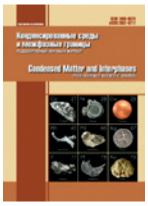

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Synthesis and study of graphene oxide obtained from waste transparent polythene bags using Modified Hummers method

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Abstract

Purpose: Plastic industry has seen explosive growth in previous decades over the world. Every sphere of day to day life revolves around plastic products. The used plastic gets accumulated on the Earth s surface and serves as a contaminant causing soil, land and water pollution. Waste plastic needs proper management and elimination from the biotic layer of the ecosystem. Most potent solution is conversion of plastic waste into a functional carbon material. Present work concentrates on upcycling of waste plastic into economically crucial material that is graphene oxide. The waste polythene bags were successfully converted to graphitic carbon which in turn serves as the base for the synthesis of graphene oxide using modified Hummers method.

Experimental: The analysis was made by studying the FTIR spectra, SEM images and XRD graphs. The FTIR confirms the presence of hydroxyl and carbonyl groups along with carbon carbon interaction. Surface morphology shows the porous and layered structure with an average particle size of 2.74 μm. X ray diffractogram illustrates the crystal structure of the graphene oxide and interlayer spacing.

Conclusions: With the characterisation results, the synthesis of graphene oxide from plastic waste was verified.

Keywords: Polythene bags, Carbon material, Graphene oxide, Modified Hummers method, Environment protection

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1. Introduction

Plastic contamination of ecosphere has become a common issue all around the Globe. Many developing countries are not only finding possible alternatives for plastic products but also researching extensively to reduce the plastic currently present in the environment as pollutant. Plastic products are sizeable components of automotive, construction, healthcare and electronic industries. Packaging industry forms one of the highest plastic consuming industry [1, 2]. Due to its increasing demand the production is increasing exponentially every year. The unique properties of plastic such as flexibility and durability have now become the disadvantage of plastic usage. The persistent plastic waste on Earth surface is the source of air, water and land pollution. Apart from this, polythene bags also cause major clogging of roadside drains in India resulting in stagnant polluted water in human colonies [2]. The dumped plastic somehow finds its way into water bodied, henceforth causing a negative impact on the marine environment. The presence of microscopic pieces of plastic termed as microplastic was reported by Surthy and Ramasamy [3] in the Vembanad Lake, Kerala, India. The results from the 10 samples of sediments collected from various latitudes of the lake reveals a range of microplastic concentration in the water body. The concentration ranges from 96 to 497 microplastics in per metre square of area with an average of 252.2 m^{-2}. Out of the all types of plastic, Low Density Polythene was found to be most abundant in the lake water. Marina beach, the longest stretched beach of India under investigation revealed the presence of microplastic of different shapes, colour and category of plastic [4]. These microplastic show detrimental effects in all strata of living organisms and thus an ideal solution is required. The methods of plastic control such as recycling, incineration and landfill were found best previously.

A recent study shows the release of chemicals and toxicants from incineration and landfill of plastic waste. The particulate emission, volatile organic compounds, polycyclic aromatic hydrocarbons are the by products of plastic combustion [5]. The mentioned paths along with Government policies are required for diminishing the plastic pollution. However, the methods fall short due to lack of awareness and public effort [6, 7]. A scientific step of creating a novel polymer with different sets of properties as of plastic would mark a difference [7].

Another alternative is conversion of plastic waste to fuel and functional carbon material. Pyrolysis and catalytic pyrolysis have been utilised for conversion of waste plastic to oil which further was experimented as transportation fuel [8, 9]. More recent study reveals the accomplished efforts of converting plastic waste into carbon material of various forms. Nanostructures such as spheres, fibres, tubes and sheets can be obtained by varying the synthesis conditions [10, 11]. Scientist have experimented the conversion of various types of plastics into diverse carbon materials. The experimental conditions play an impactful role in determining the characteristics of the material formed [12]. The most crucial allotrope of carbon is Graphene and its derivatives, it has gained great attention in the recent years since its extraction using scotch tape. In a review by Berktas and team, the importance of graphene is highlighted along with stating varied greener and cost effective raw materials for its production [13]. Graphene has exceptional conducting, mechanical and optical properties owing to which it attracts both psychists and chemists [14]. Graphene oxide has a similar layered structure as that of graphite and can be synthesised by introducing oxygen atoms in between graphitic layers. Graphene oxide can act as a precursor for the synthesis of graphene sheets namely reduced graphene oxide [15, 16].

