

## Influence of Temperature on Mechanical Properties of Jute/Biopolymer Composites

Alexandra Løvdal, Louise Løcke Laursen, Tom Løgstrup Andersen, Bo Madsen, Lars Pilgaard Mikkelsen

Department of Wind Energy, Section of Composites and Materials Mechanics, Technical University of Denmark, Risø Campus, Frederiksborgvej 399, DK-4000 Roskilde, Denmark

Correspondence to: L. P. Mikkelsen (E-mail: lapm@dtu.dk)

**ABSTRACT:** Biopolymers and natural fibers are receiving wide attention for the potential to have good performance composites with low environmental impact. A current limitation of most biopolymers is however their change in mechanical properties at elevated temperatures. This study investigates the mechanical properties of two biomass-based polymers, polylactic acid (PLA) and cellulose acetate (CA), as a function of ambient temperature in the range from 5 to 80°C. Tests were done for neat polymers and for jute fiber/biopolymer composites. Micromechanical models were applied to back-calculate the reinforcement efficiency of the jute fibers. The elastic modulus of neat PLA is constant until a temperature of about 45°C, after which it is decreased rapidly. For neat CA, the elastic modulus is almost constant in the whole temperature range. The maximum stress of the neat biopolymers is consistently reduced. For the jute fiber composites, both the elastic modulus and maximum stress are reduced when the temperature is increased. For the elastic modulus, this is shown to be due to a reduction in the reinforcement efficiency of the jute fibers; i.e., a reduction in the back-calculated effective elastic modulus of the fibers. Altogether, the results demonstrate that the thermal sensitivity parameters typically provided for polymers, e.g., the glass transition temperature and the heat deflection temperature, cannot be used as sole parameters for determining the gradual change in mechanical properties of polymers and composites. © 2012 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* 000: 000–000, 2012

**KEYWORDS:** biopolymers and renewable polymers; composites; mechanical properties; thermal properties

Received 16 April 2012; accepted 24 July 2012; published online

**DOI:** 10.1002/app.38387

### INTRODUCTION

As an alternative to conventional composites made with petrochemically based polymer matrices, and nonbiodegradable fibers, composites of biomass-based polymers reinforced by natural fibers are receiving increasing scientific and industrial attention.<sup>1–5</sup>

The advantages of natural fibers compared to man-made fibers, such as glass fibers, are low density, biodegradability, and low environmental impact, while providing composites with good mechanical properties.<sup>6,7</sup> In addition, the reinforcement by natural fibers such as jute, flax, and banana are favorable as they are low cost materials.<sup>7,8</sup>

A wide range of biopolymers based on corn, starch, cellulose, or other biomass-resources are commercial available today: e.g., polylactic acid, cellulose acetate, and starch acetate. These biopolymers are currently mostly used in the packaging industry.<sup>4</sup> A limitation of biopolymers, typically observed for polylactic

acid, is the reduced resistance to deformation, i.e., a reduction of the elastic modulus, at moderate ambient temperatures around 50°C,<sup>8,9</sup> due to a glass transition temperature of the polymers in this temperature range. This limitation is not seen for conventional polymers like polypropylene and polyethylene. Reinforcement of biopolymers with natural fibers has previously been attempted to stabilize the mechanical properties of the materials at elevated temperatures.<sup>4,8–10</sup>

The thermal sensitivity of natural fiber/biopolymer composites in terms of mechanical properties has only been limited established in previous studies,<sup>4,9,10</sup> and a quantitative understanding of the effect of the fibers and the underlying mechanisms is still not complete. Reinforcement of the biopolymer with strong natural fibers, will presumably results in stronger materials, but this might not necessarily improve the thermal sensitivity of the materials. The properties of composites are governed by the properties of the constituent fiber and matrix phases, in addition to the ability of efficient load transfer at the fiber/matrix

interface,<sup>11</sup> and all three elements contribute to the resulting thermal sensitivity of the composites.

In the present study, the mechanical properties of two widely used biopolymers, polylactic acid, and cellulose acetate, showing different thermal sensitivity behavior, are examined as function of increasing ambient temperatures in the range 5–80°C. The objective is to study if the thermal sensitivity parameters normally provided for polymers, the glass transition temperature and the heat deflection temperature, are sufficient to describe the change in mechanical properties with temperature. An analogous series of measurements are performed on jute fiber composites with polylactic acid and cellulose acetate matrices, to investigate the effect of fiber reinforcement.

## EXPERIMENTAL

### Materials

Composites were made from jute fiber fabric (type 100/16, plain weave) supplied by Nevotex, Sweden and two types of biopolymeric matrices. Polylactic acid (PLA) (type 4032D) (Nature Works LLC, USA) film with thickness 0.4 mm was supplied by ILIP, Italy. Granulates of a biopolymer blend (type Biograde C7500) based on cellulose acetate (CA) were supplied by FKUR Kunststoff GmbH, Germany and extruded into film with thickness 0.5 mm. The jute fabric was woven with 30% of the fibers in the warp direction and 70% in the weft direction.

### Processing of Composites

Composite plates with the dimensions 300 × 250 × 2 mm<sup>3</sup> were made by a film-stacking technique, followed by compression molding. For each composite plate, four layers of biopolymer film and three layers of jute fabric were used. Prior to processing, the jute fabrics were dried under vacuum at room temperature over night. The compression molding conditions were 190 or 225°C (for PLA and CA matrix composites, respectively), 20 mbar, 5 min for the initial vacuum heating step, and 30°C, 200 kN, 1 min for the final consolidation step. The process is described in details in the study by Madsen and Lilholt.<sup>12</sup> The PLA and CA matrix composites were heated at two different temperatures due to the different melting temperatures on 165–173 and >180°C for the two polymers, respectively, as given by the supplier data sheet. Neat polymer plates with the dimensions 140 × 210 × 2 mm<sup>3</sup> were made using the same processing technique as described for the composites.

