Synthesis of Nanocellulose from Sugarcane Bagasse and Rubber wood: A Comparative Analysis

Madhuri R[1], Latha M S[2], Onkarappa H S[1], Pradeep H K[3] and
Virupaxappa S Betageri [1*]

[1] *Research Centre, Department of Chemistry, G M Institute of Technology, Davangere, Karnataka, India-577006.*

[2] *R L Science Institute (Autonomous), Belagavi, Karnataka, India*

[3] *Department of Pharmaceutics, GM Institute of Pharmaceutical Sciences and Research, Davangere.*

Abstract

Sugarcane Bagasse and Rubber wood are transformed into cellulose by pre-treating with $NaClO_2$ and NaOH. The obtained cellulose is converted into its respective Nano Cellulose (NC) through acid hydrolysis method and followed by high-intensity ultra-sonication. Synthesized NCs were characterized by Fourier Transformation Infrared spectroscopy (FT-IR), X-Ray Diffractometer (XRD), Transmission Electron Microscope (TEM), and Scanning Electron Microscopy (SEM). The thermal properties of the NCs were studied by Thermogravimetric and Derivative Thermal Analysis (TGA/DTA). FT-IR spectroscopy shows the principle chemical structure with the significant conversion of cellulose to NCs. XRD shows the morphological structure of the NCs having crystalline and amorphous nature. SEM and TEM analysis depict the surface morphology. In addition, the comparative study on the properties and morphology of NCs were detailed. The current work is not only expected to provide useful information about the synthesis of NCs but also provides interesting information on the role of acid hydrolysis on morphology and its properties.

Keywords

Acid hydrolysis, Nanocellulose, Rubber Wood, Sugarcane Bagasse, Cellulose

1.Introduction

Lately, researchers are uncovering diverse ways to make use of natural resources that are not exhaustible, but also capable to replace the place of polymers. Recently, the prime focus is on versatile lingo-cellulosic biomass that can be utilised in multiple ways based on the applications. But, in many countries the challenge of using agricultural residues in industrial sectors are still unaddressed. For instance, India stands at the top of the list for many of the crops. So, naturally, there is a huge availability of an unused source of cellulose. This unexploited form of cellulose is habitually either fed to animals or burnt to ash. But latest studies have marked the authenticity and reliability of cellulose concerning its aspect ratio, precise strength, rigidity, low toxicity, surface finish, biocompatibility excellent rheological and optical properties [1,2]. This provides several opportunities for researchers to investigate and examine cellulose and make use of it to replace conventional sources.

Cellulose is a homopolysaccharide constituted with AGU (D-anhydrous glucopyranose unit) which are connected with the aid of β-1, 4-glycosidic bonds [3]. It is a natural semi-crystalline polymer that has both crystalline and amorphous regions [4]. Due to the ubiquity of hydroxyl groups, it undergoes hydrogen bonding and thus it makes it tough naturally [5]. The presence of functional groups helps to convert cellulose to nanocellulose.

'Nanocellulose' (NCs) is the most advanced form of cellulose achieved with the aid of nanotechnology. In recent times numerous methods are being experimented with and explored using cellulose to obtain nanocellulose. But, which of the most recited methods are more conveniently fit and is eco-friendly remains questionable. The current investigation focuses on the use of agricultural bio-wastes and saw mill wastes to produce nanocellulose. It is utilised in various applications like drug delivery systems, tissue engineering, pharmaceutical production etc., [6]. Biomasses gathered from industries are usually comprised of hemicellulose, α-Cellulose, and lignin [7]. α-Cellulose is utilised to prepare nanocellulose and it can be prepared traditionally by either chemical or mechanical processes. However, it is suggested that to obtain a good result, the combination of a mechanical and a chemical process is more encouraged. The chemical approach involves TEMPO (2,2,2,6,6-tetramethylpiperidine-1-oxyl) oxidation, Acid Hydrolysis, and Ionic liquid method whereas the mechanical approach includes high-pressure homogenization and ultra-sonication [8].

In this current study, nanocellulose has been achieved by the practice of the Acid Hydrolysis (AH) method which dates back to the '90s [9]. Cellulose Nano Crystals (CNC) is the name coined for the nanocellulose obtained by AH, which is rod-like in construction with dimensions smaller than 50nm [10-11]. As affirmed previously, both crystalline and amorphous regions are present in cellulose and it is essential to note the necessity to convert the amorphous region into crystalline. This conversion is made with the aid of acid hydrolysis. When cellulose undergoes AH, it selectively oxidizes in the amorphous region only. This is due to the tight packing of the polymer chains and the orderly arrangement of the crystallites in the crystalline region [12].

