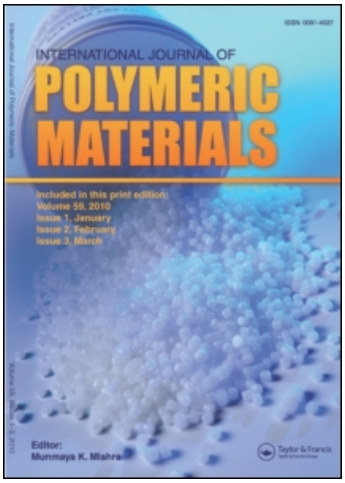

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Influence of fiber type, fiber mat orientation, and process time on the properties of a wood fiber/polymer composite

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INFLUENCE OF FIBER TYPE, FIBER MAT ORIENTATION, AND PROCESS TIME ON THE PROPERTIES OF A WOOD FIBER/POLYMER COMPOSITE

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A rapid press consolidation technique was used to produce composites from two types of air-laid wood fiber mat, incorporating either mechanically refined or bleached chemi-thermomechanically refined Norway Spruce [Picea abies (L.) Karst] and a bicomponent polymer fiber. The manufacturing technique involved pre-compression, contact heating to the process temperature under vacuum and then rapid transfer to the press for consolidation and cooling. Composites were tested to determine response to water or water vapor, porosity, fiber volume fraction and tensile properties. The composites absorbed water rapidly and showed changes in thickness with fluctuations in relative humidity. Porosity was higher in composites containing mechanically refined (MDF) fibers than in composites containing bleached chemi-thermomechanically refined (CTMP) fibers. Tensile test results suggested that fiber wetting by the polymer matrix had been maximized within a five-minute heating time. Results also indicated that porosity was not the key determinant of tensile properties in the composites.

Keywords: Norway spruce, bicomponent polymer fibers, mat forming, composites

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INTRODUCTION

As discussed in a recent review article [1], there is increasing worldwide interest in the use of natural plant fibers as alternatives to synthetics (*e.g.*, glass fiber, carbon fiber) for the reinforcement of polymers. The use of plant fibers can potentially provide a number of advantages including lower costs of manufacturing, less wear on processing equipment and environmental benefits such as easier recycling or disposal at the end of a product's useful life. Life cycle analyses [2, 3] have also indicated advantages in terms of energy use during manufacture when compared with synthetic fiber-reinforced materials. Internationally, commercial interest is being driven by the possible use of plant fiber-reinforced materials in both niche and commodity applications. For example, in Europe the automotive industry is using plant fiber-reinforced polyolefins for interior trim parts [4] and some companies are experimenting with more demanding exterior applications in automobiles. By contrast, building product applications have dominated in North America; however, the automotive industry there is also now starting to use these materials. Strong growth in demand is forecast in these markets as well as product applications in other industrial sectors [5].

Fiber-reinforced polymer composites can be manufactured by a number of processes. For example, a granulate containing a mixture of fibers and thermoplastic polymer can be produced by extrusion or thermo-kinetic mixing and this granulate can then be injection-molded into a final component. Much research has been focused on this type of process and the improvement in mechanical properties of injection-molded test bars when compared with those of pure polymer [6–9]. Alternatively, intermediate products can be created by means of fiber mat forming and then converted into composites through compression molding [10]. Plant fiber mats are commercially available in a number of countries and are already used for a variety of applications (*e.g.*, absorbents, growth media, geotextiles, intermediate products).

A key issue in the use of plant fibers to reinforce polymers, and one that has been much studied, involves methods to improve the compatibility of hydrophilic fibers with hydrophobic thermoplastics (*e.g.*, polypropylene) in the compounding process [11, 12]. The addition of a compatibilizer enhances the wetting of fibers by the polymer and should ensure good stress transfer between the plant fibers and the polymer matrix. The addition of a compatibilizer is also thought to improve the dispersion of plant fibers in the matrix (*e.g.*, during extrusion or injection molding). In the case of polypropylene composites, maleated polypropylene (MAPP) has been extensively studied as a compatibilizer [13–15]. The maleic anhydride moiety within MAPP may react with hydroxyl groups on the fibers during compounding, thereby attaching a polypropylene chain that will make the

fiber surface more compatible with the polymer matrix [16]; however, there is evidence that the occurrence of such reactions in the case of wood may depend on fiber type. Maleated polyethylene (MAPE) has also been investigated as an additive in polyethylene composites [17].

