**Effect Soaking Time of NaOH Solution on Physical, Mechanical and Thermal Properties of Chopped Sisal-Polyester Composite**

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**Abstract.** This study investigates the effect of alkali soaking time by 5 wt.% NaOH on the Sisal fiber. Chopped sisal fibers cut 10 mm to reinforce the unsaturated polyester resin (UPR) with a fraction volume ratio of 30:70. The composite product uses the press mould method with 2 bar pressure to produce the chopped sisal-polyester composite (SPC). Mechanical tests include tensile, bending, and impact according to ASTM D-638, D-790 and D-5942. The tensile test shows that SPC soaked for four hours (4h) increases the tensile, flexural and impact strength by 41%, 8%, and 38%, respectively. The Fracture observed by the Scanning Electron Microscope (SEM) image shows that untreated SPC is dominated by fibre pull-out and debonding. Treated for four hours (4h), it shows transversal fibers fracture. It occurs because the hydroxyl group (OH) of sisal fibres' cellulose bonded perfectly with the UPR matrix. The long soak for six hours (6h) causes degradation of the cellulose crystalline region, and it causes poor bonding between fibers and the UPR matrix.

1. **Introduction**

In composite material technology, natural fibre materials have the potential for reinforcement to replace synthetic fibers in the automotive industry [1]. Synthetic materials are still used in several components, such as dashboards and door trim. In the future, these synthetic fibers will be abandoned and replaced by natural materials because they cannot be recycled. Natural fibers are abundant and sustainable materials, such as Sisal, Flex, Hemp, Jute, Rami, Kenaf, bamboo, etc., which have started to be used to reinforce polymer composite material. Now, each automotive product is developed by natural fiber reinforcement on polyester resin applied to the door trim, dashboards, ceiling panel, and seat back of the car [1]. Low density, high strength-to-weight ratio, resistance to chemical content, low energy content, and recyclability are all benefits of using natural fiber composites. One of the kinds of natural fiber objects of this research is the Agave Sisalana (Sisal) is leaf fiber plant. Sisal is one of the annual tropical plants, and periodically time. These plants grow well in arid and rocky soil conditions, like in Malang (East Java), Karanganyar (Bali), and Sumenep (Madura) Province, Indonesia. These fibers are widely used for ropes, nets, brooms, mats, and other commercial craft products because they are strong, elastic, and resistant to seawater [2]. Automotive products processed from natural fibers cannot be used directly because the fibers still contain a lot of impurities, which disrupts compatibility with the matrix. The matrix and fibers' interfacial bonding must be adequately considered in reinforcing the composite products. This case relates to the compatibility between the polyester resin (hydrophobic properties) and Sisal fiber (hydrophilic properties). Hence, the chemical modification to obtain better surfaces and a highly crystalline structure of fibers has been a wide area of recent research. It could be achieved by removing the hemicellulose, lignin and wax components on the surfaces of natural fibers [3, 4, 5]. This study focuses on the chemical treatment of Sisal fiber by NaOH solution with soaking time parameters to remove impurities and impact and increase the composite's mechanical and thermal strength.

1. **Materials and Methods**

*2.1 Materials*

Raw Sisal was obtained from the Sumenep, Madura Island, Indonesia. A hand processes it manually to remove the skin from the leaves and become the fibers. The Sisal fibers were soaked in distillate water and then dried in the sun for one day to remove residual leaves sap. Sodium hydroxide (NaOH) has 98% purity. It is used for the chemical treatment of Sisal fibers. Unsaturated Polyester Resin (UPR) Yukalac BQTN 157-EX series was used as the polymer matrix. They were obtained from PT. Justus Kima Raya, Semarang Indonesia.

*2.2 Fiber Surfaces Treatment*

The Sisal fibers soaking in the 5 wt.% NaOH solution for 2, 4 and 6 hours. It dried for four hours at a temperature of 100°C in the oven to remove the moisture. Sisal fibers are chopped uniformly into 10 mm lengths and then prepared using the press mould with 2 bar pressure to produce a Chopped Sisal-polyester composite (SPC).

