Fiber-Reinforced Cellulosic Thermoplastic Composites

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ABSTRACT: Steam-exploded fibers from Yellow poplar (Liriodendron tulipifera) wood were assessed in terms of their thermal stability characteristics, their impact on torque during melt processing of a thermoplastic cellulose ester (plasticized CAB) matrix, their fiber-matrix adhesion and dispersion in composites, and their mechanical properties under tension. Fibers included water-extracted steam-exploded fibers (WEF), alkali extracted fibers (AEF), acetylated fibers (AAEF), and a commercial milled oat fiber sample (COF) (i.e., untreated control). The results indicate that the thermal stability of steam-exploded fibers increases progressively as the fibers are extracted with water and alkali and following acetylation. The greatest improvement resulted from the removal of water-soluble hemicelluloses. The modification by acetylation contributed to improved interfacial wetting that was revealed by increased torque during melt processing. Whereas modulus increased by between 0 and 100% with the incorporation of 40% fibers by weight, tensile strength either declined by $\frac{1}{3}$ to $\frac{1}{2}$ or it increased by a maximum of 10%, depending on fiber type. AAEF composites produced the best mechanical properties. Fiber–aspect ratio was reduced to an average of 25–50 from ≥ 200 during compounding. The superior reinforcing characteristics of AAEF fibers were also reflected by SEM, which revealed better fiber-matrix adhesion and failure by fiber fibrillation rather than by fiber pullout. © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 73: 1329-1340, 1999

Key words: wood fibers; composites; thermal stability; cellulose acetate butyrate; steam explosion; acetylated wood

INTRODUCTION

The use of cellulose-rich fibers from renewable resources has been gaining considerable interest in the composites field.^{1–3} A combination of properties, such as high aspect ratio, relatively high tensile strength and bending modulus, low density, and low cost, all contribute to a rising interest from the manufacturers of low-cost, low-weight composites. Although cellulose fiber-rein-

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forced biodegradable composites have been tested using thermoplastic, fermentation-produced polyesters,^{4,5} most studies on wood fiber-reinforced composites have concentrated on nondegradable polyolefins, especially polypropylene.^{6–12} However, biodegradability continues to be one of the parameters significantly in favor of cellulosebased composites.

Well-publicized constraints on the use of cellulosic materials in thermoplastic composites include the thermal instability of biomass fibers at the typical processing temperatures of thermoplastics (around and above 200°C), their hydrophilic character that is responsible for poor interfacial adhesion and poor fiber dispersion, and a modulus that is strongly dependent on cellulose

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content and fibril orientation.^{13,14} Fiber pretreatments, although having a negative impact on economics, are potentially able to overcome these limitations. Among candidate pretreatment methods are steam explosion and surface acetylation.^{7,10,15–18}

The objectives of the current study involve an examination of the impact of steam explosion, removal of water- and alkali-solubles from the steam-exploded fibers, and surface acetylation on fiber properties and qualifications for thermoplastic composites. For reasons of compatibility and biodegradability, a commercially available thermoplastic cellulose ester was selected as matrix.

EXPERIMENTAL

A. Materials

1. Fibers

All fibers were generated by steam explosion of Yellow poplar (Liriodendron tulipifera) wood chips using a severity of $\log R_0$ 4.23. The severity factor R_0 is, as defined by Overend and Chornet,¹⁹ $R_0 = \int_0^t \exp[(T_1 - T_0)/14.75] dt$, where T_0 and T_1 are base and reaction temperature, respectively, and t is retention time. The fibers were processed in the usual manner using water extraction at 60°C and extraction with 20% (based on fiber solids weight) alkali at an 8 : 1 liquor-tofiber ratio at 60°C for 30 min.^{20,21} Water-extracted fibers (WEF) and alkali-extracted fibers (AEF) were dried separately by spreading out in a laboratory fume hood. AEF fibers were also acetylated by using a suspension of dry fibers in toluene/acetic acid followed by the addition of acetic anhydride and by heating the mixture in a Parr (Moline, IL) reactor at 140°C for 8 h. This resulted in acetylated fibers (AAEF) under heterogeneous conditions, with an average weight gain of about 15% by weight. Degree of fiber modification was assessed by FTIR spectroscopy using the absorption peak centered around 1745 cm^{-1} as indicator.

