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Physical and mechanical properties of microcrystalline cellulose prepared from agricultural residues

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Abstract

Microcrystalline cellulose (MCC) was prepared from local agricultural residues, namely, bagasse, rice straw, and cotton stalks bleached pulps. Hydrolysis of bleached pulps was carried out using hydrochloric or sulfuric acid to study the effect of the acid used on the properties of the produced microcrystalline cellulose such as degree of polymerization (DP), crystallinity index (CrI), crystallite size, bulk density, particle size, and thermal stability. The mechanical properties of tablets made from microcrystalline cellulose of different agricultural residues were tested and compared to a commercial-grade MCC. The use of rice straw pulp in different proportions as a source of silica to prepare silicified microcrystalline cellulose (SMCC) was investigated. The effect of the percent of rice straw added on the mechanical properties of tablets before and after wet granulation was studied.

Keywords: Bagasse; Rice straw; Cotton stalks; Microcrystalline cellulose; Mechanical properties; Wet granulation; X-ray analysis; Thermogravimetric analysis

1. Introduction

Microcrystalline cellulose (MCC) has been widely used especially in food, cosmetic and medical industries as a water-retainer, a suspension stabilizer, a flow characteristics controller in the systems used for final products, and as a reinforcing agent for final products such as medical tablets. MCC is obtained at an industrial scale through hydrolysis of wood and cotton cellulose using dilute mineral acids. Since cellulose from different sources differs in properties (crystallinity, moisture content, surface area and porous structure, molecular weight, etc.) different properties of MCC obtained from different sources are expected. The conditions of hydrolysis also affect the properties of the obtained MCC. Preparation of MCC from materials other than wood and cotton such as water hyacinth (Gaonkar & Kulkarni, 1987), coconut shells (Gaonkar & Kulkarni, 1989), sugar cane bagasse (Padmadisastra & Gonda, 1989; Shah, Shah, & Trivedi, 1993; Tang et al., 1996), (Castro & Bueno, 1996), (Paralikar & Bhatawdekar, 1988), ramie (Castro & Bueno, 1996), wheat and rice straws (Jain, Dixit, & Varma, 1983; Chen, Yan, & Ruan, 1996), jute (Abdullah, 1991), flax fibers and flax straw (Bochek, Shevchuk, & Lavrentev, 2003), and soybean husk (Nelson, Edgardo, & Ana, 2000) has been studied. However, for the best of our knowledge, the mechanical properties of MCC tables made from agricultural wastes and the effect of kind of acid used on these properties has not been studied in details.

MCC has relatively low chemical reactivity combined with excellent compactibility at low pressures. MCC was rated the most useful filler for direct compression tableting (Shangraw & Demarest, 1993). However, a number of limitations to the use of MCC have been reported (Bolhuis & Chowhan, 1996), the most important of which were considered to be its low bulk density,

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high lubricant sensitivity, poor flow characteristics and the influence of moisture on the compression characteristics. A number of new grades of MCC have been introduced to reduce some of these problems. Most notable amongst these are high density and large particle size grades of the materials. Although these grades may have some advantages in terms of greater plasticity (Muñoz-Ruiz, Antequera, Parales, & Ballesteros, 1994) they form weaker compacts than the base material, which may reflect a reduced surface area for bonding during compression (Khan & Pilpel, 1986; Zeleznik, Virtanen, & Sherwood, 1996). In addition, the small surface area of large particle size grades makes them more susceptible to the effects of lubricants and they can form poor ordered blends with low particle size drugs (Staniforth & Tralhao, 1996).

It has been suggested that co-processing of MCC with other excipients may improve the performance of materials in direct compression. Included amongst these additives are starch, calcium sulfate (Bavitz & Schwartz, 1974), calcium carbonate (Mehra, West, & Wiggins, 1987), dibasic calcium phosphate, β-cyclodextrin and lactose (Armstrong, Roscheisen, & Alaghbar, 1996; Belda & Mielck, 1996). The beneficial properties of surface modification of MCC with silicon dioxide or silicic acid have been reported (Nürnberg & Wunderlich, 1995; Nürnberg & Wunderlich, 1996a; Nürnberg & Wunderlich, 1996b). The process of silicification leads to the deposition of silicon, presumably in the form of silicon dioxide, both on the outer envelope surface of the particle and on exposed surfaces within the particle (Staniforth & Tobyn, 1996). In addition, silicified MCC (SMCC) has been shown to possess a number of pharmaceutical advantages in terms of powder flow (Khalaf, Tobyn, & Staniforth, 1997), tablet strength (Sherwood, Hunter, & Staniforth, 1996), lubricant sensitivity and wet granulation (Staniforth & Chatrath, 1996). Preliminary data also suggest that the material performs well in direct compression formulations (Riba, Segado, & Ferrer, 1997) and roller compaction (Sheskey, Davidson, & Figtner, 1997). It was evident that when microcrystalline cellulose is silicified in the preparation of SMCC, no bulk chemical change in the MCC is observed at the resolutions tested and no observable polymorphic changes are induced (Tobyn, McCarthy, Staniforth, & Edge, 1998). It was also showed that tablets made of wet-granulated MCC had much lower tensile strength than tablets made from MCC before granulation while wet-granulated SMCC had the same tensile strength as the original MCC (Sherwood & Becker, 1998).

