Carbohydrate Polymers 80 (2010) 852–859

Contents lists available at ScienceDirect

Carbohydrate Polymers

journal homepage: www.elsevier.com/locate/carbpol

Cellulose microfibres produced from banana plant wastes: Isolation and characterization

Silviya Elanthikkal [a], Unnikrishnan Gopalakrishnapanicker [a,*], Soney Varghese [b], James T Guthrie [c]

[a] Department of Chemistry, National Institute of Technology, Calicut 673 601, India
[b] School of Nanoscience and Technology, National Institute of Technology, Calicut 673 601, India
[c] Department of Colour Science, School of Chemistry, University of Leeds, LS2 9JT, UK

ARTICLE INFO

Article history:
Received 20 November 2009
Accepted 23 December 2009
Available online 7 January 2010

Keywords:
Banana fibre waste
Cellulose microfibres
Zeta potential
Crystallinity studies

ABSTRACT

Cellulose microfibres from banana fibre waste have been isolated and characterised. Bleached banana waste fibres were hydrolysed, under different conditions, to study the effects of temperature, reaction time, and acid concentration on the properties of the resultant cellulose microfibres. As the concentration of acid used in the hydrolysis was increased, more stable aqueous suspensions of the cellulose product were obtained and the dimensions of the resulting cellulose microfibres were reduced. XRD studies reveal that cellulose prepared by such hydrolysis was more crystalline than the banana fibres. The effect of initial dimension of banana fibres on the size of cellulose microfibres was also examined.

1. Introduction

Over the past decades there has been a growing interest in the use of natural fibres as reinforcing fillers in polymer matrices (Mohanty, Misra, & Hinrichsen, 2000; Samir, Alloin, & Dufresne, 2005). The fibres can be utilised for reinforcement in the long form, short form or in a derived form.

The banana plant fibres are fibrous residue of pseudo-stems and leaves left over after banana cultivation. India is one of the larger producers of banana plants. Banana cultivation generates a considerable amount of cellulosic-based waste. The banana fibre wastes generated is lignocellulosic in nature. The comestible part, the fruit, constitutes only 12% by weight of the plant. The remaining parts become agricultural waste, causing environmental problems in banana farming regions. This residual resource is rich in cellulose, a feature that has attracted much interest due to the potential use of such materials as a reinforcing component in high performance composite materials.

The use of nanoparticles and microparticles as reinforcements in high performance composites and other structural materials has attracted interest (Gacitua, Ballerini, & Zhang, 2005). In the last decade, effort has been placed on developing nanofillers from various natural resources. Cellulose has emerged as a strong candidate for providing nanoparticles for use as a reinforcing agent.

Microcrystalline cellulose (MCC) is widely used, especially in the food, cosmetic and medical product industries. Such applications involve MCC acting as one or more of the following ways: as a water-retainer, as a suspension stabilizer, as a flow characteristics controller in the systems used for final products and as a reinforcing agent for products such as medical tablets. MCC is obtained on an industrial scale through the hydrolysis of wood cellulose and of cotton cellulose using dilute mineral acids. The preparation of MCC from wheat and cereal straws (Alemdar & Sain, 2008; Jain, Dixit, & Varma, 1983), jute (Abdullah, 1991), soybean husk (Nelson, Edgardo, & Ana, 2000), flax fibres and flax straw (Bochek, Shevchuk, & Lavrentev, 2003), sugar cane bagasse (Bhattacharya, Grminario, & Winter, 2008), mulberry barks (Li et al., 2009) and peel of pear fruits (Habibi, Mahrouz, & Vignon, 2009) has been studied.

The controlled acid hydrolysis of native cellulose fibres yields highly crystalline, rod-like particles through the selective degradation of the more accessible material. Depending on their origin, these microfibrils differ in lateral size. Upon the action of strong acids, these fibres break down into short crystalline rods or cellulose microcrystallites, having shorter lengths, ranging from a few hundreds of nanometers to a few microns.

Acidic hydrolytic cleavage is dependent on the acid species, the acid concentration, the time for hydrolysis and the temperature of the hydrolysis reaction. Under controlled conditions, cellulose microcrystals can be obtained using sulphuric acid hydrolysis. This process induces the grafting of the sulphate groups, randomly distributed on the cellulose microfibril surface, providing a negative electrostatic layer, covering the microfibrils. The different treatments of these charged microcrystallites, such as mechanical dispersion or ultrasonication, permits the dispersion of the aggregates and finally produces colloidal suspensions. Because of

* Corresponding author. Tel.: +91 495 2286560; fax: +91 495 2287250.
E-mail address: unnig@nitc.ac.in (U. Gopalakrishnapanicker).

0144-8617/$ - see front matter © 2010 Elsevier Ltd. All rights reserved.
doi:10.1016/j.carbpol.2009.12.043

their stiffness, thickness, thickness distribution, length and length distribution, these rod particles are commonly called "whiskers" (de Souza Lima & Borsali, 2004). The controlled hydrolysis conditions with sulphuric acid (temperature, time, and acid content) lead to whisker suspensions that neither precipitate nor flocculate. This effect is mainly caused by electrostatic repulsion between the negatively charged particles on their surfaces.

