ABSTRACT

The properties of eco-friendly adhesives prepared from poly(vinyl alcohol) (PVOH) can be improved by the use of cellulose microfibre (CMF). The CMF was prepared from oven dried sugarcane bagasse fibres. They were hydrolyzed with nitric acid of 10% (w/w). The cellulose content of the CMF increased from 44.10% to 88.99% after treatment. The CMF was added into PVOH adhesives in varied amounts of 0.5, 1.5, 2.5 and 3.5 wt% of total dry adhesive weight. The effects of CMF on chemical structure, thermal properties and crystallinity of the composite adhesives were investigated by Fourier-transform infrared spectroscopy and differential scanning calorimetry. The results showed the interactions of OH groups of PVOH with CMF molecular chains which led to increased melting point and crystallinity. In addition, the molecular weight and concentration of PVOH influenced the adhesion property of the composite adhesives. The results indicated that with the addition of CMF filler the shear strength of samples of PVOH (BF-17) of medium molecular weight increased from 1.55 to 2.41 MPa, while those of high molecular weight (BF-26) increased from 1.79 to 2.15 MPa. Furthermore, the incorporation of the CMF into the PVOH adhesives led to an improved drying time during adhesion of the test specimens.

INTRODUCTION

The technology of wood adhesives is an advanced field of science which integrates the technology of adhesive formulation and preparation with a multitude of sophisticated applications for different purposes.

Generally, a good bonding depends mainly on the use of an efficient adhesive. The general requirements for wood adhesives include their composition, solid content, viscosity, purity, ease of application, compatibility with additives, and cheap raw materials.

The mechanical properties of adhesives and the emission of monomers or volatile organic compounds (VOC) or formaldehyde during application are also considered important during adhesives production [1]. The adhesion bonding of thermosets such as phenol-formaldehyde (PF) and urea-formaldehyde (UF) and their derivatives may be highly durable [2]. However, there are health risks on workers posed by...
the emission of volatiles into the environment [3].

Today, there is growing awareness about the environment protection and new standards have been legislated and applied. There is a great interest in developing sustainable and environmentally friendly systems through enhanced performances, by including suitable bio-based adhesives. This aspect has been investigated for a long time that includes the search for adhesives based on cellulose, starch and poly(vinyl alcohol) (PVOH) [1,4].

PVOH is an environmental friendly and water soluble synthetic polymer. The properties of PVOH depend on its molecular weight and degree of its hydrolysis [5]. It is also used as wood adhesive. However, the bonding properties of bio-based adhesives are not of substantial strength for many types of constructions using wood substrate [3]. There is a need to modify their specifications to compete with petroleum-based adhesives in terms of performance and cost. One way to improve the properties of bio-based adhesives and altogether enhance their commercial potential is to heighten reinforcement properties of the adhesive [6].

The use of agricultural residue in bio-composites has prospects as a commercial application. There are some useful studies in the literature on agricultural fibres in bio-composites [6-10]. However, no report has been found about the biocomposite adhesive which would use cellulose microfibre as reinforcement. Cellulose is the fibrillar component of plant cells. It is the most abundant biopolymer which is also renewable. Chemically, cellulose is a linear polymer of \((1\rightarrow4)-\text{linked } \beta-D\text{-gluco-pyranosyl} \) residues [7]. Cellulose-based biofibres include cotton, flax, hemp, jute and sisal, and wood fibres. These biofibres are used to reinforce both synthetic polymeric matrixes and natural polymers because of their relative high strength, high stiffness and improved thermal properties [7,9]. Utilizing natural fillers from renewable resources not only contributes to a healthy ecosystem, but also makes them economically feasible for industrial applications [9].

The hydrophilic surfaces of the cellulose microfibres are expected to interact with the hydroxyl groups of partially hydrolyzed PVOH chains, which demand more efforts into exploring new and novel processes. Therefore, this research was designed to investigate the properties of poly(vinyl alcohol) adhesives with added cellulose microfibre from sugarcane bagasse as a filler. Sugarcane bagasse is a by-product obtained from sucrose extraction and has a high proportion of cellulose [11].

