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SYNTHESIS OF ENVIRONMENTALLY FRIENDLY, RENEWABLE, AND SUSTAINABLE NATURAL FIBER COMPOSITE BASED ON POLYLACTIC ACID/COIR WITH BENTONITE ADDITION

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ABSTRACT
A natural fiber composite based on Polylactic Acid (PLA) polymer with the addition of coir and bentonite as fillers has been synthesized using several variations of bentonite. The variations in bentonite filler were 0 % (PCB0), 2 % (PCB2), 4 % (PCB4), 6 % (PCB6), and 8 % (PCB8). The samples were compared to pure PLA (PCBx) polymer. The tensile strength of the composite increased with the increasing concentration of bentonite filler. The result of the pure polymer sample PCBx tensile test was 10.51 MPa. The addition of 6 % bentonite filler showed a higher tensile strength of 19.43 MPa. In the thermal test, the best results were obtained with the PCB8 sample, which had a decomposition temperature of 381°C. SEM testing for polymers with a mixture of bentonite in the PCB8 sample showed that the surface structure was smoother and more homogeneous compared to the PCB0 sample.
Keywords: Composite, Natural Fiber, Polylactic Acid, Bentonite, Coir, Tensile Strength.
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INTRODUCTION

The demand for natural resources has increased due to the constant advancement of science and technology. These developments raise concerns about environmental preservation and material scarcity. Researchers are now exploring new materials that are biodegradable, renewable, and recyclable due to the rapid depletion of oil reserves and the greenhouse gas emissions caused by the widespread use of petroleum-based products and their inability to biodegrade.[1] Biodegradable composites are among the materials that can help address this issue and ensure a sustainable environment. These composites are reinforced with cellulose fibers and have matrix materials made from agricultural and forestry raw materials. Composites are used because their natural decomposition does not harm the environment.[2,3] Utilizing bio-composites has special advantages, such as renewability, biodegradability, low density, and low cost, which is a crucial quality for commercial applications.[4] Additionally, they provide adequate acoustic and thermal insulation in addition to flexibility. Furthermore, since bio-composites are non-toxic and do not spread disease, they are safer to use than synthetic composites.[5,6] Because of this, bio-composites are a desirable replacement for synthetic composites. They have a wide range of applications in a variety of industries, including the automotive, aerospace, building, defense, biomedical, sporting goods, and packaging sectors, among others, because of these advantages. There are many potential candidates for the role of matrix material in bio-composites, including starch, soy resin, gelatin, polyhydroxy butyrate (PHB), polyhydroxy butyrate-co-valerate (PHBV), poly lactic acid (PLA), and others, both natural and synthetic biopolymers.[7] One of these is polylactic acid, which has been proposed as a potential substitute for synthetic plastics made from petroleum processing byproducts. This is one of the possibilities. It is made from a variety of crops, including corn and sugar beets, and is a material that is both renewable and biodegradable. It has excellent mechanical strength and a lot of stiffness, and its composites can be produced using traditional production methods.[8,9] In addition, a life cycle analysis of PLA revealed that it uses only about a tenth of the energy required to produce fossil resins alone, as opposed to synthetic polymers. These factors make polylactic

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acid (PLA) a superior substance for the creation of biodegradable composites. In the past few years, natural fibers have become a very popular material.[10,11,12,13]This new material, which is made of biodegradable polymers and plant fibers and is usually called a site biocomposite, has been given the name site biocomposite. It was found that some materials, such as coir, could be used to reinforce thermoplastics.[14] Maleic anhydride is often used in the reaction process as a "coupling agent".[15] This is done to fix the fact that fiber and starch with high polarity and hydrophobic PLA don't stick well to each other on the surface. Additionally, PLA biocomposites are made using detailed manufacturing processes.[16] According to the tensile test, TGA (thermal) DSC test, and scanning electron microscopy (SEM) test, bentonite type and shape increase biodegradability, mechanical properties, thermal properties, and morphological structure composites. These findings indicate an increasing molecule. If PLA and Coir polymers are made with bentonite filler, their mechanical properties may change.[17] In this study, PLA reinforced with coconut coir and bentonite filler is examined.[18] This study investigates how natural fiber support affects PLA's thermal, static, and dynamic mechanical properties. PLA/Coir polymer with Bentonite was used in sample formulations of 0%, 2%, 4%, 6%, and 8%.

