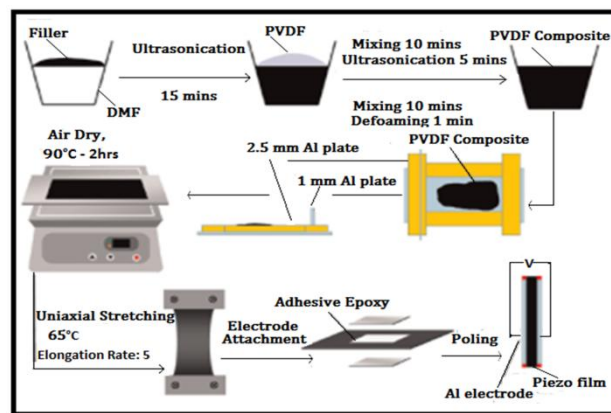


Figure 7. Fabrication process of PVDF/rGO nanocomposite films. PVDF: poly(vinylidene fluoride); rGO: reduced graphene oxide; DMF: dimethylformamide [69]

3.3. Graphene in Steel Industry

3.3.1. Steel Coating

Steels are iron alloys combined with carbon and other alloying elements of appreciable concentrations. These alloys are widely used in different industrial applications such as shells in automotive industry, supporting columns

and tableware [70]. Graphene is introduced as a coating material for different types of steels seeking improvement not only in corrosion resistance but also in the electrical properties to open the scientific research gates and provide strong chance for graphene to be used extensively in industrial applications in order to increase efficiency and quality of produced steels. Graphene exhibits superior impermeability compared to the most commonly used traditional anticorrosive coatings such as polymer coatings. Also, graphene is environmental-friendly, and offers green, alternative solutions to the dangerous coatings, such as chromium-based coatings [71].

To the best of authors' knowledge graphene possess a weak adhesion on metal substrate which affects its performance during harsh working conditions. Hence, researchers are seeking to manufacture a long-term graphene anticorrosive coating. Moreover, the growth of graphene on some metals and alloys (Fe, Mg, Al) is quite challenging due to poor catalytic activity [71]. These challenges could be avoided through using multi-layer graphene coatings (MLG) rather than SLG because in MLG the diffusion pathway of some of corrosion species is enhanced compared to SLG. Also, polymer-graphene hybrid composites could overcome some of challenges mentioned above as polymer enhances the adhesion between coating and metal surface without using very high temperature to avoid deterioration of mechanical properties. Moreover, polymer matrix acts as insulator that insulates graphene from metal surface to avoid galvanic corrosion. Also, the soft polymer helps graphene to coat rough surfaces [71].

Ludovic F. Dume   et al. [72] investigated the growth of 3D nano-flakes graphene on porous stainless steel substrate via CVD process and they observed that the graphene flakes act as a corrosion inhibitor by hindering the diffusion of corrosive agents towards sensitive surfaces. They also found that the pitting point is shifted by 50% suggesting that the graphene is acting primarily as an anodic barrier to ion diffusion towards the underlying metal surface thus decreasing the corrosion rate by reducing the kinetics of metallic ionization.

Jayanta Mondal et al. [73] developed a corrosion inhibiting composite coating composed of graphene oxide and polypyrrole for 304 stainless steel substrate. The samples were tested using G48A salt test analysis by immersing samples into 6% ferric chloride aqueous solution for six days. They observed that the coating was more stable more than 72 hrs and after 144 hrs the coating structure deteriorated in some places and a breakdown occurred. Also, from Tafel plot they found that E_{corr} of the GO/PP coating increases by nearly 6.3% compared to bare steel surface (-9 increases to 48 mV).

Tong Y. et al. [74] applied graphene and graphite onto cold rolled steel (Black Plate) as a conductive coating through EPD (Electro Phoresies Deposition) process. After investigation they found that the graphite coated steel had an electrical conductivity that was higher by 10 times

compared to that of the steel substrate. Also, the optimum thermal treatment parameters was found to be 580°C for almost five minutes and the best EPD parameters are 0.175 mg/ml iodine concentration, and 20 to 40V EPD voltage for 1-minute deposition time.

