PROCESS-CONTROLLED TENSILE PROPERTIES OF NEWLY DEVELOPED BAMBOO COMPOSITE MATERIALS

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ABSTRACT Phyllostachys edulis or Moso bamboo, which is the most common bamboo species in China, has been chosen as the basis for the newly developed bamboo reinforced composite materials. Each sample employs a defined epoxy resin composition and is fabricated by the hot press forming method. The relationship between tensile strength, pressure, temperature and pressing/holding time is being analyzed. So far, it is observed that a maximum tensile strength (~ 180 MPa) can be obtained at a certain temperature/pressure combination. Further increasing the pressure or temperature has a negative effect on the mechanical properties of the composites. Similar assumptions hold for decreasing temperatures or pressures. On the other hand, the pressing/holding time plays a minor role but may be significant for enhancing the integrity of the fibers with the resin at early pressing times. Thus, understanding of the process parameters allows us to tune the material features, which is an appreciable advantage for the future development of reliable high-performance composite materials with well-defined properties.

INTRODUCTION

Fiber-reinforced composite materials became an indispensable part of our lives. Whether consumer goods, automotive or construction industries one cannot imagine these without composites reinforced by glass fibers, carbon fibers, graphite fibers, aramid fibers etc. However, in the light of the present economic and environmental crises some significant drawbacks exist in the application of these relatively new material technologies: The primary raw material is petroleum, which is a non-renewable resource. Most of these materials are difficult to recycle. The fabrication procedures are rather strenuous because the fibers are hard-to-handle and/or toxic in nature. All these features make composite materials based on synthetic inorganic fibers extremely expensive and environmentally unfriendly. Moreover, no matter how great the benefits are in terms of mechanical properties, only a small percentage of the world’s population can actually afford and profit from them due to their high price.

Researchers all over the globe have already recognized these drawbacks and the quest for more affordable and easily processable alternatives has already taken its course some time ago with the application of, for example, natural fibers as composite reinforcement materials.\textsuperscript{1,2,3} Compared with their synthetic inorganic counterparts, natural fibers such as pineapple, jute, sisal, henequen or bamboo bear considerable advantages due to their renewability, biodegradability, and – most importantly – negligible costs.

The study presented in this paper aims to mark further progress towards the development of new high-performance composite materials, which are based on natural fibers and can be obtained at significantly lower costs. On that account, we have chosen bamboo as raw
material\(^3\) and challenge the limitations of existent and rather tedious production techniques by introducing a new method of processing the raw bamboo into an easy to handle non-toxic fibrous material. A hot-press fabrication method yields a high-tensile strength epoxy composite. By understanding and controlling the process conditions we are able to tune the tensile capacities of our composites and prove the general concept of using natural organic fiber reinforcement to achieve mechanical properties in the range of synthetic composite materials.

**EXPERIMENTAL PROCEDURES**

**Materials.** *Phyllostachys edulis* or Moso bamboo has been prepared into 35 cm long and 2 cm wide strips with a thickness of approximately 2 mm. The strips have been dried in an oven for several hours at 60°C to reduce the moisture content to less than 10%. To assess the reinforcing quality of the bamboo fibers, unidirectional composites were fabricated using epoxy as matrix. A standard two-component epoxy system with a bisphenol-A-based resin and a diamine-based hardener was employed. The composites were prepared by pre-aligning the epoxy-impregnated strips into a layered structure and placing them into the metallic mold of a hot press. Thereby, controlling the position of the nodes and minimizing the movement and deformation of the fibers turned out to be important in order to obtain void-free unidirectional fiber composites. Subsequently, the test specimens were subjected to different pressures (between 15 and 25 MPa) and temperatures (between 80°C and 140°C) at various pressing/holding times to afford highly compressed composite samples with a volume fraction of more than 70%. Figure 1 shows macroscopic photographs of the raw-bamboo strips and the resulting composites. Finally, the composites were cured for 12 hours at 40°C.