The purpose of the research described in this paper was to examine the impact of two different wood fiber types and other process variables on the properties of a wood fiber/polymer composite as a precursor to further research involving the use of fiber/polymer compatibilizing agents. New knowledge from such research can be valuable in optimizing plant fiber/polymer composite manufacturing processes. In the research, a commercially available bicomponent polymer fiber with a polypropylene core and a lower melting polyethylene outer layer was used in combination with wood fibers to prepare fiber mat intermediates. These intermediates were then converted into composites for testing and evaluation.

MATERIALS AND METHODS

Raw Materials

Three types of wood fiber were used to produce composites from mat-formed intermediate products: (1) bleached chemi-thermomechanically refined Norway spruce [Picea abies (L.) Karst] fibers (CTMP) obtained in commercial fluff grade (brightness = 70) from Waggeryd Cell AB in Vaggeryd, Sweden and then hammer-milled for use in mat forming, (2) Norway spruce fibers that had been thermomechanically refined under conditions suited to medium-density fiberboard (MDF) production and obtained from the Sunds Defibrator Inc. (now Valmet) pilot plant at Sundsvall in Sweden, and (3) a bicomponent polyethylene/polypropylene polymer fiber obtained from FiberVisions A/S in Varde, Denmark. The latter was designated AL-Special-C PHIL 65/35 with properties including 6 mm fiber length, 1.7 dtex (1.7 gms per 10,000 metres of fiber) and a concentric construction with a polyethylene sheath and a polypropylene core. The sheath/core volume ratio was 65 : 35 with a sheath softening point of 124°C and a sheath melting point of 130°C. This fiber is described commercially as a high strength/low bulk fiber for non-woven applications.

Mat Forming

The raw materials were used to prepare three types of mat on a Dan-Web air-laid mat-forming system (Fig. 1) at the Plant Fibre Laboratory of Denmark's Royal Veterinary and Agricultural University. Chosen mat compositions were 50 w/w% spruce CTMP fibers and 50 w/w% bicomponent polymer, 50 w/w% spruce MDF fibers and 50 w/w% bicomponent polymer, and 100%

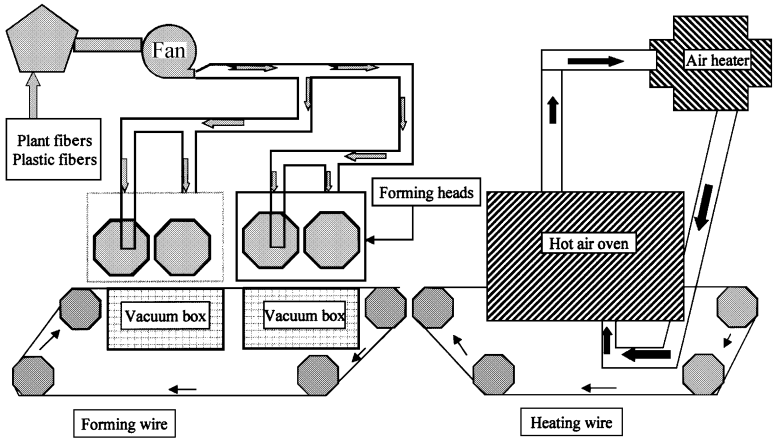

FIGURE 1 Schematic diagram of the Dan-Web air-laid mat forming machine at the Plant Fibre Laboratory of Denmark's Royal Veterinary and Agricultural University.

bicomponent polymer. The fibers or fiber mixtures were fed into the forming head at a rate of 750 – 1000 gms/min. The mat speed through the equipment was 4 – 5 metres/min. and fiber mats were formed by passing through a hot-air oven at 137°C. Dwell time of the mat material in the oven was between 30 seconds and one minute. The mats were 60 cm wide and 15 to 20 metres in length. A length of approximately two metres at the end of each mat was discarded due to poor forming. Sections measuring 200 mm × 300 mm were cut from the middle of each mat for further processing. Mat sections were then weighed to obtain an average mat basis weight.