Zhang et al., have proposed a method for the synthesis of cellulose nanospheres from cotton fibres by using mixed acid treatment (6:1:3 = water:HCl:H_2SO_4) (Zhang, Elder, Pu, & Ragauskas, 2007). It was observed that the product mixture consisted of two different particle size species, averaging approximately 500 nm and 70–200 nm. The study revealed that cellulose nanoparticles of smaller sizes could be obtained by a further acidic sonication of the initially sonicated cellulose fibres. The authors pointed out that there was a linear relationship between the size of the cellulose nanoparticles and the treatment time. Another observation was that the initial cellulose sample was cellulose I, whereas the obtained cellulose spherical particles were of cellulose II polymorphic character. Bhattacharya et al. have reported the isolation of cellulose microfibrils from bagasse, a sugar cane by-product by using 60 wt.% sulphuric acid (Bhattacharya et al., 2008). The authors reported that bagasse was more resistant to hydrolysis than were the tunicate, bacterial and wood celluloses. Dong et al., examined the effect of the preparation conditions (time, temperature and ultrasound treatment) on the resulting microcrystalline cellulose structure from the sulphuric acid hydrolysis of cotton fibres (Dong, Revol, & Gray, 1998). The authors found a decrease in microcrystalline cellulose fibre length and an increase in the surface charge of the particles with prolonged hydrolysis times.

It has been reported that cellulose whiskers prepared by sulphuric acid hydrolysis are more stable than those prepared using hydrochloric acid (de Rodriguez, Thielemans, & Dufresne, 2006). Indeed, the sulphuric acid prepared whiskers present a negatively charged surface. The hydrochloric acid prepared whiskers are not charged. Another way to achieve charged whiskers consists of the oxidation of the surface of the whiskers (Araki, Wada, & Kuga, 2001; Isogai & Kato, 1998) or the post-sulphonation of the hydrochloric acid prepared microcrystalline cellulose (Araki, Wada, Kuga, & Okano, 1999).

Zuluaga et al. have isolated cellulose microfibrils from banana rachis, using a combination of chemical and mechanical treatments (Zuluaga, Putaux, Restrepo, Mondragon, & Ganan, 2007). The chemical treatment involved treatment of bleached banana rachis residues with a mixture of 80% acetic acid solution and 70% nitric acid solution at 120 °C for 15 min. The washed and purified cellulose was sonicated for 15 min. In the mechanical process, a bleached residue was suspended in water and homogenized. It was noted that an acidic treatment resulted in shorter aggregates of parallel cellulose microcrystallites. Individualised or bundled microfibrils were obtained by homogenization. In another report, banana pseudo-stem fibres have been used for the preparation of sodium carboxymethylcellulose (Mario, Marseno, & Haryadi, 2005).

The objective of the present study was to examine the possibility of using sulphuric acid hydrolysis method for the preparation of cellulose microfibres from banana fibres and to characterise the resultant microfibres. The effect of preparation conditions such as time, temperature, acid concentration and initial dimension of the banana fibres, on the cellulose microfibre structure and characteristics has also been investigated.

2. Experimental

2.1. Materials

The banana fibres used as the source of cellulose fibrils were supplied by the Fibre Design Centre, Khadi and Village Industries Commission (KVIC), Trivandrum, India. Sulphuric acid (>98%) and the other chemicals that were used in the pre-treatment of banana fibres were of analytical purity, obtained from Fisher Scientific, UK.

2.2. Isolation of cellulose microfibres from banana fibres

The cellulose microfibres were generated from the banana fibres by an acid catalysed hydrolysis route. The banana fibres were ground to approximately 500 μm in size, using a cutting mill, (Retsch GmbH SM1, Germany), with a 500 μm stainless steel trapezium shaped sieve. The isolation of cellulose fibres required the removal of other components such as lignin, hemicellulose and pectin from the banana fibres. This step was achieved by the use of an alkaline solution treatment and bleaching. The alkaline solution treatment was designed to solubilise the pectins and the hemicelluloses. Banana fibres were treated with 1 M NaOH, at 80 °C, for 4 h (Bhattacharya et al., 2008). Solubilised components were then removed by washing with deionised water. The bleaching treatment was performed to break down phenolic compounds or molecules having chromophoric groups present in lignin and to remove the by-products of such breakdown, to whiten the pulp. This is achieved by treating the banana fibres with 5% NaOCl, at 30 °C, for 3 h (Mario et al., 2005). During bleaching, the lignin was oxidised and became soluble in the alkaline medium. After this pre-treatment, the cellulose pulp was washed with deionised water until pH of 7 was reached. The centrifuged residue was then subjected to controlled acid hydrolysis to provide the required cellulose microfibres. After hydrolysis, the mixture was diluted ten-fold with distilled water and neutralised with an alkaline solution, followed by centrifugation. The aqueous suspension of the residue was dialysed against deionised water and sonicated for 30 min in an ultrasonic bath. This suspension was freeze dried (LSL Secfroid, Switzerland). For the sake of comparison of effect of drying on the morphology, one of the sample suspensions (sample 4A45) was subjected to both freeze drying and spray drying (BUCHI 190 Mini Spray dryer, Switzerland). The dried products were then stored in a desiccator.

There exist reports concerning the hydrolysis of cotton, black spruce, flax fibres etc. by using 64 wt.% H_2SO_4 at 45 °C (Beck-Candanedo, Roman, & Gray, 2005; Cao, Dong, & Li, 2007; Viet, Beck-Candanedo, & Gray, 2007). The size reduction was shown to be slight when 64 wt.% H_2SO_4 was used to hydrolyse the banana fibres. Therefore, in the present work, the banana fibres were treated with H_2SO_4 across a range of higher concentrations by varying reaction times, under different controlled reaction temperatures.

