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The effect of viscosities of various coating solutions on the physical, mechanical and morphological properties of kenaf/epoxy composites

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ABSTRACT

Coating treatment is a new method to improve the overall properties of natural fibres. However, recent research paying less attention to the effects of coating solution viscosities towards the coating behaviour inside the fibres embedded in the composite system. Thus, this study explores the effects of coating domination to the deformation of the composite from the analysis of coating uptake, maximum tensile modulus and the Poisson's ratio. The samples were prepared by dipping the fibres in 1:4, 1:5 and 1:6 coating solutions, cured and pulverised prior moulded and tensile tested. The result shows that 1:6 coated fibre is the best since it improves about 40.1 % of coating uptake, 23.8-31.4 % of maximum tensile modulus with highest Poisson's ratio of composites properties. It also supported by its scanning electron micrograph as better fibre-matrix interaction was observed. The high coating penetration in the structure of the fibre might change the fibres natural polarity and improves the fibre-matrix interaction to reduce the deformation of the composite to give a stiff composites property. Conclusively, the optimum coating solution is needed since it influences the fibres coating behaviour to control the deformation of the composites.

Keywords: *kenaf fibre; coating solution; viscosity; physical properties; mechanical properties; morphological properties*

1. INTRODUCTION

Recently, the usage of plant-based fibres in load-bearing applications become a focus by most of the industrialist and engineers. These fibres are able to give a low impact on the environment in terms of safety in disposal, low in energy, toxicity and the carbon dioxide emission. Moreover, the plant-based materials also were chosen because of the cost aspect as well as due to their performance that consists of density, strength, and stiffness. However, despite its merits, its demerit also was considered since the said fibres have moderate practicality in hot and wet conditions. But, still, it has excellent durability to water as compared to the mineral-based fibre [1].

Nevertheless, the strength performance of plant-based products still inconsistent and it considered not fully optimised. Since the fibre naturally have variation in physical, chemical and morphological properties that influence by its natural growth (E.g. water, sunlight and minerals) and processing parameters (E.g. parts of harvested plants and materials retting process). The morphological properties getting worsen since the shape of mechanically cut fibres are variable since it depends on the types of milling technique as discovered by Legras [2]. Besides, it is becoming complicated as the shapes of fibre are getting varied after the pulverisation process that used knife-type mill [3].

Thus, a remedial new coating treatment was suggested to the fibre to reduce the effects of the variability of the fibres shapes. In general, the coating technique is a natural fibres treatment that uses diluted resin to penetrate the fibres system at a specific immersion time. Related research was conducted by Ghaztar, Ibrahim [4] shows that the coating treatment was able to reduce the variability of the fibres shape. In theory, natural fibres with fewer shape irregularities will give consistent and predictable

properties that are needed to design a product. The design of a product highly relies on the specific mechanical properties that will affect the weight and; this is crucial especially to be applied in the automotive applications.
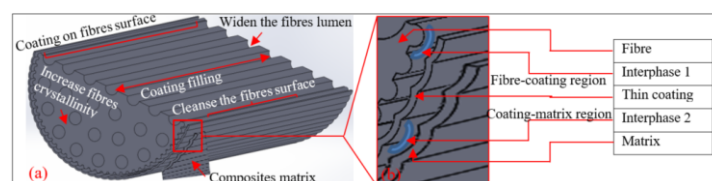

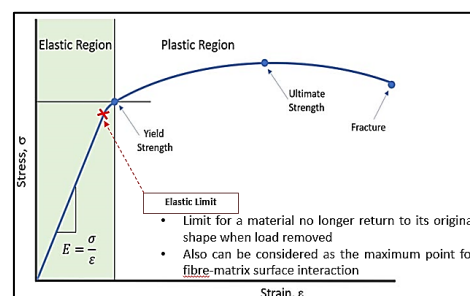**Figure 1.** The (a) in-situ coating mechanism and (b) overall features of the coated fibres.

Figure 2. The maximum elastic limit point.

The coating method was used not just to control the shape of the coated fibres but, to improve the surface interaction of the fibres. The coating creates a layer of protection to the fibre at all of its aspect ratios. Thus, after fabricated in the composite system, the coating creates two (2) new interphases compare to the uncoated composite system that has one (1) interphase. Refer to Figure 1 for features in coated fibre. In theory, the viscosity plays

a role in determining the thickness of the coating formation. An effective coating will penetrate and strongly adhere to the surface of the fibre.

