

Graphene from waste and bioprecursors synthesis method and its application: A review

Nur Fatihah Tajul Arifin ^{a,b}, Norhaniza Yusof ^{a,b*}, Ahmad Fauzi Ismail ^{a,b}, Juhana Jaafar ^{a,b}, Farhana Aziz ^{a,b}, Wan Norhayati Wan Salleh ^{a,b}

^a Advanced Membrane Technology Centre (AMTEC), Universiti Teknologi Malaysia, Skudai 81310, Johor, Malaysia

^b School of Chemical and Energy Engineering (SCEE), Universiti Teknologi Malaysia, 81310 UTM Johor Bahru, Johor, Malaysia

* Corresponding author: norhaniza@petroleum.utm.my

Article history

Received 11 Mac 2019

Revised 26 June 2019

Accepted 31 January 2020

Published Online 15 June 2020

Abstract

Recently, carbonaceous material such as porous carbon, carbon nanotubes (CNTs), graphene, graphene oxide (GO) and activated carbon has received tremendous attention from researchers. To date, the exploration of graphene is still in vast. Graphene has been applied in various applications which include polymer composites, energy storage, fuel cell and biomedical applications. This is due to its unique characteristics such as large surface area and high remarkable electronic, mechanical and thermal properties. Even though chemical vapor deposition (CVD) has been established as an effective method to synthesize graphene, but the yield is low and may not compatible in certain applications. In addition, the chemical process of the production of graphene from exfoliation of graphite oxide involves hazardous and toxic reagents. Currently, bio-waste materials have been a great source for production of carbon. Furthermore, bio-waste materials are abundant and proper disposal method is needed. Hence, preparation of graphene from waste and biomass precursors is a new alternative to overcome the afore mentioned problem. Therefore, this paper will be focused on the method of synthesizing graphene from glucose, rice husk, chitosan, corn stalk core and plastic waste. The application of graphene derived from each bioprecursor for dye removal, adsorption of toxic and heavy metals, gas storage and supercapacitors will also be reviewed.

Keywords: Graphene, bio-waste material, chemical vapor deposition

© 2020 Penerbit UTM Press. All rights reserved

INTRODUCTION

Nowadays, carbonaceous materials like porous carbon, carbon nanotubes and graphene have gathered great interest owing to their high surface area and pore size, ease of functionalization and chemically stable structure (İlbay *et al.*, 2017). Generally, graphene has remarkable electronic, mechanical, and thermal properties (Somers, 2015). In addition, graphene also has large theoretical specific surface area ($2630 \text{ m}^2\text{g}^{-1}$), high intrinsic mobility ($200,000 \text{ cm}^2 \text{ v}^{-1} \text{ s}^{-1}$), high Young's modulus ($\sim 1.0 \text{ TPa}$) and thermal conductivity ($\sim 5000 \text{ W m}^{-1} \text{ K}^{-1}$), high optical transmittance ($\sim 97.7 \%$) and good electrical conductivity (Bhuyan *et al.*, 2016). Thus, it is applied in many applications which include polymer composites, energy-related materials, sensors, 'paper'-like materials, field-effect transistors (FET) and biomedical applications (Gao, 2015). The structure of graphene is consisted of two equivalent sub-lattices of carbon that bonded together with σ bonds. Meanwhile, each carbon atom in the lattice has a π orbital that contributes to a delocalized network of electrons (Zhu *et al.*, 2010).

Conventionally, graphene is formed from a technique called micromechanical cleavage in year 2004 which obtained by extracting monolayer sheets from the three-dimensional graphite. However, a lot of methods nowadays that can be applied to synthesize graphene without the formation of graphene oxide (GO) such as epitaxial growth by ultrahigh vacuum graphitization, chemical vapor deposition (CVD), solvothermal synthesis with pyrolysis and electrochemical methods (Antolini, 2012; Zhang *et al.*, 2009). Nevertheless, the

chemical process of the production of graphene from exfoliation of graphite oxide involves hazardous and toxic reagents even it shows a route for scalable synthesis (Purkait *et al.*, 2017). Generally, preparation of synthetic graphene can be divided into two categories (i) top-down and (ii) bottom-up.

For top-down approach, graphite is exfoliated into graphene layers via overcoming van der Waals forces between the layers using oxidation or other treatments (Yi and Shen, 2015). However, during the sheet separation, graphene tends to possess surface defects which can cause re-agglomeration and low yields (Das *et al.*, 2017). In addition, chemically produced, reduced graphene oxide is not acceptable for energy storage device as it suffers from poor electrical conductivity (Purkait *et al.*, 2017). On the other hand, graphene needs to be prepared from scratch in bottom-up method. It starts with simple carbon molecules like methane and ethanol (Chua and Pumera, 2014). Bottom-up methods include chemical vapor deposition (CVD) and epitaxial growth on silicon carbide (SiC) (Bhuyan *et al.*, 2016).

Conventional method of preparing graphene

The most common conventional method to produce graphene is via mechanical exfoliation method which also known as "scotch-tape method". By using a scotch-tape, high purity graphite will be separated into few layers of graphene (Martinez *et al.*, 2011). However, it is very challenging to overcome the van der Waals attraction between adjacent graphene flakes when preparing graphene from this method (Yi and Shen, 2015). In addition, the monoatomic layer that will be formed is difficult to control (Chang *et al.*, 2010).

CVD is comparatively an easier method to produce graphene with desired features. This method involves the decomposition of a carbon feedstock either polymers or hydrocarbons (Seah et al., 2014). To synthesize a good quality of graphene via CVD, few ranges of transition metals can be used which include platinum (Pt), cobalt (Co), nickel (Ni) and copper (Cu) (Seah et al., 2014; Lee et al., 2016). By applying this technique, the number of graphene layers can be controlled by the type and thickness of the catalyst chosen. On the other hand, the size of catalyst will affect the graphene size (Seah et al., 2014).

Besides that, GO can be prepared via several methods which are modified Hummers, Brodie and Staudenmaier (Seah et al., 2014). Meanwhile, graphene is commonly produced by reduction of GO (Park et al., 2011) through the removal of the oxygen containing groups with the recovery of a conjugated structure (Pei and Cheng, 2012). GO can be reduced via chemical or thermal reduction (Sun and Shi, 2013). Chemical reduction of GO is commonly used due to several advantages such as the product can be produced in bulk and graphene with processability and functionality can be obtained (Yuan et al., 2012). In addition, this method is very suitable to be carried out under mild conditions (Yin et al., 2015) and cheaper (Pei and Cheng, 2012). The common reductant used to reduce GO includes hydrazine, sodium borohydride (NaBH₄) and hydrobromic acid (HBr) (Fernández-Merino et al., 2012). On the other hand, thermal reduction usually need high-temperature treatment (1000 – 1100 °C) (Yin et al., 2015) and this will prevent the rGO to be redispersed in solutions (Sun and Shi, 2013). Unfortunately, the reduction process can cause structural defects into the produced graphene (Seah et al., 2014). Table 1 summarizes the conventional method used to produce graphene.

