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Synthesis and Characterization of Graphene Oxide from Agricultural Waste: Cocoa Pod (*Theobroma Cacao*)

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Abstract: the study based on synthesis and characterization of graphene oxide from cocoa pod waste sample collected from Imesi-Ile in Ekiti State, Southern-west Nigeria. The raw cocoa pod waste was carbonized and synthesize using modified Hummer's method. The raw cocoa pod, the graphene form and graphene oxide obtained were characterized using X-ray Diffraction (XRD); Fourier Transform Infrared Spectrometry (FT-IR) and Scanning Electron Microscopy (SEM). The percentage yield of graphite powder produced from the raw cocoa powder (agriculture waste) was 1.290 g, which corresponded to 21.5 % yield. It can be inferred from this that irrespective of the percentage yield, it is independent of the graphene oxide formed. The SEM analysis shows well-developed agglomeration growth with significant increasing in grain size form raw cocoa powder to graphene oxide. The FT-IR analysis of graphene oxide shows availability of O-H stretch at (3772.9 cm^{-1}) with C-O-C at (1118.2 cm^{-1}) which confirms the presence of hydroxyl, epoxy, and carboxylic groups upon oxidation of graphene. While for graphene the assigned peaks are located at (3205, 1632, 2117 and 1632cm^{-1}) which was corresponded to (OH, C=C, C≡C and C-O) respectively. The XRD analysis reveals a wide range scan, the graphene oxide peaks were closed to 2θ =25 and 45°, indicating the disorderliness of carbon, while the graphene XRD results shows four peaks at (26.5), (30.4), (32.6) and (42.1). As established from the results, the cocoa pod waste has excellent prospective potential for producing high valuable adsorbent product for reducing environmental pollution.

1. Introduction

Nanotechnology has opened an opportunity for a new economically viable material (Tohamy *et al.*, 2020; Osherov *et al.*, 2023; Aldwayyan *et al.*, 2013;). Carbonaceous nano-materials such as carbon nanotubes (CNTs), graphene (G) and graphene oxide (GO) have attended great interest in the scientific community, in the last few years, because of their versatile properties, exceptional structures (Alghyamah *et al.*, 2020). Also, due to their wide range of envisaged applications across several fields (Somanathan *et al.*, 2015). Among these carbonaceous nano-materials, such as graphene and grapheme oxide have displayed excellent electronic and mechanical properties, thermal conductivity (Balandin *et al.*, 2008), good dispersion performance and strength with high surface area and aspect ratio. Graphene is an extremely two dimensional (2D) atomically thin layers of carbon-

based material with honey comb hexagonal lattice, consisting of a network of sp^2-bonded carbon atoms and which is stable in its free state. Graphene's unique properties make it an excellent candidate for modifying composite materials' structure and unlocking various applications (Shahnaz *et al.*, 2024; Song *et al.*, 2011).

The oxidized derivative of graphene is graphene oxide (GO), which contains reactive oxygen functional groups such as epoxide, carboxyl groups, phenol and hydroxyl groups at the sheet edges. These functional groups lead to the negative charge, hydrophilicity and easy dispersion of graphene oxide in aqueous solutions (Deng *et al.*, 2013). This makes graphene oxide a good candidate for the adsorption of different pollutants by degradation.

Several synthetic methods have been used over the years in the preparation of GO, these methods includes; physical method such as (micromechanical exfoliation or scotch tape method from graphite and epitaxial growth on electrically insulating surface); chemical method such as (chemical vapor deposition (CVD) from degradation of organic compound on metal surface) and bio-mass based method such as (carbonization of biomass and agricultural waste materials) (Eissa *et al.*, 2020; Aro-Mahmudunnabi *et al.*, 2020; Aro-Modiu *et al.*, 2019)

Over the years, the management of agriculture waste materials such as cocoa pod, sugarcane, rice husk, and wood wastes among others have posed a very serious environmental health hazard due to its vast volume and slow degradation rate (Aro-Modiu *et al.*, 2019). However, recent research indicates that agricultural waste materials are center of attraction due to their abundance, need for recycling and source of carbon. Also, these waste materials can be easily converted into useful carbonaceous nano-materials such as graphite (G) and graphene oxide (GO), which can be utilized in various fields of science (Taniya *et al.*, 2017), with variety of environmental potential applications such as the adsorption of organic pollutants and removal of heavy metals from water (Hafez *et al.*, 2024). Agricultural waste materials have been classified as the simple, cheap and eco-friendly sources for the synthesis and preparation of grapheme and its oxides (Chen *et al.*, 2016). Therefore, this study seek to chemically synthesize and characterized graphene and graphene oxide produced from agricultural waste such as cocoa-pod waste using modified Hummers' method.

