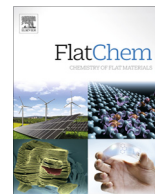




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Stretchable electronic devices using graphene and its hybrid nanostructures

Jihyun Paek, Joohee Kim, Byeong Wan An, Jihun Park, Sangyoon Ji, So-Yun Kim, Jiuk Jang, Youngjin Lee, Young-Geun Park, Eunjin Cho, Subin Jo, Seoyeong Ju, Woon Hyung Cheong, Jang-Ung Park^{*}

School of Materials Science and Engineering, Wearable Electronics Research Group, Ulsan National Institute of Science and Technology (UNIST), Ulsan 44919, Republic of Korea

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ABSTRACT

Research of flexible and stretchable electronic devices has shown remarkable development due to the remarkable characteristics of graphene. To exploit these characteristics, many devices have been developed using graphene-based hybrid materials or structures. In this paper, we present the electrical, optical, thermal, and mechanical properties of graphene and then provide information concerning the various electronic devices fabricated using graphene and its hybrid materials or structures, with special emphasis on stretchable devices.

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^{*} Corresponding author.

E-mail address: jangung@unist.ac.kr (J.-U. Park).

Introduction

Wearable devices have attracted significant attention because of their potential for use in future mobile applications, such as transistors, transparent displays, and photovoltaic sensors [1–7]. Most of these devices have semiconductor materials with structures that are specially designed for flexibility and stretchability, such as nanopillars, nanowires, and nanoribbons. However, other approaches have been attempted to fabricate these devices, including efforts to identify new materials with conductive or semiconductive characteristics that also have satisfactory mechanical characteristics, such as flexibility and stretchability. Recently, a thin, monolayer film of carbon, referred to as “graphene,” was developed by A. Geim and K. Novoselov. They prepared the graphene using the simple method of using cellophane tape to detach a monolayer of carbon from graphite and transfer it to a silicon substrate [8]. Currently, the method that is most often used to synthesize graphene is chemical vapor deposition (CVD) with a copper substrate [9]. First, carbon obtained from a mixture of CH_4 and H_2 can be directly grown on a copper film by surface-catalysed process at a temperature of about 1000 °C. Graphene produced by this method has superior properties especially on high Young's modulus, which makes it suitable for applications in stretchable devices. The technique used to transfer graphene to other substrates facilitates its use for various other applications. Therefore, many researchers have studied graphene-based, wearable electronic devices that have high stretchability.

This review reports the fundamental characteristics of graphene to illustrate the merits of using this material in various innovative devices, focusing especially on its use in stretchable devices. First, we briefly cover the electrical, optical, thermal, and mechanical properties of graphene. Then, we introduce various applications of graphene and its hybrid structures in stretchable devices, including transistors, light emitting diodes, energy devices, sensors, and others. In the last section, we provide a summary of the prospective uses of graphene in stretchable devices.

Properties of graphene and its hybrid structures

Graphene possesses considerable potential due to its outstanding electrical, optical, thermal, and mechanical properties, e.g., (a) high charge carrier mobility at room temperature ($15,000 \text{ cm}^2/\text{V}\cdot\text{s}$) [10]; (b) low light absorption of 2.3% in the visible region [11]; (c) high thermal conductivity (5000 W/mK) [12]; (d) high elastic modulus (1.02 TPa) and intrinsic strength (130 GPa) [13]; and (e) high specific surface area (calculated value, $2630 \text{ m}^2/\text{g}$) [14]. These properties offer promise for the use of graphene in a wide range of graphene-based, hybrid materials and nano devices (e.g., sensors, light-emitting diodes, and energy storage devices).