2. Matrix

Commercially available plasticized cellulose acetate butyrate (CAB),²² was provided by Eastman Chemical Company of Kingsport, TN under the designation Tenite Butyrate Formula 285. Its reported Vicat softening temperature (according to ASTM D1525) was 115°C, and the specific gravity (ASTM D792) was reported as 1.2. The resin was used as supplied.

3. Chemical Wood Components

Thermal stability tests involved powderous cellulose, xylan, and lignin that were obtained from the pilot plant of the Biobased Materials/Recycling Center, Virginia Tech, Blacksburg, VA, under the designation steam-exploded bleached yellow poplar cellulose powder, as alkali-soluble heteropolysaccharide (ca. 65% xylose content) from mildly prehydrolyzed, delignified yellow poplar wood chips²³, and as alkali-soluble lignin from water-washed steam-exploded wood chips.²⁰ Their chemical characteristics are described elsewhere in detail.²⁴

B. Methods

1. Determination of Thermal Stability

Thermal stability was assessed by either a dynamic or a static thermogravimetric analysis (TGA) experiment. In the dynamic test, fiber samples were subjected to temperature rising at 10°C min⁻¹ in an atmosphere of air from room temperature to 150°C, followed by a more gradual rise of 2°C min⁻¹ from 150 to 400°C. An arbitrary weight loss figure of 2% of the weight recorded after the moisture loss at 140°C was chosen to define thermal stability. Fiber samples then varied with regard to the temperature at which the 2% weight loss threshold was surpassed.

In the static thermal stability test, fibers were heated to 200°C using a gradient of 10°C min⁻¹ from room temperature and kept isothermally for 2 h in an atmosphere of flowing nitrogen (45 mL/min⁻¹). The total weight loss under these conditions was determined for each fiber type.

2. Melt Processing

CAB and CAB-fiber mixtures were melt processed in a Haake Rheomix 900 (Paramus, NJ) twin screw kneader at an actual temperature of 204°C. Processing conditions involved melting the thermoplastic matrix in the preheated mixing chamber of the kneader for 5 min using a rotor speed of 60 rpm. Carefully dried fibers (vacuum oven, 60°C, <5 mm, and desiccator, CaCl₂) were then added to the mixing chamber that had been disintegrated for 2 min in a Waring blender in dry state. This produced a fluffy pulp that could easily be handled and weighed. Compositions were prepared having fiber contents between 0 (control) and 40% by weight by mixing molten matrix and fibers in the kneader for 15 min while maintaining a rotor speed of 60 rpm. Continuous, on-line torque measurements provided assurance that matrix (before fiber addition) and matrix-fiber mixtures had reached a uniform mixing state so as to eliminate processing variables from the study goals. Twenty minutes after adding matrix pellets into the mixing chamber, the kneader was stopped and cooled with water. The thermoplastic fiber mixture was recovered and stored for subsequent compression molding experiments. The samples were molded in a Carver hot press at 195°C using a constant pressure of 0.5–0.75 metric tons of pressure and 2.5-mm spacers.

3. Scanning Electron Microscopy

Scanning electron microscopy (SEM) of freezefractured surfaces of composite test specimens were conducted on an Amray 180D instrument using gold-sputtering technique. The microscope was operated at 15 kV. The test specimens were attached to an aluminum mount. To avoid electron charging effects, the specimens were coated at the bottom with Ladd Silver conducting paint before being sputtered with gold in a Denton vacuum DV 515 evaporator. About 50 photomicrographs were collected from the different samples at two magnifications (\times 500 and \times 2000).

4. Determination of Fiber Dispersion

Fiber dispersion quality was evaluated using thin composite sections and image analysis. Sections $(40-\mu m \text{ thickness})$ obtained by carefully microtoming cross sections of dog bones (ca. 2×8 mm) were mounted between microscope slides and examined by light microscopy using a Zeiss microscope equipped with a metamorph image analysis system. Approximately 60-70 images were analyzed for each specimen, and the quality of fiber dispersion of the images was quantified by measuring their standard deviation of gray level according to Scott.²⁵ In brief, this test involves placing composite sections of constant thickness in the light path of an imaging microscope. Transmitted light is recorded and converted into an electronic image consisting of 512×512 pixels or picture elements. Each pixel is analyzed with regard to its location and a digitized gray level that describes its shade in 256 steps between black (0)and white (255). Fiber dispersion is quantified by determining both the average and the standard deviation of the gray level. The later provides an objective quantitative assessment of dispersion for sections having the same fiber content. Composite samples having 10 and 20% fiber content could be analyzed by this method. Test sections from composites having higher fiber contents (30 and 40%) were too dark (with average gray levels that were numerically too low) for analysis.