To study the influence of the acid concentration in the hydrolysis reaction, banana fibres were treated with solutions of sulphuric acid in water that contained 76, 70 or 64 wt.% of sulphuric acid, respectively. In the following discussion, the resulting samples have been referred to as A (76% sulphuric acid used), B (70% sulphuric acid used) and C (64% sulphuric acid used), respectively (Table 1). The influence of initial dimensions of the banana fibres on the hydrolysis reaction was also evaluated (Table 2). The ground banana fibres were sieved using test sieves of 355 μm, 250 μm, 150 μm and 38 μm aperture sizes (Endecotts Ltd., England). These sieved fibres were hydrolysed under the same conditions as those used for sample 4C45 (Table 1). The respective sieved samples have been termed as 355-4C45, 250-4C45, 150-4C45 and 38-4C45. The characteristics that were monitored were the size, the morphology, the change in crystallinity of the cellulose microfibres and the zeta potential of the aqueous suspensions of microfibres.

2.3. Characterisation

2.3.1. Zeta potential measurement

The zeta potential is an important parameter for examining dispersion stability. The zeta potential of the cellulose suspensions that

Table 1
Effect of reaction conditions on the acid hydrolysis of banana fibres.

Sample code	Acid concentration (wt.%)	Temperature (°C)	Treatment time (Hours)
4A45[a]	76	45	4
4B45[b]	70	45	4
4C45[c]	64	45	4
4C65	64	65	4
6C65	64	65	6
8C65	64	65	8

[a] 4, treatment time in hours; A, acid concentration of 76 wt.%; 45, reaction temperature in °C.
[b] 4, treatment time in hours; B, acid concentration of 70 wt.%; 45, reaction temperature in °C.
[c] 4, treatment time in hours; C, acid concentration of 64 wt.%; 45, reaction temperature in °C.

Table 2
Effect of initial dimension of banana fibres on acid hydrolysis.

Sample code	Initial size of banana fibre (μm)	Acid concentration (wt.%)	Temperature (°C)	Treatment time (Hours)
355-4C45	>355	64	45	4
250-4C45	355–250	64	45	4
150-4C45	250–150	64	45	4
38-4C45	106–38	64	45	4

were prepared by the above mentioned methods was measured using a Malvern 3000 Zetasizer. Aqueous suspensions of the cellulose samples (2.5 mg/cm^3) were prepared using the freeze dried samples. The mobility of the particles undergoing electrophoresis was measured using dynamic light scattering studies. This measured electrophoretic mobility was then converted to the zeta potential.

2.3.2. FT-IR spectroscopy

FT-IR spectroscopy was used in examination of changes in the chemical composition of the cellulose fibres after chemical treatment. The untreated banana fibres and the dried powders that were derived from the aqueous suspension of treated banana fibres were analysed. The cellulose samples that were prepared using the different reaction conditions were also evaluated by FT-IR spectroscopy. A Perkin–Elmer Spectrum One spectrophotometer was used to provide the spectrum of each sample. Spectra were taken at a resolution of 4 cm^{-1}, with a total of 100 scan for each sample. The FT-IR spectrum of each sample was obtained in the range of 4000–400 cm^{-1} in the transmission mode.

2.3.3. Thermogravimetric analysis

Thermogravimetric analysis was performed in a study of the thermal characteristics of the cellulose microfibre samples. The thermal behaviour of each freeze dried samples (~10 mg) was determined, using a TGA 2050 series thermogravimetric analyzer (TA Instruments), across a temperature range of 30–500 °C, at a heating rate of 10 °C/min, in a nitrogen environment (Purge rate details: Balance chamber flow rate = 30 cm^3/minute, furnace flow rate = 150 cm^3/minute).

2.3.4. Scanning electron microscopy

The morphology and particle size of the various cellulose microfibre samples were investigated by using scanning electron microscope (SEM) and field emission scanning electron microscope (FESEM). For the cellulose microfibre samples, freeze dried samples and spray dried samples were used. The samples were sputter coated with gold to avoid subsequent charging before measurement by SEM. Images were taken on a JEOL JSM-820 model SEM and on a Hitachi SU6600 FESEM.

2.3.5. XRD analysis

XRD measurements were performed on a PANalytical X'Pert PROMPD system. The diffracted intensity of Cu Kα radiation (0.154 nm, 40 kV and 30 mA) was measured in a 2θ range between 5° and 70°. Ground banana fibres and the hydrolysed cellulose samples were subjected to crystallinity analysis.

3. Results and discussion

3.1. Zeta potential measurement

Zeta potential measurements were carried out on aqueous dispersions of relevant microfibrous cellulose samples, prepared under the specified hydrolysis conditions. Relevant data are given in Table 3. All of the cellulose suspensions, in neutral water, showed a negative zeta potential value. Among the cellulose samples, sample 4A45, generated using the 76 wt.% sulphuric acid solution, gave the highest negative value. In the C series (cellulose samples prepared by using the 64 wt.% sulphuric acid solution) the zeta potential increases with increase in the temperature of the hydrolysis reaction. From these results, it is clear that the influence of temperature is dominant, being more significant than the reaction time at a given temperature. For the cellulose samples that were prepared using the highly concentrated acid, a greater value of zeta potential was observed. It is known that insufficient hydrolysis of cellulose may result in larger particles (less surface area per unit mass) with a lower mean surface charge, favouring particle–particle interaction (Hebeish & Guthrie, 1981).

