

# Structural Characterization and Solid State Properties of Thermal Insulating Cellulose Materials of Different Size Classifications

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This study investigated two classifications of wood cellulose of particle sizes 300  $\mu\text{m}$  to 424  $\mu\text{m}$  and 600  $\mu\text{m}$  to 849  $\mu\text{m}$ . The cellulose samples were characterized using X-ray diffraction (XRD), energy-dispersive X-ray spectroscopy (EDX), and scanning electron microscopy (SEM). The cellulose crystal revealed a preferred orientation along the (200) plane for the most prominent peak. The XRD diffractogram revealed an orthorhombic structure obtained from the powder diffractogram file (PDF). Furthermore, the crystallinity index and crystalline size were calculated and the increase in crystalline size of the isolated cellulose indicated higher thermal stability. The EDX analysis showed chemical components of carbon (C), sodium (Na), chlorine (Cl), and oxygen (O) in the isolated cellulose. The morphology of the cellulose appeared as strings of fibres. The isolated cellulose has applications in the production of biomaterial, thermal insulating devices, and domestic applications.

*Keywords:* Celtis philippensis; Pulping; Bleaching; Cellulose; Instrumental Analysis

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## INTRODUCTION

Wood is one of the most abundant natural materials on earth with a composition of cellulose, hemicellulose, and lignin. It has been widely used and found essential to human life for thousands of years as building materials and other applications (Edgar *et al.* 2001; Zhang 2001; Belgacem and Grandini 2005; Oluyamo and Adekoya 2015).

There has been concern in the environmental degeneration caused by residues and wastes from domestic and industrial activities all over the world. Therefore, the need to transform waste to wealth is vigorously being pursued by researchers to remove the constituent threats to the environment.

Sawdust, a by-product of wood, has been identified as a challenge and a waste to the environment as the material is usually disposed of in haphazard locations, which most often cause environmental pollution. Various studies have been conducted on this waste material. The results have shown that it can be used to improve insulation properties of device materials (Adekimi 2004; Oluyamo and Bello 2014).

Thermal insulation materials can prevent heat loss and provide thermal comfort. The main factor that determines an insulation material's effectiveness is its thermal conductivity. The lower the thermal conductivity of a material, the more effective it can

be as an insulator. One such material is cellulose isolated from wood, which is naturally eco-friendly and also has favourable thermal properties.

Cellulose, having the general formula  $(C_6H_{10}O_5)_n$ , can be isolated from other raw materials (cotton, sugarcane bagasse, rice straw, durra stalk, groundnut shell, *etc.*), but wood remains one of the main sources of cellulose isolation. It is a renewable resource with an exceptional strength, low density, and high durability (Draftz *et al.* 2015; Wei *et al.* 2015). Cellulose can be regarded as the major component of wood, a natural composite that has several components (Hon 1994). This natural polymer is close to one-third of plant tissue, which can be restocked by photosynthesis (Morán *et al.* 2008). Cellulose is one of the most widely used polymers in nature, mostly used by humans for thousands of years as building material, and also used by engineers due to its natural availability (Rong *et al.* 2001; Salajkova 2003; Vignon *et al.* 2004; Largerwall *et al.* 2014). It represents a naturally linear condensation polymer consisting of D-anhydroglucopyranose units joined together by  $\beta$ -1,4-glycosidic bonds (Klemm *et al.* 1998; Bledzki and Gassan 1999). The chemical, constitutional, and spatial confirmations are structural entities that aggregate the cellulose chain with a strong tendency (Sun *et al.* 2008). Van der Waals forces and stronger inter- and intra-molecular H-bonding are the tightly and compactly arranged chain (Abdullah *et al.* 2011). Various reagents can be used for the isolation of cellulose from wood, and acidified sodium chlorite ( $NaClO_2 + H_3O^+$ ) has been a widely used standard reagent (Wise *et al.* 1946). The chlorine radical,  $Cl\cdot$ , can react with the lignocellulosic material, transforming it into toxic organochlorine.

