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## Modified Composite Activated Carbon Derived from Post-Consumer Plastics and Lignocellulosic Materials

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### Authors' contributions

*This work was carried out in collaboration between all authors. Authors ANR and AB designed the study, performed the statistical analysis. Author ANR wrote the protocol, and wrote the first draft of the manuscript. Authors LBB and KJM managed the analyses of the study; author KJM corrected and performed the final manuscript. All authors read and approved the final manuscript.*

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### ABSTRACT

The composite activated carbons were produced using both lignocellulosic materials and urban waste plastics and their characteristics were investigated. The mixture of raw material was first carbonized at 600°C in N<sub>2</sub> atmosphere. The char obtained was then mixed with KOH (in the ratio 2:1 for KOH/char) before the activation process with steam at 850°C. The results show that composite carbons are micro, mesoporous and have higher carbon yield compared to the carbons from both pure raw materials, which are mainly mesoporous. These results were confirmed by their high adsorption of methylene blue. BET surface area for all the samples were over the acceptable range (991.5 - 1412.9). Moreover, the average size of the pores of carbon between in the range of 1.8 – 2.3 nm. Carbon surfaces were analyzed in detail using FTIR and SEM, exhibiting hydroxyl and

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carboxylic functional groups on the surfaces and cavities of mesopore size.

*Keywords: Lignocellulosic materials and waste plastics; impregnation; composite activated carbon; mesoporous; yield.*

## 1. INTRODUCTION

The production and utilization of activated carbon has increased for decades, especially in water treatment and in industrial applications such as in the extraction of metal ions, air handling, purification [1], the decoloration of food in the food industry and in the pharmaceutical industry [1-3]. This is because of high adsorption capacity of carbon for certain toxins and poisons, due to their porous structure and accessibility of their surface to combat certain types of bacterial ailments. In the last thirty years, activated carbons have been used as catalysts and as catalysts support [4-7]. Despite such features, the production of activated carbon on a large scale is limited due to their high production cost. Consequently, activated carbon is being replaced by cheap adsorbents in many industrial applications, despite their low removal efficiencies compared with those of AC. Over the last decade, many studies have been carried out on the development of low-cost adsorbents derived from waste materials. So AC production costs have been reduced by choosing cheap and locally available raw materials such as urban wastes and agricultural wastes instead of costly deforestation [8].

A post-consumer plastic waste, with its wide commercial applications and low biodegradability, represents a serious challenge for developing countries. The transformation of post-consumer plastics into activated carbon appears not only as an alternative solution to the problems of pollution by plastics, but also a way of recycling it. Much work has recently been done on the preparation of AC from plastics, and the results have shown that they have a better adsorption capacity and a larger surface area as compared to those from lignocellulosic materials which are widely used on a commercial scale [9-13]. But the main problem here is that of low yield, as large quantities of plastic are needed to obtain a high yield. It is then very expensive compared to AC obtained from plant origins which have a relatively high output.

The present work is focused on the preparation of composite activated carbon from post-consumer plastics (polyethyleneterephthalate) mixed with lignocellulosic material (oil palm shell). It is hope that, this will increase the yield of AC compared to that from plastics waste only. For this purpose physical and chemical activation will be combined using steam vapor and KOH to increase their porosity.