The aim of this work is to study physical properties of MCC prepared from different agricultural residues (bagasse, rice straw, and cotton stalks) and the mechanical properties of tablets prepared from the different MCC samples. Also, using of rice straw pulp as a source of silica to prepare silicified MCC was investigated through co-processing bagasse and rice straw pulps in different ratios.

2. Experimental

2.1. Raw materials

Bleached kraft bagasse and bleached soda rice straw pulps were kindly supplied by Qena Company for Pulp and Paper, Qena, and Rakta Company for Paper Manufacture, Alexandria, Egypt, respectively. Bleached cotton stalks pulp was prepared in the laboratory by pulping of cotton stalks using NaOH (15% based on dry weight of cotton stalks) at 150 °C for 2 h and bleaching the produced pulp using the sodium chlorite bleaching method (Browning, 1967). The chemical compositions of the bleached pulps were determined using the known standard methods (Browning, 1967) and were as follows in Table 1.

2.2. MCC preparation

Bleached bagasse, cotton stalks, and rice straw pulps were hydrolyzed with 2 N hydrochloric acid or 2 N sulfuric acid under reflux for 45 min (Paralikar et al., 1988); the liquor ratio was 1:10. The hydrolyzed pulps were thoroughly washed with distilled water and freeze-dried. Degree of polymerization (DP) of the different samples was determined by viscosity measurement of the samples dissolved in copper-ammonium hydroxide solution (Browning, 1967). For preparation of silicified MCC, different ratios of bleached rice straw pulp were added to the bleached bagasse pulp so that the final silica concentrations were 2%, 4%, 6%, and 8% based on the final weight of the pulp. The mixed pulps were hydrolyzed by 2 N sulfuric acid under reflux for 45 min then washed and freeze-dried.

2.3. X-ray powder diffraction studies

Diffraction patterns were obtained using a Phillips X-ray diffractometer. The diffraction patterns were recorded using Cu-Kα radiation at 40 kV and 25 mA. The samples were pressed into pellets (25 mm in diameter) by compression of 0.25 g in a mold under a pressure of 50 MPa.

The crystallite size of MCC was measured using the half-height width of the I_{002} reflection and crystallinity index (CrI) was calculated as follows (Sidiras, Koullas, Vgenopoulos, & Koukios, 1990):

$$\mathrm{CrI} = [(I_{002} - I_{\mathrm{am}})]/I_{002}$$

where I_{002} is the intensity of the 002 peak (at about 2 $\theta = 26$) and I_{am} is the intensity corresponds to the peak at about 2 $\theta = 18$.

Table 1
Chemical composition of the bleached pulps used

	α-Cellulose (%)	Lignin (%)	Hemicelluloses (%)	Ash (%)	Silica in ash (%)
Bagasse	77.6	0.87	21.4	1.3	–
Cotton stalks	75.1	0.94	19.3	1.3	–
Rice straw	71.2	1.32	17.4	13.8	71.3

2.4. Scanning electron microscopy

Scanning electron microscopy (gold coating, Edwards Sputter Coater, UK) was performed using a Jeol 6310 (Jeol Instruments, Tokyo, Japan) system running at 5–10 keV.

2.5. Particle size and particle size distribution

Particle size data were obtained using a dynamic laser light scattering particle size analyzer (type Horiba, LB-500). Three separate samples were used to determine a mean particle diameter for each material.

2.6. Bulk and tapping densities

An appropriate amount of the sample was poured in a 50 ml tarred graduate cylinder. The cylinder was lightly tapped twice to collect all the powder sticking on the wall of the cylinder. The volume was then read directly from the cylinder and used to calculate the bulk density according to the relationship: mass/volume. For tapping density, the cylinder was tapped until no change in volume. The volume of the sample was then read and used in the calculation.

2.7. Thermogravimetric analysis (TGA)

A Perkin-Elmer Thermogravimetric analyzer was used to study the thermal stability of the different MCC samples. The heating rate was set at 10 °C/min over a temperature range of 50–700 °C. Measurements were carried out in nitrogen atmosphere, with a rate of flow of 50 cm^3/min.

2.8. Preparation of tablets

Tablets were prepared by compacting powders (2 g) in a mold (25 mm diameter), using a load of 100 kN and a dwell time of 1 min using a Craver press. Tablets were tested on the same day of preparation, typically 2 h after compaction. This allows compaction and testing to be performed under comparable ambient conditions (temperature and relative humidity).

2.9. Tensile strength testing of tablets

Diametric tensile testing was performed at 5 mm/min using a LLOYD LR 10 k universal testing machine Tensile strength was calculated using the failure load over the diametric area of the compacts (Fell & Newton, 1972). The energy of failure was calculated by integration the area under the load/deflection curve of the tensile test.