*2.3 Scanning Electron Microscope (SEM) of Sisal fiber and UPR*

SEM image menggunakan mikroskop JCM -7000 JEOL/MP untuk mengobservasi bentuk morphology raw sisal and treated Sisal. The sample coated by platinum in a vacuum beam equipment. The SEM equipment operated on 5 kV to obtained clear image contrast. The micro-scale of fiber was observed by 5000 times magnification.

*2. 5 Fabrication of Chopped Sisal-Polyester Composite (SPC)*

The press-mold uses two mild steel plates with 250 x 250 x 2 mm dimensions. It press with hydroulic press with 2 bar pressure. The steel moulding were polished with a mould-releasing agent (mirror glaze) applied on the mould surfaces to remove the panel product easily. The unsaturated polyester resin (UPR) used Yukalac 157 BQTN-EX and hardener, namely Methyl Ethyl Ketone Peroxide (MEKPO). The ratio between UPR and hardener is 100:1. The volume fraction ratio between polyester and Sisal fiber is 70:30.

**Table 1.** Properties of cast-cured UPR Yukalac 157 BQTN-EX

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| Item | Unit | Typical Value | Note |
| Specific Gravity | - | 1.215 | 25oC |
| Gel Time | minutes | 20-30 | - |
| Heat Distorsion Temperature | oC | 70 | - |
| Water Absorption  (room temp.) | % | 0.188 | 24 hours |
| % | 0.466 | 7 days |
| Flexural Strength | MPa | 92 | - |
| Flexural Modulus | GPa | 2.9 | - |
| Tensile Strength | MPa | 53 | - |
| Tensile Modulus | GPa | 2.9 | - |
| Elongation | % | 1.6 | - |
| Compressive Strength | MPa | 123 | ISO-604 |

*2.6 Mechanical Characterization*

The mechanical properties of the composite are known through mechanical tests such as tensile, compressive and SEM photo tests. Several destructive tests were carried out, including tensile strength (ASTM-D638), flexural strength (ASTM-D700), and charpy impact (ASTM-D5942). The UTM (universal testing machine) is used for tensile and flexural tests. The gage length for tensile is 50 mm, 12 mm of width. The span for the flexural test is 70 mm, and 12 mm of width. The gage length for impact is 55 mm, and the weight of the Charpy pendulum is 1 Kg, with a 156° free swing arm (α). Each tests uses minimal five sample according ASTM (American Society for Testing and Materials).

*2.7 Fracture Analysis*

Fracture analysis after destructive tests observed by macro photos. The macro photos were obtained from an SEM image.

*2.8 Thermogravimetric Analysis (TGA) of Sisal fiber*

The TGA test, Mettler Toledo type, calculated the thermal degradation of materials. The temperature flow rate is 10°/min for a range temperature of 30 to 600°C through Nitrogen gas 20 ml/min. The 2-3 mg small chopped untreated, treated Sisal and polyester weighing and test. The mass loss degradation and oxidation along the temperature rise. The onset (Ton) and maximum temperature (Tmax) have been recorded.

1. **Results And Discussion**
   1. *SEM Morphology Sisal Fibers*

Morphological analysis of untreated and treated sisal observed fibers by SEM are shown on Figure 1. Figure 1a. shows micro bundles are covered by impurities, including hemicellulose, lignin and pectin. The amorphous material which is covered the cellulose causes crystallinity structure of natural fibers is low and its impact disrupt the bonding [6]. Figure 1b shows that soaking in 5 wt% NaOH for four hours is optimal for removing impurities on the surface of the Sisal fiber and increasing the CI. This soaking time causes the surface to become smooth and rough, thus expanding the cohesiveness between the fibers and the matrix. It affected decreases the diameter of Sisal from 130-180 μm to 60-100 μm. Figure 2. Diameter is measure with transversal position of single sisal fiber using the microscope optic like shown on Figure 2.

