

Article

Alkali Treatment on Hybrid Pineapple Leaf and Glass Fibre Reinforced Epoxy Composites

Syed Mohd Ridzuan Syed Pauzi¹, Syahida Suhaimi²

¹Faculty of Mechanical Engineering, Universiti Teknologi Malaysia (UTM), 81310 Johor Bahru, Johor, Malaysia

²Faculty of Science and Technology, Universiti Sains Islam Malaysia (USIM), Bandar Baru Nilai, 71800 Nilai, Malaysia E-mail: syahida@usim.edu.my

Abstract— **In this paper, the Pineapple Leaf fibre (PALF) have been previously treated with various Sodium Hydroxide (NaOH) concentration percentages in the range between 0% to 10% at 1hour Sodium Hydroxide (NaOH) solution immersion time and dried in oven at 80°C for 24 hours. The influence of Sodium Hydroxide (NaOH) treatment on Pineapple Leaf fibre (PALF) tensile properties was studied. The result of tensile test on PALF treated showed an improvement in tensile strength with highest value of 43.13MPa of treated Pineapple Leaf fibre (PALF), particularly the treated PALF with 7% Sodium Hydroxide (NaOH) concentration. The 7% treated Pineapple Leaf fibre (PALF) was then selected to use as raw materials for hybrid composites with glass reinforced epoxy matrix. The hybrid composites were subjected to distilled water immersion to study moisture absorption behavior on hybrid composites with four different layer sequences named 4P, PGGP, GPPG and PGPG. The pure Pineapple Leaf fibre (PALF) (4P) have recorded highest percentage for both moisture uptake as well as thickness swelling as compared to the other hybrid composites with value of 12.16% and 51.30% respectively. Hybrid composites are the candidate to be used a pressure vessel especially in oil gas and marine industry that deal with wet and dry environment.**

*Keywords***— PALF, water absorption; tensile test; NaOH treatment; thickness swelling test**

I. INTRODUCTION

Composite materials are widely implemented in various applications due to their unique properties and characteristics, which cannot be compared to conventional materials such as metals and rubbers. The properties of composites are strongly depending on the strength of reinforcement materials. The most common categories of composites are polymer matrix composites (PMC), metal matrix composites (MMC) and ceramic matrix composites (CMC) [1]. Fibre reinforced plastic (FRP) is a composite materials made of a polymer matrix reinforced with fibre. FRP consists of a polymer matrix implanted with highstrength fibres such as carbon, glass, aramid, basalt and natural fibres [2]. Natural fibres are usually being used as reinforcing components for thermoplastic and thermoset matrices, because of their special characteristics like renewably, biodegrability, availability and environmental friendliness that offered by natural fibres. Natural fibres also have unlimited availability, high specific properties, low cost, low density, non-abrasive and less harmful while handling [3]. In Malaysia, the unlimited availability of natural fibres is a benefit for researchers to investigate the properties of the fibre more details [4]. The mechanical performance of natural fibres is influenced by a different parameter such as the cellulose content, the microfibrillar angle, the fibre diameter, the temperature, the presence of defects and water content inside fibres. Considering the effect of diameter,

most studies conducted on natural fibres in traction showed that both the young's modulus and tensile strength increased when the fibre's diameter decreased [5]. Others parameter that gives an obvious effect on mechanical properties of fibres is the contribution of temperature. The excellent mechanical properties of PALF are correlated with its high cellulose content and comparatively low microfibrillar angle [6].

In recent years, the uses of eco-friendly materials such as pineapple leaf fibre (PALF) have increased tremendously due to unlimited resource from forest and agriculture. Compared to other natural fibres, pineapple leaf fibres (PALF) exhibit superior mechanical properties due to its high cellulose content 70% to 82% and low microfibrillar angle at 4° [5].