Prior to processing, the layers of polymer films and dried jute fabric were weighed to determine the fiber weight fraction ( $W_f$ ) of the composite plates.  $W_f$  was calculated as the weight of fibers with respect to the total weight of fibers and matrix [eq. (1)]. After processing, the density of the composite plate was determined by mass of composite ( $m_c$ ) and volume of composite ( $v_c$ ), as calculated from the measured thickness and area of the plate [eq. (2)]. From this, the fiber volume fraction ( $V_f$ ) was calculated based on  $W_f$  and the ratio of the density of the composite and the fibers ( $\rho_f$ ) using eq. (3). The density of the jute fibers was assumed to be 1.50 g cm<sup>-3</sup> as reported in the literature for natural fibers in general.<sup>13</sup>

$$W_f = \frac{m_f}{m_f + m_m} \quad (1)$$

$$\rho_c = \frac{m_c}{v_c} \quad (2)$$

$$V_f = W_f \cdot \frac{\rho_c}{\rho_f} \quad (3)$$

Because three layers of jute fabric were used for all composites, it was assumed that the amount of jute fibers is the same, and only the amount of polymer vary between the PLA and CA matrix composites. From the jute/PLA composite, the mean thickness of the jute fabric ( $t_f$ ) was calculated using the determined fiber volume fraction in the jute/PLA composite ( $V_f^{\text{PLA}}$ ) and the thickness of the composite plates ( $t_c^{\text{PLA}}$ ) [eq. (4)].

$$t_f = t_c^{\text{PLA}} \cdot V_f^{\text{PLA}} \quad (4)$$

The thickness of the jute/CA composites ( $t \approx 2.4$  mm) was larger than the jute/PLA composites ( $t \approx 2.2$  mm), because the CA film was thicker than the PLA film. The volume fraction of jute fibers in the jute/CA composites ( $V_f^{\text{CA}}$ ) was calculated using eq. (5) as the ratio of thickness of the jute fabric and thickness of each of the jute/CA composite plates ( $t_c^{\text{CA}}$ ).

$$V_f^{\text{CA}} = \frac{t_f}{t_c^{\text{CA}}} \quad (5)$$

The determined average  $V_f$  for the jute/PLA composites was (32.1% ± 1.9%), and (28.4% ± 0.3%) for the jute/CA composites. In the micromechanical calculations, the determined fiber volume fraction for the individual plates is used.

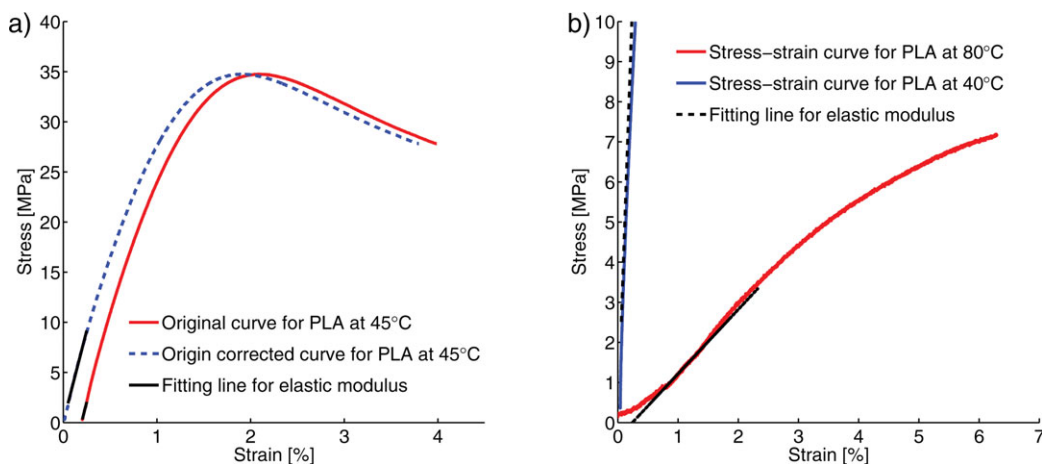
For all composite and neat polymer plates, dog bone shaped specimens were cut according to ISO 527-1 with outer specimen dimensions 180 × 25 mm<sup>2</sup>, and gauge section dimensions 20 × 15 mm<sup>2</sup>.

### Mechanical Testing

All specimens were tested in tension (Instron 115) with a 10 kN load cell (UK 562) and a displacement rate of 1 mm min<sup>-1</sup> (approximately a strain rate of 1% min<sup>-1</sup>) at temperatures ranging from 5 to 80°C. The composite specimens were tested in the weft direction of the jute fabric. An oven enclosing the clamped specimen in the tensile testing machine was used to control the temperature of the test environment. The specimens were equilibrated at a given temperature for 30 min before testing. The temperature at the surface of the specimens was monitored by a thermometer. The tensile tests of the neat PLA specimens from 45 to 80°C were performed using a video-extensometer as these specimens were too soft to allow attaching an extensometer without bending the specimens. The remaining specimens were tested using an attached extensometer centered at the gauge section of the specimens.

### Data Processing

Initially, the recorded strain values were calibrated so that the initial linear phase of the curve intercepts the origin [see Figure 1(a)]. The elastic modulus of the stress–strain curves was found in a strain interval of 0.05–0.25%. However, for the neat PLA specimens tested at 55, 60, and 80°C, the stress–strain curve did not show a distinct initial linear phase, and the elastic modulus



**Figure 1.** Stress–strain curves of neat PLA showing (a) the applied approach of origin-correcting the curves so that the initial linear phase intercepts the origin, and (b) the applied approach of determining the elastic modulus of PLA at high temperatures where the curves do not show an initial linear phase. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]

was found in the apparent linear phase of the curve above the toe region, and before the yield stress as seen in Figure 1(b).

