

# Surface Modification with Silane Coupling Agent on Tensile Properties of Natural Fiber Composite

Willy Artha Wirawan<sup>a</sup>, Sofyan Arief Setyabudi<sup>b</sup>, Teguh Dwi Widodo<sup>c</sup>, Moch. Agus Choiron<sup>c</sup>

<sup>a</sup>Mechanical Engineering Department, Brawijaya University, Malang, Indonesia

Telephone +62-341-554291, 587711, fax +62-341-554291

e-mail: willymadiun93@gmail.com

## Abstract

*Biocomposite is an innovation of renewable material in engineering made from the bark fiber of waru (*Hibiscus tiliaceus*, the bark is environmentally friendly and has the potential to be developed. The purpose of this study is to modify biocomposite by adding methacryloxypropyltrimethoxysilane-coupling agent on waru bark fiber (*Hibiscus tiliaceus*) as an effort to improve tensile properties. Waru bark fibers, as reinforcement, were prepared by alkali process using 6% NaOH solution for 120 minutes and then added 0.75% coupling agent and ordered using continuous fibers with 0°/0°, 0°/90° and 45°/45° orientation. After that, the Biocomposites were formed with a polyester matrix using Vacuum Pressure Resin Infusion (VAPRI) method. The results of SEM tested showed the quality improvement of the bonds bringing significant impact on the tensile properties of the waru bark biocomposite. On the continuous fiber 0°/90° orientated showed that the highest strength was 401.368 MPa, while biocomposites with 45°/45° orientation has lowest tensile strength of 65.243 MPa.*

**Keywords:** Natural Fiber Composite; Waru bark; Silane Coupling Agent; Tensile strength

## 1. INTRODUCTION

Natural fibers have now been widely used as reinforcements in the making of composites as they are more environmentally friendly compared to synthetic fibers [1]. Natural fibers have many advantages such as low price, abundant of amount, lightweight, high strength, and biodegradable [2]. Therefore, it has great potential to be developed as reinforcement material for polymer composites [3].



Figure 1.1 (a) *Hibiscus tiliaceus* and (b) *Hibiscus fibers*

Waru (*Hibiscus tiliaceus*) in Figure 1 is a plant that widely found in Indonesia. People uses the trees as the material for making vessel ropes because its strength and toughness. Hence, it can be developed as reinforcement in the manufacture of polymer

composites [4] [5]. The problem often arises in the manufacture of natural fibers based composites are the debonding that causes composite strength decrease [6]. One the method to overcome this problem is by chemical treatments using NaOH solution and addition of silane coupling agent. This method improve the adhesion force and entanglement bond, hence improving the compatibility of polymer matrix and natural fiber [7]. The silane will react with the cellulose on the fiber surface and bind the matrix [8].

The presence of hydroxyl group OH in natural fibers causes the waru bark fibers to be hydrophilic. It causes poor bonding quality to the polymer matrix [9]. Surface modification is necessary reducing the hydrophilic properties of the waru bark fibers, hence increasing its compatibility. The effort of this study is to improve tensile properties of waru bark fiber (*Hibiscus tiliaceus*) biocomposites by surface modification using methacryloxypropyltrimethoxysilane-coupling agent. This coupling agent is belong to anhybride group that can react with the surface hydroxyl groups of natural fiber, and successfully applied to organic filler reinforced polymer composite.

## 2. METHODOLOGY

The waru bark fiber was supplied by waru farmers in Tulungagung, Indonesia. Its specification shows in the Table 2.1. While The matrix polyester 157 is supplied from PT Justus Kimia Raya Surabaya and the chemical specification of the matrix is shows in Table 2.2.

Table 2.1 Chemical Specification of the Fibers from Waru Bark

Properties	Composition
Raw protein	17.08
Ether extract	3.45
Raw fibers	22.77
Ash (%)	10.79
Carbohydrate	45.91
Tannin (%)	8.93
Saponins (mg/g)	12.90
Cellulose	24.22

Table 2.2 Chemical Specification of the Polyester Matrix 157

Mechanical Properties	Quantity	Unit
Specific gravity	1.4	gr/cm <sup>3</sup>
Hardness	40	-
Heat distortion temperature	70	°C
Water absorption (room temperature)	0.188	% (24 jam)
	0.446	% (7 hari)
Flexural Strength	9.4	Kg/mm <sup>2</sup>
Flexural Modulus	300	Kg/mm <sup>2</sup>
Tensile strength	5.8	Kg/mm <sup>2</sup>
Elasticity modulus	300	Kg/mm <sup>2</sup>
Elongation	2.4	%

The surface modification of the waru bark fibers was carried out by a chemical treatment using NaOH solution with Methacryloxypropyl-trietoxysilane silane-coupling agent addition originated from Dow Corning. This coupling agent has specific gravity and viscosity of 1.03 and 5 mm<sup>2</sup>/s, respectively. The chemical structure of silane-coupling agent can be seen in Figure 2.1.