Graphene oxide having properties similar to graphene is utilised in diverse fields such as an adsorbent for purification, as biosensors and as membranes [16]. Graphene has multiple synthesis procedures including mechanical exfoliation and chemical vapour deposition. However, these methods are restricted to small scale synthesis. For bulk scale chemical methods are preferred, one of them is use of strong oxidising agents under the Modified Hummers method [17, 18]. Graphene is obtained in the form of the graphene oxide using this method, which can further reduced to graphene. Hummer's method although being an efficient and trusted route still has some major defects. The use of $KClO_3$ along with HNO_3 was highly exothermic as well as liberated toxic gases such as NO_2, N_2O_4 and ClO_4. [19, 20]. The

Modified Hummers method employs $NaNO_3$, $KMnO_4$ and H_2SO_4 as oxidising agents by the production of MnO_4^- ions. Addition of H_2O_2 helps in the removal of residual manganate ions and termination of the reaction [20, 21]. The modified method is considerably effective, less toxic and highly applicable [22, 23].

In this study, we convert plastic waste in the form of Polyethylene (PE) into graphitic material which acts as the substrate for Graphene oxide synthesis. The method is environment friendly and highly efficient as well as less laborious.

2. Experimental procedure

2.1. Collection of waste polythene bags

Waste plastic bags were collected from various garbage dumping areas of our city Kanpur as illustrated in Fig. 1. The bags were washed to remove the unwanted dust, dirt and other degradable waste. The cleaned bags were allowed to sun dry until extra moisture is lost and then further steps were carried on.

2.2. Preparation of cleaned polythene bags

After washing and drying, the cleaned bags underwent a manual separation on the basis of colour, transparent bags were kept separately. The bags were then cut into small sized pieces manually using a paper cutter and scissor as shown in Fig. 2.

2.3. Conversion to graphite

The small chips of polythene were weighed and mixed with $Fe(NO_3)_3{\cdot}9H_2O$ in 1 : 3 ratio. To this 5 ml of ethanol and 45 ml of deionized water were added and stirred. Temperature range was maintained between 35–40 °C and rotation speed around 250 rpm. The mixture was stirred for 2 hours and then kept at rest for 24 hrs. The mixture was then heated inside a muffle furnace with a gradual rise of 10 °C per minute till the temperature reached