The maximum tensile stress of the stress–strain curves was determined as the maximum point of the curves [see example in Figure 1(a)]. For some of the neat PLA specimens tested at 55, 60, and 80°C, no maximum stress was found before the test ended at about 6% strain [see example in Figure 1(b)]. In these cases, the maximum tensile stress was read from the curve at 5.0% strain. The stresses for these cases were found to be very low.

### Model Calculations

The elastic modulus ( $E$ ) of the composites is assumed to be given by the weighted sum of the elastic moduli in the axial direction ( $E_{c,1}$ ) and in the transverse direction ( $E_{c,2}$ ) [eq. (6)], weighted by the parameter  $\beta$ , which is the fraction of fibers in the axial direction of the fabric [eq. (7)]. Thus, it is assumed that the stiffness in each direction works as two independent layers on top of each other. Each elastic modulus is calculated according to the conventional rule of mixtures models for unidirectional composites tested in the axial and transverse directions, including parameters for the elastic modulus of matrix ( $E_m$ ) and fibers ( $E_f$ ), and their respective volume fractions [eqs. (8) and (9)]. The full model equation is shown by eq. (10).

$$E_c = \beta \cdot E_{c,1} + (1 - \beta) \cdot E_{c,2} \quad (6)$$

$$\beta = \frac{v_f^1}{v_f} \quad (7)$$

$$E_{c,1} = E_m \cdot (1 - V_f) + E_f \cdot V_f \quad (8)$$

$$E_{c,2} = \frac{E_m \cdot E_f}{(1 - V_f) \cdot E_f + E_m \cdot V_f} \quad (9)$$

$$E_c = \beta \cdot E_m \cdot (1 - V_f) + \beta \cdot E_f \cdot V_f + \frac{(1 - \beta) \cdot E_m \cdot E_f}{(1 - V_f) \cdot E_f + E_m \cdot V_f} \quad (10)$$

When the elastic modulus of the composites is measured, the effective elastic modulus of the fibers ( $E_f$ ) can be back-calculated

from eq. (10). The effective fiber modulus,  $E_f$  can be determined by solving the second order polynomial eq. (11), which is derived from eq. (10).

$$0 = E_f^2 (\beta V_f \cdot V_m) + E_f (\beta \cdot V_m^2 \cdot E_m + \beta \cdot V_f^2 \cdot E_m + (1 - \beta) \cdot E_m - E_c \cdot V_m) + \beta \cdot E_m^2 \cdot V_m \cdot V_f - E_c \cdot E_m \cdot V_f \quad (11)$$

For all cases, the composites has been tested in the weft direction where  $\beta = 0.7$ . For comparison it can be noted that by making the more simple approximation that all fibers in the transverse direction act as the matrix material, a 8–10% higher effective fiber stiffness for the investigated cases would be found.

Assuming that fiber failure is causing composite failure, the effective maximum stress of the fibers can be back-calculated from the rule of mixtures model given by eq. (12) where  $\sigma_m^*$  is the stress in the matrix at the composite failure strain. It is assumed that only the fibers in the axial direction contribute to the effective maximum stress of the composite and that the fibers in the transverse direction are included as part of the matrix volume fraction. The derived effective maximum stress of the fibers within the composite is shown in eq. (13).

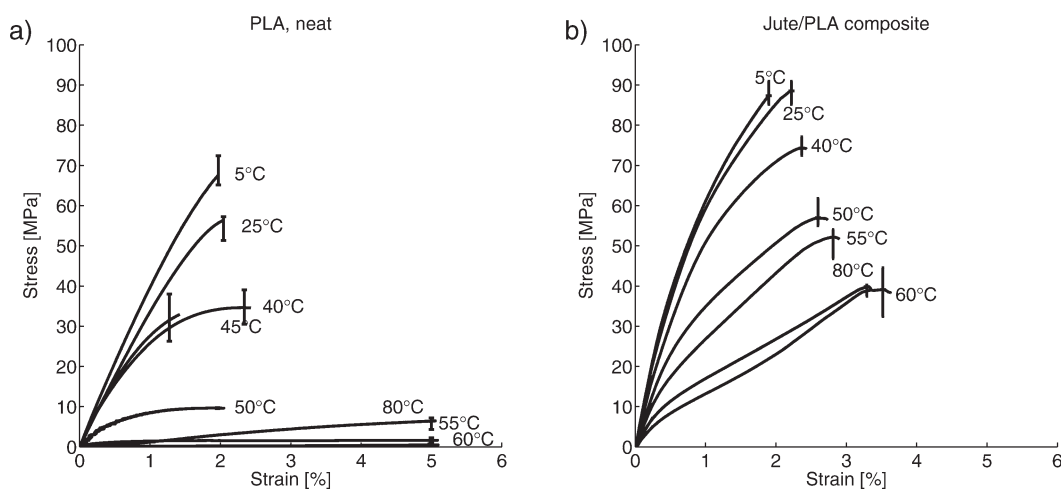
$$\sigma_c = \sigma_m^* \cdot (V_m + (1 - \beta) \cdot V_f) + \sigma_f \cdot V_f \cdot \beta = \sigma_m^* \cdot (1 - V_f \cdot \beta) + \sigma_f \cdot V_f \cdot \beta \quad (12)$$

$$\sigma_f = \frac{\sigma_c - \sigma_m^* (1 - V_f \cdot \beta)}{V_f \cdot \beta} \quad (13)$$

## RESULTS AND DISCUSSION

### PLA and Jute/PLA Composites

As seen in Figure 2(a), the stress–strain curves for neat PLA become lower as the testing temperature is increased from 5 to 80°C. The largest drop between two stress–strain curves is seen when the temperature is increased from 45 to 50°C. As the temperature is further increased and exceeds the glass transition



**Figure 2.** Stress–strain curves of (a) neat PLA and (b) jute/PLA composites at increasing temperatures from 5 to 80°C. Each curve is a selected representative of the test series at each temperature. The maximum stress value for each curve is indicated, and the error bar represents a 95% confidence interval for all samples.

temperature for PLA ( $T_g = 58^\circ\text{C}$ ),<sup>14</sup> the specimens become soft and they elongate extensively. At 55 and 60°C, the stress–strain curves show very low maximum stresses and large strains. It is seen that the stress–strain curve for 80°C is positioned above the ones for 55 and 60°C.