2. Materials and Methods

2.1 Sample Collection and Identification

The Cocoa pod was collected from a local farm in Imesi ile, Ekiti, South-west, Nigeria. The collected sample was later identified at the department of physical biology university of Ilorin.

2.2 Sample and Treatment and Preparation

The cocoa pod was washed thoroughly multiple times with a distilled water to remove all the traces of impurities on it. After washing, the cocoa pod was left to open air dry in the laboratory (Moharram *et al.*, 2015; Ouattara *et al.*, 2023). The collected sample was grounded using a ceramic metal and pestle and later ground using local grounding machine located at Mandate- Market, Adewole, Ilorin, Nigeria.

2.3 Carbonization Study

6 g of grounded cocoa pod sample with 34.53 g of the crucible was weighed and carbonized in muffle furnace at varied temperature between 300- 650ºC for 3hours. The weighed Cocoa pod sample was wrapped into aluminium foil to ensure complete deoxygenated condition during carbonization. The wrapped aluminium foil was placed into a muffle furnace. The muffle furnace was set at temperatures for varying length of times as such; 300 ºC, 350 ºC, 400 ºC, 450 ºC, 500 ºC, 600 ºC, 650 ºC, 700 ºC, 750 ºC, 800 ºC for 3 hours each (Osman *et al*.2014). The graphene obtained for each temperature and time range was weighed on the digital electronic weighing balance and a new carbonized cocoa pod weight was recorded.

2.4 Graphene Oxide Synthesis

Synthesis of graphene oxide (GO) from carbonized cocoa pod. The GO was prepared according to the modified Hummer method (Torres *et al*.2021). In detail, 5g of graphite from carbonized cocoa pod and 2.5g of $NaNO_3$ were mixed with 108 mL,H_2SO_4 and 12 mL H_3PO_4 and stirred in an ice bath for 10min. Next, 15g of $KMnO_4$ were slowly added so that the temperature of the mixture remained below 5 ºC. The suspension was then reacted for 2 hours in an ace bath and stirred for 60 mins before again being stirred in a 40 ºC water bath for 60min. The temperature of the mixture was adjusted to a constant 98 ºC for 60 mins while water was added continuously. The suspension was diluted to a volume of 400 mL by adding deionized water. After a 5-minute interval, 15 mL of H_2O_2 was added. The resulting reaction mixture was centrifuged, and the precipitate was washed repeatedly with deionized water and a 5% HCl solution. The product was then dried in an oven at 60^{o}C.

2.5 Sample Characterization Study

The raw cocoa pod powder, graphene powder and synthesized graphene oxide was characterized for elemental constituent using X-ray florescence (ThermoScientific XTRA-EFX) identification of the crystalline substance using X-ray powder diffraction (Rigaku XRD Miniflex-600); micro- structures analysis was carried out using Scanning Electron Spectroscopy (PhenomWorld-SEM Pro-X); functional groups identification using Fourier Transform Infrared spectroscopy (Agilent-ATR Calgary 620 FTIR). The characterization was source for at a Sister University, Kastina.

3. Result and Discussions

3.1 Percentage Yield Result

The image below shows the metamorphosis of cocoa pod to graphene oxide. At initial stage the brown colour of ground cocoa pod change to black after carbonization at 650 ºC after 3 hours. The oxidized graphene maintains the black colour as shown in (**Figure 1**)

The percentage yield of graphite powder produced from the carbonized raw cocoa powder (agriculture waste) was presented in Table 1. Starting with 6 g of the cocoa powder, 1.290 g of graphite which corresponded to 21.5 % yield which is a very fair result, was obtained. It can be

inferred from this that, irrespective of the percentage yield of the produced carbonized powder, it is independent of the percentage yield of the graphene oxide formed. Also, it can be observed that production of graphene powder from cocoa pod waste does not have sufficient carbon content.