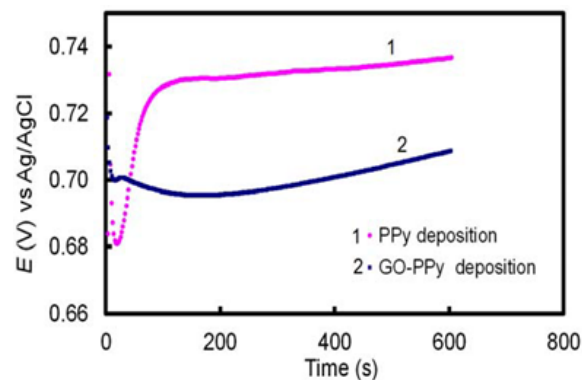


Figure 8. Galvanostatic electrochemical deposition of polypyrrole (curve 1) and graphene oxide-polypyrrole composite (curve 2) on AISI type 304 stainless steel substrates [73]

Abdulkareem Mohammed et al. [75] investigated the corrosion properties of RGO coated aluminium, copper, and stainless steel 316 at four temperatures (20 , 30 , 40 , and 50°C). They observed that all graphene coated samples showed better corrosion properties compared to the bare samples and they found that the best protection efficiencies were for SS316 type and their values reached 95% at 20°C and these values decreased with increasing temperature.

The impact of applying graphene to stainless steel surface layers using CVD was studied in [76]. It was observed that at high temperatures, the nickel barrier in steel reduces the formation of metal carbide and facilitates catalysation process of graphene precipitation. This solved the poor graphitization issue on bare SUS304 without buffer layer. In the same study, G/SUS304-900-4hr showed the worst corrosion resistance compared to other specimens. In contrast, the corrosion current of the G/SUS304-900-4hr is far away by higher ratios from other specimens. This is attributed to the inadequate graphene distribution on the surface. The graphene on the surface of the G/Ni/SUS304-900-4hr specimen acts as a barrier and slows the rate of oxidative corrosion; hence, no significant passivation was observed in the anodic polarization curve of this specimen. Another interesting outcome in [76] is that the contact resistance of the Ni/SUS304-900-4hr specimen only slightly increases after polarization testing by 20% .

Ali A. Naser et al. [77] used GO and GO derivatives such as GO-amine (GOA) and GOB as corrosion inhibitors for carbon steel alloy (C1025). They tested their specimens using electrochemical analysis by applying hydrochloric acid (0.1M) and they observed that GO, GOA, GOB efficiencies were 77.4% , 91.7% , 87% , respectively, under 100 ppm concentration. They also investigated the effect of temperature on the corrosion reaction and they observed that the increase in temperature decreased the efficiencies.

3.3.2. Steel Welding

Recently, graphene has been used as a reinforcement material during the welding processes [78-81], due to its high electrical and thermal conductivity, excellent tribological behavior [82], and low thermal expansion coefficient.

Hadi Jafarlou *et al.* [83] used graphene as reinforcement in welding low carbon steel joints via SMAW technique and they observed that the yield strength was improved by 3–45% and the ultimate tensile strength was enhanced by 5–22%. They also found that the enhancement in mechanical properties in terms of strength and hardness values was attributed to the refinement of ferrite grains made by graphene additions.

Tanmoy Das *et al.* [84] added graphene on AISI-1008 steel surface by drop-coating method and the coated steel was joined with aluminum plates (Al-1100 alloy) via resistance spot welding technique. They observed up to 124% enhancement in strength values. Also they concluded that the enhancement in hardness values was attributed to the formation of intermetallic compounds (IMCs) which couldn't be avoided by graphene additions and these IMCs could bear shear stresses only.

In an attempt to improve the mechanical properties of mild steel welded joints, the influence of adding graphene oxide and reduced graphene oxide were tested during flux cored arc welding process [85]. It was found that 10mg/ml of RGO enhanced the mechanical properties in terms of hardness and tensile strength by about 38.4% and 25% respectively due to the refinement of ferrite grains.

3.3.3. Lubrication of Steels

It was found that one third of the total energy consumption throughout the world lost due to poor friction and wear properties of material and the most common failure mechanisms of the 80% of the fabricated parts were wear and tear [86,87]. It was also demonstrated that the friction and wear have applications not only in operation of industrial machines and part life, but also in biological and human ones [88,89].

In order to enhance the tribological behavior and reduce energy losses of any application, a gaseous, liquid or solid lubricant should be used as the lubricant role is to reduce the metal to metal contact by forming a low shear boundary film between rubbing surfaces to increase the durability of the part [90].

Graphene has an ultrathin structure (about 0.34nm thick) which makes it the best candidate in some nano and micro scale systems such as electromechanical systems to decrease friction, and wear between rotating and sliding parts [90]. Several studies are conducted concerning the graphene's frictional mechanism based on some of the computational methods at different scales ranging from nano up to macro [91-96], as shown in figure (9).

Graphene has recently shown superior tribological behavior not only in nano scale but also in micro scale compared to bulk graphite [98]. However, there are a limited

number of publications concerning the tribological characteristics of graphene as a coating at macro scale [90]. This may be due to the poor adhesion of graphene as a coating to most of the engineering materials surfaces. Also, it has become a challenge to obtain a continuous graphene coating on a large area [99]. To address this issue, coupling agents such as (3-Aminopropyl) triethoxysilane have been used to improve the adhesion between graphene and silicon wafer [100].