Customarily, HCl and H_2SO_4 are the strong acids used, which generate a colloidal suspension [14]. Cellulose hydrolysis can be done efficiently using AH as it is cost-effective and time-efficient when compare to other methods. The nature of the nanocellulose obtained is based on the type of acid used in hydrolysis. NC obtained from H_2SO_4 yields a stable colloidal suspension when compared to HCl because there is more surface charge due to electrostatic repulsions. Thus, AH via H_2SO_4 is more practised when compared to methods like High-Pressure homogenization and Ionic Liquid. It is more convenient and the ability to reuse the acid makes it a green approach. The materials used here to extract cellulose are Sugarcane Bagasse and Rubber wood.

Sugarcane (SCB) is one of the significant major sources of sugar in India and is also prized for its medicinal properties and its ability to produce several by-products like cellulosic ethanol, molasses etc. [15]. Depending on the traits of climate Uttar Pradesh, Maharashtra, Karnataka, and Bihar are the top states in India to cultivate sugar cane with a total cultivation area of 52.28 lakh hectares. Sugar cane bagasse is the waste material obtained from alcohol and the sugar industries in generous quantity [16]. Rubber wood (RW) is another material that is widely grown in the different states of India - Kerala, Tamil Nadu, Assam, and Tripura mainly with a total of 648,000 tonnes of stock approximately per year. Rubber wood when used in furniture and toy making eventually results in powder or chunks of it, which is often discarded or burnt to ash. It cannot be fed to cattle, unlike sugar cane bagasse. Thus, this leads to the amplest availability of biopolymer often prone to burning. Sugar cane Bagasse and Rubber wood powder is used for cattle feeding or burning which are industrial and agricultural wastes. Both SCB and RW hold functioning optical materials, particle tracking, greenhouse plastics, and many more. They are more flexible, ultralight and transparent making them a prime focus for multiple applications [18].

In the present study, Sugar Cane Bagasse (SC), Rubber Wood (RW) and direct commercial cellulose are utilized to prepare nanocellulose through the Acid Hydrolysis process followed by high-intensity ultra-sonication. The nanocellulose are analysed for structural monography, thermal stability and comparative studies were examined by FTIR, XRD, SEM, TEM and TGA/DTA.

2. Materials and methods

Institute of Wood Science and Technology Bangalore, Karnataka, India has kindly presented rubber wood powder and Cellulose. Sugar Cane Bagasse were gathered in the vicinity of Davangere, Karnataka, India. Acetic acid (CH_3COOH), Sulphuric acid (H_2SO_4), sodium chlorite ($NaClO_2$), and sodium hydroxide ($NaOH$) were acquired from SD Fine Chemicals. All chemicals were used without any further purification and were of 98.0% pure.

2.1 Preparation of nanocellulose from Acid hydrolysis method

One of the major practised methods to secure nano-cellulose from diverse cellulosic materials is the Acid-Hydrolysis method [19]. In cellulose, there is the ubiquity of both ordered and disordered regions. When cellulose undergoes acid-hydrolysis the disordered regions present in it easily exposes themselves to hydrolysis leaving the ordered parts intact [20]. Among the many available acids, sulphuric acid is most used for acid hydrolysis, because of its ability to produce a dispersed solution with a stable colloidal system [21, 22]. It can also alienate nano-crystalline cellulose strongly. This is because of the hydroxyl groups experiencing esterification by the means of sulphate ions [22]. Acid concentration, reaction time, and temperature are the main factors affecting the properties of the synthesized cellulose.

Fig.1 shows the mechanism of cellulose undergoing acid-hydrolysis in two different paths. It is obvious from the mechanism that hydrolysis is followed by esterification. In path one, the hydrolysis of the amorphous chain cellulose involves accelerated protonation of glycosidic linkage, whereas in path two it is through the cyclic oxygen which is preceded by the separation of glycosidic bonds slowly, led by the addition of water. It is essential to note that, the innate structure of cellulose is retained while undergoing hydrolysis resulting in shorter fragments of the fibres.

