

## Review Article

# A Comprehensive Review of Graphene Nanocomposites: Research Status and Trends

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This paper provides a comprehensive review of the present trends in graphene research with an emphasis on graphene-based nanocomposites and their applications. Various synthesis routes have recently been devised for mass production of graphene to address the needs of the composite industry. This paper describes the worldwide scenario of research and patents being conducted in the field of graphene nanocomposites. It concludes with a discussion of the impact of graphene in composites and the future challenges to meeting industrial demands.

## 1. Introduction to Graphene

The single-layered atom-thick flatbed structure has revolutionized the nanotechnology platform since its discovery [1]. To date, several attempts have been made to synthesize graphene on a large scale to address the needs of various industries, particularly the composite industry, in which the use of graphene has dramatically transformed the global market for the production of state-of-the-art composite materials. The addition of graphene to a host matrix has achieved a number of enhanced properties with promising applications in many industries, such as aerospace, electronics, energy, structural and mechanical, environmental, medicine, and food and beverage. Since 2004, graphene has taken the nanotechnology platform by storm, with exponential growth in its applications. The remarkable properties of graphene make it a “magic bullet” for the composite world. Several papers on graphene and graphene-based nanocomposites have been published. According to Geim [2], graphene research has reached an unexpectedly great height and has emerged as a champion in the field of applied sciences. A simple search in Web of Science, Google Scholar, or Science-Direct yields several thousands of papers on graphene. Since 2000, there have been a total of 23,945 research papers published on various

synthesis methods and on isolation of graphene on a large scale. The numbers are still growing exponentially. An analysis using the Web of Science tool reveals that the majority of graphene publications come from countries of Asia, followed by Europe, the Americas (countries between Canada and Argentina), Australia, and Africa. Our record count analysis based on subject-wise publications found that the majority of the publications were published in the area of physics, followed by chemistry, materials science, technology-based topics, engineering, electrochemistry, polymer science, and many more.

According to the latest report released by the BBC, “Graphene: Technologies, Applications and Markets,” the graphene business is projected to boom up to \$67 million by the year 2015 and \$680 by 2020, with a compound average annual growth rate (CAGR) of 58.7% in a span of 5 years. Based on a similar report on graphene, “The World Market for Graphene to 2017” by Future Markets Inc., by the year 2017, the production volume of graphene is expected to grow to 573 tonnes from the initial growth of 28 tonnes in 2010 [3]. According to a recently published report by Sambasivudu and Yashwant [3], the annual growth of patents worldwide on graphene synthesis is becoming strong, and about 141 patents were published from 2001 to 2011. The report

also extensively analyzed the global distribution of patents (including patent cooperation treaty (PCT) and European patent organization (EPO) classified on the basis of graphene synthesis). Analysis revealed that the United States has filed over 135 patents through 2011, followed by the Republic of Korea (82), China (44), Japan (37), and Europe (26). An interesting fact to note in this analysis is that the majority of the patents have been filed through the PCT and the United States Patent and Trademark Office (USPTO). This clearly indicates the importance and commercial exploitation of graphene technology on a global scale. A report by Frazier et al. (2009) [4] described in detail the patents filed for graphene and its synthesis, publications, and applications in various patent offices worldwide. It also provided detailed data of the number of papers published with *graphene* as a central word, from 2000 to 2008. Numerous methods have been devised to produce graphene on a laboratory scale. The primary one is the “scotch-tape method” [1] used for isolation of pure defect-free graphene for research purposes, followed by other synthesis processes like exfoliation methods [1, 5–8], chemical vapor deposition (CVD) [5, 9–17], pyrolysis [18, 19], chemical synthesis [20–28], arc discharge [21, 29–33], unzipping of CNT [34–45], solvothermal [9, 21, 46–48], epitaxial growth [5], molecular beam epitaxy [10, 49–60], and electrically-assisted synthesis [61–63].

The present review presents the current research trends in graphene synthesis and graphene-based nanocomposites. It describes related publications, patent overview, synthesis, and applications as well as the several previous reviews that have been published about graphene and its composites [21, 64–66].

## 2. Synthesis Approach of Graphene

A long and coherent attempt of unsuccessful efforts to produce a single-layer graphene sheet has been made emphasizing the timely invention of a simple method presented by Novoselov’s group in 2004 [1, 67, 68]. The group repeatedly peeled a graphite crystal using an adhesive tape until a specific limit and later transferred the thinned out graphite onto an extremely thin (<300 nm) appropriately colored, oxidized silicon wafer [67]. This remarkable discovery led to the onset of mass-scale production of graphene and its utilization in various polymeric, electronic industries. Over the last forty years, various unsuccessful attempts have been made to achieve large-scale production of pure, defect-free graphene sheets [67]. Recently, the method of epitaxial growth on metal carbide, the CVD method, has shown promise for production of graphene [10, 67, 69]. Various methods have been devised and categorized into “top-down” and “bottom-up” processes. The following sections describe synthesis routes for graphene.