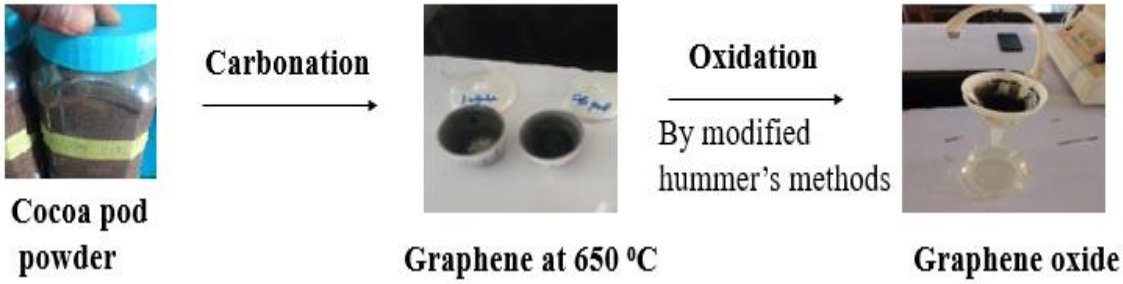

Figure 1. The Pictorial Image of Synthesized Graphene Oxide

Table 1. Percentage Yield Table of Cocoa pod sample.

Sample	Weight of Crucible (g)	Weight of Sample (g)	Weight of Crucible + Sample (g)	Weight of Crucible + Sample after ashing (g)	Weight of Sample after ashing (g)	Percentage yield (%)
Cocoa Pod	28.53	6.00	34.53	29.82	1.29	21.5

3.2 SEM Analysis

SEM micrographs (**Figures 2a-c**) revealed slightly irregular spherical particles with a high degree of agglomeration and small to large pores. Raw cocoa powder, graphene and graphene oxide size analysis was conducted using the SEM images, as depicted in the (**Figures 2a-c**) which shows well-developed agglomeration growth with significant increasing in grain size form raw cocoa powder to graphene oxide. The possible reason for increasing the grain size is that more crystal nucleus was formed with increase in carbon content. It is well known that raw cocoa powder contain carbon with small amount of donor impurities yields the material being fine-grained in the form of irregular shapes (Badapanda *et al.* 2012), as presented in the image (**Figure 2a**). In the case of graphene and graphene oxide, the SEM images shows a similar microstructure of the raw cocoa ash before oxidation, which is characterized by unpredictable shapes with protuberances and amassed surface. This is probably because of the combustion procedure, which would wipe out the water and carbon dioxide in the materials. The micro-structure of graphene and graphene oxide are presented in (**Figure. 2b, c**), where the flakes can be observed in a few-layer mass agglomeration. The SEM of graphene and graphene oxide also shows a flaky irregular shape of the particle made up of the graphene synthesis with many cavities and pores. This observation is similar comparable to the report by (Kumar *et al.*, 2017; Xu *et al.*, 2011). The existence of the cavity is more pronounced in with many polygonal shapes. This pore sizes exhibited has made the product a potential candidate for adsorption processes.

