# The effect of processing methods on the improvement of tensile properties of Random Ramie Fiber – Reinforced Tapioca Starch Biocomposites

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**Abstract.** The disposal of Glass Fiber-Reinforced Plastic (GFRP) composites is becoming an environmental issue. To overcome the problem, biocomposites made of ramie fibers and tapioca starch might become an alternative to GFRP composites, due to their renewable properties, environmental friendly and easily degraded by nature. However, mechanical properties of fiber-reinforced starch based biocomposites are quite low due to high void content and poor fiber-matrix interface. In this research, the effect of the processing methods on tensile properties of tapioca starch - reinforced random ramie fiber biocomposites were studied. Ramie fiber reinforced tapioca starch biocomposites were fabricated by means of solution casting and compression molding technique. Random ramie fiber-reinforced tapioca starch biocomposites were succesfully made with the highest tensile strength and modulus elasticity of biocomposites were 18 MPa and 959 MPa respectively. The use of 1% NaOH alkalization process on ramie fiber increased tensile strength and modulus elasticity of biocomposites 64% and 54% respectively. Application of 120 $^{o}$C compression temperature, increased tensile strength and modulus elasticity of biocomposites 18% and 74% respectively. Homogenous dispersion of ultimate fiber increased the tensile strength up to 91%. However, the addition of glycerol decreased the tensile strength and modulus elasticity for 46% and 39% respectively.

## 1. Introduction

Glass Fiber-Reinforced Plastic (GFRP) composites have been increasingly used in many applications in major industries. It is estimated that the production volume of GFRP in the European countries in 2015 increased by 2.5% to 1,069 megatons [1]. And the demand for GFRP will continue to grow rapidly worldwide. However, the growing production of GFRP caused the increasing of GFRP wastes. The GFRP wastes are commonly disposed by means landfilling and incineration which may affect the environment. To overcome this problem, the development of biocomposites derived from renewable resources are required. Ramie fibers and cassava starch have a great opportunity as an alternative to subtitute the glass fibers and the synthetic polymers on producing biocomposites.

Ramie is commonly found locally in Indonesia and belongs to the bast fiber. Ramie fibers have several advantages such as high grow rate over other bast fiber [2], high specific strength and modulus, and high cellulose content [3]. The tensile strength, modulus elasticity, and elongation of ramie fibers have been reported to be in the range of 400 – 938 MPa, 61 – 128 GPa, and 1.2 – 3.8 %,

respectively [3]. As a result, ramie fibers have been widely used as reinforcing fibers for polymer composites [3-5].

Starch is a potential raw material for the production of biodegradable plastic and have been used to fabricate biocomposites due to renewable, biodegradable, and sustainable [6]. Cassava is one of the major resources for starches [7], and Indonesia is one of the largest producer of cassava with production of 24 million tonnes in 2014 [8]. The use of cassava starch as the matrix for the biocomposites have been reported in the past decades [9-14]. However, the tensile properties of fiber-reinforced starch based biocomposites are quite low, due to high void content and poor fiber-matrix interface [9-14]. Therefore, the main purpose of this study is to improve the tensile properties of biocomposites with the various processing methods. The effect of alkalization, compression temperature application, glycerol content, and fiber dipersion distribution variation were investigated.

## 2. Experimental

### 2.1. Materials
The ramie fibers used in this study were obtained from Pesantren Darussalam Garut, Jawa Barat (Indonesia). The native casava starch was purcased from PT. Budi Acid Jaya Tbk (Indonesia). The glycerol was used as plasticizer, was supplied by PT. Brataco Bandung (Indonesia). The polyvinyl alcohol (PVA) was supplied by Central Kimia Bandung (Indonesia).

### 2.2. Preparation of Ramie Fibers
Native-ramie fibers were washed with demineralized water and dried at room temperature for 48 h. The amounts of native-ramie fibers, were kept to be used as reinforcement for the biocomposites preparation. Another portion of native-ramie fibers were prepared for alkali treatment. Native-ramie fiber were refluxed in 1 wt% NaOH solution, with ratio 1:100 for 2 h at 98 $^{\circ}$C. Last, the ramie fibers (untreated and treated fiber) were then cut into 10 mm lengths.

