



Recent advances in graphene based polymer composites

Tapas Kuilla^a, Sambhu Bhadra^b, Dahu Yao^a, Nam Hoon Kim^c,
Saswata Bose^d, Joong Hee Lee^{a,d,*}

^a BIN Fusion Research Team, Department of Polymer & Nano Engineering, Chonbuk National University, Jeonju, Jeonbuk 561-756, Republic of Korea

^b School of Polymers and High Performance Materials, The University of Southern Mississippi, 118 College Drive #10076, Hattiesburg, MS 39406-0001, USA

^c Department of Hydrogen and Fuel Cell Engineering, Chonbuk National University, Jeonju, Jeonbuk 561-756, Republic of Korea

^d Department of BIN Fusion Technology, Chonbuk National University, Jeonju, Jeonbuk 561-756, Republic of Korea

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ABSTRACT

This paper reviews recent advances in the modification of graphene and the fabrication of graphene-based polymer nanocomposites. Recently, graphene has attracted both academic and industrial interest because it can produce a dramatic improvement in properties at very low filler content. The modification of graphene/graphene oxide and the utilization of these materials in the fabrication of nanocomposites with different polymer matrixes have been explored. Different organic polymers have been used to fabricate graphene filled polymer nanocomposites by a range of methods. In the case of modified graphene-based polymer nanocomposites, the percolation threshold can be achieved at a very lower filler loading. Herein, the structure, preparation and properties of polymer/graphene nanocomposites are discussed in general along with detailed examples drawn from the scientific literature.

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Abbreviations: AFM, atomic force microscopy; CNT, carbon nanotubes; CNF, carbon nanofiber; CCl₄, carbon tetrachloride; CTEs, coefficient of thermal expansions; CVD, chemical vapor deposition; S-CCG, chemically converted graphene; CHCl₃, chloroform; CMG, chemically modified graphene; Cryo-TEM, cryogenic transmission electron microscopy; DCM, dichloro methane; DMSO, dimethyl sulfoxide; DSC, differential scanning calorimetry; DMA, dynamic mechanical analysis; DMTA, dynamic mechanical thermal analysis; EELS, electron energy loss spectroscopy; EMI, electromagnetic interference; σ , electrical conductivity; ESEM, environmental scanning electron microscopy; xGnP, exfoliated graphite nanoplatelet; FLG, few layer graphene; GNS, graphene nanosheets; EVA, ethylene vinyl acetate; EG, expanded graphite; FG, foliated graphite; FT-IR, fourier transform infra red; GPa, gigapascal; T_g , glass transition temperature; GO, graphite oxide; HMPA, hexamethylphosphoramide; HDPE, high density polyethylene; IR, infra red; GNP^{PL}, ionic-liquid-functionalized graphene; KMG, KOH treated GO; LDH, layered double hydroxide; LM, light microscopy; LLDPE, linier low density polyethylene; e'' , loss factor; MPa, megapascal; MMA, methyl methacrylate; NMP, N-methylpyrrolidone; DMF, N,N-dimethylformamide; DMAc, N,N'-dimethylacetamide; ODA, octadecylamine; PETI, phenylethynyl-terminated polyimide; PANI, polyaniline; PPS, poly(phenylene sulfide); PNIPAAm, poly(N-isopropylacrylamide); PSS, poly(sodium 4-styrenesulfonate); PVA, poly(vinyl alcohol); PS, polystyrene; PMMA, poly(methyl methacrylate); PI, polyimide; PET, poly(ethylene terephthalate); PE-g-MA, polyethylene grafted maleic anhydride; PP, polypropylene; PVC, poly(vinyl chloride); PA6, polyamide; SIS, poly(styrene-*b*-isoprene-*b*-styrene); PS, polystyrene; PU, polyurethane; PVDF, poly(vinylidene fluoride); PEDOT, poly(3,4-ethyldioxythiophene); PB⁻, pyrenebutyrate; PBA, pyrene butyric acid; SEM, scanning electron microscopy; SiO₂, silica; SiC, silicon carbide; SWNTs, single wall carbon nanotubes; SDS, sodium dodecyl sulfate; SDBS, sodium dodecyl benzene sulfonate; E' or G', storage modulus; SSSP, solid-state shear pulverization; SPANI, sulfonated polyaniline; THF, tetrahydrofuran; TMEDA, tetramethylethylenediamine; TCNQ, 7,7,8,8-tetracyanoquinodimethane; TGA, thermogravimetric analysis; TPU, thermoplastic polyurethane; XRD, X-ray diffraction.