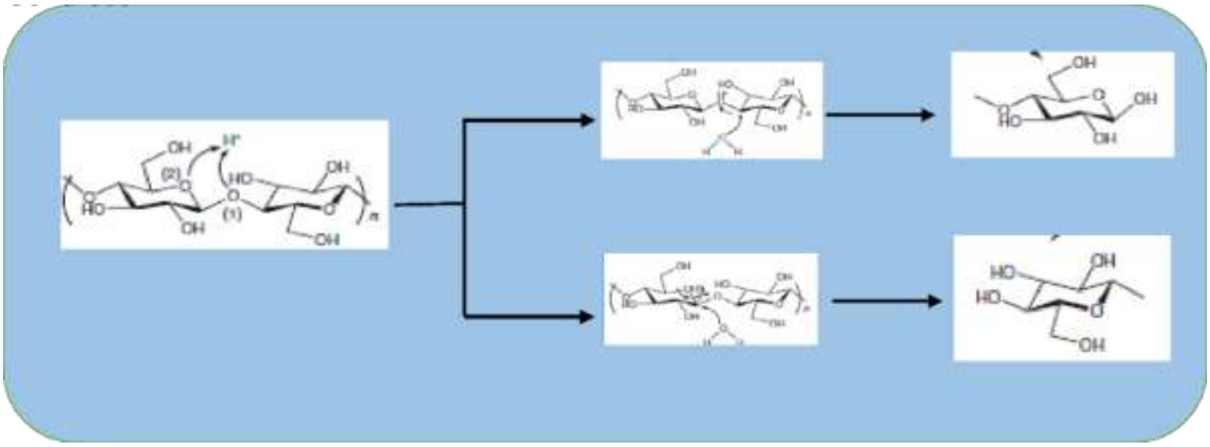

Fig.1 Mechanism of Acid Hydrolysis in Path-I and Path-II

2.2 Synthesis of cellulose from Sugar Cane Bagasse and Rubber wood

The Sugarcane Bagasse and Rubber wood fibers were thoroughly cleaned with distilled water. It was compelled into fine powder by drying and sieving. An adequate amount of RW and SCB were pre-treated in 5 % NaOH solution for about 3 hours under constant stirring, at 80 to 100 °C. The acquired celluloses were completely cleaned by washing them 4 to 5 times using distilled water till the effluent reached a neutral ph. The enhanced exposure of fibres is promoted by alkali treatment which equips the cellulose for acid treatment and bleaching. The obtained pre-treated husk was exposed to bleaching by using 5% of $NaClO_2$ solution for nearly 3 hours at 80 to 100°C in acidic ph. The residual lignin was excluded by washing repetitively with distilled H_2O. A neutral pH was achieved by repetitive filtering and washing using deionized water. The obtained cellulose was used for future study by drying it in an oven for about 24 hours and was stored in a dry place [8].

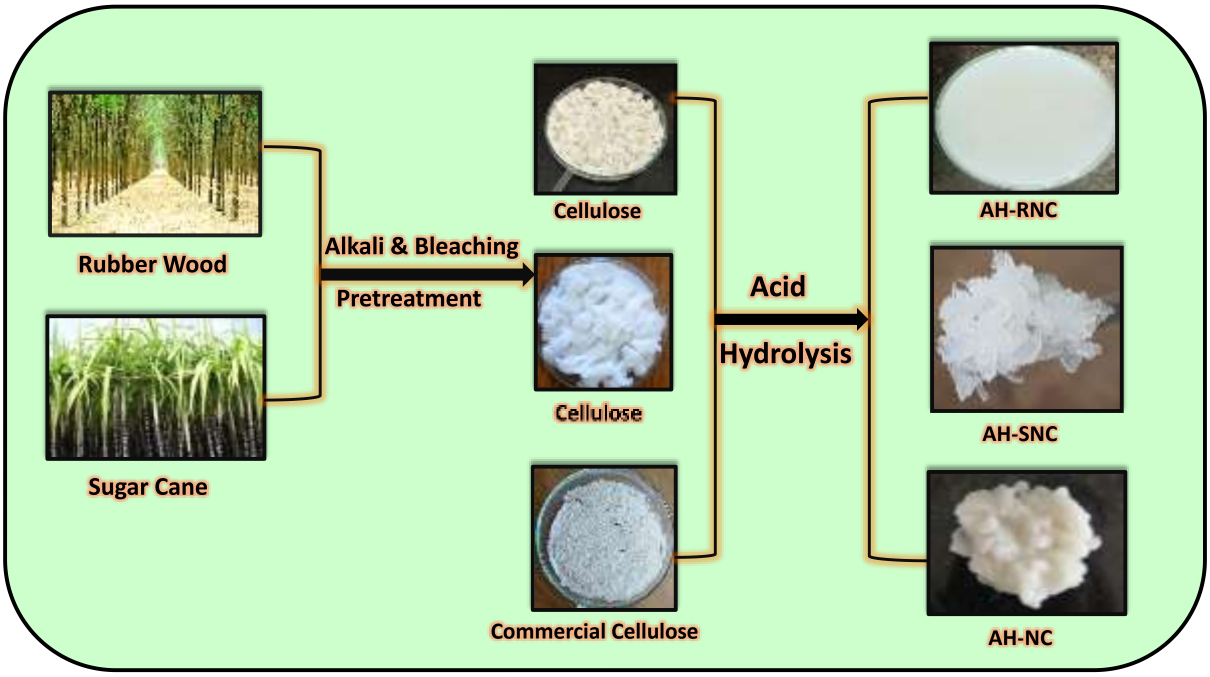

Fig.2 Schematic representation of Synthesis of nanocellulose from Sugar cane Bagasse, cellulose and Rubber wood

Nanocellulose (AH-NC), Sugarcane Nanocellulose (AH-SNC), and Rubber Wood nanocellulose (AH-RNC) were prepared according to the general procedure of acid hydrolysis.