Table 1 Conventional method to produce graphene

Method	Advantages	Disadvantages	References
Mechanical exfoliation (scotch-tape)	Able to produce a single layer of defect-free graphene. Simple method.	Difficult to overcome the van der Waals attraction. Not suitable for large-scale production.	(Singh et al., 2011; Lee et al., 2013; Yi and Shen, 2015)
CVD	Easier to control the number of graphene layer. Large surface area of graphene. High quality of graphene with minimal defects.	Not suitable for large-scale production.	(Zhu et al., 2010; Seah et al., 2014; Yin et al., 2015)
Reduction of GO	Graphene produced is suitable for energy applications.	Produced graphene with many defects. Environmental issues (hazardous reductant is used in most of the reduction processes).	(Dubin et al., 2010; Sun and Shi, 2013)

The present review is mainly focused on graphene preparation using different precursors instead of aforementioned conventional precursors. The waste generation and accumulation become inevitable, so wise use of any waste is a commendable approach. Further converting the waste into advance material-like graphene is an appreciable act. Bio-precursor such as chitosan, glucose, and alginate, also supports the preparation of graphene. Even coal has been used to

prepare graphene quantum dots. The entire review deals with the study on preparation of graphene via different unusual precursors, which will be discussed in later section with reference to literature. Every approach has its own merit and demerit, and nevertheless, each approach is admirable and novel. If graphene is successfully and economically produced using biomass or other waste, it can be used for several applications from energy to environment.

Importance to prepare graphene from different precursors

Graphene, which is prepared from conventional method may not compatible for polymer composites because it cannot produce graphene in a large amount (Kim et al., 2010). Currently, bio-waste materials have been a great source for production of carbon. In addition, bio-waste materials are abundant and need for recycling. Furthermore, the management of waste biomass is challenging especially in well-developed countries. Thus, it is suggested to convert the material into carbonaceous materials which can be used in enormous applications (Purkait et al., 2017). Hence, researchers have put a lot of effort to synthesize graphene sheets using eco-friendly biomass precursors such as waste corn shell, egg shell and gelatin (Chen et al., 2016).

Graphene derived from bioprecursors and waste material

Glucose-based graphene

Glucose or also known as sugar molecule (C₆H₁₂O₆) is abundant and renewable carbon source. Recently, researchers have started to use glucose as a starting material to prepare graphene (Priyanka and Saravanakumar, 2017). Zhang and coworkers reported that graphene sheet is obtained via carbonization and calcination of a glucose and activating agent mixture (Fig. 1) (Zhang et al., 2014). FeCl₃ is necessary to obtain high-quality graphene material. From the Raman spectrum, there are three typical bands found which are the D-band, G-band and 2D-band (Zhang et al., 2014; Wu et al., 2016). G-band indicates the existence of sp²-hybridized carbon atom. Meanwhile, the defects like disorders, edges and boundaries of the graphene are shown by the D-band and the 2D-band provides the number of layers of the graphene material. Without the addition of activating agent, the quality of the graphite material is disappointing due to the amorphous carbon structure. This is proven by the weakening 2D-band while the intensity of the D-band is increased. Upon the addition of activating agent, the properties of graphene are getting better (Zhang et al., 2014). For instance, the electrical conductivity is found to be at 768 S/cm, which is similar to the graphene that is formed via CVD method (Chen et al., 2011). In addition, the peak of the amorphous carbon is decreased and the graphite carbon is increased (2θ = 26°) when being analyzed with X-ray Diffraction (XRD). Hence, with the addition of FeCl₃ it can be concluded that a high quality of graphene is obtained. (Zhang et al., 2014).

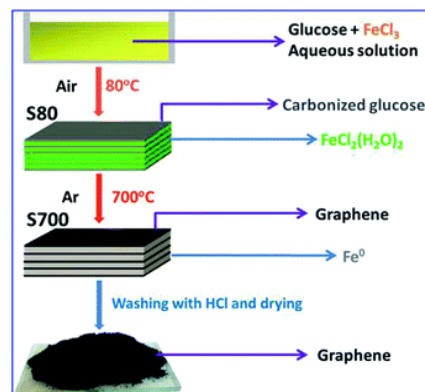


Fig. 1 Illustration of preparation of graphene via carbonization and calcination (Zhang et al., 2014).

Besides that, graphene quantum dots (GQDs) are also derived from glucose. GQDs which are luminescent carbon nanomaterials, have received massive attention in various areas of analytical chemistry. This is because GQDs have higher surface area, larger diameters and better surface grafting properties (Li *et al.*, 2013). Hallaj and coworkers reported that the GQDs are prepared via simple pyrolysis of glucose for chemiluminescence (CL). XRD spectrum of GQDs shows a broad peak around $2\theta = 20^\circ$, which indicated that graphite structure is obtained (Hallaj *et al.*, 2014). Two peaks around 1370 and 1705 cm^{-1} are found in the Raman spectrum which corresponded to the D and G band of graphene, respectively (Tang *et al.*, 2013). It is reported that GQDs shows a small intensity (I) of I_D/I_G which is about 0.5-0.6. This indicates that the produced material is made up of high quality crystalline of graphitic system (Hallaj *et al.*, 2014; Shehab *et al.*, 2017).

Rice-husks derived graphene

Approximately, 120 million tons of rice husks are produced annually in the world. Due to the massive production of rice husk, it receive abundant of attention as a starting material to generate high value-added materials such as silica and porous carbon. To prepare graphene from rice husk, typically potassium hydroxide (KOH) is used to activate the agricultural waste. This is because, KOH can greatly helps in the formation of pores in carbon materials including carbon nanotubes, graphene, and carbon fibers, thus enhancing their electrochemical performance (Muramatsu *et al.*, 2014). On top of that, KOH also assists in the formation of high purity graphene material containing stable and clean edges (Priyanka and Saravanakumar, 2017).

Singh and coworkers reported that graphene is synthesized by activating rice husk with KOH and annealing at 900°C (Fig. 2). The results show that the produced graphene has few layers of graphene with agglomeration of silica particles when it is observed using TEM (Fig. 3(a)) (Singh *et al.*, 2017). Using the same method, Muramatsu and colleague claimed that they have produced graphene with high crystallinity and corrugated containing pores and active edges (Muramatsu *et al.*, 2014). Meanwhile, the I_D/I_G ratio (0.55) on Raman spectrum confirms that the synthesized sample consists of few layers of graphene (Singh *et al.*, 2017). The growth of crystalline graphene is confirmed using XRD. Silica peak around 23° on RHA is disappeared after chemical activation with KOH. On the other hand, weak peaks around (002) and (100) are observed in the graphene sample, which indicated the absence of regularly stacked graphitic structure (Fig. 3 (b)) (Muramatsu *et al.*, 2014).

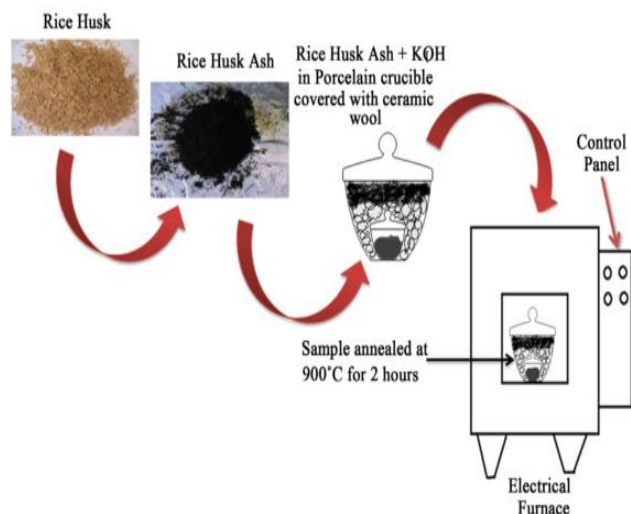


Fig. 2 Experimental set-up for synthesis of rice husk ash derived graphene (Singh *et al.*, 2017).