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| **Figure 1.** SEM image of Sisal fiber a) untreated, b) treated 4h, c) treated 6h |

The long soaking time for six hours causes the degradation of cellulose fibers (Figure 1c). Ismail et al. [7] investigates the effect of concentration and soaking time to influence on the diameter and surfaces roughness and amount of cellulose of the Kenaf fiber. It has been resulted a decrease on diameter caused removal the large content of impurities (lignin and hemicellulose) and wax on the outer layer that covered the cellulose.

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| **Figure 2.** The diameter of single Sisal fiber a) untreated, b) treated 4h |

The Agave Sisalana fiber has the same genus as Agave cantala fiber, which has similar chemical components and physical properties. Yudhanto et al. [8] stated the same results that the optimum treatment for Agave Cantala fibers is soaking them in 5 wt.% NaOH for four hours causes an increase in the percentage of composition, reaching cellulose 23%, reducing lignin up to 28% and hemicellulose 24%. That treatment decreases diameter smaller than untreated and it increasing the crystallinity index.

* 1. *Mechanical Properties of SPC*

The highest tensile strength and elongation at break were obtained on soaking time for four hours (4h) by an average of 31.2±3 MPa and 1.20%, respectively. Compared with untreated Sisal fibers, it increases tensile and elongation by 41% and 93%, respectively. A suitable concentration and soaking time are essential to get suitable surfaces to bind with UPR. After two hours of soaking (2h), the impurities were partially present on the surfaces, so it didn't result in a tensile stress optimum, resulting in 28.5±4 MPa. Treated for four soaking times, alkaline impacts the significant toughness properties of SPC. The treated fiber gave excellent intermolecular bonding between fiber and matrix. A prolonged soaking (6h) of Sisal fiber degrades the crystalline structure of cellulose, and it causes a decrease in the tensile strength of SPC to 21.1±2 MPa. Besides affecting roughness on fibers surfaces, the treated Sisal improves the aspect ratio (L/d). The high aspect ratio, suitable roughness surfaces and high crystallinity were essential factors that influenced the mechanical properties of fibers as reinforcement [9]. Likewise, the flexural strength on SPC-treated untreated Sisal by 59±3 MPa increases to 64±4 MPa. The treated 6h causes decreased flexural strength to 34±2 MPa. The best Impact strength on treated 4h is 116±3 kJ/m2, which causes the bundle Sisal fibers to have good elasticity. The prolonged treated 6h causes damage to the surfaces of fibers, affecting the mechanical strength of SPC (Figure 3). The previous research of Ayalew et al. [10] uses treated Sisal fibers from Debanke-mountain, Bahir Dar, Ethiopia. It was immersed in NaOH solution with 2, 6 and 10 wt.% concentrations to reinforcement polyester matrix with a ratio of 30:70. The optimum tensile and flexural strength of the composite was found at 44.0 MPa and 50.8 MPa, respectively, for concentration 6 wt.% NaOH. The chemical treatment for Paederia Foetida fibers (PFs) has been studied by Sari et al. [11]. They use 5wt.% NaOH for two hours of soaking time. The results increased the crystallinity index to 79%; thermal stability improved at a temperature of 330°C, and tensile strength increased by 42% higher than untreated PFs. The research by Nimanpure et al. [12] uses the same fibers, Sisal from Bhopal, India, for lightweight composite with resin polyester with a volume fraction ratio Sisal and polyester resin is 30:70. Treated Sisal with 5wt.% NaOH at room temperature for 72 hours increases the tensile and flexural strength by 41% and 30%, respectively.