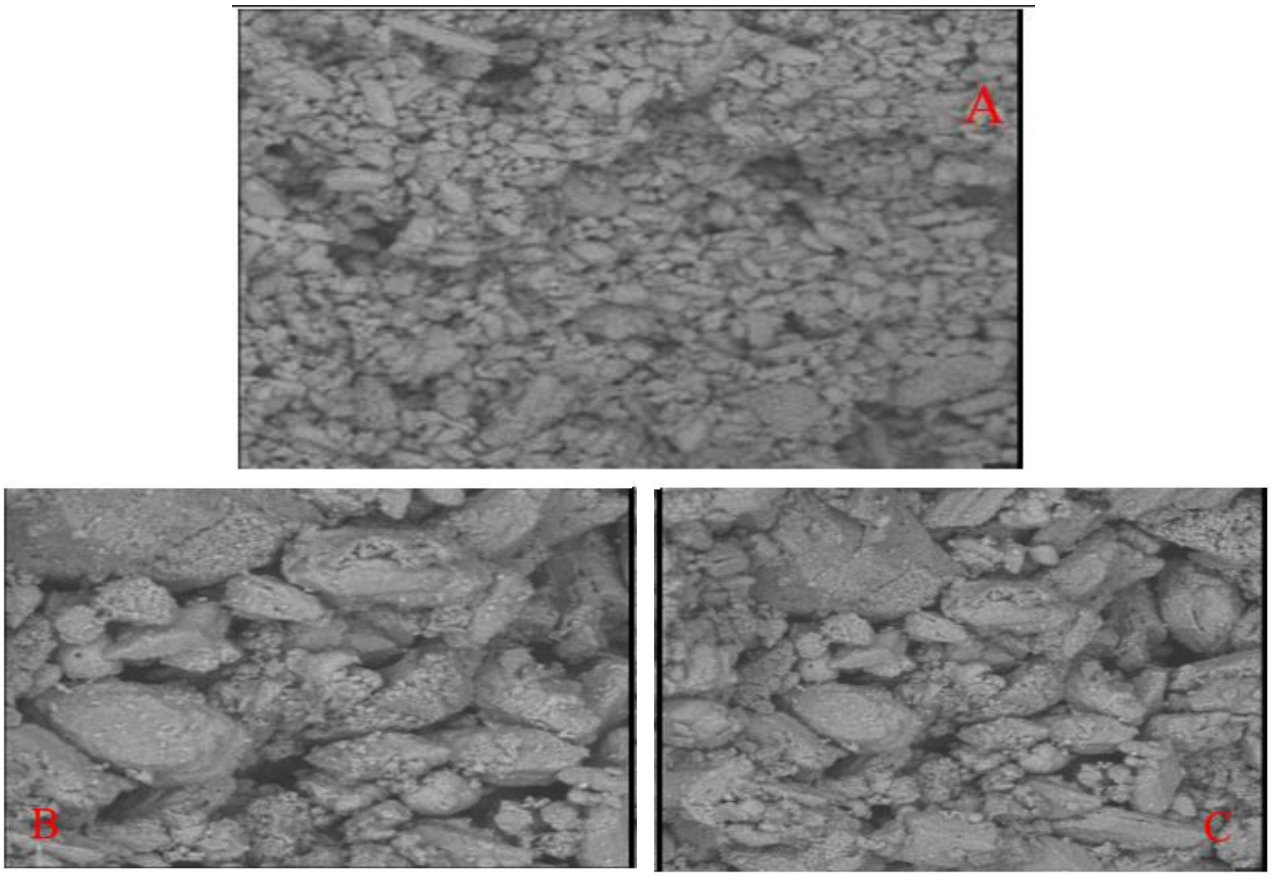

Figure 2. The micro structural images:
a) Raw cocoa pod biomass, **b**) Graphene and **c**) synthesized graphene oxide.

3.3 XRD Analysis

The XRD result of raw cocoa powder, graphene and graphene oxide is depicted in (**Figure 3a-c**). The XRD spectra for graphene from cocoa pod showed that the peaks were not very sharp indicating amorphous nature (**Figure 3a**), but that of graphene oxide showed slightly sharp peaks which indicated crystalline nature (**Figures 3b**). In the XRD result of characterized graphene oxide as shown in (**Figure 3b**), using a wide range scan. The observed peaks are in the range between 2 θ =20 – 50, which shows that 8 peaks are located respectively at 18.18º, 19.05º, 25.32º, 27.25º, 29.85º, 35.68º, 42.86º and 47. 08º. Some peak was closed to 2 θ = 25 and 45º, respectively, indicating the presence of disordered carbon with a turbostratic structure (Lious *et al.* 2010). This result observed the crystalline structure of graphene oxide sample (Kennedy *et al.*2004, Tung *et al.*2009). This is similar to what is observed in the graphene (**Figure 3a**), the carbonized cocoa pod shows less crystalline structure due to observed noise in the spectra. XRD results shows four peaks at 26.5º, 30.4 º, 32.6 º and 42.1º all assigned in synthesized graphene. The graphene oxide sample shows a sharp peak at the 2θ =25.32º which may attributable to an increase in the interlayer spacing due to oxygen containing functional groups on the graphene oxide (Grace *et al.*2020).