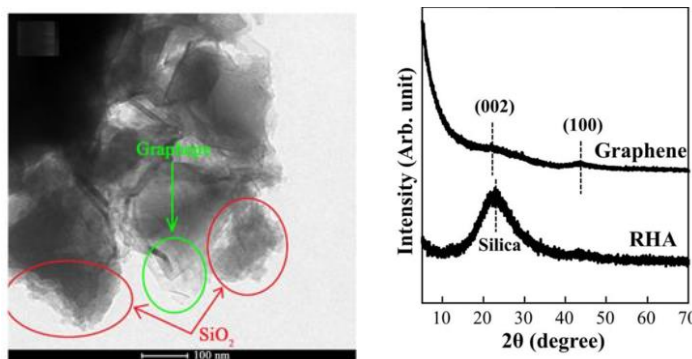


Fig. 3 (a) TEM image of graphene (Singh *et al.*, 2017) (b) XRD spectrum of graphene and rice husk ash (Muramatsu *et al.*, 2014)

Chitosan-based Graphene

After cellulose, chitosan is found to be the second most abundant biopolymer in nature and it has been used in a broad range of applications such as biomedical, pharmaceutical and industrial applications because of its good biocompatibility, biodegradability, and multiple functional groups of amino (NH_2) at C-2 and hydroxyl (OH) at C-3 (Kumar and Koh, 2014; Anandhavelu and Thambidurai, 2013). Interestingly, chitosan can adhere to negatively-charged surface or adsorb negatively-charged materials when it is dissolved and carried with the positive charge of $-\text{NH}_3^+$ groups. Thus, chitosan has been used widely to disperse nanomaterials and immobilize enzymes for constructing biosensors (Yin *et al.*, 2010). In addition, chitosan is a natural nitrogen containing biopolymer which able to produce graphene that is suitable for electrocatalyst. The presence of nitrogen atoms inside the graphene sheet will greatly enhance their electronic bands and become semiconductor (Lavorato *et al.*, 2014).

Cobos and teamwork reported a novel study on the effect of nanofiller (graphene and plasticizer (glycerol)) on chitosan (CS) matrix. Based on the FT-IR spectrum analysis, plasticized CS-graphene has showed that NH_3^+ band is shifted from 1552 cm^{-1} to 1539 cm^{-1} as compared to pristine plasticized CS. This is because, when graphene is introduced into the matrix, there is an electrostatic interaction between negatively-charged surface of graphene with positively-charged chitosan (Cobos *et al.*, 2018). The crystallinity of plasticized CS-graphene is observed under XRD analysis. It shows that plasticized CS-graphene has only a single peak at $2\theta = 23^\circ$ which indicated that the incorporation of graphene has resulted in the decrease of the crystallinity of CS (Ogawa *et al.*, 1984). Besides that, the SEM image confirms the existence of graphene as it shows a rough fractured surface and wave-like morphology without any aggregation which indicated that the graphene sheets are dispersed homogeneously in the CS matrix (Figure 4) (Han *et al.*, 2011).

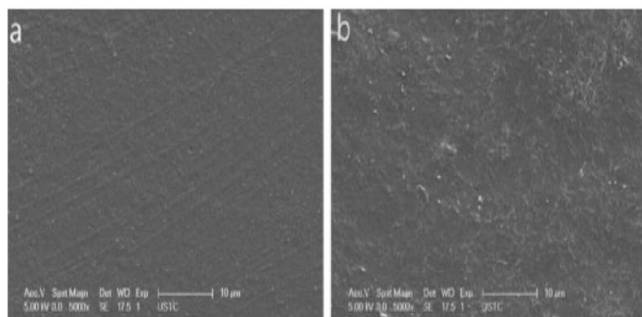


Fig. 4 SEM images of (a) CS (b) CS/GO (Han *et al.*, 2011).

Another novel method of producing composites with low defect graphene sheets by direct chemical exfoliation of graphite in chitosan solution (Cs-Ac) is reported by Jagiello and coworkers (2014). FT-IR spectrum of Cs-Ac shows α -stretching vibrations of symmetrical and asymmetrical ether (C-O-C) bonds at 1068 cm^{-1} and 1036 cm^{-1} , respectively. Meanwhile, peaks at 1650 cm^{-1} to 1100 cm^{-1} are

indicated to the chemical binding that associated with amine and amide groups (Akar *et al.*, 2016). Raman spectrum illustrates that there are G-band at 1582 cm^{-1} and 2D-band at $\sim 2700\text{ cm}^{-1}$. The 2D-band suggests that the graphene is formed in few layers. On the other hand, the I_G/I_{2D} and I_G/I_D shows that graphene sheets are defected lesser and thinner in this study (Yang *et al.*, 2010). Around 23° on RHA is disappeared after chemical activation with KOH. On the other hand, weak peaks around (002) and (100) are observed in the graphene sample which showed the absence of regularly stacked graphitic structure (Figure 3 (b)) (Muramatsu *et al.*, 2014).

Corn stalk core-derived graphene

Cellulose biomass is a promising material that can be used to generate sustainable bio-fuels as alternative energy for fossil fuel. Besides that, it can also supply reproducible raw materials for production of chemical products like surfactants, pigments and polymers (Xu *et al.*, 2016). Corn stalk is one of the ample agricultural wastes that is made up of a cortex and a core. The cortex is made up of cellulose and lignin while corn stalk core (CSC) is primarily hemicellulose and lignin and cellulose. CSC is naturally porous, which can form a large surface area of carbon, which is an excellent property for electron transfer and easier for ion diffusion in carbon matrix (Cao *et al.*, 2016).

Liu and coworkers have synthesized CSC-GO via assembly mechanism (freeze-dried process) where GO is firstly prepared by using modified Hummer's method (Figure 5) (Liu *et al.*, 2018). From the SEM images, it shows that the freeze-dried GO exhibits monolithic structure and after the addition of CSC, the surface of CSC is covered with GO (Figure 6) (Karim *et al.*, 2006). After the addition of CSC, the layers of GO are distracted and stripped into thinner layer (Cong *et al.*, 2011).

On the other hand, Ge and teammates have prepared citric acid functionalized magnetic graphene oxide coated corn straw (CA-mGOCS) via solvothermal system (Ge *et al.*, 2016). It is reported that CA has successfully grafted into mGOCS due to the formation of peak around 1756 cm^{-1} on FT-IR spectrum, which is belong to CA (Li *et al.*, 2013). Raman spectrum indicates that the I_D/I_G ratio is reduced when the amount of CS added into the mixture is increased, concluding that the order of carbonaceous material structure and sp^2 cluster size are increased (Ouyang *et al.*, 2015).

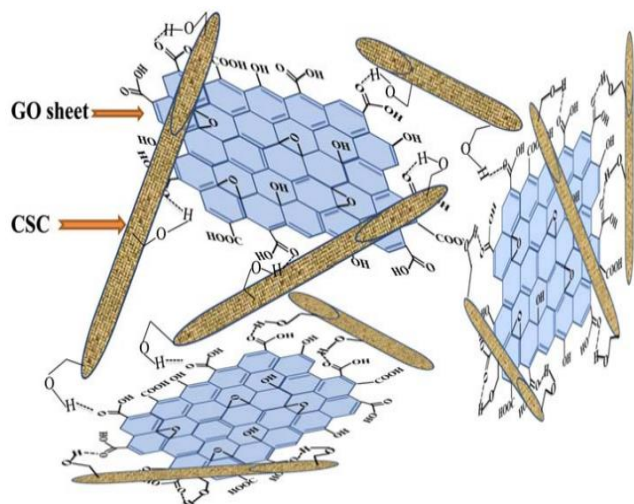


Fig. 5 Illustration of assembly mechanism between GO and CSC (Liu *et al.*, 2018).