5. Strength Determination

Compression-molded composite test specimens were cut from molded sheets $(100 \times 100 \times 2.5 \text{ mm})$ using a die punch cutter with a gauge length of 7.5 mm and a width and thickness of approximately 2.75 and 2.20 mm, respectively. All test specimens were kept in a desiccator prior to testing to avoid moisture absorption. The tests were conducted at room temperature $(23 \pm 2^{\circ}\text{C})$ with a strain rate of 1.25 mm/min. From the stressstrain curves the modulus of elasticity (E), the maximum tensile stress (δ) , and the elongation at break (ε) were determined. At least five specimens were tested for each set of samples and the mean values as well as standard deviations were calculated.

RESULTS AND DISCUSSION

1. Thermal Stability of Fibers

The determination of thermal stability by a static and dynamic assay involving TGA (Fig. 1) provides information on the weight loss of fiber samples on a temperature gradient (dynamic experiment) or on a time scale under isothermal (200°C) conditions. The results of the dynamic weight loss experiment reveal that all fibers experience their most rapid thermal decomposition between around 300 to 400°C, and that the onset of decomposition varies between wood (i.e., control) and the three steam-exploded fiber samples (Table I). Using a 2% weight loss threshold in a dynamic experiment (Fig. 1), the only sample less thermally stable than wood is untreated, unwashed, steam-exploded yellow poplar fiber (SEF) (Table I). Although WEF matches the thermal stability of wood, AEF and AAEF both surpass the thermal stability of untreated wood (Table I). When examining the dynamic thermal stability of wood in comparison to isolated wood components, cellulose, lignin, and xylan (Table I), the data reveal



Figure 1 Typical thermogravimetric curves of cellulose fiber samples on a temperature gradient (dynamic test) and on a time scale (isothermal test). Note: a 2 wt % loss threshold following the loss of moisture was selected to compare the thermal stability of fibers.

that chemical cellulose is significantly more stable than wood, but not as stable as acylated cellulose (Table I) and that both lignin and xylan are significantly less stable than untreated wood. It is apparent that the hemicellulose component (i.e., xylan) represents the thermally most unstable wood component. Removal of both xylan and lignin boosts fiber stability during melt processing and reduces the formation of volatile degradation products.

These results from the dynamic testing of thermal stability parameters are in general agreement with the results obtained in static tests (Table I). Comparing weight loss figures of different fiber types in a 2-h isothermal (200°C) nitrogen atmosphere reveals that AEF is somewhat more stable than untreated wood, WEF, and acetylated fiber (AAEF), which are virtually identical, and that unwashed SEF is significantly more unstable (Table I).

Comparing the weight loss figures of untreated wood and wood components in the static experiment reveals the significantly greater thermal instability of xylan compared to sawdust, and this is followed by lignin (Table I). Cellulose is more stable than untreated wood in the static experiment.

In summary, the thermal analysis tests of various steam-exploded wood fibers, wood components, and control (untreated wood) reveal that water-extracted, steam-exploded wood fibers are at least as stable as untreated wood under two assay procedures and that alkali-extracted and acetylated fibers produce fewer volatile degrada-

Fiber/Wood Component	Static Test Weight Loss at 200°C in 2 h (wt %)	Dynamic Test (2 wt % Loss Threshold)	
		Temperature at 2 wt % Loss (°C)	Time at > 200 °C for 2 wt % Loss (min)
Yellow poplar	1.2	250	23
COF	0.6	245	23
SEF	7.1	190	8
WEF	1.4	250	28
AEF	0.4	270	36
AAEF	1.2	280	42
Cellulose	0.4	280	44
Xylan	13.6	190	4
Lignin	4.3	200	18
CAB	4.1	215	19

Table I Thermal Stability of Fibers and Wood Components^a

^a See Experimental section.