In addition, the maximum elastic limits point for each composite were taken since in theory, it indicates the maximum fibre-matrix interfacial adhesion before microcracks generated [5, 6]. Refer to Figure 2 the maximum elastic limit. The Poisson's ratio of the composites also was taken as it analyses the lateral and axial strain deformation during a tensile force being applied.

2. MATERIALS AND METHODS

2.1. Materials.

The raw kenaf natural fibre was obtained by the Kenaf and Tobacco Board. The received fibre was in the continuous form (more than 30 cm in length). The epoxy resin (CP362A) and hardener (CP362B) was manufactured by Oriental Option Sdn. Bhd. The physical and chemical characteristics of epoxy resin and hardener are tabulated in Table 1. Acetone was supplied by Classic Chemicals Sdn. Bhd. with 95% purity with the molecular weight of 58.08 g/mol. The viscosities of the diluted 1:4, 1:5 and 1:6 epoxies coating solutions are 6.4, 6.0 and 5.6 cps respectively.

Table 1. The curing characteristics of CP 362 epoxy resin.

Part	Viscosity (cps)	Colour	Gel Time	Post Cure	Final Viscosity (cps)
Epoxy (CP362A) DGEBA	13 000	Transparent			8500
Hardener (CP362B) Modified Polyamine	400	Transparent	35 Minutes (25°C)	9.5 Hours (25°C)	

Source: The physical characteristics of CP 362 epoxy resin [4, 7].

2.2. Fibre coating uptake test.

The Kenaf fibres were combed prior conditioned at 24 °C for 48 hours. Then, the fibre bundles with an average weight of 0.04 gram and length of 5 cm each were soaked in 1:4, 1:5 and 1:6 coating solutions separately at 25 °C. The soaking time was varied from 0.17 minute to 5 minutes. The dilution and coating process were conducted in a controlled condition due to the high evaporation rate of acetone.

Upon completing the soaking process, the fibres were cured in an air circulated oven at 80 °C for 24 hours. Each fibre that cured in each coating solution and immersion time was weighed to measure the coating uptake by the fibres. The coating uptake (CU) was calculated using Equation (1).

$$Coating\ Uptake, CU\ (\%) = \frac{W_i - W_o}{W_o}\ x\ 100\% \qquad (Eqn.\ 1)$$

Where W_i and W_o is the weight after and before the coating process respectively.

2.3. Fibre treatment process.

The Kenaf fibres were combed prior conditioned at 24 °C for 48 hours. The coating solutions were prepared by diluting the epoxy solution with acetone at 1:4, 1:5 and 1:6 w/w epoxy to acetone ratios. The fibres were immersed instantly after prepared using 3:7 fibre to coating ratio at room temperature. The processes were conducted in a controlled condition to prevent evaporation of acetone. After 5 minutes of soaking process, the fibres were tossed and individually separated to promote a single fibre coating during the curing process at 80 °C for 24 hours in an air circulated oven.

The untreated and chemically treated fibre batches become control 1 and 2 samples respectively. The fibres were combed and cleaned using water and dried prior treatments. For the first treatment, the fibres were immersed in 6 w/v % of NaOH-water solution with 7:3 immersion ratio at 25 °C. The fibres were washed and dried at 70 °C at 96 hours. The fibres were treated again with 1 (v/v %) of APS, 95 (v/v %) of ethanol, 4 (%) water for 30 minutes and air-dried for 1 hour.

2.4. Pulverisation process.

All fibres were cut using a pulveriser machine (Fritsch Power Cutting Mill PULVERISETTE 15) passing through 5.0 mm, 1.0 mm, 0.5 mm until 0.25 mm pulverise mesh size aperture in consecutively. The fibre was added gradually and collected in receiver bowl.

2.5. Fabrication process.

The composite was prepared by mixing the fibres with epoxy at 50 °C on a hotplate for 30 minutes. Prior mixed with the hardener for 5 minutes. The fibres were varied by 10, 20, 30, 40 and 50 (w/w %) fibre loadings. The samples coding is in Table 2.

