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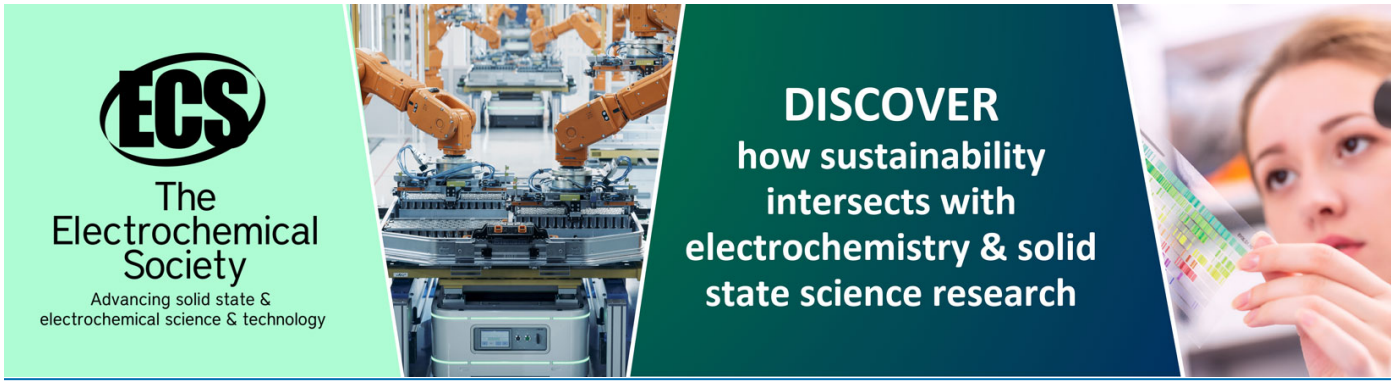

The Manufacturing Process of Recycled Polymer Composites reinforced with Natural Fibers – A Systematic Literature Review

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Abstract: In the light of growing global awareness in environmental sustainability, it is essential to employ more "environmentally friendly technology" which preserve natural resources. In the past decade, natural fiber composites with thermoplastic and thermoset polymers have been applied by several industrial sectors in various parts of the world. Natural fibers such as kenaf, hemp, sisal, abaca, and sugar palm have been widely used because they provide several advantages such as low cost, environmentally friendly, and reducing dependence on the use of materials that cannot be recycled. The use of recycled material for the development of natural fibre reinforced polymer (NFRP) has seeking current attention. This article reviewed several literatures on the manufacturing process of recycled polymer in NFRP material. The five-step systematic literature review (SLR) was chosen as the approach in reviewing the literature. The analysis of the selected articles revealed that a successful process for making recycled natural fiber reinforced polymers begun by appropriate treatment of natural fibers. Treatment of natural fibers using alkaline treatment (sodium hydroxide (NaOH) and silane treatment is the most influential factor in the success of the process of making recycled polymer composites reinforced with natural fibers. This study implies a new manufacturing trend for natural based fibre composites in recycled polymer matrix.

Keywords: *Natural Fibers, Manufacturing process, Polymer Composites, Systematic Literature Review.*

1. Introduction

Composites are materials that consist of a strong load-carrying material (known as reinforcement) combined with a weaker material (known as a matrix). Natural fiber-reinforced polymer composites (NFRP) derived from renewable resources have received attention from various sources circles for use in several applications, including in the transportation sector.

Natural fiber is a renewable resource and currently, it has received a good response in the community. Natural fibers also offer many advantages as an alternative to polymer fillers in the composite manufacturing process. Natural fibers have many advantages in the manufacturing process: a) It has a high modulus of breakdown elasticity, b) has a high breakdown strength, c) is safe for health d) low density, e) is renewable, f) its availability in nature is very abundant around we [1].

The use of plastic materials has a positive impact on technological advances that are beneficial for the society in meeting their daily needs. [2]. Indonesia is the 2nd country in the world as the largest producer of plastic waste after China, which produces 197.2 million tons of plastic waste in the sea [3].

The processing of recycled plastic waste through physical and chemical processes to produce reusable plastic, types of plastics that can be recycled: PETE (Polyethylene Terephthalate), PVC(Polyvinylchloride), LDPE (Low-Density Polyethylene), PP (Polypropylene), PS (Polystyrene), and HDPE (High-Density Polyethylene) [3].