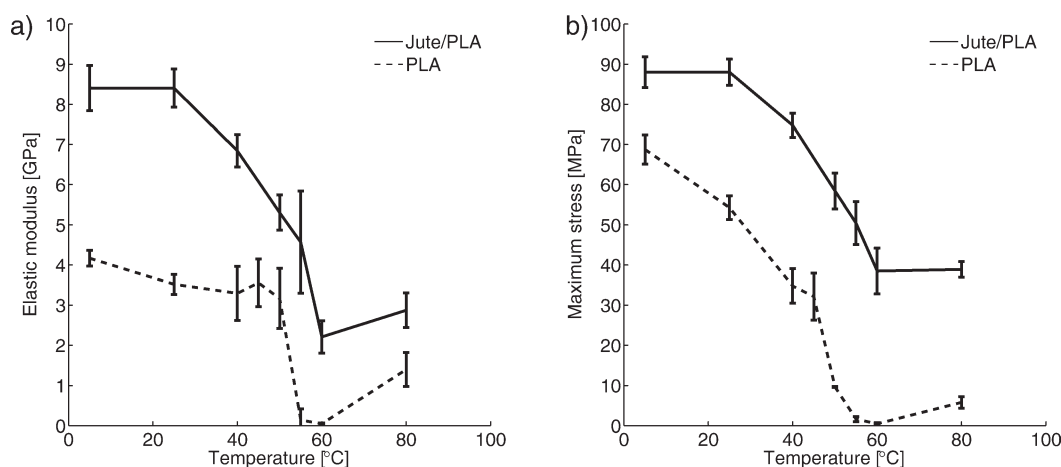
As seen in Figure 3(a), the elastic modulus for neat PLA is sustained at about 3.5 GPa at testing temperatures up to 45°C, and then, when the temperature is increased above the glass transition temperature of PLA, the elastic modulus reduces towards zero. In contrast to the change of the elastic modulus, the maximum stress ( $\sigma$ ) of PLA gradually decreases from about  $\sigma^{5^\circ\text{C}} = (69 \pm 4)$  MPa to  $\sigma^{45^\circ\text{C}} = (32 \pm 6)$  MPa as the temperature is increased from 5 to 45°C, as seen in Figure 3(b). When the temperature approaches 60°C, the maximum stress is  $\sim 0$ , which is consistent with the results of the elastic modulus.

The neat PLA exhibits a somehow surprising behavior with a slightly increased elastic modulus and maximum stress for the temperature above 60°C. Even though this has not been further

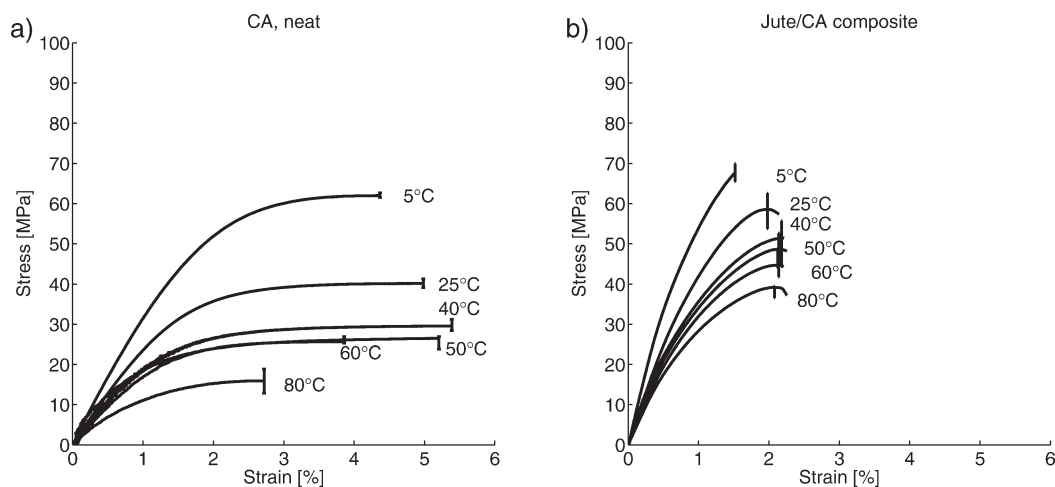
investigated in the present study, a thermally activated crystallization has earlier been reported for PLA<sup>15</sup> at high temperature and low strain rates, and this might explain the observed behavior.

For the jute/PLA composites, a gradual drop in the stress–strain curves is seen when the temperature is increased [Figure 2(b)]. In addition, the specimens are consistently showing an increased strain at fracture when the temperature is increased. Accordingly, as seen in Figure 3(a,b), the mechanical properties of jute/PLA composite show a gradual decrease as function of temperature. The elastic modulus is decreased from  $E^{5^\circ\text{C}} = (8.5 \pm 0.5)$  GPa to  $E^{50^\circ\text{C}} = (5.3 \pm 0.4)$  GPa when the temperature is increased from 5 to 50°C, and the maximum stress is decreased from  $\sigma^{5^\circ\text{C}} = (86 \pm 4)$  MPa to  $\sigma^{50^\circ\text{C}} = (58 \pm 5)$  MPa in the same temperature interval. Minimum values of elastic modulus and maximum stress on about  $E^{60^\circ\text{C}} = (2.3 \pm 0.4)$  GPa and  $\sigma^{60^\circ\text{C}} = (38 \pm 6)$  MPa are seen at 60°C.