EXPERIMENTAL

Material and Methods

Materials Polylactic Acid (manufactured by Nature Work Co.), Coir (also known as coconut coir), and Bentonite sourced from the North Aceh district were utilized throughout this research. It took 18.2 grams of CTAB to produce 0.05 moles of CTAB solution when it was dissolved in 250 milliliters of distilled water. After that, the heating process was utilized to treat this solution for an hour at a temperature of 80 degrees Celsius. After that, up to 20 grams of bentonite were put into a volume of aquadest that was equal to 500 milliliters. The previous CTAB solution was stirred together with this solution, but this solution was stirred separately. Following that, the bentonite dispersion was added to the CTAB solution, and the mixture was stirred for an additional hour. Following that, the filtrate was titrated with silver nitrate to remove any chloride or bromide that might have been present. Bentonite was allowed to dehydrate in an oven set to 60 degrees Celsius before being strained through a sieve tray with a 100-micron aperture. To obtain nanoscale bentonite, the aforementioned process needed to be carried out. Other additives include Cetyl Trimethyl Ammonium Bromide or CTAB, which can be purchased from Sigma-Aldrich, and $(Na_2PO_3)_6$ from Merck

General Procedure

The synthesis of the material was carried out by mixing the matrix and reinforcement. The matrix in the form of *polylactic acid* was first melted, and then the mixing process was carried out manually by inserting the fiber into the matrix through a stirring process. Several samples were prepared according to the mixture, i.e. pure PLA, PLA/Coir, and PLA/Coir and Bentonite. Pure PLA without Coir and Bentonite labeled as PCB_x, PLA/Coir without Bentonite (PCB_0), PLA/Coir + Bentonite 2% (PCB_2), PLA/Coir + Bentonite 4% (PCB_4), PLA/Coir + Bentonite 6% (PCB_6), and PLA/Coir + Bentonite 8% (PCB_8). Samples were melted and blended at a temperature of 150 ºC and they will harden on their own to form PLA/Coir without and with Bentonite addition composite. The samples were cut into granules and placed in specimen molds according to ASTM 638 D Type IV Standard and compacted by the hot press at 180 ºC. The samples were dried in a vacuum oven at 65 degrees Celsius for 24 hours.

Synthesis of Bentonite

Bentonite was taken from North Aceh Regency. Bentonite is then dried at 105ºC and stored in a desiccator, then partially separated as a pure bentonite sample. Then purification of the sample was done to get montmorillonite. The process of purification is through 3 stages, grinding, dispersion, and centrifugation. Raw materials of bentonite were smoothed using a grinder for 60 minutes, then sifted with a 230 mesh sieve. 3 grams of fine bentonite and dissolved in 36 grams of free water mixed with 0.03 gr $(NaPO_3)_6$ or 1% of the weight of bentonite. The mixture is then stirred using a magnetic stirrer for 24 hours. After stirring, then centrifuging at a speed of 700 rpm for 2 minutes. The precipitate obtained is then heated in an oven at a temperature of 60ºC to a constant weight. Furthermore, the bentonite sample was reprocessed with the addition of CTAB (Cethyl Trimethyl Ammonium Bromide) as a surfactant to modify bentonite to form organo bentonite.

Synthesis of Coir

The fiber must undergo a special treatment that will both clean and alter its size before beginning the process of material synthesis. This treatment is an alkalinization procedure. Coconut fiber is alkalized by soaking it for two hours in a solution of aquades and 5% NaOH which produces the highest mechanical value of coconut fiber. After the soaking process was complete, the coconut fibers were blended for twenty minutes with a lye solution. Then the fiber was dried in an oven at 100 degrees Celsius for one hour after removing all of the alkaline liquid.

Detection Method

The tensile properties of PLA/Coir with and without bentonite nanocomposite molded according to ASTM D 638 standard specimen were tested by axial force from the tensile test equipment until it broke.[19,20] Engineering tensile tests provided strength data for material specifications and basic design information.[21] The test object's elongation is measured as the axial tensile force load increases. The Shimadzu DTG-60 TGA instrument was used to study sample mass change as a function of temperature, time, and atmosphere. This method measured material mass loss from room temperature to 800 C at 20 C/min.[22] Raising the sample's temperature gradually and calculating the large temperature change is the analysis. All specimens were nitrogen gas-tested. The Scanning Electron Microscope (SEM) is an equipment used to determine the morphology and surface structure of materials, as well as their chemical composition.[23] Electromagnets and electrostatics were utilized by this microscope to control the incoming light and the appearance of the produced image.