Composite Production

Following production of mats on the Dan-Web system, samples were taken to Risø National Laboratory in Roskilde, Denmark for conversion into trial composites. This work was undertaken on a press that had originally been designed for research on synthetic fiber/polymer composites. The equipment has been described in detail in an earlier paper [18]. The press consolidation process used in this equipment, shown as an example in Figure 2 for a three-dimensional molded component, consisted of the following steps: (1) pre-compression, (2) contact heating of the mat under vacuum, (3) rapid transfer to a press for consolidating and cooling, and (4) removal of the finished part from the press.

Stacks of mats of each type were prepared to a total weight in the range of 120 – 125 gms to ensure each final composite panel had approximately the same weight. Each stack was placed between two Teflon sheets in a metal

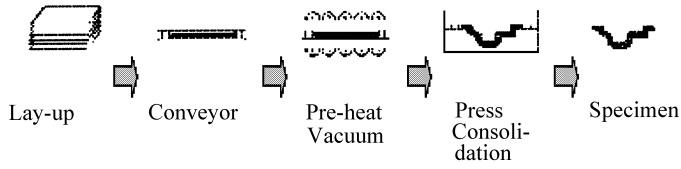

FIGURE 2 Schematic diagram of the pressing system at Risø National Laboratory.

frame, pre-pressed for 30 seconds at 3.3 MPa and then rapidly conveyed (maximum conveyor speed of 1.5 metres/second) into the vacuum pre-heating stage. In this stage, the compressed mats were pressed for one, five or 10 minutes at a temperature of 190°C, a platen pressure of 3.3 MPa and a minimum vacuum of two mbar. The vacuum was included to assist in the mat consolidation process and to minimize porosity in the consolidated composite. The removal of air should also help to limit the thermal degradation of wood fibers at elevated temperatures. Finally, the frame containing the mat sandwiched between Teflon sheets was conveyed into the press where the composite was consolidated at a platen temperature of 30°C and a pressure of 3.3 MPa for one minute. Each composite panel was then removed from the press and weighed when cool. This process worked well in the case of the MDF/polymer and CTMP/polymer matrices but problems arose in creating a pure bicomponent polymer panel as a result of adhesion of the melting polymer mix to the Teflon sheets. These problems were solved by employing a more rigid Teflon sheet material to make these panels. Once panels of a satisfactory appearance had been made, two of each type made at each press time were prepared and cut up for testing and evaluation as shown in Figure 3. This cutting pattern was designed to allow the variation in tensile properties with mat direction (i.e., warp *vs.* weft) to be evaluated. In the case of the pure polymer panels, no variation in properties with pressing time was anticipated and therefore production of samples for evaluation was limited to two panels prepared using a five-minute heating time.

Composite Testing

The composites were tested for weight and dimension changes on water soaking, weight and dimension changes in response to cyclic changes in relative humidity, porosity, fiber volume fraction and tensile properties.

The procedure adopted for testing water absorption was based on ASTM Standard D 570-98 [19] using specimens measuring 20 mm in width, 90 mm in length and two mm in thickness and cut from composites manufactured with a 10-minute heating time. Samples were tested in sets of five. Initial thickness was measured at three locations, width at three locations and length at two locations using a Mitutoyo Digimatic caliper. The samples

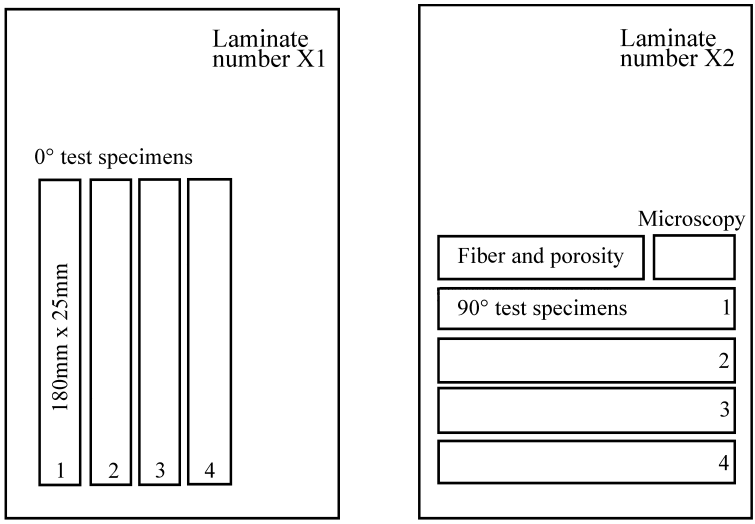

FIGURE 3 Cutting pattern for tensile tests, fiber volume fraction, and porosity tests and later microscopic examination of two samples from each composite type.

were weighed and fully immersed in water at $23+/-1°C$. Sample weights and dimensions were then measured on a weekly basis.