In addition to the law on environmental awareness, several sectors of the transportation industry reduce the use of carbon-based materials. In this context, NFRP composite materials are a good choice because they are low cost, less polluting to the environment, and have relative properties equivalent to metal materials. The purpose of using natural fibers as reinforcement for recycled polymers is one step that can be taken to reduce the increase in plastic waste and the utilization of abundant natural resources by combining the two properties of these components to produce reinforcing components that can improve bio composite properties with low cost and strength the good one.

2. Research question

In the manufacturing process used for natural fiber-reinforced polymers, several factors can play an important role in determining the mechanical properties of natural fiber-reinforced polymers such as processing factors, the temperature used, thread speed used, manufacturing process time, and fiber treatment before the manufacturing process, such as chemical methods and physical methods. Also, the manufacturing processes used, such as the Injection Molding, Compression Molding, Extrusion Molding, and Continuous Pultrusion Molding processes can also affect the mechanical and physical properties of polymers reinforced by natural fibers [4].

The manufacturing process of natural fiber reinforced polymers must select the correct composite processing techniques and conditions so that it will result in the transformation of polymers and natural fibers into composites without causing structural damage to the composites formed.

Thermally, the natural fibers are unbalanced when compared to the polymer matrix which has a high processing temperature. Where the thermal degradation of the polymer starts at 2000°C [5]. This can cause a decrease in the mechanical performance of the resulting composites because differences in thermal stability can limit the efficiency of the fibers in the polymer matrix. The main challenges faced in the manufacture of polymer composites are natural fibers which have hydrophilic properties and polymers that have hydrophobic properties which cause poor interface adhesion. This can lead to poor load transfer as the mismatch reduces adhesion between the fiber and polymer.

The method in making natural fibers as polymer reinforcement can be done by increasing the adhesion of natural fibers in the matrix and increasing the mechanical strength of the composites by increasing the hydrophobicity of the fibers in the fiber surface treatment. Usually, modification of natural fibers can be done by using alkalization of 5% NaOH solution which can increase the interface bond between the fiber and the polymer matrix. However, it also depends on the type of fiber to be used as polymer reinforcement [18].

3. Methodology

The Systematic Literature Review (SLR) was chosen as a systematic approach methodology to review the literature on natural fiber-reinforced polymers. Systematic Literature Review is carried out with a systematic approach where research questions exist based on the process of identification, assessment, and interpretation of all studies. This review is the Systematic Literature Review (SLR) [6]. According to Khan, Kunz, Kleijnen, and Antes, the process in this Systematic Literature Review is divided into five stages [7]. These five stages are formulating questions, identifying appropriate/relevant articles, assessing the quality of literature studies, summarizing some evidence, and interpreting the final findings [8].

The research objectives were based on the research questions that had been prepared. The procedure for searching for relevant articles is adjusted based on predefined inclusion and exclusion criteria. After that, a quality assessment is carried out and an in-depth evaluation is carried out on each article. The evaluations that have been carried out are then used as a basis for summarizing the evidence and avoiding the risk of evidence bias. The selected articles are then collected and can be used as data to answer the

research questions that have been compiled. There is data synthesized and presented consistently to answer research questions.

The final stage is to interpret several findings, where the findings are based on answers to research questions that have been developed previously. The conclusions presented must also match the available data.

4. Result and Discussion

Based on the five stages and the SLR methodology, the results of a study of the natural fiber reinforced polymer manufacturing process are described in the following sections:

4.1 First Step (Farming the question)

This Systematic Literature Review will discuss what researchers have done in previous research on natural fibers as polymer reinforcement. These following research questions will be used for guiding the literature review:

- P1: How to manufacture the Natural Fiber Reinforced composite using Recycled Polymer?
- P2: What are the challenges in the manufacturing process of Natural Fiber Reinforce composite using Recycled Polymer?
- P3: How to overcome the challenges in processing Natural Fiber Reinforce composite using Recycled Polymer?

