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STRUCTURE OF CELLULOSE AND ITS USE; A REVIEW

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ABSTRACT

Cellulose is one of the natural resources in abundance and is easily obtained from plants and animals. One of the uses of cellulose in the industry is by utilizing the fiber content in plants to be used as composite materials, paper materials, and bioethanol. To be able to utilize the cellulose contained in plants, researchers must be able to know and analyze the characteristics of the basic materials to be used. With a qualitative approach, it is hoped that data related to cellulose can be obtained so that it can be the basis for further research. From the results of a literature study, it can be seen that cellulose has a unique structure, so it has almost the same mechanical ability as Kevlar and aramid fibers, so it can be used as body armor. This paper is structured to review cellulose and its uses in life.

KEYWORDS: cellulose, composites, natural fibers, bioethanol, body armor

INTRODUCTION

Cellulose is one of the components that make up living cells, especially plant cell walls. This shows that cellulose is a compound contained in every organism. Cellulose is composed of bonds between glucose and hydrogen, insoluble in various solvents, and resistant to chemicals, due to these bonds, cellulose can only be broken down with strong acids and also with the help of the cellulase enzyme [1].

Based on the type of cellulose is divided into 4 components, namely: wood cellulose, non-wood cellulose, cellulose derived from marine fauna, and derived from bacterial activity [2]. Based on this, it shows that the cellulose contained in every living thing has different levels and purity, depending on where it comes from. The right knowledge and the right method will be the basis for the proper and good use of cellulose.

The use of cellulose is divided into 2, namely micro cellulose and nano cellulose, this is based on the size of the cellulose strands produced. Cellulose contained in plant cell walls needs to be isolated to obtain pure cellulose in the form of molecular aggregate chains or combined strands of polysaccharide chains with a width of 3-10 nm [3]. Based on this, the size of the pure cellulose produced is influenced

by several factors, including the source of the raw material, the isolation method, and the characterization process.

Several previous studies conducted research related to the use of cellulose in everyday life, such as the use of cellulose for nanocomposite applications, based on the size produced, in addition, several studies were also carried out on the mechanical properties of the cellulose produced. Based on this, this paper will review the characteristics and utilization of cellulose.

MATERIALS AND METHODS

In this study, researchers used a qualitative method approach. Qualitative research is a type of research that focuses on understanding meaning or action, so that research will lead to the stages of observing and interpreting. Meanwhile, according to Sugiyono, qualitative research views the object as something dynamic and is the result of the construction of thought, so it is holistic because the object and the result of thought are interconnected entities.

Data collection in this study was carried out using a literature study, where researchers collected data from various sources, such as research journals, books, magazines, and others. After the data is collected, a comparative analysis is carried out between one source and another. So that after the analysis, conclusions can be drawn.

RESULTS AND DISCUSSION

Cellulose Source

The most abundant source of cellulose is found in woody plants, this is because cellulose is a component of cell walls. Based on the leaf structure, woody plants are divided into two major groups, namely: broadleaf wood (hardwood) and needle leaf wood (softwood). In needle wood, the structure is uniform, while in broadleaf wood the structure is more complex. Based on this, it will determine the physical properties of the wood, so it can be concluded that the anatomical structure and physical properties of the wood are interrelated [4].

In a woody tree, it is composed of bio composite, with a matrix composed of lignin and hemicellulose, while the matrix is crystalline cellulose. Based on this structure, lignocellulosic bio composite material can support the existence of trees to become a strong and resilient material [3].

Sources of cellulose in addition to being obtained from woody trees, also obtained from agricultural waste other than wood, such as cotton, kapok, empty fruit bunches of oil palm, grasses, rice straw, sugar cane, and others. There are several advantages of non-wood cellulose sources compared to wood cellulose, including lower lignin content, environmentally friendly, renewable, and lower energy used for insulation. However, apart from having several advantages, agricultural products and wastes are not competitive to be used as raw materials for pulp and cellulose due to 2 things, namely: high

production costs for harvesting and high silica content in some raw materials such as rice husk, wheat straw, and bamboo bark [5].