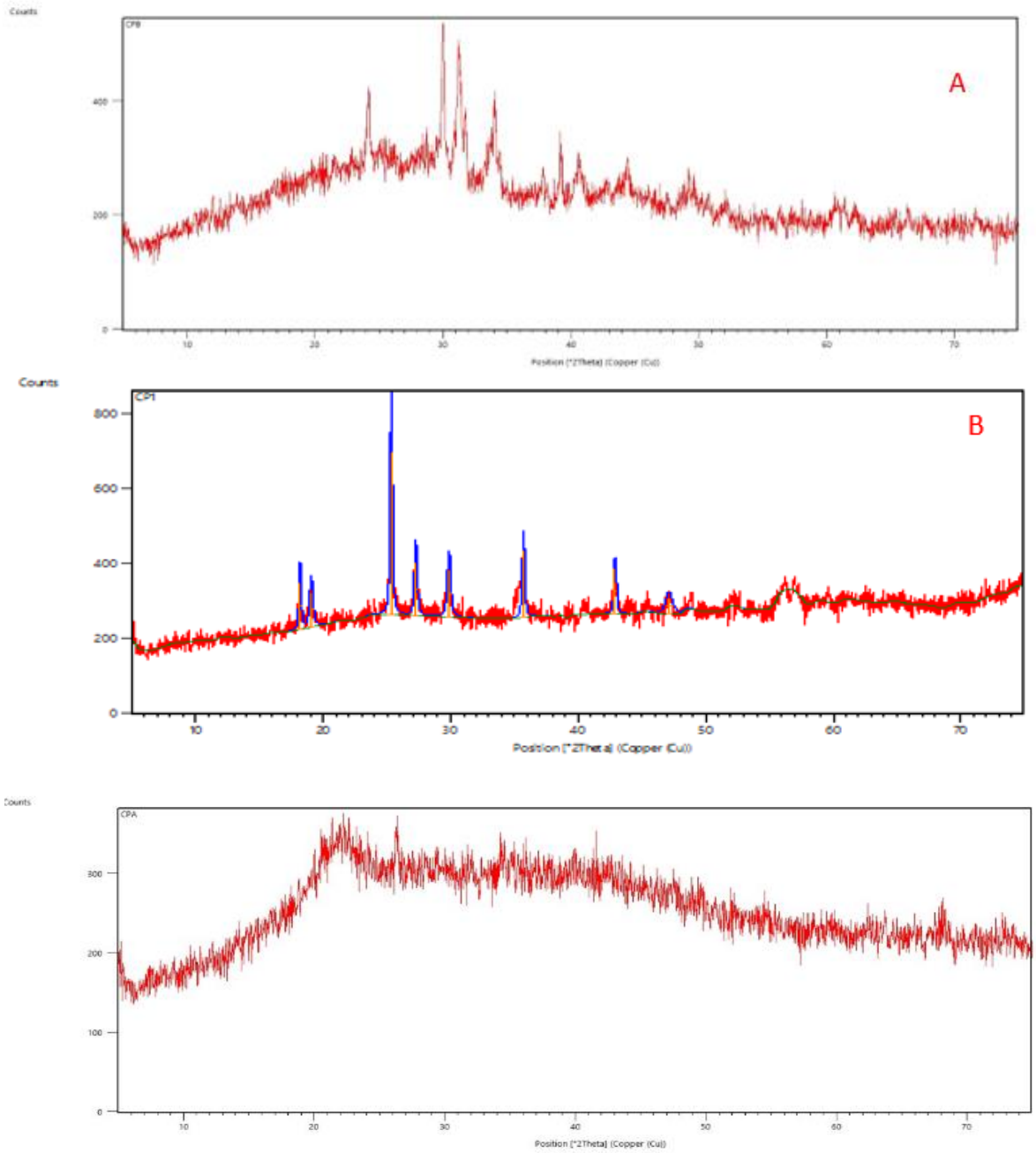

Figure 3. The XRD spectral **a**) graphene oxide **b**) synthesized graphene and **c**) Raw cocoa pod.