3.2. FT-IR spectroscopic analysis

The FT-IR spectroscopic analyses of the untreated banana fibre, the alkaline solution treated banana fibre, the bleached banana fibre and the hydrolysed banana fibre samples (Fig. 1a) reveal that compositional changes occur in the fibre structure during hydrolysis. The prominent peak at 1731 cm^{-1} that is seen in the spectrum of the untreated banana fibre can be attributed either to the acetyl and uronic ester groups of the hemicelluloses or to the ester linkage of carboxylic group of the ferulic and p-coumeric acids of lignin and/or hemicelluloses. No equivalent peak is displayed in the FT-IR spectra of the chemically treated banana fibres, indicating the removal of most of the hemicelluloses and lignin from the fibres, by the applied chemical extraction. The peaks at 1513 cm^{-1}, in the spectrum of the untreated fibres represent the aromatic —C=C— stretch of the aromatic rings of lignin. The lack of this peak in the treated fibres is attributed to the removal of lignin during treatment. The lack of bands at 1731 cm^{-1} and at approximately 1513 and 1239 cm^{-1} implies the effective removal of lignin, pectin and hemicelluloses in the treated fibres. The peak at 1425 cm^{-1} is due to the —CH_2— bending. The sharp peak observed at 1313 cm^{-1} indicates —C—H asymmetric deformations. The peaks in the region from 1200–950 cm^{-1} are due to —C—O— stretching. The —C—O—C— pyranose ring skeletal vibration gives a prominent band at 1022 cm^{-1}. The increase in intensity of the band at 1022 cm^{-1} shows the increase in the cellulose content. The

Table 3
Zeta potential values of aqueous suspensions of cellulose.

Sample	Zeta potential (mV)
4A45	−41.32
4B45	−22.35
4C45	−20.72
4C65	−28.53
6C65	−29.67
8C65	−32.01

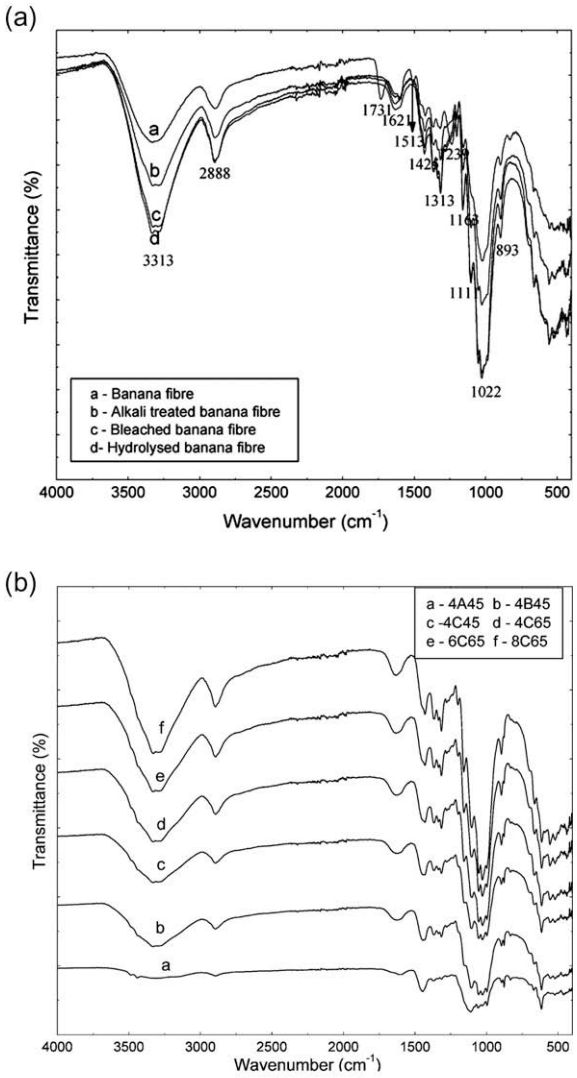

Fig. 1. (a) FT-IR spectra of banana fibres after different treatments. (b) FT-IR spectra of cellulose microfibres produced under different reaction conditions.

893 cm^{-1} band in the FT-IR spectrum of the chemically treated banana fibres is typical of the structure of cellulose. The small, sharp band at 893 cm^{-1} represents glycosidic —C_1—H deformation, with a ring vibration contribution and —O—H bending. These features are characteristic of β-glycosidic linkages between the anhydroglucose units in cellulose (Alemdar & Sain, 2008).

Fig. 1b shows the FT-IR spectra of the cellulose microfibres that were prepared by using the different reaction conditions. All the samples prepared under varying reaction conditions gave very similar spectral patterns to that of cellulose. The only difference observed was in the peak intensity, especially the intensity at 3600–3000 cm^{-1}, which corresponds to the –O–H group; varying for each sample. This implies either that the moisture content in each of the samples may differ, or that, as a result of hydrolysis, cellulose chain scission occurs to various extents, leading to different amounts of free –O–H groups being present. This effect can be appreciated from consideration of the decrease in the broadening of the hydroxyl peak.

3.3. Thermogravimetric analysis

Fig. 2 gives the consequences arising from the heating and subsequent pyrolytic degradation of the cellulose microfibre samples that were produced from banana fibres of different dimensions. There was a weight loss of about 3%, during heating to 120 °C. This corresponds to the vaporisation and removal of bound water in the cellulose samples. The cleavage of the glycosidic linkages of cellulose, leading to the formation of H_2O, CO_2, alkanes and other hydrocarbon derivatives, occurs in the temperature range of 230–370 °C. After 380 °C, the residual decomposition products maintain a slow degradation profile (El-Sakhawy & Hassan, 2007). Differences in the decomposition profiles, for the four samples, indicate the slight differences in the thermal stability of the samples. However, the final residue values are very similar.

3.4. Scanning electron microscopy

Fig. 3a shows an SEM image of banana fibres after they had been subjected to alkaline solution treatment and bleaching. Fig. 3b and c displays the SEM images of a spray dried cellulose sample and freeze dried cellulose sample respectively. These samples were obtained by acid hydrolysis, using 76 wt.% sulphuric acid, as described earlier. The spray dried sample, (4A45), shows doughnut shaped species that are well defined and separated. The freeze dried sample, (4A45), appears as an aggregation of flake assemblies of microfibres.