**EXPERIMENTAL**

**Materials**

Poly(vinyl alcohol) (CCP brand) was 98.5-99.2% hydrolyzed, with different molecular weights of 22,000 (BF05), 75,000-80,000 (BF17) and 112,000-120,000 (BF26) Dalton. The samples were purchased from Chang Chun Petrochemical Co. Ltd., Taiwan. Sugarcane bagasse fibres were obtained from local producers in Songkhla Province, Thailand.

**Preparation of Cellulose Microfibre**

Cellulose microfibres (CMF) were isolated from an oven dried sugarcane bagasse, and the samples had moisture content of about 11 g/100 g. Initially, the dried bagasse was cut into 4-5 cm lengths and then hydrolyzed with 10% (w/w) of nitric acid. The ratio of dried bagasse to acid liquor was 1/20 and heated at 90°C for 30 min [12]. The pulp was bleached by sodium hypochlorite with 10% active chlorine. The ratio of dried bagasse pulp to the bleaching agent was 5/1 [13]. Bleached fibre was then reduced in size by a blender (Philips model HR 2021, China) and screened with a sieve of mesh size number 200 (diameter size 0.074 μm).

**Adhesive Preparation**

Poly(vinyl alcohol) powder was added into de-ionized water. The mixture was heated to about 90-95°C with constant stirring until the solid was completely dissolved. The concentrations of the medium (75,000-80,000 Dalton, BF-17) and high molecular weight PVOH adhesives (112,000-120,000 Dalton, BF26) were each 12% (w/w). The volume of each sample was adjusted by the addition of water at this time to yield final concentrations as indicated above. Subsequently, the CMF powder was added as 0.5, 1.5, 2.5 and 3.5 wt% of total dry adhesive weight. Then, the mixtures were stirred at 80 rpm for 3 h to
allow the CMF filler to disperse in the PVOH adhesive. The samples of the adhesive were stored in closed containers and used for analysis.

**Characterizations**

**Chemical Analysis of Sugarcane Bagasse Fibres and CMF**
The sugarcane bagasse fibres and CMF were chemically analyzed for moisture and ash contents by using the A.O.A.C (1999) standard. The cellulose content of the fibres was measured by using Kurschner and Hoffer method [13].

**Scanning Electron Microscopy**
Scanning electron microscopy (SEM) was used to study the morphology of the bagasse fibre and CMF. The samples were mounted on bronze stubs and sputtered with gold (Sputter coater SPI-Module, PA, USA) in order to make the samples conductive. The micrographs were recorded at selected magnifications at 10.0 kV with FEI model Quanta 400, Japan.

**X-Ray Diffraction and Crystallinity Measurement**
The crystallinity of the cellulose fibre was examined by using a Philips X’Pert MPD system (Philips Inc., Netherland). The diffracted intensity of Cu Kα radiation was assessed at a voltage of 40 kV and 30 mA. The samples were dried and measured in a 2θ range between 10 and 50 degrees. Crystallinity was commonly measured as a ratio between the diffraction portion from the crystalline part of the sample, $A_c$, and the total diffraction from the same sample, $A_{total}$. The values of $A_c$ could be obtained after an appropriate subtraction of the scattering portion from the background, $A_b$. The relative crystallinity index was calculated by eqn (1) [14] as follows:

\[
\text{Crystallinity (\%)} = \frac{A_c \times 100}{A_{total}} \tag{1}
\]

and,

\[
A_{total} = A_c + A_b \tag{2}
\]

**Fourier Transform Infrared Spectroscopy**
FTIR spectroscopy was used to trace any changes in the chemical structure of the bagasse fibre, CMF and composite adhesives. The FTIR spectra were recorded with a Bruker Model Equinox 55 (Bruker Co., Ettlingen, Germany). The samples were prepared by mixing the fine powder with KBr and compressing the mixture. The spectra were obtained at a resolution of 4 cm⁻¹ in the range 4000 to 400 cm⁻¹.

**Differential Scanning Calorimeter**
Differential scanning calorimeter (DSC) thermograms were recorded by a Perkin Elmer DSC 7 instrument (Norwalk, CT, USA). The sample (7-10 mg) was placed in an aluminum pan. The first heating scan was run in the temperature range between 25 and 260°C at a heating rate of 10°C/min. The DSC melt thermograms showed the melting temperature ($T_m$) of the samples. The second heating run was performed with samples which were suddenly cooled to 6°C and heated to 300°C. This depicted the glass-transition temperature ($T_g$) of the samples.

