

WATER AND AIR PERMEABILITY OF MELT-BLOWN POLYPROPYLENE FILLED RECOVERED CARBON GRAPHENE OXIDE

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Abstract. This research used Modified Hummer's approach to synthesize Graphene Oxide (GO) from recovered carbon from waste tires and combine it with Polypropylene (PP) fabric. PP melt-blown cloth was employed for the basic medical masks due to its non-absorbent qualities. This environmentally friendly method contributes to the transformation of waste into wealth. The Sonication method was used to spread recovered carbon GO powder on a polypropylene fabric. The Raman analysis peak confirmed the effective synthesis of GO from a waste carbon tyre. with the value of the D band peak at 1367 cm^{-1} and the G band peak at 1593 cm^{-1} , respectively. The D band intensity ratio to the G band (ID/IG) is 0.87 represent multilayer GO were produced. FESEM images reveal that GO particles clustered together on the sample structure's surface, bumping pieces and forming coarse surfaces. Carbon (C) has a higher elemental content than oxygen (O), indicating that graphene oxide has a favored composition. While the coated PP sample shows that GO adhered well to the fabric fibres and demonstrated superior air and water permeability when compared to the control PP fabric. This result established that graphene oxide might be employed as a filler to enhance the filterability of PP fabric.

Keywords: graphene oxide, melt blown polypropylene, recovered carbon black, water permeability, air permeability

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Introduction

Waste carbon tyre was used as a carbon source for the production of graphene oxide. As most countries' legislation to ensure their environmentally safe disposal comes into force, the tire recycling industry is experiencing tremendous growth. Malaysia too has implemented regulations restricting or outright prohibiting the dumping of tyres in landfills, as part of the 1974 Environmental Quality Act (Act 127). The recycling of waste tyres into value-added goods increases economic leverage.

The big challenge of commercializing reproducible graphene oxide and producing it at a large-scale, high-quality material at a low cost is the things to look forward to now. Therefore, there is a need to develop graphene oxide by environmentally friendly approaches with a low cost of production [1]. Many researchers used Hummer's method to produce graphene oxide due to its cost-effective and scalable preparation [2]. Chen et al. [3] used Hummer's method to synthesize graphene oxide, which then fabricated polymer graphene nanocomposite.

GO was evaluated and reported in 2017 to inhibit bacteria *Bacillus subtilis* growth by rupturing the bacteria's thick cell walls [4-5], whereas (PP) has been successfully used to block droplets and microbes, and is thus used in surgical masks and protective clothing for health care workers [6]. PP fibre is always used for basic medical masks since it is non-absorbent and does not absorb moisture, making it ideal for use as a face mask to prevent droplets and viruses. However, research conducted in 2020 found that due to the pore size of the fabric and its non-antibacterial qualities, face masks cannot entirely stop the distribution of cough particles to the exterior face mask surface and environment. GO possesses antibacterial characteristics, inhibiting the growth of Gram-positive and Gram-negative bacteria. The PP fabric face masks were filled with GO in this study to enhance the permeability of the PP fabric face masks while incorporating the benefits of GO. The breathability of the fabric is closely related to the wearer's comfort, which can be determined using air permeability and water vapor permeability of the fabric [7]. As far as the authors are aware, no research on the breathability of the GO-PP fabric has been conducted. Thus, this project focused on improving the permeability properties of (PP) fabric by adding GO synthesized from recovered carbon of waste tyre for face mask materials. Hence, studies on air permeability and water vapour permeability were conducted in this research work.