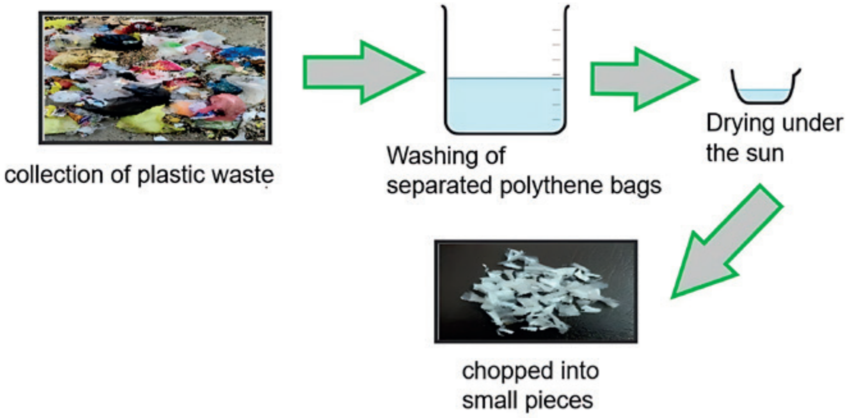

Fig. 1. Graphical Abstract showing the preparation of waste polythene

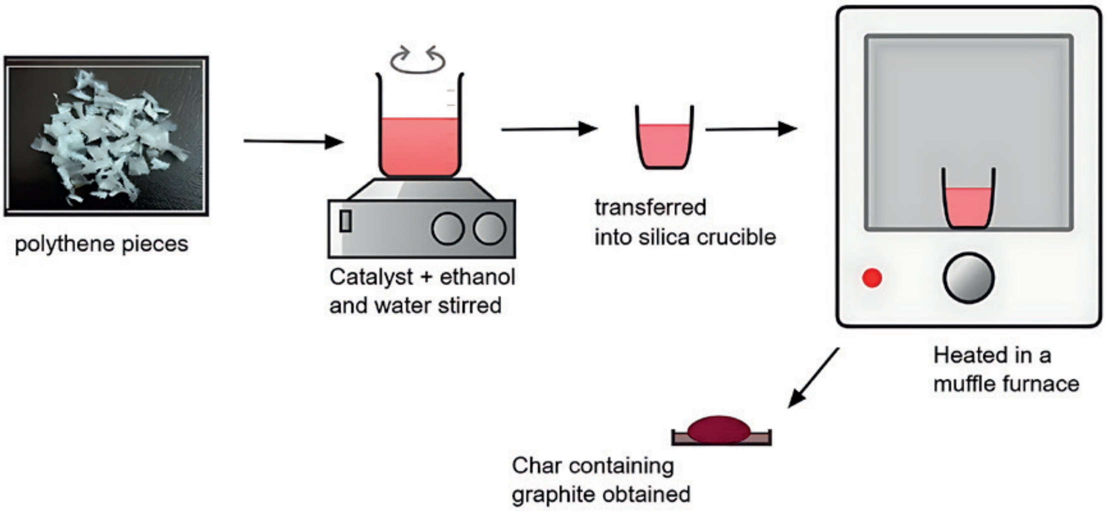

Fig. 2. Graphical Abstract show casing the formation the char

700 °C. The furnace was allowed to cool, then the char was obtained and weighed.

2.4. Graphene oxide synthesis

2.4.1. Acid wash of char

The 1g of char obtained in the previous step was acid washed to remove the catalyst and other impurities. The char was sonicated for an hour in 0.1 M HCl and kept for 24 hours in 0.1 M NaOH. The obtained graphitic carbon (GC) was used for further conversion to graphene oxide. This step helps in removal of all the excess ions of iron present in it.

2.4.2. Conversion to Graphene oxide

The Modified Hummers method employs $NaNO_3$ and $KMnO_4$ as the agents to introduce oxygen in between the layers of GC. The three components were mixed in a 1:1:5 ratio respectively as GC : $NaNO_3$: $KMnO_4$. The mixture was tardily added to 46.6 ml of H_2SO_4 and then stirred for an hour at a speed around 200 rpm as shown in Fig. 3.. After an hour the temperature was raise to 35–40 °C and again stirred for another half an hour. 5.3 ml of H_2O_2 and 80 ml of deionized water was added to the mixture continuing the stirring for another 30 minutes. On addition of H_2O_2, occurrence of yellowish brown colour confirms the formation of graphene oxide. On completion of the two hour process, the mixture was allowed to settle for a few minutes and then 10 ml HCl was added to balance the pH of the mixture. The mixture was centrifuged to obtain the graphene oxide (TGO) in semi liquid state which was allowed to dry at 60 °C in an oven for 24 hours to obtain a brownish and blackish powder. The TGO powder was further utilised for various characterisations.

3. Material characterisation

To confirm the formation of Graphene oxide the bonding was analysed by FTIR PerkinElmer Spectrum IR Version 10.6.1 with a range of 4000–400 $cm^{-1.}$ The surface morphology was studied using FE Scanning Electron Microscopy (FESEM). To further detect the structure of the graphene oxide formed X Ray diffractometer XPERT-PRO with K alpha value of 1.540598 was brought into use.