**Testing of Adhesive**
The adhesives were subjected to analysis to determine their physical properties. These included the adhesive viscosity according to TIS.181-2530 using a Brookfield viscometer (model DV-II, USA); the solid content determination according to A.O.A.C, 1999; and the setting speed of the adhesives. The mechanical properties of PVOH adhesives were measured as the shear strength and elongation-at-break (EAB) according to TIS.360-2523 and ASTM D1002-72, 1982. The morphology of the CMF dispersion was investigated using an optical microscope (Olympus model CHS 3N 0182, Japan).

The specimens for mechanical testing were prepared from a 0.5 cm thick veneer which was cut to a dimension of 2.5×10 cm² (Figure 1). About 0.030-0.040 g of the adhesives per glued area (6.25 cm²,
approximately) were applied to one side of the veneer under contact with another piece of veneer for at least 10 test specimens. Initial grip separation was 80 mm and crosshead speed was set at 1.3 mm/min. The glued veneers were then pressed together under a pressure of 3.45 MPa (500 Psi) at ambient temperature. Test samples were maintained and conditioned at 23°C with relative humidity of 50% RH for 2 days before testing.

Shear strength and elongation-at-break of the veneer specimens were examined by a Universal tester (Lloyded Instrument, model LR30K, England). Shear strength and elongation-at-break were calculated by eqns (3) and (4), respectively.

\[
\text{Shear strength} = \frac{\text{maximum force}(N)}{\text{glued area}(\text{mm}^2)} \tag{3}
\]

\[
EAB(\%) = \left(\frac{d_{\text{after}} - d_{\text{before}}}{d_{\text{before}}}\right) \times 100 \tag{4}
\]

where, \(d_{\text{after}}\) and \(d_{\text{before}}\) are the distances between the grips holding the specimen before or after their breaking points, respectively.

RESULTS AND DISCUSSION

Effect of Chemical Treatment on Fibre Characterizations

Chemical Compositions of the Sugarcane Bagasse Fibre and CMF

The sugarcane bagasse fibres and CMF were chemically analyzed to measure the moisture, ash, cellulose and lignin contents. Before testing, the fibres were prepared by grinding and screening with a sieve, mesh No. 40. Table 1 shows some of the chemical compositions of the fibres. The moisture and ash contents of sugarcane bagasse fibres are 11.05% and 1.76%, respectively. The CMF showed moisture and ash contents of 5.85% and 1.46%, respectively.

Normally, the hydrogen bonds between cellulose molecules are aligned in a regular system resulting in an ordered system with crystal-like properties [14]. The fine structure of the cellulose materials is composed of crystalline and non-crystalline regions [15]. Crystalline cellulose consists of long chains while non-crystalline cellulose has shorter chains. As chemical treatments had effects on the crystallinity of the cellulosic fibres, it was considered desirable to retain pure cellulose, whose crystalline form and high packing density would result in a stronger composite.

Table 1 shows that the cellulose content of the fibres was initially 44.10% in sugarcane bagasse fibres and it had been increased to 88.99% after acid treatment. As it was expected the CMF showed higher cellulose content compared to sugarcane bagasse fibres, because acid treatment had removed most of the hemicellulose and lignin contents from the sugarcane bagasse fibres. This implies that the crystalline regions of cellulose in fibres must have increased after acid-treatment.