The graphene oxide is more crystalline and gives sharper peaks compared to that from carbonized cocoa pod. The graphene oxide synthesis from cocoa pod compared more favourably with the graphene oxide reported by (Alam *et al.*, 2017). In the case of raw cocoa pod powder (**Figure 3c**), the diffraction peaks are not clear and sharp due to observed noise in the spectra. A diffraction peak was observed at around 23.57^{o} and are referred to as an amorphous like carbon material which comprises many folding layers, defects, impurities and hybridization structures

3.4 FTIR Analysis

The FTIR spectrum of raw cocoa powder, grephene and graphene oxide from cocoa-pod was presented in (**Figure 4a-c**). The three samples have similar peaks. The O-H stretching vibration were observed at peaks 3295 cm^{-1}, 3285 cm^{-1} and 3775 cm^{-1} for raw cocoa pod, graphene and graphene oxide respectively, this can be due to the presence of moisture or hydroxyl or their derivatives (Aliyev *et al.*, 2019).

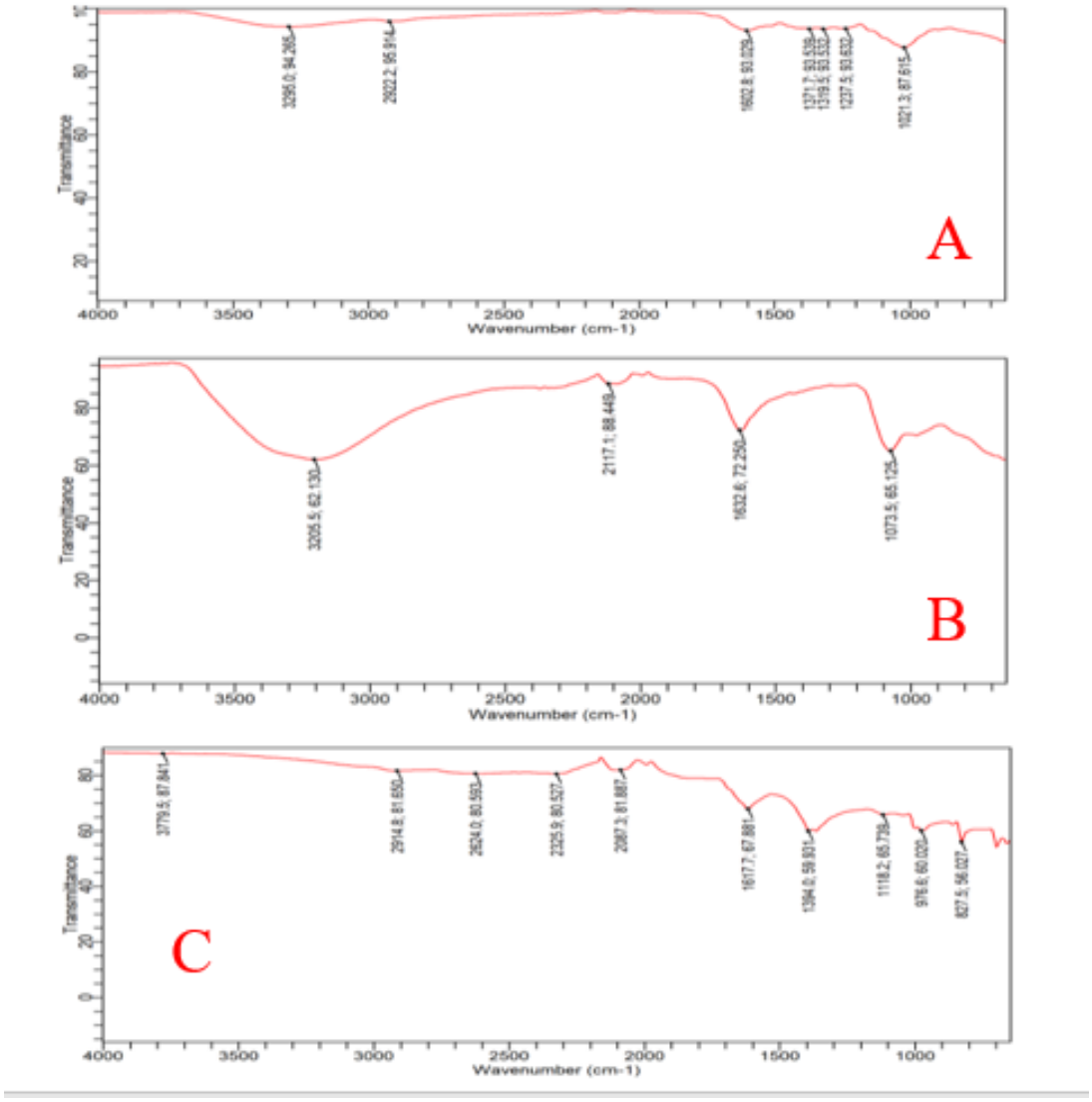

Figure 5. FT-IR: This shows the **a**) Raw cocoa pod biomass, **b**) Graphene and **c**) synthesized graphene oxide

The FTIR spectrum of graphene oxide confirms the introduction of oxygen-containing groups such as functional hydroxyl, epoxy, alcohol and carboxylic groups upon oxidation of cocoa pod. This observation was similar compared to a study by (Bhattacharyya *et al.*, 2017), where they obtained

peaks around 3407 - 3457 cm^{-1} corresponded to stretching vibrations of O-H group found in almond shell. A weak peak around 2117-2914 cm^{-1} was observed, this is due to asymmetric stretching vibration of C-H bond in all the samples, with C=C=O stretching vibrations at 2262.9 cm^{-1} was observed in graphene oxide spectra. The peaks seen at 1608cm^{-1}, 1632 cm^{-1} and 1617 cm^{-1} for raw cocoa pod, graphene and graphene oxide samples respectively, were corresponding to a C=C stretching vibration for aromatic which can be assigned to the skeletal vibrations of graphitic domains (Eluyemi *et al.,* 2016).

