# Chapter 4

# RESULTS AND DISCUSSION

# 4. Results and Discussion

#### 4.1 Preliminary Assessment of Processing Conditions (Pre-trials)

In searching for processing conditions that are optimal for the performance of the composites produced, there are several parameters that could be manipulated, such as mixing temperature, mixing time, rotor speed and fiber fraction. In pre-trial experiments, tensile properties of CAB/AAEF composites at a fiber content of 20% by weight (arbitrarily chosen) were evaluated at different combinations of mixing temperature and mixing time. The composites were prepared at 187 and 204 ° C for 10, 15, and 20 minutes at a rotor speed of 60 rpm. The latter was chosen as a compromise between fiber breakage and poor fiber dispersion, which tend to occur at high and low rotor speeds, respectively.

Generally, there are two questions involved in determining mixing time, (1) how soon to add fibers and (2) how long to knead. Fibers should be added once the polymer melt reaches a state that is close to a 'steady' state condition. This is because at this state, incorporation of fibers is easier and severe fiber breakage may be avoided due to low shear stress of the system. Kneading, which follows fiber addition, should also be carried out until the fiber/matrix mixture approaches a state that is also close to a 'steady' state condition. This will lead to a homogeneous mixture of components. Kneading should not be too long. The matrix may degrade; fiber off-gassing may lead to void formation; and the fibers may undergo fragmentation. As a result, composites may suffer loss of desirable properties, such as strength. Referring to the torque versus mixing time graph of pure CAB (Figure 4.1), the polymer melt was found to reach a state close to a 'steady' state after approximately 5 minutes of mixing (a total of 3 minutes of matrix addition and 2 minutes of melting) regardless of mixing temperatures. It was therefore decided to add fibers 5 min. after the introduction of matrix polymer. Fibers could not be added at a true 'steady' state since true 'steady' state is never reached. Matrix polymer depolymerizes with prolonged exposure to high temperature kneading leading to a progressive reduction in melt viscosity and torque. Similar arguments apply to kneading time after fiber addition (Figure 4.2), which was set to 15 minute regardless of mixing temperatures.

As for mixing temperature, this should represent a compromise between melt flowability (viscosity) and thermal stability of the fibers and the matrix. At low mixing temperature, polymer melt viscosity as well as shear stress were high. Kneading was difficult. Although fibers and matrix are unlikely to degrade thermally, the tendency of the fibers to undergo fragmentation was high. In contrast, at high mixing temperature, kneading was easier due to improved flowability of the polymer melt and low shear stress. The fibers may not fragment, but there exists a considerable risk of thermal degradation of the fibers as well as the matrix. Fiber fragmentation and thermal degradation of the fibers and the matrix need to be avoided particularly during processing since they may result in composites with inoptimal mechanical properties.

Effect of mixing time and mixing temperature on tensile properties of CAB/AAEF composites with 20% (by weight) fiber content is shown in Table 4.1. The table reveals a small effect (that can be ignored) of mixing time on properties. Mixing temperature on the other hand has a positive effect on stress and elongation at break.

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Figure 4.1 Mixing torque curves for pure CAB processed at 187 and 204 ° C for 10, 15, and 20 minutes at rotor speed of 60 rpm. CAB pellets were added in the mixing chamber within the first 3 minutes. After 2 minutes of melting, the melt reached a state that is closer to a 'steady state' condition (regardless the mixing temperatures). Addition of fibers was decided to begin at this particular moment.



Figure 4.2 Mixing torque curves for CAB/AAEF composites prepared at 187 and 204  $^{\circ}$  C in 10, 15, and 20 minutes at 60 rpm. CAB pellets were added into the mixing chamber in about 3 minutes. Once the polymer melt reached to a state close to a 'steady' state (~2 min.), fibers were added. Kneading then was done for 5, 10, and 15 minutes, which corresponds to mixing time of 10, 15, and 20 minutes.