In Fig. 3a, the pre-treated banana fibres appear as long narrow fibrils of reasonably uniform length of size between 100 μm and 1 mm, approximately. The fibre diameters are reasonably consistent at a value of ~15–20 μm.

Fig. 3b represents the cellulosic component as discs that are reasonably uniform in shape and size. The doughnut shaped assemblies are between 2 μm and 15 μm in diameter. However, the proportion of larger particles is small. The doughnut shaped particles, shown in Fig. 3b (sample 4A45), arise as a consequence of the spray drying process (Chaubal & Popescu, 2008). In this process, droplets of cellulose–water dispersion are sprayed from an orifice into the drying chamber. These very small droplets soon lose their aqueous component. However, the rules of surface phenomena for a fluid composition must apply (smallest volume per unit mass). This leads to the formation of the spherical array that becomes distorted on further drying.

At a first glance, Fig. 3c, (4A45), representing freeze dried cellulose species, shows the particles as stacked flakes. However, the fibrillar structure of each of the particles can be discerned, as can the hollow nature of ends of fibril bundles. Fig. 3d shows magnified image (FESEM) of the same, which contains cellulose fibrils of width 40–100 nm. From the figures it is clear that each stage of

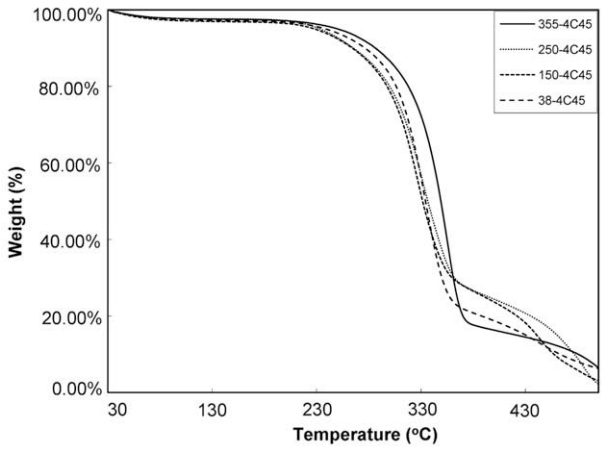

Fig. 2. TGA curves of cellulose microfibres derived from banana fibres of different dimensions.