Cellulose Structure

The trunk of a tree has a certain structure, this is because the tissue that makes up plants is not only cellulose but there are other issues such as Xylem, Phloem, Vascular Cambium, etc. The following is the structure of the tissue that makes up the plant cell wall.

Based on the picture, we can see that if a cell wall in a plant has a unique structure, it includes a primary layer, a secondary layer consisting of 3 layers (S1, S2, S3), and in the middle, there is a lumen. In the secondary layer, especially S2, cellulose is more dominant than lignin and hemicellulose [6]. In the second layer, there is a connecting network between S1, S2, S3, called microfibrils, wherein these microfibrils there are crystalline regions that have a regular arrangement of elements and are amorphous with an irregular structure arrangement. As seen in the image 1. These crystalline and amorphous phases can be seen when using atomic force microscopy so that the crystalline phase is very regular and the distance between the molecules is so tight that water molecules cannot enter.

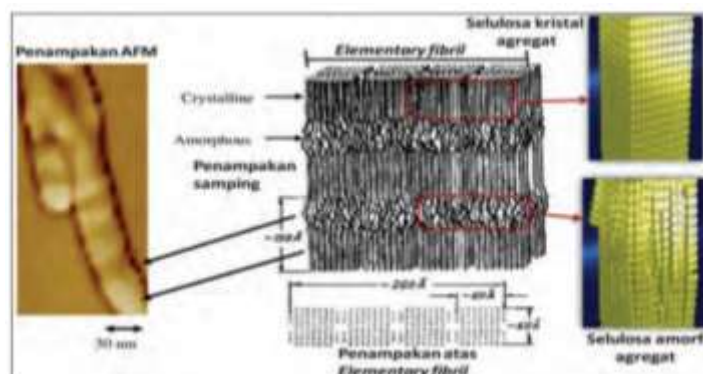


Figure 1 crystalline and an amorphous phase shown by the appearance of atomic force microscopy

Source: Battacharya et al, 2008

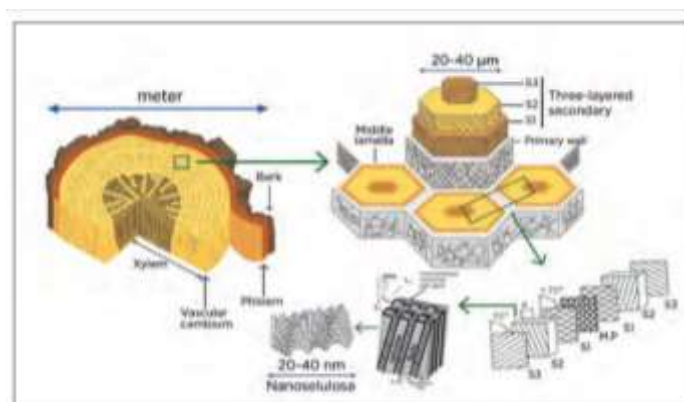


Figure 2. Plant cell wall structure
Source: Fratzl and Weinkamer, 2017

Cellulose is a strong linear chain bond, which is formed from intermolecular and intramolecular hydrogen bonds [7]. In addition to the presence of bonds between glucose and hydrogen, there is also an OH group that will determine the physical and chemical properties of cellulose [8]. Hydrogen bonds called intermolecular bonds are bonds that occur between hydroxyl groups and hydrogen groups located on adjacent cellulose molecules, while intramolecular bonds occur between hydroxyl groups and hydrogen groups on adjacent cellulose molecules, as shown below. As a result of these two bonds, it will produce energy, so by knowing the energy bonds formed, it can be determined the minimum energy needed to convert cellulose from macro to nano size [9].