Mixing temperature (° C)	Mixing time (min.)	Stress at <sup>1</sup> break (MPa)	Elongation <sup>1</sup> at break (%)	Modulus <sup>1</sup> (MPa)
187	10	24.73(3.37)	5.88(0.30)	660.35(61.17)
	15	27.57(3.14)	6.65(1.05)	755.26(37.41)
	20	26.88(2.20)	5.93(0.51)	789.94(47.23)
204	10	20.23(2.03)	5.06(0.31)	628.37(34.12)
	15	25.64(3.58)	5.73(0.75)	740.21(36.90)
	20	23.88(3.11)	4.86(0.43)	778.52(44.51)

Table 4.1 Results of Pre-trials: Tensile properties of CAB/AAEF composites at 20% fiber content by weight at different processing conditions.

<sup>1</sup>Mean values of five samples. Values in parentheses are standard deviations.

Both properties were found to increase as the mixing temperature was increased from 187 to 204  $^{\circ}$  C. As for modulus, the effect of mixing temperature was found to be irrelevant since the composites studied were all reinforced with the same amount of fiber content, i.e., 20% by weight.

Based on these pre-trial experiments, the processing conditions selected for further study were set at 204  $^{\circ}$  C for mixing temperature and 20 minutes for mixing time (time taken from the beginning of polymer addition to end of kneading time with fibers).

#### 4.2 Assessment of Fiber Types

In this study various types of cellulose fibers were used to reinforce cellulose acetate butyrate (CAB), a commercially available melt-processable, plasticized cellulose ester. The fibers used were steam-exploded fibers from Yellow Poplar wood chips and commercial oat filler. The steam exploded fibers had first undergone fiber fractionation, which involved water-washing to remove water-soluble degradation products from hemicelluloses and alkali extraction to separate alkali-soluble lignin. The resulting fibers, water-extracted (WEF) and alkali-extracted steam exploded fibers (AEF), had virtually identical geometry (i.e. size, shape and size distribution). AEF was then esterified by reacting the fibers with acetic anhydride to produce (surface) acetylated steam exploded fibers (AAEF). The objective of this work was to study the effect of these treatments on the performance of steam exploded fibers as reinforcements of thermoplastic composites. The composites were prepared using a Haake Rheomix 900 kneader at a mixing temperature of 204 ° C, and this was selected as a compromise between fiber stability and melt flowability. Mixing time and rotor speed was set at 20 minutes and 60 rpm, respectively, based on several pre-trials at other conditions. Fiber content ranged from 10% to 40% by weight. Changes in torque during processing, mechanical properties, fracture surfaces as well as fiber dispersion were evaluated.

#### 4.1.1 Mixing torque

The mixing torque behavior of the CAB/fiber melt suspensions studied were found to vary depending on the type and the amount of fibers blended with the matrix (Figures 4.3, 4.4, 4.5, and 4.6). Two peaks are observed. The first broad, irregular peak, which extends from approximately 0 to 180 sec, is due to sequential feeding of the matrix material in pellet form. The second peak, which is observed between approximately five and ten minutes of mixing, is due to fiber addition. Torque then decays gradually. Compounding was terminated after a total mixing time of 20 minutes, or 15 minutes after the fibers were added to the matrix melt.

The results indicate that torque (at the end of compounding) increases simultaneously with fiber content regardless of the type of fibers added (Figure 4.3-4.6). This effect, which is clearly shown in Figure 4.7 may be explained with the change in melt viscosity of the polymer-fiber suspension as a result of fiber addition. Fibers, when added into polymer melts tend to perturb the normal flow of the pure polymeric liquid by



Figure 4.3 Mixing torque curves for CAB/WEF composites at different fiber content processed at 204  $^{\circ}$  C for 20 minutes.



Figure 4.4 Mixing torque curves for CAB/AEF composites at different fiber content processed at 204  $^{\circ}$  C for 20 minutes.



