Emerging Analysis on the Preparation and Application of Graphene by Bibliometry

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Abstract

Electroplastic graphene has been recently attracting considerable interest due to its intrinsic electrical, mechanical and thermal properties. This review has been presented the recent progress in preparation, application and bibliometric data of graphene published over twenty-eight years from 1986 to 2014. Recent researches clearly confirmed that graphene is one of very useful promising materials with applications ranging from electronics to biomedical systems such as biosensors, electrodes, and electromagnetic interferences. In addition to graphene, this article has been also introduced the synergistic effects of hybrid graphene-polymer on the properties of its composites. The emerging analyses of graphenes and their nano-composites have been finally discussed and proposed for the advances of electronics and biomaterials science and technologies.

Keywords: Emerging technology; Graphene; Nanocomposites; Electroplastic; Biosensor; Bibliometric

Introduction

Electroplastic graphene is attracting much consideration owing to its intrinsic mechanical, thermal and electrical properties such as band gap, mobility, quantum electronic transport, etc. [1-7]. Graphene is also a comparable non-cytotoxic and biocompatible nanomaterial which serves as a powerful platform for cell growth, differentiation and fate conversion [8]. The 2-dimensional nanomaterial of graphene has been recently investigated as one of emerging materials for biomaterials and electronic materials [9]. The chemically stable graphene also exhibits excellent optoelectronic and thermal properties [10]. The unique feature of the graphene nanomaterial has been exploited in tissue engineering and regenerative medicine as a scaffold of biological tissues. Graphene-based substrates can be supported neuronal differentiation of stem cells and hence may be potentially emerged into nerve regeneration. In the present paper, recent advances and progress have been reviewed on the nanomaterials of graphene by bibliometry. The emerging analysis on the preparation and application of graphene has been accomplished from the bibliometric data of 48,608 articles published from 1986 to 2014 in various aspects.

Review on Graphenes

The journal articles for bibliometric analyses were collected using search query made of keywords using WoS (Web of Science) database provided by Thomson Scientific (Philadelphia, PA, USA). The used search query is shown in Table 1. The dataset contains SCI and SCI-Expanded (SCIE) with a document type limited to the ‘article’ reflecting the accurate R&D trends and the search period was set to 1986 ~ 2014. The basic analyses were carried out using COMPAS (COMPetitive Analysis System) developed by KISTI (Korea Institute of Science and Technology Information) and VantagePoint* provided by Search Technology, Inc. Figure 1 shows the number of publications in the year of 1986 ~ 2014. Since several articles were formally published on graphene since 1986, the number of annual journal articles on graphenes increases continuously from 1991 to 2014. Finally in 2014, the accumulative number of publications reached to 48,608. One can see the interesting growth level of the graphene sciences and technology by curve fitting to a suitable model. The accumulative data are able to interpret by a logistic formula (1) written as follows [11-13].

\[
y = \frac{L}{1 + e^{-\beta t}}
\]

where L is the upper limit of technology growth, t means time, and the coefficients α and β are the parameters which determine the curve shape of the growth. We can estimate the year of growth reflection point to present by the fitting curve as shown in the inset of Figure 1, which means the current research trends on graphene and its composites in the midst of active time.

In order to observe the progress of graphene related research by the investigators in many countries on the world, we have extracted the countries information from the addresses of authors-affiliation. The researches on graphene and its composites have been performed by 115 countries from 1986 to 2014. Among the countries, China recorded 18,333 articles published to show 37.72% of total 48,608 articles. USA is shown 10,583 (21.77%), South Korea 4,115 articles (8.47%) recorded as shown in Figure 2. The figure shows a share of the number of publications of top-ten countries which take a share of about 98.85% of the total publications. Japan ranks the fourth with 3,127 articles (6.43%). Germany (2,735), India (2,115), Singapore (1,880), UK (1,867), France (1,562), and Spain (1,485) are come along after South Korea and Japan. For the whole countries, the number of articles increases annually as shown in Figure 3. The figure shows gradually the increase of published articles related to graphene and its nanocomposites on the journals from 1986 to 2014 in several countries.