The desired quantity of the respective celluloses was combined with 60% w/v 50 ml of sulphuric acid separately. The obtained mixtures were hydrolysed for 60 minutes at 45°C under continuous stirring. 100ml of cold water was added to the reaction mixture to arrest the hydrolysis. The residual slurry was washed by centrifuging it for 10 minutes repetitively. The resulting supernatant from the sediment was removed and substituted with distilled water. The procedure was followed until the pH of water reached 7 using 2% of NaOH. To achieve AH-RNC, AH-SNC and AH-NC the suspensions were ultra-sonicated for about 10 minutes and was subsequently stored at 4°C in a refrigerator for future use. The total yield of nanocellulose was marked to be 85% [8].

3. Characterization

AH-RNC, AH-SNC and AH-NC were measured as KBr pellets to infer the Fourier transform infrared spectra (FT-IR) by NICOLET 370 model in the range 4000-400 cm-1. Rigaku Miniflex 600 (5th gen) was used to record X-Ray Diffractogram. The measurement was performed using Cu Kβ with a step size of 0.02, and the crystallinity index (CI) was calculated by using Scherer's formula. Scanning electron microscopy (SEM) images were recorded on JOEL JSM 6390LV, with an acceleration voltage of 10 kV and a Secondary Electron (SE) detector was used to capture the images. Thermal analysis was done by using Perkin Elmer STA 6000 instrument for Differential Thermal Analysis and Thermogravimetric analysis. Model-Perkin Elmer, diamond TG/DTA, Temperature Range-Ambient +1200 °C, TG Measurement Range-200 mg, TG Sensitivity 0.2 mg, DTA measurement Range-1000 mV, DTA Sensitivity-0.06 mV, Programmable rate 0.01–100 °C/min, Sample pan volume-45 ml or 90 ml, single beam Temperature range 15 to 900 °C Temperature accuracy –b ± 0.5 °C Thermocouples-PT-PT/Rh (Type R).

3.1. Results and Discussion

3.1.1 Fourier Transformation and Infrared Spectroscopy (FT-IR)

FTIR spectroscopy was a non-fatal method for monitoring the physiochemical properties of lignocellulose materials to acknowledge the functional groups present in them. **Fig.3** depicts FT-IR spectra of Cellulose. AH-RNC, AH-SNC and AH-NC revealed the organic group presence. The absorption peaks showed by all the three nano celluloses were alike to each other. The absorption peaks at 3442, 3429 and 3352 cm^{-1} assigned to the CH2 groups, due to the attraction of intermolecular Hydrogens and stretching of the O-H group. The results also insinuated that there is the presence of polysaccharide aromatic ring and β-glycosidic linkage. The glucose units in cellulose registering that there was no remodification in the principal chemical structure. Thus, the employed technique did not affect the principal groups. The peaks at 2901, 2962 and 2923 cm^{-1} correspond to C-H groups. The peaks observed at 2140 and 2124 cm^{-1} which is not present in AH-SNC due to the excessive stretching experienced between C-C bonds. Furthermore, the peaks at 1637, 1635cm^{-1} are attributed to the conjugated C=O stretching vibration and H-O-H deformation of absorbed water. Small shoulder peaks at 1370, 1376 and 1380 cm^{-1} vibrations are corresponding to C-H groups. The intense peaks at 1113, 1109 and 1059cm^{-1} attribute to a strong C-O stretching.

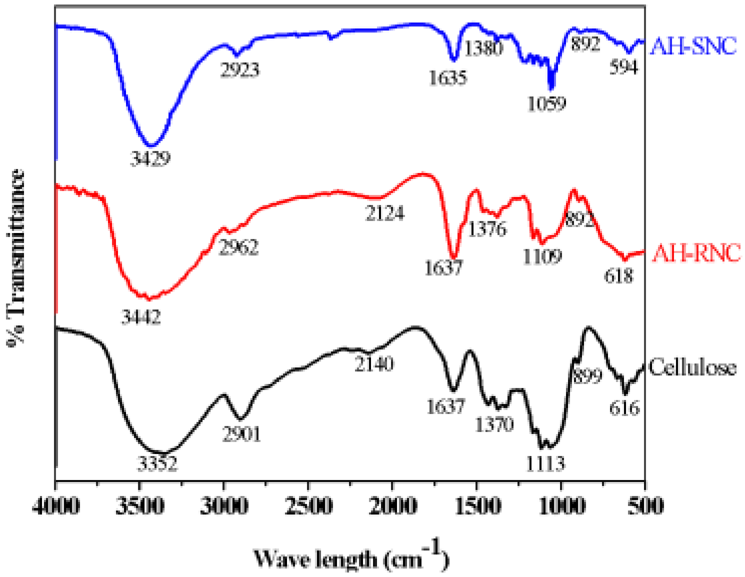

Fig.3. FTIR of Cellulose, AH-RNC and AH-SNC.