Table 2. The Samples Coding for the Various Kenaf Fibres/ Epoxy Composites.

Fibre Loadings (wt %)	Untreated Samples (Control 1)	Epoxy Coated Samples			NaOH/ APS Treated Samples (Control 2)
		1:4 Ratio	1:5 Ratio	1:6 Ratio	
10 %	Unt 10%	Ctd 1:4 10%	Ctd 1:5 10%	Ctd 1:6 10%	Trtd 10%
20 %	Unt 20%	Ctd 1:4 20%	Ctd 1:5 20%	Ctd 1:6 20%	Trtd 20%
30 %	Unt 30%	Ctd 1:4 30%	Ctd 1:5 30%	Ctd 1:6 30%	Trtd 30%
40 %	Unt 40%	Ctd 1:4 40%	Ctd 1:5 40%	Ctd 1:6 40%	Trtd 40%
50 %	Unt 50%	Ctd 1:4 50%	Ctd 1:5 50%	Ctd 1:6 50%	Trtd 50%

Then, the fibres were poured into the mould and pressed using the hydraulic hot press at 1200 psi at 50 °C for 2 hours. Afterwards, the mould was post-cured in an air circulated oven at 50 °C for 24 hours.

2.6. Testing and characterization.

All the samples were tested for a tensile test. The fractured surface of tested samples was further analyzed using a scanning electron microscope (SEM).

2.7. Tensile test.

The tensile test was conducted as in ASTM D3039/3039M-08 [8] by Shimadzu Universal Testing machine AG-X series. The conditioned samples (15 cm length by 2 cm

Thus, this study explored the physical, mechanical and morphological properties of various fibres that cured using various coating solution viscosities. The ideal coating solution would be able to penetrate the fibres system to change the polarity of the fibre and suppressing the formation of the voids to form a compact composite system. It improves the fibre-matrix surface interaction as well to make the bio-composites able to perform at its optimum properties specifically in automotive applications.

width by 0.3 cm thickness) that coupled with an extensometer was tested with speed of 5 mm/min. In addition, 8 mm gauge length and 3.5 cm grip length for the end of each sample was set for all tested samples. The tensile Poisson's ratio can be calculated as in Equation (2) to (4).

$$\varepsilon_{Lat} = \frac{d_1 - d_o}{d_o} \qquad \text{(Eqn. 2)}$$

Where, ε_{Lat} is the composites lateral strain, d_1 is the width of the samples after deformation (mm), d_o is the width of the initial samples (mm).

$$\varepsilon_{Long} = \frac{L_1 - L_o}{L_o} \qquad \text{(Eqn. 3)}$$

Where, ε_{Long} is the tensile strain, L_1 is the length of the samples after deformation (mm) and L_o is the length of the initial samples (mm).

$$\nu = \frac{\varepsilon_{Lat}}{\varepsilon_{Long}} \qquad \text{(Eqn. 4)}$$

Where, ν is the Poisson's ratio, ε_{Lat} is the composites lateral strain and ε_{Long} is the composites longitudinal strain.

2.8. Surface morphology characterisation.

The morphology of tensile samples fractured surfaces was observed by using a Scanning Electron Microscope (SEM) (Hitachi TM3030 Plus, USA) at 5.0 kV acceleration voltage.

3. RESULTS
3.1. Fibre coating mechanism.

In general, the coating process happened as the epoxies molecule moves towards the fibre during the initial immersion process. With time, the epoxies gradually pull along its neighbouring chains and stick onto the surface of the fibre. The effect of gravity is not as effective as the coating build-up is still small. By prolonging the immersion time, the formation of the coating layer reaches the critical mass resulting to the possible gravity to take effect and pulling-down the mass [9]. This will affect the coating thickness deterioration which random resulting in a non-linear correlation between coating thickness and time.