* Corresponding author at: BIN Fusion Research Team, Department of Polymer & Nano Engineering, Chonbuk National University, Jeonju, Jeonbuk 561-756, Republic of Korea. Tel.: +82 63 270 2342; fax: +82 63 270 2341.

E-mail address: jhl@chonbuk.ac.kr (J.H. Lee).

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1. Introduction

The field of nanoscience has blossomed over the last twenty years, and the importance for nanotechnology will increase as miniaturization becomes more important in areas, such as computing, sensors, biomedical and many other applications. Advancements in these disciplines depend largely on the ability to synthesize nanoparticles of various materials, sizes and shapes, as well as to assemble them efficiently into complex architectures [1]. Currently, nanomaterials have an enormous range of applications owing to their structural features. However, material scientists are examining materials with improved physicochemical properties that are dimensionally more suitable in the field of nanoscience and technology. In this regard, the discovery of graphene and graphene-based polymer nanocomposites is an important addition in the area of nanoscience, playing a key role in modern science and technology [2].

The discovery of polymer nanocomposites by the Toyota research group [3] has opened a new dimension in the field of materials science. In particular, the use of inorganic nanomaterials as fillers in the preparation of polymer/inorganic composites has attracted increasing interest owing to their unique properties and numerous potential applications in the automotive, aerospace, construction and electronic industries [4–11]. Thus far, the majority of research has focused on polymer nanocomposites based on layered materials of a natural origin, such as a montmorillonite type of layered silicate compounds or synthetic clay (layered double hydroxide) [5–17]. However, the electrical and thermal conductivity of clay minerals are quite poor [18–20]. In order to overcome these shortcomings, carbon-based nanofillers, such as carbon black, EG, CNT, and CNF have been introduced to the preparation of polymer nanocomposites [21–47].

Among these, CNTs have proven to be very effective as conductive fillers [22,33–39]. The only drawback of CNTs as a nanofiller is their higher production cost [48]. Therefore, the mass production of CNT based functional composite materials is very difficult. As Nicholas A. Kotov wrote in his review in *Nature* [49] “When carbon fibers just won’t do, but nanotubes are too expensive, where can a cost-conscious materials scientist go to find a practical conductive composite? The answer could lie with graphene sheets”. Graphene is considered a two-dimensional carbon nanofiller with a one-atom-thick planar sheet of sp^2 bonded carbon atoms that are densely packed in a honeycomb crystal lattice. It is regarded as the “thinnest material in the universe” with tremendous application potential [50,51]. Graphene is predicted to have remarkable properties, such as high thermal conductivity, superior mechanical properties and excellent electronic transport properties [52–56]. These intrinsic properties of graphene have generated enormous interest for its possible implementation in a myriad of devices [57]. These include future generations of high speed and radio frequency logic devices, thermally and electrically conducting reinforced nanocomposites, ultra-thin carbon films, electronic circuits, sensors, and transparent and flexible electrodes for displays and solar cells [57–71]. Graphene, as a nanofiller, may be preferred over other conventional nanofillers (Na-MMT, LDH, CNT, CNF, EG, etc.) owing to high surface area, aspect ratio, tensile strength (TS), thermal conductivity and electrical conductivity, EMI shielding ability, flexibility, transparency, and low CTE [52–56,72]. Table 1 gives a comparative chart on the mechanical, thermal and electrical properties of graphene with CNT, steel, plastic, rubber and fiber. The tensile strength of graphene is similar or slightly higher than CNT, but much higher than steel, Kevlar, HDPE and natural rubber. The thermal conductivity of graphene is higher than

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