## 2. EXPERIMENTS

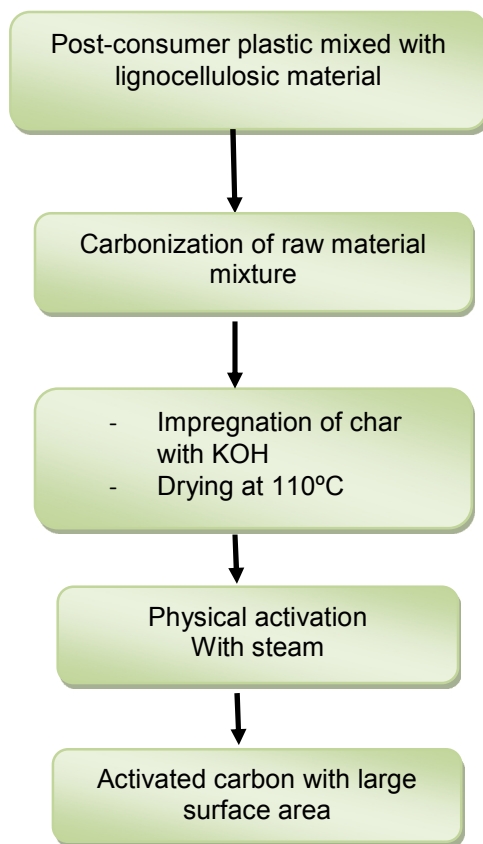
### 2.1 Raw Materials and Sample Preparation

The raw materials for the preparation of ACs are synthetic plastic and Oil Palm Shell (OPS), as lignocellulosic materials. The composites ACs were obtained by mixing both PET and OPS in the ratios 2:1 and 1:1 respectively.

Oil palm shells (OPS) were granulated into particle sizes ranging from 0.5 to 1.5 mm in diameter and Polyethyleneterephthalate (PET) samples were sliced into similar particles sizes.

OPS and PET were mixed in the ratios 2:1 and 1:1 respectively, the mixtures were dried in the oven at 110°C and then, subjected to carbonization using a cylindrical furnace. The heating started from room temperature with a heating rate of 10°C/min up to the final temperature of 600°C. The heating process was stabilized with nitrogen at a flow rate of about 100mL/min. The residence time of the samples at the final stabilized temperature of 600°C was 3hrs. After heat treatment, the nitrogen flow was stopped and the charcoals were cooled down to room temperature.

Before the activation step, 5–6g of the carbonized samples were mixed with Potassium hydroxyl in the ratio 1:2 (char/KOH) and dried in the oven overnight at 110°C. The mixtures were activated with steam (0,13mL/min) in a furnace by heating the reactor from room temperature to 850°C at a heating rate of 10°C/min, with a residence time of 2hrs. After the activation, the steam flow was stopped and the obtained ACs were cooled down to room temperature and washed with distilled water to a pH of 7. For comparisons, activated carbons were also prepared from the pure raw materials (PET and OPS) in the cylindrical furnace under the same conditions (Fig. 1).



**Fig. 1. The procedure for preparing composite activated carbon from post-consumer plastic and lignocellulosic material mixture**

## 2.2 Characterization of Activated Carbons by Nitrogen Gas Adsorption

Adsorption of gases at low temperature provides considerable information on the determination of specific surface area and pore size distribution of nanoporous adsorbents. Within the framework of this work, carbon samples were characterized for their surface and pore characteristics using an adsorption apparatus (Micromeritics TriStar surface Area and porosity analyzer). Analyses of adsorption and desorption of nitrogen were conducted at -195.8°C. Before the experiments, the sample was degassed at 200°C overnight. Total surface areas were determined by applying the BET equation [13]. External surface areas were determined by applying the *t*-plot model between the range 7–9 Å [15]. Pore size distributions were determined using both BJH methods.

## 2.3. Aqueous Adsorption Characteristics

### 2.3.1 Iodine number

The iodine number was determined by using the sodium thiosulfate volumetric method. Standard iodine solution was added over activated carbon (10 mg) and after an equilibration time of 4h, the residual iodine concentration was determined by titration with standard sodium thiosulfate using starch as an indicator. The iodine number was defined as the adsorbed quantities of iodine (in mol g<sup>-1</sup> of carbon) obtained by subtracting the residual concentration at equilibrium  $C_e$ , from the initial concentration  $C_o$ .

$$Q_o = \frac{C_o - C_e}{m} V \quad (1)$$

Where  $m$  and  $V$  are the mass of AC sample and the volume of adsorbing solution respectively.