2.10. Hardness test of tablets

The hardness of tablets was measured using a Wilbert Hardness tester HT 2004 according to the DIN 53 456 standard.

2.11. Wet granulation of MCC

Wet granulation of the different MCC samples was carried out by moistening the sample with water (~100% of mass) and passing the wet mass through 12-mesh screen. The granules were dried in air till constant weight.

3. Results and discussion

3.1. Hydrolysis of bagasse, rice straw, and cotton stalks pulps

Hydrolysis of the different kinds of pulps to prepare MCC was carried out using sulfuric or hydrochloric acids. Preliminary experiments showed that these pulps reach constant weight loss and level-off degree of polymerization (LODP) after their reflux with acids for 30–45 min. Degradation of cellulose by acids to reach LODP is known to occur through the degradation of the gylcosidic bonds of cellulose chains. Table 2 shows the LODP of the MCC prepared from the different pulps using HCl or H_2SO_4. The results are compared to a commercial Avicell MCC.

The LODP obtained for bagasse was higher than that of cotton stalks and rice straw MCC and was comparable to that of the commercial Avicell MCC. Rice straw and cotton stalks MCC had similar LODP values at each kind of the acid used. The LODP values were higher for MCC samples prepared using HCl than in the case of using H_2SO_4. Theoretically, no difference in DP is expected for using both acids. But, hydrolysis via H_2SO_4 is known to cause esterification of cellulose and introduction of sulfate groups (Revol, Bradford, Giasson, Marchessault, & Gray, 1992). The sulfate groups on MCC will be ionized in solution and repulsion between chains may cause easier flow than MCC prepared using HCl, i.e., shorter flow time and calculated DP in case of MCC prepared using H_2SO_4.

3.2. Crystallinity and crystallite size

The X-ray diffraction pattern of MCC samples prepared from bagasse, rice straw, and cotton stalks along with that of the commercial Avicell sample is shown in Fig. 1. The calculated crystallinity index and crystallite size of the different MCC samples are given in Table 3.

As shown in Fig. 1 all samples have a typical crystal lattice for cellulose I (Nelson & O'Connor, 1964). Also, all MCC samples had similar CrI values with slightly lower values for bagasse MCC regardless the kind of acid used.

Table 2
LODP of the bagasse, rice straw, and cotton stalks MCC using HCl or H_2SO_4 acids

	Avicell PH 101	Bagasse		Rice straw		Cotton stalks	
Acid used	None	HCl	H_2SO_4	HCl	H_2SO_4	HCl	H_2SO_4
LODP	317	317	299	237	224	245	232