Because the elastic modulus and maximum stress of the neat PLA specimens are  $\sim 0$  at 60°C, it is indicated that the



**Figure 3.** Diagrams showing (a) elastic modulus and (b) maximum stress of neat PLA and jute/PLA composites as a function of temperature. The error bars represent 95% confidence intervals.



**Figure 4.** Stress–strain curves of (a) neat CA and (b) jute/CA composites at increasing temperatures from 5 to 80°C. Each curve is a selected representative of the test series at each temperature. The maximum stress value for each curve is indicated, and the error bar represents a 95% confidence interval for all samples.

resistance to deformation of the jute/PLA composite at this temperature is sustained by the jute fibers. Altogether, the results clearly show that the jute fibers function as reinforcement of the PLA polymer, and this leads to a higher elastic modulus and maximum stress of the composite at all temperatures, compared to the neat PLA.

Shih and Huang<sup>8</sup> showed that the heat deflection temperature (HDT) for neat PLA was 62°C, and it was increased to 130°C, when PLA was reinforced with banana fibers. This finding can however not be supported by the results in the present study, which shows that the thermal sensitivity of the mechanical properties of PLA is not improved by reinforcement with natural fibers, only the elastic modulus and maximum stress of the PLA composites are at a higher level, due to the reinforcement effect of the fibers. From this study it is observed that the temperature impact is seen gradually, rather than taking place at a well-defined temperature interval. The HDT and the glass transition temperature cannot be used as the sole thermal sensitivity parameters for determining the gradual change in mechanical properties of polymers and composites.

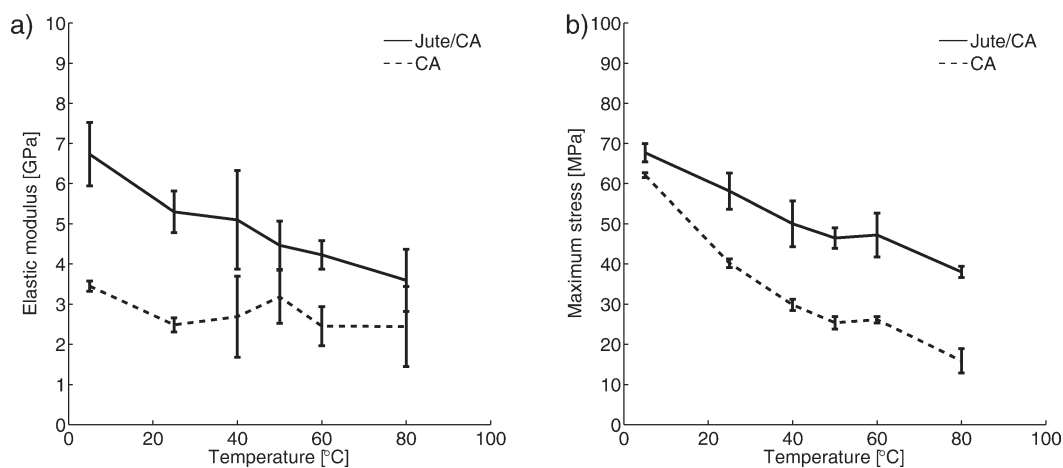
As already mentioned, the results show that the elastic modulus for neat PLA does not change at temperatures from 5 to 45°C, whereas the elastic modulus of the jute/PLA composite is reduced gradually in the same temperature interval. This points toward that the reinforcement efficiency of the jute fibers changes as a function of temperature. The cell wall of jute fibers is consisting of cellulose, lignin and hemicellulose, which all have glass transition temperatures well above 100°C.<sup>16</sup> In addition, a previous study of jute fiber composites<sup>17</sup> estimated that the elastic modulus of the fibers barely changes within the temperature range used in present study. Thus, based on these previous findings, it is indicated that the reduction of the elastic modulus seen for the jute/PLA composite, but not seen for neat PLA, are not caused by the fibers themselves. Instead, it is indicated that the reinforcement efficiency of the fibers is reduced due to changes in the interaction between matrix and fibers within the composite. Even though the matrix sustains a con-

stant elastic modulus, the ability of the matrix to transfer the forces to the fibers is apparently reduced when the temperature is increased. To validate this hypothesis, further investigations are needed of the thermal induced change of interface bonding properties in natural fiber composites with biopolymeric matrices.

#### CA and Jute/CA Composites

The stress–strain curves for neat CA tested at temperatures between 5 and 80°C are shown in Figure 4(a). It can be observed that the curves are lowered as a function of temperature. A relatively large effect of temperature is seen between 5 and 25°C. The glass transition temperature or HDT for the commercial CA-based Biograde C7500 used in the present study was not listed by the supplier. The glass transition temperature for pure CA is 69°C.<sup>18</sup> The HDT for CA (CA 398-30, Eastman Chemical) was determined to be 62°C by Wibowo et al.<sup>19</sup> Based on the values of these thermal parameters, it can be expected that a large difference in the stress–strain curves for CA should be seen between 60 and 80°C. This is supported by the stress–strain curve for CA at 80°C showing a clearly lower maximum stress and strain. However, as seen in Figure 5(a), the elastic modulus of CA is almost constant at about  $E = 2.7$  GPa when the temperature is increased from 25 to 80°C, and only showing a larger value of  $E^{5^\circ\text{C}} = (3.4 \pm 0.1)$  GPa at 5°C. In contrast, the maximum stress for neat CA is more dependent on the increase in temperature with values of  $\sigma^{5^\circ\text{C}} = (62 \pm 1)$  MPa and  $\sigma^{80^\circ\text{C}} = (16 \pm 3)$  MPa at the two extreme temperatures of 5 and 80°C, respectively. A reduction in maximum stress can be observed between 5 and 40°C, after which the maximum stress reaches a plateau, but decrease further as temperature is increased from 60 to 80°C.