### 2.3.2 Methylene blue adsorption test

The samples of carbons were investigated for their aqueous adsorption of dyes using methylene blue. Carbon samples (10 mg) were mixed with MB in 100 mL stoppered Erlenmeyer flasks. Batch experiments were carried out in the shaker (200 rpm) at room temperature for 4 h. The solutions were filtered and the concentrations of solutions were then determined by spectrophotometer at 660 nm. Then the quantity adsorbed was also calculated (mol g<sup>-1</sup> of carbon) using equation 1.

## 2.4 Scanning Electron Microscopy and Infra-Red analysis

The carbon samples were analyzed using FTIR spectroscopy. The samples were mixed with potassium bromide and the mixture was pressed into pellets to be used in the analysis. The spectra were recorded from 4000 to 400 cm<sup>-1</sup>. Microstructures of the samples were examined using a Scanning Electron Microscope.

## 3. RESULTS AND DISCUSSION

The sample obtained from the mixture of raw materials are COM1I and COM2I respectively ratio 1:1 and 2:1 (PET/OPS), those obtained from polyethyleneterephtalate and oil palm

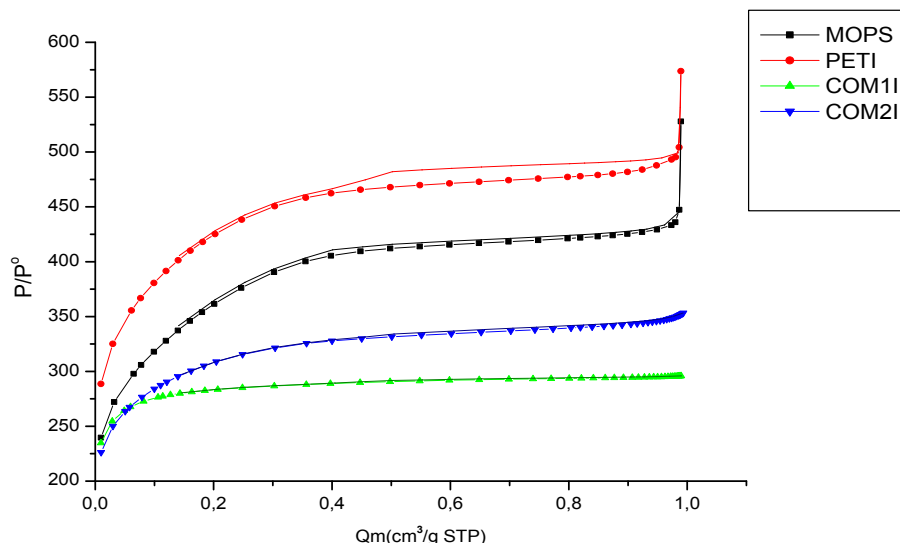
shell are called PETI and MOPS respectively. The process for the preparation of the composite adsorbent is presented in Fig. 1. The characteristics of prepared carbon samples are presented in Table 1.

**Table 1. Characteristics of different ACs obtained**

Activated carbons	Raw materials	Yields%	
		Carbonization	Activation
OPS	<i>Oil palm shell(OPS)</i>	30	45.7
PETA	PET	15	33.3
COM1	OPS +PET; ratio 1/1	24	58.7
COM2	OPS +PET; ratio 2/1	21	40.7

