

# Nano-Graphene and Its Derivatives for Fabrication of Flexible Electronic Devices: A Quick Review

Wee Siang Koh<sup>1</sup>, Kiat Moon Lee<sup>1</sup>, Pey Yi Toh<sup>2</sup>, Swee Pin Yeap<sup>1,\*</sup>

<sup>1</sup>Department of Chemical & Petroleum Engineering, Faculty of Engineering, Technology & Built Environment, UCSI University, 56000, Cheras Kuala Lumpur, Malaysia

<sup>2</sup>Department of Petrochemical Engineering, Faculty of Engineering and Green Technology Universiti Tunku Abdul Rahman, Kampar, Perak, Malaysia

\*Corresponding author: E-mail: yeapsw@ucsiuniversity.edu.my

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## Abstract

Along with technology development, the demand for flexible, foldable, and portable electronic devices has grown over the past few years. Successful fabrication of these flexible electronic devices relies on the internal electronic components which are also flexible and lightweight. In this regard, researchers are now working on using nanomaterials which exhibit the desired electronic properties to replace the conventional electronic components. Graphene nanosheet and its derivatives are known for their intrinsic electrical behaviour. Meanwhile, they are lightweight and consume small space in any design. Hence, recent research has been focussing on fabricating flexible and foldable electronic components by attaching the graphene and its derivatives on a thin film/substrate. In fact, this idea has been realized in year 2017 on the first flexible OLED panel that uses transparent graphene-based electrode. In view of the positive impact of this nanomaterial towards future design of electronic devices, the present paper aims to provide a quick review on the current stage of research, the challenges encountered, as well as the future outlook in the use of graphene nanomaterials for designing flexible electronics. Copyright © VBRI Press.

**Keywords:** Graphene, nanotechnology, flexible, foldable, electronic devices.

## Introduction

Flexible electronics are specifically a class of electronic devices which can be bent, folded, twisted, stretched, or even deformed into arbitrary shapes but retain its electrical performances [1]. It is different from the conventional electronic devices which were built of integrated circuit planted on a rigid wafer. The problem with the conventional electronic components is that they are hardly incorporated into an irregular or soft moving object [2]. On the other hand, the current digital and electronic industries have been moving into fabrication of thin and flexible devices for both human and industrial daily usage. This includes the fabrication of E-skin in robotics, biosensing devices, smart-wearable human healthcare, environmental monitoring devices, energy conversion and storage devices, daily communication devices, and various display devices. In fact, along with the competitive growth in smart phones markets, the technologies of thin and flexible smart phones are of big interest in worldwide companies.

Nanotechnology plays an important role in the design of these flexible electronic devices. In particular, nanomaterials with intrinsic electronic properties are

employed to replace the conventional rigid electronic components. Graphene and its derivatives (namely graphene oxide (GO) and reduced graphene oxide (rGO)) are the nanomaterials that known for electrical characteristic. Graphene, a 2-dimensional nanomaterial, is regarded as the best conductor for electricity [3]. The conductive behaviour of graphene is ascribed to its chemical structure which consisted of many  $sp^2$  hybridized carbon atoms arranged in honeycomb hexagonal lattice. The  $\pi$ -electrons of the carbon atoms freely mobile within the crystal plane and thus make the graphene an excellent electrical conductor [4]. On the other hand, the GO is an insulator due to disruption of  $sp^2$  bonding networks by the oxygenated group. To restore the electrical conductivity and recover the honeycomb hexagonal lattice, the oxygenated group shall be removed via reduction method [5-7]. The derivative produced from this reduction is termed as rGO; nevertheless, the rGO still retains some of the oxygenated group and thus make it less conductive than the pristine graphene [8]. **Fig. 1** illustrates the chemical structure of graphene, GO, and rGO.

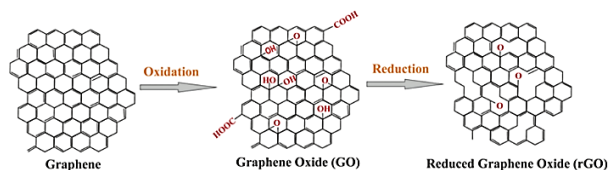


Fig. 1. Chemical structure of graphene, GO, and rGO [9].

Owing to the unique properties, graphene, GO, and rGO have been the centre of research study ranging from fundamental investigation on their physical and chemical properties, synthesis methods, mechanism simulation, application in wastewater treatment processes, application in biomedical field, up to fabrication of electronic devices and etc. **Table 1** highlights the several areas of studies on graphene and its derivatives over the past three years.