Figure 4.5 Mixing torque curves for CAB/AAEF composites at different fiber content processed at 204  $^{\circ}$  C for 20 minutes.



Figure 4.6 Mixing torque curves for CAB/COF composites at different fiber content processed at 204  $^{\circ}$  C for 20 minutes.



Figure 4.7 Change in torque (compared to pure CAB) vs. fiber content for Cellulose fibers-CAB composites processed at 204  $^{\circ}$  C for 20 minutes.

hindering the mobility of chain segments in flow [40]. This results in a fiber-filled system with a higher melt viscosity than the pure polymer. Increasing fiber content will further restrict polymer flow. Thus, an increase in melt viscosity will be observed with fiber content rising, and this requires higher torque for compounding.

The results of torque vs. fiber content (Figure 4.7) also reveal that torque, at the end of compounding, varies distinctly with fiber type. AAEF generates the highest torque increase relative to CAB-melt, approximately four times that of pure polymer. This is followed by COF, AEF, and WEF. COF may not be comparable as it has much different fiber geometry. Torque is seen to increase progressively to approximately 421% of pure CAB when 40% AAEF is added to the matrix melt. This compares to 310%, 260%, and 190% for COF, AEF, and WEF, respectively. Since all steam exploded fibers, i.e., WEF, AEF, and AAEF, have approximately the same fiber geometry (Figure 4.8), the different effect of the fibers on torque during compounding must be attributed to differences in surface character resulting from the specific post-treatment undergone by each fiber. Comparing torque differences between WEF and AEF (Figure 4.7 and Table 4.2) shows that WEF consistently generates lower torque values than AEF at the same weight fraction. This must be due to the presence of lignin in WEF. As for AAEF, the significant rise in torque observed must be attributed to fiber (surface) acetylation, which was carried out to promote fiber-matrix interaction between the fibers and the CAB matrix.

Chemically treated (i.e., esterified) fibers tend to adhere strongly to a thermoplastic polymer matrix as a result of surface modification. Since this contributes to increased friction between the fibers and the polymer melt, it limits the mobility of

80



400 μ

Figure 4.8 Optical micrographs of cellulose fibers (before compounding).

A: WEF, B: AEF, C: AAEF, and D: COF. Note the similarities between all steam exploded (ie, A-C), and the different fiber geometry for COF (ie, D).

Fiber content (wt %)	CAB/WEF	Torque difference (%) <sup>1, 2</sup> CAB/AEF CAB/AAEF CAB/COF		
10 20 30	$79 \pm 7$ 117 ± 2 138 ± 2	76 ± 2 124 188 + 2	$64 \pm 2$ $148 \pm 2$ $252 \pm 5$	$62 \pm 2$ 114 ± 2 209 + 2
40	$190 \pm 2$	259±2	$421 \pm 5$	$309 \pm 5$

Table 4.2 Torque difference (compared to neat CAB) for cellulose-based composites processed at 204  $^{\circ}$  C for 20 minutes.

<sup>1</sup> Torque difference compared to neat CAB.

<sup>2</sup> Mean values of two samples.

polymer chain segments relative to fiber-free melts. As a consequence, an increase in melt viscosity during processing is observed. Since viscosity is directly related to torque (at constant temperature, rotor speed, etc.), this explains the distinct increase in torque when acetylated fibers were blended with the CAB matrix relative to WEF and AEF.

Thomas *et al.* (1996) [43] also observed increases in viscosity when chemically treated fibers (treatments based on poly(methylene)-poly(phenyl)isocyanate (PMPPIC), silane and peroxide) were melt compounded with low-density polyethylene. This was also attributed to improved fiber-matrix interaction between the two components.

In summary, fiber type as well fiber content significantly affect mixing torque. AAEF contributes the highest increase in torque, and this is attributed to improved interaction between the fibers and the CAB matrix. The increase in torque was followed by AEF and WEF. COF also showed an increase in torque, and this was higher than either AEF or WEF. This difference must be explained with differences in fiber geometry (i.e., smaller dimensions, more fines).