Similar specimens to those used in the water absorption tests were also used to evaluate response of the composites to cyclic changes in relative humidity. The samples were weighed, the dimensions measured and the samples then placed in a Sanyo Gallenkamp environmental chamber. The temperature in the chamber was set at 20°C and relative humidity was cycled between 65% and 95% on a weekly basis. Sample weights and dimensions were measured weekly.

The use of plant materials in mats allows for non-straight fibers, low stiffness fibers and branched fibers. A result of the limited compaction ability of such mats is that high fiber contents can lead to high porosity. A porosity of zero is ideal in terms of fiber/polymer interaction and the overall structural integrity of the composite. The content of fiber, polymer matrix and porosity in the composite was therefore determined by gravimetric measurements using a slightly modified version of ASTM Standard D 3171-76 [20]. In this procedure, composite samples were weighed, the polymer matrix dissolved in hot xylene and the residual wood fibers then weighed. With assumed densities for the wood fibers (1.5 gms/cc) and for the bicomponent polymer (0.9 gms/cc), it was possible to calculate the weight and volume fractions of fiber and polymer matrix as well as the porosity of the composite samples.

Four tensile test specimens were cut from two samples of each composite type as shown schematically in Figure 3. Tensile testing of dogbone-shaped specimens, measuring 180 mm in length, 25 mm in maximum width and 15 mm in the narrowest section, was performed on a mechanical Instron testing machine with a 5 kN load cell. The machine was operated in displacement control at a speed of 2 mm/min. Longitudinal strain of each specimen was recorded with two back-to-back extensometers. Readings of load and strain were sampled at 4 Hz with a PC-based data acquisition system (Labtech Notebook). The average strain was used and the stress was calculated as load divided by initial cross-section. Results were expressed as plots of stress (MPa) against strain (%). The ultimate tensile strength or tensile strength at yield and the tensile stiffness, as determined by the slope of the tangent to the stress–strain curve drawn through the origin, were established from the stress–strain plots.

RESULTS AND DISCUSSION

Mat Weights

Mat basis weights were 244 gms/m^2, 278 gms/m^2 and 200 gms/m^2 for CTMP, MDF and bicomponent polymer mat samples respectively after storage for four weeks under ambient laboratory conditions. Oven-dry basis weights were 218 gms/m^2, 263 gms/m^2 and 200 gms/m^2 respectively. Differences in weight between each stack of mats and each consolidated composite were in the range 3.3 to 4.1% for the composites containing MDF or CTMP fibers. This decrease in weight after pressing is largely attributed to moisture loss from the wood fibers during the pressing process.

Water Absorption and Response to Cyclic Humidity Changes

Results of the water absorption and cyclic humidity tests, expressed in terms of changes in mean sample thickness, are shown in Figures 4 and 5 respectively. As shown in Figure 4, water absorption occurred relatively rapidly in samples containing wood fibers and reached near-maximum values within one to two weeks. A plot of sample weight increases against time appeared very similar to that for thickness increases and reflects findings by others when examining properties of plant fiber-reinforced polypropylene composites [21]. Thickness variations in response to changes in relative humidity (Fig. 5) are also in line with expectations with the exception of the measurements at week three. The anomalous result in week three was later found to be due to a temporary malfunction of the climate chamber during that week. In both tests, composite samples containing CTMP fibers picked up more water than composite samples containing

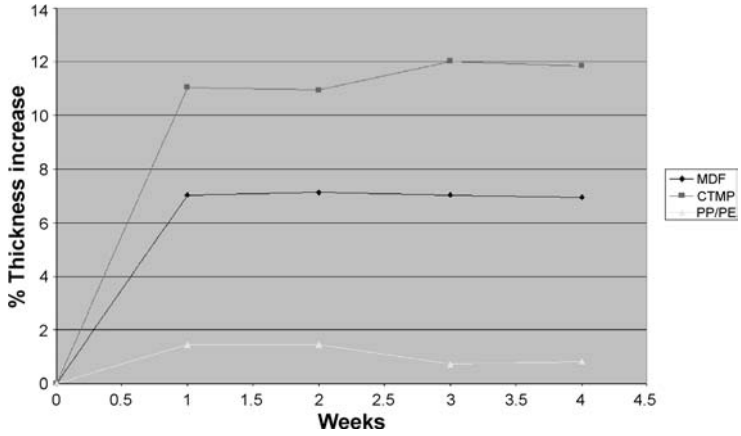

FIGURE 4 Mean increase in composite sample thickness as a function of soaking time in water.