**2.1. Top Down.** Top-down approaches commence with an existing form of the bulk material and process it to create the final product. This approach may be cost efficient, depending on the material used. In general, it is limited to a lab scale and has limited quality control [4]. In this approach, graphene or altered graphene sheets are produced by either separation, peeling, cleaving, or exfoliation of graphite or

its derivatives (graphite oxide (GO) and graphite fluoride (GF)) [26]. Researchers have been successful in fabricating a few layers of free-standing graphene sheets on both micro- and nanoscales [4]. However, since this approach involves great investment and produces relatively low yields, the need remains for mass scaled-up processes to address the needs of industries economically. Various mechanical processes have been involved in producing high-quality, defect-free graphene: mechanical exfoliation of graphite, sonication, functionalization, electrochemical exfoliation, super acid dissolution of graphite, alkylation of graphene derivatives, chemical reduction of aqueous/organically treated graphene oxide (GO), thermal exfoliation, and chemical reduction of GO [26, 49]. A detailed account of synthesis of graphene by the exfoliation method, functionalization, and reduction along with its utilization in the fabrication of nanocomposites has been extensively reviewed by Potts et al. (2011) [70], providing thorough insight into the procedures followed by various authors. Similarly, Daniel et al (2012) [71] reviewed and extensively outlined the synthesis of graphene from various sources using several similar approaches. Several other papers are available online that review the synthesis of graphene using the top-down approach [34] in detail.

**2.2. Bottom Up.** The bottom-up approach consists of standard techniques such as epitaxial growth using metallic substrates by means of CVD [10, 50–54, 72, 73] or organic synthesis [23, 72, 74, 75], which depend on the choice of precursor chemicals and thermal degradation and decomposition of the SiC [19, 51, 53, 54, 73, 76]. Several other processes, such as arc discharge [29, 30, 77], chemical conversion [23, 27, 77, 78], CO reduction [79], CNT unzipping [35, 38, 80–82], and self-organization of surfactants [83–85] have also been tried for synthesis of graphene and its derivatives. Of all these processes, CVD and epitaxial growth, which produce bantam quantities of flawless graphene sheets with larger size [49], may in future be attractive for mass-scale graphene production, in contrast to mechanical cleaving. Using CVD and epitaxial methods, graphene sheets find their way into fundamental research with a multitude of applications ranging from electronics to polymeric nanocomposites [1]. Also, production of large quantities of graphene sheets is dependent on the chemical precursors used during synthesis. In particular, graphite oxide (GO), chemically reduced graphite (CRG), and thermally reduced graphite (TRG) are ideal candidates for polymer nanocomposite applications [1, 24, 26, 86–90]. In the bottom-up approach, as discussed earlier, the small molecule chemicals and catalysts are determining factors for the specific control of morphology, crystallinity, and structure of graphene [91]. There are several accounts of using hydrocarbons as the source of graphene production and using metal catalysts through the CVD process [91–94]. Currently, an Ni (111) surface is considered the best template for deposition of graphene due to the small variance in its lattice heterogeneity [91]. The control and stability in the graphene scale are potentially high, which makes CVD the most appealing method for device assembly and fabrication [91]. Nevertheless, this method faces a major

challenge in the control of edge structure and topology [91]. Epitaxial growth of graphene on a SiC substrate is another common technique, in which decomposition of SiC results in the formation of graphene layers. The silicon is desorbed off the surface leaving highly pure defect-free graphene sheets [51, 91, 95, 96]. This process has several advantages, such as (i) there is no transition or transportation of the resulting material from the metal substrate to the dielectric-type substrate; (ii) the resultant graphene film is free from impurities; and (iii) controlled initiation and growth of the product can be tailored through the correct choice of substrate [91]. Recently, self-assembly processes, such as layer-by-layer assembly (LLB), have been extensively employed to fabricate nanocomposite thin films using graphene. The resulting composite structure is expected to have well-aligned components [97–99]. Though the bottom-up approach to graphene synthesis presents less defects compared to the top-down approach, the operation and procedures are much harder, making it difficult to realize mass production for practical applications, and it is an expensive affair [72, 91]. Still, the most commonly chosen route of graphene synthesis is a bottom-up strategy because it offers incredible possibilities to tailor the atomic size, composition, shape, stability, and edge structure in graphenes [72]. Researchers around the globe are making strong efforts to develop a reliable strategy to produce defect-free, high functional quality and large quantity graphene using synthetic and processing protocols compatible with standard fabrication procedures at low cost [72, 91].

### 3. Worldwide Research on Graphene and Graphene Nanocomposites

Since its discovery, graphene has revolutionized and completely redefined modern day technology with its remarkable properties. The research has exponentially grown by numerous universities, R&D establishments, and many more private and governmental bodies around the world. Research on graphene can be found in every discipline, as of today. In the present section, looking into current prospects, we provide an overview of a number of published works, which illustrates the impetus in graphene research around the globe. The following sections briefly address the present scenario and the importance of graphene research. The several review articles published through 2010 highlight the quality and quantity of graphene research on the basis of publications with an aim towards patenting. Here, we attempt to update this trend and showcase the current growth of publications and patents.