Morphologies of Sugarcane Bagasse Fibre and CMF

The morphological structure and size of the sugarcane bagasse fibres and CMF were observed using SEM, which are shown in Figures 2 and 3, respectively. A definite change in the morphological structure and size of the cellulose fibres occurred upon acid-hydrolysis. Both fibres seem to have had a long rod shape. Figure 2 shows the SEM micrograph of the sugarcane bagasse fibres which tended to be smooth. Some of small fractures on the fibres surface are resulted from grinding process. The morphology of the CMF surface is shown in Figure 3. The outer layer of the CMF was obviously destroyed and some chemical components were removed. At larger magnification as shown in Figure 3b, there are many terraces, steps and kinks formed on the fibre surfaces after acid hydrolysis.
In addition, the acid treatment had effect on the size of the cellulose. The CMF had lengths of 100-300 μm and diameters of 5-10 μm, which were lower than the sizes of the macrofibrils of sugarcane bagasse fibres. The sugarcane bagasse fibres had a length higher than 1000 μm and diameters of 100-150 μm (Figures 2 and 3). We have assumed that the morphological structure and size of the fibres might have changed due to the removal of the amorphous regions of the cellulose by acid treatment.

FTIR Characterization of Sugarcane Bagasse Fibre and CMF

The chemical structures of the bagasse fibres and CMF components are shown in Figure 4. The FTIR spectra of bagasse fibres show a strong characteristic carbonyl absorption peak at 1733.44 cm⁻¹. This was attributed to the acetyl and uronic ester groups of the hemicellulose or the carboxylate groups of the ferulic and p-coumeric acids of lignin and hemicellulose [14,16,17]. The peaks at 1513.89 and 1426.65 cm⁻¹ in the bagasse fibres show the aromatic C=C stretch of the aromatic rings of lignin [14,18]. The FTIR spectra of CMF shows the removal of pectins, lignin and hemicellulose resulting from the vanishing characteristic band at 1733.44 cm⁻¹ (carboxylate groups), 1605.51 cm⁻¹ (acetyl groups) and 1252.30 cm⁻¹ (methyl ester groups), 1513.89 and 1426.65 cm⁻¹ (aromatic C=C stretch), indicating that acid treatment had removed large amounts of pectin, lignin and hemicellulose. Equally this was in accordance with the chemical changes in the fibre surfaces and its crystallinity which was determined using XRD analysis.

Crystallinity Measurements of Sugarcane Bagasse Fibre and CMF

X-Ray crystallography was used to investigate the
crystallinity of the samples at different stages. X-Ray diffraction patterns of sugarcane bagasse and CMF are shown in Figure 5. There are 3 peaks for each treatment ($2\theta = 15.5, 22$ and $34.5$ degree). The peak at $2\theta = 22^\circ$ of the CMF is sharper than that of the sugarcane bagasse fibres. The sharper diffraction peak indicated higher degree of crystallinity in the structure of the treated fibres.

The crystallinity values were estimated as 30% and 50%, for the sugarcane bagasse and CMF, respectively. The crystallinity of the sample increased after each stage of the chemical treatment. The increase in the crystallinity of the CMF was due to the partial removal of the hemicellulose and lignin [14]. During the acid-treatment, cellulose chains breakage must have occurred primarily in the amorphous regions. The amorphous regions in the cellulose chains are susceptible to water or chemical penetration and degrade sooner than crystalline regions [15]. An increase in the number of crystalline regions increased the rigidity of cellulose. The higher crystallinity in the CMF is associated with the higher tensile strength of the fibres.

**Effect of CMF Addition on PVOH Composite Adhesives Properties**

**Morphology of PVOH Composite Adhesives**

Figure 6 shows the appearance of the composite adhesives with medium MW PVOH at a concentration of 12% (w/w) (BF17+1.5 wt% CMF). These images were recorded using an optical microscope at a magnification ×4. The CMF phases seem to consist of rod-like particles with the dimensions as given previously. The results indicated that CMF were evenly dispersed in the PVOH adhesive matrix.

**FTIR Characterization of PVOH Composite Adhesives**

FTIR spectroscopy was used to investigate the differences in chemical structures of PVOH adhesives and PVOH/CMF composite adhesives. Figure 7 shows the FTIR spectra of the following: PVOH BF-17 adhesive (spectrum a); PVOH BF-26 adhesive (spectrum b); PVOH BF-17+1.5 wt% CMF adhesive.
In FTIR spectra of PVOH composite it is found that the transmittance variations and wavenumbers are shifted. The bands of O-H groups and C-H bending in PVOH BF-17 and BF-26 structures are observed at 3440.7, 3434.8 cm\(^{-1}\) and 859.8, 855.5 cm\(^{-1}\), respectively [13]. Broader bands for the above groups have emerged and shifted to lower wavenumbers of 3406.5, 3421 cm\(^{-1}\) and 851.2, 853.2 cm\(^{-1}\), for the same composite adhesive structure.