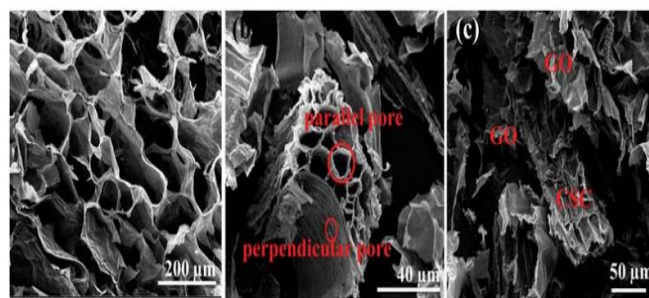


Fig. 6 SEM images of (a) freeze-dried GO (b) $ZnCl_2$ treated CSC (c) CSC-GO (Liu *et al.*, 2018).

Plastic waste-based graphene

Plastic is one of the abundant wastes that found on the earth which includes plastic bottles, plastic bags and plastic sheets, which can cause a serious waste of resource and environmental pollution (Cui *et al.*, 2017). Therefore, there are a lot of alternatives to convert this waste into carbon based material. Study conducted by Essawy and teammates reported that, graphene is successfully obtained from polyethylene terephthalate (PET), which is used enormously in pharmaceutical, food and soft drink bottles and containers industries (El Essawy *et al.*, 2017). Usually, PET waste is eliminated using incineration, chemical recycling or feedstock recycling to produce gaseous and liquid products and carbon-enriched materials (Mishra *et al.*, 2003). However, as PET contains high amount of carbon and negligible content of mineral impurities, it is favorable to turn this material into carbon based material such as carbon nanotubes, carbon microspheres and activated carbon (Mendoza-Carrasco *et al.*, 2016).

Based on the study, the SEM image exhibits a fiber network morphology with void space, which can provide access for porosity (figure 7) (El Essawy *et al.*, 2017). The graphene produced shows a diffraction pattern at $2\theta = 26, 42.3$ and 44.3° which corresponded to (002), (100) and (101). The result indicates that the graphene is highly amorphous (Zhang *et al.*, 2016). On the other hand, the Raman spectrum indicates that there are two bands found which are G-band (1596 cm^{-1}) and D-band (1360 cm^{-1}). G-band is formed by the interaction with other layers or due to external disorder that breaks the hexagonal symmetry of graphene. Meanwhile, the formation of D-band indicates a highly disordered graphite arrangement (El Essawy *et al.*, 2017). Thus, the intensity ratio (I_D/I_G) obtained is relatively low, 1.13 which means that the prepared graphene is highly disordered (Sergiienko *et al.*, 2009).

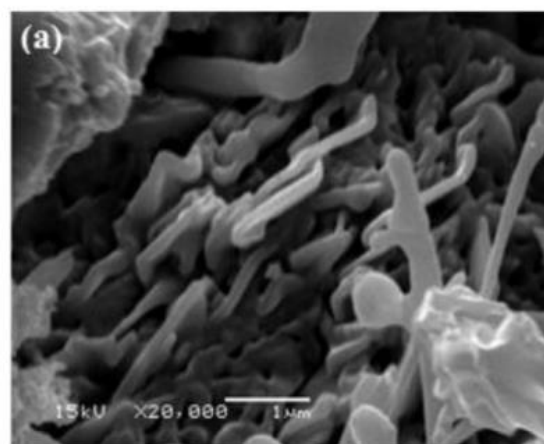


Fig. 7 SEM image of graphene derived from PET (El Essawy *et al.*, 2017).

On the other hand, Sharma and colleagues have produced a high quality single crystal graphene on polycrystalline Cu foil using solid waste plastic as carbon source in an ambient pressure (AP) chemical vapor deposition (CVD) process (Figure 8) (Sharma *et al.*, 2014). They claimed that, graphene obtained via this method provides much

better opportunity to grow large-area high quality monolayer graphene (Li *et al.*, 2009, 2011). This statement is proven by the Raman spectrum where a very small defect on the graphene is shown by the low intensity of D-band. Meanwhile, G-band and 2D band are found at 1590 cm^{-1} and 2700 cm^{-1} , respectively. Based from the results, it confirms that the graphene produced in this study is single layer as the 2D-band has a higher intensity than G-band (Sharma *et al.*, 2014).

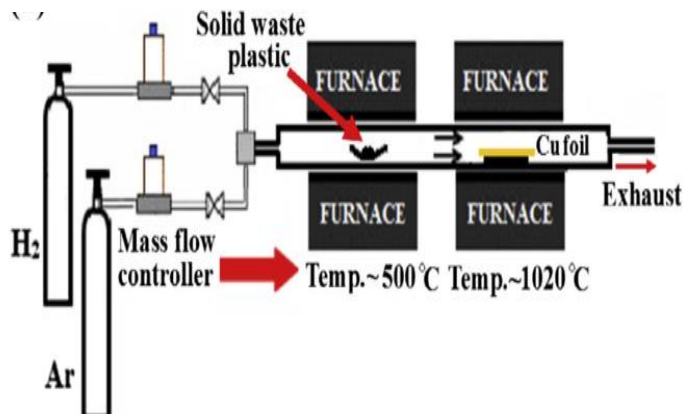


Fig. 8 Schematic diagram of preparation of graphene via CVD method (Sharma *et al.*, 2014)

Besides that, Chui and coworkers reported that they have prepared graphene foil (GF) via solid-state CVD (Cui *et al.*, 2017). Based on the characterization, Raman spectrum shows that the G-band of the material is intense and narrow, indicating a good crystallinity and high graphitization (Pimenta *et al.*, 2007). On the other hand, low I_D/I_G shows that the defect on the sample is relatively less. The XRD analysis further supports the Raman results where peak at $2\theta = 26.7^\circ$ reveals a high graphitization of graphene (Cui *et al.*, 2017).

Application of waste and biomass derived graphene

Nowadays, it is vital for researchers to develop energy-related materials and devices to meet the global energy demand (Dawoud *et al.*, 2007). Graphene has received tremendous attention and its properties have been studied extensively such as room temperature quantum Hall effect (Bhuyan *et al.*, 2016), optical (Nasrollahzadeh *et al.*, 2015) and mechanical properties (Potts *et al.*, 2011). Owing to its unique properties, graphene has been attracted to be applied in various applications which include dye removal (Elsagh *et al.*, 2017), adsorption of heavy metals (Matilainen *et al.*, 2010), supercapacitors (Chen *et al.*, 2014) and many more. Herein, we summarize some of the recent studies concerning the applications of waste and biomass derived graphene in dye removal, adsorption of toxic and heavy metals, gas storage, high performance supercapacitors, electrochemical sensor and fuel cell.

Dye removal

Water pollution and inadequate access to clean water are some of the most pervasive environmental problems that afflicting people throughout the world. Synthetic dyes contain some components or moieties that can be toxic, carcinogenic, teratogenic or mutagenic to aquatic life and humans, that consequently will cause danger (Konicki *et al.*, 2017). For an effective dye removal, many studies have suggested that adsorption process is preferred compared to other methods of waste water treatment (Rajamohan, 2009; Elsagh *et al.*, 2017).