The composite manufacture process was conducted by Vacuum Assisted Resin Infusion (VARI) method is shown in Figure 2.3.

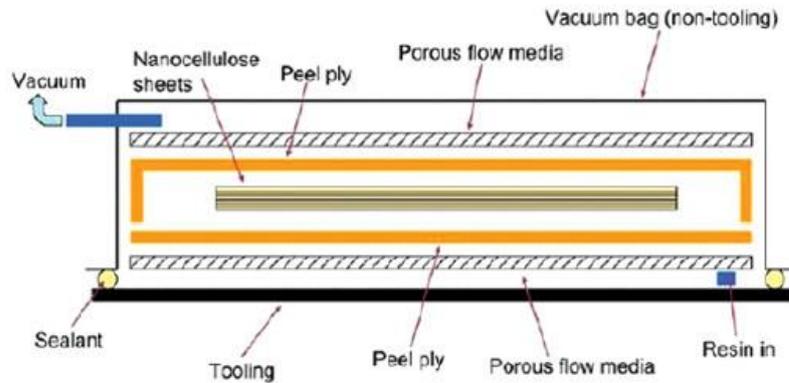


Figure 2.3. The Systematic Diagram of VARI [11]

Table 2.3 Treatment of Fibers

Treatment Type	Code	Description
Untreated	SS	Fiber without treatment
Alkali (SN)	SN 0-0	Fiber treated with alkali continous fiber composite
	SN 0-90	Fiber treated with alkali orientation 0-90
	SN 45-45	Fiber treated with alkali orientation 45-45
Alkali Silane (SSN)	SSN 0-0	Fiber treated with alkali and silane continous fiber composite
	SSN 0-90	Fiber treated with alkali and silane orientation 0-90
	SSN 45-45	Fiber treated with alkali and silane orientation 45-45

### 3. RESULT AND DISCUSSION

#### 3.1 Interface Morphology

The SEM result on Figure 3.1 shows the density improvement of surface. Figure 3.1a shows that untreated bark (SS) has rougher surface because it has more cellulose and hemicellulose content [2]. Whereas Figure 3.1b shows that the fiber interface with alkali treatment (SN) is proven cleaner and has a flatter and smoother fiber texture. The alkali treatment is very effective on reducing cellulose, lignin, and hemicellulose content [12].

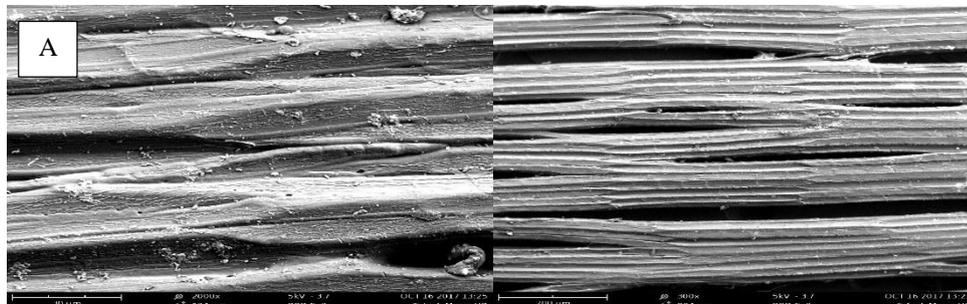


Figure 3.1 (a) Untreated SS (b) Alkali Treatment SN (c) Alkali and Silane Treatment SSN