In the case of jute/CA composites, the stress–strain curves are gradually lowered when the temperature is increased [Figure 4(b)], and with a smaller differences between the curves than for the neat CA. In addition, the overall shape of the stress–strain curves changes less for the jute/CA composites than for the neat CA. As shown in Figure 5, both the elastic modulus



**Figure 5.** Diagrams showing (a) elastic modulus and (b) maximum stress of neat CA and jute/CA composites as a function of temperature. The error bars represent 95% confidence intervals.

and the maximum stress of the jute/CA composites consistently decrease, when the temperature is increased in the range from 5 to 80°C with values of  $E^{5^\circ\text{C}} = (6.7 \pm 0.8)$  GPa to  $E^{80^\circ\text{C}} = (3.6 \pm 0.8)$  GPa, and  $\sigma^{5^\circ\text{C}} = (68 \pm 2)$  MPa to  $\sigma^{80^\circ\text{C}} = (38 \pm 1)$  MPa, respectively.

#### Comparison between PLA and CA

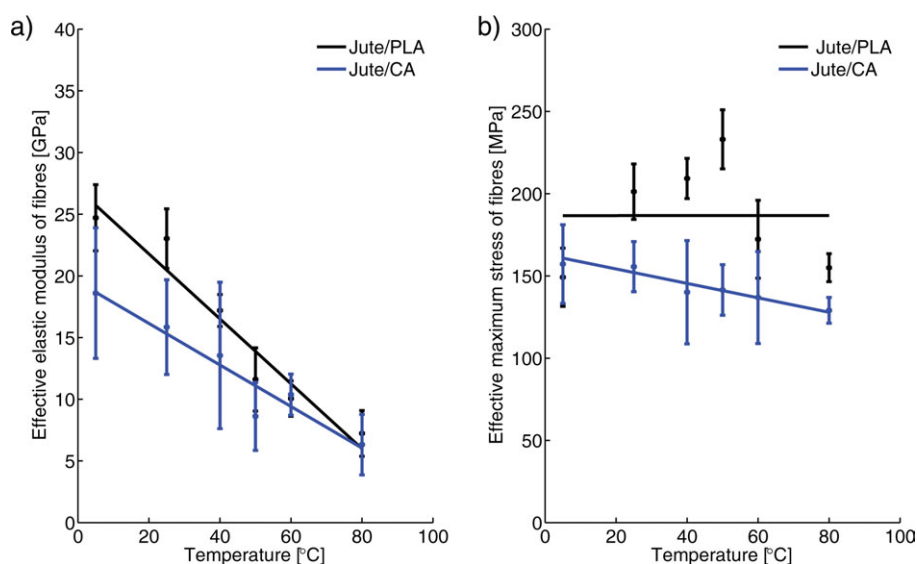
The comparison between neat PLA and neat CA show that their basic mechanical properties at 25°C are in the same range with  $E^{25^\circ\text{C}} = (3.5 \pm 0.2)$  GPa and  $E^{25^\circ\text{C}} = (2.5 \pm 0.2)$  GPa, and  $\sigma^{25^\circ\text{C}} = (54 \pm 3)$  MPa and  $\sigma^{25^\circ\text{C}} = (40 \pm 1)$  MPa, respectively (see Figures 3 and 5). However, the mechanical properties of their respective composites show a larger difference. Because the same fibers are used for the CA and the PLA matrix composites, it is expected that the reinforcement efficiency of the fibers should be the same, and this should lead to composites with almost equal properties. This is however not supported by the results showing values of  $E^{25^\circ\text{C}} = (8.4 \pm 0.4)$  GPa and  $E^{25^\circ\text{C}} =$

$(5.3 \pm 0.5)$  GPa and  $\sigma^{25^\circ\text{C}} = (89 \pm 3)$  MPa and  $\sigma^{25^\circ\text{C}} = (58 \pm 4)$  MPa of the jute/PLA and jute/CA composites, respectively, at 25°C. The difference in reinforcement efficiency of the jute fiber in the two types of composites is analyzed by micromechanical models in the following section.

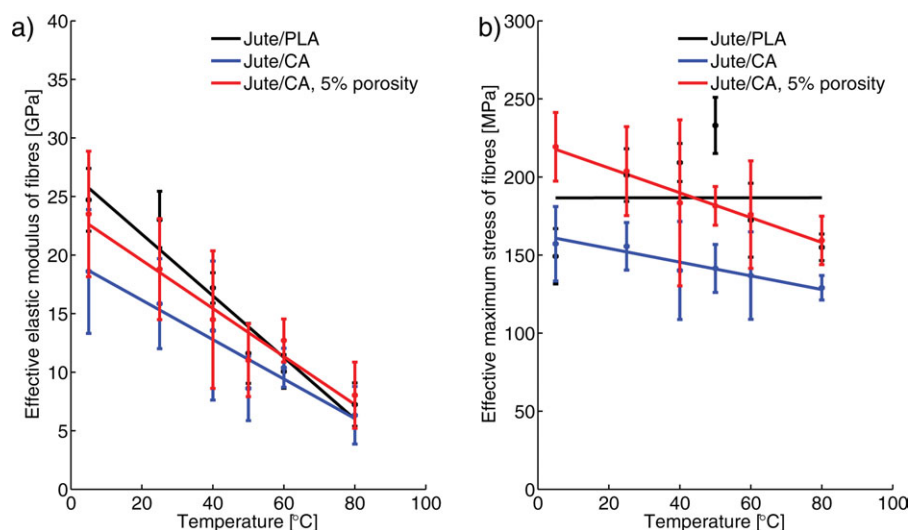
Furthermore, the comparison between neat PLA and CA show that the mechanical properties of neat CA is less sensitive toward changes in temperatures in the investigated temperature range of 5–80°C. Consequently, the mechanical properties of the jute/CA composites exceed the ones of the jute/PLA composites when the temperature is increased above 50°C.