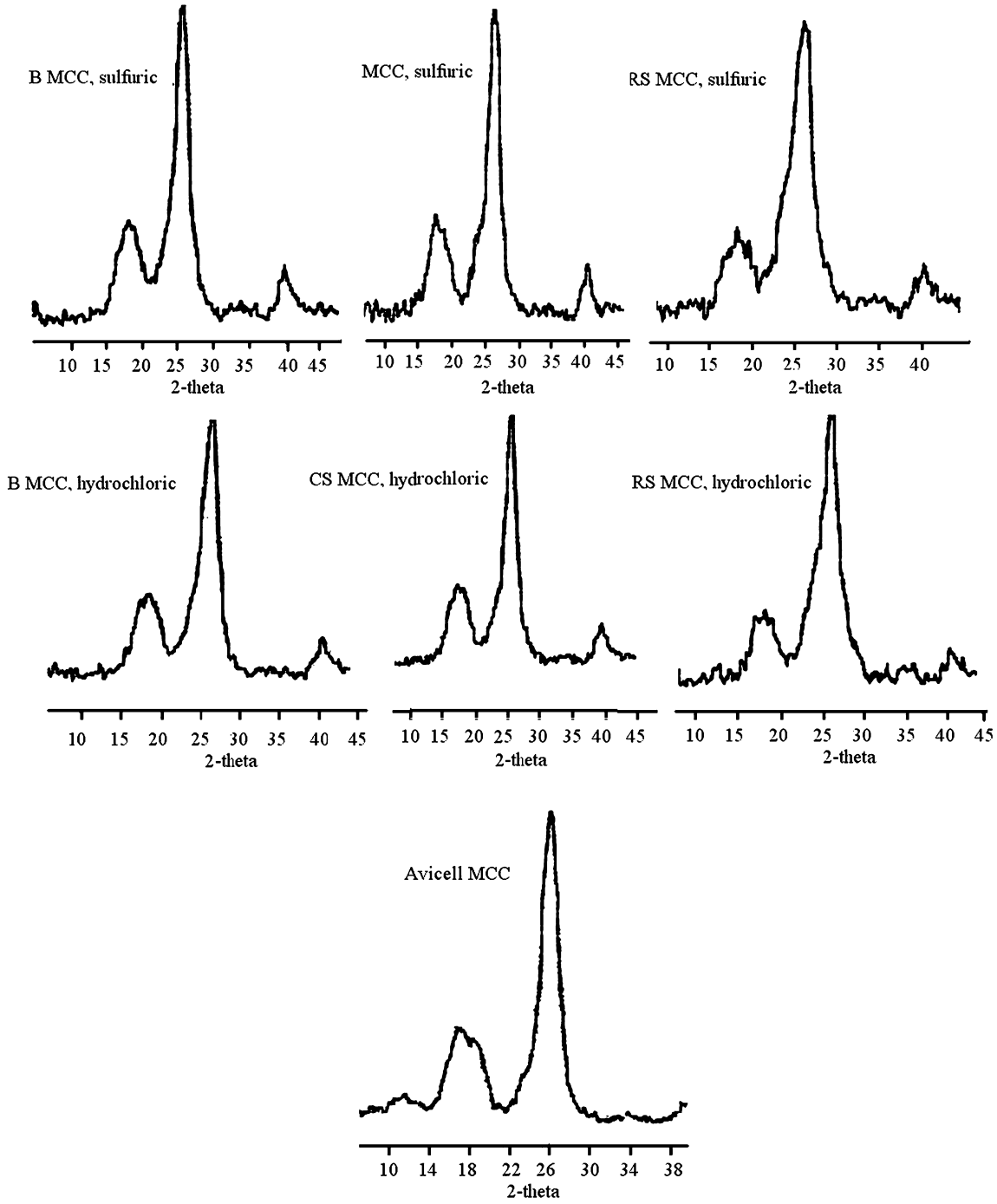

Fig. 1. X-ray diffraction patterns of Avicel, cotton stalks (CS), bagasse (B), and rice straw (RS) MCC samples prepared using HCl or H_2SO_4 acids.

However, rice straw had the smallest crystallite size followed by bagasse and cotton stalks. Generally, the kind of the acid used had no effect on crystallinity and crystallite size of the prepared MCC.

3.3. Scanning electron microscopy (SEM)

Fig. 2 shows SEM graphs of the different MCC samples. As seen from the figure showing hydrolysis of bleached pulps (bagasse is given as an example and the other pulps were similar), shortening of fibers occurred and rod-shaped MCC formed. There is no difference between the different MCC samples except for the large amount of pith (a non-fibrous material) seen in case of cotton stalks MCC. The source of this debris is the fine pith core of cotton stalks. No significant differences were observed between samples prepared using different kind of acids was found. Some strands of cellulosic microfibers were observed in the different MCC samples prepared.

Table 3
Crystallinity index (CrI) and crystallite size (D_{002}) of MCC samples

	Acid used	CrI	D_{002} (nm)
Avicell PH 101	None	0.78	5.52
Bagasse	HCl	0.76	4.42
Bagasse	H_2SO_4	0.75	4.42
Rice straw	HCl	0.78	3.97
Rice straw	H_2SO_4	0.77	3.32
Cotton stalks	HCl	0.77	5.31
Cotton stalks	H_2SO_4	0.77	4.65

3.4. Bulk and tapping densities

Density of MCC reflects its porosity, which is an important factor in the application of MCC. Table 4 shows the bulk and tapping densities of the different MCC samples. As shown in the table cotton stalks MCC had significantly higher tapping and bulk densities than the other samples. This may be due to the presence of the large amounts of pith – non-fibrous materials – as shown in the SEM micrographs. No significant effect of the kind of acid used on the density of the different MCC samples was observed. The density of rice straw and bagasse MCC is generally comparable to the commercial Avicell MCC sample.

3.5. Particle size and particle size distribution

The results of particle size analysis of the different MCC samples are summarized in Table 5. As shown in the table the mean particle size of all prepared samples are compara-