In our environment contains a lot of water but its action on composite materials is not fully understood. It is found that the wet conditions cause acceleration to the rate of fatigue cracks growth in glass fibre reinforced plastic under cyclic loading, but decrease the rate of growth under static loading. Water is found to be the greatest enemy to adhesive joints, including those used in hybrid composites. There are problems with the technical characteristics of reinforced materials that absorb moisture where usually have high moisture absorption and relatively low impact strength. Natural fibres absorb water from the air and direct contact from the environment. This absorption deforms the surface of the composites by swelling and creating voids. The result of these deformations is lower strength and an increase in mass [7].

An alternative to minimize the problems is the devolvement of hybrid composites which combining natural fibres and synthetics fibres. Synthetic fibre like glass and carbon can improve the stiffness, strength as well as moisture resistant behaviour of the composites and hence a balance between environmental impact and performance could be achieved [8]. Thus to enhance the adhesion property of fibres, it need surface modification by using appreciate chemical like NaOH chemical treatment [9]. This study is focusing on effect of alkali treatment on the water absorption of hybrid pineapple leaf / glass fibre reinforced epoxy composites.

II. MATERIAL AND METHOD

Sample preparation is required for this project and also as a starting point. The material used are untreated pineapple leaf fibre, PALF which is supplied by Jolly Enterprise, Kolkata, India in 2 times 2 basket weave form (tex=300, density =1.53 g/cm³), glass woven roving (tex = 4400, density $=2.54g/cm^3$) which is supplied by S&N Chemicals Sdn., Bhd., Johor, and the matrix used is Epoxy resin (1006) which is supplied by Ehsan Enterprise, Johor, Malaysia. These resin's density is 1.16 g/cm³.

NaOH solution is prepared using sodium hydroxide pellets and distilled water. The untreated pineapple leaf fibre, PALF was soaked in different percentage (0%, 5%, 6%, 7%, 8%, and 10%) of NaOH solution in the water bath for 1 hour at room temperature. The ratio of the fibres and solution was 1:20 (weight/volume). After treatment the PALF were washed and rinsed several time with distilled water. Afterwards the PALF were dried in an oven at 80° C for 24 hours.

Tensile test was conducted according to ASTM D3039 and the size samples for tensile test was 250mm long and 25mm wide with thickness of treated PALF as shown in Fig. 1 below.

Fig. 1 Treated PALF samples according to ASTM D3039

The treated PALF samples were pulled by using Universal Test Machine (Model: Instron 3382). The samples was then pulled at a rate of 2mm per minute (2mm/min) and it is necessary that the sample to be positioned vertically in the grip of the testing machine. Extensometer is used to measure elongation inside the sample. At the end of this procedure, a load displacement curves were generated for each sample.

The hybrids composites are used in this research are 2 by 2 baskets weave PALF and glass woven roving fibres reinforced composites. The moisture content of these fibres was removed by drying in an oven at 80°C for 30minutes.

After removing moisture, hardener is mixed into the epoxy resin with appropriate ratio. The mixed resin is then placed into vacuum chamber to remove the air bubble inside the resin. Then, the fibres is soaked into resin using hand lay-up and compression moulding with mould size of 250 x 200mm² and let to cure at room temperature for 24 hours. Fig. 2 shows the layering sequence of hybrid PALF (denote by P) and glass fibre (denote by G) composites.

Water absorption studied was performed following the ASTM D5229. Every samples of each composites were immersed need cut into a coupon size of $50 \times 50 \text{ mm}^2$. All coupon samples are sealed with epoxy coating paint as a water resistant paint at every edge cutting. The sample is dried in an oven, weighted and label as as-received weight, *MR*. Every sample is dried in an oven for 24 hours at temperature of 50° C, this is to make sure all the moisture in the specimen is dried out. Then, every specimen is left to cool at room temperature. After that, all specimens is weighted and label as oven-dried weight, *Mo*. Specimen is immersed immediately into water bath at temperature of 60C. Weight percentage is measured periodically until it reached saturation state. The weight percentage of moisture absorb are calculated using Equation 1.

Water Uptake,

$$
M_t = \frac{M_t - M_0}{M_0} x \ 100 \tag{1}
$$

Where M_t is the weight of specimen at time *t*, and M_0 is the weight or specimen before immersion (dry).