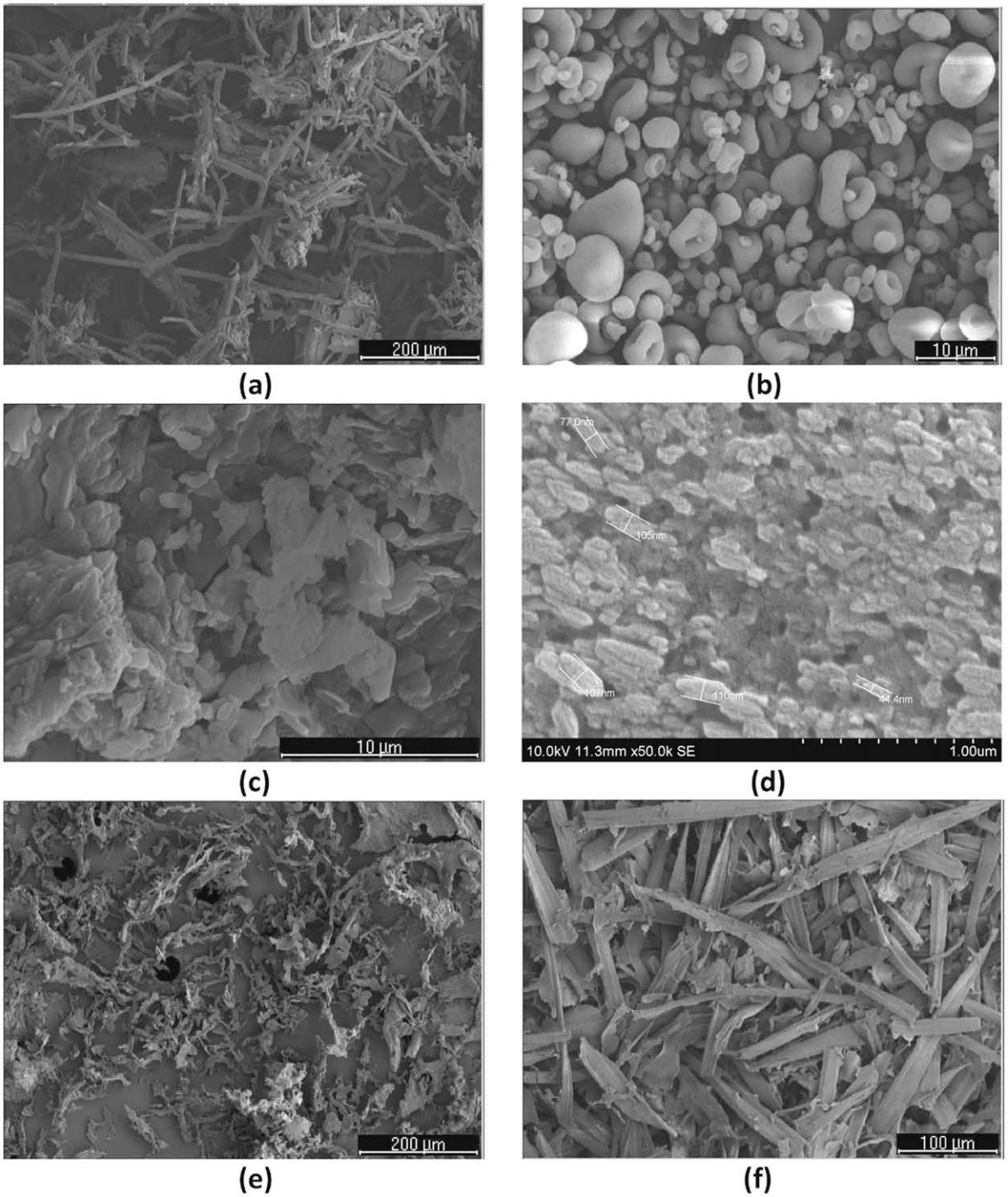

Fig. 3. (a) SEM image of pre-treated banana fibres. (b) SEM image of spray dried cellulose sample – 4A45. (c) SEM image of freeze dried cellulose sample – 4A45. (d) FESEM image of freeze dried cellulose sample – 4A45. (e) SEM image of freeze dried cellulose sample – 4B45. (f) SEM image of freeze dried cellulose sample – 4C45.

the treatment produces cellulosic fibrils that are markedly different in assembly. However, the fundamental physical and chemical nature of each fibril may not be compromised. The particles are seen as different assemblies of cellulose fibrils.

Fig. 3c, e and f shows the SEM images of cellulose fibres that were generated by the hydrolysis reaction, using sulphuric acid at different concentrations (76, 70 and 64 wt.%), with the reaction temperature being 45 °C. From the images, it is clear that the sulphuric acid concentration has much influence on the dimensions of the resulting cellulose fibres. As the concentration of the acid is increased, under otherwise identical conditions, considerable particle size reduction, coupled with a loss of "fibrous character" identity occurs, indicating the "surface etching and erosion" nature of the hydrolysis process.

An attempt was made to ascertain the effects of the time of hydrolysis and initial dimension of banana fibres, on the products of the hydrolysis reactions. Thus, Fig. 4a–c shows the SEM images of cellulose fibres produced by the acid hydrolysis of banana fibres at a temperature of 65 °C for a period of 4 h, 6 h and 8 h, respectively. From the images it is clear that as the duration of hydrolysis

is increased the size of the resulting cellulose fibres is reduced and that the rods become more flattened or disordered. Fig. 4d–f is SEM images of cellulose fibres that were produced by the acid hydrolysis of banana fibres of different initial dimensions. Fig. 4d shows the SEM image of cellulose fibres created by the hydrolysis of banana fibres obtained after sieving in a sieve of 355 μm aperture size. Fig. 4e shows the SEM image of cellulose fibres created by the hydrolysis of banana fibres obtained after sieving in a sieve