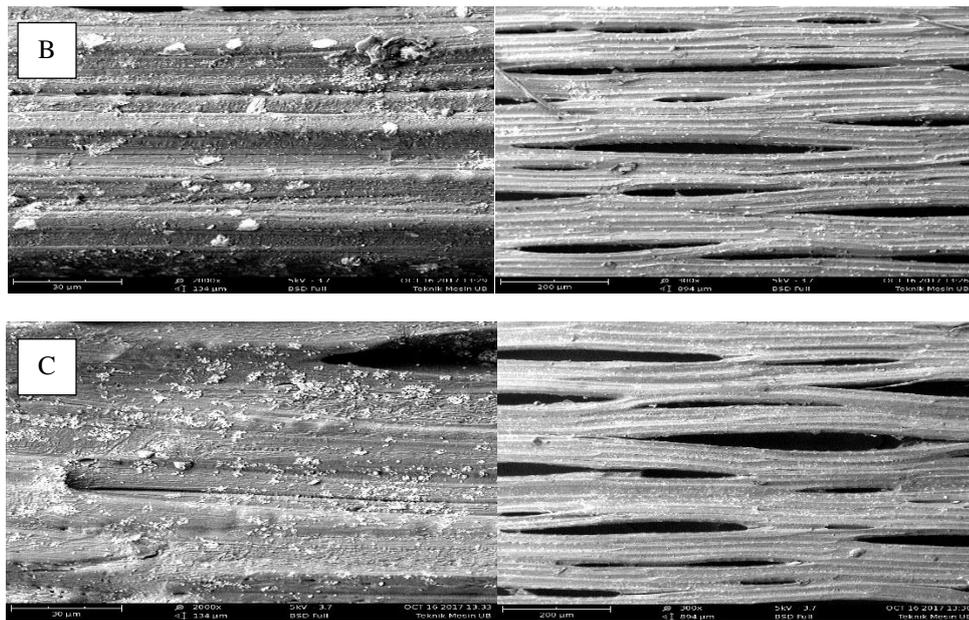


Figure 3.1 (a) Untreated SS (b) Alkali Treatment SN (c) Alkali and Silane Treatment SSN (continued)

Fibers with alkaline and silane treatment (SSN) shows a better surface morphology [10]. The fiber surface becomes very clean, flat, dense, and very smooth compared to the untreated one (SS) and alkali treated (SN) surfaces. With better interfaces, it is possible to have compatibility between the bark fiber and polymer matrix [12]. The matrix and the bark fibers will make a good binding, hence the biocomposite can receive the load evenly and provide a very significant impact to increase its tensile strength [13]

### 3.2 Tensile Properties

The surface modification of the *waru* bark fibers has a significant effect on the tensile strength of the biocomposite and it can be seen in Figure 3.2, 3.3 and 3.4.

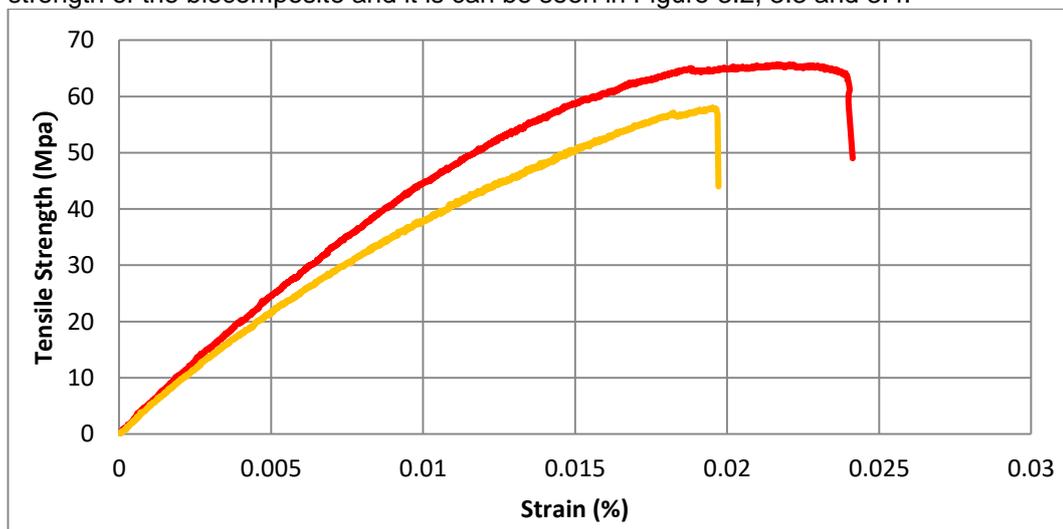


Figure 3.2 Strain-Stress (a) Fiber Treated with Alkali Orientation 45-45 (b) Fiber Treated with Alkali and Silane Orientation 45-45

Figure 3.2 shows the biocomposite strain treated by the alkali (SN) and alkali silane (SSN) form a 45-45 orientation angle of the fiber arrays. It can be seen in Figure 3.2 that biocomposite treated with alkali silane has higher strength and strain than biocomposite

treated with alkali, with 65.24 Mpa 0.023% and 58.005 Mpa 0.023%, respectively. The biocomposite on 45-45 oriented has higher strain compared to unidirectional composites on 0-90 orientation, meanwhile the strength is lower [14].