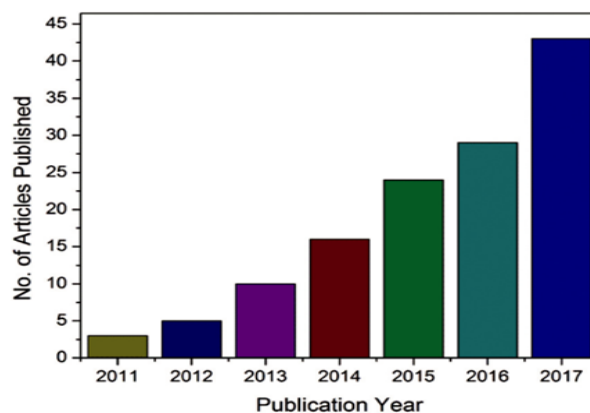


Figure 9. Number of research articles published in literature on the topic “graphene nanolubricants” [97]

Qi, Shaojun *et al.* [101] used aminosilan to modify a stainless steel substrate prior to add a GO coating. They found that aminosilan as an intermediate layer improved the tribological properties of the GO coating by reducing friction to a very low value (0.1), increasing the durability by about 20 times, and reducing wear by 10-folds, as shown in figure (10).

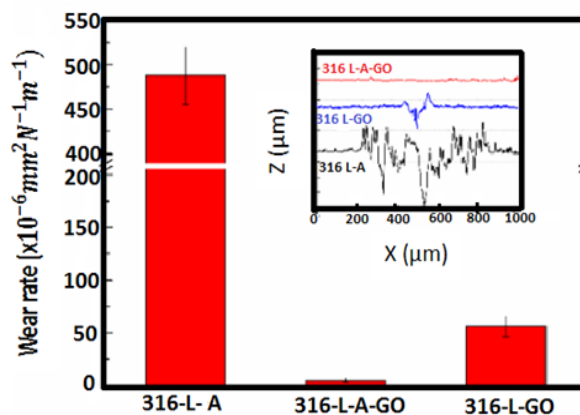


Figure 10. A comparison between the wear rates and the corresponding wear tracks (inset) [101]

Berman *et al.* [102, 103] used multi-layer graphene flakes as a solid lubricant for stainless steel and they observed a reduction in coefficient of friction by 6 times while using a low concentration of graphene. They also showed that the wear rate decreased by two orders of magnitude and the previous tribological behaviour was attributed to the ease of shearing between graphene layers within sliding contact interfaces.

In automotive industry, an improvement was made on the tribological behaviour on internal combustion engine cylinder liners by applying graphene coating to grey cast iron surface. A significant decrease in friction coefficient by up to 53% compared to uncoated samples was achieved and the wear resistance increased by up to 5 folds [104].

Recent attempts in metal cutting operations explored the ability of graphene to act as a cutting fluid additive in improving the tribological behaviour of cutting tool life made from alloy steel [97]. Yi et al. [105] used graphene oxide as a dispersed phase in a coolant used for drilling Ti alloys and it was found that the GO phase succeeded to decrease drilling forces and reduce flank wear occurred on the cutting tool during the cutting operation.

D.K. Das et al. [106] have conducted a comparison between TiN and graphene as a coating for cutting tool tips. They observed a huge percentage reduction of cutting force and friction force by up to 64% and 98% respectively.

4. Conclusions

Graphene is a promising material that reached high apex in energy storage applications. Moreover, it is used in strain sensors as it enabled considerable high gauge factors and sensitivity. Recently, graphene is introduced as a coating material for different types of steels seeking improvement not only in corrosion resistance but also in the electrical properties which provides strong chance to increase efficiency and quality of produced steels. Another track of research investigated the use of graphene as a reinforcement material through addition to flux during the welding processes to enhance mechanical properties of steel. Considerable interest is currently given by researchers to the efficiency of graphene as an additive to lubricants to enhance tribological properties in different applications such as metal cutting and lubrication of internal combustion chamber liners.

Future work should solve the following points:

- The inadequate dispersion or agglomeration of graphene which leads to inhomogeneous structure where agglomerated GNPs can act as stress concentrators. Moreover, this also impairs the efficiency of lubrication.
- The unclear strain sensor mechanism and this is because the difference in graphene film quality produced by different fabrication processes.
- CVD-grown graphene sensors are fabricated through an expensive and complicated process that is unsuitable for practical applications.
- Controlling the sensitivity of the sensor for measuring diverse human body motions remains challenging.
- The effect of graphene on steel weldments needs more investigations.
- There is a need to develop a correlation between wear volume and friction coefficient for graphene as a nano lubricant.

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