#### 4.1.2 Mechanical properties

The mechanical properties of CAB-based composites reinforced with various cellulose fibers were evaluated by tensile testing at a strain rate of 0.125 mm/min.. Strain-stress curves of the composites produced are shown in Figure 4.9-4.12. Maximum tensile



Figure 4.9 Tensile stress-strain curves for CAB/WEF composites at fiber contents rising from 10 to 40% by weight.



Figure 4.10 Tensile stress-strain curves for CAB/AEF composites at fiber contents rising from 10 to 40% by weight.



Figure 4.11 Tensile stress-strain curves for CAB/AAEF composites at fiber contents rising from 10 to 40% by weight.



Figure 4.12 Tensile stress-strain curves for CAB/COF composites at fiber contents rising from 10 to 40% by weight.

stress of CAB/WEF, CAB/AEF, and CAB/COF composites was found to decline dramatically (i.e., by 42-48%) with fiber content (Figure 4.13 and Table 4.3). This is consistent with the observations of other investigators, such as Kokta et al. (1987) [38] and Gatenholm et al (1991) [36], who also observed decreases in maximum tensile stress with fiber loading while studying cellulose-based composites. These authors attributed the observation to poor interfacial adhesion between the cellulose fibers and the matrix. This conclusion was supported by SEM micrographs of fracture surfaces of the composites studied in which smooth fiber pull-out was observed. Since WEF, AEF, and COF are hydrophilic (cellulose fibers have typically a high equilibrium moisture content) while CAB is hydrophobic, limited interfacial adhesion between the fibers and the CAB matrix is expected. This results in composites with poor mechanical properties. Other possible reasons that may lead to a continuous decline of maximum tensile stress with fiber content are voids (Figure 4.14), which may result from the release of volatile substances from the fiber (i.e. fiber "off-gassing") during compounding or compression molding. Voids would contribute to stress concentration effects as well as reduced density. The results (Figure 4.14) indeed reveal significant differences in void volume between the four fiber types, with AAEF having the lowest void volume.

Another factor influencing the mechanical properties of composites involves fiber length and aspect ratio. While the original fibers typically had lengths of up to 1.4 mm (Figure 4.8) at aspect ratios of nearly 1000, fibers recovered from finished composites (by matrix removal by dissolution in acetone) typically had maximum lengths of only 200-400  $\mu$ m (Figure 4.15).



Figure 4.13 Effect of fiber content on stress at break of CAB reinforced with various cellulose fibers.

Fiber	ber Maximum tensile stress (MPa) <sup>1</sup>						
content					Statistical difference		
(wt. %)	WEF (a)	AEF (b)	AAEF (c)	COF (d)	at 95% confidence		
0	30.5 (0.5)	30.5 (0.5)	30.5 (0.5)	30.5 (0.5)	none		
10	24.5 (1.8)	21.8 (0.4)	19.5 (1.6)	18.7 (0.4)	a, b vs. c vs. d		
20	20.8 (1.9)	18.7 (1.2)	22.6 (1.1)	14.2 (1.1)	a, b vs. c vs. d		
30	18.9 (1.5)	17.5 (3.3)	24.7 (4.9)	15.3 (1.1)	a, b vs. c vs. d		
40	17.7 (3.2)	15.9 (0.5)	33.7 (1.9)	16.8 (1.3)	a, b vs. c vs. d		

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Table 4.3 Stress at break of cellulose fiber-based composites.

<sup>1</sup> Mean values of five samples. Values in parentheses are standard deviations.



 $400\,\mu m$ 

Figure 4.14 Optical micrographs of cellulose-based composites showing voids.

A: CAB/WEF, B: CAB/AEF, C: CAB/AAEF, and D: CAB/COF.