Table 1. Research Question and Motivation

Code	Research Question	Motivation
P1	How to manufacture the Natural Fiber Reinforced composite using Recycled Polymer?	Identify the NFRP manufacturing process and the steps in the NFRP manufacturing process
P2	What are the challenges in the manufacturing process of Natural Fiber Reinforced composite using Recycled Polymer?	Identify challenges that occur during the NFRP creation process
P3	How to overcome the challenges in processing Natural Fiber Reinforced composite using Recycled Polymer?	Identification of suitable methods for NFRP production and how to overcome challenges in the NFRP manufacturing process

4.2 Second step (Identify relevant articles)

The search for articles was carried out on five suitable scientific databases, namely the Sage Journal, Springer, Science Direct, Elsevier, and IEEE. Several search procedure techniques were used in collecting research articles that were suitable for this study. The first step taken is to identify research that fits the objectives of this study. The next step is to conduct an article search using the following search terms: Natural fiber composites. A total of 124,407 hits were obtained from three scientific databases. The search results were filtered using the following advanced search terms: ("Natural fiber-reinforced polymer" AND "Natural fiber-reinforced recycled polymer") which resulted in 355 hits.

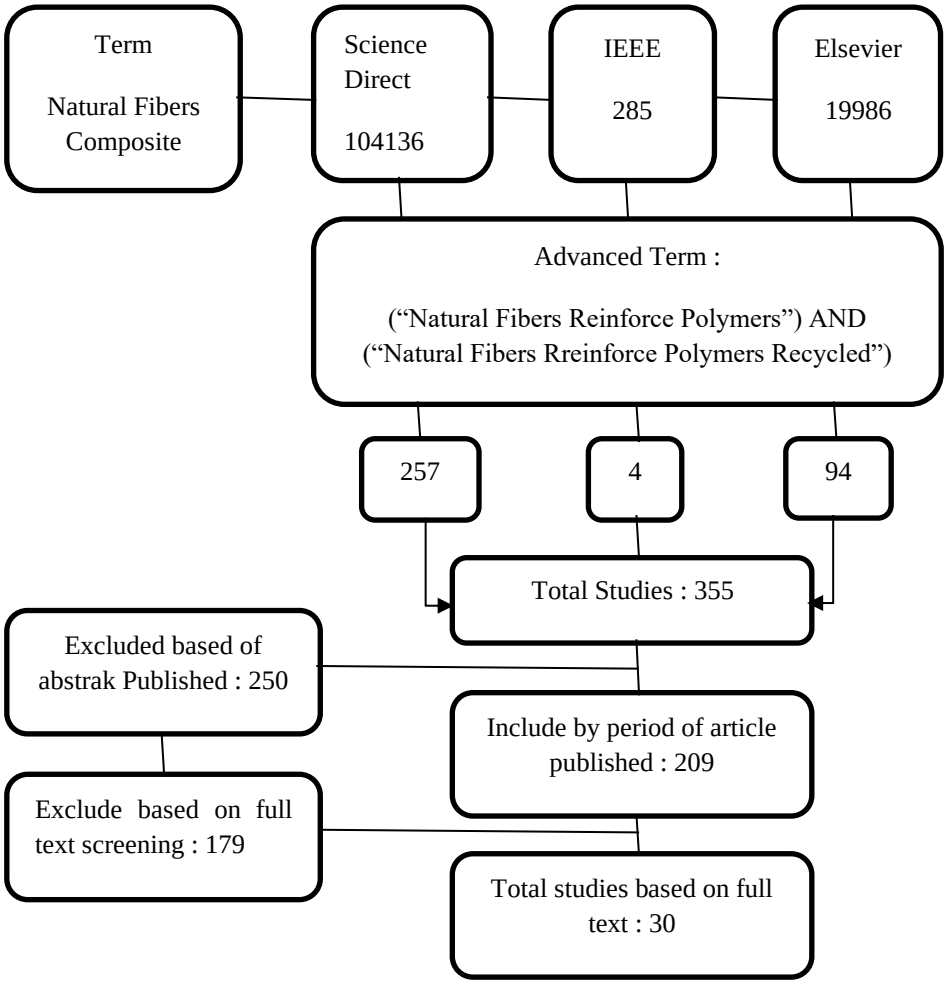

Figure 1. Literature Search Procedure

Article searches will be excluded if there are: (a) an incomplete text article is available to the author, (b) the article comes from a book review. The authors will select relevant articles by assessing the full text against the criteria for inclusion in the literature review. Therefore, the last 30 articles were included in the literature review. An overview of the literature search procedure is shown in Figure 1.