Cellulose is unique with respect to its versatility in usage, which can be affected by the method of its isolation, the number of inter- and intra-molecular hydrogen bonds, chain lengths, chain length distribution, crystallinity, and distribution of functional groups within the repeating units and along the polymer chains (Åkerholm *et al.* 2004; Klemm *et al.* 2005; Oh *et al.* 2005).

*Celtis philippensis* Blanco is a flowering plant that includes the elms (genus *Ulmus*) and the zelkovas (genus *Zelkova*). It is from the family Cannabaceae, which is common in the research area. The tree is harvested from the wild for local use as medicine, food, and as a source of wood (Denk and Grimm 2005). Relevant research has been conducted to determine its thermal properties and applications of the material.

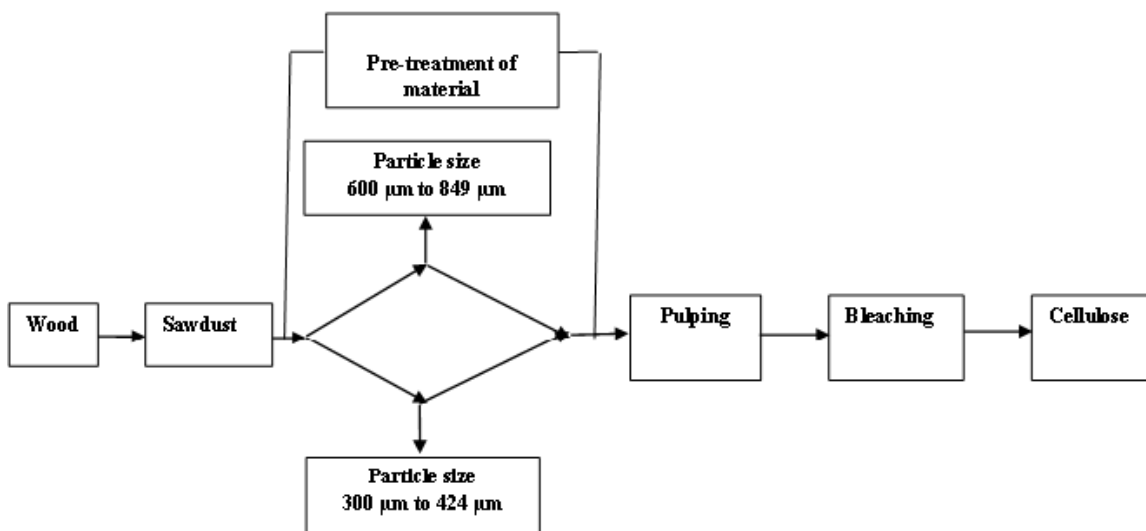
The main objective of the present work is to produce thermal insulating cellulose materials from *Celtis philippensis* and characterize the structural and solid-state properties for domestic and industrial applications. The crystalline properties were studied by X-ray diffraction (XRD), the morphology was analyzed using scanning electron microscopy (SEM), and the elemental composition was analyzed using energy-dispersive X-ray spectroscopy (EDX).

## EXPERIMENTAL

### Materials

The *Celtis philippensis* samples were collected from a sawmill in Akure South Local Government Area of Ondo State, South Western Nigeria. All the samples were authenticated, processed into saw dust at the Department of Forestry and Wood Technology, and sieved into different particle sizes (300  $\mu$ m to 424  $\mu$ m and 600  $\mu$ m to 849  $\mu$ m) at the Department of Materials and Metallurgical Engineering of The Federal University of Technology, Akure Nigeria. Sodium chlorite ( $NaClO_2$ ) (Sigma-Aldrich,

Steinheim, Germany), sodium hydroxide (NaOH) (British Drug House, Darmstadt, Germany), and acetic acid (Sigma-Aldrich, Steinheim, Germany) of analytical grades were the chemicals used in the analysis.