Figure 2 Mixing torque curves for CAB/AAEF composites at different fiber content processed at 204°C for 20 min.

tion products under conditions resembling melt processing. This superiority becomes even more important when considering that modulus generally correlates with cellulose content and that acetylation produces a fiber surface with significantly reduced surface free energy (i.e., improved compatibility with the matrix melt). The removal of both hemicelluloses and lignin contributes markedly to the thermal stability of fibers.

2. Fiber Pretreatment Effects on Melt Processing

When different cellulose fibers from steam explosion and fractionation and untreated biomass filler [commercially available oat filler (COF)] are added to the melt of the cellulose ester matrix while it is undergoing melt processing in a twin screw kneader, an increase in torque is recorded (Fig. 2). The initial torque spike experienced upon fiber addition, approximately 3 min after fibers are added, subsides as fiber dispersion improves (Fig. 2). It is apparent that uniform flow conditions are reached approximately 5-10 min after fiber addition has commenced (Fig. 2). The data reveal that torque varies with time and fiber content and, in addition, with temperature and fiber type. Whereas fiber dimension will also influence torque during melt processing, differences observed for steam-exploded fibers from the same source, but different in postproduction treatment (extraction with water, alkali, and acetylation), must be attributed to surface characteristics. When examining the impact of fiber content on torque changes (compared to control) (Fig. 3), distinct differences are revealed for different fiber types. Whereas WEF fibers are responsible for a torque increase of about 200%, AAEF fibers raise

torque by 421%. COF and AEF fibers raise torque by 309 and 259%, respectively. Considering that dimensional factors can be ruled out in case of the fibers from steam explosion (but not COF), surface differences must explain the observed results. Torque can be considered to be related to fiber-matrix interaction and adhesion. A fiber with a highly compatible matrix surface produces a large interfacial surface area in a mixture with a fluid matrix, and, as a consequence, this mixture resists deformation and contributes to an increase in torque during melt processing. Although the torque difference between AEF and WEF must be attributed to the presence of lignin, the acetylated fiber substrate clearly displays the greatest impact on torque behavior of all fibers used (Fig. 3). This observation is consistent with earlier observations on the free energy of cellulosic surfaces in relation to OH derivatization.²⁶

In summary, significant differences in the effect of fiber addition on melt processing properties (i.e., torque behavior) are revealed, and these vary with both fiber content and type. Fiber acetylation contributes to the most dramatic increase in torque, especially at the 40% fiber content level, and this is attributed to better fiber-matrix interaction and adhesion by the fluid matrix. Lesser torque increases are seen for AEF and WEF. COF, having substantially different particle geometry (more filler than fiberlike), also displays a highly positive effect on torque (Fig. 3); however, this is attributed to the increased surface area caused by fiber/particle dimensions.



Figure 3 Change in torque (compared to pure CAB) versus fiber content for various cellulose fiber/CAB composites processed at 204°C for 20 min.



Figure 4 Scanning electron micrographs of tensile fracture surfaces of 20% cellulose fiber/CAB composites at a magnification of \times 500. A: CAB/WEF, B: CAB/AEF, C: CAB/AAEF, and D: CAB/COF.

3. Fiber Pretreatment Effects on Composite Properties

a. Scanning Electron Microscopy

Freeze-fracture surfaces of all composites were studied by SEM. A comparison between WEF, AEF, AAEF, and COF at $\times 500$ and $\times 2000$ magnification, respectively, reveals significant differences in how fibers are imbedded in the matrix (Figs. 4 and 5). Whereas WEF, AEF, and COF indicate substantial interfacial delamination between fiber surface and matrix, with $1-2 \mu m$ gaps developing around most fibers, AAEF exhibits a substantially greater amount of interfacial adhesion by the matrix to the fiber surface (Fig. 5). Micrographs taken at $\times 2000$ magnification show distinct signs of pullout of undamaged fibers for WEF, AEF, and COF, and, in contrast, substantial fiber fracture with cell wall fibrillation for the case of the AAEF specimens (Fig. 5). The adhesion-promoting effect of fiber acetylation becomes

even more apparent when micrographs are compared at all fiber volume fractions (not shown). Whereas interfacial delamination is apparent in all AEF-based composites, fiber failure with extensive fibrillation is detected for all AAEF-based composite samples.