4. Results and discussions

4.1. FTIR

The FTIR graph of TGO Fig. 5 are the FTIR graphs obtained for graphite carbon and graphene oxide. Fig. 5 b shows the presence of carbon carbon

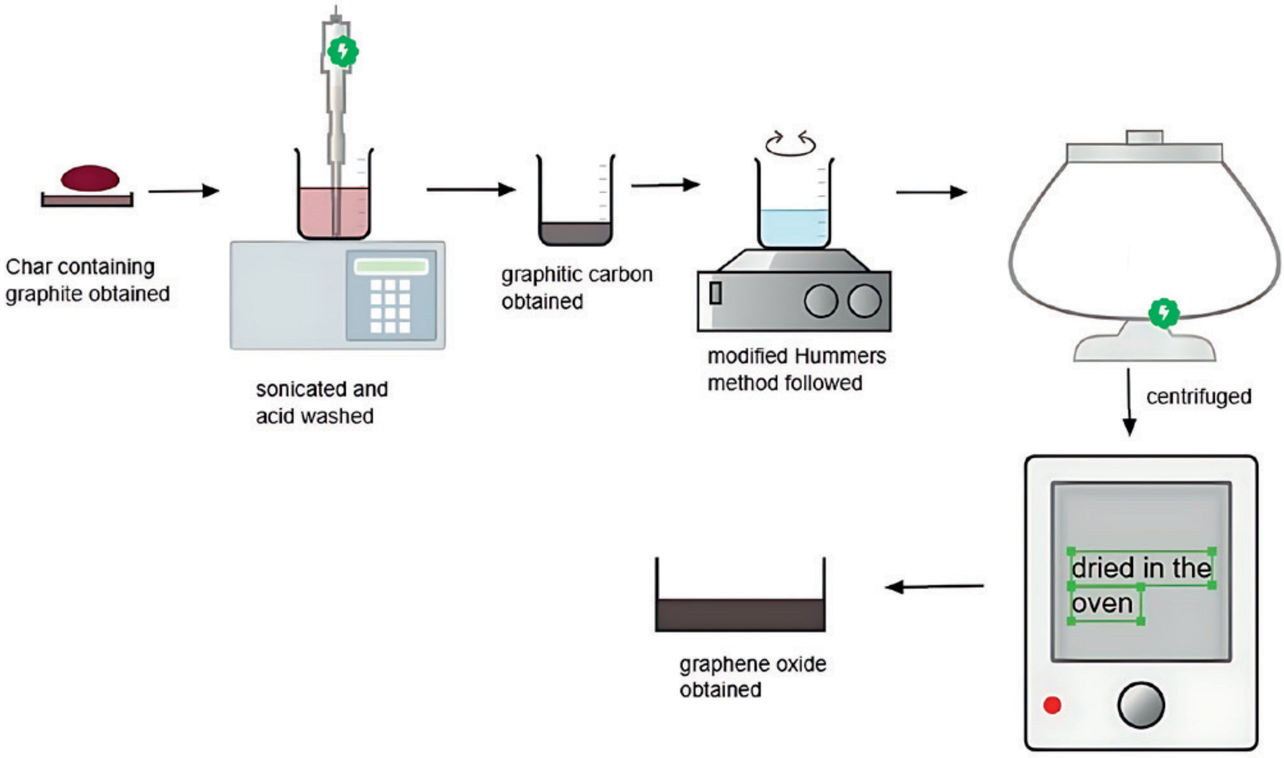

Fig. 3. Graphical Abstract for the preparation of graphene oxide

Fig. 4. Graphene oxide obtained from graphitic carbon

bond and carbon oxygen bonds to varying degrees. The spectra illustrates the vibrational group bands that include hydroxyl, carbonyl and aromatic double bond. The band around 3261.01 cm^{-1} is attributed to hydroxyl and carbonyl groups present in TGO. This confirms the addition of oxygen to the carbon atoms providing it the characteristic properties. The band in the region of 1644.94 cm^{-1} is due to the aromatic sp^2 carbon present in the form of C=O roups. The band around 1127.6 cm^{-1} and 1019.20 cm^{-1} confirms the presence of carbon oxygen stretching and C-O-C epoxy group in the TGO. The band at 974 cm^{-1} corresponds to the carbon carbon bending stretch. The obtained