Materials and Methods

Preparation of graphene oxide (GO). 23 ml of sulphuric acid (H_2SO_4) and 1.0 g of sodium nitrate ($NaNO_3$) were added and stirred for several minutes. Then, 1.0 % by weight of waste carbon tyre powder was added into the solution under stirring conditions. Next, the mixture was transferred to the ice bath, and the stirring process continued for 2 hours at a temperature below 10 °C. After that, 3.0 g of potassium permanganate ($KMnO_4$) was slowly added into the mixture, and the stirring process continued for another 1 hour. The ice bath was removed after 1 hour of stirring, and the mixture was left for 16 hours. After 16 hours, the mixture was heated up to 35 °C and stirred for another 1 hour before adding 46 ml of distilled water and continue stirring for another 2 hours and heated to temp 95 °C . 100 ml distilled water (H_2O) were added followed by 10 ml hydrogen peroxide (H_2O_2) into the mixture and stirred for 1 hour to eliminate the excess of $KMnO_4$. The mixture was left cool to room

temperature. After that, 10 ml of hydrochloric acid (HCl) and 30 ml of deionized water (DIW) were added into the solution and centrifuged for 10 minutes at 8500 rpm. The supernatant was decanted away and rewashed with the residuals HCl and DIW three times. Then, washed GO solution was dried in the oven at 70 °C for 24 hours [8-9].

Preparation of GO-PP fabric. 0.01 wt% GO and a non-woven (PP) melt blown with a mass density of 9.623 mg/cm² were used in this part. The PP fabric was immersed in boiled deionized water at 100° C for 30 min for scouring treatment. At the same time, GO solutions were sonicated for 1 hour [10]. Then, the fabric was immersed into the dispersed GO solution and continued sonicated for 30 min. Following that, the fabric-coated GO was rinsed with distilled water and dried for 2 hours in an oven set at 80 °C. After two hours in the oven, the PP fabric-coated GO was washed with deionized water and ethanol to remove any remaining GO and contaminants. The coated PP fabric (GO-PP fabric) was then dried overnight at 35 °C in a vacuum oven.

Weight analysis of PP coated GO. The weight of coated GO on the PP fabric was calculated from the difference in weight before and after attachment. Neat PP fabric was weighed using weight balance before mixing with the GO process. Coated PP-GO again was weighed after scouring, mixing with GO, and drying process.

Raman spectroscopy analysis. The confirmation analysis for GO was characterized by Raman spectroscopy LabRAM HR Evo, Horiba. The structural properties of the GO were characterized with a Raman spectrometer based on the D and G band information. Raman images have characterized the appearance in 3-D for graphite whereas in 2-D for graphene oxide, which is useful to determine the D and G bands assigned bonding states of sp² and sp³.

Field emission scanning electron microscopy (FESEM). The surface morphology of the GO powder and PP fabric-coated GO was characterized using a field emission scanning electron microscope (FESEM) (Hitachi S-4800).

Water vapour permeability and air permeability tests. The SDL Atlas vapor permeability tester was used to determine the water vapour permeability of control PP and GO-PP textiles. The experimental work followed the ISO 8096 standard procedure. The permeability of all samples to water vapour was determined using equation (1):

Water vapor permeability:

$$(WVP) = \frac{24 M}{At} \quad (1)$$

where M is the water vapor loss (g) in expose period t (h), and A is an area of the sample (m²).

The air permeability of the control PP and GO-PP fabrics was conducted using MESDAN Air-Tronic based on the ASTM D737–18 standard method. The test area of each sample was 10 cm², with the amount of pressure being about 200 Pa.

Results & Discussion

Weight analysis. Figure 1 refers to the weight of the fabric before and after coated with GO. The result shows an increase in coated fabric weight after 24 hours of drying. This data proves that GO was successfully coated on PP fabric. The increment of weight is about 18.34% of the fabric. Results are also supported with the FESEM image in Figure. 3(b) and 3(c). Attachment of Graphene oxide is supported by a statement from Thema et al. [11] where graphene oxide (GO) has oxygen-containing functional groups, making it more compatible and uniformly dispersed in polymer matrices. This condition also contributes to the stronger interactions with the polymer matrix compared to graphene. Ullah et al. [12] stated PP fabric is hydrophobic; hence it is naturally repelling water. However, the scouring treatment is applied to the hydrophobic PP fabric helps the GO particles to adhere to the fabric. Scouring is the process of removing natural and external contaminants from fabrics to prepare them for the next step of procedures. This treatment makes the fabric hydrophilic, allowing it to absorb chemicals and dyes adequately.