4.3 Third Step (Study Quality Assessment)

The purpose of this study is to examine what previous researchers have done regarding the manufacturing process that implements NFRP manufacturing, the challenges, and constraints experienced during the NFRP manufacturing process, and the appropriate methods in the manufacture of NFRP composites regarding the process of making polymers reinforced by natural fibers. The full text is selected based on the article corresponding to that component. The assessment should describe the manufacturing process for recycled polymers reinforced by natural fibers, such as the processes used in polymer manufacturing, how the fibers are treated during the manufacturing process, and the appropriate method for the type of composite to be made because the different types of composites are different and the manufacturing methods used. Furthermore, there is a process of examining how appropriate methods of fiber treatment are to improve the chemical and mechanical properties of composites and whether those methods are supported by relevant data or theory. All articles used as references in this literature review met all of these criteria.

4.4 Fourth Step (Summarizing the evidence)

At this stage, it begins by providing an overview of the substance of the article that has been selected, accompanied by an explanation of the results for each of the three research questions. The research background of the 30 articles examined in this study were natural fiber polymers and recycled natural fiber polymers.

The majority of the studies used extrusion molding methods (50%), followed by compression molding methods (25%), and injection molding methods (25%). An overview of the method of making recycled natural fiber reinforced polymers is shown in figure 2.

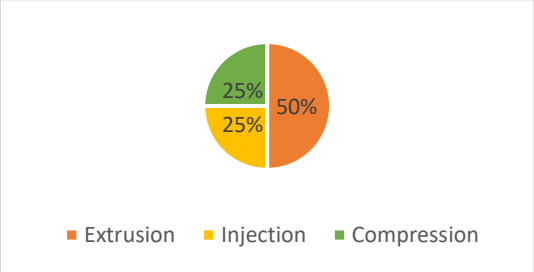

Figure 2. Polymer Manufacturing Methods

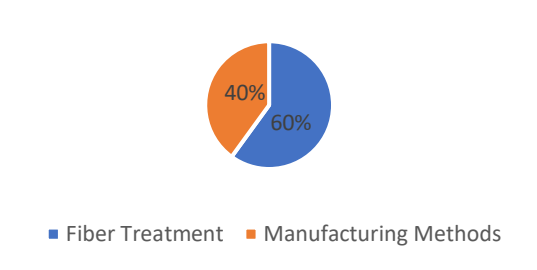

Figure 3. Manufacturing Success Factors

This literature study identifies several aspects that can affect the success of the natural fiber reinforced polymer manufacturing process. Fiber treatment is a major factor influencing the success of making recycled polymers reinforced with natural fibers. The description of the factors for manufacturing success is shown in Figure 3.

4.5 Step Five (Interpretation of findings)

4.5.1 How to manufacture Natural Fiber Reinforced composite using Recycled Polymer?

Determining the process of making recycled polymer composites is an important factor in determining the mechanical, physical, chemical, and thermal properties of the polymer to be produced. The process of making Natural Fiber Reinforced Polymers has many choices, depending on the type and use of the composite that will be produced later. Setyanto [5] The process of making composite materials consists of two ways, namely a) open printing process, including Hand, lay-up process, Vacuum bag, Pressure bag, Spray-up, and Filament winding, b) closed molding process, including compression printing, molding Injection, and Continuous Pultrusion.

Table 2. Classification of Composite Manufacturing Techniques based on the type of mold

Composite Manufacturing Process	
Open Molding	Closed Molding
Hand Lay-Up	Compression Moulding
Filament Winding	Pultrusion
Spray Up	Resin Transfer Moulding (RTM)
	Centrifugal Casting
	Vacuum Infusion Processing
	Vacuum Bag Moulding Vacuum Bagging Injection Moulding
	Continuous Lamination Vacuum-assisted RTM (Va-RTM) Co-Injection RTM (CIRTM)
	Reinforced reaction injection moulding (RRIM) Structural reaction injection moulding (S-RIM)

Daniel [8] The process of making polymer reinforced bamboo fibers using Hand Layup, called the Lay-up process, is a method of making polymers by filling the matrix into the mold by hand on the bamboo fibers that are already in the mold. then woven, stacked, and tied. The emphasis in this method is using rollers so that the interface bond of the matrix and bamboo fibers will be tight.

The bio composite manufacturing process by Chaitanya [9] carries out a bio composite manufacturing process developed by an Extrusion-Injection molding process. Commercial-scale extrusion and injection molding machines are used to make test specimens. The dry Aloe Vera fiber and PLA pellets were mixed with a weight ratio of 30:70. The fiber-matrix mixture is then liquefied with the compound at 180℃ using a single screw extruder. The extrudate obtained in the form of strands is then pelletized using a pelletizer and then injection molded into the desired specimen.