Overall there are three (3) coating mechanisms proposed in this research as the fibres being coated and cured in different coating viscosities and immersion time. The first mechanism, due to the fast immersion, the coating formed only on the fibres surface. The second mechanism, as the immersion time extended, the coating formed on the fibres surface and starting to leach the fibres constituents. The third mechanism, at the optimum immersion time, the fibre continuously leached its constituent and the coating form on its surface and penetrated into its system. Refer Figure 3 for illustration of all coating mechanisms.
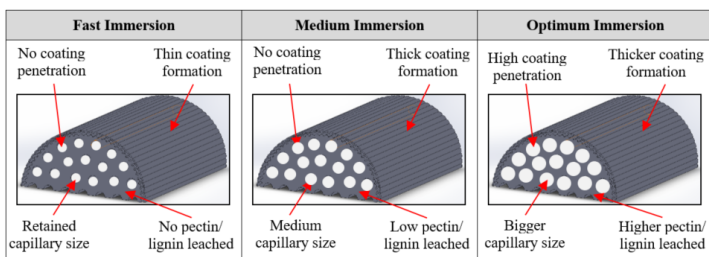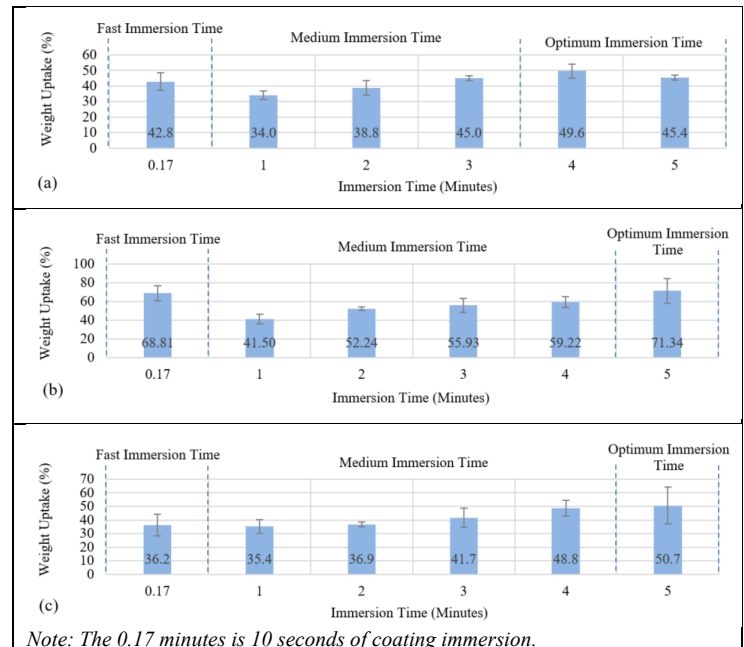

Figure 3. Illustration on the possible coating mechanism occurred during fast, medium and optimum immersion time.

3.2. Fibre coating uptake analysis.

Figure 4 shows the coating uptake for cured coated fibres in 1:4, 1:5 and 1:6 coating solutions. In general, an increasing trend also was observed by prolonging the immersion periods for all coating solutions. It might be due to the effect of epoxies coating uptake as the coating adhere more onto the fibres once it cured. It forms a thin layer of coating and penetrates the fibres structure as well to fill the fibres capillary.

All the coated fibres experience reduction of coating uptake after 1 minute of coating immersion for all coating solutions. The sample reduces about 20.56%, 39.69% and 2.2% compared to the

0.17 minute sample for each 1:4, 1:5 and 1:6 coating solutions respectively. It might be due to the effect of acetone since it cleansed and leached pectin, lignin and hemicellulose from the fibres system.

Note: The 0.17 minutes is 10 seconds of coating immersion.

Figure 4. The coating uptake of coated fibres bundle after cured using (a) 1:4, (b) 1:5 and (c) 1:6 coating solutions at 0.17 to 5.0 minutes of immersion time.

For 1:4 coating solution, due to the low amount of acetone, the constituents might not completely leach-out from the fibre for the 1 minute sample. As for 1:5 coating solution, due to the increment of 3.33% of acetone, the fibre still leached-out the constituents at a greater volume. Vice versa for 1:6 coating solution, since the coating begins to dominate the fibre thus resulting in an increment in weight uptake thus reduces its weight difference as compared to 0.17 minute sample.

However, for 1:4 coated fibres, the percentage of coating uptake for 5 minutes sample is dropped slightly compared to the 4 minutes sample. It might due to the maximum effect of acetone that leached the fibres constituent and hence lowers the weight of the fibres but still, the coating formed simultaneously throughout the structure of the fibres.