Preparation and Properties of Graphenes

The pioneering research on graphene was conducted by Geim...
and Novoselov who received 2010 Nobel Prize in Physics [14]. First patent on the production of graphene was filed as US Patent 7071258 in October, 2002 and granted in 2006 [15]. Geim and Novoselov extracted a single-atom-thick crystallite from bulk graphites [4]. The first observation of anomalous quantum Hall effect in graphene provided directly to show the evidence of Berry’s massless Dirac fermions phase for graphene [3,16-18]. The SSA (specific surface area) of graphene is theoretically about 2630 m$^2$/g. The graphene is an allotrope of carbon with 2-dimensional crystalline structures. Each four carbon atoms, which are about 1.42 Å apart, have four bonds including one sigma ($\sigma$) bond with each of their three neighbors and one pi ($\pi$) bond that is oriented out of the plane [19,20]. The hexagonal lattice of graphene may be regarded as two interleaving triangular lattices. In order to prepare the single- or multi-layer graphene, a solution-based process had demonstrated by chemical exfoliation [21-23]. The merit of the chemical exfoliation method is very simple solution-based process and can be produced graphene largely at low cost.

Another possible method of the preparation is the epitaxial growth process for the production of large-area and high-quality for graphene on silicon carbide (SiC) [24-26]. SiC wafers may be utilized as a substrate the electronic devices of graphenes. However, SiC is relatively expensive and may be limited in size. On the other hand, the thermal chemical vapor deposition (T-CVD) method has been utilized to prepare graphene from carbon sources such as CH$_4$, C$_2$H$_4$ and polymers [27,28]. The growth of graphene on polycrystalline nickel thin films was reported to show high up to 3,650 cm$^2$/V$^{-1}$s$^{-1}$ of electron carrier mobility [28,29]. By using the combination of T-CVD with plasma energy, plasma-assisted CVD methods has been applied to decompose hydrocarbon at lower temperature (<1,000 C) rather than high temperature (>1,000 C) in the method of T-CVD [29,30]. The mechanical and physical properties of grown graphene films on the basis of plasma-assisted CVD method are comparatively controlled well by varying the plasma power, growth time and temperature [31]. To reduce the defects produced during preparation, a polymer supporting layer is needed on the grown graphene films as following two methods: One is the wet-transfer method and the other is dry-transfer method. In the wet-transfer method, the spin-coated PMMA (polymethylmethacrylate) or PDMS (polydimethylsiloxane) has been used as a supporting and protecting layers [31]. In the dry-transfer method, the thermal releasing tape has been allowed for the large-area graphene transfer onto the rigid substrate and flexible polymers [32,33].

## Emerging Analysis on the Application of Graphenes

Recently, the emerging technologies are used to be detected by bibliometric techniques [13,34]. We applied the keyword mapping on this study for emerging analysis. The research period from 2000 to 2014 is divided into three parts. Figure 4 shows keyword maps of graphene in Period I (2000~2005) which is beginning time of the graphene research. The word ‘graphene’ just begins to appear since the extraction of graphene was achieved in 2004 [14]. As shown in Figure 5, The Period II (2006-2011) was the glory days of graphene when the research based on graphene was performed very actively in the world. During Period II, the physical properties are studied importantly, which is reflected in the keyword map. Also, new application research area has been formed and the word ‘biosensor’ has shown. Meanwhile, the numbers of publications on graphene reached its peak in 2010 and then steadily decreased after a few years. The numbers of publications seem to be well correlated with the growth of graphene research in the world. The publication numbers of graphene research are counted by the number of articles related to graphene published in journals each year. The fitting curves of accumulative number of articles are shown in Figure 1. Figure 2 shows the distribution of articles related to graphene and its composites published in each country.
in the Period III (2012-2014), the research scale is getting bigger and bigger, and the application area is more various shown in Figure 6. The remarkable keywords are ‘supercapacitor’, ‘fuel cell’, ‘biosensor’, ‘MoS2’, ‘metamaterial’, and so on.