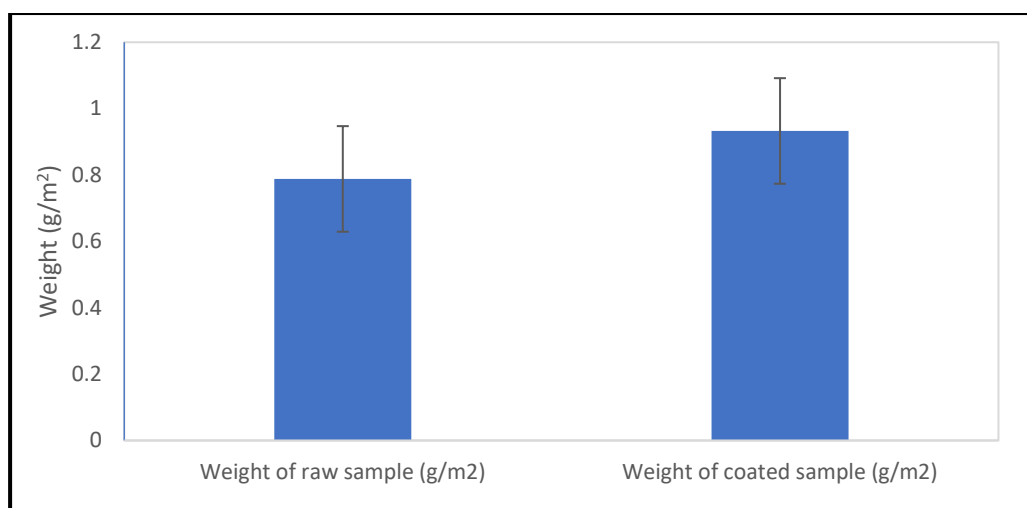
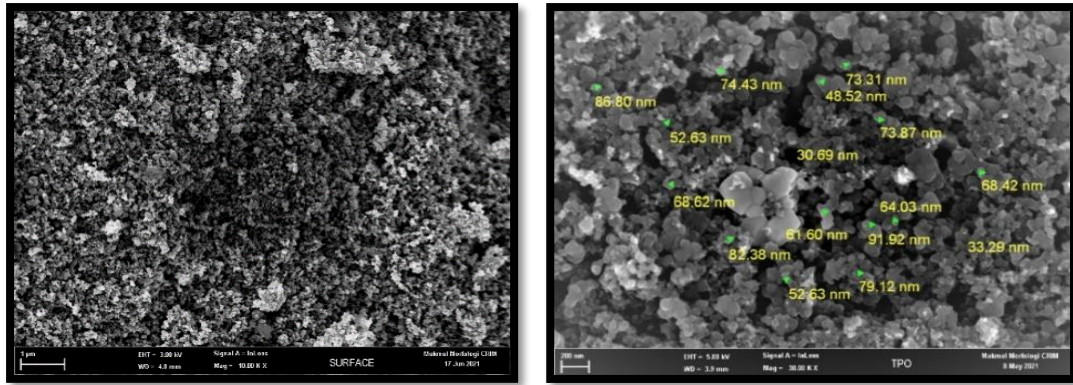
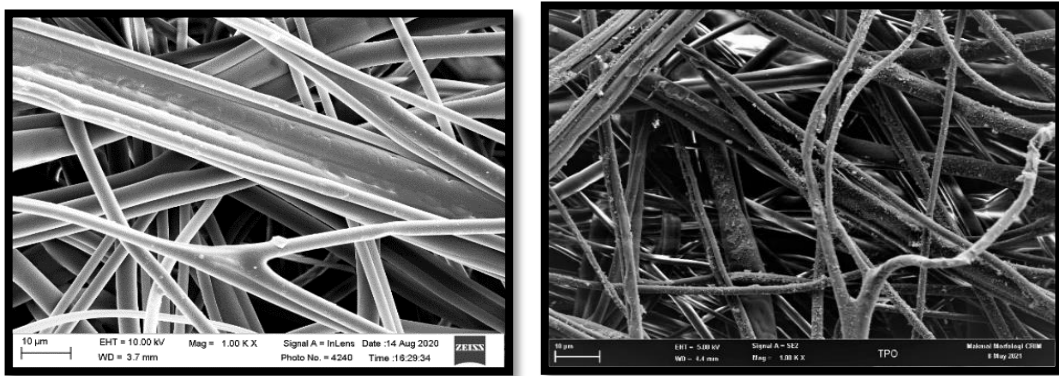