**Fig.1.** Schematic diagram of the isolation process

#### *Pre-treatment of material*

Prior to the pulping of the samples, the materials were sieved using a Wiley mechanical sieve shaker (Pascal Engineering, Sussex, England) and sawdust with two different particle sizes ranges (300 µm to 424 µm and 600 µm to 849 µm) were obtained.

#### *Pulping procedure*

The pulping method followed the procedure developed by Oluwasina *et al.* (2014) with a few modifications. Specifically, the wood particle was pulped under atmospheric pressure in a water bath at 90 °C with a wood to liquor ratio of 1:20, using 20 % NaOH for 90 min. After digestion, the pulp was obtained by filtration and washed thoroughly with water until it was free of residual alkali. The pulp yield was oven-dried at 105 °C to a constant weight.

#### *Bleaching procedure*

An amount of 1000 mL of hot distilled water, 10 g of NaClO<sub>2</sub>, and 3 mL of acetic acid were added to approximately 20 g of oven-dried pulp sample in a 2-L Erlenmeyer flask. The flask was covered, and the mixture heated in a water bath at 70 °C for 30 min with intermittent stirring. After the first 30 min in the water bath, another 10 g of NaClO<sub>2</sub> and 3 mL of acetic acid was added with intermittent stirring and sustained for another 30 min before switching the bath off. The sample was allowed to settle down for 24 h in the water bath. After digestion, the bleach was obtained by filtration and washed thoroughly with water until it was free of residual alkali and chlorine. The obtained sample was dried at 80 °C to a constant weight.

## Methods

### Characterization

The crystallinity resulting from different processing procedures was determined using the XRD techniques, which enabled verification of the phases present in the samples. The XRD patterns were obtained using a Philips PW 3710 X'pert Pro diffractometer (Philips Analytical, Almelo, Netherlands) with a Cu-K $\alpha$  monochromator of wavelength,  $\lambda = 1.540598 \text{ \AA}$ , in the range of  $5^\circ$  to  $50^\circ$  ( $2\theta$ ) generated at 15 kV. The surface morphology and atomic composition of the cellulose were studied using SEM (FEI Company, Hillsboro Oregon, USA) with an accelerating voltage of up to 30 kV and a resolution of up to 1 nm. The elemental composition was determined with an energy-dispersive X-ray (EDX) using a FEI NOVA 200 NanoSEM equipment (FEI Company, Hillsboro Oregon, USA).

### Theoretical considerations

The Interplanar spacing ( $d$ -spacing) was calculated from the Bragg equation (Wada and Okano 2001; Kim *et al.* 2010),

$$n\lambda = 2d\sin\theta \quad (1)$$

where  $n$  is the order of reflection,  $\lambda$  is the wavelength of the incident X-rays (m),  $d$  is the interplanar spacing of the crystal, and  $\theta$  is the Bragg's angle ( $^\circ$ ).

The orthorhombic lattice parameter ( $a \neq b \neq c$ ) for the orthorhombic phase structure was calculated by Eq. 2 as,

$$d = \sqrt{\left(\frac{a}{h}\right)^2 + \left(\frac{b}{k}\right)^2 + \left(\frac{c}{l}\right)^2} \quad (2)$$

where  $d$  is the interplanar spacing(m),  $a$ ,  $b$ , and  $c$  are lattice parameters, which are measure in (m),and  $h$ ,  $k$ , and  $l$  are the miller indices.

The crystallinity index (CrI, %) was estimated from the XRD diffraction pattern. The patterns were engaged to determine the crystallinity parameters of cellulose derived from different particle sizes of the wood dust samples. The crystallinity index was calculated according to Segal *et al.* (1959), as follows in Eq. 3,

$$CrI = \frac{I_{200} - I_{am}}{I_{200}} \times 100 \quad (3)$$

where  $I_{200}$  is the maximum intensity of the lattice diffraction and  $I_{am}$  is the low intensity peak of the amorphous region of the baseline at  $2\theta$ , approximately  $18^\circ$ .