### 2.3. Preparation of Starch-Based Biocomposites
Tapioca starch-based biocomposites with 15 wt% ramie fiber were prepared by solution casting and compression molding technique. PVA 29 wt% was dissolved in demineralized water and stirred on magnetic stirer. The mixture was then heated to 85 $^{\circ}$C. Thereafter, the mixture was cooled down to 55 $^{\circ}$C. At 55 $^{\circ}$C cassava starch, without and with addition of 20 wt% glycerol, were added under constant stirring, and was then heated to 70 $^{\circ}$C. Ramie fibers (untreated and treated fiber) were spread randomly into the molds. The fiber distribution was varied and coded as D0, D1 and D2 reflect the unhomogeneous until homogeneous fiber dispersion respectively. After that, the resin mixture was removed from the heat and pouring it on the surface of ramie fibers and then dried at room temperature for 72 h. Other biocomposite specimens were fabricated by means of hot compression molding and were heated at 120 $^{\circ}$C for 1 h. Table 1 shows the sample of biocomposites with the various processing methods.

**Tabel 1**. Sample of Biocomposites.

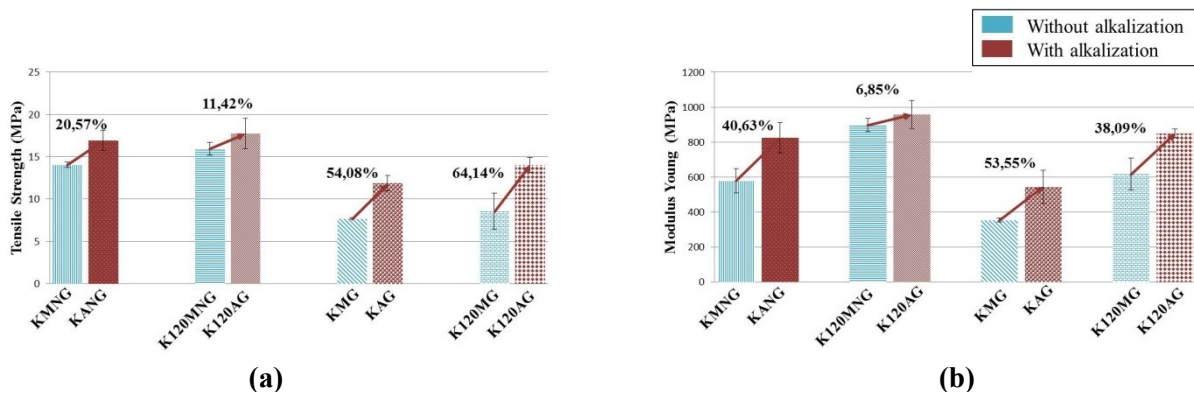| Wihout Compression | With 120$^{\circ}$C Compresion Temperature |
|---|---|
| KMNG | K120MNG |
| KANG | K120ANG |
| KMG | K120MG |
| KAG | K120AG |
| N: Without alkalization | NG: Without addition of glycerol |
| A: With alkalization | G: With glycerol content |

*2.4. Tensile Test*
Tensile test were carried out following ASTM D-3039. Tensile properties was measured using Tensilon RTF-1310, equipped with a 1 kN load cell at a crosshead speed of 5 mm/min. Three specimens were tested for every sample. The fractured surface of biocomposites were observed by optical microscope.