The thickness swelling of PALF/ Glass hybrid composites was calculated using the following Equation 2.

Relative thickness,

$$
\mathbf{T_e} = \frac{\mathbf{T_t} - \mathbf{T_0}}{\mathbf{T_0}} \, x \, 100 \tag{2}
$$

Where T_t is the thickness at time t, and T_0 is the initial thickness before immersion (dry).

III. RESULTS AND DISCUSSIONS

PALF have been treated with NaOH solution of different percentage concentration. The effect of NaOH concentration from 0, 5, 6, 7, 8, to 10 percentages for testing PALF fibres revealed that maximum tensile strength resulted from 5% NaOH treatment. ASTM D3039 standard were used for tensile test and the ASTM D3039 standard procedures were

used for tensile strength determination. Six specimens were tested to failure under tension at a crosshead speed of 1mm/min using Instron Universal Tester. An extensometer was attached to the gauge section of the samples for strain measurement. Table I shows the maximum tensile stress and tensile strain.

TABLE I MAXIMUM TENSILE STRESS AND MODULUS

Samples	Tensile Modulus (GPa)	Tensile Stress (MPa)	Strain $(\%)$
0	0.9415	35.9244	2.763
$\overline{5}$	1.2342	36.9048	3.463
6	1.3677	40.9705	3.682
	1.8613	43.1336	3.428
8	1.74978	35.1619	2.8149
10	1.2394	32.1251	2.484

Fig. 3 shows that 7% NaOH treated Palf fibres improved the tensile stress and tensile modulus 20% and 80% respectively, compared with untreated Palf fibres.

Tensile Modulus (Gpa)

Fig. 3 Tensile stress and tensile modulus at different NaOH concentration percentage

The 7% percentages NaOH PALF have better tensile strength compared to 0% and 10% concentration. The tensile properties of composites were increased after alkali treatment and become better for 7% percentage NaOH. The significance of NaOH treatment is the disruption of hydrogen bonding in fibre surface, thus increasing surface roughness. The NaOH treatment improved fibre matrix

interaction by removal of lignin and hemicellulose, which led to better incorporation of fibre with matrix [3].

This is because at high concentration, their delignification of natural fibre taking place caused to damage on the fibre surface. The tensile strength of palf decreased drastically after certain optimum NaOH concentration. Sodium hydroxide is used to break the hydrogen bonding in network structure of the fibre celullose and therefore increasing the fibre surface roughness. Alkalization within limit induced better interfacial bonding and yielded improvement of the mechanical properties [10]. On the other hand, when concentration of NaOH is used lower for treatment, the complete removal of lignin on the fibres is not possible [11]. From Fig. 4, the 7% NaOH treated Palf fibre is selected as materials for hybrid composites.

Physical properties of hybrid composites are shown in Table II. Fibre volume fraction is depending on the ratio between weight density of fibre and weight of the matrix epoxy.

TABLE II PHYSICAL PROPERTIES OF HYBRID COMPOSITES

Samples	Fibre Volume Fraction (%)	Thickness (mm)
PGGP		$2.5 + 0.03$
GPPG	$37 + 2.5$	$2.5 + 0.05$
PGPG		$2.5 + 0.02$

Five samples for each layering configuration were used and the mean of measured result were used for analysis. The moisture absorption behaviour of four different layering configuration hybrid composites is plotted in Fig. 4. In initial stage of moisture absorption test, all specimen absorb moisture are drastically but when saturated stage reached the rate become decreased. For this present study, will focus on main materials is PALF (refer Table III).

TABLE III SUMMARY OF MOISTURE ABSORPTION PROPERTIES OF HYBRID **COMPOSITES**

Samples	Maximum Moisture Uptake, M_{max} %	Diffusivity, $.10^{5}$ mm ² /s	Time for $M_{max}(hr)$
4P	12.26	4.11	160
PGGP	7.22	4.16	268
GPPG	8.27	0.91	232
PGPG	7.13	0.84	268

Exposure time, $\sqrt{\text{time}}$, min^{1/2} Fig. 5 Comparison between moisture absorption behavior of hybrid composites PALF and Woven Roving Glass with different layering sequences.