RESULTS AND DISCUSSION

Tensile Test Analysis

The tensile test results for pure PLA composite, PLA/Coir with and without Bentonite at varying Bentonite concentrations.

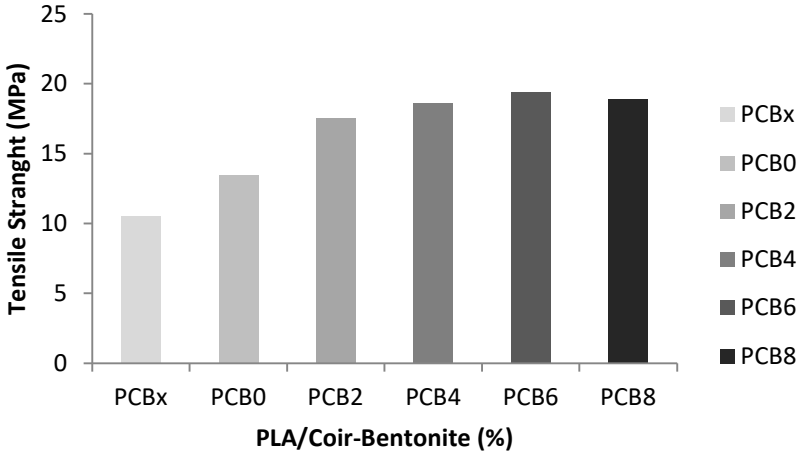


Fig.-1: Tensile Strength of the Samples Formulation

The addition of bentonite, which forms a composite with PLA/Coir, results in a significant increase in tensile strength compared to composites without bentonite. The resulting new material exhibits an improved quality of its strength properties. The percentage of filler influences the value of the tensile strength proportionally (Fig.-1). The greater the percentage of bentonite mixed into the PLA matrix, the greater the tensile test value produced. However, if the filler is overloaded, the tensile strength value will decrease. The cause of the decrease in the tensile strength value can be attributed to the occurrence of dispersion and the interaction between the plasticizer and the cellulose ring structure through hydrogen bonds, without interaction between the plasticizer and the cellulose chain (glycosidic bonds). This incident resulted in the cellulose ring not moving much because there was little steric hindrance so the chain bonds became strong. The tensile test results for pure PLA samples without the addition of coir-bentonite filler obtained a tensile strength value of 10.51 MPa. It can be seen that pure PLA samples have brittle chain bonds. In the sample without the addition of Bentonite, the tensile strength value is 13.45 MPa and brittle. With the addition of

Bentonite the tensile strength of PCB_2, PCB_4, PCB_6, and PCB_8 samples, 17.54, 18.63, 19.43, and 18.87 MPa, respectively. The highest tensile value was achieved on the PCB_6 sample with the addition of 6% bentonite, which was 19.43 MPa. Along with limitations, technical solutions to the traditional problems of natural fiber composite processing, tensile properties improvement, durability, thermal stability, and interfacial incompatibility enhancement are discussed. [24] The alternately arranged structure with thin and thick layers contributes substantially to the mechanical strength of the bamboo fiber [25], so thermoset polymer-based coir composites typically show higher mechanical strength than thermoplastic-based coir composites.

Thermal Degradation

The Thermogravimetric analysis is carried out to study the thermal stability of natural fibers and their composites. The physical and chemical properties can be determined as a function of increasing temperature while maintaining a steady heating rate.[26] The thermal degradation of natural fiber occurs consequently from hemicellulose, cellulose, lignin, wax, and other components with an increment of temperature.[27] The decomposition process, which is the breakdown of chemical bonds, is what causes the mass to be lost during the heat test. A graph of the TGA test results for 6 samples, including the PCB_6 sample with the addition of 6% bentonite filler (Fig.-2). This sample had the highest value in the prior test.