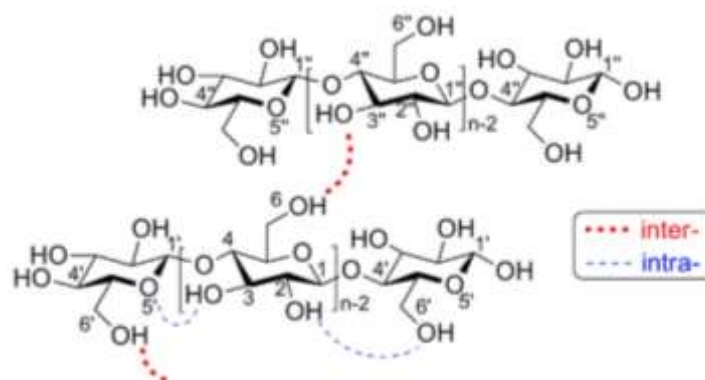


Figure 3. molecular and intermolecular hydrogen bonding schemes in cellulose
Source: Liu et al, 2015

Based on the picture above, it can be seen that each strand of cellulose has a different length and short length, this is called the degree of polymerization. The short length of the cellulose chain is also influenced by the ratio between the molecular weight of cellulose and the weight of 1 unit of a glucose molecule. The degree of polymerization in each plant part is different, where the highest DP is found in cells located close to the cambium and decreases towards the nucleus [10]. To be able to determine

the DP, two tests were carried out, viscometry and gel permeation chromatography. In the viscometric method, the steps carried out are isolation and dissolution of cellulose then measurement of viscosity, then the viscosity results are analyzed to show the value of the degree of polymer. Knowing the degree of polymerization will help in the manufacture of rayon, paper, cellulose acetate, nitrocellulose, and absorbent products.

Cellulose Insulation

Cellulose isolation is a process used to extract cellulose content by extracting plants. In general, there are 3 methods used to isolate cellulose, namely mechanical processes, biological and enzymatic processes, and chemical processes.

Mechanical Process

This method can be done in 3 ways to isolate cellulose, namely: High-Pressure Homogenizer, milling, and ultrasonication. The principle of using the HPH machine is to flow the mixed solution in a narrow gap, where the width of the gap is adjusted based on the viscosity of the solution and at this stage, a high pressure (100-2000 MPa) is applied [2]. At this stage there will be a fibrillation process against the cellulose fiber, this happens because of a change in pressure so that micro gas bubbles will form, frictional forces between fibers, collisions between fibers, and turbulence currents will appear around the gap. From this stage, it will produce a degree of fibrillation which indicates the change of cellulose to nanocellulose, which is based on the suspension cycle as it passes through the HPH. [11].

In addition to using HPH machines, the second mechanical process can be done by milling. In this method, a stone grinder will be used, if the HPH method uses pressure, then in this grinding method a cellulose suspension solution that has been flowed through the narrow gap between the two discs, where one disc will be able to rotate at a speed of 1500 rpm [12]. As a result of the contact between the suspension solution and the disc, it will cause a fibrillation process, where the degree of fibrillation produced can be adjusted by adjusting the width of the disc gap and the suspension cycle when passing through the stone grinder.

The third mechanical process that can be done is ultrasonication, in this method will utilize energy from ultrasound, by converting sound energy into physical energy and chemical energy. The working principle of this method is different from the two previous methods, because, in this method, the cellulose suspension liquid does not flow through the gap, but is placed in a container, then an ultrasonic rod is placed inside. From this ultrasonic rod will produce microbubbles, which will meet the surface of the fiber, as a result, will produce hot spots and cavitation processes, wherefrom this there will be a delamination process of the fiber layer, the energy generated is 10-100 kJmol⁻¹ [13]

Biological and enzymatic processes

In biological and enzymatic processes, the method used to isolate cellulose using enzymes or chemicals, the fiber fibrillation process will require total energy of 2MWh ton⁻¹ which is lower than

the energy requirement using mechanical treatment (70 MWh ton⁻¹) [2]. Enzymes that can be used in the isolation of cellulose are cellulase enzymes. Zhou & Ingram, 2000 explained that this cellulase enzyme is divided into 3 types of enzymes, namely: Endoglucanase, where this class of enzymes can degrade the amorphous phase present in cellulose, Exoglucanase, which in this class of enzymes can progressively degrade the crystalline and amorphous phases so that cellulose converted into disaccharides and B-glucosidase, this enzyme can hydrolyze disaccharides and tetra saccharides into glucose.