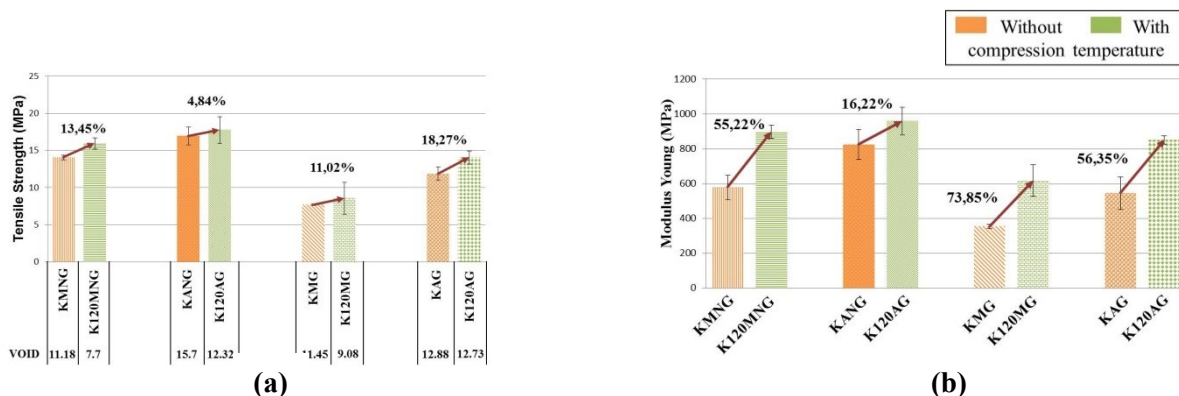## 3. Results and discussion

*3.1. Effect of Alkalization*
Figure 1 shows the effect of alkalization on tensile properties of the biocomposites. It can be seen that alkalization improves the tensile strength and modulus elasticity of the biocomposites, with the maximum improvement by 64% and 54% respectively. Alkalization increased the fiber surface roughness and the number of free hydroxyl groups in the fiber surface, resulting in better mechanical interlocking and chemical bonding which leads to an improvement in fiber-matrix interface [15-17]. The improvement in fiber-matrix interface results in improvement the tensile properties of biocomposites.



**Figure 1.** The effects of alkalization on tensile properties of the biocomposites: (a) tensile strength and (b) modulus elasticity.
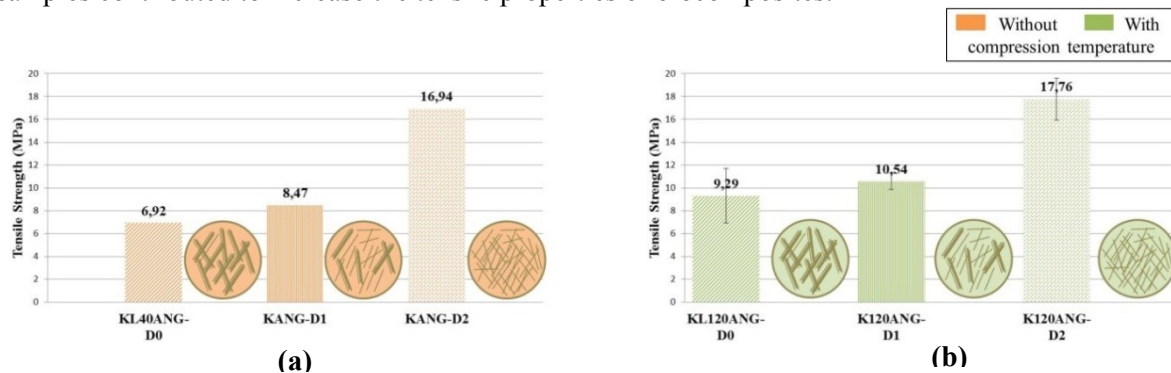
*3.2. Effect of Compression Temperature*
Figure 2 shows the effect of compression temperature on tensile properties of the biocomposites. The tensile strength and modulus elasticity increased after the application of compression temperature at 120 $^{\circ}$C, with the improvement by 18% and 74% respectively. This improvement was mainly due to the decrease of void content on the biocomposites (see Figure 2.a).



**Figure 2.** The effects of compression temperature on tensile properties of the biocomposites: (a) tensile strength and void content and (b) modulus elasticity.