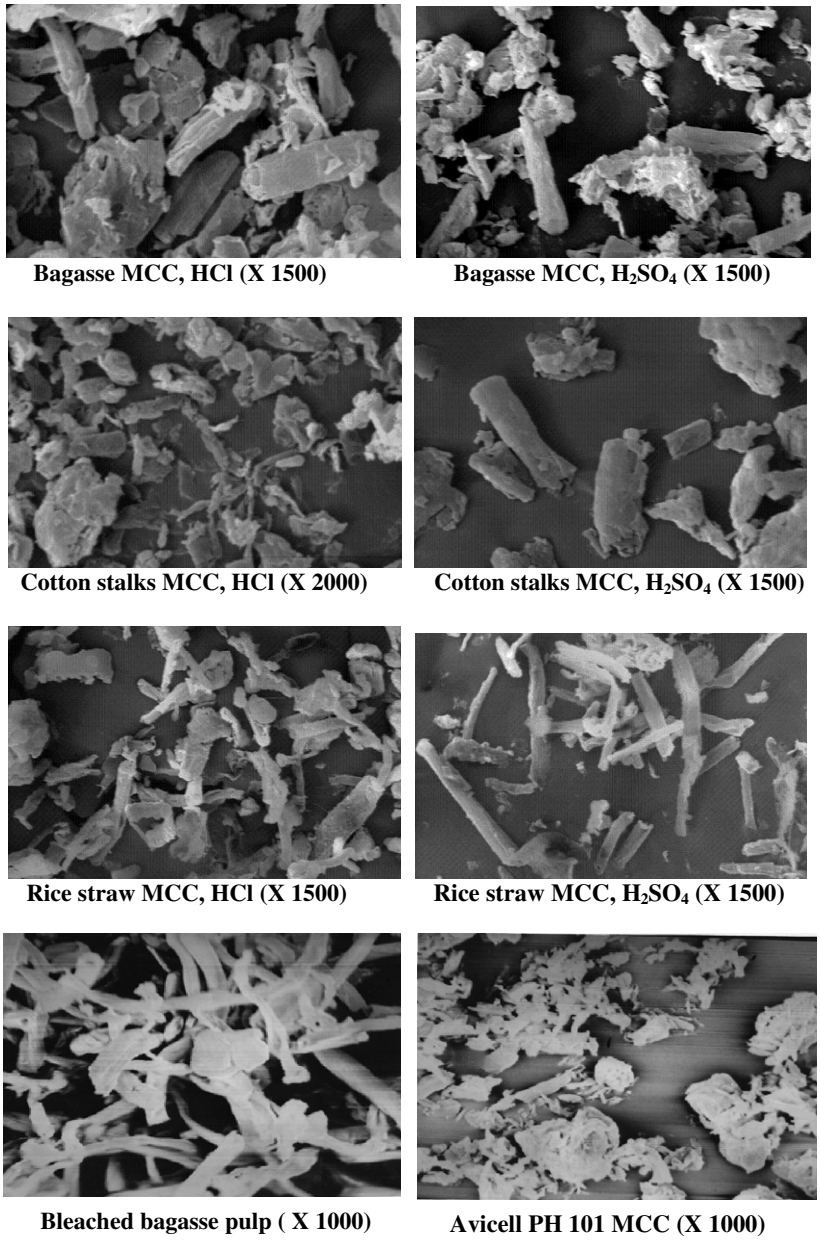

Fig. 2. SEM graphs of bleached bagasse pulp and the different MCC samples.

Table 4
Bulk and tapping densities of bagasse, rice straw, cotton stalks, and Avicell MCC samples

	Acid used	Bulk density (g/cm^3)	Tapping density (g/cm^3)
Avicell PH 101	None	0.37	0.51
Bagasse	HCl	0.32	0.49
Bagasse	H_2SO_4	0.36	0.54
Rice straw	HCl	0.36	0.54
Rice straw	H_2SO_4	0.34	0.52
Cotton stalks	HCl	0.59	0.87
Cotton stalks	H_2SO_4	0.54	0.78

Table 5
Particle size and particle size analysis of bagasse, rice straw, cotton stalks, and Avicell MCC samples

	Acid used	Mean particle size (μm)	Particle size range (μm)
Avicell PH 101	None	5.53	5–6
Bagasse	HCl	4.38	3–6
Bagasse	H_2SO_4	4.34	3–6
Rice straw	HCl	3.66	3–5.2
Rice straw	H_2SO_4	4.06	3–6
Cotton stalks	HCl	4.07	3–6
Cotton stalks	H_2SO_4	5.49	4.5–6

ble except for rice straw MCC prepared using HCl, which had smaller particle size, and consequently higher specific surface area, than the others. All prepared samples had slightly lower particle size, than Avicell MCC but with wider particle size distribution. The kind of acid used did not affect the particle size in case of MCC prepared from bagasse. In case of cotton stalks and rice straw MCC samples with larger particle size was obtained in case of using H_2SO_4 in the hydrolysis/of the prepared MCC samples.

3.6. Thermogravimetric analysis

Thermal stability of the different microcrystalline cellulose samples was studied using Thermogravimetric analysis (TGA). The samples selected were: bagasse MCC prepared using H_2SO_4, bagasse MCC prepared using HCl, rice straw MCC prepared using H_2SO_4, and cotton stalks MCC prepared using H_2SO_4. Fig. 3 shows the TG curves of these samples and Table 6 gives the data obtained from these curves. As shown in the figures, the degradation of the different MCC samples involves two main degradation stages. The onset degradation of cellulose is believed to be due to the evolution of non-combustible gases such as carbon dioxide, carbon monoxide, formic acid, and acetic acid while the second degradation stage is believed to be due to pyrolysis and evolution of combustible gases (LeVan, 1989). As shown in Fig. 3, there is no significant difference between the TG curve of bagasse MCC prepared using H_2SO_4 and that of bagasse MCC prepared using HCl. Both samples had nearly the same onset degradation temperatures for the two stages and also the same maximum weight-loss temperatures of the two stages (the maximum weight-loss temperatures are obtained from the first-derivatives of TG curves). However, the rate of thermal degradation of the first stage, in case of bagasse MCC prepared using H_2SO_4, was higher than that in case of bagasse MCC prepared using HCl. Using H_2SO_4 in the preparation of MCC results in formation of sulfate groups onto cellulose chains (Revol et al., 1992); splitting of the sulfate groups during MCC thermal degradation may be the reason for the higher rate of degradation via weight loss and the possible attack of detached sulfate groups to the cellulose chains. Cotton stalks MCC showed slightly higher onset degradation temperature and maximum weight-loss temperature for both degradation stages than bagasse and rice straw MCC samples. Also, complete degradation of cotton stalks (ash formation temperature) occurred at higher temperature than bagasse and rice straw MCC samples. The TG curve of rice straw MCC shows its high ash content, which is rich in silica.