FTIR spectra suggest that the hydrogen-bonding interactions between the hydroxyl groups of PVOH and the cellulose in CMF has led to a hybrid formation [19,20]. The peaks at 2920.61, 2924.4 cm\(^{-1}\) (C-H stretching) and 906.5, 914.6 cm\(^{-1}\) (C-C stretching) are shifted to higher wavenumbers such as:

**Figure 6.** The optical microscope images of: (a) surface morphology of PVOH adhesive (BF-17, 12%) and (b) CMF fibres composite PVOH adhesive (BF-17-12+1.5% CMF).

**Figure 7.** FTIR Spectra of adhesives: (a) PVOH BF-17, (b) PVOH BF-26, (c) PVOH BF-17+1.5% CMF, (d) PVOH BF-12+0.5% CMF and (e) CMF.
2941.2, 2939.4 cm\(^{-1}\) and 919.2, 917.6 cm\(^{-1}\), respectively. Furthermore, the 500-900 cm\(^{-1}\) fingerprint region is an indication of structural changes in the PVOH [21]. The composite adhesive spectra show that the absorption peak at 705 cm\(^{-1}\) has disappeared and the peaks at 512 cm\(^{-1}\) and 628-648 cm\(^{-1}\) are weakened. This region is similar to that of the CMF spectra.

The weakness, disappearance, and shift of the characteristic absorption band might have resulted from the interactions of different OH groups in the PVOH and CMF molecular chains [22]. This may indicate the development of new inter- and intramolecular hydrogen bonds and a change in the conformation between PVOH and CMF [23]. These results may suggest that chemical binding between PVOH and CMF molecules must have improved the compatibility and other properties of the composite adhesives.

**Thermal Properties of PVOH Composite Adhesives**

DSC is a useful method of characterizing polymers based on their exothermic and endothermic thermal transitions, such as glass transition temperature (\(T_g\)), melting temperature (\(T_m\)) and enthalpy of melting (\(\Delta H_m\)). The thermograms of PVOH and CMF/PVOH composite adhesives are shown in Figure 8. The values obtained for these parameters (Table 2) reflect the effect of CMF content used in preparation of the two types of composite adhesives.

As shown in Figure 8, the thermograms of the composite adhesives have two transitions. These are initial endothermic transition peaks which one corresponds to the \(T_g\), and another to the endothermic peak corresponding to their melting temperature; similar to the thermogram of the PVOH adhesives.

**Table 2. Thermal properties of PVOH and composite adhesives.**

<table>
<thead>
<tr>
<th>Adhesives</th>
<th>(T_g) (°C)</th>
<th>(\Delta C_p) (J/g)</th>
<th>(T_m) (°C)</th>
<th>(\Delta H_m) (J/g)</th>
<th>(X_c) (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PVOH BF17</td>
<td>40.15</td>
<td>0.869</td>
<td>220.20</td>
<td>50.84</td>
<td>32.80</td>
</tr>
<tr>
<td>PVOH BF26</td>
<td>41.34</td>
<td>0.749</td>
<td>219.03</td>
<td>46.89</td>
<td>30.25</td>
</tr>
<tr>
<td>BF17+1.5% CMF</td>
<td>39.37</td>
<td>0.330</td>
<td>224.70</td>
<td>53.82</td>
<td>34.72</td>
</tr>
<tr>
<td>BF26+0.5% CMF</td>
<td>41.18</td>
<td>0.662</td>
<td>227.20</td>
<td>48.77</td>
<td>31.46</td>
</tr>
</tbody>
</table>
The first scan shows wide endothermic peaks around 60-130°C (data not shown). They were presumably associated with the evaporation of free water and bound water that were absorbed in the adhesive samples [24,25]. The presence of $T_g$ and $T_m$ in the PVOH adhesive reflects its partial crystalline structure. The $T_g$ of the pure PVOH at a concentration of 12% (w/w) (BF17-12 and BF26-12) are similar, around 40.15 and 41.34°C, respectively. It is smaller than what was reported by other researchers [25]. Because PVOH adhesives already contained some water as a plasticizer, the thermogram showed a broad $T_g$ which might be due to the commercial PVOH with a wide distribution in terms of its molecular weight [24]. The DSC curves of PVOH composite adhesives compared with those of PVOH adhesives. It was observed that the addition of CMF showed no impact on $T_g$ in both of these PVOH adhesive types (BF17 and BF26, each with the same concentration of 12% (w/w)).