**Table 1.** Recent research studies on graphene and its derivatives.

Year	Area of studies	References
2019	Physical and chemical studies	10-18
	Synthesis methods improvement	19-22
	Water purification	23-24
	Morphology study	25-28
	Electronic applications	29-31
	Reaction kinetic theory and simulation	32-34
	Biomedical application	35-39
	Energy conversion	40-43
2018	Physical and chemical studies	44-47
	Biological study	48-51
	Synthesis methods improvements	52-57
	Water purification	58-64
	Morphology	65-67
	Biomaterial synthesis	68
	Reaction kinetic theory and simulation	69-73
	Electronic applications	74-77
	Architected printing	78
	Power generation	79-81
	Biomedical applications	82-88
	Photo-catalysis applications	89-97
	Air filtration	98
2017	Physical and chemical studies	99-100
	Biological studies	101-102
	Synthesis methods improvement	103-107
	Water purification	108-117
	Morphology study	118-121
	Reaction kinetic theory and simulation	122-125
	Electronic applications	126-132
	Biomedical application	133-134
	Energy conversion	135-136
	Photo-catalysis applications	137-139

### Studies on electronic properties of graphene and its derivatives

Many underlying electronic properties of graphene, GO, and rGO have been disclosed along with research studies. For instance, Kuc *et al.*, 2010 found that the electronic properties of graphene nanoflakes is dependent on their size and topology, whereby its Homo-Lumo gap decreased when the size of the flakes increased [140]. On the other hand, Özçakır *et al.*, 2016 reported that GO flakes of larger building blocks normally resulted in three-fold better mechanical and electrical properties [141].

While for rGO, its electrical conductivity may vary by the synthesis condition and/or by the starting material. Pavoskit *et al.* 2017 reported that rGO produced from graphite flakes exhibited better electrical conductivity than the one produced from an expanded graphite. This could be due to the many defects formed in the rGO produced from the expanded graphite. More interestingly, few layers of rGO was found to exhibit ten times superior in electrical conductivity than the starting graphite [142].

Phiri *et al.*, 2018 reported that the incorporation of graphene, GO, or rGO into a cellulosic material may alter the electrical and thermal properties of the material in different manners. The graphene and rGO-based composites showed highest conductivity value with  $1.66 \text{ S m}^{-1}$  and  $0.57 \text{ S m}^{-1}$  but as expected, GO-based composite is an insulator. However, in term of the thermal stability, it was found that the graphene-based composite is more stable compared to both GO and rGO-based composites. Such observation may ascribe the oxygen groups in GO and rGO that decompose when temperature increase; the weight loss is higher in GO due to higher oxygen moieties present [143].

### Substrates used for fabrication of flexible electronic film

The unique electronic characteristics of graphene and its derivatives have made them a good candidate for the fabrication of flexible electronic component. Basically, a flexible electronic component consists of a dielectric substrate, an electrical conductor, an adhesive material, as well as a protective layer. The substrate for electronic applications must be highly flexible (*i.e.* low Young's Modulus), high thermal stability, high thermal dimensional stability, low water vapour and oxygen transmission rate, and surface smoothness. The flexible substrates are categorized into three main types: polymer films (such as plastic), thin glass, and metal foils. Each material has its own pros and cons as listed in **Table 2**. Meanwhile, **Table 3** provides a summary of substrates used in fabricating graphene-based flexible electronic film.

**Table 2.** Comparison of plastic, thin glass, and metal-foil substrate [144].

Substrate	Advantages	Disadvantages
Polymer film (e.g. Plastic)	Rugged Bendable and Rollable Transparent	Poor dimension stability Low process temperature and chemical resistance, High H <sub>2</sub> O and O <sub>2</sub> permeation.
Thin glass	Conformable Transparent, Low H <sub>2</sub> O and O <sub>2</sub> permeation.	Low mechanical stability Low process temperature.
Metal foil	Rugged and Conformable Low H <sub>2</sub> O and O <sub>2</sub> permeation High process temperature. Good dimension stability	Opaque Rough surface Capacitive effect