**3.1. Quantity of Publications in Graphene Since 2000.** Using the Web of Science tool, a clear representation of the publication trend for graphene is shown, indicating the quantity of graphene research being carried out globally. Figure 1 shows the publication trend since the year 2000 (January) that the amount of research has grown exponentially from 106 articles in the year 2000 to 8,169 articles in the year 2012 (December).

A total of 23,945 articles were retrieved through this search using the syntax string <graphene>. Using the same

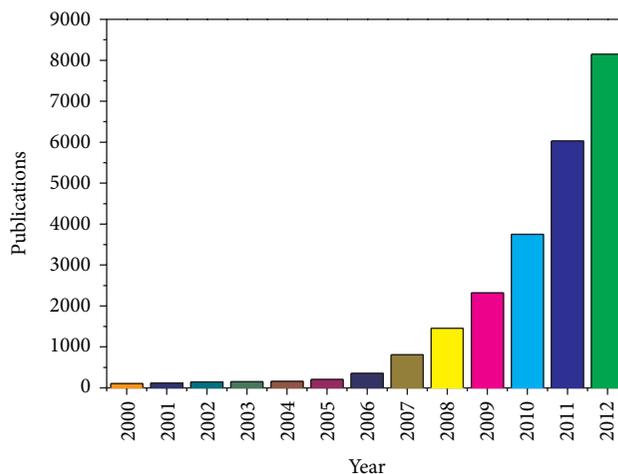


FIGURE 1: Publication trend in graphene chronology since 2000–2012.

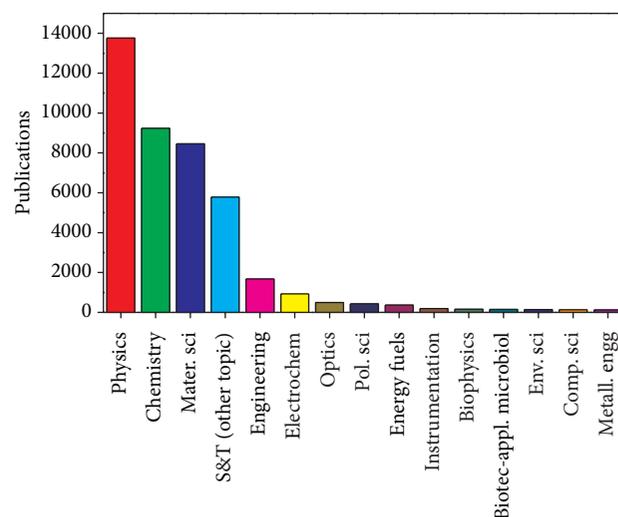


FIGURE 2: Top 15 research areas in which graphene is extensively used since 2000–2012.

tool (Web of Science), the results were further refined to assess the top research areas in which graphene is extensively used and the number of articles published since 2000.

As Figure 2 shows, physics (record count: 13,756) is the most often researched area, followed by chemistry (9,231), materials science (8,458), science and technology (5,779), engineering (1,677), electrochemistry (932), and so on. Research in the latter area is expected to grow at a rapid pace and in the future to compete with the forerunners. A country-wise refined search was conducted to assess the publication order in graphene research since 2000. Results (Figure 3) show that the United States tops the chart with the highest number of publications, 6,500, followed by China with 6,400, Japan with 1,979, South Korea with 1,648, and so on. This indicates the importance given to graphene research and the quantity of research around the globe. Considering the present scenario, it is estimated that by the year's end (2013),

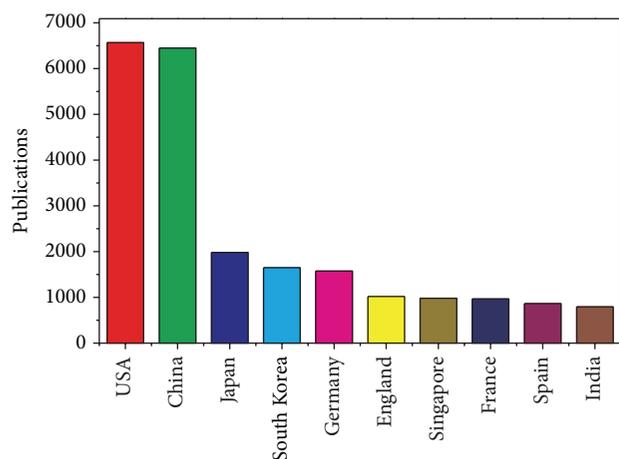


FIGURE 3: Country wise publications in graphene since 2000–2012.

competition will intensify among various countries for the highest position, quality, applications, patents, commercial products, and future markets of graphene.

**3.2. Patents on Graphene Since 2000.** Patents play a major role on the scientific and industrial platform. According to the data provided by Lv et al. (2011) [100], more than 50 countries have filed nearly 823 patents on graphene from January 2000 to December 2010, in 15 different disciplines. These data show that the top countries obtaining and applying for patents are the USA, followed by Asian nations. China, Japan, and South Korea contribute almost 25% of all the patents published in Asia. In the last ten years, Asian countries have recorded an explosive patent growth of almost 87% in the field of graphene [100]. Recently, Shapira et al. (2012) [101] used the Thompsons Reuters-Derwent Index to study graphene-based patents over the period 2000–2010. Their search found 4,787 graphene publications and 911 patents, with 97% of these patents being application oriented, indicating strong emergence of commercial applications. An increasing number of patents are regarded as indicators of breakthroughs in chronological performance [100].