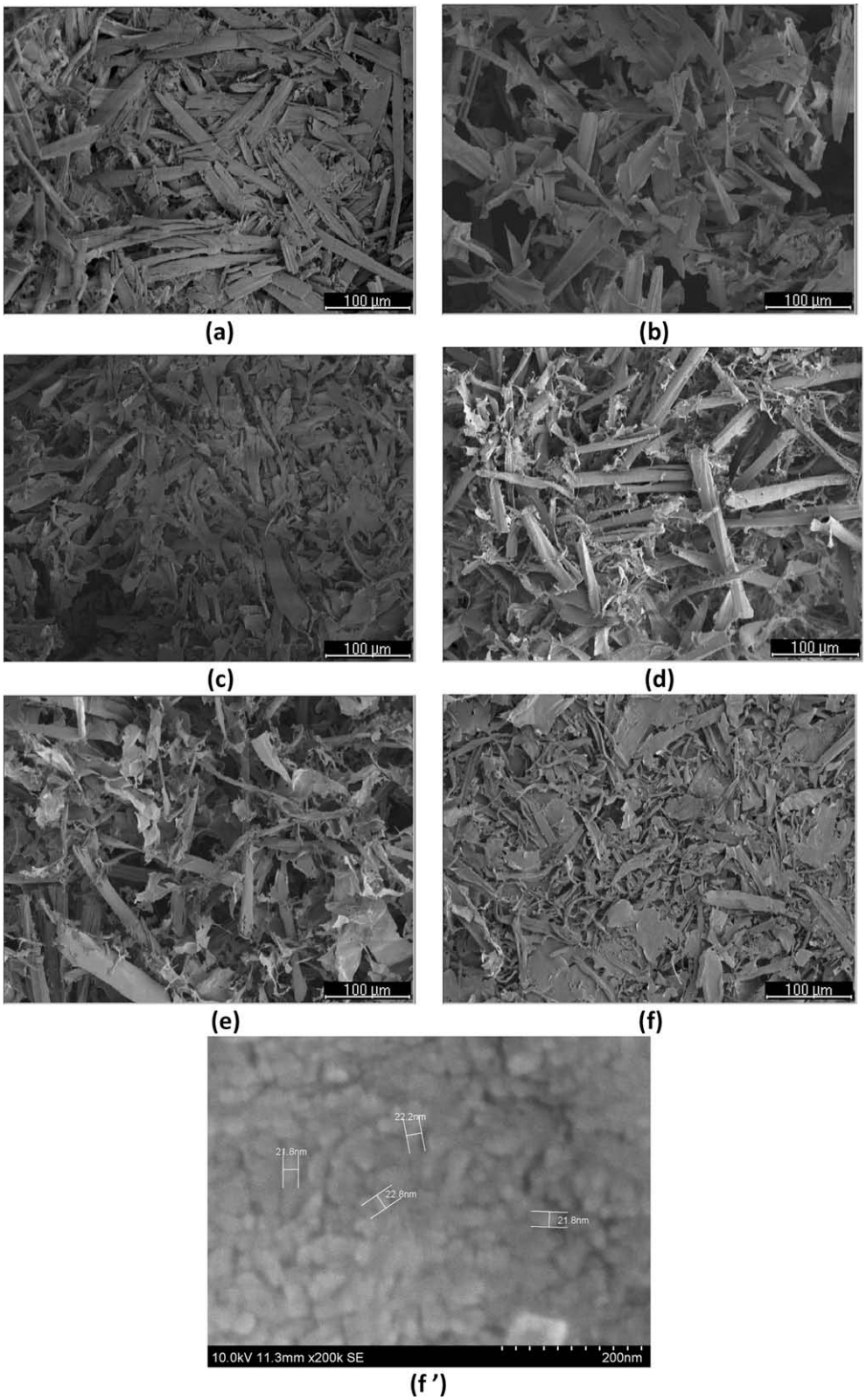

Fig. 4. (a) SEM image of freeze dried cellulose sample – 4C65. (b) SEM image of freeze dried cellulose sample – 6C65. (c) SEM image of freeze dried cellulose sample – 8C65. (d) SEM image of freeze dried cellulose sample – 355-4C45. (e) SEM image of freeze dried cellulose sample – 150-4C45. (f) SEM image of freeze dried cellulose sample – 38-4C45. (f') FESEM image of freeze dried cellulose sample – 38-4C45.

of 150 μm aperture size. Fig. 4f′ shows the SEM image of cellulose fibres created by the hydrolysis of banana fibres obtained after sieving in a sieve of 38 μm aperture size. Fig. 4f′ shows the FESEM image of the same sample (sample 38-4C45), which illustrates the presence of cellulose particles of 20–25 nm in size. From the figures it is clear that, the less the dimensions of the starting material (banana fibre), the easier it is to obtain cellulose microfibres with smaller dimensions.

3.5. XRD analysis

Studies were made of the influence of the hydrolysis process on the crystalline or ordered nature of the resulting cellulose fibrous species. From Fig. 5a, it can be seen that the cellulose that was subjected to the hydrolysis procedure is more crystalline than the initial banana fibres. This indicates that the banana fibres contain both crystallites and amorphous/disordered regions. These disordered regions were removed by the acid hydrolysis. Thus, during the hydrolysis process, the banana fibres were converted into more crystalline cellulose species. The increase in the overall order of the hydrolysed fibres can be attributed to the removal of the hemicelluloses and lignin during the chemical treatment. The diffractograms, probably representing typical cellulose I diffractograms, show a peak at $2\theta = 22^\circ$ and a shoulder in the region $2\theta = 14^\circ$–17° (Bondeson, Mathew, & Oksman, 2006).

Fig. 5b shows the XRD profiles of cellulose microfibres that were prepared under the different conditions, described earlier. From the figure, it is clear that all of the samples give similar diffraction patterns. The only difference concerns a slight intensity changes in the peaks, representing minor changes in the level of order in the samples.

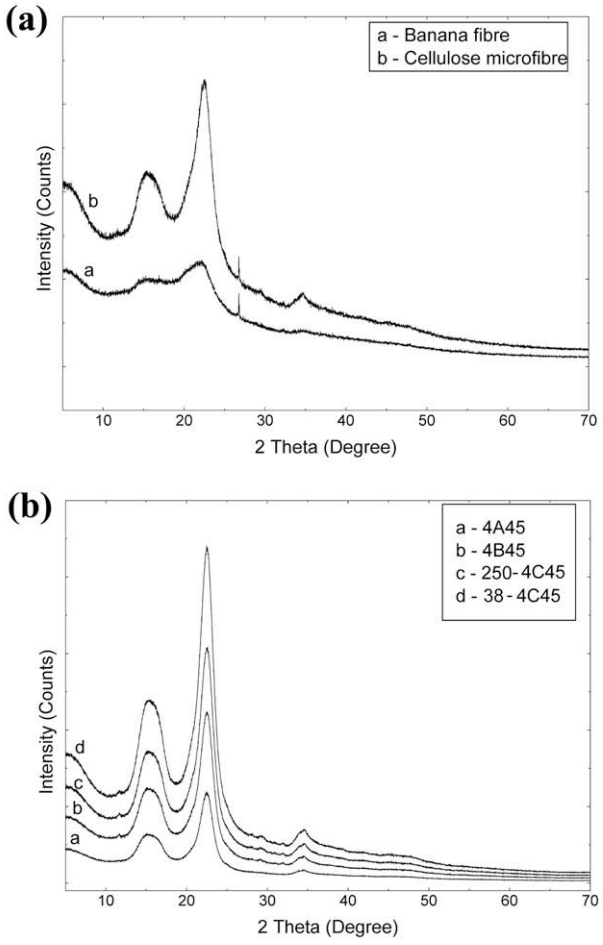

Fig. 5. (a) XRD curves of banana fibre and cellulose microfibres. (b) XRD curves of cellulose microfibres prepared under different conditions.

4. Conclusions

Cellulose microparticles that possess different structural and morphological characteristics can be prepared from the controlled hydrolysis of fibres derived from banana plant waste. The properties show a dependence on the conditions used in the hydrolysis process, such as the concentration of the sulphuric acid, the temperature of the hydrolysis system and the method used to dry the microfibrous species. The extent and the nature of the hydrolysis influence the dimensions and the surface charge of the resulting particles and the level of disorder that is created. Significant size reductions were obtained when banana fibres of initially smaller dimensions were used. The materials that have been developed show potential for inclusion in a variety of polymer composite systems, for use across a range of biomedical applications.