Munoz [10] carried out a bio epoxy composite manufacturing process with a liquid composite molding process (LCM) consisting of the manufacture of a composite from a polymer matrix by impregnating dry fibrous cloth in a closed mold with low viscosity resin. The resin is injected into the mold with a pressure difference between the resin inlet and the air outlet. The pressure difference can be positive as a resin transfer molding (RTM) process shown in Figure 1 or negative as a vacuum injection process. The process starts with the injection of the resin into the mold and ends when the resin completes the chemical curing reaction and the part is removed. The polymerization reaction is highly exothermic and varies in duration depending on the type of resin, the inhibitor used, the temperature of the resin, and especially the ratio of resin to an inhibitor. The injection pressure should be low enough to provide proper fabric impregnation and to ensure that it is not displaced by resin flow in the mold. This pressure varies between 0.5 bar for vacuum injection and 3 bar for pressure injection.

Gorbunova [11] carried out a production process using a hot-pressing method for polymer composites heated at 180℃ with a pressure of 10 MPa for 20 minutes to produce polymer composites.

Siaotong [12] conducted a study on the manufacture of polymers by extrusion experiments using a co-rotating twin-screw extruder with a screw diameter of 38 mm and a length/diameter ratio of 38: 1. The extruder is electrically heated (with five heating zones) and water-cooled. The screw speed is adjustable. The blend of hemp fiber and polymer matrix was extruded based on a factorial design of three experimental process variables. In this case the extrusion parameters such as screw speed 110 and

150 rpm, barrel zone temperature (75, 110, 120, 130, and 140℃ [T1] and 75, 120, 130, 140, and 150℃ [T2]), and fiber content (0%, 12.5%, and 25%). The final product is an extrudate, which is pelletized and ground before rotational molding.

Favaro [13] researched the process of making composites using a sisal fiber reinforced HDPE matrix with two variables (modified and unmodified). Components are mixed in a laboratory made mono screw extruder with screw specifications 25/700 mm (diameter / length) L / D = 28. Cylindrical temperature profiles set to 110, 155, and 185℃ for three heating zones with an average rotation of 40 rpm for all formulations. After extrusion, the material was cooled with water and then allowed to stand at room temperature. The extrusion results are then molded using an injection machine with a screw diameter of 42 mm at a temperature of 180℃ and an injection pressure of 35 bar.

Tabarsa [14] carried out polymer production using the Extrusion-Hot Press method with double screws. Wood powder mixtures with variations of 30, 35, and 40% and Recycled PP 6% are manufactured using extrusion machines with temperature parameters of 165, 170, 175, and 180℃ and the mold temperature is maintained at 185℃. The screw speed is 60 rpm. The extruded product is passed through a water bath and pelletized. The resulting granules were then placed in a hot press at 175℃ for 10 minutes. then cooled to room temperature under pressure. The pressure for heating is controlled at 35 bar. The method above can be seen in the picture 4.

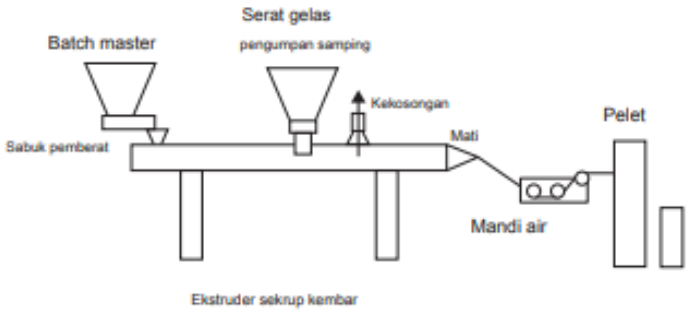

Figure 4. Extruder with double screws

In the process of making polymers using recycled polymers, mechanical properties, physical properties, and thermal properties are statistically comparable and even have better strength than composites made of pure plastic. The use of recycled polymers is thought to allow for expanding use in the manufacture of polymer composites. The process of making recycled natural fiber reinforced polymers using the Extrusion-Injection process is a process capable of producing good polymer composites, the extrusion process is used to homogenize polymers and fibers and produce composites that are easy to work with. The injection process is used to reduce extrudates which can then be used for the manufacture of new composite products [15].