For 1:5 coated fibres, the maximum coating uptake is at 5 minutes of immersion but still, the increment is slightly (3.68 %)

as compared to the 0.17 minute sample. The 5 minutes sample might begin to dominate by the coating solutions, whereas, for 0.17 minute sample, the fibre still dominated by its constituents in its system.

Vice versa for the 1:6 coated samples, since it recorded a higher weight uptake from 2 to 5 minutes of weight uptake compare to the 0.17 minute sample. It might be due to the effect of acetone that enlarges the capillaries size as its constituent leached thus allows the coating to penetrate efficiently and dominate the fibre structure. Its 5 minutes sample shows the highest coating uptake as it indicates the maximum domination of the 1:6 coating solution.

Thus, by averaging the results between all coated fibres at various viscosities but different immersion time, the 5 minute sample recorded the highest coating weight uptake compare to the rest of the samples. It might be due to the highest coating uptake that dominated the fibre system thus it was selected for the immersion time in the fibres bulk coating process for all coating solutions to fabricate a composite sample.

3.3. Tensile properties.

Figure 5 shows the Poisson's ratio and a maximum modulus of unfilled and filled samples at various fibre loadings. For maximum modulus, in general, the filled samples recorded higher stiffness than the pristine epoxy especially the treated and coated 1:6 samples.
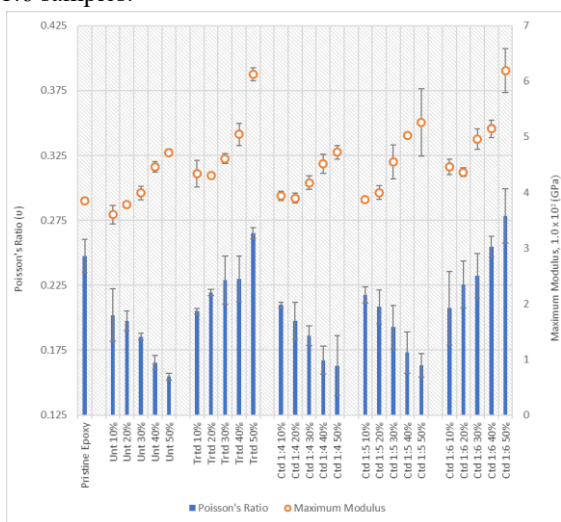

Figure 5. The Poisson's ratio and maximum modulus of pristine epoxy, untreated, NaOH/ APS treated, 1:4, 1:5 and 1:6 coated kenaf/ epoxy composites in 10, 20, 30, 40 and 50 wt% fibre loadings.

Both samples also display the highest Poisson's ratio by the addition of fibre loading. It indicates that the said samples have better fibre-matrix surface interaction than the other samples. For 1:6 samples, the high penetration of coating inside the fibre might contribute to the improvement of its stiffness [10] in the composite system higher than the treated samples.

For the treated samples, the mercerise treatment able to remove the foreign content and roughen the surface to promote mechanical interlocking once in the composite system. The said treatment also able to change its cellulose I to cellulose II that make the fibre chain in helical shape [11]. The removal of pectin, lignin and hemicellulose making the interfibrillar region loosely bonded thus promote for fibrils movement to shift to the load loading direction as force applied.

For the silanisation treatment, it creates a strong covalent bond [12] between fibre and matrix that naturally incompatible and results in an improvement in the chemically treated samples tensile properties. For coated 1:6 samples, the high penetration of the coating throughout the system of the fibre might contribute to the improvement of its tensile properties. Thus, as both samples have better surface interaction, the force was well-distributed throughout the region of the samples and efficiently retain in the stiff fibre system.

For coated 1:4 and 1:5 samples, slight property increase is observed for both tensile maximum slope and Poisson's ratio as compared to the untreated samples. It might be influenced by its coating behaviour that deteriorated during the pulverisation process that reduces its properties. The coating might dominate only outer and half part of the fibre system for 1:4 and 1:5 coated samples respectively. Thus, after pulverisation process, the coating detached from the fibre system and unable to strengthen the fibre.