3.1.2 X-ray diffraction analysis

To study the crystalline structure of three NC samples obtained by Acid Hydrolysis X-ray diffraction was utilized. **Fig.4** shows the X-Ray Diffractogram of AH-RNC, AH-SNC and AH-NC. All three samples show the intense peak 2θ at AH-RNC-22.630, AH-SNC- 29.890 and AH-NC-22.690 which corresponds to the structure of Cellulose-I. This concludes that acid hydrolysis with ultra-sonication does not change the principal chemical structure of the cellulose. Besides, there is no selectivity in the cavitation effect when the samples underwent ultra-sonication. But it affects both amorphous and crystalline regions. Therefore, there is only a slight increase in the crystallinity of AH-RNC, AH-SNC and AH-NC which is chargeable to the distortion of the amorphous regions only because crystalline regions are inert to the chemical treatments [8, 20].

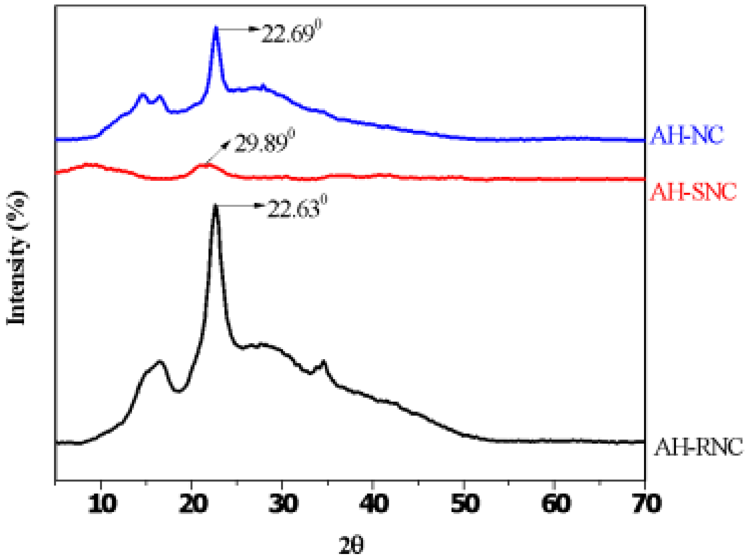

Fig.4. XRD pattern of AH-RNC, AH-SNC and AH-NC

3.1.3 Scanning Electron Microscopy

Change in surface morphologies of the AH samples were studied using SEM images. This lead to the revealing of the rod-like pointy structures of NC. The SEM images shown in **Fig.5** indicate good agglomeration and fibrous nature. All the samples were acid-hydrolysed at 45°C for 60 minutes.

Fig 5a and **5b** show the images of AH-RNC. It is evident from the monographs that the obtained nanocellulose is free of lignin and, its size has been reduced to nano-level. Furthermore, notes can be drawn that it has a low aspect ratio [17]. There are lesser impurities observed due to the removal of hemicellulose and lignin along with the cluster of micro-fibrils in huge number [18]. It has a non-uniform surface with lumens and an irregular cross-section. There is the presence of long fibrils which are tangled with the smaller ones creating a webbed assembly. But it can be marked that the size of the fibres is in nano-level.

Fig 5c and **5d** monographs of AH-SNC is shown. The particles are having a more uniform and smoother surface with no presence of impurities hence lacking any valley formation [19]. The reason being the repetitive washing of the nanocellulose and ultra-sonication. The size distribution has increased commendably because of the subtraction of the amorphous regions in the crystallinity chain. It should be noted that, even though RW and SCB were hydrolysed in similar conditions and temperatures, the resulting yield is slightly different in the surface study.

Fig 5e and **5f** is related to the AH-NC showing the formation of some crystallites and is similar in shape. To conclude, it can be undoubtedly stated that all the samples were successfully converted in nano terms with the aid of the employed method that gave commendable results. The three samples have a different shape owing to the nanoscale and are fibrous in nature.