Among the many manufacturing processes available for producing natural fiber reinforced plastic products, injection molding (IM) techniques are used to make at least 50% (by weight) SFRP. Molded parts produced by the IM technique have the advantages of economical, quantity, and no post-finishing operation. IM is primarily a mass production method and provides the best returns on use [16].

Each natural fiber reinforced polymer matrix composite manufacturing technique has important value and unique operating features. Each Polymer Composite Manufacturing technique adjusts to the

volume of products produced and the type of structural application in the industry indicated by the composite product.

4.5.2 What are the challenges in the manufacturing process of Natural Fiber Reinforced composite using Recycled Polymer?

In the process of making recycled polymers reinforced by natural fibers, several challenges occur during the manufacturing process. Malkapuram [17] conducted a study on the main problems that occur during the manufacturing process of the incompatibility between hydrophilic natural fibers and hydrophobic thermoplastic matrices. This requires the use of a compatibilizer or coupling agent to increase adhesion between the fiber and matrix.

Saba et al [18] suggested that the challenges experienced in the natural fiber manufacturing process with polymer composites such as hydrophilic properties, poor wettability, poor adhesion of the fiber/matrix interface, the poor thermal stability of natural fibers, and incompatibility with multiple polymer matrices.

Poletto [19] noted a deficiency in the process of making natural fiber-reinforced polymers in that the heat stability of natural fibers decreases with increasing processing temperature. Degradation of fibers occurs due to chemical and physical changes with heating between 100-300℃.. Some of these important changes include dehydration, fiber discoloration, recrystallization, hydrolysis, oxidation, decarboxylase, and depoliticization. In this study, hemp and banana fibers showed that temperatures above 170℃ significantly reduced the mechanical and polymeric structure of these fibers.

Shah [20] pointed out the shortcomings in composite manufacturing processing using Injection Molding (IM) machines, where the average temperature used is 175-190℃. When the granules are heated and compressed using extruder screws, in the mixing stage, the maximum natural fibers are damaged due to friction (interfiber, matrix fibers, and extruder fibers).

The challenges experienced in the polymer manufacturing process generally lie like natural fibers. Where the incorporation of hydrophilic natural fibers in a polymer matrix results in a heterogeneous system, having lower properties due to the lack of adhesion between the fiber and the matrix. Therefore, it is important to increase the fiber-matrix adhesiveness for the development of natural fiber-reinforced composites. Problems in the development of natural fiber-reinforced composites can be grouped into three categories, namely thermal degradation, water absorption, and biodegradation.

4.5.3 How to overcome the challenges in processing Natural Fiber Reinforced composite using Recycled Polymer?

These natural fiber-reinforced composites are a potential replacement for glass fiber reinforced composites. Therefore, the fiber treatment method in the process of making Natural Fiber Reinforced Polymer is a method that can be used to produce natural fiber reinforced polymers that have good thermal, physical, and mechanical properties and are by the needs of later composites.

Yan et al [21] conducted a study to improve the mechanical properties of natural fiber composites by using an alkaline treatment of bamboo fibers. They used 5% NaOH for 30 minutes, and then made a flax epoxy composite. The result is a significant increase in tensile strength (21.9%) and flexural strength (16%) along with some increase in transverse strength. In the same way, Van de Weyenberg et al. [22] used the same alkaline treatment technique for the flax/epoxy composites, but they used 4% NaOH, treated time 45 seconds, and saw a significant improvement in mechanical properties.

Nadine [23] conducted a study on fiber treatment methods which were carried out with four different fiber treatment variations. Rosella fiber was immersed in NaOH solution with three different concentrations (3%, 6%, 9%) for 2 hours at room temperature, then the next treatment used a silane linking agent, namely by soaking the fiber in silane solution for 24 hours. After chemical fiber treatment, the fibers were washed with clean water and dried in an oven at 100℃ for 48 hours to remove the moisture effect of the fibers. Alkalization methods and silane linking agents have been shown to provide improved chemical, physical, mechanical, and thermal properties.

Krishna [24] presented the results of his research on the treatment of processed kenaf fiber using a separate solution of glutamic acid and lysine base solution. The solution used in the kenaf fiber treatment is 10% of the total kenaf fiber to be used. Kenaf fibers were soaked in glutamate solution that had been prepared and left at room temperature for 24 hours and soaked again in lysine base solution for 24 hours, after soaking the fibers were washed three times using H_2O solution and dried for 2 days at room temperature used for further polymer manufacturing process. The results of the kenaf fiber treatment showed that the treatment by soaking the lysine base solution was more effective than the soaking with the glutamic acid solution.

