

PREPARATION OF LIGNOCELLULOSE-BASED COMPOSITES

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Introduction

Lignocellulose-based composites are widely used in conventional applications (i.e., automobile components) because lignocellulose is abundant, lightweight, mechanically resistant, non-hazardous and relatively inexpensive [1]. Moreover, the use of lignocellulose in the form of residual products (e.g., sawdust) provides an incentive for recycling and renewing this resource with the consequent reduction in environmental impact and increase in life-span and added value. These composites are commonly prepared by using thermoplastic polymers of a low curing temperature [2]. This is because lignocellulose decomposes at temperatures below 350 °C [3]. The widespread use of these composites is limited, therefore, by the thermal stability of the lignocellulose. In order that lignocellulose-based composites can be used in applications where the material needs to operate above 350 °C, the composition of the lignocellulose must be modified to make it more resistant to temperature.

The present work deals with the preparation of lignocellulose-based composites using petroleum pitch as the matrix precursor and a curing temperature of 900 °C. The structural and mechanical properties of the composites are related to the pre-treatments applied to the precursors.

Experimental

A petroleum pitch (PP) and sawdust from Scandinavian pine wood (SD) were used as the matrix precursor and filler, respectively. In order to improve the structure and properties of composites, the petroleum pitch and the sawdust were treated under different conditions. PP was treated at 350 °C for 3 h in the presence of 10 wt.% of magnesium oxide (PP-MgO). On the other hand, SD was heated at 150 °C for 2 h with an 8 N solution of sulphuric acid and then washed (SD-H₂SO₄). The main properties of the composite precursors are summarized in Table 1.

The composites were prepared by mixing, molding and carbonizing to 900 °C blends consisting of 50:50 wt.% of pitch and sawdust, before and after treatment. By this procedure four series of composites were obtained: SD/PP, SD-H₂SO₄/PP, SD/PP-MgO and SD-H₂SO₄/PP-MgO. The materials were characterized in terms of microstructure (optical microscopy and SEM) and mechanical properties (four-point flexural strength).

Table 1. Characteristics of composite precursors.

| Sample | Treatment | Elemental Analysis (wt.%) | | | | | SP ¹ | CY ² |
|-----------------------------------|--|---------------------------|-----|-----|-----|------|-----------------|-----------------|
| | | C | H | N | S | O | | |
| SD | None | 51.2 | 6.0 | 0.4 | 0.0 | 42.4 | - | - |
| SD-H ₂ SO ₄ | H ₂ SO ₄ , 150 °C, 2 h | 64.0 | 1.9 | 0.4 | 0.2 | 33.5 | - | - |
| PP | None | 93.2 | 5.8 | 0.0 | 0.1 | 0.9 | 112 | 52 |
| PP-MgO | 350 °C, 3 h, 10 wt.% MgO | 94.4 | 5.3 | 0.0 | 0.0 | 0.3 | 288 | 88 |

¹ Softening point (Mettler, °C)

² Carbon yield (Alcan, wt.%)

Results and Discussion

The combination of untreated pitch (PP) with untreated sawdust (SD) results in a highly distorted composite (Figure 1a). The massive release of volatiles that come from both the petroleum pitch and the sawdust, generates a significant amount of porosity, which adversely affects the flexural strength of the composite (11 MPa). Moreover, pitch and sawdust react with each other during processing, giving rise to a totally isotropic material (Figure 2a).

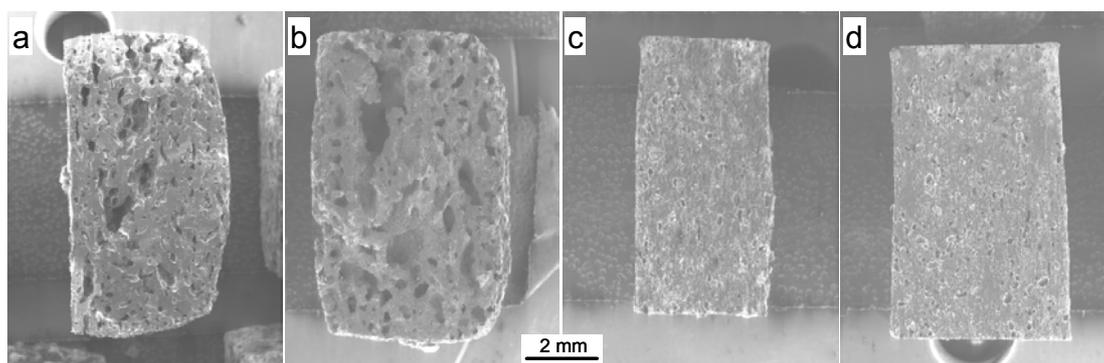


Figure 1. SEM images of (a) SD/PP, (b) SD-H₂SO₄/PP (c) SD/PP-MgO and (d) SD-H₂SO₄/PP-MgO composites.