Figure 1. Polypropylene fabric before and after the dispersion process

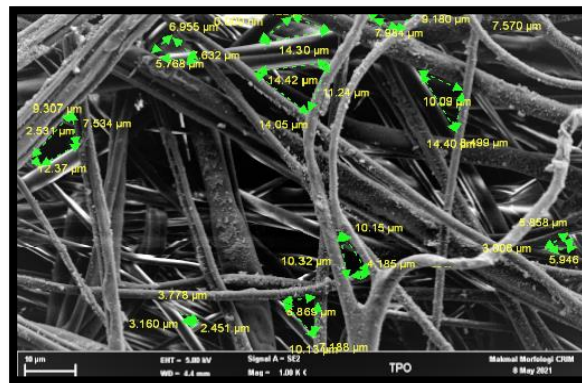
Morphological analysis. The FESEM images in Figures 2(a) and (b) show graphene oxide powder made from recovered carbon from a waste carbon tyre. According to Figure 2(b), the pictures of GO reveal a circular structure trailed together on the sample's surface. Additionally, it exhibits bumping pieces of small particles and a rough surface with a diameter of between 30 and 80 nm. Figure 3a depicts an uncoated PP fabric, while Figures 3(b) and (c) depict GO added to a melt-blown polypropylene fabric. There were significant differences in image quality between uncoated and coated PP – GO. This conclusion was further corroborated by the results of the weight analysis in Figure 1 and the Raman analysis of coated fabric in Figure 6, which showed a rise in weight after coating and a peak representing the GO structure in the PP-GO Raman spectrum.



(a) (b)
Figure 2. The SEM micrograph of graphene oxide obtained a) 10k x magnification and b) 30k x magnification.



(a) (b)



(c)

Figure 3. The SEM micrograph of (a) Polypropylene fabric, (b) Coated graphene oxide (GO) on the Polypropylene fabric and (c) Coated PP with fiber measurement size

Energy Dispersive X-Ray Spectrometer (EDX) was used to measure the elemental analysis of the composites [2]. Figure 4 shows the result of EDX of the coated sample confirmed the existence of Carbon and Oxygen elements on the coated fiber representing the GO successfully adhering to the fabric.