**Table 3.** Summary of substrates used in fabricating graphene-based flexible electronic film.

Nanomaterials	Substrate	Reference
Graphene	Rubber, polyimide	145
	Polypropylene	146
	None	147
	Polyethylene terephthalate	148
	Polyimide	149
	Copper foil/ Poly (methyl methacrylate)	150
	Photographic paper AP27020A4	151
	Kapton foil with Quartz, Silicon and High resistivity Silicon.	152
	Polyethylene terephthalate	153
	Polyethylene terephthalate	154
	Polyimide	155
	Polyethylene terephthalate	156
Polyvinyl alcohol	157 - 159	
GO	Polyethylene terephthalate	160
	Polyethylene terephthalate	161
	Silicon	162
rGO	Silicon	163
	Paper coated with Polyvinyl alcohol	164
	Polyethylene terephthalate	165
	Indium tin oxide/ Polyethylene terephthalate	166
	Polydimethylsiloxane	167
	Polyvinyl alcohol	168
	Polypropylene	169
	Fluorine doped tin oxide Glass	170
Kapton sheets and Polyethylene terephthalate	171	

The coating of graphene or its derivatives onto the flexible substrate is commonly done via roll to roll printing methods (inkjet printing). Deposition, gravure, flexographic, flatbed, soft lithography, laser ablation technique are the typical types of roll to roll printing. The advantages of the roll to roll printing method are in-line hot drying, non-vacuum low temperature deposition, and high-volume continuous production [172-176].

### Challenges and future outlook in this research area

As prescribed, the flexible OLED launched recently is an indication of realization of graphene-based research study into real world application. It is envisaged that the future OLED display will comprise rollable touch screen with unparalleled resolution and contrast. However, the cost associated to the material/substrate, the stacking process, the barriers protection to oxygen and water (encapsulation process), as well as the solvent selection are the main challenges in OLED research and development. Meanwhile, researchers also concern on the thermal and chemical instability of certain substrates [177]. Apparently, more research studies are required to be done in this area.

Stretch ability is another requirement for flexible electronic devices. To achieve this objective, the

adhesion force between the conductive layer (nanomaterials) and the underlying substrate need to be enhanced. Unfortunately, the existing graphene sheets and other nanomaterials are weakly bound to the substrate via van der Waals forces [178]. The poor adhesion may subsequently lead to peeling-off of the nanomaterials during the folding or stretching process. Furthermore, such a poor adhesion will develop into cracks or localised rupture upon stretched. The crack network diminishes the electrical properties of the conductive layer [179] and subsequently led to material failure [180]. To cope with this challenge, research study shall venture into the development of chemical bonding to strongly bind these conductive nanomaterials onto the substrate. For instance, Guo *et al.*, (2015) introduced covalent bond between the Au nanoparticles and PDMS substrate [181]. The strength of chemical bonding is at least two order of magnitude stronger than van der Waals forces. Meanwhile, the authors have introduced a pre-strain process to suppress the localized rupture. The resultant flexible films were found to exhibit a better scratch resistance and durability. The same technique may be tested on graphene-based flexible electronics.

Graphene has been a wonder material and a perfect ingredient for the fabrication of flexible electronics. Nevertheless, it is not without limitation, for instance, graphene possesses with no intrinsic bandgap, huge energy barrier, as well as comparatively high sheet resistance [182]. These limitations have hindered its application in electronics industry. To solve this issue, recent studies have utilised doping technique to improve the electronic properties of graphene [27, 182].

More researches are to be done to further disclose and expand the electronic properties of graphene and its derivatives. For instance, introducing magnetization properties onto graphene and its derivatives (via chemical doping, ion irradiation, or by synthesizing graphene nanoribbons with perfect zigzag edge) may expand their applications to the spintronics field [183-185]. Meanwhile, Piao *et al.*, 2017 [186] reported that the amphiphilic GO sheet able to prevent electric short in the electrorheological measurement, thus, it is a suitable candidate for electrorheological applications. Nevertheless, an ideal flexible electrode shall have good resistance to the electrorheological fluid [187]. Hence, future work may venture into the impact of electrorheological fluid on the flexible materials.

While the usage of graphene and its derivatives have grown significantly over the past few decades, people are now concern about their toxicological impact. It was reported that the toxicity of graphenic materials may vary by their production methods and concentration [188, 189]. Wright *et al.*, 2018 reported that GO with high oxygen content contributed to oxidative stress, cytotoxicity, and pulmonary toxicity. In view of this, future study shall focus on the toxicological studies of graphene-based nanomaterials and ways to reduce it [190]. This piece of information is important especially for flexible electronics that are to be implanted in human bodies.

## Conclusion

Application of graphene and its derivatives have been a hot topic due to their tiny sizes and unique electronic properties. Despite being a promising candidate for the fabrication of flexible electronic devices, more research studies are needed, in particular, to boost the electronic properties of the graphene-based nanomaterials, to further improve the adhesion strength of these nanomaterials onto the flexible substrate, as well as to optimise the application performance. In addition, toxicological study is needed prior to large scale incorporation of this nanomaterial in human life.

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