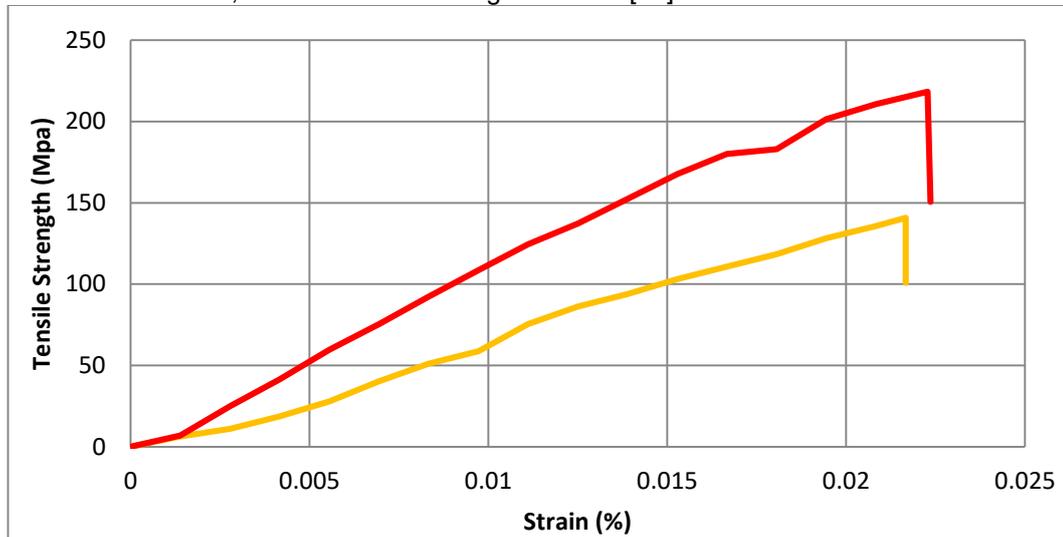


Figure 3.3. Strain-Stress (a) Fiber Treated with Alkali Orientation 0-90  
 (b) Fiber Treated with Alkali and Silane Orientation 0-90

Figure 3.3 shows the biocomposite strain treated by the alkali (SN) and alkali silane (SSN) form a 90-90 orientation angle of the fiber arrays. It can be seen in Figure 3.3 that biocomposite treated with alkali silane has higher strength and strain than biocomposite treated with alkali, with 218.246 Mpa 0.22% and 140.861 Mpa 0.021%, respectively. The biocomposite on 0-90 oriented has higher strain compared to composites on 45-45 orientation [14].

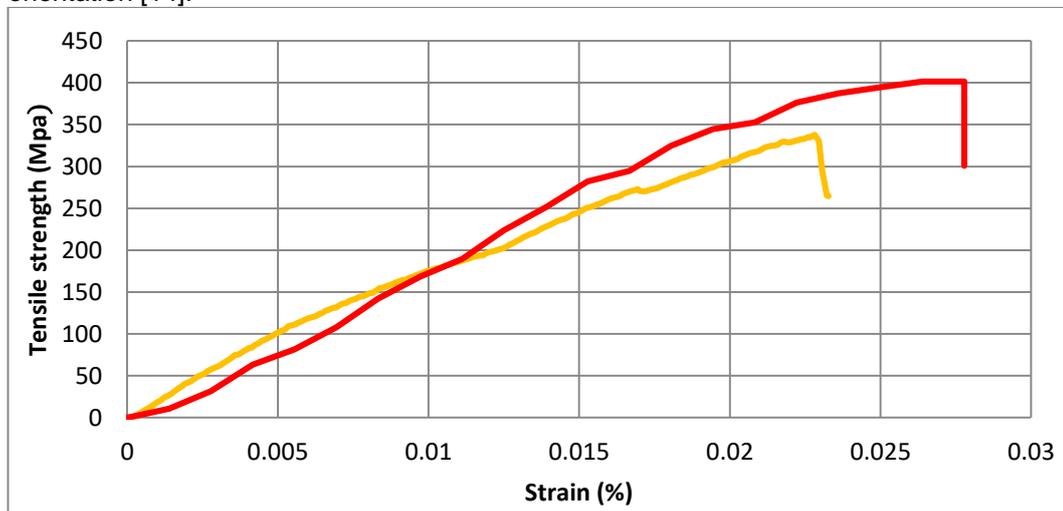


Figure 3.4 Strain-Stress (a) Fiber Treated with Alkali Continuous Fiber Composite  
 (b) Fiber Treated with Alkali and Silane Continuous Fiber Composite