Chemical Process

In the chemical process, there will be a change in the surface properties of cellulose to be anionic or cationic, because the formation of this group will cause a repulsive force between charges, as a result, the fiber will experience repulsion more easily. delamination process [14]. This chemical process will reduce energy and increase the yield of nanocellulose, there are several methods for chemical processes, including:

Acid Hydrolysis

Acid hydrolysis is a cellulose degradation process under acidic conditions and high temperatures, as a result, the amorphous phase will be degraded and only the crystalline phase will remain. Using different types of acids will produce different CNCs. The use of concentrated H₂SO₄ will produce a crystalline phase with a negatively charged sulfate group, this can occur due to a chemical reaction between the two resulting in the substitution of OH groups, but the use of HCl will produce a CNC without charge. The presence of sulfate groups causes CNC to disperse when in an aqueous solution, but the heat resistance of cellulose will decrease [15].

Carboxymethylation

This method aims to give a negative charge to cellulose, the next step of this carboxymethylation process is: homogenization, ultrasonication, and centrifugation, this aims to separate the insoluble fiber so that it will produce CNF with a diameter of 5-15 nm and a length of up to 1 micron. CNF produced through the carboxymethylation process will have smaller and uniform dimensions compared to CNF produced from the enzymatic process [16].

Oxidation Tempo

Using this method will produce a negatively charged CNF, with a zeta potential of TEMPO-CNF in the water of -75 mV. This reaction will occur when the hydroxyl group at C6 is modified so that the dimensions and crystallinity of cellulose do not change, but depolymerization can still occur if the oxidant concentration is excessive so that the oxidation reaction takes longer. The oxidation system used requires several parameters, including TEMPO/NaOCl/NaBr at a pH of 10.5, at room temperature, during the reaction the pH must remain constant. To maintain the pH value, NaOH was added, so that a hydroxyl group content of 1.7 mmol g⁻¹ was achieved and a small aldehyde group

appeared indicating an intermediate phase. By performing the ultrasonication step, 3-5 nm in diameter of cellulose will be produced. [17]

Periodic Chlorite Oxidation

In the periodate-chlorite oxidation process which aims to change the hydroxyl group in secondary alcohol (OHC3C2), this change process includes the addition of sodium periodate which aims to convert it to an aldehyde group, and the addition of sodium chlorite which aims to convert it into a carboxyl group. To be able to produce CNF without mechanical treatment, the amount of carboxyl must be maximized, this is because in 1 hydro glucose unit there are 2 carboxyl groups, where this will cause a repulsive force on the cellulose chain. [18]

Cellulose Characterization

As a natural material, of course, cellulose has several characteristics, which are generally divided into 3 properties, namely the physical properties of cellulose, the mechanical properties of cellulose, and the chemical properties of cellulose. By studying these three properties, cellulose will be able to be utilized optimally, for example, cellulose wants to be used as body armor, of course, there must be certain conditions such as having a high hardness value and low density, this can be seen from the mechanical properties of the cellulose produced. The following is an explanation of each of these properties.

Physical properties of cellulose

In general, cellulose has the following physical characteristics: white, degradable, non-toxic, and has high tensile and compressive strength [19]. There are several physical properties of cellulose such as morphology of cellulose, thermal and electrical properties.

Cellulose morphology

Cellulose morphology is the appearance of the shape and condition of the specimen's containing cellulose, to be able to make observations and analyzes it is done with the help of a microscope, both SEM and TEM, the following is an example of the display of some test specimens.