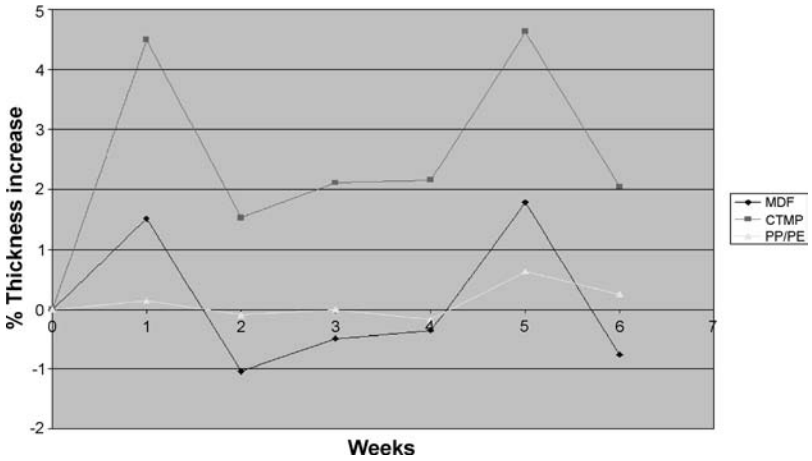

FIGURE 5 Mean increase in composite sample thickness on exposure to weekly changes in relative humidity between 65% and 95%.

MDF fibers. Since lignin has relatively few hydroxyl groups when compared with cellulose or hemicelluloses, the high lignin concentration on the surface of MDF fibers would produce a slightly less hydrophilic character than the relatively cellulose-rich surface of the CTMP fibers and might partially explain these results.

Porosity and Fiber Volume Fraction

Results of porosity testing are shown in Table 1 and Figure 6. As shown, longer pre-heating times led to greater composite consolidation and therefore lower porosity. Mean porosity values were consistently lower for the CTMP composites than for the MDF composites and this might be expected considering that the CTMP fibers had been chemically treated,

TABLE 1 Density, fiber weight fraction, fiber volume fraction, and porosity for composites containing MDF or CTMP fibers at one-, five-, or 10-minute pre-heating times

Material	*Pre-heat time* [*min*]	*Composite density* [g/cm^3]	*Fiber content* [*weight-%*]	*Fiber content* [*volume-%*]	*Porosity* [*volume-%*]
MDF	1	0.97	47.4	30.8	12.4
	5	1.03	47.8	32.9	7.2
	10	1.05	43.1	30.1	3.8
CTMP	1	1.01	45.6	30.8	7.9
	5	1.07	47.1	33.6	3.6
	10	1.07	42.8	30.6	1.0