In the present study, we used Thompson's Derwent Innovation Index to retrieve patent data for the strings <graphene> and <graphene nanocomposites> over the search time span from January 2000 to December 2012. The data retrieved on the total number of patents published on graphene during the years 2000–2012 was approximately 2,306 patents; thus, in a span of two years (2010–2012), compared to the results of Shapira et al. (2012), a strong jump occurred of nearly 1,395 patents. The results for the second search on graphene nanocomposites patents published during 2000–2012 retrieved only 80 records. Both searches showed that, during 2000–2007, there were no records of published patents on graphene and graphene-based nanocomposites. This can be attributed to the onset of trials to investigate the commercial importance of graphene in modern industry, with many researchers carrying out various trials to demonstrate high-end applications that

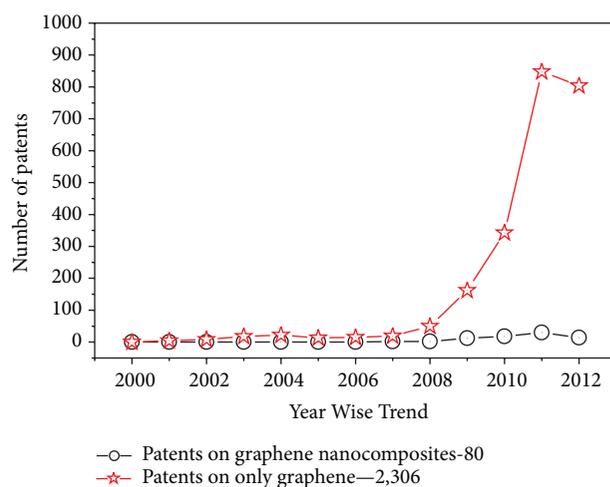


FIGURE 4: Patent trends of graphene and graphene nanocomposite since 2000–2012.

are likely to become available in the forthcoming years. We also conducted a cumulative search to see the trends and competition among the standalone graphene patents and graphene-based nanocomposites during January 2000–December 2012.

From the graph (Figure 4), it is evident that the future of R&D for graphene-based nanocomposites is very bright with high prospects. Since the average annual number of patents and publications for graphene is very high and increasing dramatically, it appears that investigations of graphene's properties are likely to focus in the near future on nanocomposites as the primary area of application. The growing interest in graphene in various disciplines is depicted in Figures 5 and 6. Analysis of the various patents on graphene in the time frame January 2000–December 2012 reveals that the most examined subject area is chemistry (Figure 5). This shows that most of the graphene research continues to be based on the principles and properties of its chemistry. These results are consistent with those shown in Figure 2. Analysis (Figure 6) of the patents for the year 2012 found that most of the patents published in this year were related to the engineering field. This shift in subject area from chemistry to engineering indicates that more application-based patents were published. A refined analysis shows that most of the applications were based on mechanical and structural designs, consistent with the use of nanocomposites in engineering applications. These results are consistent with the findings described at the beginning of this section. It is remarkable that, in the year 2012, 789 patents on graphene and 14 patents on graphene nanocomposites were published. A recent estimate shows that the over years (2004–till date) the funding in graphene-based composites has also increased several folds. It is estimated that the United States has spent more than \$760 million on graphene composite research. Also, more recently, a business week magazine reported that the European Union (EU) has invested around \$1.35 billion in graphene industry with an 18-month target to release its first graphene-based product in the market. Also, parallel

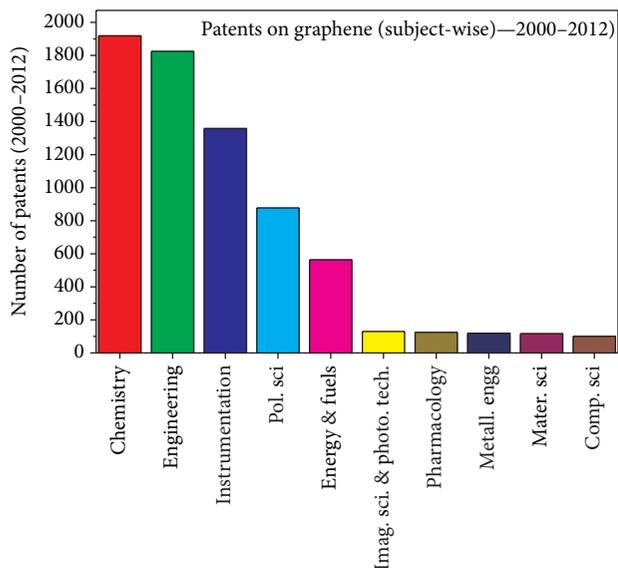


FIGURE 5: Subject-wise patent trend of graphene since 2000–2012.