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| a) | |
| b) | c) |
| **Figure 3.** The Mechanical properties of Chopped SPC; a) Tensile strength,  b) Flexural strength and c) Impact strength | |

* 1. *Fracture Analysis*

The results of tensile, bending and impact mechanical tests are shown in Figure 4. with different fracture shapes. Fracture tensile test shown on Figure 4a, flexural test on Figure 4b and charpy impact test on Figure 4c. After the destructive test, the fiber pull-out and debonding fracture mechanism test show that the fibers can’t bond well to the matrix because the cellulose is encased in lignin and hemicellulose on untreated Sisal fiber. The two-hour (2h) soaking time treatment produces a delamination type of fracture due to partial fibers still covered by lignin. The well-bonded fibers are found in the SPC-treated four-hour (4h) fracture mechanism. This happens because SPC-treated 4h is dominated by cellulose with a rough and smooth surface well bonded to the UPR-matrix. Crystalline cellulose has many hydroxyl groups (OH), which give intermolecular links to polyester resin [13,14, 15]. Fibers with a long soaking time (6h) gave poor bonding, as indicated by the brittle matrix cracking mechanism; it caused partial crystalline structure damage by alkaline. The SEM image shows in Figure 5 the excellent and poor interfacial strength between Sisal fibers and matrix. Figure 5a shows that alkali treated for four hours (4h) makes the surfaces rough and crystallinity cellulose increase; these impact excellent mechanical and chemical bonds between fibers and matrix. Figure 5b shows that alkali treated for six hours (6h) affected the damages or degraded the cellulose’s crystalline structure.

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| **Figure 4.** Macro image of specimen fractures after a) Tensile, b) Flexural, d) Impact test |

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| **Figure 5.** SEM image of chopped Sisal-polyester a) treated 4h and b) treated 6h |

* 1. *Thermal Gravimetric Analysis (TGA)*

Figure 6 shows TGA analysis on three material components: raw fiber (untreated), treated 4h and polyester (UPR). It has high resistance at an initial temperature (Tonset) of 30 to 220°C and does not experience evaporation or decomposition. Degradation occurred after a temperature of 220°C by 12 wt.%. Meanwhile, raw and treated sisal fiber experienced the highest evaporation in raw fibers by 7 wt.% and treated fiber by 6 wt.% at a temperature range of 30-100°C. In this range, water evaporation and volatile organic compounds often occur in natural fibers [16, 17]. The Tonset of treated Sisal (290°C) is higher than raw Sisal fiber (250°C). It happens because treated fiber has a higher crystallinity value than raw fibre because of the removal of the impurities [18]. The hemicellulose content quickly degrades in the 30-100°C temperature range and lignin in 100-290 °C. It causes the hemicellulose dominant water content and lignin to have hydrophilicity and structural plasticity. The maximum temperature (Tmax) of UPR has the highest resistance at a temperature of 400°C with a weight loss of 86 wt.%. It is similar to what was stated by Teh et al. [19] and Calabrese et al. [20], that UPR starts degradation in the temperature range of 220-400°C, and final degradation occurs at a temperature of 460°C. Meanwhile, raw and treated fiber has the same of Tmax at a temperature of 360°C with a lower weight loss of around 36-38 wt.%.

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| **Figure 6.** TGA curve on the raw, treated 4h and polyester |

1. **Conclusion**

Chopped treated sisal soaking in 5% wt. NaOH solution decreases the diameter, causing the removal of impurities components such as hemicellulose and lignin. The optimum treatment on the four hours soaking time to optimum good surface roughness, high crystallinity index, and hydroxyl group (OH) makes excellent inter-mechanical bonding between the cellulose and UPR resin. Adding 30% chopped treated fiber causes the SPC to improve the mechanical properties such as tensile, flexural, and impact strength by 41%, 8%, and 61%, respectively. The SEM image shows fractures of untreated SPC dominate the fibers pull-out, which indicates poor bonding. The treatment of fiber increases the cellulose content and removes the amorphous material. It causes good mechanical properties. The thermal stability of untreated fiber is lower than treated. The UPR is a hydrophobic matrix with high heat resistance on the initial temperature stable from 30 to 180 °C, and it has a maximum temperature of 400°C.

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