The results are consistent with the presence or absence of interfacial adhesion, interfacial delamination, fiber pullout as opposed to fiber fragmentation and failure by fibrillation. Whereas no significant difference is detected between WEF, AEF, and COF, fiber acetylation (i.e., AAEF) clearly produces fibers that exhibit superior adhesion to the matrix.

b. Fiber Dispersion

A method for quantitatively determining the quality of fiber dispersion in composites by image analysis has recently been introduced by Scott.²⁵ Using light microscopy in conjunction with image



Figure 5 Scanning electron micrographs of tensile fracture surfaces of 20% cellulose fiber/CAB composites at a magnification of \times 2000. A: CAB/WEF, B: CAB/AEF, C: CAB/AAEF, and D: CAB/COF.

analysis of thin (microtomed) sections, transmitted light is sensed by optical imaging. By detecting dark versus light regions of the image, a gray level is defined around which a standard deviation of gray is computed. A low standard deviation of gray level thereby defines uniform dispersion, and this becomes less uniform as standard deviation of gray level values increase. This is illustrated in Figure 6 where the optical images of two composite sections are given with the corresponding gray level images. Because of the difficulties of distinguishing between light and dark contrasts in the images of high-fiber volume fraction composites (i.e., >20%), and compounded by the difficulties associated with the microtoming of highly brittle, high-fiber content composites, image analysis was limited to composites having 20% or less fiber volume fraction.

The relationship between the standard deviation of gray level for the four composites with 20% fiber volume fraction, representing WEF, AEF, AAEF, and COF fibers, and torque differences compared to neat CAB matrix, reveals a significant improvement in fiber dispersion with surface acetylation (Fig. 7). Uniformity of dispersion (at 20% fiber content) increases from COF (least uniform) to WEF, AEF, and AAEF (most uniform). Uniformity of fiber dispersion is viewed as a defining quality criterion of composites with discontinuous fibers, and this has previously been found to be correlated with melt viscosity.²⁵

c. Mechanical Properties

The mechanical properties in tension that were obtained with dog bone test specimens produced by die cutting are illustrated in Figure 8(a-c). Data for maximum stress, modulus, and elongation at break are related to fiber content. Maximum stress [Fig. 8(a)] reveals significant strength losses for all fiber composites at low-fiber volume fraction (<10%). This is consistent with the be-



Figure 6 Images of (a) good and (b) poor fiber dispersion. (c) and (d) Variations of gray level in the plane from X to Y for images (a) and (b), respectively.

havior predicted by the rule of mixtures (Fig. 9). The rule of mixtures model assumes unidirectional composite architecture with continuous fibers and perfect interfacial stress transfer which can be compensated by applying length and orientation correction factors. However, whereas



Figure 7 Standard deviation of gray level (representing fiber dispersion) versus mixing torque for cellulosebased composites at 20% fiber content by weight. Composites with higher fiber weight fractions were too dark for scanning by image analysis.

WEF-, AEF-, and COF-based fiber composites experience a continued strength decline with fiber content rising beyond the critical fiber content (i.e., 10-40%), the AAEF-based composites exhibit a distinct strength recovery, up to 110% of the matrix, as fiber content rises to 40% (Fig. 8). This strength recovery is consistent with the rule of mixtures model (Fig. 9). Using a product of length correction factor, α_1 , and orientation efficiency factor, α_2 , of 0.5, predicted and observed behavior reveal that actual composite data parallel those of the model (Fig. 10). 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, whereas the model predicts approximately 17%. Maximum strength at 40% fiber content is predicted to be 55 as opposed to 34 MPa for the actual composites (Fig. 10). 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 (Fig. 11). 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 critical fiber



Figure 8 Effect of fiber content on (a) stress, (b) modulus, and (c) elongation at break of CAB reinforced with various cellulose fibers. (\diamondsuit) WEF, (\Box) AEF, (\triangle) AAEF, and (\bigcirc) COF.

concentration may be 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.^{11,18} investigated the effect of fiber orientation on tensile properties of sisal fiber-reinforced polystyrene composites and found a similar variation in maximum tensile stress with fiber content. The authors achieved a modest increase in strength with fiber content 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 (Fig. 8). Stiffness (or modulus) was found to increase consistently with fiber content (Fig. 8). 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. 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.²⁷⁻²⁹ 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 found not to con-



Figure 9 Variation of tensile stress with fiber fraction as predicted using the rule of mixtures. The model is based on the maximum tensile strength, 30 and 250 MPa for matrix and fiber, maximum elongation at break, 60 and 0.86%, and tensile modulus, 640 and 1000 MPa, respectively. tribute noticeably to modulus regardless of fiber content.