3.7. Mechanical properties of MCC tablets

Microcrystalline cellulose is a widely used tableting excipient. In terms of tableting technology, the material is described as a filler/binder in that it is usually added to formulations to enhance compactibility. Although the preparation of MCC from agricultural residues and their physical properties have been reported as mentioned above the mechanical properties of tablets pressed from MCC of these agricultural residues have not been studied.

The different MCC samples were pressed into tablets and their mechanical properties (tensile strength, energy of failure, and hardness) were measured. The results are given in Table 7. One important aspect of pharmaceutical mechanical testing is to prepare and test samples using the same protocols. Additionally, it is essential that compacts of comparable density are prepared since porosity has a marked effect on strength.

As shown in the table, tablets made from the prepared MCC samples had similar densities except for cotton stalks, which produced tablets of higher density. As seen in the SEM graphs a lot of pith was found in case of cotton stalks MCC. The pith is non-fibrous and its existence leads to higher density and, at the same time, weaker mechanical properties. Usually, higher density MCC tablets have higher mechanical properties, but in case of cotton stalks the presence of pith decreases the fiber–fiber bonding and thereby the mechanical properties.

Tablets made from rice straw MCC had higher tensile strength and energy of failure than that made from bagasse MCC in spite of the high silica content of the former. Silica presents in rice straw fibers are impeded in the fiber lumen and do not affect the fiber–fiber bonding. Tensile strength is often used to describe the strength of a compact. However, this measurement does not fully reflect inter- and intraparticle cohesion within a compact. The cohesion (integrity or binding capability) in a compact may be further represented by the energy