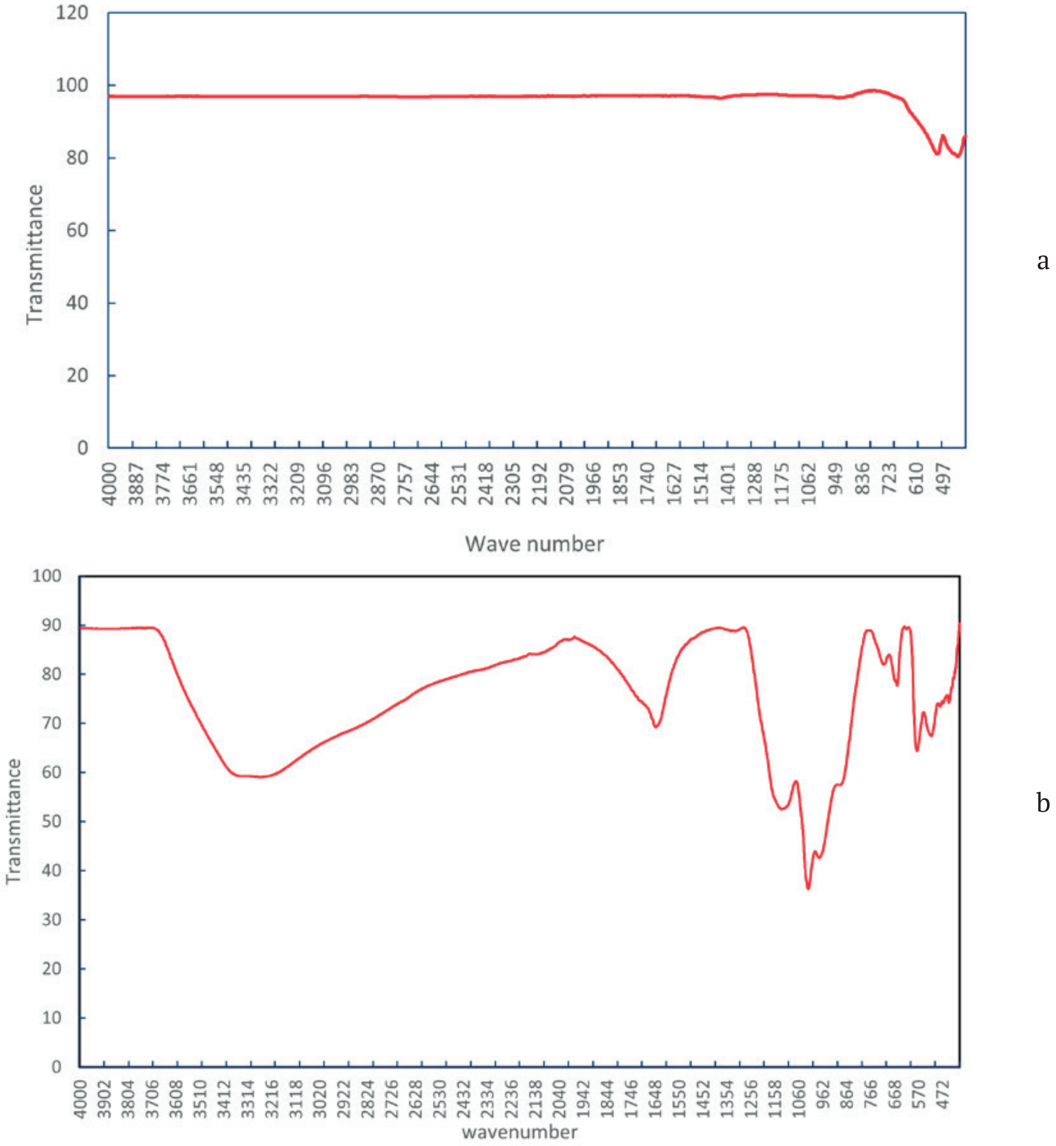

Fig. 5. The FTIR graph obtained for (a) graphitic carbon (b) graphene oxide synthesised

Table 1. Encloses the various peaks obtained from FTIR spectra for TGO and their analysis

S.no	(cm^{-1})	Bonding
	3261.01	-O-H stretching
	1644.94	-C=C-
	1127.6	-C=O- stretching
	1019.20	-C=O- stretching
	974.98	-C=C Bending

major peaks shown in Table 1 are consistent with the results for graphene oxide synthesized by the Hammer method in the previous literature [24, 25]. Thus, the graph is in favour of oxidation of graphitic carbon using Modified Hummers