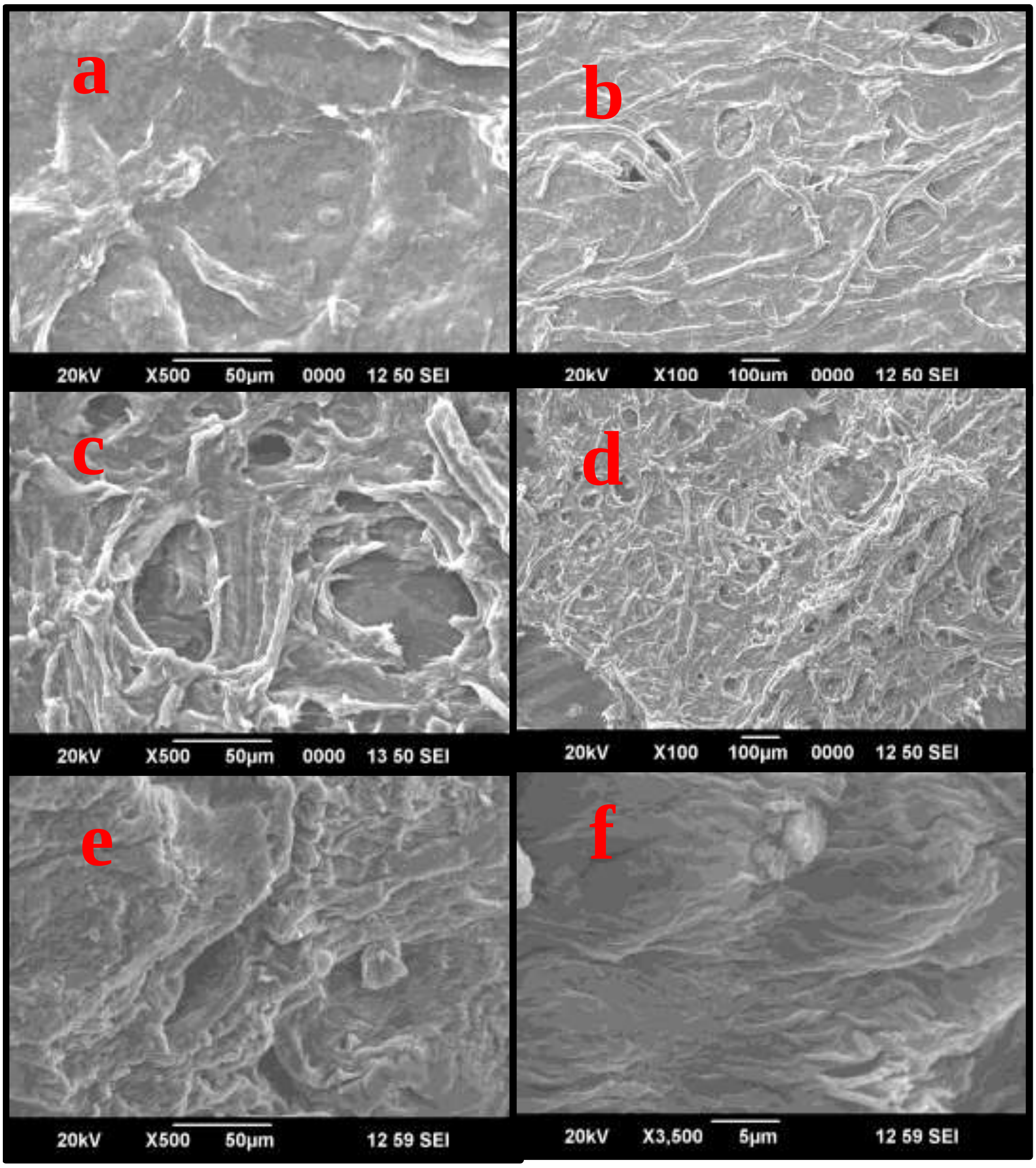

Fig.5. SEM Monographs of (a & b) AH-RNC (c & d) AH-SNC and (e & f) AH-NC

3.1.4 Transmission Electron Microscopy

To study the internal arrangement and surface morphologies of the prepared nanocelluloses TEM analysis was used. **Fig 6(a, b), 6(c, d)** and **6(e, f)** are the TEM images of AH-RNC, AH-SNC and AH-NC respectively. It is observed that, the formation of elongated chains fall in nano-scale. The nano-fibres obtained are finely separated from one another.

They have several nano-fibrils that are rod-like structure. The size of the prepared nanocelluloses of AH-RNC, AH-SNC and AH-NC are in the range of 33-45nm, 21-23nm and 38-40nm respectively thus, proving that they are in nano-scale. Over the SEM results TEM clearly show the strong agglomeration of the nano-cellulose compounds. The rod-like structure of the nano-cellulose is due to the fact that the hydrogen bonds present in the compound have overcome the repulsions experienced due to surface negative charges.