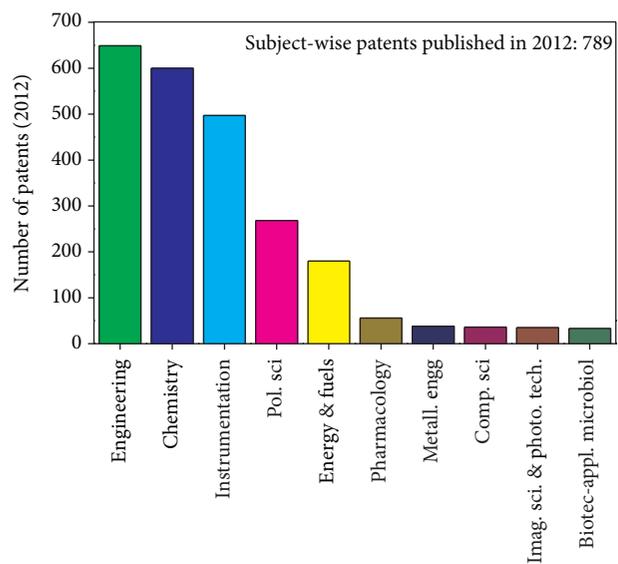


FIGURE 6: Subject-wise patent trend of graphene in 2012.

to this, research has shown increased citations in graphene nanocomposites over the years. Figure 7 shows the citation trend of graphene nanocomposite since 2000 till date with a total count of 22,500.

The increasing trend of citation shows the tremendous output in graphene nanocomposite research which may in future completely revolutionize and conquer the global market.

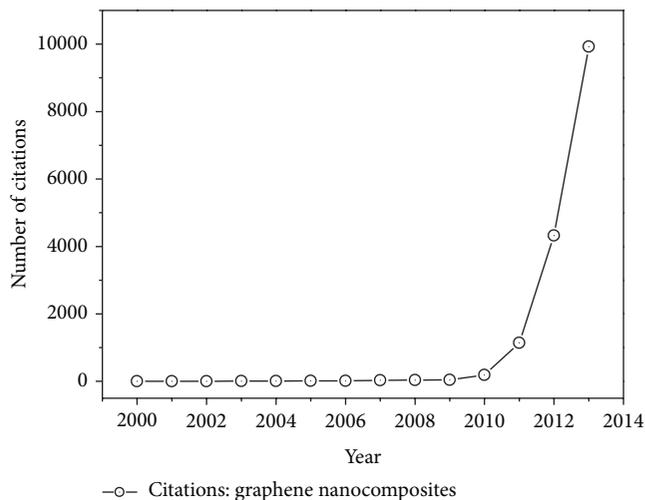


FIGURE 7: Citation trend of graphene nanocomposites since 2000–2013.

#### 4. Graphene-Based Nanocomposites and Their Importance

Because of graphene's exceptional thermal, mechanical, and electronic properties [70, 102, 103], it stands out as the most promising candidate to be a major filling agent for composite applications [70]. Graphene nanocomposites at very low loading show substantial enhancements in their multifunctional aspects, compared to conventional composites and their materials. This not only makes the material lighter with simple processing, but also makes it stronger for various multifunctional applications [70, 104, 105]. As stated in previous sections, the remarkable properties of graphene are able to improve the physicochemical qualities of the host matrix upon distribution. This helps in strengthening and increasing the interfacial bonds between the layers of graphene and the host matrix. It is this bonding that dictates the emergence of the cumulative properties of graphene in reinforced nanocomposites [106]. Kuilla et al. (2010), in their extensive review article on graphene-based polymer nanocomposites, have systematically explained the importance and use of graphene in various host materials. They also carried out a remarkable comparison of various nanofillers and listed their important applications in detail [106]. In the world of composites, theoretical analysis plays a crucial role in understanding their mechanisms, molecular interactions, and physical properties and their potential applications. A number of simulation tools provide cumulative or specific results for composite analysis. With the aid of these computational tools, a broad understanding and guide to successful research can be systematically achieved. Such theoretical investigations help the investigators to precisely optimize their case study to perfect the composite product [107]. In recent years, there has been some significant research papers on graphene-based composites with a polymer matrix. Jang and Zhamu (2008) reviewed the processing of graphene nanoplatelets (GNPs) for fabrication of composite materials [108]. Mack et al. (2005) prepared nanocomposites of