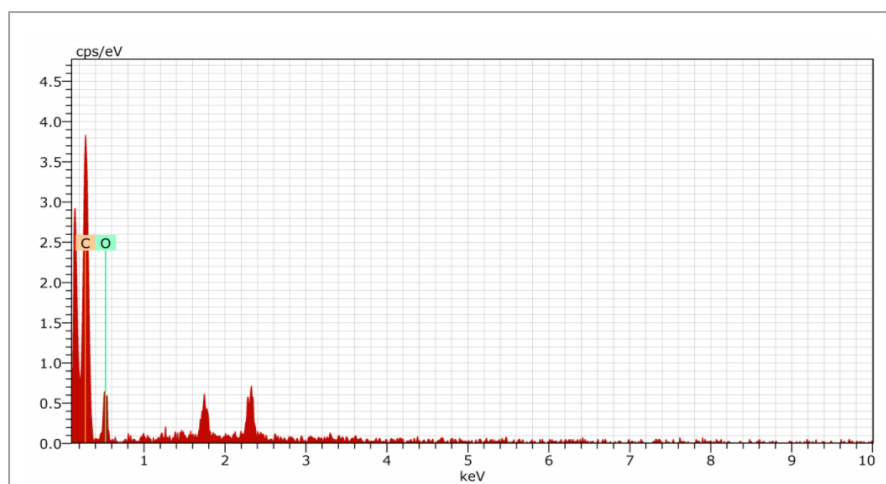


Figure 4. The EDX image of graphene oxide

The carbon and oxygen content in graphene oxide is 77.26 % and 22.40 % respectively, representing GO. Wang et al. [13], stated that graphene oxide is easy to disperse in water and other organic solvents and in different matrices due to oxygen functionalities. This is an important property of GO to act as a filler in composite materials for improving their mechanical properties with good adhesion between filler and matrix.

Raman spectroscopy analysis. Raman spectroscopy explains the conversion of recovered carbon to GO. Raman analyses are essentially identical to the characteristic peak of graphene oxide by two peaks around 1300 cm^{-1} and 1500 cm^{-1} (D band and G band) [14].

Table 1. Raman Spectroscopy

Sample	D peak (cm^{-1})	G peak (cm^{-1})	I_D/I_G ratio
GO - RCW	1367	1593	0.86
GO - PP	1382	1594	0.87

Table 1 shows relevant data of the Raman spectrum, such as the wavenumbers of D and G peaks and I_D/I_G ratios. The most prominent feature in the graphite spectrum is an acute G peak at around 1593 cm^{-1} corresponding to the vibrations in the graphite structure plane and a peak at around 1367 cm^{-1} corresponding to the disorder band, D.

In this work, the values of the I_D/I_G ratios were calculated using intensities value. From two samples, Graphene oxide (GO - RCW) and (GO - PP), the I_D/I_G ratio presented by GO RCW is 0.86, and the GO-PP is 0.87. The I_D/I_G ratio measures the degree of disorder and is inversely proportional to the mean size of the sp^2 clusters present in the material. Thus, the higher the I_D/I_G ratio in carbon materials, the greater the structural disorder. The GO reduction process can be expressed in Raman spectra by changes in the relative intensity of the D and G bands. As already mentioned, the I_D/I_G ratios are higher for the GO than GO-PP, which suggests that new (or more) graphite domains are formed, and the number of sp^2 clusters is

increased during the reduction. This indicates an efficient reduction, collaborating with the results obtained by the surface energy characterizations. In this research, the value of (I_D/I_G), represents the multilayer of GO. This value also indicated the graphene oxide with higher degree graphitization, and less defect produced [15-16].