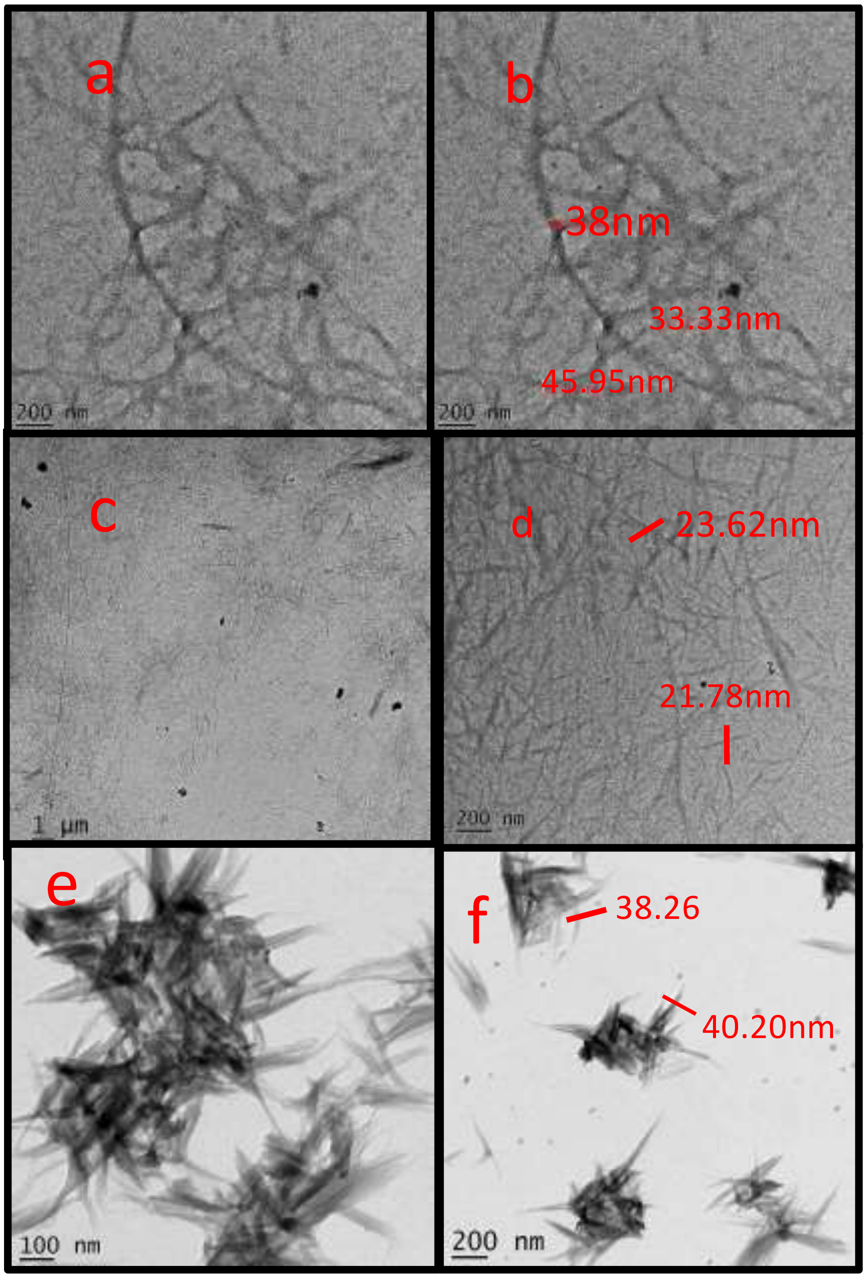

Fig.6. TEM Images of (a & b) AH-RNC (c & d) AH-SNC and (e & f) AH-NC

4.5 Thermogravimetric analysis and Differential thermal analysis (TGA/DTA).

By making use of Derivative Thermogravimetric (DTA) and Thermogravimetric Analysis (TGA) the thermal and morphological stability along with the properties of the three samples AH-RNC, AH-SNC and AH-NC were investigated. The curves obtained from TGA and DTA are shown in **Fig.7** and **Fig.8** respectively. Due to the evaporation of water present in the nanocellulose, around 100^{o}C all three samples show evident weight loss [8]. The degradation temperature of the three samples was observed around 98°C, 330°C and 210°C for AH-RNC, AH-SNC and AH-NC respectively. This degradation can be observed in the small troughs to the left of the graph in **Fig.7.** AH-RNC displays loss of weight substantially in the region of

90-100°C this is due to the rupture of the glycosidic linkages and de-polymerization of the hemicellulose. Whereas the onset temperature of the AH-RNC is at 90°C which is hydrolysed using H_2SO_4 is seen in the deep troughs of the DTA curve. AH-SNC shows a major loss in its weight at 330°C marking the excellent thermal stability. AH-NC shows weight loss at 210°C. Hence from the outcomes of the TGA analysis, it is evident that AH-SNC has greater thermal stability that is superior to AH-RNC and AH-SNC with higher degradation temperature.