3.4. Characterisation analysis of the samples fractured surface.

Figure 6 shows the fibre-matrix interaction of coated 1:4 and coated 1:6 samples. For 1:4 sample, the interaction between fibre-matrix is low since gaps appeared to be formed between the surfaces between the fibre and matrix. It might be due to the effect of low coating formation on the surface of the 1:4 coated fibre sample. Since the coating detached during the pulverisation process and exposing the uncoated fibre area that naturally incompatible with the matrix [7].
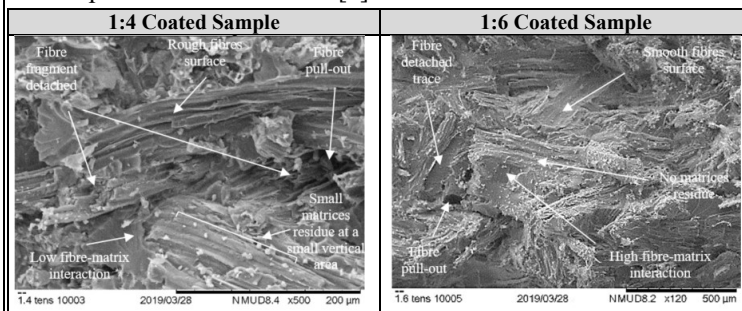

Figure 6. The SEM micrograph of fibre-matrix surface interaction of tensile tested samples fractured surfaces at 120x to 500x magnifications

But, for 1:6 coated sample, the surface interaction is better since the matrix appeared to be efficiently attached to the fibre without any gap formation. It might be due to the effect of the existence of thin coating layer that altered the fibres to become hydrophobic polarity similar to the polarity of the matrix that led to better interaction between coted fibre and matrix in the composites system as agreed by Yan [13]. Nevertheless, from the micrograph, it noticed that both 1:4 and 1:6 samples experience a fibre pull-out failure.
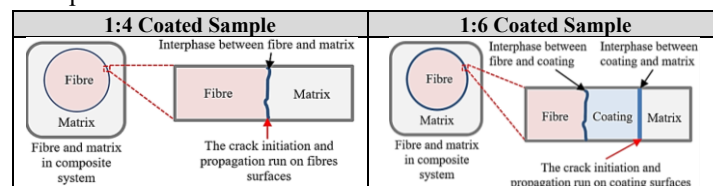

Figure 7. Illustration of the possible crack initiation and propagation between coated 1:4 and 1:6 fibres in the composites system.

This signifies that, at some point, matrices deficiency at certain fibre rich area can causing intratow voids formation [14] that easily caused a fibre pull-out failure. It happened even though

for a composite that has better surface interaction. It is also observed that on 1:4 coated sample fractograph, small matrices residue left at a certain area on the fibres. This might indicate that at this area the coating was not deteriorated by the pulverisation process.

The undamaged coating layer gives better interaction with the matrix thus, creating few residues once it failed. However, for 1:6 sample, no matrix residue is observed on the surface of the fibre.

It might be due to the thin layer of coating that controls the crack initiation and propagate at the coating-matrix interphase as illustrated in Figure 7. Thus, the fibre area was untouched leaving the fibre fragment still intact inside embedded fibres.

By comparing to 1:4 coated sample, the uncontrol crack runs between the fibre-matrix interphase thus this resulting in few fibre fragments detached. The formation of the coating can be observed from the topography of the fibres. For 1:4 coated sample, the fibre appeared to have a rough surface between the bulk fibril-fibril cleavage. But, for 1:6 coated sample, the fibre forms smoother surfaces which indicate the presence of coating that fills the fibril-fibril cleavage of the fibre bundle.

4. CONCLUSIONS

The physical, mechanical and morphological properties of various coated treated fibres composite as the effect of various coating viscosities were studied. It is perceived that the new and simple coating treatment able to improve the properties of the short Kenaf fibres in the composite system. For physical properties, the coating uptake increases as the coating immersion increase. The presence of acetone able to leach and enlarge the fibres capillary to promote coating domination. The 5 minutes of immersion period was selected to be used for all different viscosities of coating solutions as it is an ideal duration for fibres to be cleansed and leached its constituent to promote coating penetration. The 1:6 coated fibre recorded the highest mechanical properties better than the combination of NaOH/ Sil treated samples properties. The improvement is caused by the presence of coating that controls the crack initiation and generation that runs through the coating-matrix surfaces as discovered in fractographic analysis.

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6. ACKNOWLEDGEMENTS