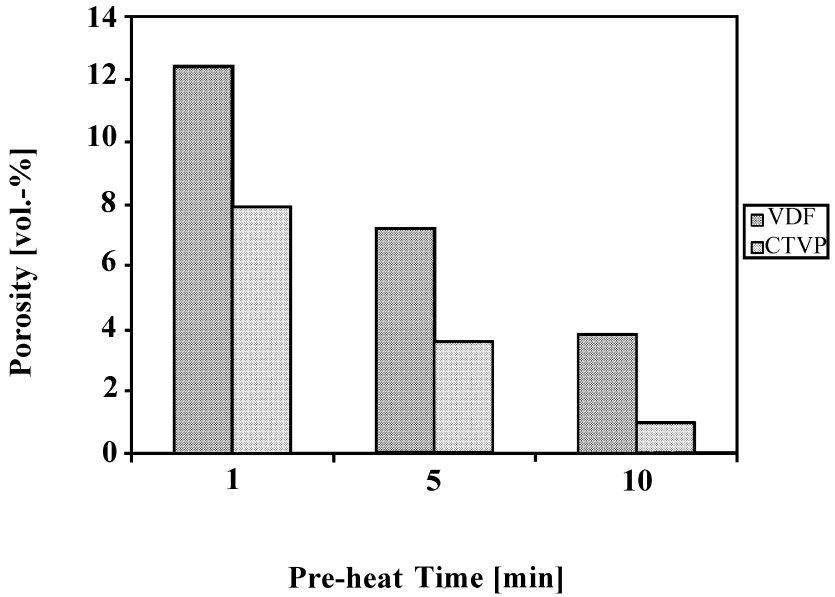

FIGURE 6 Porosity as a function of pre-heat time for composites containing MDF or CTMP fibers.

refined and hammer-milled while the MDF fibers had only been subjected to mechanical refining. As a result, it would be reasonable to conclude that CTMP fibers should be more flexible and compressible and produce composites with lower porosity values than MDF composites under the same conditions. Fiber weight fractions (Tab. 1) were found to be slightly below the target value of 50% and this was attributed to moisture loss and loss of volatiles from wood fibers during the pressing stage.

Tensile Properties

Results of tensile testing are shown in Figures 7 and 8. Data from tensile testing were statistically analyzed using a pairwise *t*-test approach to establish the significance of the influence of each variable (*i.e.*, process time, fiber mat orientation and fiber type) on composite strength and stiffness at the 99% probability level ($p = 0.01$).

The influence of fiber type on composite tensile strength was significant with MDF fibers providing better tensile strength than CTMP fibers in all cases; however, differences in composite stiffness between the two fiber types were only significant when composites were prepared with a 90° fiber mat orientation at a heating time of one minute.

The influence of fiber mat orientation only had a significant effect on composite tensile strength for materials based on MDF fibers heated for five or 10 minutes. These results may reflect a tendency for fibers to align slightly more in the mat direction in these two situations. The influence of fiber mat orientation on tensile stiffness was not significant in any material/process variable combination.

Visual inspection of the tensile test results for composites prepared using five- or 10-minute heating times strongly suggested a pooling for comparison with the one-minute results. When this was done, the influence of process time on tensile strength and stiffness was found to be significant at the 99% probability level in all cases with the exception of CTMP composite stiffness properties based on 90° fiber mat orientation. Since porosity values (Fig. 6) clearly varied significantly between composites prepared using five- or 10-minute process times, the results indicate that porosity is not the controlling factor in determining tensile properties. This is in contrast to previous findings with jute/polypropylene and synthetic composites [22].

Both tensile strength and stiffness were clearly improved in the fiber/polymer composites in relation to those properties for unreinforced bi-component polymer matrix. Literature values for polypropylene indicate tensile strengths and tensile moduli in the ranges 30 – 40 MPa and 1.0 – 2.0 MPa respectively with corresponding values of 7.0 – 8.0 MPa and 0.1 – 1.4 GPa respectively for polyethylene, depending upon polymer density [23].