### 3.1 Nitrogen Adsorption Isotherms Pore Size Distributions

The N<sub>2</sub> adsorption isotherms at 77 K for the composite activated carbons produced from the mixture of post-consume plastics and lignocellulosic materials by physico-chemical activation are presented in Fig. 2. All the samples gave type II isotherms, characteristic of microporous and mesoporous solids. The adsorption isotherms show adsorption-desorption hysteresis, indicating the presence of mesopores. Besides contributing significantly to the adsorption of the adsorbate, Mesoporous predominance could be attributed to the interaction of functional groups at the surface of char which favors the evolution of molecules during the heating process of activation, then probably creating external pores. These pores act as conduits leading the adsorbate molecules to the micropore cavity [16]. The impregnation of char by KOH generates a large external surface area for samples MOPS and MPET compared to their internal surface (micropores surface area). Although the micropores surfaces of composites ACs are greater than external surface, it can be noticed that the amount of their external surface are significant, in the case of COM2I where micropores and external surface are 570.3 and 514.7 respectively. The overall porosity development determined by N<sub>2</sub> adsorption at 77 K using BET and BJH methods are shown in Table 2. It shows that all the samples have a large surface area between 1000 and 1500 m<sup>2</sup>/g with highly developed microporosity. The mesopore accumulate in a small size range (around 2-4 nm) and this is more clearly seen in the MOPS and MPET samples. In the micropore size distributions, only COM1I sample showed an important amount of micropore below 1nm. Mesoporous predominance could be attributed to the interaction of functional groups at the surface of char which favors the evolution of molecules during the heating process of activation, then probably creating external pores.



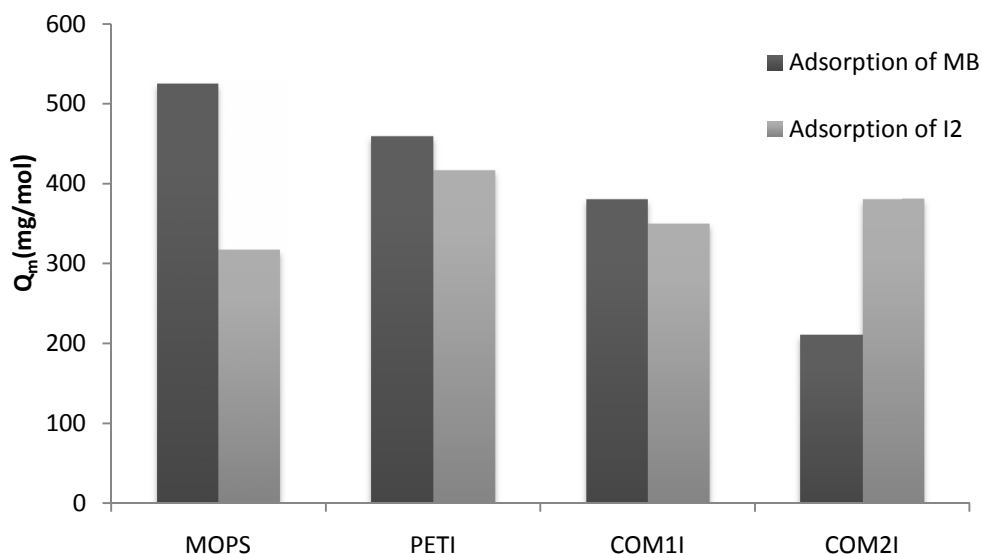
**Fig. 2.** Typical nitrogen adsorption/desorption isotherm of the four samples of activated carbon prepared by physical activation

**Table 2.** Porous parameters of the activated carbons

Samples	$S_{BET}$ ( $M^2/G$ )	$S_{MI}$ ( $M^2/G$ )	$S_E$ ( $M^2/G$ )	Pores volumes( $CM^3/G$ )		Average pores sizes (NM)		
				$V_{MICRO}$	$V_{CUM}$	$BET_{ADS}$	$BJH_{DES}$	$BJH_{ADS}$
MOPS	1221.7	312.6	909.1	0.1602	0.4909	2.2655	3.7693	3.7441
PETI	1412.9	516.7	896.1	0.2634	0.4553	2.2085	3.7252	3.6253
COM11	991.45	829.93	161.52	0.3674	0.4574	1.8453	2.8920	2.8846
COM2I	1085.0	570.26	514.7	0.2507	0.5439	2.0053	2.8875	2.9176