The adsorption efficiency can be evaluated by knowing the amount of adsorbent (Mittal *et al.*, 2009). Liu and colleague reported that increasing of GO loading in CSC can lead to higher adsorption of methylene blue (MB) (pH = 8, 298 K), where the adsorption efficiency is increased to 34.11% when 50 wt% GO is used. This is because, higher GO amount will reduce the adsorbent concentration to achieve the equilibrium adsorption (Liu *et al.*, 2018). Meanwhile, magnetic/chitosan/graphene oxide (MCGO) shows the highest

adsorption efficiency, 92.9% at 90°C using Disperse Blue 367 (DB367). This indicates that, the internal structure of MCGO is influenced by higher temperature which then facilitated the DB367 distribution in the adsorbent's interspaces structure. In addition, the adsorption process is endothermic (Taher *et al.*, 2018). Adsorption of crystal violet (CV) is evaluated using graphene oxide nanoplatelets (GONp) which are synthesized from rice straw biomass. The result shows that adsorption of CV is increased (99.73%) along with higher amount of adsorbent (0.75 g/L). However, no significant improvement of adsorption is observed when higher amount of GONp is added (Figure 9 (a)) (Goswami *et al.*, 2017)

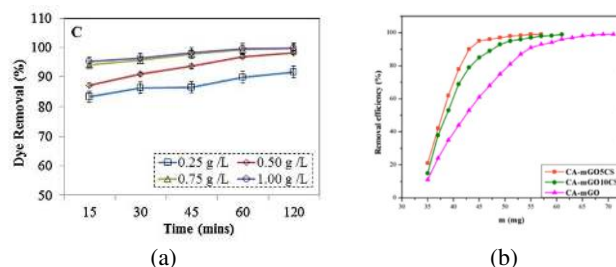


Fig. 9 Effect of adsorbent dosage on the removal of (a) CV (Goswami *et al.*, 2017) (b) MB (Ge *et al.*, 2016)

Citric acid functionalized magnetic graphene oxide coated corn straw (CA-mGOCS) is synthesized to study the effectiveness for methylene blue (MB) adsorption. It is reported that CA-mGO5CS (5g CS) shows the fastest speed in the adsorption process (Figure 9 (b)). This reveals that CS has porous structure and abundant reactive groups which will promote the adsorption of MB solution (75 mL, pH = 8, 298 K) (Ge *et al.*, 2016).

Adsorption of toxic and heavy metals

Waste water from agricultural, landfill and industrial may contain various heavy metals such as mercury (Hg), lead (Pb), silver (Ag), zinc (Zn) and copper (Cu) that will contribute to health problem and ecosystem issues because of their toxicities and danger (Mashhadzadeh *et al.*, 2016). Recently, study shows that adsorption becomes a promising method to overcome the aforementioned problem due to its low cost, simplicity of design, ease of operation, insensitivity to toxic pollutants and smaller amounts of harmful substances (Peng *et al.*, 2017).

Chitosan-gelatin/graphene oxide (CGGO) has been synthesized and the adsorption performance on lead ion (Pb^{2+}) is studied. Zhang reported that, higher GO content can gradually increase the adsorption capacity of Pb^{2+} (up to 90 %) (Zhang *et al.*, 2011). Khasbataar and teammates claimed that this is likely due to the existence of carboxyl groups which are effective chelating groups for metal ions (Khasbataar *et al.*, 2008).

Gas storage

Carbon dioxide (CO_2) has been the main greenhouse gas that leads to global warming and climate change (Nowrouzi *et al.*, 2018). To date, various of efforts are being undertaken to reduce the greenhouse gas concentration that is mostly emitted from the combustion of fuel in vehicles and burning of coal in power plants (Yoon *et al.*, 2011). Among all techniques, adsorption is considered as an efficient alternative due to its low energy requirement and cheap (Shafeeyan *et al.*, 2010).

Ekhlesi and colleague have studied the graphene that derived from populus wood biomass (PWB) as adsorbents for CO_2 capture using different amounts of KOH as activating agent. Based on the report, they stated that the optimum KOH/C ratio is found to be at 3:1 as the maximum CO_2 adsorption capacity can be reached up to 9.86 mmol CO_2/g (Ekhlesi *et al.*, 2018). This can be explained by the fact that the increased in activation ratio will cause the micropores and mesopores to be enlarged. Hence, the adsorption capacity of CO_2 will be enhanced (Cazorla-Amorós *et al.*, 1996).

High performance supercapacitors

Supercapacitors is one of the energy storage devices that received tremendous attention due to its high power densities, long cycling life and fast charge-discharge rate (El-Kady and Kaner, 2013). Supercapacitors can be classified into two categories: electrochemical double-layer capacitors (EDLCs) and pseudocapacitors (Simon *et al.*, 2014). Hence, Fan and teammates have conducted a study on N-doping cotton derived carbon frameworks incorporated with reduced graphene oxide (NCCF-rGO) that is prepared from direct carbonization of commercial cotton for supercapacitors. NCCF-rGO shows that it has excellent capacitance retention as no changes is observed when it is bent under different angles (Figure 10). In addition, it also shows a better energy storage capability and displays a high energy density of 20 Wh/kg at a power density of 4000 W/kg (Fan *et al.*, 2017).

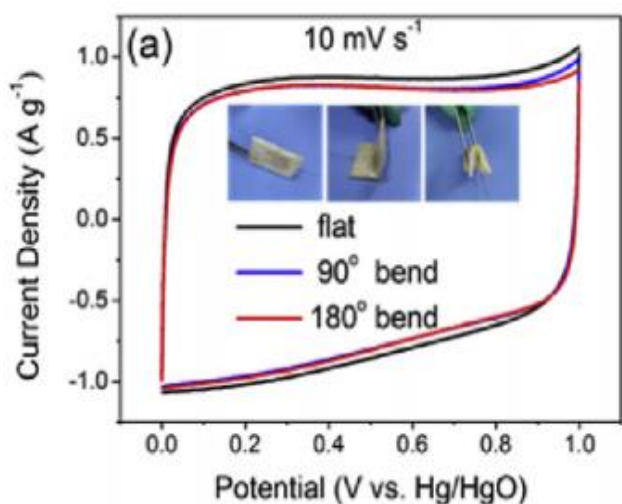


Fig. 10 CV curves of the flexible devices at different bending angles (Fan *et al.*, 2017).

Graphene which is synthesized from biomass phytic acid (PAGH) has been studied for supercapacitors. It is reported that PAGH has outstanding flexibility due to excellent mechanical strength which can be revealed by CV curves (Figure 11). The energy density shows the highest value of 26.5 Wh/kg at power density of 132 W/kg (Liu *et al.*, 2018).

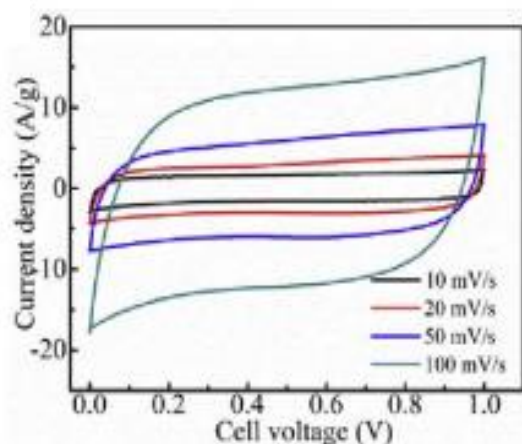


Fig. 11 CV curves of PAGH (Liu *et al.*, 2018).

CONCLUSION

Waste and biomass derived graphene is another promising material that can be applied in many applications, yet the synthesis method is still challenging. Therefore, several studies need to be carried out to justify the best method in order to produce graphene with the most satisfying characteristics. In addition, production of graphene using biomass precursors will possibly reduce the disposal issue of the material. Furthermore, based on this review, most of the approaches are scalable, efficient and cost effective, making graphene as suitable to be applied in many applications such as water filtration, supercapacitors, fuel cell and gas adsorption.