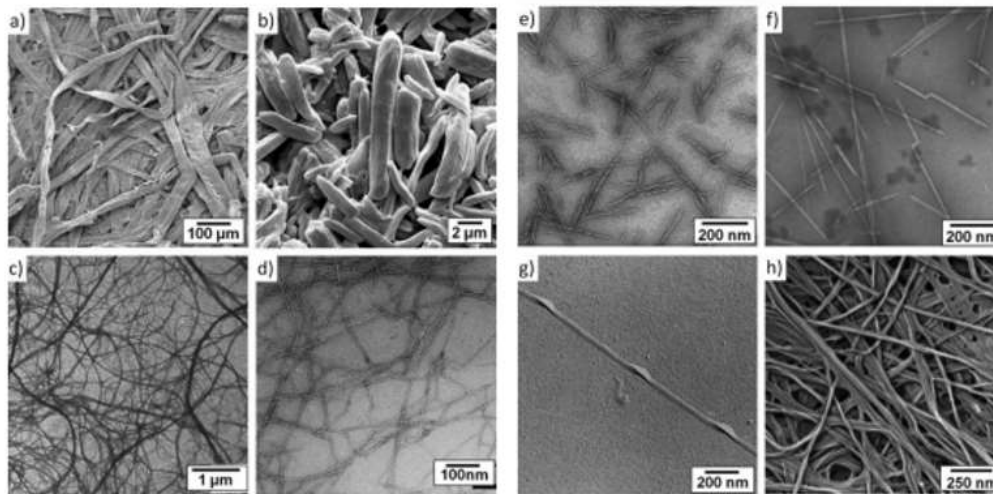


Figure 4 scanning electron microscopy observations
Source: Moon et al, 2011

The picture above shows 8 test specimens that received different treatments. In specimen a is the morphology of wood pulp fibers with a scale of 100 microns, specimen b is the result of SEM from microcrystalline cellulose (MCC) cellulose specimens, where the fabrication process is through an acid hydrolysis process, but the results of the SEM show the occurrence of agglomeration. during the drying stage due to the large surface area, the particles are easy to agglomerate so that the size is close to nano. Specimen c is cellulose in the form of CNF, where the fabrication process is a mechanical process using a homogenizer. Specimen d is cellulose in the form of CNF produced from the TEMPO oxidation process with nano dimensions and uniform length. Specimen e is cellulose in the form of CNC with a needle-like shape and looks cylindrical where the amorphous phase has disappeared so that only the crystalline phase remains. Specimen f is cellulose in the form of CNC taken from the sea animal tunicin, looks smaller in diameter than CNC from wood, and has a tensile strength value of 3-6 GPA [20]. This shows that the CNC of tunicin animals can be used as a candidate for composite reinforcement materials. Specimen G is cellulose derived from CNF produced from algae through acid hydrolysis. Specimen H is cellulose derived from bacteria and can be seen to have the form of interlocking webs.

Thermal and Electrical Properties

Material can conduct heat, but this depends on the chemical content of the material, this can occur because of the transmission of molecular vibrations from one atom to another [6] Thermal testing on test specimens generally uses thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) [21]. TGA is a heat test that aims to analyze the heat stability of a component by measuring changes in sample weight with an increase in temperature, while DSC This study aims to analyze the amount of energy absorbed and released when the test specimen is heated at a certain temperature. Cellulose itself is a material that has good heat stability, this is because cellulose has a crystalline phase, where the presence of this phase can improve the thermal stability of wood [22]

The MCC degradation process occurs through several stages, namely: the weight of cellulose will be lost at a temperature of 600C and 140C, followed by dehydration, decarboxylation, and depolymerization of glucose units which will eventually produce charcoal residue at a temperature of 2500-4500C [23]. The MCC degradation process requires high energy, due to its high molecular weight, this is due to the presence of a crystalline phase in MCC. Intrinsically, the crystalline phase has resistance to fire so that the resulting residue weight is high [21]