#### Effective Elastic Modulus and Maximum Stress of Fibers

Figure 6 shows the back-calculated effective elastic modulus and maximum stress of the jute fibers, as calculated by the micromechanical models shown by eqs. (10) and (12). From Figure 6(a), it is seen that the effective elastic moduli of the jute fibers within



**Figure 6.** Diagrams showing the back-calculated effective (a) elastic modulus and (b) maximum stress for jute fibers within jute/PLA composites and jute/CA composites. Lines are trend lines. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]



**Figure 7.** Same as in Figure 6, but including estimated effective (a) elastic modulus and (b) maximum stress of jute fibers within jute/CA composites containing 5% porosity. [Color figure can be viewed in the online issue, which is available at [wileyonlinelibrary.com](http://wileyonlinelibrary.com).]

the two composites are not identical; the values are generally higher for the jute/PLA composites. At 25°C, the effective elastic modulus is  $E^{25^\circ\text{C}} = (23.4 \pm 2.4)$  GPa and  $E^{25^\circ\text{C}} = (16.0 \pm 3.8)$  GPa for the jute/PLA and jute/CA composites, respectively. These values are in the lower end of previously estimated effective elastic moduli of natural fibers in composites.<sup>20</sup> In Figure 6(a), the difference between the two composites is decreased when the temperature is increased. At 60 and 80°C, there is no difference between the composites, and the effective elastic modulus of the fibers is about  $E = 5.8$  GPa. As already noted and discussed above, the consistent reduction of the reinforcement efficiency of the fibers with temperature is probably due to changes in the interaction between matrix and fibers within the composite, and this requires further experimental investigations to document the underlying parameters and mechanisms. As shown in Figure 6(b), the effective maximum stress of the fibers is generally larger for the jute/PLA composites than for the jute/CA composites. At 25°C, the effective maximum stress is  $\sigma^{25^\circ\text{C}} = (201 \pm 16)$  MPa and  $\sigma^{25^\circ\text{C}} = (156 \pm 17)$  MPa for the jute/PLA and jute/CA composites, respectively. However, in contrast to the effective fiber elastic modulus, no clear decreasing effect on the effective fiber maximum stress with increasing temperature can be seen.

The calculation of the effective elastic modulus by Eq. (10) does not account for porosity in the composites. It might be speculated that the porosity content in the jute/CA composites is larger than in the jute/PLA composites due to the slightly lower melt flow rate index of CA (17–21 g/10 min, as listed in supplier datasheet) compared to the index for PLA (20 g/10 min).<sup>21</sup> The lower flow rate index for CA could lead to less efficient impregnation of the fibers, and thereby a larger porosity content. This could (partly) explain why the back-calculated effective mechanical properties of the jute fibers are higher for the jute/PLA composites. To address the effect of porosity, the micromechanical models can be modified to take account of this additional phase in the composites.<sup>12,20</sup> The porosity corrected models are shown by eqs. (14) and (16), where  $E'_c$  and

$\sigma'_c$  are the elastic modulus and maximum stress of the (perfect) composites with no porosity, and  $V_p$  is porosity. The derived equations for the back-calculation of the effective elastic modulus and maximum stress of the fibers, in the case of composites containing porosity, are shown by eqs. (15) and (17).

$$E_c = E'_c \cdot (1 - V_p)^2 \quad (14)$$

$$0 = E_f^2 (\beta \cdot V_f \cdot (V_m - V_p)) + E_f \left( \beta \cdot (V_m - V_p)^2 \cdot E_m + \beta \cdot V_f^2 \cdot E_m + (1 - \beta) \cdot E_m \cdot \frac{E_c}{(1 - V_p)^2} \cdot (V_m - V_p) \right) + \beta \cdot E_m^2 \cdot (V_m - V_p) \cdot V_f - \frac{E_c}{(1 - V_p)^2} \cdot E_m V_f \quad (15)$$

$$\sigma_c = \sigma'_c \cdot (1 - V_p)^2 \quad (16)$$

$$\sigma_f = \frac{\frac{\sigma_c}{(1 - V_p)^2} - \sigma_m^* (1 - \beta \cdot V_f - V_p)}{\beta \cdot V_f} \quad (17)$$

Figure 7 shows how an estimated porosity of 5% ( $V_p = 0.05$ ) in the jute/CA composites will affect the back-calculated effective elastic modulus and maximum stress of the fibers within the composites. A porosity content of 5% is considered to be realistic based on the results of previous studies of porosity in natural fiber composites.<sup>12,22</sup> The figure shows that the applied approach of porosity correction leads to that both the effective elastic modulus and the maximum stress of the fibers are approximately at the same level in the two types of composites, irrespectively of the used matrix type.

Another explanation for the difference in the reinforcement efficiency of the fibers in the jute/CA and jute/PLA composites could be the different composite processing temperature used for the two composites. Because CA has a higher melting temperature than PLA, the jute/CA composites were processed at 225°C, whereas the jute/PLA composites were processed at

190°C. In other studies of jute fiber composites,<sup>3,23</sup> the maximum stress of the composites was found to be reduced when the composite processing temperature was increased in the range from 190 to 220°C. Similar findings were shown in a study of hemp fiber composites.<sup>24</sup> Thus, thermal degradation of the jute fibers during processing of the composites might also (partly) explain why the jute fibers in the jute/CA composites show lower reinforcement efficiency.