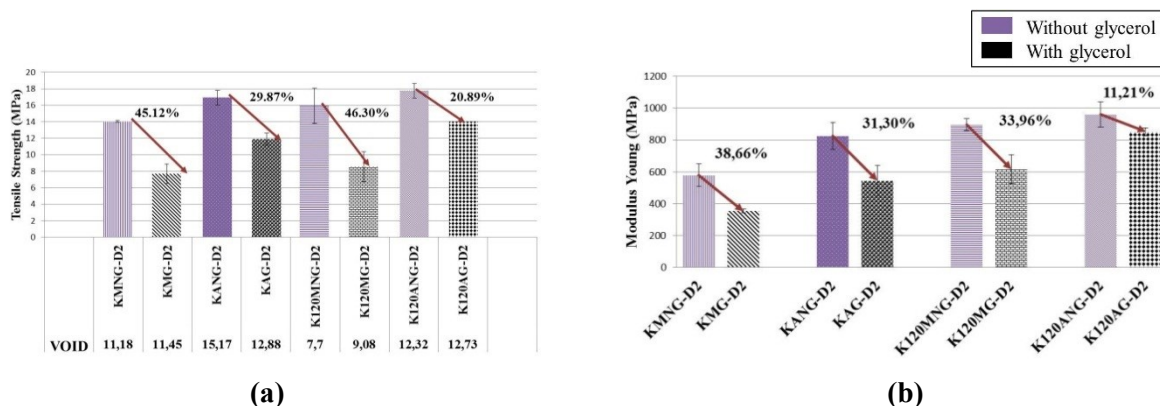### 3.3. Effect of Fiber Dispersion

Figure 3. shows the effect of fiber dispersion on tensile properties of biocomposites. The tensile strength and modulus elasticity increased significantly with homogeneous fiber dispersion. The highest tensile strength (17.76 MPa) was obtained from the biocomposites made with 120 $^{o}$C compression temperature using homogeneous ultimate fiber distribution, K120ANG-D2, see Figure 3.b. Homogeneous distribution of the ultimate fiber made the matrix easily penetrated into the fiber mats and resulting in better fiber-matrix interface. The tensile strength of ultimate fibers used in D2 sample which is much higher than fiber bundles (938 MPa  vs 238 MPa [3,14]) used in D0 and D1 samples contributed to increase the tensile properties of biocomposites.



**Figure 3.** The effect of fiber dispersion on tensile strength of biocomposites: (a) without and (b) with 120 $^{o}$C compression temperature.

### 3.4. Effect of Glycerol Content

The addition of glycerol content decreased the tensile strength and modulus elasticity for 46% and 39% respectively as shown in Figure 4. It shows that the addition of glycerol increased the void content on biocomposites (see Figure 4.a), resulting in lower tensile properties of biocomposites.
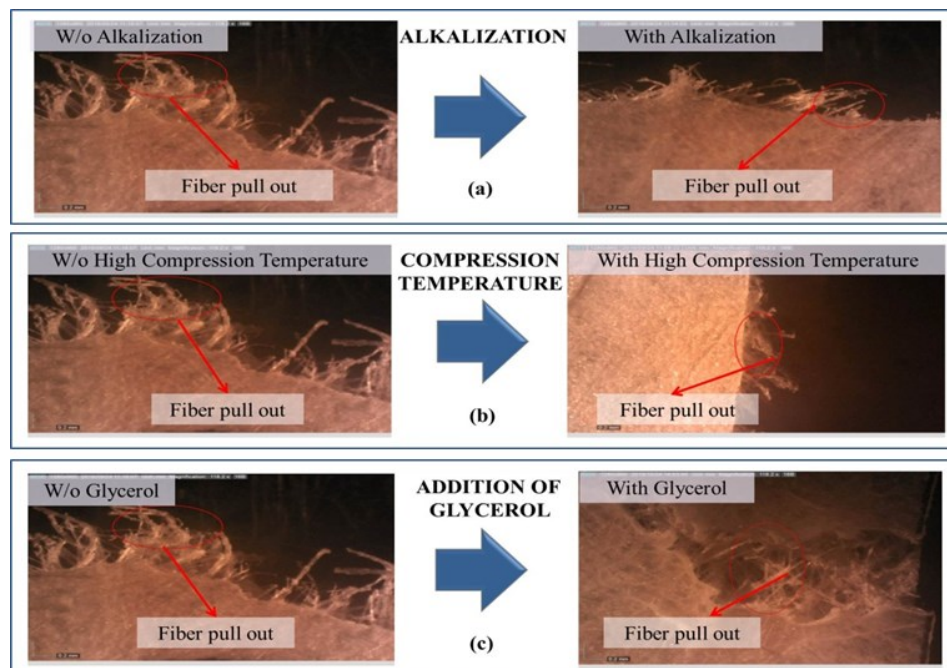


**Figure 4.** The effects of glycerol content on tensile properties of biocomposites: (a) tensile strength and void content and (b) modulus elasticity.

