PROCESS FOR MANUFACTURING A HIGH PERFORMANCE NATURAL FIBER COMPOSITE BY SHEET MOLDING

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Abstract

In the past few years, natural fibers are finding an increased interest in polymer matrices. The natural fibers serve as reinforcement by enhancing the strength and stiffness to the resulting composite structure. In this study, a novel processing technique has been developed for water based thermoset polymers to prepare resin-impregnated mats, which can be used for sheet molding process to manufacture complex automotive semi-structural and structural parts. In order to optimize the curing conditions the mechanical properties of composites at different curing temperature and the crosslink density of the composites cured at different times were evaluated. The optimum curing cycle was obtained at 180 °C for 10 min. Composites with one and two layers of impregnated mat with 40 % resin and 60 % fiber were manufactured and their performance were evaluated. The mechanical properties of the cured pure resin and hemp fiber acrylic based composites with two different fiber lengths were measured and the effect of fiber content and fiber length were investigated. The flexural strength was found to be around 94 MPa and the flexural modulus was 14 GPa for the composite.

Introduction

In recent years, there has been a growing interest for the use of natural fibers in composite applications, especially in the automotive industry. These types of composites present many advantages compared to synthetic fiber reinforced plastics such as low tool wear, low density, cheap cost, availability and biodegradability. Although the mechanical properties of natural fibers are much lower than traditional fibers such as glass fiber, their specific properties, especially stiffness, are comparable to the values of those fibers [1,2].

Currently, different agricultural species like hemp, kenaf, flax, pineapple leaf, have been considered for use in the production of natural fiber composites and mechanical properties have been investigated. The mechanical properties of hemp fiber poly propylene composite using film stacking method were studied by Pervaiz et al. [3]. The results demonstrate that hemp-based natural fiber mat thermoplastics have comparable or even higher strength properties as compared with conventional flax-based thermoplastics. This study show that hemp based thermoplastic composites is good candidates for automotive applications.

Panthapulakkal et al. developed high performance injection molded hybrid composites using natural fibers along with glass fibers with thermoplastic polyolefins for automotive interior applications. The mechanical properties were found to be superior to the engineering plastics [4].

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In the thermoset resins, the fiber mats are impregnated with resins and then exposed to high temperature for curing. Resin Transfer Molding (RTM) technique is the most common method used for natural fiber thermoset composites. More recently vacuum assisted resin transfer molding was applied to manufacture composites based on plant oil thermoset resin and natural fibers [5]. These natural composites were found to have mechanical strength suitable for automotive applications. Moreover, Rouison et al. investigated the manufacture of hemp/kenaf and pure hemp fiber-unsaturated polyester composites and the effect of surface treatment of hemp fibers on the performance of the composite [6].

One of the major issues in development of composites is dispersion of the fibers into the matrix. The strong inter-fiber bonding, which keeps the fibers together, causes poor dispersion of the fibers which lead to inferior performance of the composite. In this work, resin impregnation of fiber has been investigated with especial emphasis to improve stiffness property development.

Experimental

Materials

In this study, a commercial water-based acrylic resin (viscosity at 23 °C = 400-4000 mP.s), an environmentally friendly binder for natural fiber composites, was obtained from a commercial source. It contains polycarboxylic acid with a polyhydric alcohol as a crosslinking agent. Hemp fiber used in this work was kindly supplied by the Hempline Company.

Resin Impregnation and Sheet Molding Process

In one of our earlier patent application, hemp fibers were randomly oriented in a perforated screen and the acrylic resin solution was circulated to impregnate the fibers with the solution [US pat. APPL. No. 60/562,444].