In dry conditions, cellulose can be useful as an electrical insulator, this is because when water is absorbed, electrical conductivity will occur [6]. Based on this, cellulose has low electrical conductivity and high resistivity, but the water content in cellulose can reduce resistivity. The interaction between materials and electric fields will result in the dielectric properties of a material. The dielectric constant is the ability of a material to absorb and store energy quantitatively. The dielectric constant in wood can increase when the moisture content increases and the constant will decrease when the plane frequency increases, this is also influenced by: temperature, density, and orientation of the structural plane [24]

Mechanical properties of cellulose

The phase contained in cellulose is a crystalline phase and amorphous phase, these two phases interact with each other, because of this interaction cellulose has stiffness or modulus of elasticity [25]. In addition, due to the crystalline structure of cellulose, it affects the strength of cellulose, where crystalline cellulose has a modulus of elasticity of 120-140 GPa [26]. In addition to the modulus of elasticity, the ductility of cellulose is also measured by testing the tensile strength, this is due to the presence of cross-links in the cellulose crystals [19]. Several things that affect the mechanical properties of cellulose include: cellulose content in the fiber, DP cellulose, and microfibril angle (MFA), when the cellulose content, DP cellulose is high, and MFA is low, the tensile strength and modulus will be high [27]

Several previous studies conducted tests related to the manufacture of test objects and testing the mechanical properties of cellulose. The process of measuring the mechanical strength of the test object is also influenced by the size of the crystals in the cellulose, which is to determine the size using the XRD method [28]. Other methods have been used such as Raman spectroscopy, in this method, the modulus of elasticity is measured by measuring the intensity of molecular deformation which will be seen from the vibration of the carbonyl bond [29]. In addition, there is another method, namely the atomic force microscopy method, in this method the test object is placed on a specimen holder which has a basin width of 227 nm, which will then be subjected to a pressure test [30].

Chemical properties of cellulose

Fiber which is one of the components in cellulose can experience swelling and dissolve, the process of dissolving the fiber is called the ballooning phenomenon [31]. This development is also influenced

by the quality of the solvent used, but it is also influenced by the reaction that occurs between cellulose and the developer. The development of cellulose is a change in the size of cellulose, which is an important step to obtain regenerated fibers such as viscose and lyocell, mercerization and functionalization of cellulose [32].

Mercerization according to the KBBI is a process of changing cotton to be heavier, stronger and viscous, and easy to color, to produce cotton it needs to be dissolved in a 25% NaOH solution. In addition, the result of the mercerization process using an alkaline solution is called basic cellulose, where this basic cellulose can also be produced from the manufacture of viscose fiber with cellulose ether derivatives [33]. The mercerization process can cause several things, such as a decrease in the size of the cellulose crystals, hydrogen bonds between chains, the degree of crystallinity of the cross-section of the cellulose fiber, the expansion of the outer surface of the fiber but causes an increase in the volume of the pores. [34]

Cellulose degradation aims to produce cellulose derivatives, this degradation process can be carried out by peeling reactions using alkaline solutions and oxidation-reduction reactions on cellulose. In addition to these two reactions, there are two other reactions aimed at oxidative degradation, namely the esterification reaction and the cellulose etherification reaction. From this reaction, derivative compounds such as sulfonate esters, cellulose acetate, and cellulose ether will be produced [33].

The process of hydrolysis of cellulose can occur completely, and imperfectly complete hydrolysis will produce glucose, while incomplete hydrolysis will produce oligosaccharides and disaccharides. So, it can be said that by carrying out a hydrolysis reaction, cellulose can be converted into monomers and the process is assisted by the cellulase enzyme which functions as a biocatalyst.

Cellulose Utilization

As previously explained, the results of the isolation of cellulose into monomers can be used in everyday life, such as pulp and paper, bioethanol, and composite reinforcement.