200 µm

Figure 4.15 Optical micrographs of cellulose fibers recovered from finished composites by matrix dissolution in acetone. A: CAB/WEF, B: CAB/AEF, C: CAB/AAEF, and D: CAB.COF. Note the relative uniformity in fiber dimension in contrast to the beforecompounding differences (Figure 4.8).

This indicates that fibers are broken during compounding and/or compression molding, and that all fibers that effectively reinforce the matrix are of the same order of magnitude in size as the COF-filler. Preservation of the original fiber dimension would supposedly require changes in matrix melt viscosity.

As for CAB/AAEF composites, maximum tensile stress is found to increase (by 10.5%) with fiber content rising to 40% after showing an initial decrease of 36% at 10% fiber by weight (Figure 4.13, Table 4.3). The initial decrease in maximum stress is expected since at low fiber loading the fibers tend to act as flaws instead of as reinforcements. This reduces the strength of the composites. As the fiber content increases, however, the maximum stress increases up to 110% of the matrix at 40% fiber content by weight (Figure 4.13). This positive effect of AAEF on the maximum tensile stress of CAB composites, which also indicates the ability of the fibers to act as reinforcement, is expected and must be attributed to superior fiber-matrix interfacial adhesion as a result of fiber acetylation. Improved interaction between AAEF and CAB matrix is the consequence of molecular compatibility between the two acetylated components. The variation of maximum tensile stress with fiber content observed is consistent with the behavior predicted according to the 'Rule of Mixtures' (Figure 4.16) [6]. The model assumes unidirectional composite architecture with continuous fibers and perfect interfacial stress transfer. In order to take into account the influence of fiber length as well as fiber orientation,  $\alpha_1$  (length correction factor) and  $\alpha_2$  (orientation efficiency factor) may be introduced. The assumed mechanical properties of the fibers



$$\sigma_{ll}^{*} = \alpha_{1}\alpha_{2} \sigma_{f}^{*}V_{f} + \sigma_{m}^{'} (1 - V_{f})$$
(Fiber)  

$$\sigma_{ll}^{*} = \sigma_{m}^{*} (1 - V_{f})$$
(Matrix)  

$$\sigma_{m}^{'} = \varepsilon_{f}^{b} E_{m}$$

Where,

 $\sigma_{ll}^*$  = Stress at break of the composite parallel to fiber orientation (MPa)

 $\sigma_m^*$  = Stress at break of the matrix (MPa)

 $V_f$  = Fiber volume fraction (%)

 $\alpha_1$  = Length correction factor

 $\alpha_2$  = Orientation efficiency factor

 $\epsilon_{\rm f}{}^{\rm b}=Elongation$  at break of the fiber

 $E_m = Modulus of the matrix$ 

Figure 4.16 Variation of tensile stress with fiber fraction as predicted using the 'Rule of Mixtures'. See Table 4.4 for assumed values for fibers and matrix.

(AAEF) and the CAB matrix used are listed in Table 4.4. The fiber content beyond which the fibers can reinforce, i.e., sustain a fully transferred load, is called the critical fiber concentration (by volume fraction) (V<sub>crit.</sub>). V<sub>crit.</sub> is shifted towards higher values as the diminishing effects of fiber length and fiber orientation become increasingly important (Figure 4.16). The maximum stress increases with fiber content beyond the critical fiber concentration if fiber length and fiber orientation are adequate for reinforcement. The model predicts a substantial increase in strength with fiber content rising when  $\alpha_1 \alpha_2$ equals 1.0. This assumes (a) that the average fiber length is infinite or very much larger than the critical fiber length; (b) that all fibers are oriented in a single direction; and (c) that interfacial stress transfer is perfect. These conditions are not given and deductions must be made to compensate for short fiber lengths, random orientation and less than perfect interfacial stress transfer (i.e.,  $\alpha_1 \alpha_2 \ll 1.0$ ). A comparison between the results of the "Rule of Mixtures" assuming  $\alpha_1 \alpha_2 = 0.5$  and those obtained experimentally reveal that the actual composite data parallel those of the model (Figure 4.17). The critical fiber concentration for CAB/AAEF composites in which the fibers begin to carry the tensile stress is found to be 10% by weight (Figure 4.17) while the model predicts ca. 17%. Maximum strength at 40% fiber content is predicted to be 55 MPa as opposed to 34 MPa for the actual composite (Figure 4.17). Based on the model and the experimental results, AAEF can be assumed to orient randomly in the CAB matrix, and the fibers do show a broad distribution in fiber length (Figure 4.15 C). This explains the reason for the relatively small increase in maximum tensile stress with fiber content in the case of the experimental data. A greater increase in stress with fiber content and a lower V<sub>crit</sub> may be