The wet mat was dried in the oven to remove all the moisture content and to complete thickening process. After drying cycle, the impregnated mat was cured at 175 °C, 180 °C, 185 °C for different curing times from 2 to 12 minutes using a hydraulic press with 1500psi pressure. In order to prevent any blister formation due to any moisture content, the composite was cooled inside the press to around 70 °C using water cooling system. Some composites were prepared using 2 layers of impregnated mat instead of using 1 layer. Also, two different fiber lengths (2.5 cm and 9 cm) were used to investigate the effect of fiber length on mechanical properties. The final composites were prepared for mechanical properties measurements.

Crosslink Density Measurements

The crosslink density of the matrix of the hemp fiber acrylic based composite was measured using the Flory-Rehner equation. The samples of approximately 1 cm square shape and 2 mm thickness were cut, weighed accurately. Then they were kept in a suitable solvent at room temperature to swell for 24 hr. After reaching equilibrium, samples were removed and the solvent adhered to the surface was wiped out and weighted. The weight of the samples after complete drying was measures as well. Volume fraction of resin (v) was then calculates using the equation:

$$\upsilon = \frac{(D - FT)\rho_r^{-1}}{(D - FT)\rho_r^{-1} + A_0\rho_s^{-1}} , \qquad (1)$$

Where *T* is the initial weight of the sample, *D* is the de-swollen weight of the samples, *F* is the weight fraction of insoluble components, ρ_r is the density of the resin, A_0 is the weight of the solvent absorbed by the sample and ρ_s is the density of the solvent.

Mechanical Properties

The tensile properties of the composites and pure cured resin were measured following the ASTM standard method D-638. The flexural properties were obtained according to the ASTM standard method D-790. Also the ASTM D-256 was applied to measure the notched impact properties of samples.

Results and Discussion

In the previous study, the optimum cure conditions for pure acrylic resin were studied [7]. Temperature and time are the most important parameters to select in a cure cycle. It was found that the resin cured at 180 °C for 10 min had highest crosslink density. In order to make sure that these conditions are optimum parameters to manufacture the composite with the same thickness, the mechanical properties of the composites cured at different curing temperatures 175 °C, 180 °C, 185 °C for 12 min were evaluated to obtain the optimum curing temperature.

Figure1 and 2 show the effect of cure temperature on the performance of the composite. As it can be noticed, the composite cured at 180 °C has superior mechanical properties compare with others. That means at this temperature, strong adhesion between fiber and matrix due to higher crosslinking resulted in higher shear stress strength at the fiber/matrix interface. Stronger force must be used to overcome the shear strength interface which resulted in higher tensile strength. Moreover, higher crosslinking of matrix support to transfer load from matrix to the fiber which has a pronounced effect on the flexural properties.

To optimize the curing time for the composite the crosslink density of the composites cured at 180 °C for different curing times were evaluated. Crosslink density is a quantitative measure of the number of crosslinks that exist in a given volume in the thermosetting polymer. A higher crosslink density means more dimensional stability and high mechanical properties. According to the theory of Flory and Rehner, for a perfect network, crosslink density v_x is calculated by using the following equation:

$$\upsilon_x = -\frac{\ln(1-\upsilon) + \upsilon + \chi_1 \upsilon^2}{\varphi_1 \left(\upsilon^{1/3} - \upsilon/2\right)} , \qquad (2)$$

Where v_x is the number of moles of elastically effective network chains per cubic centimeter, v is the volume fraction of the polymer in swollen matrix, φ_1 is the molar volume of the solvent and χ_1 is the Flory-Huggins interaction parameter between solvent and polymer. Using χ_1 equal to 0.498 from the literature, data for volume fraction of swollen sample v, crosslink density v_x of the composites cured at different curing times between 0 min to 12 min at 180 °C were calculated an illustrated in figure 3. It is clear that the curing time has a significant effect on the crosslink density of the cured sample. The crosslink density of the matrix increases from 2.93×10^{-3} to 6.9×10^{-3} mol ml⁻¹ with increasing the curing time from 2 min to 10 min at 180 °C. It can be noticed that the rate of increasing of the crosslink density decreased after 5 min. Also, as the curing time increased from 10 min to 12 min the crosslink density decreased from 6.9×10^{-3} to 4.32×10^{-3} mol ml⁻¹, which can be due to decomposition of the resin. Therefore, the optimum curing time was considered to be 10 min for manufacturing the composite.