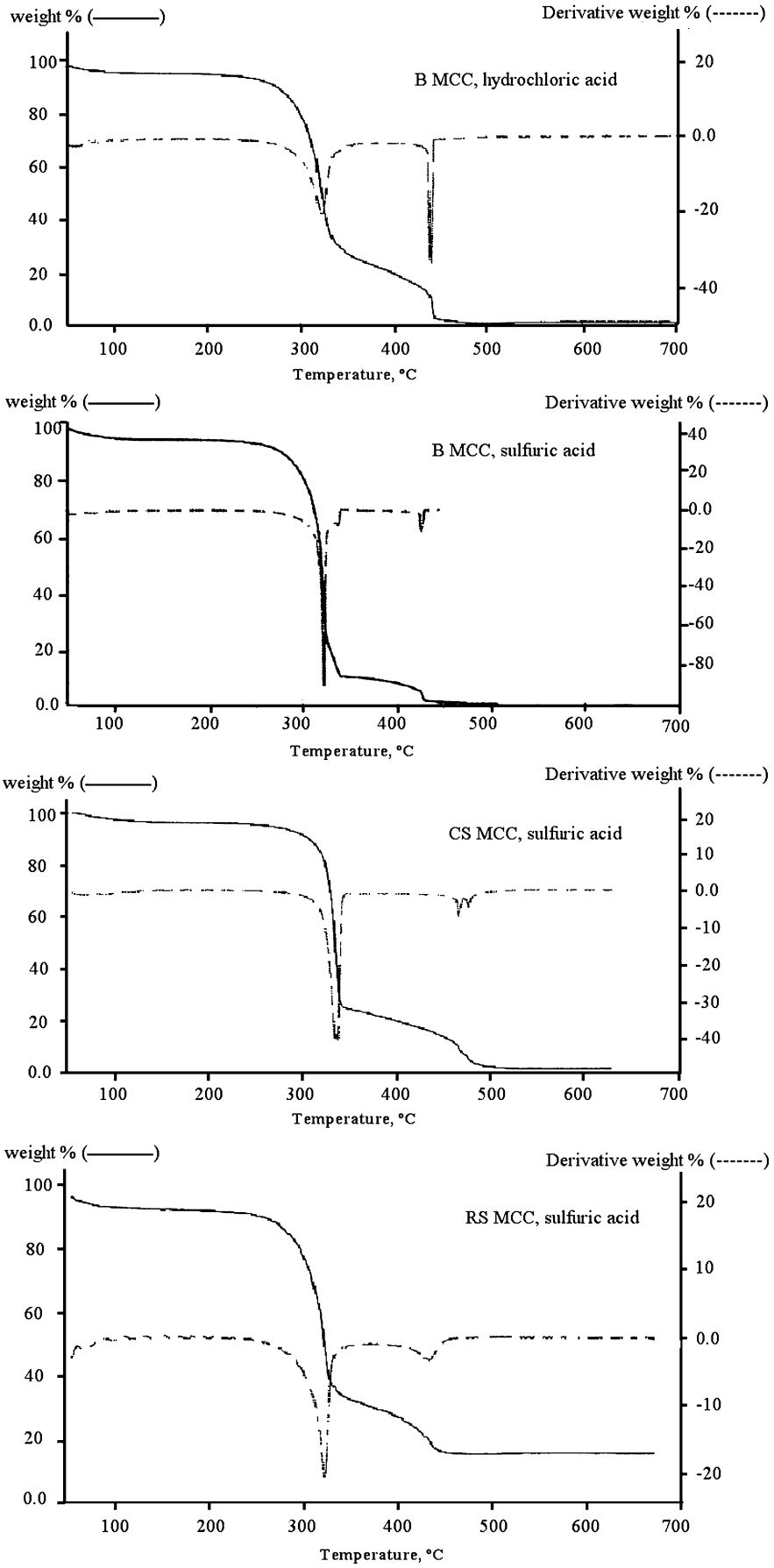

Fig. 3. TGA curves and differential TG curves of cotton stalks (CS), bagasse (B), and rice straw (RS) MCC samples prepared using HCl or H_2SO_4 acids.

Table 6
TGA data obtained for bagasse, rice straw, and cotton stalks MCC samples

	Acid used	First stage onset degradation temperature (°C)	First stage maximum weight-loss temperature[a] (°C)	Second stage onset degradation temperature[a](°C)	Second stage maximum weight-loss temperature (°C)	Ash formation temperature (°C)
Bagasse	HCl	251	325	–	439	475
Bagasse	H_2SO_4	252	322	335	426	460
Rice straw	H_2SO_4	250	323	335	434	450
Cotton stalks	H_2SO_4	270	335	340	465	500

[a] Obtained from the first-derivatives TG curves (DTG curves).

Table 7
Mechanical properties of tablets made from the different MCC samples

	Acid used	Tensile strength (MPa)	Energy of failure (N mm^2)	Hardness (MPa)	Density (g/cm^3)
Avicell PH 101	None	6.5	870	81.1	1.28
Bagasse	HCl	3.82	769	71.7	1.20
Bagasse	H_2SO_4	4.32	873	75.6	1.21
Rice straw	HCl	4.98	876	73.6	1.22
Rice straw	H_2SO_4	5.18	905	74.9	1.22
Cotton stalks	HCl	2.22	244	26.2	1.27
Cotton stalks	H_2SO_4	3.46	321	39.8	1.26

of failure (Edge, Steele, Chen, Tobyn, & Staniforth, 2000). The hardness of bagasse and rice straw MCC tablets were close to each other. Regarding the effect of kind of acid used, tablets made from MCC prepared using H_2SO_4 had generally higher mechanical properties than tablets made from MCC prepared using HCl. This may be attributed to the presence of sulfate groups on MCC particles prepared using H_2SO_4 (Revol et al., 1992). The presence of sulfate groups may increase the polar–polar interaction between the MCC particles and consequently increases the mechanical properties of tablets made from these MCC samples. Tablets prepared from the commercial Avicell MCC were of higher density than those obtained using the different MCC samples. Due to its higher density, the tensile strength of Avicell MCC tablets was remarkably higher than the other tablets prepared from the different MCC samples. The energy of failure, which reflect the extent of the cohesion within the tablets, was higher in case of rice straw and bagasse MCC samples than that in case of tablets made from the Avicell MCC when the density of the tablets is taken into consideration.

3.8. Effect of wet granulation on the properties of MCC tablets

Microcrystalline cellulose is used as a compression aid in directly compressed tablet formulations and as diluents in wet-granulated products. It has been reported that wet granulation of microcrystalline cellulose deteriorate the compression properties (Sherwood et al., 1996), i.e., it does not retain compaction properties after wetting and drying. It has been also reported that tablets made of wet-granulated MCC had much lower tensile strength than tablets made from MCC before granulation (Sherwood & Becker, 1998). This phenomenon is due to hornification as a result of wetting and drying of cellulose.