Pulp and paper

The manufacture of pulp and paper utilizes cellulose derived from lignocellulosic biomass. In the manufacture of pulp and paper, it is necessary to select the right raw materials and pulp processing processes to produce the desired characteristics of pulp and paper. What is done in pulp processing is to remove lignin and get as much cellulose as possible. To get the cellulose, carried out three processes, namely; mechanical, chemical, and semi-chemical processes. In the mechanical process, using the stone groundwood pulping and refiner mechanical pulping methods, while in the chemical process, chemicals are needed for dissolving lignin [35]

The chemical process in the manufacture of pulp and paper is divided into 3 stages, namely: alkaline, acid, and organosolv stages. There are 2 types of solutions used, namely: NaOH is used to dissolve

lignin, and Na₂S is used for the delignification process. While the semi-chemical process is carried out because this process will use two methods, namely the use of chemicals and mechanics. Both of these processes aim to soften and remove lignin, besides that in the semi-chemical process there is another variable, namely temperature. At this stage the processing process is divided into 2, namely the cold soda process and a hot soda, which is based on the temperature used. In addition, the pulping process also allows the use of fungi from the Basidiomycota class which functions for the lignin delignification process, this process is called bio pulping. [36]

Bioethanol

Bioethanol can be used as an alternative energy source to replace fuel. Bioethanol is the last product produced from the hydrolysis of cellulose to produce glucose, which is then fermented by yeast [37]. Based on this, the challenge in utilizing cellulose as bioethanol lies in the length of the conversion process from cellulose to produce bioethanol.

When converting biomass into bioethanol, there are two problems, namely the presence of lignin and the crystalline structure of cellulose. In addition, several other factors might be an obstacle in the process of converting cellulose into bioethanol, including particle size, an area that can be utilized, and cellulose depolymerization [38]. The crystal structure of cellulose in addition to being used is a problem because it inhibits the work of enzymes in breaking the beta-1,4 glycoside bonds between glucose monomers, but with this crystal structure, thermal stability is good.

Composite reinforcing material

Composites are one of the advanced materials that are still experiencing rapid development, one of which uses cellulose as a reinforcing material in composites, such as body armor. This is because the crystals that contain cellulose, have mechanical properties equivalent to Kevlar and aramid fibers, both of which are commonly used as body armor materials. Materials that can be used as composite reinforcement must have certain characteristics such as high hardness value, low-density value, tensile value, compressive value, and others. The characteristics of the composite are influenced by several things, such as the nature of the matrix, the reinforcing material, and the interaction of the two.

Cellulose can be used as a reinforcement in composites, where the matrix used is polymer, the interaction of the two shows a weak bond this is due to the different properties of the cellulose functional group (hydrophilic) with the polymer matrix (hydrophobic) [39]. To utilize cellulose as a reinforcing material, it is necessary to modify it with several methods such as acetylation, etherification, and amidation. This is done to bind the hydroxyl groups of cellulose so that it can utilize cellulose as a reinforcing material [40]

Several previous studies have utilized natural fibers as composite reinforcement materials used as body armor materials. Some of these tests have been successful and some require further testing. As research conducted by Topan Asmoro Aji et al, in 2018, succeeded in making test specimens with water

hyacinth reinforcing materials, were from the results of ballistic tests, test specimens with a thickness of 10 mm were able to withstand projectiles so that the specimens were not penetrated. Meanwhile, the research conducted by Mercy Iskandar Fajri et al in 2013, only carried out the tensile test, so this research can be continued with other tests, such as the compression test and ballistic test.

CONCLUSIONS

Based on several literature studies conducted, cellulose is one of the natural resources that is very possible to be used as a basic material for the manufacture of paper, bioethanol, and composites. This is in terms of the structure, characteristics, and isolation methods used, wherewith the use of different isolation methods the results obtained will be different. So, to be able to utilize cellulose in the industry, further research is needed.

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