![Figure 1. Macroscopic photograph of the oven-dried bamboo strips (left) used for the fabrication of the composites and a corresponding composite specimen (right).](image)

**Tensile Testing of Bamboo Reinforced Composites and Bamboo Strips.** The unidirectional composite specimens were prepared into dog-bone shapes as per the ASTM D3039-D3039M-08 for tensile properties of polymer matrix composite materials, which was chosen due to the lack of a specific standard for bamboo composite materials. In order to be able to control the limiting values of the mechanical properties of the composite it is important to know the tensile strength of the reinforcing material. Therefore, the tensile strength of the raw oven-dried bamboo strips has been evaluated, as well. Tensile tests were carried out using a Shimadzu AG-IC 100kN machine in accordance with the ASTM D3039-08 standard at a strain rate of 1 mm/min. The tensile strength was calculated from the ultimate load and the cross-sectional area of fibers. From each composite sample at least five specimens have been tested and results exceeding a 10% standard deviation range, which has been statistically set as confidence interval, have been discarded.
Figure 2. Photograph of the “dog-bone”-shaped test specimens for the tensile tests after tensile failure.

Microscopy. The tensile fracture surface of the composite samples were analyzed with a Carl Zeiss SteREO Discovery.V12 optical stereomicroscope and then coated with silver for a field emission scanning electron microscopy (FESEM) analysis with a JEOL, JSM-6700F.

RESULTS AND DISCUSSION

Mechanical Properties of Natural Bamboo Strips. The intrinsic strength of single fibers of the Moso bamboo species is known to exceed 1.4 GPa.\textsuperscript{5,6} However, due to the natural brittleness of the lignin network that the fibers are embedded into the tensile capacity of fiber bundles drops to about 300 to 400 MPa.\textsuperscript{7} In the herein presented bamboo strips the fiber bundles are embedded into a holocellulose network, which further decreases their tensile strength as compared with single fibers. Our tests have shown that tensile failure of the raw oven-dried strips occurs already between 120 and 160 MPa. It is worth mentioning, however, that earlier studies have demonstrated that heat and pressure treatment as occurring when applying the hot press method may to a certain extent vary the natural strength of the fibrous material by affecting the fiber matrix.\textsuperscript{8} Thus, considering the high pressures and elevated temperatures applied throughout this study the tensile features of the strips might not necessarily constitute the limiting factor for the mechanical properties of the resulting composite material.

Tensile Strengths of the Bamboo Reinforced Composites. Table 1 summarizes the tensile strengths of the composites for different pressures and temperatures of the hot press fabrication process.

\begin{table}[h]
\centering
\begin{tabular}{|c|ccc|}
\hline
\textbf{T} & \textbf{P} & 15 MPa & 20 MPa & 25 MPa \\
\hline
80°C & 135 MPa & 147 MPa & 182 MPa  \\
& SD: 9% & SD: 7% & SD: 8% \\
100°C & 150 MPa & 181 MPa & 163 MPa  \\
& SD: 7% & SD: 3% & SD: 4% \\
120°C & 132 MPa & 158 MPa & /  \\
& SD: 5% & SD: 6% & / \\
140°C & / & 92 MPa & /  \\
& & SD: 54% & / \\
\hline
\end{tabular}
\caption{Tensile strengths of the Moso composites with the corresponding standard deviations (SD) in percent as obtained at different pressures and temperatures with a pressing/holding time of 15/15 minutes. At 140°C and at 120°C with 25 MPa the samples started to carbonize and it was difficult to obtain meaningful results.}
\end{table}
Table 1 reveals the highest tensile strengths when applying pressures of 20 MPa and 25 MPa at temperatures of 100°C and 80°C, respectively. Notably, with the 25 MPa/80°C setting the results were less consistent. At temperatures exceeding 120°C an increasing carbonization of the bamboo strips and the decomposition of the test samples has been observed. Samples fabricated with 20 MPa at 140°C have been tested for their tensile strength but the very broad mean variation of the results did not allow for any meaningful conclusions.