polyacrylonitrile (PAN) nanofibers strengthened by GNP, which they demonstrated to have improved mechanical qualities [109]. Research by Hansma et al. (2007) showed successful fabrication of graphene-based nanocomposites. They successfully optimized the amount and combination of adhesives and high-strength nanostructures (graphene) needed to yield a strong, low-density, lightweight, and damage resistant composite material [110]. Ramanathan et al. (2008) reported an unprecedented shift in the glass transition temperature ( $T_g$ ) of a polymer nanocomposite with functionalized graphene sheets. They observed that, by the addition of functionalized graphene sheets (1wt%) to the polyacrylonitrile (PAN), the  $T_g$  of the composite material increased by 40°C, whereas, when only 0.5 wt% was added to polymethyl methacrylate (PMMA), they observed a 30°C rise in  $T_g$  [111]. Also, they observed that by addition of 1wt% graphene to PMMA, an 80% increase resulted in the elastic modulus and a 20% increase resulted in the ultimate tensile strength. Their comparative research concluded that monolayered functionalized graphene serves as the best nanofiller among all examined nanofillers [111]. Das et al. (2009) employed the nanoindentation technique to the graphene-reinforced nanocomposite fabricated by using polyvinyl alcohol (PVA) and PMMM. The results showed significant improvement in crystallinity, elastic modulus, and hardness through the addition of only 0.06 wt% of graphene. The authors suggested that the enhancement was due to close mechanical interaction between the host (polymer) matrix and the layers of graphene. This interaction successively provides and dictates a better load transfer within the host matrix and the nanofiller [112]. Yu et al. (2007) identified that epoxy-based few-layer graphene nanocomposites show fascinating properties for the electronics industry, ideal for development of thermal-interface-based materials [113]. Zhang et al. (2009) and Liu et al. (2009) successfully synthesized graphene-fullerene-based hybrid nanocomposites [114, 115]. Booth et al. (2008) demonstrated the successful synthesis of robust 100  $\mu\text{m}$  thick macroscopic graphene membranes that can bear heavy loads [116]. According to Luechinger et al. (2008), the introduction of metal nanoparticles in the host matrix along with graphene has attracted a lot of researchers due to the advantage it provides by facilitating an improved interparticle contact (i.e., contact between the metal particle and the host matrix) [117]. Watcharotone et al. (2007) fabricated a transparent, electrical conductor by employing a simple sol-gel, spin-coating, chemical reduction, and thermal-curing route. The group used graphene oxide (GO) sheets mixed in the silica solution to obtain metal-encapsulated graphene nanocomposites [118]. In 2008, Chen et al. [119] fabricated graphene conducting paper that was electrically conducting, mechanically strong, and biocompatible. They uniformly dispersed graphene sheets in a solution using vacuum filtration followed by moderate thermal annealing. Recently, Cheng et al. (2013) synthesized carbon-coated SnO-graphene sheet composites in a green approach via a single-pot hydrothermal route. The composite was fabricated as an anode material for an Li-ion battery, and it exhibited high storage capacity and improved cyclic

performance [120]. Similarly, Perera et al. (2013) synthesized  $\text{V}_2\text{O}_5$  nanowire-graphene nanocomposite as an electrode material. The composite electrode exhibited an equilibrated electric double layer (EDL), energy density of 38.8  $\text{Wh kg}^{-1}$ , and pseudocapacitance at a power density of 455  $\text{W kg}^{-1}$ , and, also, the composite material showed a high specific capacitance of 80  $\text{F g}^{-1}$ . These results clearly indicate that the composite electrode was capable of effective storage and deliverance of charges toward high energy applications [121]. Lee et al. (2013) used cryomilling to synthesize fine particles of graphene and chitosan. The mixture was sonicated and layered to form a composite. The graphene particles conferred a cumulative effect in improving the mechanical attributes of the composite and also decreased the agglomeration quotient of graphene during mixing [122]. Guo et al. (2011) prepared a water-dispersed graphene-tryptophan-PVA nanocomposite for improving tensile strength, modulus, and thermal stability. There was a 23% increase in tensile strength when only a small loading of graphene (0.2 wt %) was introduced in the PVA matrix [123]. Ansari et al. (2013) studied the DC electrical conductivity retention of their indigenously prepared graphene-Pani-MWCNT nanocomposite in air and also assessed the cyclic aging. They found that Pani-graphene showed higher electrical conductivity and good stability for the DC electrical conductivity retention under isothermal conditions [124]. Jeon et al. (2012) prepared an exfoliated-grapheme- (EG-) cellulose acetate nanocomposite using the melt compounding method. They found that exfoliated graphene (EG) was uniformly dispersed in the host matrix with lower loadings. They also found that the composite had high thermal stability and improved conductivity and modulus [125]. The great number of application- and property-oriented possibilities suggests that future research and prospects for graphene-based nanocomposites are likely to expand tremendously in every discipline, with many surprises and products in store.

## 5. Applications of Graphene-Based Nanocomposites

Graphene has a great number of applications encompassing engineering, electronics, medicine, energy, industrial, household design, and many more [126, 127]. A previous review search yielded several review papers that examined field-oriented and specific applications of graphene. Majority of the papers dealt with electronic/sensor-oriented applications, to generalize the broad applications of graphene and graphene-based nanocomposite into their respective disciplines. Shen et al. (2012) extensively reviewed the biomedical applications of graphene including drug delivery, gene delivery, cancer therapy, biosensing and bioimaging, GO-based antibacterial materials, and scaffolds for tissue/cell culturing [128]. Similarly, Huang et al. (2011) and Choi et al. (2010) explained various phenomena associated with graphene and graphene-based materials and their applications in the field of memory devices for electronics, ranging from electrochemical sensors to instrumentation [127, 129].