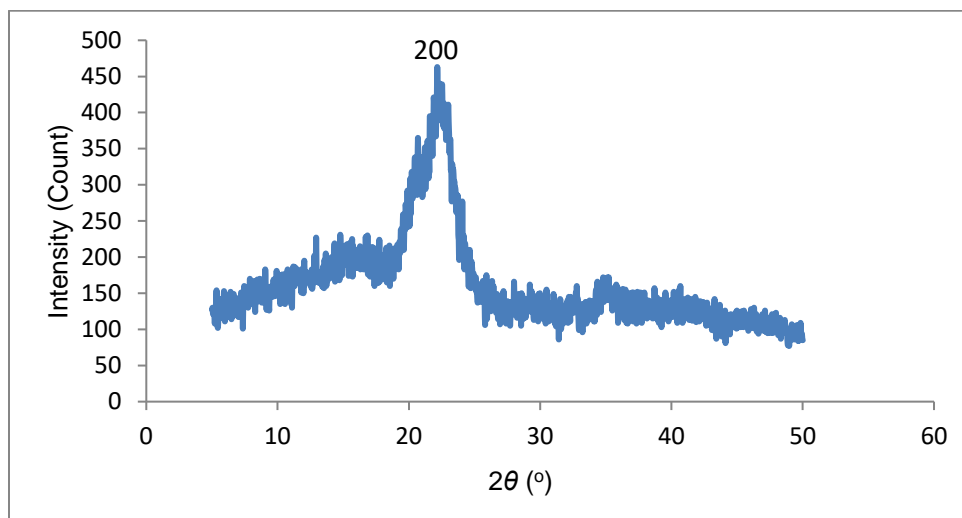
The crystalline size ( $L$ ) was calculated using the Scherrer equation (Gümüskaya *et al.* 2003; Popescu *et al.* 2011), as

$$L = \frac{K \times \lambda}{H \times \cos\theta} \quad (4)$$

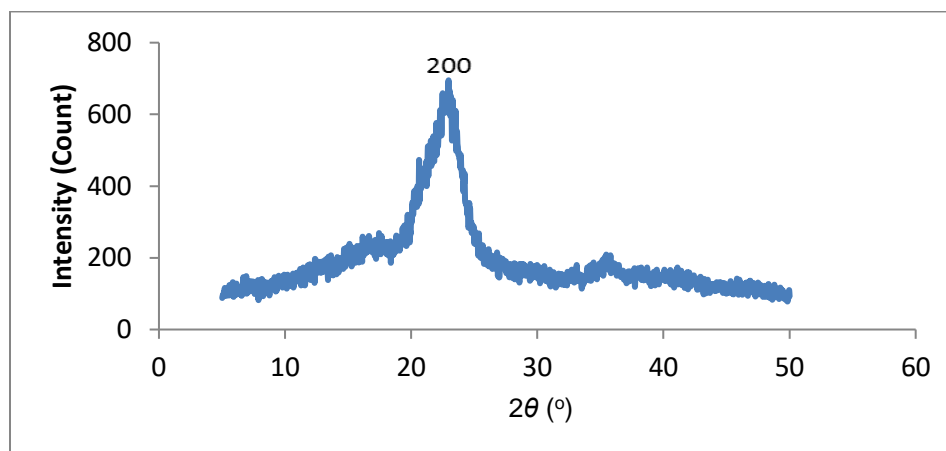
where  $K$  is a constant whose value is given as 0.91,  $\theta$  is the Bragg's angle ( $^\circ$ ), and  $H$  is the intensity of the full width at half maximum (FWHM) ( $^\circ$ ) corresponding to a high intensity peak of the diffraction plane.

## RESULTS AND DISCUSSION

To determine the crystallinity, the XRD patterns of the isolated cellulose were analyzed. Figures 2a and 2b show the XRD diffractogram pattern of cellulose with two ranges of particle sizes, 300  $\mu\text{m}$  to 424  $\mu\text{m}$  and 600  $\mu\text{m}$  to 849  $\mu\text{m}$ .