Tablets were made from bagasse and rice straw MCC samples after wet granulation and their mechanical properties were tested; the results are given in Table 8. As shown in the table, wet granulation of MCC samples produced tablets with higher density than those made from MCC before wet granulation. The tensile strength of the tablets did not significantly affected by the wet granulation but the energy of failure remarkably decreased. A slight decrease in the hardness occurred as a result of the wet granulation. The decrease in the energy of failure was the lowest in case of tablets made from rice straw MCC. The only significant difference between bagasse and rice straw MCC is the presence of high percent of silica in rice straw MCC, i.e., rice straw MCC is naturally in situ silicified. The presence of silica within the MCC particles reduced the negative effect of the wet granulation on cohesiveness of tablets, i.e., on the energy of failure. Tablets made from wet-granulated Avicell MCC showed also significant decrease in their mechanical properties as a result of wet granulation. No trend was found for the effect of the kind

Table 8
Mechanical properties of tablets made from wet-granulated MCC samples[a]

	Acid used	Tensile strength (MPa)	Energy of failure (N mm^2)	Hardness (MPa)	Density (g/cm^3)
Avicell PH 101	None	4.78 (6.5)	351 (870)	74.8 (81.1)	1.43 (1.28)
Bagasse	HCl	4.78 (3.82)	396 (769)	73.2 (71.7)	1.36 (1.20)
Bagasse	H_2SO_4	4.23 (4.32)	364 (873)	71.9 (75.6)	1.32 (1.21)
Rice straw	HCl	5.07 (4.98)	695 (876)	68.8 (73.6)	1.28 (1.22)
Rice straw	H_2SO_4	4.86 (5.18)	629 (905)	71.4 (74.9)	1.35 (1.22)

[a] Values between brackets are the values before wet granulation.

Table 9
Mechanical properties of tablets made from SMCC samples before and after wet granulation (WG)

Kind of MCC and % silica in prepared SMCC	Tensile strength (MPa)		Energy of failure (N mm^2)		Hardness (MPa)		Density (g/cm^3)	
	Before WG	After WG	Before WG	After WG	Before WG	After WG	Before WG	After WG
Bagasse MCC[a]	4.32	4.23	873	364	75.6	71.9	1.2	1.32
SMCC bagasse–rice straw, ~2%	4.25	4.93	827	381	75.5	75.0	1.22	1.32
SMCC bagasse–rice straw, ~4%	4.39	4.62	889	399	73.6	74.6	1.23	1.33
SMCC bagasse–rice straw, ~6%	4.92	5.12	843	484	74.1	70.0	1.21	1.34
SMCC bagasse–rice straw, ~8%	4.89	5.08	822	467	75.4	69.4	1.2	1.34
Rice straw MCC[a]	5.18	4.86	905	629	74.9	71.9	1.22	1.35

[a] Prepared by hydrolysis using H_2SO_4.

of acid used on the properties of the wet-granulated MCC samples.

3.9. Using rice straw pulp to prepare in situ silicified MCC

Rice straw pulp is characterized by high silica content (~14%). The high silica content may be an obstacle in using MCC prepared from rice straw in pharmaceuticals. On the other hand, as mentioned before, silicification of MCC with silicon dioxide or silicic acid leads to deposition of silica on the surface and within MCC particle. The silica added is usually about 2%. The silicified MCC (SMCC) has some advantages over MCC such as better tablet strength (Sherwood et al., 1996), better performance in direct compaction (Riba et al., 1997), and retaining of tensile strength of tablets after wet granulation (Sherwood & Becker, 1998). As shown from the above results, rice straw MCC tablets have better tensile strength than bagasse and cotton stalks MCC. Also, wet granulation of rice straw MCC resulted in a decrease in the mechanical properties of the tablets but lower than in case of bagasse MCC. Processing of mixtures of rice straw and bagasse pulps using H_2SO_4 was carried out to prepare MCC with different silica contents, i.e., naturally in situ SMCC. The properties of tablets made from different SMCC sample before and after wet granulation are given in Table 9. As shown in the table, the mechanical properties of the different SMCC tablets prepared from co-processed bagasse and rice straw at different ratios are generally comparable to that the MCC tablets made from bagasse or rice straw. Increasing the percent of rice straw pulp (increasing the silica content) resulted in a slight increase in tensile strength and a slight decrease in the energy of failure compared to that of bagasse or rice straw MCC samples. The effect of the wet granulation of the different SMCC samples on the mechanical properties of tablets was studied. As shown in Table 9, the density of the tablets was increased as a result of wet granulation of SMCC samples. Also, the energy of failure was remarkably deteriorated and the addition of rice straw pulp to bagasse pulp resulted in a decrease of the negative effect of the wet granulation on cohesiveness of tablets.

4. Conclusions

- Rice straw, bagasse, and cotton stalks could be used for the preparation of MCC using either HCl or H_2SO_4. However, the kind of acid used was found to affect particle size, thermal stability, tensile strength, and cohesiveness of the tablets made from the different MCC samples.
- Although of its high silica content, rice straw MCC tablets showed better tensile strength and cohesiveness than those made from bagasse and cotton stalks MCC. Rice straw MCC showed also the highest resistant toward the negative effect of wet granulation on cohesiveness of tablets.
- Co-processing of bagasse pulp with rice straw pulp to prepare SMCC having different silica contents produced SMCC that had better resistance to the negative effect of wet granulation but did not significantly affect the tensile strength of tablets.

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