Treatment with sulphuric acid causes dehydration (Table 1) and an increase in the thermal and structural stability of the sawdust. The composite prepared from treated sawdust (SD-H₂SO₄) and untreated pitch (PP) still shows a distorted, porous structure (Figure 1b). However, the pitch is able to progress, giving rise to an anisotropic matrix (Figure 2b, position 2A), while the sawdust preserves its honeycomb structure (Figure 2b, position B). These changes in optical texture are accompanied by a slight increase in flexural strength (15 MPa).

The treatment of the pitch with magnesium oxide causes polymerization/condensation reactions via dehydrogenative processes, producing an increase in the softening point and carbon yield of the pitch (Table 1). The combination of treated pitch (PP-MgO) with

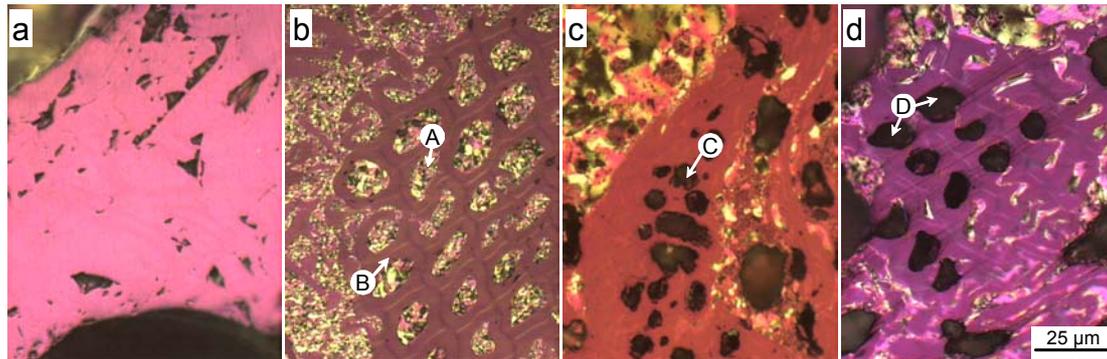


Figure 2. Optical micrographs of (a) SD/PP, (b) SD-H₂SO₄/PP (c) SD/PP-MgO and (d) SD-H₂SO₄/PP-MgO composites.

untreated sawdust (SD) gives rise to an undistorted composite (Figure 1c). Porosity is substantially reduced and the material undergoes an important volumetric contraction (Figure 1c). The treatment with magnesium oxide also produces a decrease in the reactivity of the pitch components, allowing the pitch to be transformed into an anisotropic matrix (Figure 2c). The honeycomb structure of the sawdust is partially destroyed in this case. This might be due to the volumetric contraction of the material. Moreover, the increase in pitch softening point reduces the ability of the pitch to penetrate into the sawdust channels. Consequently, empty channels can be observed in the composite (Figure 2c, position C).

The composite prepared from treated pitch (PP-MgO) and treated sawdust (SD-H₂SO₄) acquires its most positive characteristics from PP-MgO and SD-H₂SO₄ (Figures 1d and 2d). The carbon matrix is anisotropic while the sawdust preserves its cellular structure, despite the empty channels observed (Figure 2d, position D). The flexural strength of the composite increases to values of 30 MPa.

Conclusions

The combination of sawdust and petroleum pitch results in highly distorted composites with a low mechanical resistance. However, treatment of the sawdust with sulphuric acid and treatment of the pitch in the presence of magnesium oxide gives rise to composites with an anisotropic matrix that engulfs the honeycomb structure of the filler and improves the mechanical properties.

References

- [1] English B; Youngquist JA, Krzysik, AM. Lignocellulosic composites. In Gilbert RD, editor. Cellulosic Polymers, Blends and Composites, New York: Hanser Publishers, 1994:115-130.
- [2] Bledzki AK, Gassan J. Composites reinforced with cellulose based fibres. Prog. Polym. Sci. 1999;24:221-274.

[3] Álvarez P, Blanco C, Santamaría R, Granda M. Improvement of the thermal stability of lignocellulosic materials by treatment with sulphuric acid and magnesium oxide. *Journal of Analytical and Applied Pyrolysis*, submitted.