### 3.2. Adsorption Tests in Aqueous Solution

Aqueous adsorption tests were conducted on the four adsorbates compounds with the aim of confirming their porous structure and also to test the adsorption capacity of inorganic and organic species from the aqueous phase. With regards to gas adsorption results, the samples are expected to adsorb higher quantities of methylene blue due to their large external area. Methylene blue is adsorbed by filling mechanisms due to the large amount of mesopores and adsorption by iodine proceeds by capillary condensation in the mesoporosity. Indeed the high adsorption capacity of methylene (between 350 at 525 mol/g) was found compared to the adsorption of iodine (300 - 420 mol/g), Fig. 3 [17]. Adsorption of methylene blue for COM2I sample is lower than that of the other samples, this can be justified by the fact that it contains micropores and mesopores, so the existing mesopores at the external surface is lower and serves only as the entrance to micropores.



**Fig. 3. Adsorption of methylene blue and iodine**

### 3.3 Surface Chemical and Morphologic Characteristics

Infrared spectroscopy (IR) (Fig.4) is an important and forceful tool that can provide useful information about surface functional groups on carbons. The adsorption capacity of active carbons is determined by their physical or porous structure but is strongly influenced by the chemical structure [18-19]. The spectra show a weaker shoulder between 1550 at 1600  $\text{cm}^{-1}$  which can be attributed to C=C aromatics cycles stretching vibration. The dominant absorption peak around 1120  $\text{cm}^{-1}$  indicated the existence of C=O bond stretch. The region between 501 and 600  $\text{cm}^{-1}$  for all the samples are due to deformation vibrations out of the aromatic C-H bonds. The intense absorption band at 3400  $\text{cm}^{-1}$  was attributed to absorbed water. It can be noticed in the spectra that the peaks appear at the same frequency but their intensity vary with sample showing that functional structure on the carbons are identical. The Scanning Electron Microscopy is used for imaging the morphologies of ACs. Examination of these carbons (Fig. 5) exhibit external surface structure indicating that in some parts of the carbon impregnation has produced cavities of mesopores size [20-21].