**Fig. 2a.** XRD pattern of cellulose with particle size ranging from 300  $\mu\text{m}$  to 424  $\mu\text{m}$



**Fig. 2b.** XRD pattern of cellulose with particle size ranging from 600  $\mu\text{m}$  to 849  $\mu\text{m}$

The band positions of angle  $2\theta$  and the observed interplanar spacing ( $d$ -spacing) values of the isolated cellulose spectra closely matched the reported values in the powder diffractogram file (PDF) indicated in Table 1. Other parameters obtained from XRD measurements, such as particle sizes, FWHM, plane of orientation, and isolated cellulose phase are also included in Table 1 for the particle sizes examined (300  $\mu\text{m}$  to 424  $\mu\text{m}$  and 600  $\mu\text{m}$  to 849  $\mu\text{m}$ ). The Scherrer's equation was used to estimate the crystalline sizes for each sample.

It was observed that the isolated cellulose with the particle sizes considered had the same pattern. This was in agreement with Zugenmaier (2008) in the study of crystalline cellulose and derivatives.

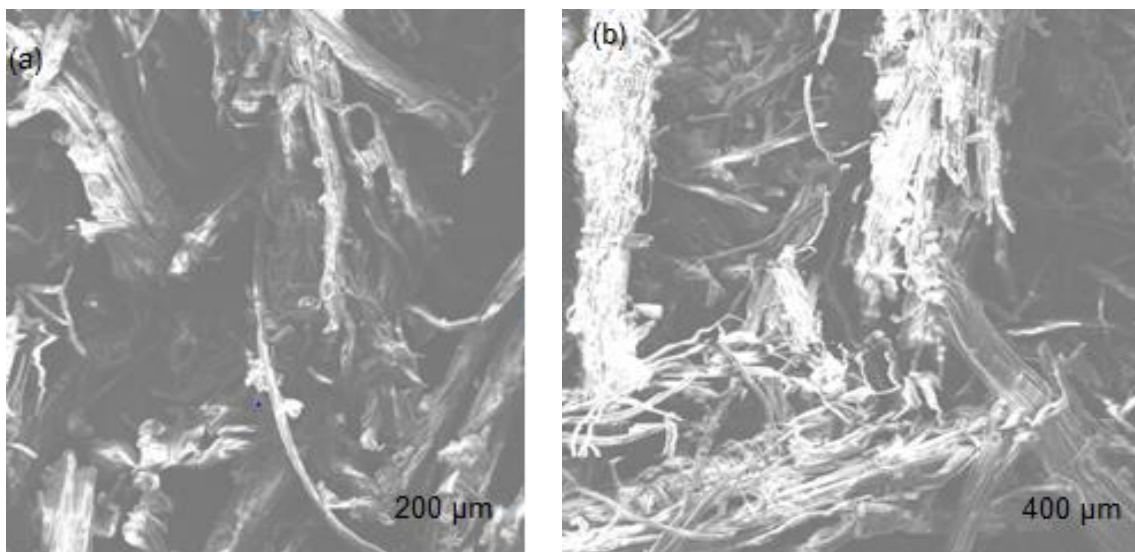
**Table 1.** Summary of XRD Analysis on the Particle Size Range of Plane (200)

Particle Size ( $\mu\text{m}$ )	Angle ( $2\theta$ ) $^\circ$	d - Spacing ( $\text{\AA}$ m)	FWHM ( $^\circ$ )	Crystalline Size (nm)	Crystalline Index (%)	Phase	Ref. Code Marching
300 to 424	22.289	3.985	0.6336	0.226	60.91	Orthorhombic	00-008-0109
600 to 849	22.860	3.889	1.1088	0.130	64.89	Orthorhombic	01-072-1026