ACKNOWLEDGEMENT

Authors would like to express their gratitude for the supports and funds by Ministry of Education (MOE) Malaysia and Universiti Teknologi Malaysia through Fundamental Research Grant Scheme (R.J130000.7846.4F929), GUP grant (Q.J130000.2546.16H29), PRGS ICC-UTM (R.J130000.7746.4J329) and Higher Institution Centre of Excellence (HiCOE) grant (R.J090301.7846.4J180 and R.J090301.7846.4J179).

REFERENCES

- Akar, E., Seki, Y., Cem Yılmaz, Ö., Çetin, L., Özdemir, O., Şen, İ., Sever, K., Gürses, B., Sarikanat, M. (2016) *Electromechanical performance of chitosan-based composite electroactive actuators*. in: *Composites Science and Technology*, 108-115.
- Anandhavelu, S., Thambidurai, S. (2013) *Electrochimica Acta* Single step synthesis of chitin / chitosan-based graphene oxide – ZnO hybrid composites for better electrical conductivity and optical properties. *Electrochimica Acta*, 90, 194–202.
- Antolini, E. (2012) Graphene as a new carbon support for low-temperature fuel cell catalysts. *Applied Catalysis B: Environmental*, 123-124, 52–68.
- Bhuyan, M.S.A., Uddin, M.N., Islam, M.M., Bipasha, F.A., and Hossain, S.S. (2016) Synthesis of graphene. *International Nano Letters*, 6, 65–83. Springer Berlin Heidelberg.
- Cao, Y., Wang, K., Wang, X., Gu, Z., Fan, Q., Gibbons, W., Hoefelmeyer, J. D., Kharel, P. R., Shrestha, M. (2016) Hierarchical porous activated carbon for supercapacitor derived from corn stalk core by potassium hydroxide activation. *Electrochimica Acta*, 212, 839–847.
- Cazorla-Amorós, D., Alcañiz-Monge, J., and Linares-Solano, A. (1996) Characterization of Activated Carbon Fibers by CO₂ Adsorption. *Langmuir*, 12, 2820–2824. American Chemical Society.
- Chang, Y.M., Kim, H., Lee, J.H., and Song, Y.W. (2010) Multilayered graphene efficiently formed by mechanical exfoliation for nonlinear saturable absorbers in fiber mode-locked lasers. *Applied Physics Letters*, 97, 1–4.
- Chen, F., Yang, J., Bai, T., Long, B., and Zhou, X. (2016) Facile synthesis of few-layer graphene from biomass waste and its application in lithium ion batteries. *Journal of Electroanalytical Chemistry*, 768, 18–26. Elsevier B.V.

- Chen, Y.-R., Chiu, K.-F., Lin, H. C., Chen, C. -L., Hsieh, C. Y., Tsai, C. B., and Chu, B. T. T. (2014) Graphene/activated carbon supercapacitors with sulfonated-polyetheretherketone as solid-state electrolyte and multifunctional binder. *Solid State Sciences*, 37, 80–85.
- Chen, Z., Ren, W., Gao, L., Liu, B., Pei, S., Cheng, H.-M. (2011) Three-dimensional flexible and conductive interconnected graphene networks grown by chemical vapour deposition. *Nature Materials*, 10, 424.
- Chua, C. K., Pumera, M. (2014) Chemical reduction of graphene oxide: a synthetic chemistry viewpoint. *Chemical Society Reviews*, 43, 291–312.
- Cobos, M., González, B., Jesús Fernández, M., Dolores Fernández, M. (2018) Study on the effect of graphene and glycerol plasticizer on the properties of chitosan-graphene nanocomposites via in situ green chemical reduction of graphene oxide. *International Journal of Biological Macromolecules*, 114, 599–613.
- Cong, C., Yu, T., Saito, R., Dresselhaus, G.F., Dresselhaus, M. S. (2011) Second-order overtone and combination raman modes of graphene layers in the range of 1690–2150 cm⁻¹. *ACS Nano*, 5, 1600–1605.
- Cui, L., Wang, X., Chen, N., Ji, B., and Qu, L. (2017) Trash to treasure: converting plastic waste into a useful graphene foil. *Nanoscale*, 9, 9089–9094.
- Das, V.K., Shifrina, Z.B., and Bronstein, L.M. (2017) Graphene and graphene-like based materials in biomass conversion: Paving the way to the future. *Journal of Materials Chemistry A*, 5, 25131–25143. Royal Society of Chemistry.
- Dawoud, B., Amer, E., and Gross, D. (2007) Experimental investigation of an adsorptive thermal energy storage. *International journal of energy research*, 31, 135–147.
- Dubin, S., Gilje, S., Wang, K., Tung, V.C., Cha, K., Hall, A.S., Farrar, J., Varshneya, R., Yang, Y., and Kaner, R.B. (2010) A one-step, solvothermal reduction method for producing reduced graphene oxide dispersions in organic solvents. *ACS Nano*, 4, 3845–3852.
- Ekhlasli, L., Younesi, H., Rashidi, A., and Bahramifar, N. (2018) Populus wood biomass-derived graphene for high CO₂ capture at atmospheric pressure and estimated cost of production. *Process Safety and Environmental Protection*, 113, 97–108.
- El-Kady, M.F. and Kaner, R.B. (2013) Scalable fabrication of high-power graphene micro-supercapacitors for flexible and on-chip energy storage. *Nature Communications*, 4, 1475. Nature Publishing Group, a division of Macmillan Publishers Limited. All Rights Reserved.
- Elsagh, A., Moradi, O., Fakhri, A., Najafi, F., Alizadeh, R., and Haddadi, V. (2017) Evaluation of the potential cationic dye removal using adsorption by graphene and carbon nanotubes as adsorbents surfaces. *Arabian Journal of Chemistry*, 10, S2862–S2869.
- El Essawy, N.A., Ali, S.M., Farag, H.A., Konsowa, A.H., Elnouby, M., and Hamad, H.A. (2017) Green synthesis of graphene from recycled PET bottle wastes for use in the adsorption of dyes in aqueous solution. *Ecotoxicology and Environmental Safety*, 145, 57–68.
- Fan, Y.M., Song, W.L., Li, X., and Fan, L.Z. (2017) Assembly of graphene aerogels into the 3D biomass-derived carbon frameworks on conductive substrates for flexible supercapacitors. *Carbon*, 111, 658–666.
- Fernández-Merino, M.J., Paredes, J.I., Villar-Rodil, S., Guardia, L., Solís-Fernández, P., Salinas-Torres, D., Cazorla-Amorós, D., Morallón, E., Martínez-Alonso, A., and Tascón, J.M.D. (2012) Investigating the influence of surfactants on the stabilization of aqueous reduced graphene oxide dispersions and the characteristics of their composite films. *Carbon*, 50, 3184–3194.
- Gao, W. (2015) The chemistry of graphene oxide. *Graphene Oxide: Reduction Recipes, Spectroscopy, and Applications*, 61–95.
- Ge, H., Wang, C., Liu, S., and Huang, Z. (2016) Synthesis of citric acid functionalized magnetic graphene oxide coated corn straw for methylene blue adsorption. *Bioresource Technology*, 221, 419–429.
- Goswami, S., Banerjee, P., Datta, S., Mukhopadhyay, A., and Das, P. (2017) Graphene oxide nanoplatelets synthesized with carbonized agro-waste biomass as green precursor and its application for the treatment of dye rich wastewater. *Process Safety and Environmental Protection*, 106, 163–172.
- Hallaj, T., Amjadi, M., Manzoori, J.L., and Shokri, R. (2014) Chemiluminescence reaction of glucose-derived graphene quantum dots with hypochlorite, and its application to the determination of free chlorine. *Microchimica Acta*, 182, 789–796.
- Han, D., Yan, L., Chen, W., and Li, W. (2011) Preparation of chitosan/graphene oxide composite film with enhanced mechanical strength in the wet state. *Carbohydrate Polymers*, 83, 653–658.
- İlbay, Z., Haşimoğlu, A., Özdemir, O.K., Ateş, F., and Şahin, S. (2017) Highly efficient recovery of biophenols onto graphene oxide nanosheets: Valorisation of a biomass. *Journal of Molecular Liquids*, 246, 208–214.
- Jagiello, J., Judek, J., Zdrojek, M., Aksienionek, M., and Lipinska, L. (2014) Production of graphene composite by direct graphite exfoliation with chitosan. *Materials Chemistry and Physics*, 148, 507–511.
- Karim, M.M., Das, A.K., and Lee, S.H. (2006) Treatment of colored effluent of the textile industry in Bangladesh using zinc chloride treated indigenous activated carbons. *Analytica Chimica Acta*, 576, 37–42.
- Khasbaatar, A.D., Chun, Y.J., and Choi, U.S. (2008) Trivalent Metal Adsorption Properties on Synthesized Viscose Rayon Succinate. *Polymer Journal*, 40, 302. The Society of Polymer Science, Japan.
- Kim, H., Abdala, A.A., and MacOsco, C.W. (2010) Graphene/polymer nanocomposites. *Macromolecules*, 43, 6515–6530.
- Konicki, W., Aleksandrak, M., Moszyński, D., and Mijowska, E. (2017) Adsorption of anionic azo-dyes from aqueous solutions onto graphene oxide: Equilibrium, kinetic and thermodynamic studies. *Journal of Colloid and Interface Science*, 496, 188–200.
- Kumar, S. and Koh, J. (2014) International Journal of Biological Macromolecules Physicochemical and optical properties of chitosan based graphene oxide bionanocomposite. *International Journal of Biological Macromolecules*, 70, 559–564.
- Lavorato, C., Primo, A., Molinari, R., and Garcia, H. (2014) N-doped graphene derived from biomass as a visible-light photocatalyst for hydrogen generation from water/methanol mixtures. *Chemistry - A European Journal*, 20, 187–194.
- Lee, H.C., Liu, W.-W., Chai, S.-P., Mohamed, A.R., Lai, C.W., Khe, C.-S., Voon, C.H., Hashim, U., and Hidayah, N.M.S. (2016) Synthesis of Single-layer Graphene: A Review of Recent Development. *Procedia Chemistry*, 19, 916–921.
- Lee, S., Eom, S.H., Chung, J.S., and Hur, S.H. (2013) Large-scale production of high-quality reduced graphene oxide. *Chemical Engineering Journal*, 233, 297–304.
- Li, L., Mak, K.Y., Leung, C.W., Chan, K.Y., Chan, W.K., Zhong, W., and Pong, P.W.T. (2013a) Effect of synthesis conditions on the properties of citric-acid coated iron oxide nanoparticles. *Microelectronic Engineering*, 110, 329–334.
- Li, L., Wu, G., Yang, G., Peng, J., Zhao, J., and Zhu, J.-J. (2013b) Focusing on luminescent graphene quantum dots: current status and future perspectives. *Nanoscale*, 5, 4015.