**5.1. Biological Applications of Graphene and Graphene-Based Nanocomposites.** Graphene in various derivatives and in its precursor form has also shown potential applications in biological/medical fields, especially related to toxicity. Hu et al. (2010) demonstrated the antibacterial activity of two types of water dispersible graphene against *E. coli* with minimum cytotoxic effects on the human participants. The group concluded that GO paper can one day be effectively used in various environmental and biological applications [130]. Liao et al. (2011) demonstrated the cytotoxicity effect of graphene and graphene oxide (GO) materials under controlled physicochemical parameters. The results showed that GO was more severely hemolytic than graphene and showed high activity under extremely small size. They observed that, when chitosan was coated on GO, the hemolytic activity disappeared completely, showing the biocompatibility of the composite for erythrocytes. They concluded that the biological or toxicological responses of the material were dependent on the particle size, quality, and state, the surface charge, and the oxygen threshold [131]. Similarly, Liu et al. (2011) compared four different types of graphene materials (graphite (Gt), graphite oxide (GtO), GO, and reduced graphene oxide (rGO)) against *E. coli*, to study the toxicity effects. The membrane and oxidative stress signals were used to measure the intensity of toxicity. Their results showed that GO was the most severely toxic, followed in descending order by rGO, Gt, and GtO [132]. Santos et al. [133, 134] reported the design, fabrication, and antimicrobial application of a graphene-poly-N-vinyl carbazole (PVK) nanocomposite, resulting in more than 80% microbial inhibition and toxicity toward a broad array of bacteria. Carpio et al. (2012) studied the toxicity effects of PVK-GO nanocomposite on planktonic microbial cells, *E. coli*, *C. metallidurans*, *B. subtilis* and *R. opacus*, biofilms, and mammalian fibroblast cells (NIH 3T3). Their results showed that PVK-GO presented a stronger antimicrobial effect than pristine GO. They also found that the PVK-GO was significantly neutral toward the fibroblast cells, indicating a huge potential of the composite material in biomedical and industrial applications [135]. Peng et al. (2012) studied an Mn-ferrite ( $\text{MnFe}_2\text{O}_4$ )-decorated GO nanocomposite for biomedical applications. They observed that the magnetic property of the ferrites can be effectively used as an ideal hyperthermia and  $T_2$  contrast MRI agent. The nanocomposite when PEGylated showed excellent biocompatibility [136]. Recently, Liu et al. (2013) synthesized a hydroxyapatite-GO nanocomposite as biocompatible prosthetic. They found that the (300) and (002) plane hydroxyapatite nanorods in the graphene matrix played a crucial role in maintaining the composite's mechanical properties. Given its superior mechanical property, the authors suggested the nanocomposite's potential in composite and biomedical industries [136]. Many other applications of graphene nanocomposites in the field of electronics and other disciplines have been reported. Generalized applications of various kinds of graphene-based nanocomposites have been described in the literature. These include sensors [137–141], Li-Ion batteries [120, 121, 142–147], fuel cells [148–152], solar cells [153–158], field emission [159, 160], super capacitors [161–172], thermal transport and stability [173–175], packaging industry [176–178], corrosion

[179, 180], fire packaging and resistance [181], and many more. We expect that, very soon, all these applications will be available from manufacturers to end users at common commercial stores.

**5.2. Ceramic Reinforced Graphene Nanocomposites and Their Application.** The recent use of ceramics in graphene-based nanocomposite has sparked a global interest. The introduction of ceramic materials in few-layered graphene results in the formation of a composite yielding exceptional electrochemical performance with high charge carrier properties. The exploitation of such properties is a boon to the energy industry [182]. Several ceramic-graphene composites like SiC-Graphene [183],  $\text{Si}_3\text{N}_4$ -graphene [184, 185],  $\text{Al}_2\text{O}_3$ -graphene [186],  $\text{ZrB}_2$ -graphene [187],  $\text{ZrO-Al}_2\text{O}_3$ -graphene [188], BN-Graphene [189], and many more are known to enhance not only electrical properties but also thermal conductivity, refractory, mechanical, antifriction, anticorrosive and biocompatibility properties for diverse applications. Use of ceramics within graphene matrix can help overcome the brittle nature, lower fracture toughness, and limited thermal shock resistance in the composite industry. The use of  $\text{ZrB}_2$ -graphene [187] is presently known to be used in aerospace industry as a high temperature barrier for space vehicle during the reentry event. These materials (ultrahigh temperature ceramic composites) are consistently used as the primal infrastructure for the nose caps in space shuttles and military ballistic equipment. Several other ultrahigh temperature ceramic composites have shown promising results. A few ultrahigh temperature ceramic composites are known to exist, for example, carbides of Ta, Zr, Hf, Nb, and borides of Hf, Zr, and Ti, respectively [187]. Recently, Lahiri et al. (2013) have shown that with the introduction of short CNTs as reinforcement within the TaC ceramics, one can induce the formation of multilayers of graphene within the host matrix during the spark plasma sintering. This procedure helps in offering high resistance to pullout which results in higher strength material with delayed fracture [190]. Similarly, Pejaković et al. (2010) reported the synthesis of carbon rich-hafnia thin films using PLD technique. The NMR results showed that the sample contained graphene aromatically bonded carbon atoms presumably in graphene phase [191]. TiN-graphene composites, on other hand, have shown promising results as a selective permeable membrane for hydrogen. The composite material according to Kim and Hong (2012) was prepared by hot press process. The disc obtained was used to study the hydrogen gas permeability between 0.1 and 0.3 MPa and at 473, 573 and 673 K, respectively, using a Knudsen diffusion model. The results obtained showed that the hydrogen permeability of TiN-graphene composites was better than the Pd-Ag amorphous membrane at 1.67, 2.09, and  $2.83 \times 10^{-7}$  mol/msPa<sup>1/2</sup> at 673 K under 0.3 MPa, respectively [192]. Almost similar results ( $2.62 \times 10^{-7}$  mol/msPa<sup>1/2</sup> at 673 K under 0.3 MPa) of hydrogen permeation were obtained recently by Lee et al. (2013) with the use of  $\text{Al}_2\text{O}_3/\text{CeO}_2$ /graphene (ACG) composite membranes prepared by hot-press method. By exploiting the pore size distribution, surface area, and elasticity, one can