The crystal plane of preferred orientation was along the (200) orthorhombic plane for the most prominent peak. These peaks were observed at  $2\theta = 22.289^\circ$  for cellulose obtained from sawdust with particle size 300  $\mu\text{m}$  to 424  $\mu\text{m}$  and  $2\theta = 22.860^\circ$  for cellulose with particle size 600  $\mu\text{m}$  to 849  $\mu\text{m}$ . The calculated orthorhombic lattice parameters ( $a \neq b \neq c$ ) of the isolated cellulose were:  $a = 4.990\text{\AA}$ ,  $b = 10.990\text{\AA}$ , and  $c = 7.110\text{\AA}$  for cellulose with particle size 300  $\mu\text{m}$  to 424  $\mu\text{m}$ , and  $a = 4.947\text{\AA}$ ,  $b = 11.032\text{\AA}$ , and  $c = 7.108\text{\AA}$  for cellulose with particle size 600  $\mu\text{m}$  to 849  $\mu\text{m}$ .

The crystallinity index (CrI) calculated according to Segal *et al.* (1959) showed that cellulose with particle size 600  $\mu\text{m}$  to 849  $\mu\text{m}$  had a higher index than that obtained from that with particle sizes 300  $\mu\text{m}$  to 424  $\mu\text{m}$ . The CrI values were 60.91% and 64.89% for cellulose with particle size 300  $\mu\text{m}$  to 424  $\mu\text{m}$  and 600  $\mu\text{m}$  to 849  $\mu\text{m}$ , respectively. In a previous study, 60.4% and 62.6% were recorded for native celluloses of *Eucalyptus grandis* and *Pinus taeda* (Poletto *et al.* 2013). However, the particle sizes of the celluloses in the study were not specified.

This high percentage in crystallinity index might be associated with the reduction in the corresponding amorphous state of the material due to the probable dissociation of the bonds as a result of pulping.



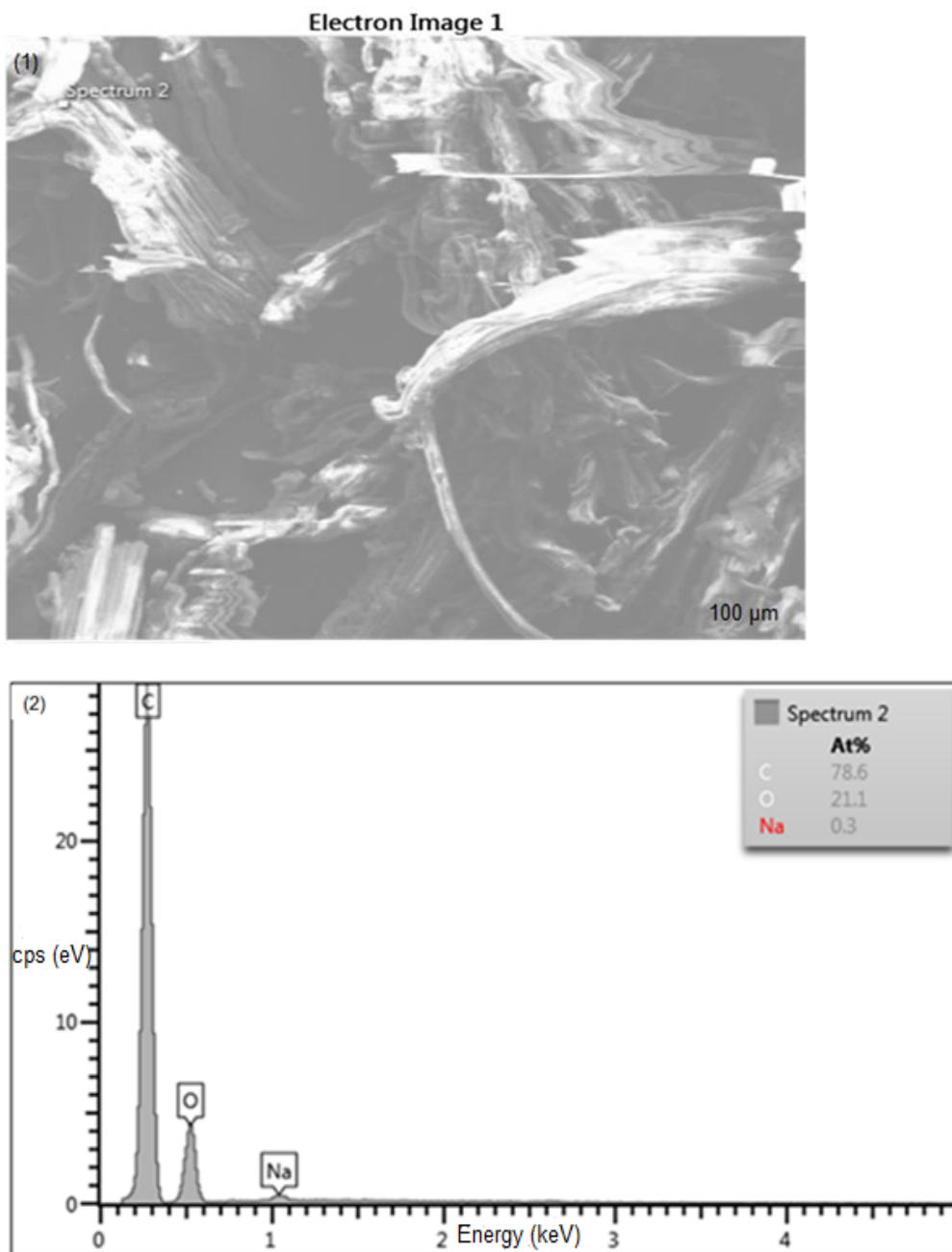
**Fig. 3.** SEM image of cellulose from sawdust with particle size (a) 300  $\mu\text{m}$  to 424  $\mu\text{m}$  and (b) 600  $\mu\text{m}$  to 849  $\mu\text{m}$