Fig. 6 Maximum moisture uptakes for different layer sequences of hybrid

Fig. 5 shows the comparison moisture absorption between 4 types of layering sequences. This graph is shows moisture absorption versus function square of times. From Fig. 6, the Hybrid composites with pure PALF (4P) layering sequences have recorded highest value of percentage of moisture uptake compared to other hybrid composites. At exposure time, 15.49 min^{1/2}, the 4P and PGGP absorb water far more than other hybrids due to hydrophilic nature and PALF is used as skin layer for 4P and PGGP hybrid. The maximum moisture uptake for GPPG is slightly higher than PGGP and PGPGP as shown in Fig. 6.

The formation of void and micro-voids in samples, which the formation of voids is one of the main cause rapid absorption of moisture. The formation void in samples during process lamination is caused by air entrapment during the initial stage of laminations that is because air trapped in resin formulation or too poor wetting of the filaments [12]. The poor wetting tends to be aggravated by high viscosity resins, which make it difficult for the matrix to penetrate fibres and displace the air. The effect of voids content is so strong, which with 1% voids in a composite can double the amount of water uptakes. In addition, cellulosic materials like fibres contain hydrixyl group (-OH) and lead to moisture sensitivity [13].

Fig. 7 below shows the comparison between theoretical and experimental, which GPPG and PGGP will consider non-fickian due to the experimental curves is not aligned with fickian theoretical. Mostly researchers used the Fickian law of diffusion to identify the model water absorption behaviour in hybrid composites. Mostly, diffusion behaviour of fibre composites is follows fickian behaviour but certain case its follow non-fickian behaviour [14].

Fig. 7(a-d) Comparison theoritical (fickian) experimental for different hybrid composites

TABLE IV PERCENTAGE THICKNESS SWELLING

Samples	Thickness Swelling (%)
4P	51.30
PGGP	25.76
GPPG	25.92
GPGP	20.44

Table IV shows the thickness swelling behavior of hybrid composites with different layer sequences. 4P hybrid composites showed highest rate of thickness swelling 51.30% compared to the others hybrid [15].When hybrid composites started to swell, they exert stress on the matrix through the fibre/matrix interface. This stress caused the matrix to stretch and make more space for swelling of fibres. According Reza Masoodi, the stress exerted by the fibres will make the swelling process to continue until it is balanced by center ailing stress on matrix [16].

IV. CONCLUSIONS

The effect moisture absorption of hybrid treated PALF / Glass fibres reinforced with epoxy matrix have been studied with distilled water immersion at 60° C. The PALF at 7% NaOH concentration have recorded the highest value of tensile properties test compared to other concentration. The PALF fibre 7% NaOH was selected and is used for manufacturing of hybrid composites together with glass fibre. Compared to the untreated PALF fibre composite, the increase in tensile strength of treated PALF composites were respectively of about 20%, which were due to high enhancement in the interfacial adhesion of the fibre with the epoxy matrix. Alkaline treatment of PALF fibres with epoxy matrix can improve mechanical properties of composites.

The effect of moisture absorption test on hybrid composites, based on result the water absorption and thickness swelling increased with immersion time for all types of hybrid composites. Hybrid composites with layer sequences GPGP have lower moisture absorption percentage and lower thickness swelling percentage compared to other hybrid composites.

ACKNOWLEDGEMENTS

The authors wish to thank Centre for Composites and Mechanics of Material Laboratory and the Universiti Teknologi Malaysia for supporting this research activity.

REFERENCES

[1] D. Hull and T.W. Clyne, *An Introduction to Composite Materials*,

2nd ed., Cambridge, United Kingdom: Cambridge University Press, 1996.