A correlation between the applied pressure and temperature is found when comparing the tensile strengths obtained for 15, 20 and 25 MPa at 80°C and 100°C as well as 20 and 25 MPa at 120°C and 100°C, respectively. The tensile strength at the higher temperature but lower pressure is comparable to the strength at lower temperature but higher pressure. Moreover, a maximum tensile strength is found at a certain pressure/temperature combination. The correlation suggests that it is possible to tune the mechanical properties of the composite by either varying the temperature or the pressure within a specific range. Thereby, the maximum tensile strength holds not only for one single pressure/temperature combination but may be also obtained when lowering the temperature and simultaneously increasing the pressure or vice versa. On the other hand, variation of both temperature and pressure at a time may significantly alter the mechanical properties of the composite, which is related to different crystallization behavior of the resin and/or a modification of the intrinsic fiber matrix. Figure 3 displays the relationship between the tensile strength and temperature at different pressures.

Remarkably, at 100°C the tensile strength reaches a maximum for both 15 and 20 MPa. At 25 MPa, on the other hand, the maximum strength is already obtained at 80°C, which insinuates a shift of the 25 MPa graph to lower temperatures. This trend implies that above a certain temperature the mechanical properties are mainly controlled by the pressure, which is well in line with the fact that the epoxy matrix requires a certain activation temperature to form a solid network. Consequently, the processing temperature will most likely impact the resin properties and the pressure will affect the interactions between the bamboo itself and between the bamboo and the epoxy matrix.
A variation of the press/hold time between 5/5 minutes and 60/60 minutes (Table 2) appears to affect the tensile strength of the composites only in a time range below 30 minutes. At this point it is important to mention that after the hot press processing the samples were cured for 12 h in order to achieve full strength of the resin. Hence, the press/hold time is considered to mainly affect the interactions between the bamboo fibers and the resin and not to impact the curing of the resin. The interactions formed between the bamboo fibers and the polymer in the 5/5-minute press/hold composites are weaker as for 15/15 minutes samples and the tensile stress is less favorably transmitted from the fibers onto the resin matrix, which is reflected by the lower tensile strength. After 30 minutes processing time, however, no significant improvement of the tensile strength is achieved by elongating this time.

Table 2. Tensile strengths of the Moso composites with the corresponding standard deviations (SD) in percent at different pressing/holding times as obtained at a pressure of 20 MPa and a temperature of 100°C.

<table>
<thead>
<tr>
<th>Time (press/hold)</th>
<th>5 / 5 min</th>
<th>15 / 15 min</th>
<th>30 / 30 min</th>
<th>60 / 60 min</th>
</tr>
</thead>
<tbody>
<tr>
<td>167 MPa</td>
<td>181 MPa</td>
<td>175 MPa</td>
<td>180 MPa</td>
<td></td>
</tr>
<tr>
<td>SD: 10 %</td>
<td>SD: 3 %</td>
<td>SD: 3 %</td>
<td>SD: 4 %</td>
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</table>

Important to point out is also the fact that in the hot press fabricated composites the intrinsic tensile strength of the bamboo strips is not only preserved but seems to be even enhanced. In other words, the given epoxy resin interacts with the fibers in such way that the formed polymer network is able to spatially distribute the locally applied tensile stress over the entire composite structure and, thus, enhance its load bearing capabilities. Further insight was provided by means of microscopic methods.

**Microstructure of the Bamboo Reinforced Composites.** Images of the traverse section and the top view of the composites fabricated at 20 MPa and 100°C are shown in Figure 4, as an example. Both images show the parallel alignment of strips and fibers and the lack of any voids or fiber contact that might contribute to a loss in composite strength.