### 3.5. Fracture Surface of Biocomposites

Figure 5 shows the fracture surface of biocomposites. It can be seen that alkalization and high 120 $^{o}$C compression temperature (see Figure 5.a and 5.b) decreased the existence of fibers pull out due to the improvement of fiber-matrix interface. These results confirm the improvement of tensile properties on biocomposites due to the alkalization and 120 $^{o}$C compression temperature. On the other hand, the addition of glycerol content (see Figure 5.c) increased the number of fibers pull out due to deterioration of fiber-matrix interface, resulting in lower tensile properties of biocomposites.
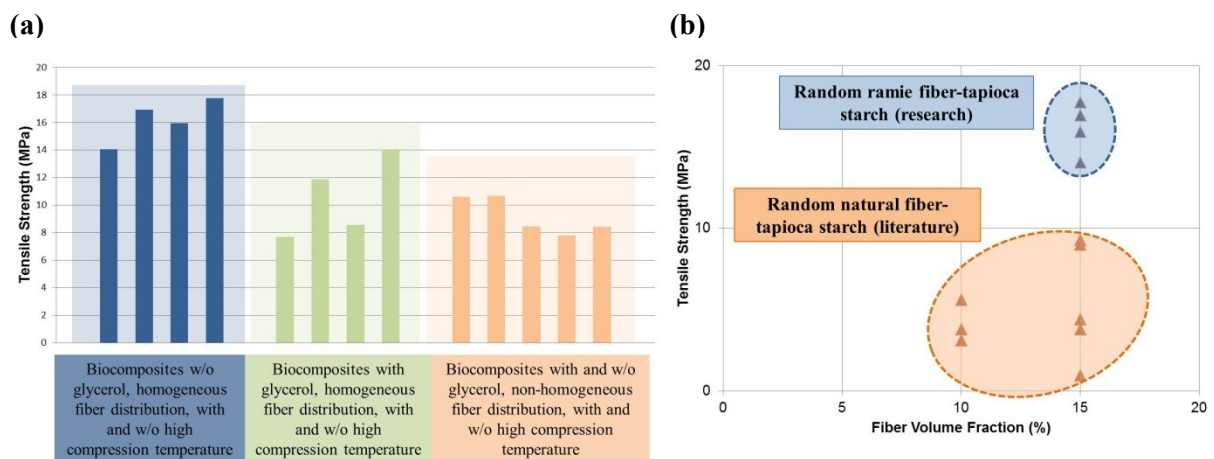
**Figure 5.** The fracture surface of biocomposites: (a) effect of alkalization, (b) effect of compression temperature, and (c) effect of addition of glycerol content.

*3.6. Comparison of Tensile Strength Fiber Reinforced Based Starch Biocomposites*
Figure 6 shows the comparison of tensile strength fiber reinforced based starch biocomposites. It can be seen, in Figure 6.a that the best processing method to obtain biocomposites with the highest tensile strength was by using homogeneous fiber distribution without glycerol addition. The tensile strength of random ramie fiber-tapioca starch developed in this research is higher compared to the literature results as shown in Figure 6.b [9-14].

**(a)**

**(b)**



**Figure 6.** Comparison of Tensile Strength Fiber Reinforced Based Starch Biocomposites (a) with different processing methods and (b) with literatures [9-14].

**4. Conclusions**
The highest of tensile strength and modulus elasticity of biocomposites developed in this research were 17.76 MPa and 959 MPa respectively. The use of 1% NaOH alkalization process on ramie fiber

increased tensile strength and modulus elasticity of biocomposites 64% and 54% respectively. The application of 120 $^{o}$C compression temperature, increased tensile strength and modulus elasticity of biocomposites 18% and 74% respectively. Homogenous dispersion of ultimate fiber distribution increased the tensile strength up to 91%. The addition of glycerol decreased the tensile strength and modulus elasticity for 46% and 39% respectively. The results indicate that renewable resources such as ramie fibers and cassava starch, have potential ability to replace glass fibers and syntetic polymers as the reinforcement and the matrix for the biocomposites application.

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