- Li, X., Cai, W., An, J., Kim, S., Nah, J., Yang, D., Piner, R., Velamakanni, A., Jung, I., Tutuc, E., Banerjee, S.K., Colombo, L., and Ruoff, R.S. (2009) Large-Area Synthesis of High-Quality and Uniform Graphene Films on Copper Foils. *Science*, 324, 1312 LP – 1314.
- Li, X., Magnuson, C.W., Venugopal, A., Tromp, R.M., Hannon, J.B., Vogel, E. M., Colombo, L., Ruoff, R.S. (2011) Large-Area Graphene Single Crystals Grown by Low-Pressure Chemical Vapor Deposition of Methane on Copper. *Journal of the American Chemical Society*, 133, 2816–2819. American Chemical Society.
- Liu, S., Ge, H., Wang, C., Zou, Y., and Liu, J. (2018a) Agricultural waste/graphene oxide 3D bio-adsorbent for highly efficient removal of methylene blue from water pollution. *Science of the Total Environment*, 628-629, 959–968.
- Liu, X., Zou, S., Liu, K., Lv, C., Wu, Z., Yin, Y., Liang, T., and Xie, Z. (2018b) Highly compressible three-dimensional graphene hydrogel for foldable all-solid-state supercapacitor. *Journal of Power Sources*, 384, 214–222.
- Martinez, A., Fuse, K., and Yamashita, S. (2011) Mechanical exfoliation of graphene for the passive mode-locking of fiber lasers. *Applied Physics Letters*, 99, 97–100.
- Mashhadzadeh, A.H., Vahedi, A.M., Ardjmand, M., and Ahangari, M.G. (2016) Investigation of heavy metal atoms adsorption onto graphene and graphdiyne surface: A density functional theory study. *Superlattices and Microstructures*, 100, 1094–1102.
- Matilainen, A., Vepsäläinen, M., and Sillanpää, M. (2010) Natural organic matter removal by coagulation during drinking water treatment: A review. *Advances in Colloid and Interface Science*, 159, 189–197.
- Mendoza-Carrasco, R., Cuerda-Correa, E. M., F Alexandre-Franco, M., Fernández-González, C., Gómez-Serrano, V. (2016) Preparation of high-quality activated carbon from polyethyleneterephthalate (PET) bottle waste. Its use in the removal of pollutants in aqueous solution. in: *Journal of Environmental Management*. 522-535.
- Mishra, S., Goje, A.S., Zope, V.S. (2003) Chemical Recycling, kinetics, and thermodynamics of poly (ethylene terephthalate) (PET) waste powder by nitric acid hydrolysis. *Polymer Reaction Engineering*, 11, 79–99.
- Mittal, A., Kaur, D., Malviya, A., Mittal, J., Gupta, V. K. (2009) Adsorption studies on the removal of coloring agent phenol red from wastewater using waste materials as adsorbents. *Journal of Colloid and Interface Science*, 337, 345–354.
- Muramatsu, H., Kim, Y.A., Yang, K.S., Cruz-Silva, R., Toda, I., Yamada, T., Terrones, M., Endo, M., Hayashi, T., and Saitoh, H. (2014) Rice husk-derived graphene with nano-sized domains and clean edges. *Small*, 10, 2766–2770.
- Nasrollahzadeh, M., Babaei, F., Fakhri, P., and Jaleh, B. (2015) Synthesis, characterization, structural, optical properties and catalytic activity of reduced graphene oxide/copper nanocomposites. *Rsc Advances*, 5, 10782–10789.
- Nowrouzi, M., Younesi, H., and Bahramifar, N. (2018) Superior CO₂ capture performance on biomass-derived carbon/metal oxides nanocomposites from Persian ironwood by H₃PO₄ activation. *Fuel*, 223, 99–114.
- Ogawa, K., Hirano, S., Miyaniishi, T., Yui, T., and Watanabe, T. (1984) A new polymorph of chitosan. *Macromolecules*, 17, 973–975.
- Ouyang, K., Zhu, C., Zhao, Y., Wang, L., Xie, S., and Wang, Q. (2015) Adsorption mechanism of magnetically separable Fe₃O₄/graphene oxide hybrids. *Applied Surface Science*, 355, 562–569.
- Park, S., An, J., Potts, J. R., Velamakanni, A., Murali, S., Ruoff, R.S. (2011) Hydrazine-reduction of graphite- and graphene oxide. *Carbon*, 49, 3019–3023.
- Pei, S., Cheng, H.M. (2012) The reduction of graphene oxide. *Carbon*, 50, 3210–3228.
- Peng, W., Li, H., Liu, Y., Song, S. (2017) A review on heavy metal ions adsorption from water by graphene oxide and its composites. *Journal of Molecular Liquids*, 230, 496–504.
- Pimenta, M. A., Dresselhaus, G., Dresselhaus, M. S., Cancado, L. G., Jorio, A., Saito, R. (2007) Studying disorder in graphite-based systems by Raman spectroscopy. *Physical Chemistry Chemical Physics*, 9, 1276–1290.
- Potts, J. R., Dreyer, D. R., Bielawski, C. W., Ruoff, R. S. (2011) Graphene-based polymer nanocomposites. *Polymer*, 52, 5–25.
- Priyanka, M., Saravanakumar, M. P. (2017) A short review on preparation and application of carbon foam. *IOP Conference Series: Materials Science and Engineering*, 263, 246–254.
- Purkait, T., Singh, G., Singh, M., Kumar, D., Dey, R.S. (2017b) Large area few-layer graphene with scalable preparation from waste biomass for high-performance supercapacitor. *Scientific Reports*, 7, 1–14.
- Purkait, T., Singh, G., Singh, M., Kumar, D., Dey, R.S. (2017a) Large area few-layer graphene with scalable preparation from waste biomass for high-performance supercapacitor. *Scientific Reports*, 7, 15–18.
- Rajamohan, N. (2009) Equilibrium studies on sorption of an anionic dye onto acid activated water hyacinth roots. *African Journal of Environmental Science and Technology*, 3, 399–404.
- Seah, C.-M., Chai, S.-P., Mohamed, A. R. (2014) Mechanisms of graphene growth by chemical vapour deposition on transition metals. *Carbon*, 70, 1–21.
- Sergiienko, R., Shibata, E., Kim, S., Kinota, T., and Nakamura, T. (2009) Nanographite structures formed during annealing of disordered carbon containing finely-dispersed carbon nanocapsules with iron carbide cores. in: *Carbon*. 1056-1065 pp.
- Shafeeyan, M. S., Daud, W. M. A. W., Houshmand, A., Shamiri, A. (2010) A review on surface modification of activated carbon for carbon dioxide adsorption. *Journal of Analytical and Applied Pyrolysis*, 89, 143–151.
- Sharma, S., Kalita, G., Hirano, R., Shinde, S. M., Papon, R., Ohtani, H., Tanemura, M. (2014) Synthesis of graphene crystals from solid waste plastic by chemical vapor deposition. *Carbon*, 72, 66–73.
- Shehab, M., Ebrahim, S., Soliman, M. (2017) Graphene quantum dots prepared from glucose as optical sensor for glucose. *CrystEngComm*, 184, 110–116.
- Simon, P., Gogotsi, Y., Dunn, B. (2014) Where do batteries end and supercapacitors begin? *Science*, 343, 1210 LP – 1211.
- Singh, P., Bahadur, J., Pal, K. (2017) One-step one chemical synthesis process of graphene from rice husk for energy storage applications. *Graphene*, 06, 61–71.
- Singh, V., Joung, D., Zhai, L., Das, S., Khondaker, S. I., Seal, S. (2011) Graphene based materials: Past, present and future. *Progress in Materials Science*, 56, 1178–1271.
- Somers, L. (2015) High yield preparation of macroscopic graphene oxide membranes. *Journal of the American Chemical Society*, 131(3)9–11.
- Sun, Y., Shi, G. (2013) Graphene/polymer composites for energy applications. *Journal of Polymer Science, Part B: Polymer Physics*, 51, 231–253.
- Taher, F. A., Kamal, F. H., Badawy, N. A., Shreshr, A. E. (2018) Hierarchical magnetic / chitosan / graphene oxide 3D nanostructure as highly effective adsorbent. *Materials Research Bulletin*, 97, 361–368.
- Tang, L., Ji, R., Li, X., Teng, K. S., Lau, S. P. (2013) Size-dependent structural and optical characteristics of glucose-derived graphene quantum dots. *Particle & Particle Systems Characterization*, 30, 523–531.