Arsyad [25] researched the processing of natural fibers using chemical treatments on the surface morphology of coconut coir. The treatment of coco fiber is divided into 3 stages. The first treatment was soaking coconut husks in NaOH solution for 3 hours at a concentration of 5%, 10%, 15%, and 20%, respectively. The second treatment was coconut husk soaked in $KMnO_4$ solution with concentrations of 0.25%, 0.5%, 0.75%, and 1% for 3 hours. The third is soaking coconut husks in H_2O_2 solution with a concentration of 5%, 10%, 15%, and 20% for 3 hours. The results of the treatment of the surface fiber morphology of coconut coir become coarser.

Srinivasa [26] describes the cultivation of hydrophobic properties in abaca fiber by a chemical treatment which is suitable for alkaline treatment where the first step of the fiber is cleaned by immersing the fiber in a 1: 2 ethanol and benzene mixture for 72 hours at 50℃, deionized water and then air drying. The dewaxed fibers were immersed in a NaOH solution with different concentrations, where the total volume of the solution was 15 times the weight of the fiber. The fibers were stored in an alkaline solution for 72 hours at 30℃; clean fibers with tap water and then neutralized with a 2% acetic acid solution. Finally, the fiber is returned in tap water to remove residual adhering acids, so the pH of the fiber is approximately 7. Then, it is dried at room temperature for 48 hours to obtain an alkaline treated fiber. The results of the treatment showed that the fiber treated with alkaline was less than the raw fiber that was not treated.

Xie [27] conducted a study on the treatment of natural fibers by using silane as a treatment applied to natural fibers to make natural fibers bond with a matrix. The processing method uses silane hydrolysis to form reactive silanol which is adsorbed and condensed to the surface of the fiber at a certain pH and temperature. The treatment given to natural fibers with silane can improve the interface bonding to the matrix and improve the mechanical performance of the natural fiber-reinforced polymer.

Ojahan [28] conducted a study on extraction steps for the treatment of banana stalk fibers, where the steps are as follows, taking the banana stem midrib, then cutting it to a length of 40 cm, and then drying it in the sun for 3 days to reduce the moisture content, then heating the water until 20℃ with a ratio of 32.5 grams of NaOH to 1 liter of water, then add the kapok banana stalk and boil it for 1 hour in a boiling NaOH solution. After the extraction process is complete, the frond fibers are heated at 80℃ for 10 hours to dry the fibers, then brush them to avoid clumping fibers..

Selke et al. [29] have researched the use of multiple additives with possible effects on fiber/matrix adhesion or fiber dispersion into the matrix. They found that maleate-modified polypropylene looked

promising because its use at a 5% concentration in composites with 30% woodgrain resulted in increased tensile strength and elongation at break.

Ansell [30] has researched the surface of natural fibers by processing hemp, sisal, and kapok fibers in various concentrations of NaOH, and it was found that the 6% NaOH concentration was the optimal concentration in terms of cleaning the surface of the fiber bundle, but still maintained a high crystallinity index high.

5. Future trends and Conclusion

Over the past decade, much effort has been made to use natural fiber reinforced polymer composites due to their unique properties, biodegradability, environmental friendliness, essential accessibility, flexibility, easy processing, and impressive physico mechanical properties. The use of recycled polymer base materials and natural fibers is also an effort to reduce plastic waste and utilize natural fibers which are in abundance.

Composite material manufacturing methods that affect mechanical properties are closely related to the type of matrix, size, length and type, and direction of fiber reinforcement. The challenge in the process of making polymer composites reinforced by natural fibers is natural fibers that have hydrophilic properties and polymers that have hydrophobic properties that cause poor interface adhesion. The suitable manufacturing method for polymer composites can improve the adhesion of natural fibers and with modification of natural fiber, treatment can improve the bonding of the natural fiber interface with the matrix.

The treatment that can be used to remove the properties of natural fibers is to use chemical treatments because with this treatment the natural properties of the fibers will be removed/reduced to increase the adhesion of the fibers and the matrix interface. The manufacturing process using the Extrusion-Injection process is a process that can be used in the manufacture of recycled polymer composites reinforced by natural fibers because in this process there is a good mixing process between natural fibers and the matrix.

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