As shown in Fig. 3, SEM images of the isolated cellulose revealed strings of fibres, which was in agreement with other studies (El-Sakhawy and Hassan 2007; Adel *et al.* 2010; Ibrahim *et al.* 2010; Morgado and Frollini 2011; Pereira *et al.* 2011; Oluwasina

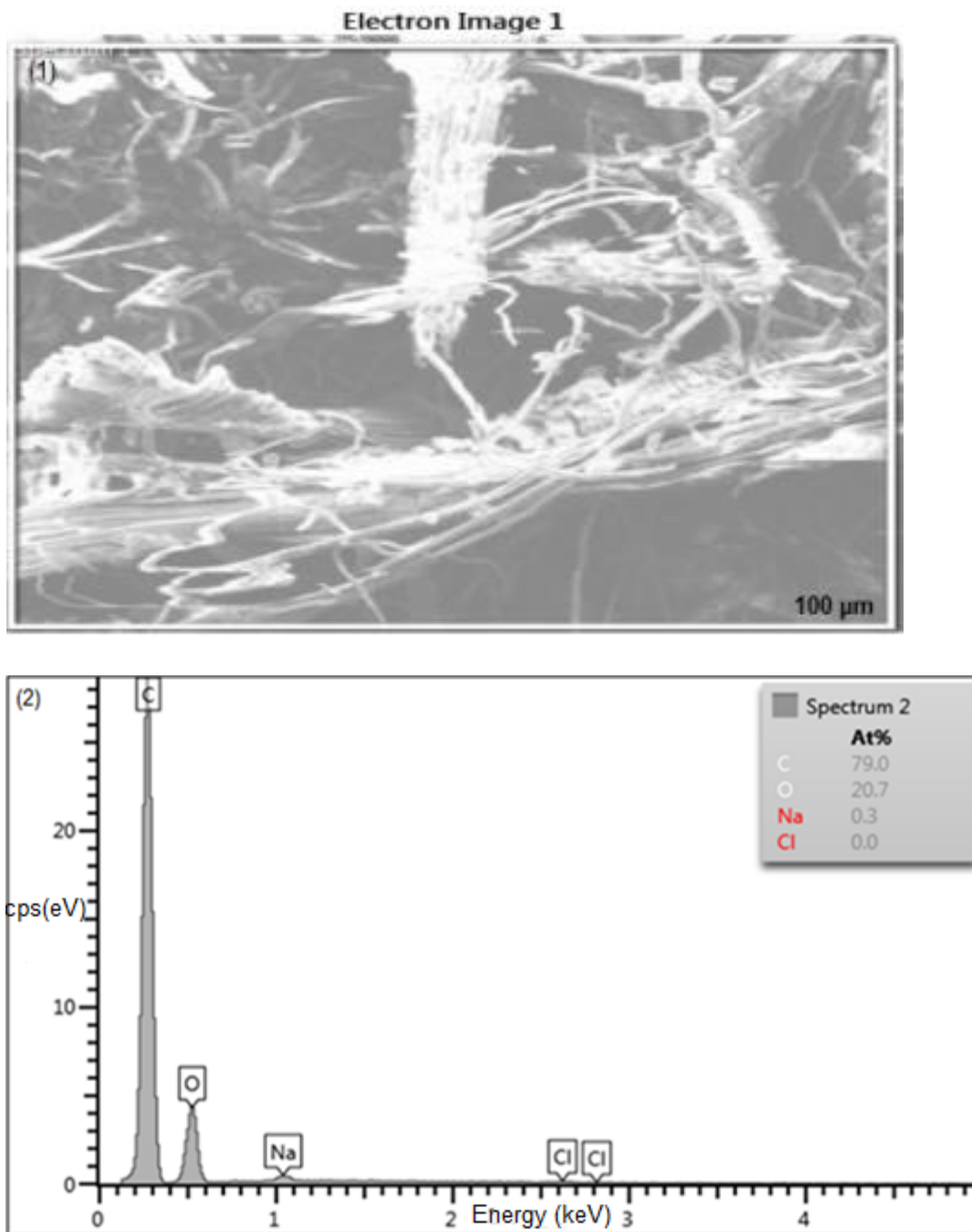
*et al.* 2014). This property suggests the strong potential of the cellulose in thermal insulating devices and domestic applications.