The optical micrographs of the fractured surface of the composites produced at 100°C and 20 MPa shown in Figure 5b disclose that the surface of the composite with the highest tensile strength is rather smooth with a very homogenous layer of the epoxy resin. Hence, at this pressure and temperature the polymer network crystalizes as a very thin layer which homogenously interacts with the surface of the bamboo and improves the adhesion to the fibers. Moreover, the outer surface of the strips seems to be infiltrated with the resin due to this high pressure. Reducing the pressure to 15 MPa (Figure 5a) results in the formation of rather large resin crystals on the bamboo surface, which in turn reduces the adhesion to the fibers. In comparison with 20 MPa a pressure of 15 MPa appears too low for the epoxy to infiltrate the fibers. This is well in line with the reduced tensile properties of the composites.
shown in Figure 5a. Eventually, increasing the pressure to 25 MPa (Figure 5c) has two effects: On one hand, the polymer forms even larger crystals than those found at 15 MPa and on the other, the infiltration is inhibited by a propagated carbonization of the fibers. The latter is proposed by the significantly darker color of the composite and the strips. Similar, effects are responsible for the lower tensile strengths of the composites fabricated at 140°C and 120°C – see Figure 6. At 140°C the fibers and the resin become extremely brittle, which is represented in Figure 6a by the dark color of the fibers and the localized, large resin crystals. At 120°C (Figure 6b) both effects are present to a slightly lower extent.

Figure 5. Optical micrographs of the fractured surfaces of the composites produced at 100°C with pressures of 15 (a), 20 (b) and 25 (c) MPa at different magnifications.

Figure 6. Optical micrographs of the fractured surfaces of the composites produced with pressure of 20 MPa and a temperature of 140°C (a) and 120°C (b) at different magnifications.

FESEM micrographs of the lateral fractured surface in Figure 7 compare the composites produced at 20 and 15 MPa and 100°C. The 20 MPa sample shows a very smooth surface with a homogenous adhesion of the resin to the fibers. At the fracture position single fibers
are seen that have been pulled out of the resin. This indicates nearly perfect bonding between both and confirms the infiltration of the epoxy resin into the strips. On the other hand at 15 MPa – apart from an inhomogeneous crystallization behavior of the resin – a debonding of the polymer from the fibers is observed at higher magnification. This implies a lower degree of infiltration as compared with the higher-pressure samples.

**Figure 7. FESEM micrographs of the fractured surfaces of the composites produced at 100 ºC with pressures of 15 (a) and 20 (b) MPa at different magnifications.**

**CONCLUSIONS**

High-strength bamboo strip reinforced composite materials with a maximum tensile strength of ~ 180 MPa were fabricated using the hot press method. Processing conditions such as temperature, pressure and pressing/holding time, have been varied and evaluated for their impact on macroscopic mechanical properties of the composites and microscopic interactions at the interface between the resin and fiber surface.

We were able to show that processing the bamboo into strips provides a raw material in which the natural fiber alignment of bamboo is easily controlled throughout the fabrication of the composite. Moreover, the fabrication process was demonstrated to not only preserve the tensile strength of the raw material but also slightly enhance its properties in the composite. Importantly, within a specific temperature/pressure regime a maximum tensile strength was obtained, which was tunable by adjusting the pressure and temperature. Microscopic analysis provided insights into the crystallization behavior and infiltration of the polymer into the fibers, which strictly depends on the fiber-resin interface. In conjunction with the degree of carbonization these factors were proven to determine the ultimate tensile capacity of the resulting composites. Hence, understanding and subsequent optimization of the process parameters – especially at the microscopic level of the fiber-resin interfaces – is crucial for controlling and improving the mechanical properties to reach regimes that might in the future compete with tensile capacities of synthetic composites at significantly lower costs. However, the lack of uniformity of natural materials in comparison to synthetic products is certainly drawback for our composites as well and needs to be controlled. We are currently addressing this issue by optimizing the processing technique and carefully choosing the raw material.
REFERENCES