Graphene-based biosensors for biomedical application

Graphene nano-composites (GN) are very interesting in applying to use electrochemical biosensors for biomedical application in new field. GN-metal nanoparticles show catalytic properties, which use suitable for acting as biosensors [34]. The conductivity measurement of the systems can be evaluated the effect on electron transfer between the active centers and electrodes such as biosensors. Graphene can be incorporated into nano-composites to couple its unique property in the other nanocomposites. The electrochemical effect of the GN-particles may be possible evaluated for sensitive and electroactive biomolecules, as different enzymes and biomolecules can be oxidized or/and reduced at different potentials. Graphenes are excellent conductors of electrical charge. The electron transfer occurs at the edges of the graphenes in their basal planes. The large surface area of graphene provides a large number of electroactive sites that enhance direct electron transfer and electrochemical biosensor [35,36]. In addition to biomedical application of graphene for biosensors, there are many strategies cases such as nano-sensors, nano-medicine, regenerative medicine of stem cell treatments and tissue engineering, robotic surgery, synthetic biology of genomics, virotherapy of oncolytic virus, etc.

Graphene-based electronics

The special properties of graphenes lead to make them promising candidates essential components for next generation electrodes, energy storage and conversion devices [37,38]. The electrical characteristics of graphene and its nanocomposites show very outstanding properties such as lower carrier concentration (10¹¹/cm²) and high field-effect mobility (~10⁴ cm²/V s) at room temperature, flexible and good mechanical properties. The novel electrical properties of graphenes make them graphene-based electronics due to massless Dirac fermions, Berry’s phase, ballistic transport, and the fast speeds [3,17]. Top-gated flexible and stretchable transistors can be built up in near future. There are the other emerging areas on electronic textile, flexible electronics, molecular electronics, spintronics, electronic nose, etc.

Graphene-based LEDs and OLEDs

Graphene is the most attractive substances for use as flexible LEDs. Graphenes are the potential to be applied to the variety of different LED components such as electrodes and active charge transport layers. To prepare highly efficient OLEDs by using a doped graphene multilayer, the graphene can be applied to the conducting composition of the polymers [39-41]. OLEDs with modified graphene have been applied by Han et al. for higher luminous efficiencies [42]. Using the enhancement of graphene’s electrical properties, the fabrication of flexible OLEDs while lighting electronics can be made on PET surface. It has been recently reported that the flexible light-emitting electrochemical cells are built up based on graphene as a cathode [40]. The foldable circuits on graphenes have been successfully prepared to create LED chips [43]. For displays, there are interested in emerging areas such as LPD, FLD, FED, Laser, OLED, QD-LED, SED, TPD, TPE, TDEL, TMOS, iMoD, etc.

Graphene-based batteries

One of successful graphene-based batteries is litium-ion battery (LIB). Graphene has been applied to make the flexible and stretchable energy storage device for the development of LIBs as soft and active electrode material. Graphene is a potential material for the electrode of LIBs which is theoretically its excellent electrical properties such as large specific surface area of 2,630 m²/g, and its good mechanical properties such as high Young’s modulus of about 1 TPa. The soft and flexible LIBs are very useful due to their density of high energy and rate capability of long-run cycle for electronics [44,45]. VACNTs (Vertically aligned carbon nano-tubes) grown onto graphene paper (GP) electrode shows the good performance of rate and electrochemical stability due to their structures of many transport paths [46]. The LIBs have excellent flexibility, long-term life-cycle performance, high capacity and rate [47]. In addition to graphene-based storage of batteries, there are emerging areas such as compressed air energy storage, grid energy

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**Figure 4:** Keyword map in Period I (2000~2005); the red color corresponds the highest density and the blue color corresponds the lowest density.
storage, molten salt batteries, nano-wire battery, lithium air battery, silicon air battery, thermal energy storage, smart grid, contactless energy transfer, etc.