- [2] J. R. Correia, S. Cabral-Fonseca, F. A. Branco, J. G. Ferreira, M. I. Eusébiob, and M. P. Rodriguesb (2005). "Durability of Glass Fibre Reinforced Polyester (GFRP) pultruded profiles used in civil engineering applications, composites in construction," in *Proc. CCC'05*, 2005, pp. 11–13.
- [3] A. Benyahia, A. Merrouche, M. Rokbi, and Z. Koudri, "Study the Effect of Alkali Treatment of Natural Fibers on the Mechanical Behavior of the Composite Unsaturated Polyester-fiber," in *Proc. ALFA'13,* 2013, pp. 69-73.
- [4] M. Asim, K. Abdan, M. Jawaid, M. Nasir, Zahra Dashtizadeh, M. R. Ishak, and M. Enamul Hoque. "A review on pineapple leaves fibre and its composites," *International Journal of Polymer Science* vol. 16, pp. 2-16, 2015.
- [5] J. Andersons, E. Sparniņš, R. Joffe, and L. Wallström, L. "Strength distribution of elementary flax fibres," *Composites Science and Technology,* vol. 65(3–4), pp. 693–702, 2005.
- [6] L. Devi, S. Bhagawan, and S. Thomas, "Mechanical Properties of Pineapple Leaf Fiber-Reinforced", *Journal of Applied Sciences,* vol. 64, pp. 1739–1748, 1996.
- [7] U. Hujuri, S. K. Chattopadhay, R. Uppaluri, and A. K. Ghoshal, "Effect of maleic anhydride grafted polypropylene on the mechanical and morphological properties of chemically modified shortpineapple-leaf-fiber-reinforced polypropylene composites," *Journal of Applied Polymer Science,* vol. 107 (3), pp. 1507-1516, 2008.
- [8] S. Nunna, P. R. Chandra, S. Shrivastava, and A. K. Jalan, "A review on mechanical behavior of natural fibre based hybrid composites," *Journal of Reinforced Plastics & Composites,* vol. 31(11), pp. 759- 769, 2012.
- [9] J. P. Siregar, S. M. Sapuan, M. Z. A.Rahman, and H. M. D. Zaman, "The effect of alkali treatment on the mechanical properties of short pineapple leaf fibre (PALF) reinforced high impact polystyrene (HIPS) composites," *Journal of Food Agriculture and Environment,* vol. 8(2), pp. 1103-1108, 2010.
- [10] H. P. S. Abdul Khalil, M. Jawaid, and A. Abur Bakar, "Woven hybrid composites: Water absorption and thickness swelling behaviours," *BioResources,* vol. 6, pp. 1043– 1052, 2011.
- [11] J. L. Thomason, "The interface region in glass fibre-reinforced epoxy resin composites: 3. Characterization of fibre surface coatings and the interphase," *Composites,* vol. 26, pp. 487– 498, 1995.
- [12] R. Masoodi, "A study on moisture absorption and swelling in biobased jute epoxy composites," *Journal of Reinforced Plastics and Composites,* vol. 31(5), pp. 285-294, 2012.
- [13] K. Senthilkumar, N. Rajini, S. Naheed, C. Muthukumar, M. Jawaid, and S. Siengchin. "Effect of alkali treatment on mechanical and morphological properties of pineapple leaf fibre/polyester composites". *Journal of Polymers and the Environment,* vol. 27(6), pp. 1191-1201, 2019.
- [14] D. Saikia, "Studies of water absorption behavior of plant fiber at different temperatures," *International Journal of Thermophysics,* vol. 31(4), pp. 1020-1026, 2010.
- [15] M. Asim, M. Jawaid, K. Abdan, and M. Nasir, "Effect of alkali treatments on physical and mechanical strength of pineapple leaf fibres," *IOP Conference Series: Materials Science and Engineering,* vol. 290(2018) 01 2030, pp. 1-6, 2018.
- [16] M. Mittal and R. Chaudhary, "Biodegradability and mechanical properties of pineapple leaf/coir fiber reinforced hybrid epoxy composites," *Materials Research Express,* vol. 6(4), 2019.