Effect of Sodium Silicate on the Dimensional Stability and Mechanical Behaviour of Non-Woven Flax Reinforced Acrylic Based Polyester Composites

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ABSTRACT

This paper investigated the effects of Sodium Silicate on the dimensional stability and mechanical behaviour of non-woven flax reinforced acrylic based polyester composites (NWFRAC) with 10, 20 and 30 wt% sodium silicate as filler which have been fabricated using the impregnation method. Water absorption tests were conducted by immersing specimens in a deionized water bath at 23 °C for different time durations. The tensile and flexural were investigated before and after water absorption. The percentage of moisture uptake increased as the increasing of sodium silicate content. The tensile and flexural properties of NWFRAC were found to decrease with increase in water uptake. The phase morphology of non-woven flax reinforced acrylic based polyester composites were investigated by using confocal laser microscope model Zeiss LFM 700 MATSEM. Higher sodium silicate content exhibits an irregular structure with many air gaps.

Keywords: water absorption, mechanical properties, polyester composites.

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Introduction

Moving toward 21 century, the awareness toward green and sustainable product and technologies have increase significantly especially in construction, packaging and automotive due to the environmental legislation as well as consumer demand have forced manufacturing industries to seek for a new materials which can replace conventional and synthetic nonrenewable reinforcing materials such as glass and aramid fibre. The advantages of Natural fibers as a reinforcement for composites over the synthetic fibre are high specific strength to density ratio, lighter, cheaper, biodegradable, good acoustic properties. Furthermore, it's easy to process and have good life cycle.

Flax (Linum usitatissimum) fibres is one of the most widely utilised natural fibres especially in polymer composites since they present good mechanical properties (Young's modulus of 12–85 GPa, for instance), low density and non-abrasive qualities [1–4]. Besides that, flax is also a natural renewable resource cultivable in most of tempered areas such as in Europe. However, the high hydrophilic properties, high flammability and poor resistance toward water absorption of flax fibres are nonetheless a main drawback that limits their application in many fields. In order to solve this problem, adhesion between the fiber and the matrix for both thermoplastic and thermoset compounds need can be enhance using chemical modifications of the flax fibres [10–16], functionalisation of the polymer matrix or addition of coupling agents like silane.

Sodium silicates belong to the family of soluble silicates. Soluble silicate (sodium silicate) is a viscous liquid with about 21 to 34 wt.-% SiO_2 and 6 to 18 wt.-% Na_2O [5–7]. Sodium silicate reacts in aqueous solutions to polysilicates. The aim of this studies was to study the water absorption and mechanical behaviour of flax reinforced acrylic based polyester composites with sodium silicates using impregnation method.

Experimental Procedure

Materials

The matrix material used in this study was acrylic based polyester resin, trade name "Acrodur 950L" supplied by BASF. Non-woven flax fibre (FF) mat with areal density of 1300 g/m² was used as the reinforcement supplied by EcoTechnilin, France. Table 1 exhibits the physical properties of Acrodur 950L resins. Sodium silicates (SS) were supplied by Sigma Aldrich with a density of 1.39 g/ml.

| Table 1: Properties of Acrodur 950L | | | | |
|-------------------------------------|------------------------------|-------------------|-----|--|
| Solid content (%) | Density (g/cm ³) | Viscosity (mPa.s) | рН | |
| 50 | 1.20 | 900 - 2500 | 3.5 | |

Composite preparation

The processing begins with drying the non-woven FF mats at $60 \pm 5^{\circ}$ C for 3h in an air circulating oven. Then, the mats were impregnated with various matrix systems consisting of 10, 20 and 30 wt% sodium silicate (Table 2) using an impregnation process as shown in Figure 1. The obtained prepregs were then dried in a vacuum oven at 70°C for 1h. The prepreg were press molded in a conventional hot press at 210°C for 270 sec at 10 bar. After curing, the matrix content $M_{matrix}(\%)$ is determined using the following equation:

$$M_{\rm fibre} = \frac{M_{\rm flax}}{M_{\rm composite}} \tag{1}$$

$$M_{\text{matrix}}(\%) = \frac{(M_{\text{composite}} - M_{\text{fibre}})}{M_{\text{composite}}} \times 100$$
(2)

Where M_{matrix} (%) is the matrix content as percentage, M_{composite} is the mass of composite, M_{flax} is the mass FF mat and M_{fibre} is the mass of fibre.

| Composite sample code | Acrodur Resin (wt%) | Sodium silicate (wt%) | Denotation |
|-----------------------------|---------------------------|-----------------------------|----------------|
| A100 | 100 | 0 | A 100% + SS 0% |
| A90S10 | 90 | 10 | A 90% + SS 10% |
| A80S20 | 80 | 20 | A 80% + SS 20% |
| A70S30 | 70 | 30 | A 70% + SS 30% |
| S100 | 0 | 100 | A 0% + SS100% |