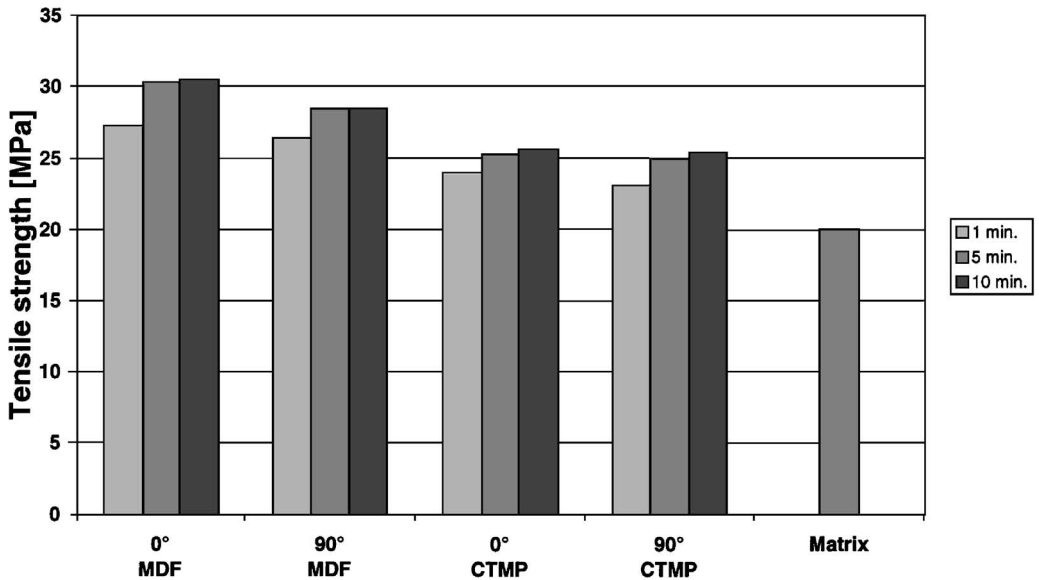

FIGURE 7 Tensile strength of composites prepared from bicomponent polymer, bicomponent polymer plus MDF fibers, or bicomponent polymer plus CTMP fibers (0° indicates samples cut in the direction of the mat forming line, 90° indicates samples cut perpendicular to the direction of mat forming) at one-, five-, and 10-minute pre-heating times. Matrix indicates the sample prepared using the bicomponent polymer alone.

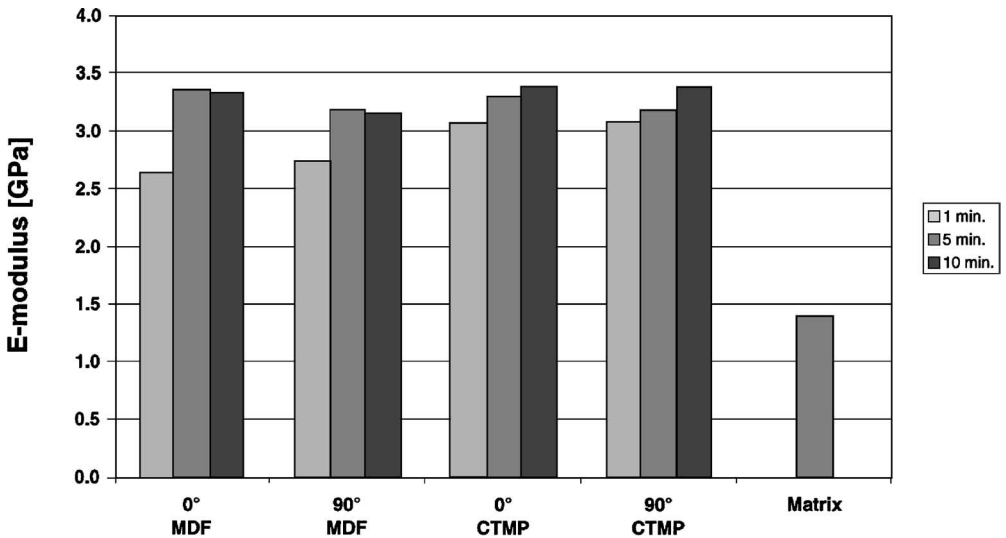

FIGURE 8 Tensile modulus of composites prepared from bicomponent polymer, bicomponent polymer plus MDF fibers, or bicomponent polymer plus CTMP fibers (0° indicates samples cut in the direction of the mat forming line and 90° indicates samples cut perpendicular to the direction of the mat forming line) at one-, five-, and 10-minute pre-heating times. Matrix indicates the bicomponent polymer alone.

CONCLUSIONS

A study was performed to determine the influence of spruce fiber type, fiber mat orientation and process time on the properties of a wood fiber/polymer composite. Composites were prepared from intermediate air-laid fiber mats incorporating bicomponent polyethylene/polypropylene fibers as a binder.

The major conclusions from this study were:

(1) The method of fiber production can significantly influence the properties of fiber/polymer composites. For example, in this study, CTMP fibers provided composites that were more susceptible to water absorption but had lower porosity than composites prepared using MDF fibers. MDF fibers consistently gave composites with higher tensile strength while this property was clearly not controlled by composite porosity.
(2) Results of tensile strength testing as a function of heating time suggested that fiber wetting had been maximized within five minutes.
(3) Tensile strength values indicated that, depending on fiber type, there may be a tendency for fibers to align slightly more in the mat direction than across the mat direction in some manufacturing situations. This is an important consideration to bear in mind when preparing test cutting patterns for composites based on use of fiber mats.

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