Figure 3.4 shows the biocomposite strain treated by the alkali (SN) and alkali silane (SSN) form a unidirectional fiber arrays. It can be seen in Figure 3.4 that biocomposite treated with alkali silane has higher strength and strain than biocomposite treated with alkali, with 401.368 Mpa 0.027% and 3387.78 Mpa 0.022%, respectively. The unidirectional biocomposite has the highest strength. Because it has longitudinal direction toward uniaxial load, hence it minimize the shear failure [14].

#### 4. CONCLUSION

Fiber treatment using alkali and silane improve the surface, hence increasing tensile strength of biocomposite. Biocomposites with unidirectional alkaline silane treatment have the highest tensile strength of 401.368 MPa, while the lowest tensile strength is obtained on 45°/45° orientation angle biocomposites of 65.243 MPa.

#### REFERENCES

- [1] Kumari, M., Kumar, R., & Kumar, V. (2014). *Surface modification of cellulose using silane coupling agent. Carbohydrate Polymers*, 111, 849–855. <https://doi.org/10.1016/j.carbpol.2014.05.041>
- [2] Xue. Li., & Canada, A. (2007). *Chemical Treatments of Natural Fiber for Use in Natural Fiber-Reinforced Composites: A Review* *Chemical Treatments of Natural Fiber for Use in Natural Fiber-Reinforced Composites: A Review*, (May 2014). <https://doi.org/10.1007/s10924-006-0042-3>
- [3] Nishino T. (2004). *Natural Fiber Sources*, In: *Bailie C, editors, Green composites Polymer composites and the environment, England, Woodhead Publishing Limited* , pp:49
- [4] Malkapuram R, Kumar V, Negi YS. (2009). *Recent development in natural fiber reinforced polypropylene composites*, *Journal of Reinforced Plastics and Composites*, 8 (10), 1169-1189
- [5] Palungan, M. B., Soenoko, Y. S. Irawan, A. Purnowidodo (2015). *Mechanical properties of king pineapple fiber (Agave Cantala Roxb) as A result of fumigation treatment. Australian journal of basic and applied sciences* 9:560-63
- [6] Akil H.M., Omar., Mazuki A.A.M., Safiee S., Ishak Z.A.M., Abu Bakar A. 2011. *Kenaf Fiber Reinforced Composites: A review, Material and design*, 32:4107-4121
- [7] Dittenber D.B. and H.V.S. Ganga Rao. 2012. *Critical review of recent publications on use of natural composites in infrastructure. Composites part A*43 (8):1419-29
- [8] Hermanson G.T., 2008. *Bioconjugate Techniques, Second edition, Elsevier's Science & Technology Rights*. United Kingdom
- [9] Islam, M. R. & Beg, M. D. H. (2010). *Effect Of Coupling Agent On Mechanical Properties Of Composite From Kenaf and Rcyclcd Polypropylne. National Conference In Mechanical Engineering Research and Postgraduate Studies*. Malaysia: University Malaysia Pahang, pp 871-875
- [10] Raharjo, W. W., Soenoko, R., Irawan, Y. S., & Suprpto, A. (2017). *The Influence of Chemical Treatments on Cantala Fiber Properties and Interfacial Bonding of Cantala Fiber / Recycled High Density Polyethylene (rHDPE). Journal of Natural Fibers*, 0(0), 1–14. <https://doi.org/10.1080/15440478.2017.1321512>
- [11] Thakur, K. V., Thakur, K. M., Gupta , K. R. (2014). *Review: Raw Natural Fiber-Based Polymer Composites. International Journal of Polymer*. 19: 256-271
- [12] Zhou F., G. Cheng and B. Jiang. (2014). *Effect of silane treatment on microstructure of sisal fibers, Applied Surface Science* 292:806-12
- [13] Gan, H. L., L. Tian, and C.H.Yi. (2014). *Effect of sisal fiber surface treatments on sisal fiber reinforced polypropylene (PP) composites. Advance Materials Research* 906:167-77

- [14] Setyabudi, S. A., Makabe, C., Fujikawa, M., Tohkubo, T. (2011). *Fatigue and Static Fracture of Machineable C/C Composites*. *Journal of Solid Mechanics and Materials Engineering* Vol. 5, No. 11, 640-654.