Property	CAB	AAEF	
Maximum tensile stress, $\sigma$ (MPa)	30	250	
Elongation at break, $\varepsilon$ (%)	60	0.86	
Modulus, E (MPa)	640	1000	

Table 4.4 Assumed mechanical properties of AAEF and CAB.



Figure 4.17 Comparison of composite tensile strength property as related to fiber fraction for model ('Rule of Mixtures' at  $\alpha_1 \alpha_2 = 0.5$ ) and experiment (CAB/AAEF).

achieved with CAB/AAEF composites if longer fibers were used (or the original fiber length was preserved), or if the fibers were aligned during processing to be oriented in a single direction. Thomas *et al.* (1996) [43] investigated the effect of fiber orientation on tensile properties of polystyrene composites reinforced with sisal fibers, and they and found a similar variation in maximum tensile stress with fiber content (Figure 4.18). The authors achieved a modest increase in strength with fiber content rising when the composites were reinforced with benzoylated fibers that were unidirectionally aligned.

All cellulose fiber-reinforced composites revealed declining elongation at break values with fiber content increasing. An approximately 80-93% reduction was recorded at 40% fiber content compared to the pure CAB (Figure 4.19, Table 4.5). Stiffness (or modulus) was found to increase consistently with fiber content (Figure 4.20). AAEF again produced the highest modulus increase, followed by AEF and WEF. Increases ranged between 47 and 103% at 40% fiber content as compared to pure CAB (Table 4.6). According to Cox [6], the modulus of a short fiber composite depends on several factors, including fiber length, fiber orientation, fiber and matrix volume fractions as well as modulus of both components. Since the fiber treatments are suspected to have only a minor impact on fiber properties, and the matrix properties are assumed to remain the same regardless of composite system, it is expected that all observed differences in the composite modulus with different fiber types must be attributed to other factors, such as void content, fiber length and fiber orientation. COF was not found to contribute noticeably to modulus regardless of fiber content.



Figure 4.18 Ultimate tensile strength of benzoylated sisal fiber reinforced polystyrene composites as a function of fiber orientation according to Thomas *et al.* [43].



Figure 4.19 Effect of fiber content on elongation at break of CAB reinforced with various cellulose fibers.

Fiber content (wt. (wt. %)	. %) WEF (a)	Elongation AEF (b)	at break (%) <sup>1</sup> AAEF (c)	COF (d)	Statistical difference at 95% confidence
 		. ,			
10	8.2 (0.7)	7.0 (0.6)	6.2 (0.9)	7.6 (0.8)	a, d vs. b, c
20	5.0 (0.4)	5.0 (0.5)	5.7 (0.7)	5.1 (0.7)	a, b vs. b,c
30	3.7 (0.2)	4.1 (0.8)	4.2 (0.7)	4.4 (0.8)	a vs. b, c, d
40	3.2 (0.7)	3.5 (0.4)	3.9 (0.4)	4.3 (0.2)	a, b, c vs. d

Table 4.5 Elongation at break of cellulose-based composites.