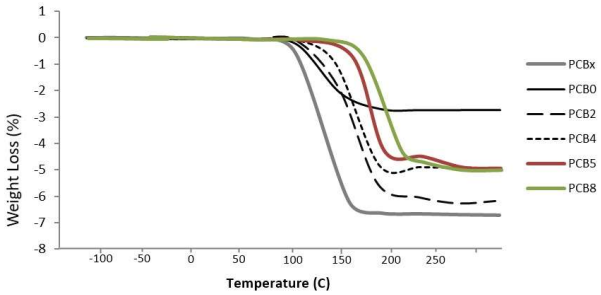


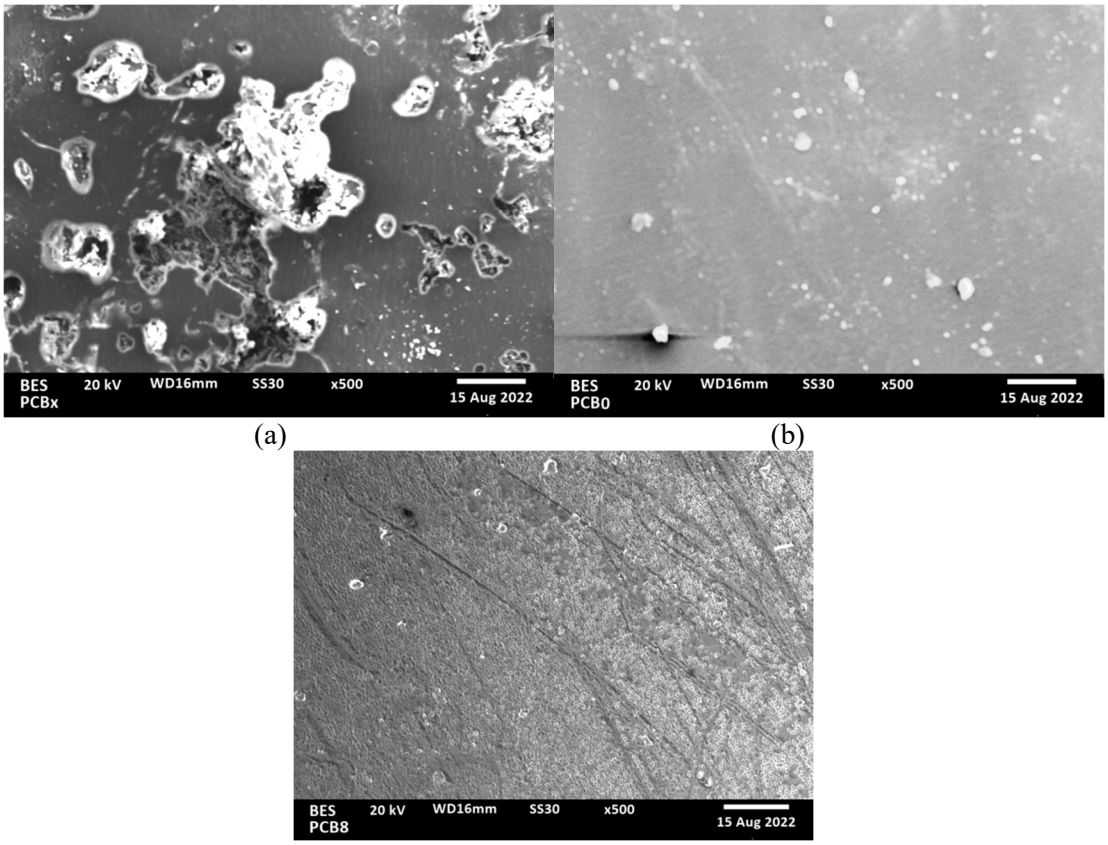
Fig.-2: Thermogravimetric Analysis of the Sample

The Figure demonstrates that there is only one breakdown for all samples. The degradation temperature range is between 96.89 and 181.94 °C; at temperatures between 100 and 200 °C. The mass of the samples is lost due to the evaporation of their water content. The PLA/Coir Bentonite composite mixtures have better thermal stability than those without a mixture of coir-bentonite filler. PCBx sample was degraded at a temperature of 96.89 °C. PCB_0 and samples with the addition of filler-coir-bentonite were degraded at a temperature of 102.21°C. Meanwhile, PCB_2, PCB_4, PCB_6, and PCB_8 samples begun to degrade at temperatures of 126°C, 141.76°C, 165.64°C and 181.94° C. These results show that the inclusion of this bentonite filler mixture has been successful in enhancing the thermal stability of the polymer without bentonite, which is characterized by an increase in decomposition temperature. The increase in degradation temperature is caused by the polymer and filler bonds which melt more powerfully, hence it is hard to break and the decomposition of the material becomes slower. In samples with the addition of bentonite, the best results were obtained on PCB_8 samples because the more additional bentonite filler in the sample the better the thermal stability. The increase in temperature degradation is triggered by the bonding of the polymer and filler which merged hence it is difficult to break and the decomposition of the material becomes slower.