use such kinds of membranes for high purity separation and filtration of chemicals, biomolecules, petroleum products, and many more [193].

## 6. Conclusions and Outlook

Graphene- and polymers-based nanocomposites show promising growth in technology and applications [70]. However, a few key challenges must be addressed and resolved to realize the potential of graphene-based nanocomposites regarding synthesis methods, costs, and applications. For instance, if we consider the physical synthesis methods like sonication, exfoliation, and cleaving of GO, the resultant product (graphene) can have a reduced aspect ratio, which can drastically degenerate the reinforcement, bonding interactions, and thermal and electrical properties [86, 194] of both the graphene and the nanocomposites [70]. From the present review, it is clearly evident that both graphene and its derivatives have demonstrated their potential as promising candidates as reinforcements for high-performance nanocomposites [44]. Various reports have discussed the effects of the lower loadings of graphene and its derivatives, which result in high levels of strength and stiffness and superior mechanical properties [44]. Many reports have also discussed the good dispersion quality of graphene and its derivatives in different host matrices. As stated earlier, certain challenges remain to be resolved in obtaining large-scale, defect-free exfoliation of graphene with high quality and good properties. Since graphene tops the charts with its exceptional properties, the graphene-based product showcases itself in various day-to-day applications. Due to the quality and quantity of work, graphene has attracted worldwide attention in the mere five-year span since its emergence. The intensity and number of publications and patents arising from various countries and related subject areas have triggered a chain reaction in the field of electronics and other applications. It is expected that, by the year 2020, the graphene market will rise by a CAGR of 60%. This remarkable growth in the coming five years predicts an exponential boom in graphene research and development worldwide. In fact, polymer-based graphene nanocomposites have already paved their way in global markets and are expected to rise even more in the future [195]. Various attempts are being made to further improve and enhance the properties of nanocomposites by altering the chemical structure of graphene and its derivatives through functionalization and encapsulation techniques at the production level [196]. In the recent applications of graphene-based nanocomposites, some researchers have used graphene as the only nanofiller, which has shown improved dispersions and properties over other nanofillers. Recently, Yadav et al. (2013) introduced graphene oxide as a nanofiller for synthesis of a biodegradable polymer nanocomposite made of carboxy methyl cellulose (CMC). The results showed strong adherence of GO and CMC via hydrogen bonding. This interaction resulted in a commensurate improvement of mechanical properties with excellent dispersion of GO within the CMC matrix. The composite was reported to be more thermally stable and mechanically robust [197].

According to Segal (2009) [198], there has been a significant output in producing mass-scale production of graphene oxide (GO), and, similarly, platelets of graphene (GNP) have also gathered interest as nanofillers; they have also been in mass production since the onset of graphene-based nanocomposite [70]. Graphite is relatively affordable and is abundantly available around the globe [198], and it is the only chief source (precursor) for the production of graphene and graphene-derived nanofillers. This is due to the affinity of GO and its derivatives toward the host matrix, resulting in strong interaction and bonding. This has made graphene a promising nanofiller candidate, just next to carbon nanotubes, which it is likely to replace in the future. The present day composite industry is utilizing more and more of these GO nanofillers in their products on large and commercial scales. Use of ceramic reinforced graphene composites can be highly advantageous in the fields of chemical, biological, and petroleum products since ceramics-graphene composites are highly stable, robust, recyclable and chemically inert. Their use as thermal barriers and refractory applications paves their way into space science helping the engineers/scientist to understand the thermodynamics and stability of material during reentry events of space vehicles and military equipment or ballistics. Since most of the ceramic materials are biocompatible they can find their use in modern surgical equipment and as prosthetics in biomedical engineering area. Due to the high diversity, properties, and advantages of graphene, a multitude of nanocomposite-based applications have been envisioned to be practical. These multifunctional graphene composites coupled with affordable cost will soon be seen in the global market.

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