The peak at 219-220°C is related to the melting point of PVOH adhesives. When CMF was added, the melting temperature increased and the height of the melting peak increased. Samples containing CMF consume higher energy for material fusion and therefore this resulted in an increase in enthalpy of melting ($\Delta H_m$) and percentage of crystallinity when the CMF contents were 0.5 and 1.5 wt% of PVOH BF17 and BF26, respectively. This is in agreement with Luz et al. [26] who found that addition of lignocellulose introduces nucleating agents and changes the crystallization of the matrix around the fibre. Moreover, this may be because of strong interactions between the CMF and PVOH matrix. However, these strong interactions or cross-linking structures can be broken, resulting in a decrease in $T_g$ [9]. Because of the strong interaction between the PVOH molecules, few of them were destroyed by the addition of CMF, leading to an enhancement of the PVOH molecular mobility. However, the degree of crystallinity ($X_c$) of the material depends on calculation of the $\Delta H_m$ values [8]. The crystallinity of the PVOH adhesive was determined by using the following equation:

$$X_c = \frac{\Delta H_m \times 100}{\Delta H_m^c \times W} \tag{5}$$

We calculated the enthalpy $\Delta H_m$ values through the numerical integration of areas covered by the melting peaks and normalized by samples masses. We used the enthalpy of 155 J/g for a theoretical 100% crystalline PVOH ($\Delta H_m^c$) [27,28], and $W$ is the mass fraction of PVOH in the composite [26,29]. We estimated that the pure PVOH exhibited a crystallinity of about 32.80% and 30.25% for PVOH BF17 and PVOH BF26, respectively. In composite adhesives these figures slightly increased to 34.72% and 31.46% for PVOH-BF17+1.5 wt% CMF and PVOH-BF26+0.5 wt% CMF, as the above given order. The increase in the PVOH crystallinity observed indicated that CMF promoted crystallization of PVOH in the CMF/PVOH composite adhesives.

Shear Properties of the PVOH Composite Adhesives
Shear strength and elongation-at-break are the principal parameters relating to the mechanical properties of adhesives. The shear strength of adhesives also affects failures in veneers. If there is higher shear strength, there is also higher veneer failure [30]. The improvement of the mechanical properties of PVOH adhesives by CMF were investigated in this research. The criteria selected for judging PVOH adhesives for mechanical properties improvement were shear strength, viscosity and compatibility with additives.

Figure 9 shows the shear strength of PVOH adhesives containing different quantities of CMF (0-3.5 wt%). The shear strength of the medium MW PVOH (BF-17) adhesives increased from 1.55 to
2.41 MPa when increasing the CMF filler content from 0 to 1.5 wt%. The high MW PVOH (BF-26) adhesives increased in shear strength from 1.79 to 2.15 MPa when increasing the CMF filler content from 0 to 0.5 wt%. The results showed that the maximum shear force that was capable of separating or de-bonding joined hard-wood pieces glued together was an indication of strong interactions between the CMF and PVOH adhesive through hydrogen bonding. Similar results have been reported in PVOH/CMF composite films by Bhatnagar et al. [16]. However, the addition of CMF at 2.5 and 3.5 wt% could induce an accumulation which actually decreases the effective content of the CMF. Thus, PVOH adhesives with 2.5 and 3.5 wt% CMF contents exhibited lower shear strength compared to samples reinforced with 0.5 and 1.5 wt% CMF.