The significantly positive effect of AAEF fibers on the tensile strength of CAB composites must be attributed to superior interfacial adhesion. Using the rule of mixtures model (Fig. 10) that relates tensile strength to fiber fraction (with the assumptions stated above) of the AAEF composite, agreement is achieved between the model and the experimental data in several points: Critical fiber fraction is at around 10%, and stress at break increases with fiber fraction both increasing and decreasing from $V_{\rm crit}.$ Although the overall strength gains by fiber addition to matrix are modest (10%), the results reveal that AAEF fibers are indeed qualified to provide true reinforcement for the CAB matrix. Combined with a 100% increase in modulus [Fig. 8(c)], this result supports the validity of fiber surface modification for cellulose fiber-reinforced composites.

d. Fiber Damage

Composite properties are greatly dependent on fiber length and aspect ratio (l/d). Aspect ratio thereby is a critical parameter determining reinforcement potential, which depends on fiber strength (σ_f) and interfacial stress (τ) . Critical fiber length, l_c , is then defined as

$$l_c / d = \sigma_f / 2\tau \tag{1}$$

where d represents diameter. It is often assumed that discontinuous fiber composites behave like continuous fiber composites if l approaches 20 $\times l_c$.

The fibers used in this study are shown in Figure 11(A-D). It is apparent that the steamexploded fiber fraction consists of a mixture of very long and short fibers, and that this is contrasted by the commercial, hammermilled product (i.e., COF). The latter is considerably shorter in length and more uniform. The fibers isolated from the ultimate composites (by matrix dissolution in solvent) [Fig. 11(E-H)] reveal that all steam explosion-derived fibers were damaged by fiber breakup into shorter fiber segments. The results reveal that fibers consist of sections approximately equal in length to those of COF, and that they approach an average aspect ratio of 25-50. The reduced cellulose fiber length following compounding in the twin screw extruder and compression molding apparently produces a fiber that exceeds the critical



Figure 10 Comparison of tensile strength properties of the 40% AAEF/CAB composite with the rule of mixtures model assuming the product of α_1 and α_2 equals 0.5.

fiber length (l_c) only in the case of AAEF by virtue of superior interfacial stress transfer property. It is only at $l > l_c$ that fibers can provide true reinforcement for composites.

CONCLUSIONS

- 1. The effective removal of hemicelluloses and lignin from wood fibers by steam explosion/fractionation contributed to improved thermal stability of cellulose-based fibers. Improvements in thermal stability associated with the stepwise removal of hemicelluloses and lignin following steam explosion, and of acetylation, are clearly indicated by two assays involving thermogravimetric analysis.
- 2. Fiber preparation by steam explosion, water washing, alkali extracting, (surface) acetylating, or alternatively, hammermilling produces fillers/fibers that influence composite processing as well as product parameters.
- 3. Shear viscosity of thermoplastic melts (as indicated by torque) increases with filler/ fiber content, and this increase varies with interfacial adhesion. It was found to be greatest for AAEF.



200 µ

Figure 11 Optical micrographs of cellulose fibers before compounding (A, B, C, D) and recovered from compression molded composites (after compounding with CAB) by matrix dissolution in acetone (E, F, G, H). Note the similarities between all steam exploded fibers (i.e., A–C) and the different fiber geometry of COF (i.e., D) before compounding, and the relative uniformity in fiber dimension after compounding in all cellulose fibers (i.e., E–H).

- 4. Mechanical composite properties varied for different fiber types. Whereas modulus remained unchanged or doubled depending on filler/fiber type, elongation declined consistently, and maximum strength decreased for all fibers except AAEF, which revealed strength gains beyond the critical volume fraction of 10%.
- 5. The superior reinforcing characteristics of AAEF fibers were found to be supported by SEM, which revealed that all fibers except AAEF exhibit substantial interfacial delamination and fiber pullout. AAEF-based composites revealed matrix-fiber adhesion and fiber failure by fibrillation.
- 6. Uniformity of fiber dispersion varied with fiber type and was found to be directly related to melt viscosity as determined by torque.

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