- Wu, L., Song, J., Zhou, B., Wu, T., Jiang, T., Han, B. (2016) Preparation of Ru/Graphene using Glucose as Carbon Source and Hydrogenation of Levulinic Acid to γ -Valerolactone. *Chemistry - An Asian Journal*, 11, 2792–2796.
- Xu, H., Zhang, H., Ouyang, Y., Liu, L., Wang, Y. (2016) Two-dimensional hierarchical porous carbon composites derived from corn stalks for electrode materials with high performance. *Electrochimica Acta*, 214, 119–128. Elsevier Ltd.
- Yang, X., Tu, Y., Li, L., Shang, S., Tao, X. (2010) Well-Dispersed Chitosan/Graphene Oxide Nanocomposites. *ACS Applied Materials & Interfaces*, 2, 1707–1713. American Chemical Society.
- Yi, M., Shen, Z. (2015) A review on mechanical exfoliation for the scalable production of graphene. *Journal of Materials Chemistry, A*, 3, 11700–11715.
- Yin, H., Ma, Q., Zhou, Y., Ai, S., Zhu, L. (2010) Electrochemical behavior and voltammetric determination of 4-aminophenol based on graphene-chitosan composite film modified glassy carbon electrode. *Electrochimica Acta*, 55, 7102–7108.
- Yin, P. T., Shah, S., Chhowalla, M., Lee, K. B. (2015) Design, synthesis, and characterization of graphene-nanoparticle hybrid materials for bioapplications. *Chemical Reviews*, 115, 2483–2531.
- Yoon, H. J., Jun, D. H., Yang, J. H., Zhou, Z., Yang, S. S., and Cheng, M. M.-C. (2011) Carbon dioxide gas sensor using a graphene sheet. *Sensors and Actuators B: Chemical*, 157, 310–313.
- Yuan, J., Chen, G., Weng, W., and Xu, Y. (2012) One-step functionalization of graphene with cyclopentadienyl-capped macromolecules via Diels–Alder “click” chemistry. *Journal of Materials Chemistry*, 22, 7929.
- Zhang, B., Song, J., Yang, G., Han, B. (2014) Large-scale production of high-quality graphene using glucose and ferric chloride. *Chemical Science*, 5, 4656–4660.
- Zhang, H. P., Luo, X. G., Lin, X. Y., Lu, X., Tang, Y. (2016) The molecular understanding of interfacial interactions of functionalized graphene and chitosan. *Applied Surface Science*, 360, 715–721.
- Zhang, L., Liang, J., Huang, Y., Ma, Y., Wang, Y., Chen, Y. (2009) Size-controlled synthesis of graphene oxide sheets on a large scale using chemical exfoliation. *Carbon*, 47, 3365–3368.
- Zhang, N., Qiu, H., Si, Y., Wang, W., Gao, J. (2011) Fabrication of highly porous biodegradable monoliths strengthened by graphene oxide and their adsorption of metal ions. *Carbon*, 49, 827–837.
- Zhu, Y., Murali, S., Cai, W., Li, X., Suk, J. W., Potts, J. R., and Ruoff, R. S. (2010) Graphene and graphene oxide: Synthesis, properties, and applications. *Advanced Materials*, 22, 3906–3924.