<sup>1</sup> Mean values of five samples. Values in parentheses are standard deviations.



Figure 4.20 Effect of fiber content on modulus of CAB reinforced with various cellulose fibers.

Fiber content (wt.%)	WEF (a)	Modulus (M AEF (b)	Pa) <sup>1</sup> AAEF (c)	COF (d)	Statistical difference at 95% confidence
0	640.0 (62.4)	640.0 (62.4)	640.0 (62.4)	640.0 (62.4)	none
10	640.6 (33.4)	611.2 (33.7)	523.8 (14.5)	478.1 (30.0)	a,b vs. c vs. d
20	732.0 (23.5)	620.2 (33.9)	685.6 (30.1)	527.5 (60.8)	a vs.b vs. c vs. d
30	840.5 (55.5)	736.9 (91.6)	1027.6 (82.8)	449.6 (54.4)	a vs. b vs. c vs. d
40	956.2 (86.4)	839.7 (29.0)	1298.9 (122.1)	663.0 (75.7)	a vs. b vs. c vs d

Table 4.6 Effect of fiber content on modulus of cellulose-based composites.

<sup>1</sup> Mean values of five samples. Values in parentheses are standard deviations.

### 4.1.3 Scanning electron microscopy

The fracture surfaces of the 20% composites studied by tensile testing were examined using scanning electron microscopy (SEM) at magnifications of 500x and 2000x (Figures 4.21 and 4.22). Other composites were examined as well (i.e., 40% fiber content), but the results were found to be most apparent at the 20% fiber content level. Whereas a rather smooth fracture surface is seen in the CAB/AAEF composite, fiber pull out as well as holes resulting from fiber delamination are observed for the composites with WEF, AEF, and COF fibers. The fracture surfaces of the CAB/WEF, CAB/AEF, and CAB/COF composites (Figure 4.21) reveal extensive interfacial delamination between the fibers and the matrix. Fiber pull-out of each composite is shown in Figure 4.22. It can be seen that WEF, AEF, and COF are hardly wetted by CAB matrix with a gap of approximately 2 to 4  $\mu$ m developing around the fibers (see arrow in Figure 4.22). The fibers also stay intact after tensile testing. Normally, pulled-out fibers and gaps are observed when the adhesion between fibers and matrix is not sufficient, and this suggests poor fiber-matrix compatibility. Poor fiber-matrix compatibility was expected for unmodified cellulose fibers, and this improves with surface acetylation.

In contrast to unmodified fibers, AAEF is found to have been coated by the matrix. This indicates that the fibers are compatible with the CAB matrix. The fibers are also severely fractured into fibrils after the tensile test suggesting that the fibers did get strained up to their maximum stress values and that the average fiber length (after processing) was above the critical fiber length ( $l_c$ ). The critical fiber length is influenced by several parameters including interfacial shear stress according to equation # 2.1.

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20 µm

Figure 4.21 Scanning electron micrographs of tensile fracture surfaces of 20% cellulose fiber/CAB composites at magnification of 500 X. A: CAB/WEF, B: CAB/AEF, C: CAB/AAEF, and D: CAB/COF.



10 µm

Figure 4.22 Scanning electron micrographs of tensile fracture surfaces of 20% cellulose fiber/CAB composites at magnification of 2000 X. A: CAB/WEF, B: CAB/AEF, CAB/AAEF, and D: CAB/COF.

Improving fiber-matrix interaction, i.e.,  $\tau_y$ , through acetylation, improves interfacial stress transfer between fibers and matrix and leads to a reduction in the value of the critical fiber length. The observation of AAEF fiber failure by fibrillation suggests improved interfacial adhesion between AAEF and CAB. This must be attributed to the similarity between the acetyl groups present in both components.