Method. Fig. 5a shows a linear graph confirming the synthesis of graphitic carbon. A linear FTIR is often attributed to carbon rich compounds. The results as per the study of literature [26] are assuring the carbon carbon linkage.

4.2. Surface studies

The SEM (Fig. 6) images reveal the flaky and layered structure of the synthesised graphene oxide. In the images (a) and (b) under low magnification show the aggregated crystaline forms with pores on the surface. A similar image is obtained in the previous studies along

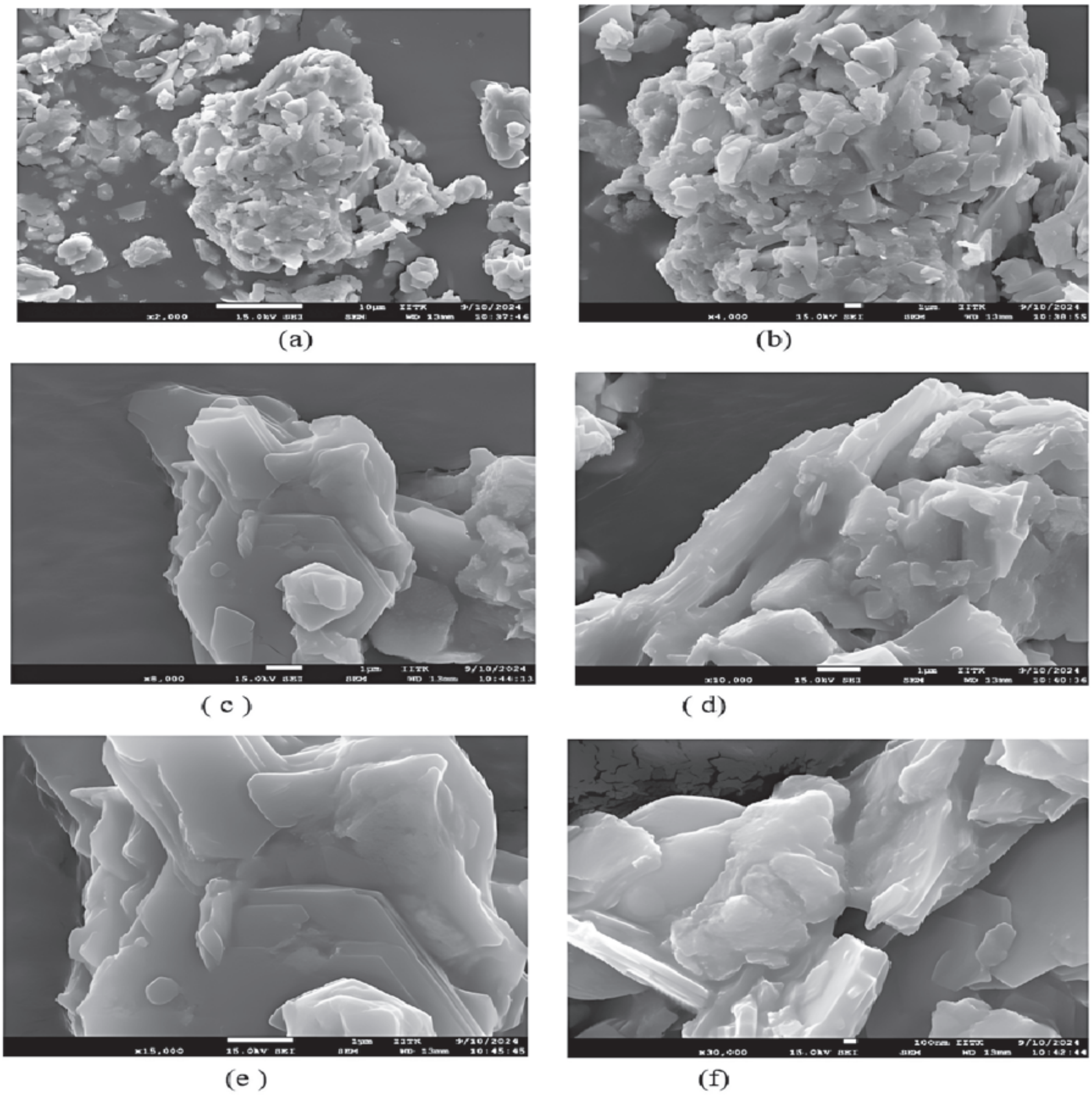
(a) (b) (c) (d) (e) (f)