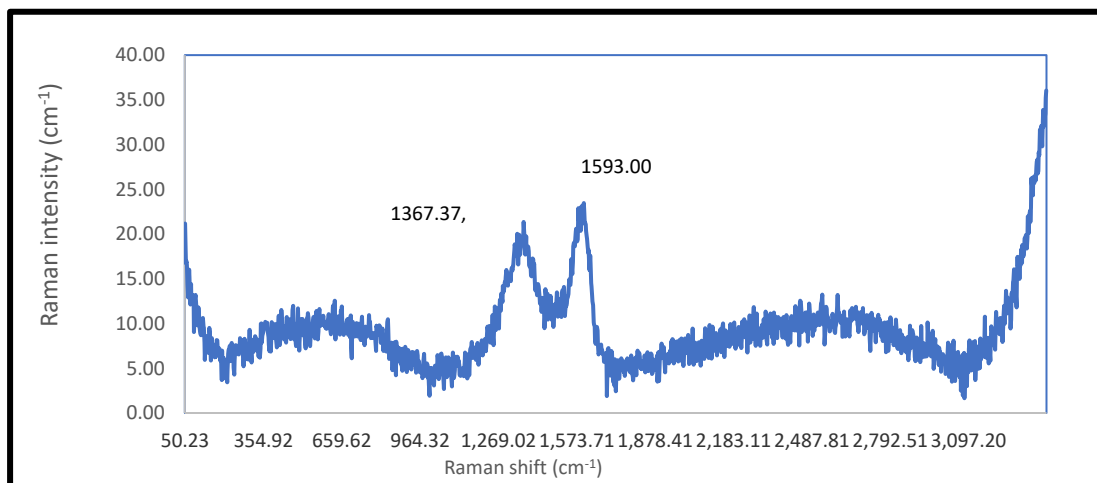


Figure 5. The Raman spectra of graphene oxide from carbon waste tyre

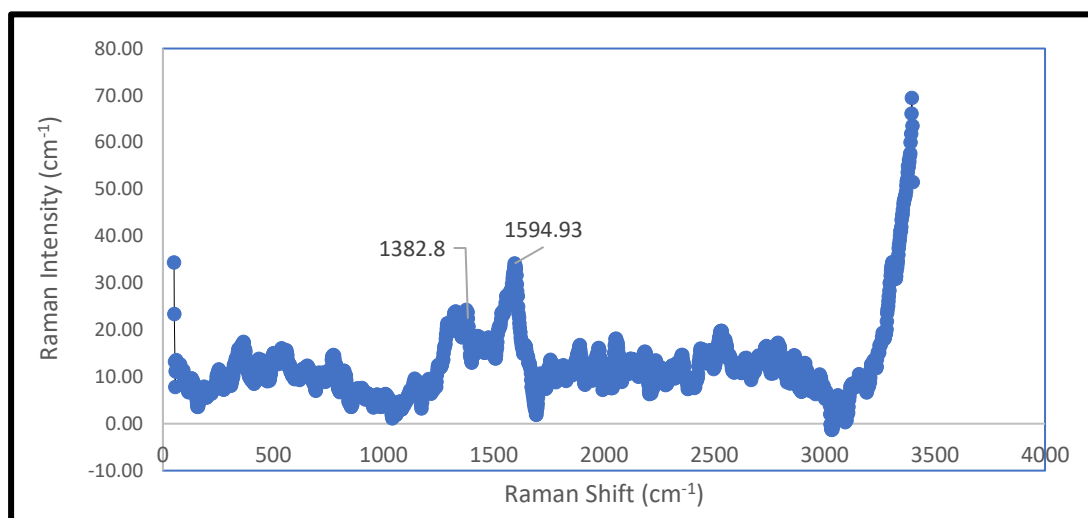


Figure 6. Raman Analysis result of GO PP Fabric.

Based on Figure 5, the Raman spectra of the obtained GO display two peaks at 1367 cm^{-1} and 1593 cm^{-1} , corresponding to the D-band and G-band. Results show that GO was successfully synthesized from recovered carbon of waste tyre (RCW). I_D/I_G ratios can be determined by the heights or areas of the peaks. Some groups use the Lorentzian function to calculate the ratio using the peak intensities, while others use the Gaussian function to make the relation between peak areas. In the latter case, the widening of the peak D is related to the distribution of agglomerates with different orders and dimensions; thus, the information about the aromatic rings is less distorted in the maximum intensity than in width, which depends on the disorder [17].

Figure 6 shows the same pattern of Raman peak for coated GO onto the PP fabric. The attachment of GO was successfully proven by the D band and G band peak of Raman spectroscopy from the sample of PP-coated GO fabric.