Table 2. NWED & C formal atter

Water absorption tests

Water absorption and thickness swelling tests were conducted in accordance with ASTM D570-98, in which the specimens were immersed in water for 2 h and 24 h at a temperature 23 ± 1 ⁰C. The weight gain and thickness increase were then measured 20 min after being removed from the water. The water uptake was evaluated using the following equation:

$$M_m(\%) = \frac{(M_t - M_o)}{M_o} \ge 100$$
(3)

Where M_m (%) is the water uptake as a percentage, M_t is the weight of the wet sample at a time (t), and M_o is the initial weight of the sample. In the water immersion tests, thickness of each specimen was also measured for determination of the thickness swelling (TS) by using the following equation:

$$TS(\%) = \frac{(\delta_i - \delta_t)}{\delta_t} \ge 100$$
(4)

In which δ_i and δ_t are the panel thickness after and before the water immersion, respectively.



Figure 1: Schematic diagram of impregnation system

Mechanical properties

Tensile test

The tensile tests are carried out according to ASTM D3039 by using INSTRON 3366 Universal Testing Machine. The tensile strength and tensile modulus were measured at a crosshead speed 0f 2 mm/min. Gauge length was measured 25 mm. The tests are performed at temperature of $23^{\circ}C \pm 2^{\circ}C$ and relative humidity was $50\% \pm 5\%$.

Flexural test

The flexural strength and modulus composites are measured according to ASTM D790 by using INSTRON 3366 Universal Testing Machine at a crosshead speed of 2 mm/min, 5 kN load cell and 48 mm in span length.

Thermal and Morphological Studies

Flax composite samples were characterized for their thermal properties which is the degree of cross linking using Differential scanning calorimeter (DSC) from Perkin Elmer. A comparison of curing enthalpy between uncured resin and cured matrix composites was made to determine the degree of crosslinking of the required sample. The heating rate used was 10°C/min. Surface appearance of NWFRAC with sodium silicate content was obtained using confocal laser microscope model Zeiss LFM 700 MAT.

Results and discussion

The composite's matrix content are presented in Table 3 with respect to various sodium silicate content in the matrix system. Our data demonstrated that the matrix content in the composite decreases with the increasing of sodium silicate content. This trend can be presumably explained by an interference of polyester formation between the reaction of polycarboxylic acid and the polyalcohol with a reaction of one or even both matrix components with sodium silicate preventing the generation of the polyester system.

| Composite sample | Sodium silicate (%) | Composite's Matrix Content (%) | |
|------------------|---------------------|-----------------------------------|--|
| A100 | 0 | 51 ± 3 | |
| A90S10 | 10 | 49 ± 2 | |
| A80S20 | 20 | 48 ± 2 | |
| A70S30 | 30 | 47 ± 3 | |
| S100 | 100 | 42 ± 2 | |

Table 3: Composite's matrix content of different sodium silicate contents.

Moisture absorption and thickness swelling

Results of composite density, moisture absorption test and thickness swelling are given in Table 4. It is found that the density of the composites ranges

from 0.809 kg/m³ for the 100A and 0.809 to 0.968 kg/m³ for the A90S10. A80S20 and A70S30. The composites with sodium silicate have lower density and higher void content as compared to composites with no sodium silicate which indicate the dispersion of the matrix reduce with the increase of sodium silicate content due to the reduction of matrix quantity and higher compound viscosity as mention above. The water absorption shows a clearly dependence of the sodium silicate contents in the compound and it increases with increasing sodium silicate fraction in the matrix system. While composites with just thermoset matrix show a water absorption value of approximately 24.95%, the compounds with sodium silicate exhibit water absorption up to 46.55%. Press plates that were impregnated just with sodium silicate show water absorption values of 54.81%.

| Table 4: Water absorption and thickness swelling of NWFRAC. | | | | | | |
|---|------------|---------|-------|--------|--------|--------|
| Composite | Density | Void | Wa | ater | Thic | kness |
| sample | (kg/m^3) | content | Uptak | te (%) | swelli | ng (%) |
| code | | | 2h | 24h | 2h | 24h |
| A100 | 1.098 | 9 | 6.91 | 24.95 | 4.16 | 15.08 |
| A90S10 | 0.8098 | 12 | 8.42 | 26.24 | 4.52 | 18.40 |
| A80S20 | 0.9706 | 21 | 18.19 | 34.81 | 11.26 | 21.91 |
| A70S30 | 0.9682 | 22 | 21.42 | 46.55 | 30.83 | 49.37 |
| S100 | 1.2985 | 10 | 38.19 | 54.81 | 39.16 | 50.08 |

The thickness swelling (TS) of the NWFRAC increases with the water absorption and thus has similar trend to the water absorption regarding the sodium silicate contents (Table 4). The TS values for the 2 h water immersion vary from 4.16% to 30.83%, and these values are increased after 24 h immersion, varying from 15.08% to 49.37%.