## 4.1.4 Fiber dispersion

The quality of fiber dispersion of a composite material, as discussed in section 2.3, can be quantified by the use of image analysis. In this technique, light or another source of radiation, is passed through thin sections of a composite. Digitized radiographic images are processed using a computer equipped with an image analysis package to determine 'standard deviation of gray level' (see Section 2.3). 'Standard deviation of gray level', which is used to represent quantitatively fiber dispersion, is related to pixel-to-pixel variation of light intensity within an image of interest. The more evenly dispersed the fibers, the more uniformly gray will be the digitized image. The values of 'standard deviation of gray level' then will be small for samples with uniform fiber dispersion.

In this study, composite sections of 40  $\mu$ m thickness were observed under a light microscope using the same amount of light and a computer equipped with the Metamorph Image Analysis System.

Images of AAEF-composites progressively reveal darker images with fiber content rising (Figure 4.23). Due to the difficulties of obtaining images with distinct

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Figure 4.23 Images of CAB/AAEF composites at various fiber content at magnification 0f 10x. A: 10% fiber content, B: 20% fiber content, C: 30% fiber content, and D: 40% fiber content.

contrast between light and dark in the composites with high fiber content, only composites with 20% fiber content were analyzed (Figure 4.24). At 20% fiber content, the best fiber dispersion is achieved with AAEF followed by AEF, WEF, and COF (Table 4.7). These differences can also be attributed to the particular post-treatment of each fiber. AAEF showed the lowest 'standard deviation of gray level' or the best fiber dispersion as a result of fiber acetylation. WEF and AEF were not significantly different. This suggests that water-washing and alkali-extraction do not sufficiently alter dispersability in the CAB matrix. Improved fiber dispersion can only be achieved if steam exploded fibers are water-washed, extracted with alkali and then acetylated to produce acetylated fibers, AAEF.

Scott (1997) [10, 11] quantified fiber dispersion as well as fiber loading in glass fiber-reinforced nylon composites in terms of 'standard deviation of gray level' and 'average gray level', respectively [10, 11]. Melt viscosity, a factor suspected to be important to fiber dispersion, was found to correlate well with fiber dispersion, i.e., standard deviation of gray level' [10]. Fiber dispersion was found to increase, i.e., the 'standard deviation of gray level' decreased, as melt viscosity of the matrix increased. Scott attributed this behavior to better wetting of the glass fiber by the matrix.

Since melt viscosity is related to torque, with torque varying directly with melt viscosity at constant temperature and shear rate, an attempt was made to correlate the observed 'standard deviation of gray level' with torque (Table 4.7 and Figure 4.25). At a

fiber content of 20%, the 'standard deviation of gray level', or fiber dispersion, exhibits a significant correlation with mixing torque (coefficient of correlation,  $R_2$  of 0.87). Therefore instead of melt viscosity, fiber dispersion or 'standard deviation of gray level' can also be correlated with torque. This correlation, however, is specific to the conditions selected, i. e., a mixing temperature of 204  $^{\circ}$  C, a mixing time of 20 minutes, a rotor speed of 60 rpm and a fiber length of approximately 200  $\mu$ m.



Figure 4.24 Images of cellulose-based composites at 20% fiber content at magnification of 10x. A: CAB/WEF, B: CAB/AEF, C: CAB/AAEF, and D: CAB/COF. Results of image analysis are given in Table 4.7.

	Fiber content (% by weight)	Mixing torque (Nm)	'Std. deviation of gray level'	Statistical 'Std. deviation of gray level'- difference at 95% confidence
CAB/WEF (a)	20	2.45	55.972	a, b, d vs. c
CAB/AEF (b)	20	2.60	55.286	
CAB/AAEF (c)	20	3.10	50.407	
CAB/COF (d)	20	2.40	59.855	

Table 4.7 Mixing torque and 'standard deviation of gray levels' of cellulose-based composites having different fiber content.



Figure 4.25 'Standard deviation of gray levels' (fiber dispersion) versus mixing torque for cellulose-based composites at 20% fiber content by weight.