Water vapour permeability. The current study has investigated water vapor permeability and air permeability of the sample to evaluate the breathability of the GO-PP fabric. Vapour permeability is a material's ability to allow vapour such as water vapour or any gas to pass through it. Gao et al. [18] stated that it is generally believed that a higher rate of water vapour transmission can increase wear comfort because it keeps the mask dry and clean. To achieve a higher water vapor transmission rate, the fabric must have lower water vapour permeability [19]. The testing took 5 hours to be completed because the maximum duration of wearing a face mask is approximately 6 hours to prevent wet, soiled, splashed, damaged, or difficult to breathe through. Figure. 7 shows the water vapor permeability of control PP fabric is approximately $798 \text{ g h}^{-1}\text{m}^{-2}$. Although the control PP is hydrophobic, pores in the fabric allow the diffusion of water vapor through the fabric.

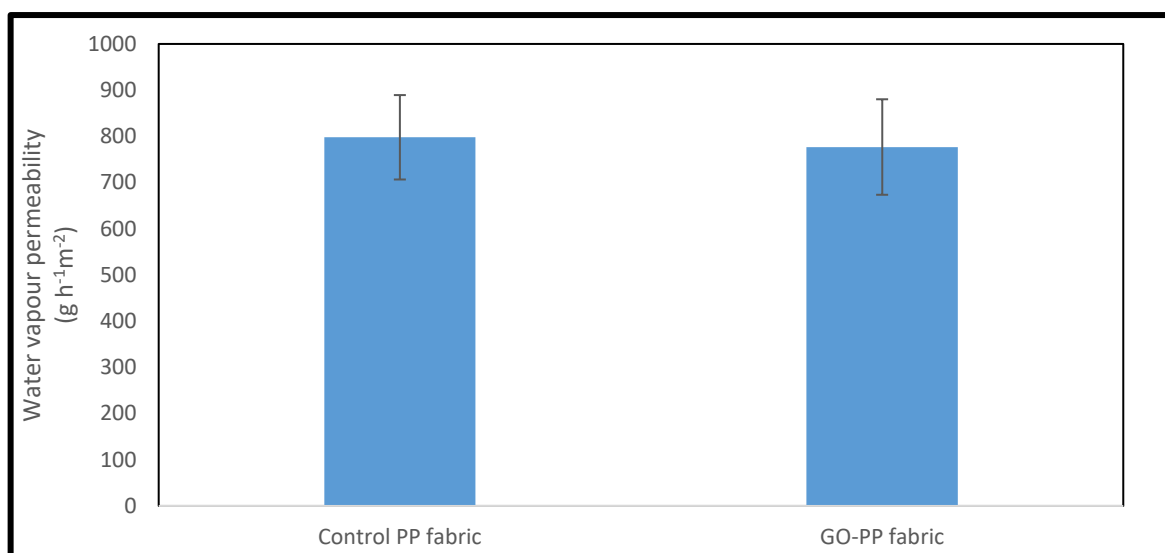


Figure 7. Water vapor permeability of control PP and GO-PP fabric.

The diffusion of water vapour through the hydrophobic PP fabric was also reported by another study [19]. A slight decrement in water vapour permeability was observed for the GO-PP fabric. After 5 hours of testing, the coated PP fabric showed a slight decrease in water vapour permeability of about 2.6% compared to the control PP fabric. GO obtained will be filled in existing PP fabric face masks to enhance the effectiveness of PP fabric face masks in blocking microbes. Kumar et al. [20] stated the first reported antibacterial activity of GO and reduced graphene oxide (rGO) were in 2010 revealed the inhibition of E. coli bacterial growth. The addition of GO with 0.01wt% has reduced 2.6% permeability properties of GO. Understandably, the presence of GO in PP fabric can slow down the transmission of water vapour through the fabric. However, the reduced water vapor permeability rate does not affect the GO-PP fabric's breathability. The GO-PP fabric continues to exceed the facemask material minimum, the minimal water vapour permeability for facemask cloth was estimated to be approximately $150 \text{ g h}^{-1}\text{m}^{-2}$.

Air Permeability. Permeability of the air is a critical indication of human respiration [19]. Air permeability was measured in the current investigation to ascertain the amount of air moving through the control PP and GO-PP textiles.