Acknowledgement

The authors are thankful to the Government of India and UK Government for awarding a project through UKIERI programme (UK-India Education Research Initiative).

References

Abdullah, A. B. M. (1991). Production of jute microcrystalline cellulose. *Journal of Bangladesh Academy of Science, 15*(2), 85–87.
Alemdar, A., & Sain, M. (2008). Biocomposites from wheat straw nanofibers: Morphology, thermal and mechanical properties. *Composites Science and Technology, 68*, 557–565.
Araki, J., Wada, M., & Kuga, S. (2001). Steric stabilization of a cellulose microcrystal suspension by poly(ethylene glycol) grafting. *Langmuir, 17*, 21–27.
Araki, J., Wada, M., Kuga, S., & Okano, T. (1999). Influence of surface charge on viscosity behavior of cellulose microcrystal suspension. *Journal of Wood Science, 45*, 258–261.
Bhattacharya, D., Grminario, L. T., & Winter, W. T. (2008). Isolation, preparation and characterisation of cellulose microfibres from bagasse. *Carbohydrate Polymers, 73*, 371–377.
Beck-Candanedo, S., Roman, M., & Gray, D. G. (2005). Effect of reaction conditions on the properties and behavior of wood cellulose nanocrystal suspensions. *Biomacromolecules, 6*, 1048–1054.
Bochek, A. M., Shevchuk, I. L., & Lavrentev, V. N. (2003). Fabrication of microcrystalline and powdered cellulose from short flax fiber and flax straw. *Russian Journal of Applied Chemistry, 76*(10), 1679–1682.
Bondeson, D., Mathew, A., & Oksman, K. (2006). Optimization of the isolation of nanocrystals from microcrystalline cellulose by acid hydrolysis. *Cellulose, 13*, 171–180.
Cao, X., Dong, H., & Li, C. M. (2007). New nanocomposite materials reinforced with flax cellulose nanocrystals in waterborne polyurethane. *Biomacromolecules, 8*, 899–904.
Chaubal, M. V., & Popescu, C. (2008). Conversion of nanosuspensions into dry powders by spray drying: A case study. *Pharmaceutical Research, 25*(10), 2302–2308.
de Rodriguez, N. L. G., Thielemans, W., & Dufresne, A. (2006). Sisal cellulose whiskers reinforced poly(vinyl acetate) nanocomposites. *Cellulose, 13*, 261–270.
de Souza Lima, M. M., & Borsali, R. (2004). Rodlike cellulose microcrystals: Structure, properties, and applications. *Macromolecular Rapid Communications, 25*, 771–787.
Dong, X. M., Revol, J. F., & Gray, D. G. (1998). Effect of microcrystallite preparation conditions on the formation of colloid crystals of cellulose. *Cellulose, 5*, 19–32.
El-Sakhawy, M., & Hassan, M. L. (2007). Physical and mechanical properties of microcrystalline cellulose prepared from agricultural residues. *Carbohydrate Polymers, 67*, 1–10.
Gacitua, E. W., Ballerini, A. A., & Zhang, J. (2005). Polymer nanocomposites: Synthetic and natural fillers – a review. *Maderas: Ciencia y Tecnología, 7*(3), 159–178.
Habibi, Y., Mahrouz, M., & Vignon, M. R. (2009). Microfibrillated cellulose from the peel of prickly pear fruits. *Food Chemistry, 115*, 423–429.
Hebeish, A., & Guthrie, J. T. (1981). *The chemistry and technology of cellulosic copolymers*. New York: Springer-Verlag.
Isogai, A., & Kato, Y. (1998). Preparation of polyuronic acid from cellulose by TEMPO-mediated oxidation. *Cellulose, 5*, 153–164.
Jain, J. K., Dixit, V. K., & Varma, K. C. (1983). Preparation of microcrystalline cellulose from cereal straw and its evaluation as a tablet excipient. *Indian Journal of Pharmaceutical Science, 45*, 83–85.

Li, R., Fei, J., Cai, Y., Li, Y., Feng, J., & Yao, J. (2009). Cellulose whiskers extracted from mulberry: A novel biomass production. *Carbohydrate Polymers, 76*, 94–99.

Mario, A. P., Marseno, D. W., & Haryadi. (2005). Synthesis and characterization of sodium carboxymethylcellulose from cavendish banana pseudo stem (*Musa cavendishii* LAMBERT). *Carbohydrate Polymers, 62*, 164–169.

Mohanty, A. K., Misra, M., & Hinrichsen, G. (2000). Biofibres, biodegradable polymers and biocomposites. *Macromolecular Material Engineering, 276*(277), 1–24.

Nelson, Y. U., Edgardo, A. G., & Ana, A. W. (2000). Microcrystalline cellulose from soybean husk: Effects of solvent treatments on its properties as acetyl salicylic acid carrier. *International Journal of Pharmaceutics, 206*, 85–96.

Samir, M. A. S. A., Alloin, F., & Dufresne, A. (2005). Review of recent research into cellulosic whiskers, their properties and their application in nanocomposite field. *Biomacromolecules, 6*, 612–626.

Viet, D., Beck-Candanedo, S., & Gray, D. G. (2007). Dispersion of cellulose nanocrystals in polar organic solvents. *Cellulose, 14*, 109–113.

Zhang, J., Elder, T. J., Pu, Y., & Ragauskas, A. J. (2007). Facile synthesis of spherical cellulose nanoparticles. *Carbohydrate Polymers, 69*, 607–611.

Zuluaga, R., Putaux, J. L., Restrepo, A., Mondragon, I., & Ganan, P. (2007). Cellulose microfibrils from banana farming residues: Isolation and characterization. *Cellulose, 14*, 585–592.