Fig. 6. FESEM was analysised under different magnifications (a) the image obtained at 2000× (b) image under 4000× (c) image obtained at 8000× (d) image under 10,000× (e) image obtained under 15,000× (f) image under 30,000×

with a highly compact structure [27]. Image (c) is revealling a symetrical form with layered arranged sequentially giving it a high degree of orderness. Different magnifications reveal various forms of structures, the images obtained for TGO coincide with previous literature. A layered morphology can be noted in many studies and can thus be regarded as a significant feature of graphene oxide [28, 29]. The higher magnification also show case the presence of benzene ring like structure along with some hollow spaces. In images (e) and (f), the arrangement of layers can be seen vivdly. The average particle size 2.74 μm calculated from the image (b) asures the nanosclale particle formation. Particle size can influence the properties and application of graphene oxide [30]. The aggregation of particles and solubility depend upon the size of the grain.

4.3. X Ray Diffraction spectroscopy

XRD analysis was utilised to determine the crystal structure and inter layer distance of the graphene oxide. The peak observed at around 13° (as shown in Fig. 7(b)) is due the graphene oxide (110) with inter layer spacing calculated to be 0.337 nm using the Bragg' s Law:

$$n\lambda = 2d\sin\theta.$$

A small peak at around 13° (as shown in Fig. 7(b)) shows the presence of graphitic carbon which was utilised as the substrate for the synthesis of graphene oxide. The interlayer spacing shows us the degree of oxidation that has occurred between the layers of graphite. A minute variation in the values of the diffraction peaks can be attributed to the amount of oxygen molecules that have occupied the inter layer space of graphitic carbon.