Figures 4a and 4b show the EDX spectra of the isolated cellulose particle sizes (300  $\mu\text{m}$  to 424  $\mu\text{m}$  and 600  $\mu\text{m}$  to 849  $\mu\text{m}$ ). The EDX spectra peaks correspond to the energy levels for which the carbon, oxygen, chlorine, calcium, and sodium were identified.

Carbon had the highest percentage among the elements observed in the spectra and sodium had the lowest.



**Fig. 4a.**(1) SEM image of isolated cellulose with particle sizes 300  $\mu\text{m}$  to 424  $\mu\text{m}$ , and (2) EDX spectrum of isolated cellulose with particle sizes 300  $\mu\text{m}$  to 424  $\mu\text{m}$



**Fig. 4b.** (1) SEM image of isolated cellulose with particle sizes 600 μm to 849 μm, and (2) EDX spectrum of isolated cellulose with particle sizes 600 μm to 849 μm

The elemental compositions for the sample with particle sizes 300 μm to 424 μm were 78.6% carbon, 21.1% oxygen, and 0.3% sodium. While, that obtained from the samples with a particle sizes 600 μm to 849 μm were 79.0% carbon, 20.7% oxygen, and 0.3% sodium.



## CONCLUSIONS

The isolation and characterization of cellulose of *Celtis philippensis* of two different size classifications were studied using XRD, EDX, and SEM. The results of the study revealed that:

1. The elemental composition of the isolated cellulose consisted of C, O, Ca, and Na.
2. The CrI values were 60.91% and 64.89% for cellulose with particle sizes 300  $\mu\text{m}$  to 424  $\mu\text{m}$  and 600  $\mu\text{m}$  to 849  $\mu\text{m}$ , respectively.
3. The samples revealed a crystal plane of preferred orientation along the (200) plane for the most prominent peak.
4. The crystal structure observed was orthorhombic for both ranges of particle sizes.
5. The isolated cellulose can find useful applications in the production of biomaterials, thermal insulating devices, *etc.*

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