Morphology

The natural fibers/coir are utilized as reinforcement material when producing composites. The fiber surface inconsistencies may be caused during the extracting process, because of the fiber's age, ecological effects, and variation of soil/bentonite. Therefore, the manufacturer should be keen on sample preparation.[28] Further, the understanding of the behavior of acoustic and thermal properties, and studying surface morphology of coir is very significant.[29] To visualize the surface of a sample, an electron microscope known as a scanning electron microscope employs a high-energy electron beam in a raster scan pattern. As a result of electron-atom interactions, the sample produces signals that can be used to determine its surface topography, chemical makeup, and other characteristics like electrical conductivity.[14] The image below depicts the analysis using Scanning Electron Microscopy (SEM). Scanning Electron Microscopy (SEM)

results on PLA/Coir without the addition of bentonite filler with PLA/Coir/Bentonite during the polymerization process for 2 hours revealed that the surface structure of polylactic acid (PLA) was widespread due to sunlight electromagnetic.



(a) (b)

(c)

Fig.-3: SEM Test Results on the Sample (a) PCB_x (b) PCB_0 (c) PCB_8

Internal structure Observations utilizing SEM in the fault location revealed the existence of voids and fiber breaks within the micro-composite material's internal portion (Fig.-3). Certain fibers exhibit pull-out during tensile and bending testing. elongation at break instead of PLA In general, the elongation at break of coir fibers is significantly larger (15-40%).[15] Compared to other natural fibers such as jute and sisal, which have comparatively low elongation values of 1.16-1.5% and 3-7%, respectively,[16] at 500 x magnification the elongation of hemp is significantly greater. Based on the results of the characterization of the surface morphology seen from the SEM results, the surface structure of a single polymer has a different surface. On the surface structure of a single sample PCB_0. In sample PCB_8, the morphology image only has a few cavities and pores. This shows that the sample with the addition of 8% bentonite filler has better quality than the previous sample because it can be seen that there are only a few cracks and the bond between the matrix and the aggregate is getting better.

CONCLUSION

The results showed that the tensile strength of the composite material comprising coconut coir fiber and Poly Lactic Acid (PLA) with the highest addition of bentonite filler was achieved in the PCB_6 sample, which had a tensile strength of 19.43 MPa. Meanwhile, the lowest tensile strength was observed in the composite material with a mixture of bentonite filler in the PCB_2 sample, with a value of 17.54 MPa. The sample without bentonite reinforcement (PCB_0) had a low tensile strength value of 13.45 MPa. In addition, the interfaces between the filler and fiber are not adhesive, which means that the stress distribution from the matrix to the fiber does not occur effectively. Furthermore, the presence of excess filler can be seen in the PCB_8 sample, where the addition of excess filler can cause the stress in the matrix to be ineffective (i.e., overloaded). In the TGA test, it was found that the PLA/Coir composite mixture with the addition of

bentonite filler exhibited better thermal stability capabilities. Specifically, the PCB_8 sample demonstrated a thermal degradation temperature of 181.94 °C, whereas the PLA/Coir mixture without bentonite filler on the PCB_0 sample degraded at 102.21°C. These results indicate that the use of bentonite filler in the fiber modification process has successfully increased the thermal stability of the composite. The better thermal stability of the material is characterized by an increase in temperature degradation. Observations made using SEM revealed that there were voids and fiber breaks present in the internal morphology of the micro-composite material produced. In comparison to PCB, the surface of the PCB_8 sample appears more uniform, smooth, and scattered.

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CONFLICT OF INTERESTS

The authors declare that there is no conflict of interest.

AUTHOR CONTRIBUTIONS

All the authors contributed significantly to this manuscript, participated in reviewing/editing, and approved the final draft for publication. The research profile of the authors can be verified from their ORCID IDs, given below:

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