Data regarding the elongation-at-break is shown in Figure 10. Increasing the CMF filler content from 0 to 0.5 wt% provides an increase in elongation-at-break for high MW PVOH (BF-26) adhesive from 2.02 to 4.29%. When the CMF filler content was increased from 0 to 1.5 wt%, the elongation-at-break of the medium MW PVOH (BF-17) adhesives was increased from 1.43 to 4.31%. However, the addition of CMF filler of more than 2.5 wt% resulted in decrease elongation-at-break which can be explained by phase separation and the stiffness of the CMF [15].

Viscosity and Solid Content of the PVOH Composite Adhesives
The amount of cellulose microfibres influences the viscosity and solid content of PVOH adhesives as shown in Table 3. The viscosity and solid contents of the medium MW PVOH (BF-17) adhesives containing CMF (0-3.5 wt%) varied from 1,468 to 2,390 cps and 11.05 to 14.52%, respectively. PVOH adhesives with CMF filler contributed the higher viscosity compared to just PVOH adhesive. However, the viscosity had no effect on surface spreading efficiency of the veneer specimens.

After adding the CMF by 0 to 3.5 wt%, the viscosity and solid content of high MW PVOH (BF-26)
Adhesives also increased from 8,570 to 10,989 cps and 11.36 to 14.75%, respectively. In this case, increasing the adhesive viscosity had an effect on the spreading efficiency of test specimens, especially at 2.5 and 3.5 wt% CMF. The viscosity values of adhesives were over 10,000 cps thus the adhesives were difficult to spread. Therefore, incorporating CMF into PVOH adhesives led to increased viscosity and solid content of the adhesive. These effects might have been due to swelling of the cellulose polymer in PVOH adhesives.

**Adhesive Setting Time**

The quick setting time of adhesives is another requirement for the application of wood adhesives. This is the result of the property whereby solvents evaporate in adhesives. The PVOH adhesives consist of PVOH dissolved in water. Thus, the water is used as a solvent. The adhesives are "set" when a certain amount of water is evaporated [31]. Figure 11 shows the effect of the amount of cellulose microfibre on the setting speed of PVOH adhesives. The setting time of PVOH adhesives has been accelerated with increased amount of CMF. The decrease in the adhesive setting time is calculated as a percentage. The setting time of medium MW PVOH (BF-17) adhesives containing CMF contents of 0, 0.5, 1.5, 2.5 and 3.5 wt% are 100, 67, 33, 22 and 22%, respectively. In addition, incorporating CMF by 0, 0.5, 1.5, 2.5 and 3.5 wt% in high MW PVOH (BF-26) adhesives has given rise to setting times of 100, 67, 67, 67 and 33%, respectively. A maximum content of CMF in PVOH adhesives has displayed the lowest adhesive setting time. It may be concluded that the CMF is able to improve the setting speed of PVOH adhesives. Since cellulose is a hydrophilic polymer and a water-absorbing material, there are strong interactions between hydroxyl groups at the cellulose surface, PVOH polymer and water or solvent [15]. The adhesion of glues has been thus improved. However, the other potential effects of setting speed still depend upon the type of solvents, air-blowing steps and the evaporation temperature [32].

**CONCLUSION**

Many properties of the adhesive could be improved by the addition of CMF. The conditions set for CMF preparation were as follows: 10% (w/w) nitric acid and a cooking temperature of 90°C for 30 min. The diameters and the lengths of CMF were 5-10 μm and 100-300 μm, respectively. Chemical analysis as well as FTIR and XRD measurements of the fibres performed. These revealed that some chemical components of the fibres, such as hemicellulose, lignin and amorphous regions of cellulose were removed by acid treatment. The cellulose content of the CMF increased from 44.10% to 88.99%. The crystallinity value of the CMF was about 50%.

The CMF was well dispersed into PVOH adhesive. The FTIR spectra confirmed that chemical binding occurred between PVOH and CMF molecules. This is evident in improvements of the compatibility, thermal properties and the mechanical properties of the composite adhesives. The amount of CMF influenced the viscosity and solid content of the adhesives. The optimal viscosity of the adhesive for spreading on the wood surface was between 1,000 to 10,000 cps. From the DSC results it was established that the CMF could promote crystallization of PVOH in the composite adhesives and reduce the setting time of PVOH adhesives. It may be finally concluded that CMF made from sugarcane bagasse could improve the principal properties of PVOH adhesives.

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