The DTG curves obtained for the three samples AH-RNC, AH-SNC and AH-NC are shown in **Fig.8.** The nanocelluloses AH-RNC and AH-NC show only one main stage of cellulose decomposition whereas AH-NC shows two stages of decomposition. This factor is due to the total degradation of nanocellulose during its chemical pre-treatment. The DTG curves of AHRNC, AH-SNC are observed at around 95-100°C and 280-340°C whereas AH-NC undergoes decomposition twice thus has two falls in the graph at 200-280°C and 430-490°C. Here, the first and second peaks are assigned to degradation of sodium anhydroglucuronate units and decomposition of cellulose chains respectively [19].

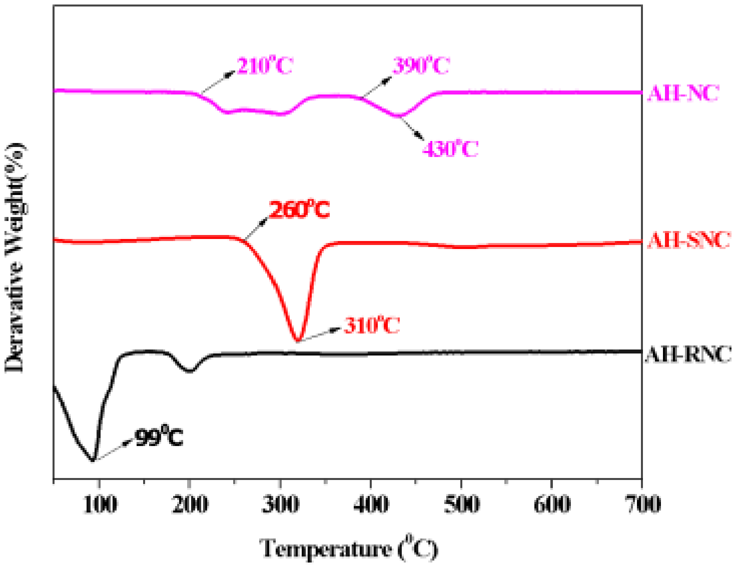

Fig.7. DTG Thermogram of AH-RNC, AH-RNC and AH-RNC

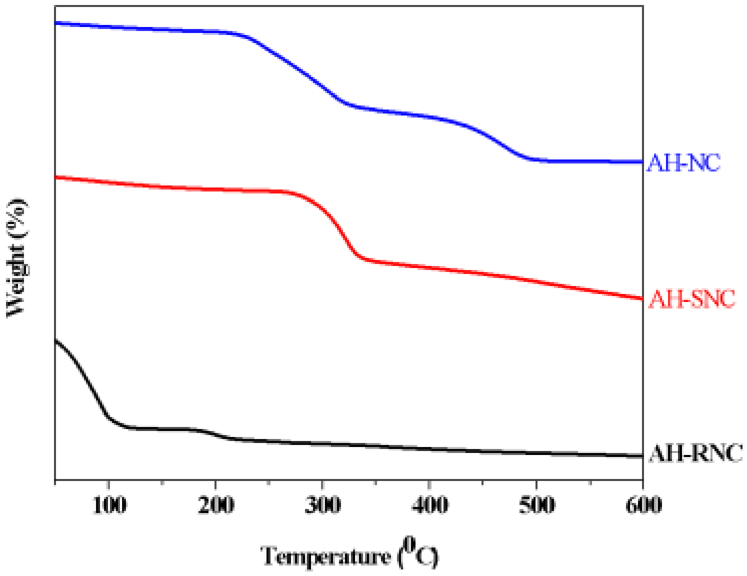

Fig.8. DTA Thermogram of AH-RNC, AH-SNC and AH-NC

Conclusion

In the present work, an effort has been made to study the nanocelluloses (NC) isolated from Rubber-wood (RW) and Sugarcane Bagasse (SCB) through Acid Hydrolysis. A comparative study of nanocelluloses obtained from standard nanocellulose has been studied. The properties and morphologies of the produced nanocellulose are investigated by the application of the synthetic method. FTIR spectra attribute the functional region of AH-RNC, AH-SNC and AH-NC shows characteristic cellulose peaks. AH-SNC showed greater crystallinity compared to the other cellulose which underwent acid-hydrolysis. The fibrillar and the fibrous regions are seen in all the SEM monographs of the samples. AH-SNC excelled the other nanocellulose in terms of good aspect ratio, thermal stability with increased absorption efficiency and stability in dispersion. Among the three obtained nanocellulose, AH-SNC is a more convincing material fit for further use in the production of nanomaterial composites and value-added products. This gives a new way to make use of the naturally available biomass in nanomaterial applications. The obtained nanocellulose can be effectively used in the production of thin films with antimicrobial activity, which can be used in water purification, pharmaceuticals, nanochip production, biodegradable nano-composites and many more. Among the three synthesized NC, AH-SNC has superior thermal stability and a commendable aspect ratio.

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