The presence of C=C groups showed that even graphene had been oxidized into graphene oxide; the main structure of layer graphene was still retained. Peaks were also observed at 1217 cm^{-1}, 1073 cm^{-1} and 1118 cm^{-1} for raw cocoa pod, graphene and graphene oxide samples respectively, corresponding to a C-O-C group for an epoxy or carbonyl group. The FTIR spectra results from this study were similar with the FTIR results of Aro-Modiu *et al.*, 2019 on synthesis of graphene and its oxide from wastes using Hummer's method. The presence of oxygen-containing functional groups on the synthesized graphene oxides from agriculture waste reveals that the graphite has been oxidized. The indication of polar groups (surface hydroxyl groups), enhances formation of hydrogen bonds between graphene and water molecules, making the produced graphene oxide to be hydrophilic in nature (Qiu *et al.*2023).

Conclusion

The study involves carbonization and synthesis of graphene oxide using modified Hummer's method. from cocoa waste pod. The raw cocoa pod, graphene and graphene oxide obtained was characterized using x-ray fluorescence (XRF); X-Ray Diffraction (XRD); Fourier Infrared Spectrometry (FT-IR) and Scanning Electron Microscopy (SEM). The SEM results reveals a well-developed agglomeration growth with significant increasing in grain size form raw cocoa powder to graphene oxide. The FT-IR analysis of graphene oxide reveals the presences of O-H stretch at (3772.9 cm^{-1}) peak with C-O-C at (1118.2 cm^{-1}) peak which confirms the presence of hydroxyl, epoxy, and carboxylic groups upon oxidation of graphene. While for graphene the assigned peaks are located at (3205, 1632, 2117 and 1632cm^{-1}) which was corresponded to (OH, C=C, C≡C and C-O) respectively. The XRD analysis reveals a wide range scan characterize peaks for both graphene and its oxide indicating the disorderliness of carbon. As established from the results, the cocoa pod waste has excellent prospective potential for producing high valuable adsorbent product for reducing environmental pollution.

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