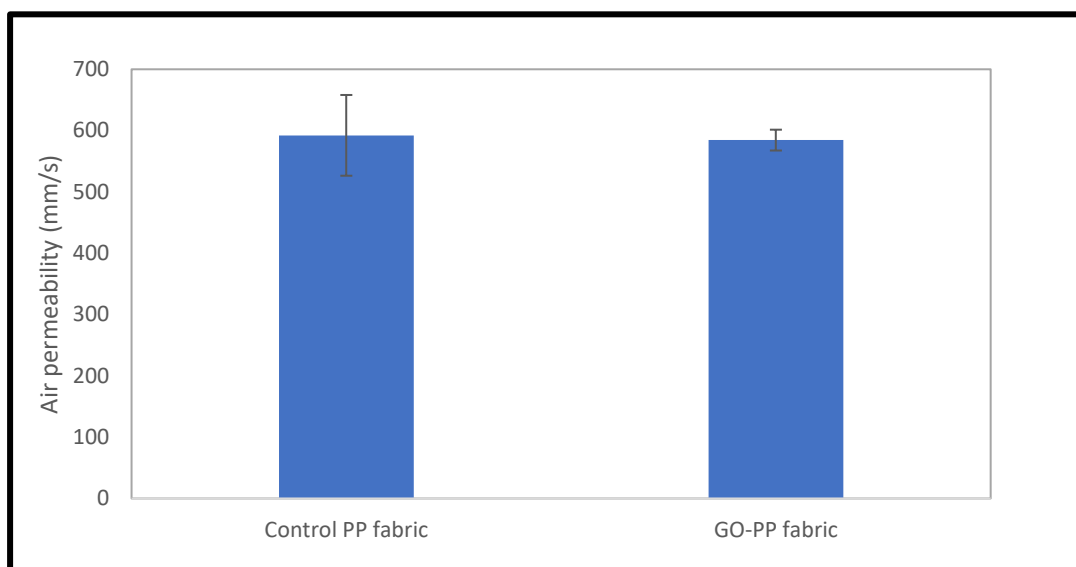


Figure 8. Air permeability of control PP and GO-PP fabric.

The air permeability of a control PP fabric and a GO-PP fabric is depicted in Figure 8. The GO-PP fabric had a 13% lower air permeability than the control PP fabric, following the same pattern as the water vapor permeability. The deposition of GO on the PP fabric surfaces decreased the cloth's airflow. While the GO-PP fabric has a decreased air permeability, it is appropriate for use as a facemask material. It shows that oxygen on the GO structure helps improve GO and PP fabric compatibility. According to Moradi & Ahmadi [19], the lowest air permeability value for a face mask material was around 27.2 cfm (138.07 mm/s). The air permeability of the GO-PP fabric used in the investigation was approximately 584 mm/s.

Conclusion

The Modified Hummers method successfully generated graphene oxide from recovered carbon from scrap tyres. FESEM pictures reveal that graphene oxide is composed of spherical particles stalked together, coarse surfaces, and flake uniformly. Carbon, C, has a higher elemental composition than oxygen, O, indicating the composition of graphene oxide. Raman spectroscopy confirmed the formation of graphene oxide by identifying the D- and G-band peaks at 1367.37 cm^{-1} and 1593.8 cm^{-1} , respectively. FESEM photos demonstrate that GO was successfully disseminated and attached to the polypropylene fabric, as demonstrated by the emergence of D and G band peaks on the PP – GO sample in the Raman spectra. Additionally, the results indicated that the presence of GO on the PP fabric results in a high level of air and water permeability, which meets industry standards for facemask materials.

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Author Contributions

All authors contributed toward data analysis, drafting and critically revising the paper and agree to be accountable for all aspects of the work.

Disclosure of Conflict of Interest

The authors have no disclosures to declare

Compliance with Ethical Standards

Not Applicable

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