## CONCLUSIONS

The influence of ambient temperature on the mechanical properties of jute fiber composites with two different biopolymeric matrices, polylactic acid (PLA) and cellulose acetate (CA), were examined, and the following conclusions are made:

1. The elastic modulus of neat PLA is almost constant until at temperature of about 45°C, after which the elastic modulus is decreased rapidly. For neat CA, the elastic modulus is almost constant in the whole temperature range of 5–80°C. However, for both PLA and CA, the maximum stress is clearly decreased when the temperature is increased.
2. When the two biopolymers are reinforced with jute fibers, both the elastic modulus and maximum stress of the composites are consistently reduced when the temperature is increased.
3. The thermal sensitivity parameters normally provided for polymers, the glass transition temperature, and the heat deflection temperature, provide insufficient information of the gradual change in mechanical properties as a function of temperature as observed for the neat biopolymers and the jute fiber/biopolymer composites.
4. By the use of micromechanical models, it is established that the reinforcement efficiency of the jute fibers in the composites (i.e., the back-calculated effective elastic modulus of the fibers) is decreased when the temperature is increased.
5. The micromechanical calculations show lower reinforcement efficiency of the jute fibers within the jute/CA composites than the jute/PLA composites. This difference is considered to be due to an expected higher porosity in the jute/CA composites, a mechanism which is supported by model calculations, and/or because the jute/CA composites are processed at a higher temperature. The latter leads to thermal degradation of the jute fibers, and thereby to a reduction of their mechanical properties.

## ACKNOWLEDGMENTS

This work was supported by the Danish funding organization Regional Growth Forum Zealand through a development and research project “Bio/Bio composites.”

## REFERENCES

1. Mohanty, A. K.; Misra, M.; Drzal, L. T.; Selke, S. E.; Harte, B. R.; Hinrichsen, G. In *Natural Fibers, Biopolymers, and Biocomposites*; Mohanty, A. K., Misra, M., Drzal, L. T., Eds.; T. Taylor and Francis Group: Boca Raton, 2005; Chapter 1, p 1.
2. Plackett, D.; Andersen, T. L. In *Sustainable Natural and Polymeric Composites—Science and Technology*; Lilholt, H.; Madsen, B.; Toftegaard, H. L.; Cendre, E.; Megnis, M.; Mikkelsen, L. P.; Sørensen, B. S., Eds.; Risø Natural Laboratory: Roskilde, Denmark, 2002; p 299.
3. Plackett, D.; Andersen, T. L.; Pedersen, W. B.; Nielsen, L. *Compos. Sci. Technol.* **2003**, *63*, 1287.
4. Satyanarayana, K. G.; Arizaga, G. C. G.; Wypych, F. *Prog. Polym. Sci.* **2009**, *34*, 982.
5. Kang, J. T.; Kim, S. H. *Macromol. Res.* **2011**, *19*, 789.
6. Mohanty, A. K.; Wibowo, A.; Misra, M.; Drzal, L. T. *Compos. A* **2004**, *35*, 363.
7. Mathew, A. P.; Oksman, K.; Mohini, S. *J. Appl. Polym. Sci.* **2005**, *97*, 2014.
8. Shih, Y.; Huang, C. *J. Polym. Res.* **2011**, *18*, 2335.
9. Petersson, L.; Kvien, I.; Oksman, K. *Compos. Sci. Technol.* **2007**, *67*, 2535.
10. Nyambo, C.; Mohanty, A. K.; Misra, M. *Biomacromolecules* **2010**, *11*, 1654.
11. Hull, D.; Clyne, T. W. In *An Introduction to Composite*, 2nd ed.; Clarke, D. R., Suresh, S., Ward, I. M., Eds.; Press Syndicate of the University of Cambridge: United Kingdom, 1981; Chapter 7, p 133.
12. Madsen, B.; Lilholt, H. *Compos. Sci. Technol.* **2003**, *63*, 1265.
13. Bismarck, A.; Mishra, S.; Lampke, T. In *Natural Fibers, Biopolymers, and Biocomposites*; Mohanty, A. K., Misra, M., Drzal, L. T., Eds.; T. Taylor and Francis Group: Boca Raton, 2005; Chapter 2, p 37.
14. Bondeson, D.; Oksman, K. *Compos. A* **2007**, *38*, 2486.
15. Mulligan, J.; Cakmak, M. *Macromolecules* **2005**, *38*, 2333.
16. Salmén, L. *J. Mater. Sci.* **1984**, *19*, 3090.
17. Cichocki, F. R.; Thomason, J. L. *Compos. Sci. Technol.* **2002**, *62*, 669.
18. Riley, R.; Gardner, J. O.; Merten, U. *Science* **1964**, *143*, 801.
19. Wibowo, A. C.; Misra, M.; Park, H.; Drzal, L. T.; Schalek, R.; Mohanty, A. K. *Compos. A* **2006**, *37*, 1428.
20. Madsen, B.; Thygesen, A.; Lilholt, H. *Compos. Sci. Technol.* **2009**, *69*, 1057.
21. Kang, J. T.; Kim, S. J. *Macromol. Res.* **2011**, *19*, 789.
22. Madsen, B.; Thygesen, A.; Lilholt, H. *Compos. Sci. Technol.* **2007**, *67*, 1584.
23. Ma, H.; Joo, C. W. *J. Compos. Mater.* **2011**, *45*, 1451.
24. Madsen, B.; Lilholt, H. *Compos. A* **2007**, *38*, 2204.