Electrical properties

The most special and useful properties of graphene are its high electrical conductivity and its mobility by massless charge carrier due to its special 2D hexagonal lattice [8,15]. In graphene, three of the four electrons in the outer shell of each carbon atom form sigma bonds in the 2D lattice, and the rest one of the four electrons (π electron) makes graphene conductive. There are two π -orbitals in the unit cell of graphene, which form bonding and anti-bonding orbital bands, and these two bands are bordered at the corners of the Brillouin zone, and they are referred to as Dirac points (K and K' in Fig. 1a). The structure of the bands near the Dirac points have linear dispersion relation, so charge carriers can move like massless, relativistic carriers (Dirac fermions) through the lattice with little scattering due to the unique zero-gap band structure, result-

ing in the half-integer quantized quantum Hall effect [16–18]. This property of zero bandgap also gives graphene an ambipolar electric field effect with mobility up to $15,000 \text{ cm}^2/\text{V}\cdot\text{s}$ at room temperature. In addition, suspended graphene (mechanically-exfoliated graphene) has an electron mobility of $250,000 \text{ cm}^2/\text{V}\cdot\text{s}$ at room temperature, the theoretical limit is reached due to minimized scattering [10]. As shown in Fig. 1b, the carrier density can be tuned continuously between holes and electrons by the electric field effect. This allows novel structures of devices in which the Fermi levels are changed dynamically by gating, which is not the case in conventional semiconductors such as silicon [11,19]. The ambipolar property also means that the adsorption of electron donating or withdrawing groups can tune the charge of the carriers. These superlative properties of conductivity and carrier mobility can be achieved by high-quality graphene that has few defects on the 2D crystal lattice, and this high-quality, pristine graphene can be obtained by the mechanical exfoliation method [20]. However, this method has clear limitations concerning large-area scalability and throughput. Also, graphene has an intrinsic zero bandgap, which hinders its ability to obtain a high on-off ratio. Thus, there have been many studies aimed at making it possible to use the superior electrical properties of graphene effectively by hybridizing graphene with other materials.

Due to graphene's high conductivity in the 2D plane, its optical transparency and mechanical robustness are used actively to produce stretchable conductors or electrodes by hybridizing the graphene with other materials, such as polymers, metal nanostructures, and oxides [21–27]. For example, as a stretchable conductor, graphene and carbon nanotube (CNT) networks have been integrated with poly(dimethylsiloxane) (PDMS). As shown in Fig. 1c, CNT networks grow vertically on graphene. After PDMS is infiltrated with the CNTs, the top ends of the CNTs are exposed for electrical contact. This CNT/graphene hybrid conductor has 45% stretchability that is reversible [21]. As transparent stretchable electrodes (TCEs), Park's group demonstrated a hybrid structure consisting of graphene/silver nanowires (AgNWs). The hybrid electrode has a sheet resistance of $33 \Omega/\text{sq}$, 94% transmittance in 550 nm, and 100% stretchability. Also, it has superb stability against electrical breakdown and oxidation when compared with graphene-only or AgNW-only electrodes [23]. In additional research, a graphene-metal nanotrough hybrid electrode was developed, and the graphene functioned as transparent, conductive fillers in the metal nanotrough's random network structures. This approach resulted in a low sheet resistance of $1 \Omega/\text{sq}$, a transmittance of 91% in 550 nm, stretchability of 80% in tensile strain, and high uniformity with a standard deviation of $\pm 0.1 \Omega/\text{sq}$ on the whole area, as shown in Fig. 1d [24]. Pei's group demonstrated a TCE using graphene oxide (GO) as a soldering material of the AgNW-AgNW junction. Due to graphene's highly conductive property, monatomic thickness, and mechanical flexibility, AgNW junctions held by graphene oxide can decrease its contact resistance and prevent sliding-induced failure (Fig. 1e). The AgNW-GO hybrid conductor had a sheet resistance of $14 \Omega/\text{sq}$, 88% transmittance at 550 nm, and stable resistance when subjected to repeated 40% tensile strain [26].

The high carrier mobility and ambipolar nature of graphene maximize its utility in field effect devices. Extensive research has been conducted to fabricate field effect devices with graphene, organic semiconductors, and hybrid structures for superior mechanical properties [28,29]. Also, due to the large and highly conductive surface area of graphene, the analyte molecules can be adsorbed effectively onto the surface of the graphene. Therefore, graphene hybridized with various nanomaterials can be used in many kinds of chemical sensors [30,31]. The details of additional applications of graphene's electrical properties using hybrid structures are presented in the 3. *Graphene-based Applications* section.

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