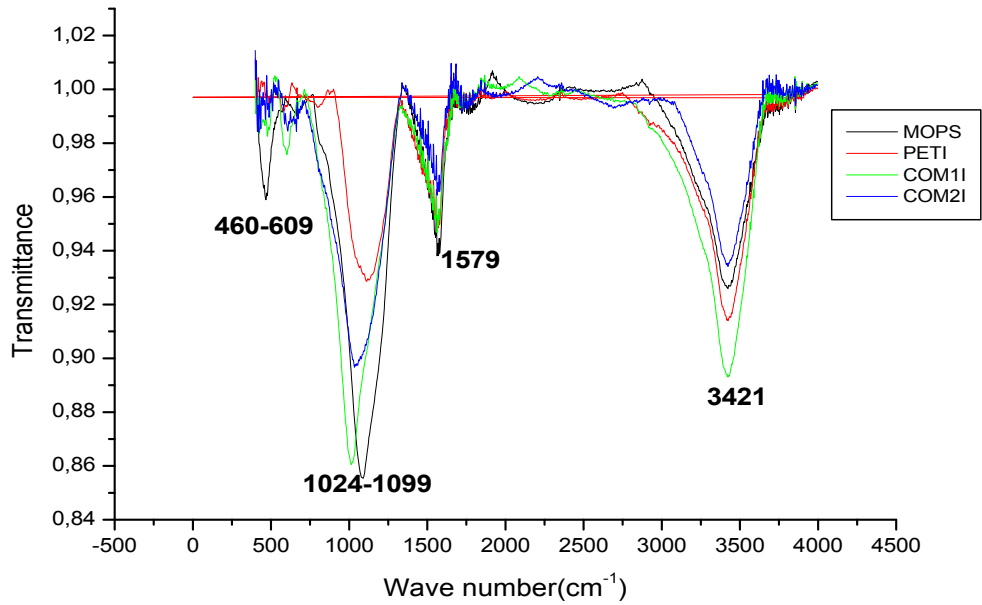


Fig. 4. FTIR spectra of four activated carbons obtained by physical activation

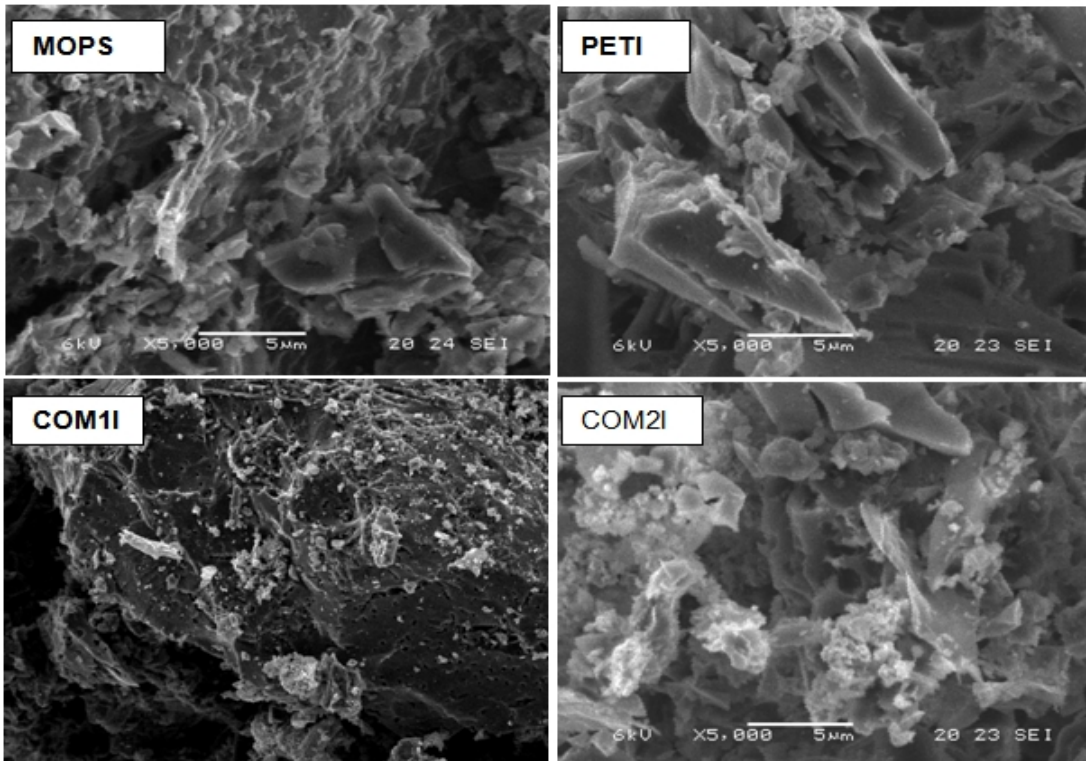


Fig. 5. SEM photographs of the four activated carbons prepared



#### 4. CONCLUSION

Activated carbons with high surface area can be produced by physico-chemical activation from a mixture of both waste lignocellulosic material and post-consumer plastics using KOH for impregnation. They have high methylene blue numbers compared to the adsorption of iodine. As revealed above, impregnation of char by KOH before activation contributed a significant influence on development of mesopores into the Composite adsorbent. The yield of composite ACs (24 and 21% for COM1I and COM2I respectively) is higher compared to the sample prepared from plastics (15%). Using low energy conditions of preparation, the specific area of the entire samples were over the acceptable range (991.45- 1412.9 m<sup>2</sup>/g). Moreover, the size of the pores on the pore wall of the carbon is in the range of 1.8 – 2.3 nm. Hydroxyl and carboxylic functional groups on the surfaces and cavities of mesopores size were also found at the adsorbent surface using FTIR and SEM analysis. Therefore, the composite ACs prepared developed porous adsorbents with a greater adsorption capacity; hence they can be used as low-cost adsorbent and as an alternative for environmental pollution remediation.

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#### COMPETING INTERESTS

Authors have declared that no competing interests exist.

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