**Graphene-based super-capacitors**

One of the more emerging and promising devices for energy storage is flexible and stretchable super-capacitors. Graphene can be applied to emerge future stretchable electronics due to their high density of power, and good strength of mechanical properties [48,49]. The graphene and its derivatives are very potential and emerging to electronics such as flexible electrodes, stretchable solid-state superconductors, and others. However, the values for specific capacitance, density of power and energy are still remained lower than expecting values because of the restacking of GO (Graphene oxides) sheets by van der Waals interactions among the individual sheets. It has been demonstrated that the good electrical conductivity (1,738 S/m) and high specific surface area (1,520 m²/g) of laser-scribed graphene (LSG) by using a standard Light Scribe DVD optical drive to reduce oxygen groups such as carbonyl and epoxy in the GO [50]. These LSG-based superconductors under high mechanical stress will be utilized on high power density, ultrahigh energy density, and long-term life cycle stability [50,51]. In addition to super-capacitors, the emerging areas of energy productions are airborne wind turbine, biofuels, artificial photosynthesis, carbon negative fuel, concentrated solar cell power, fusion power, home fuel cell, hydrogen economy, methanol economy, molten salt reactor, antenna, solar roadway, space-based solar power, ultra-capacitor, vortex engine, etc.

**Graphene-based energy conversion**

The conversion of solar energy and other mechanical energies to
electrical energies can be provided a wonderful renewably sustainable source of power on the world. One of the most emerging devices for overcoming the silicon photovoltaic technical problems may be recommended graphene as a prime candidate for improving charge transport and extraction from the systems. In advance, graphene and its derivatives, graphenes, can be applied to the systems either the active materials or the interfacial materials. OPVs used CVD-graphene films were demonstrated with a sheet resistance at 72% transparency ($2300\Omega/cm^2$) and minimal surface roughness ($\sim 0.9$ nm) [52]. The results achieved a power conversion efficiency of 1.18% to show the outstanding capability to operate under bending up to 138 degree, whereas, ITO-based OPVs failed to bend more than 60 degree. Nano-generator may be applied to use in body-implanted devices as a flexible stretchable graphene. Such flexible devices can be effectively charged by the movement of human body [53]. In addition to graphene-based energy conversion, ambient intelligence as internet of things and artificial intelligence as machine vision, semantic web, fingering recognition may be applied in near future.

Graphene-based materials for IT and communication

Memory for FRAM, MRAM, NRAM, PRAM, RAM, SONOS, Racetrack and GPGPU, optical computing, quantum cryptography, and three-dimensional integrated circuit can be applied to the systems as a graphene-based material for IT and communication. Manufacturing 3-D printing, claytronics, molecular assembler, utility fog, conductive polymers and high-temperature superconductive materials can be applied to the systems as graphene-based materials in near future.

Summary and Conclusion

The recent progress and development on the preparation and application of electroplastic graphene and its nano-composites have been reviewed with 48,608 articles collected from WoS database of Thomson Scientific related to graphene published over twenty-eight years from 1986 to 2014 by using a bibliometric technique. The preparation and application of graphene and its derivatives have been comprehensively analyzed on the basis of bibliometric data. Firstly, the trends and numbers of publications related to graphene have been pointed out each year from 1986 to 2014. Secondly, the distribution of articles related to graphene published in each year has been analyzed graphically to see the research trends on the numbers of publication in each country. Thirdly, the emerging analyses on the preparation and application of electroplastic graphene have been made in detail from biomedical applications to electronics for IT and communications. In conclusion, the graphene and its derivatives are very useful and promising for the flexibly stretchable materials of the devices in electronics and biomedical applications. There are also expecting a lot of items to develop for new applications analyzed by emerging techniques on the basis of bibliometric data. These promising electroplastic materials are many applicable emerging sources into new fascinating devices of stretchable-flexible systems for electronics and biomedical applications.

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