Tensile properties

The impact of sodium silicate content on the tensile strength and tensile modulus of the composites is shown in Figures 2 and 3 respectively. It is observed that tensile strength of all specimens decreased with increasing of sodium silicate content and the same trend also seen after immersed for 24 hours sample (Figure 2). These phenomena due to the water penetration inside polymers decreasing the connection between fiber and polymer material. Furthermore, flax fibers in the composite had many channels and capillary tube which allowed for water molecules to penetrate inside the materials and acting along the interface between polyester and flax fiber causing swelling in the samples. These reactions will cause the bonds between resin and fibers will break. So the strength of the composite material will decrease. Medina et al. [8] has reported similar results in their research for hemp/polyester composites. The tensile stress drops by 75% in their study. The tensile modulus for the composites with sodium silicate ranges from 5.4 to 6.0 GPa (Figure 3), which is much less variable than the tensile strength values. Composite of 30 wt% sodium silicate exhibits the lowest tensile modulus. The results also show that the composites with 100 wt% Acrodur have highest modulus.



Figure 2: Dependence of the tensile strength of the composites with the sodium silicate content.



Figure 3: Dependence of the tensile modulus of the composites with the sodium silicate content.

Flexural properties

The characterizations of flexural properties of the composites were carried out and the outcomes are shown in Figure 4 and 5. It can be seen that the flexural strength and modulus decrease by increasing the sodium silicate content in the matrix system. The flexural strength exhibits a similar trend to the tensile strength. The composites with 10 to 30 wt.% Sodium silicate has a flexural strength varying from 72.22 to 89.52 MPa whereas those made of 100 wt. % Acrodur have the corresponding value 86.88 MPa. Similarly, the flexural modulus for the composites varies from 6.4 to 7.2 GPa.



Figure 4: Dependence of the flexural strength of the composites with the sodium silicate content



Figure 5: Dependence of the flexural modulus of the composites with the sodium silicate content

In order to determine the degree of crosslinking of the matrix with respect to various sodium silicate content, DSC analysis was conducted to detect the enthalpy of residual crosslinking capability (Δ H residual) in the

composites samples and infer the degree of crosslinking by comparing it to the enthalpy of crosslinking reaction (Δ H resin) of an uncured Acrodur resin. In order to obtain DSC accurate result, the uncured Acrodur resin was first dried under vacuum oven at 50 °C for 3h to eliminate excess moisture without initiating the crosslinking reaction. A typical DSC scan of NWFRAC at 10°C/min is shown in Figure 7. The peak in the range of 25-130°C corresponding to the evaporation of residual moisture within the composite while the second peak at 200°C can be corresponded to the post-curing of the Acrodur matrix with a reaction enthalpy of Δ H_{Acrodur} = 140 J/g. The typical DSC scans of the NWFRAC at different sodium silicate content of 0, 10, 20 and 30% are shown in Figure 7 (b-e).

| Table 5: Degree of crosslinking of NWFRAC | | | |
|---|-----------------------------|----------------------------|--|
| Composites | $\Delta H_{residual} (J/g)$ | Degree of Crosslinking (%) | |
| A100 | 18.73 | 86.62 | |
| A90S10 | 30.41 | 78.28 | |
| A80S20 | 60.50 | 56.79 | |
| A70S30 | 78.29 | 44.08 | |

The degree of crosslinking values for the composites with the variation in sodium silicate content is presented in Table 5. These results show that the degree of crosslinking was found to decrease with the increment of sodium silicate content due to the presence of sodium silicate possibly interferes with the curing process. Figure 8 shows the microstructure analysis of the composite surfaces using confocal microscope. By increasing the sodium silicate content in the matrix system, an open surface which is not covered by matrix can be observed on the composite surface. Furthermore, a composite without sodium silicate (A100) exhibit even and homogenous surface. Figure 8c) and 8d) shows the appearance of air gap on the composite surface which result in the reduction of fibre matrix bonding as well as properties of the natural composite.



Figure 7: DSC curve for uncured Acrodur resin and composites (a) Uncured Acrodur (b) A100 (c) A90S10 (d) A80S20 and (e) A70S30.



Figure 8: Composites surfaces (a) A100 (b) A90S10 (c) A80S20 and (d) A70S30

Conclusion

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Within this research work sodium silicate was used as environmentally friendly inorganic additive in a thermoset matrix system. The water absorption and thickness swelling of the composite increase with the sodium silicate contents. The mechanical properties of the NWFRAC also decreased with increasing of sodium silicate content due to the chemical incompatibility of both systems in which the interference of sodium silicate on the curing reaction of the polymer avoids the generation of the polyester.

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