In order to evaluate the resin impregnation through the mat, the mechanical properties of the composites which prepared by one layer of impregnated mat was compared with the composite which prepared with two layer mats with the same quantity of raw materials. Both composites with the final thickness of 2.2 mm have 40 % resin and 60 % fiber. Table I indicates the effect of number of layers on the mechanical properties of the composites.

The results show that the composite, which prepared by two layers of impregnated mat has higher strength properties compare with another one. It can be concluded that the resin solution can impregnated through the fiber uniformly with less amount of fiber content. As the amount and thickness of the mat increased it will be more difficult to wet the entire surface of the fibers. Besides, figure 4a and 4b show the SEM micrographs of the fractured surface of the two composites with 2.2 mm thickness manufactured with 40 % resin and 60 % fiber, one prepared by one layer of impregnated mat and the other one with two layers of mat. Figure 4a shows the presences of fiber pull out due to lack of resin impregnation. It is obvious from figure 4b that the interfacial adhesiveness of the two layers composite is much better than that of the one layer composite. When stress is applied the fibers broke due to stronger adhesion between fiber and matrix.

The mechanical properties of cured pure resin were evaluated and the effect of fibers on the performance of the polymer was shown in figure 5, 6 and 7. As it can be noticed adding 40 % fiber to the pure resin has a pronounced effect on the mechanical properties of the resin. When pure resin stressed, random flaws in physical structure of the resin will cause the material to crack and fail. Introducing the fibers to the resin will overcome this problem and reinforce the material.

In order to investigate the effect of fiber length on the performance of the composite, two different fiber lengths were selected (2.5 cm and 9 cm) and the composites were manufactured with two layers of impregnated mat at 180 for 10 min with 40 % resin and 60 % fiber with 2.2 mm thickness. For the same amount of fiber content, as the fiber length increases the number of stress concentrating fiber ends decreases which assist to transfer load from matrix to the fiber and thus contributes toward the entire composite and consequently improvement in mechanical properties.

Conclusion

In this work, a new environmentally friendly acrylic resin was used to manufacture hemp fiber composite using resin impregnation and sheet molding process. The effect of curing temperature and time on the performance and crosslink density of the composite were investigated to optimize the cure cycle. It was found that the composite cured at 180°C for 10 min has highest amount of crosslinking and consequently better performance. The composites were prepared using one layer of impregnated mat and two layers. The effect of number of layers on mechanical properties of the composite was studied.

Moreover, the effect of fiber on the performance of the cured pure resin and the effect of fiber length on mechanical properties of the composite was investigated. Increasing the fiber length leads to an improvement in mechanical properties of the composite.

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Figure1: Effect of curing temperature on tensile and flexural strength



Figure2: Effect of curing temperature on tensile and flexural modulus



Figure 3: Effect of curing time on the crosslink density of the composite





Figure 4: SEM micrographs for hemp fiber acrylic composites manufactured with different layers mat. a) One layer and b) Two layers



Figure 5: Tensile and flexural strength of pure resin and hemp composites with different fiber lengths



Figure 6: Tensile and flexural modulus of pure resin and hemp composites with different fiber lengths



Figure 7: Impact strength of pure resin and hemp fiber composite with different fiber lengths

Performance property	Composite with one layer	Composite with two layer
Tensile strength (MPa)	26.6 ± 5	33.5 ± 7
Tensile modulus (GPa)	5.1 ± 0.67	5.3 ± 0.58
Flexural strength (MPa)	70.6 ± 14	79 ± 11
Flexural modulus (GPa)	11.6 ± 2	12.3 ± 1.35
Impact Strength (KJ/m ²)	4.2 ± 0.97	3.7 ± 0.46