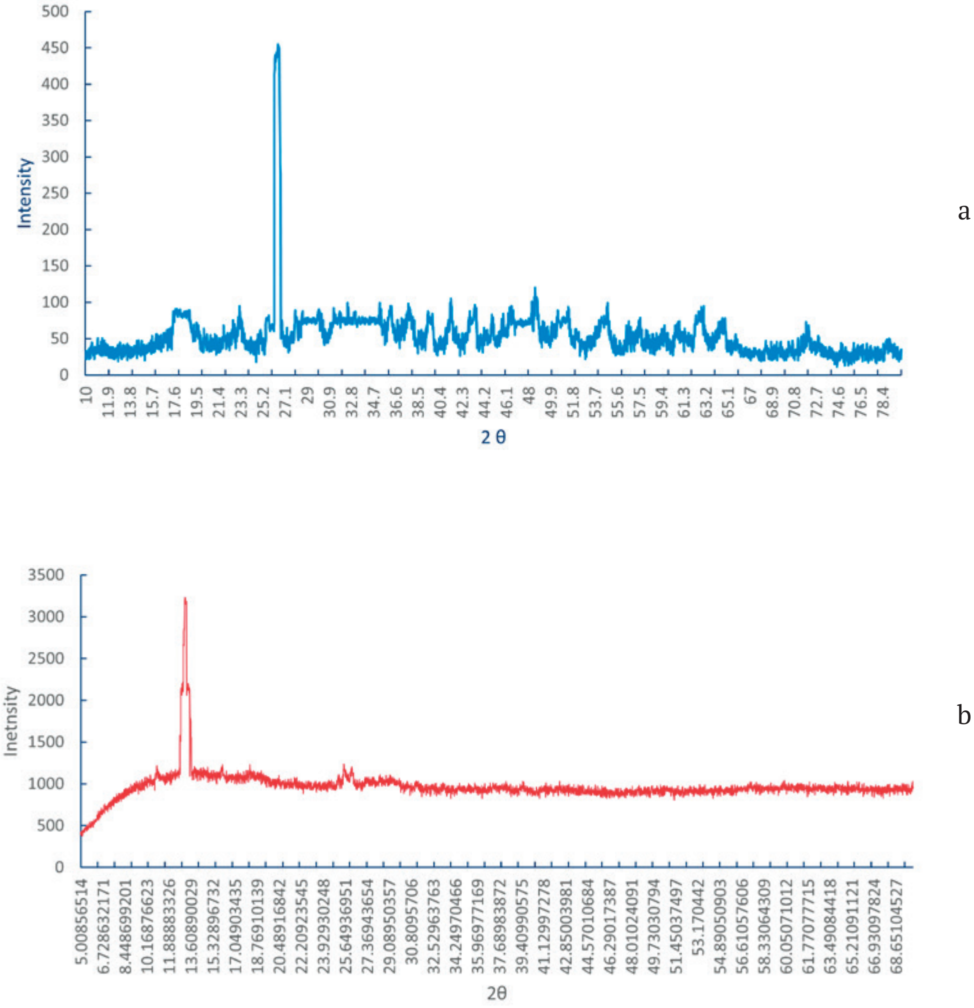

Fig. 7. Diffractogram of the (a) Graphitic carbon (b) graphene oxide synthesised using Modified Hummers method

The degree of oxidation can be a determining factor for the peak obtained, different reagents offer different extent of oxidation. As per the study of literature, synthesis conditions provide a set of results [31, 32]. Some studied also show variable results due the difference in nature of oxidising reagent used and peaks are in a scale of 10° to 12° for 2θ values [33, 34].

5. Conclusion

In the present experiment, Graphene oxide was successfully synthesised from the waste plastic that is Polyethylene using the Modified Hummers method. The graphitic carbon obtained from the pyrolysis act as the precursor of the oxidation method. Modified Hummers method applied is less tedious and time consuming along with satisfactory product formation. The characterisation including FTIR, SEM and XRD confirm the synthesised product is graphene oxide. The graphene oxide with particles of nano scale has various application in the fields of medicine and electronics. The research promotes the conversion waste to functional materials. Overall procedures are environment friendly and helps in the field of sustainable development.

Contributions of the authors

Shireen Shukla – Investigation, Methodology development, writing, Final conclusions; Meet Kamal – Supervision, Research concept, review and editing.

Conflict of interests

The authors declare that they have no known competing financial interests or personal relationships that could have influenced the work reported in this paper.

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