Production Technology and Utilization of Nano Cellulose

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Abstract
Indonesia has a very abundant potential source of cellulose raw based materials. One of the opportunities for utilization of cellulose is by reducing it to have a nano meter size structure. Nano cellulose production technology can be done through two stages, namely by top down or reduce the size from macro to nano-sized meters. The second alternative is by reforming a monomer structure to the size of a polymer with a nanometer meter. Some studies show positive opportunities for both methods. The product from the optimization of the implementation of the technology can be accomplished by the method of forming the final quality of the product resulting from its implementation. Optimal product characterization is very important to do with the specifications of the final product implementation. Nano cellulose products have the opportunity to use a wide range of implementations, both for aspects of food, pharmacy, medicine, packaging, coatings and other implementation alternatives. In this paper, we try to explore the opportunities for optimal technological development so that it can be feasible to be implemented for other alternative products, related to the unique specifications of nano cellulose products. Based on the results of this study, it is hoped that it can increase the added value of raw material products and the final products produced by the implementation of nano cellulose.

Key Words: technology, production, utilization, nano cellulose

1. Introduction

Hydrocolloid is a polymer component derived from vegetables, animals, microbes or synthetic components that generally contain hydroxyl groups. This polymer component can dissolve in water, is able to form colloids, and can thicken or form a gel from a solution. Based on its characteristics, hydrocolloid is used as a gelling agent, thickener, emulsifier, adhesive, stabilizer, and film layer forming [1]. One of the potential hydrocolloids is cellulose derivatives.

Cellulose is a polymer structure consisting of a series of glucose monomer units connected by the β-1,4-Dglycosidic chain [2,3]. Cellulose hydrolysis techniques can be enzymatically using acid or enzymatically using cellulase enzymes. Cellulose composition in plants can reach 40-50% of the plant mass so that cellulose is the most abundant renewable biopolymer in nature [4]. Cellulose is one of the hydrocolloid which can be processed from raw materials of agricultural waste. Some potential wastes that can be utilized are solid or liquid waste. Some of the potential wastes include: sugarcane bagasse, corn husks, wood, paper and other waste materials. Whereas the potential of the wastewater is the waste of the national juice processing process, coconut water, tofu making waste and so on.

Cellulose modification technology into various alternative processed products began to be implemented widely developed in the community. The utilization opportunity is by modifying cellulose to nano-meter size. Modification technology can be done either top down by reducing or hydrolyzing macro components into nano-sized particle structures. While the second method is by forming polymer chains from glucose monomers in general. One potential for developing bottom up methods is through the technique of fermentation of liquid waste using cellulose polymer chain forming bacteria.
The use of nano cellulose is widely applied to food, pharmaceuticals, medicine, cosmetics, packaging, coating and so on. This is related to the specific functional properties that can be applied from nano cellulose products. In this paper try to examine alternative methods of production technology and opportunities for further utilization of nano cellulose products. Based on the results of this study, it is hoped that it can open up opportunities to increase added value based on technology and the opportunities for its implementation of nano cellulose products.

2. Process Technology

The technology for the production of nano hydrocolloid can be done top down or by reducing the size of particles from macro particles into nanostructures. Some sources of raw materials that can be used for the production of nano cellulose as listed in the table 1 below.

Table 1. The source of cellulose for nano cellulose production technology is top down

| No | Sourced of Main Based Raw Material | Process Method | Authors |
|----|----------------------------------|----------------|---------|
| 1  | Wheat bran                       | Chemical and enzymatic process | [5]     |
| 2  | Corn stalk                       | Chemical process           | [6]     |
| 3  | Sugarcane baggase                | Chemical hydrolysis         | [7]     |
| 4  | Raw material, non processed waste, processed waste | Physical and chemical process | [8]     |
| 5  | Cotton                           | Chemical Process            | [9]     |
| 6  | Pineapple peel waste             | Bioprocess                  | [10]    |

Furthermore in Howard et al. [11] mentioned the cellulose composition of hemicellulose and lignin, from various sources including 45% corn cobs, 32.1% rice straw, 30% wheat straw, 40-55% hardwood, 45-50% soft wood and as much grass 45%. Its potential as main based material could be followed by cellulose concentration.

The second category is by forming structures from monomers to forming nano-meter-sized polymers or often termed bottom up methods. Some sources of raw materials that can be used for the production of nano cellulose as listed in the table 2 below.

Table 2. The cellulose source for nano cellulose production technology is bottom up

| No | Sourced of Main Based Raw Material | Process Method | Authors |
|----|----------------------------------|----------------|---------|
| 1  | Pineapple Juice                  | Acetobacter xylinum fermentation | [11]    |
| 2  | Tofu Waste Water                 | Acetobacter xylinum fermentation | [12]    |
| 3  | Sugar Cane Waste                 | Acetobacter xylinum fermentation | [13]    |
| 4  | Tapioca Waste                    | Acetobacter xylinum fermentation | [14]    |
| 5  | Dragon Fruit Skin Waste          | Acetobacter xylinum fermentation | [15]    |
| 6  | Cocoa Pulp Waste                 | Acetobacter xylinum fermentation | [16]    |

In processing pineapple juice for nata production process, optimal conditions according to Majesty et al. [11] using 50 grams of sucrose and 15 days of fermentation time can produce 1.776% of...
the highest fiber content. Nuraini and Sari's research [15] showed that the addition of 12% sucrose produced a fiber content of 4.44%. The use of onggok as a fermentation medium produced optimally with the addition of 7.5% sucrose and 0.75% sprout extract with a fermentation time of 10 days obtained 41.4% wet yield and 99% nata fiber [14].

While Fifendy et al. [16] optimizing the manufacture of nata using cocoa pulp waste and obtaining optimal conditions, namely the addition of a toge of 225 grams produced a fiber content of 17.93%. Addition of nitrogen sources from both urea and toge can increase the fiber content of the resulting nata.

Some of the other bacteria that produce cellulose include Acetobacter sp, Gluconoacetobacter xylinus, Lactobacilus mali, and some Acetobacter xylinum with several strains that produce several yields with various carbon and nitrogen sources [17]. Corral et al [18] uses Gluconacetobacter xylinus NRRL B-42 to produce nano cellulose.

3. Cellulose Structure

Lignocellulose can be obtained from several agricultural, animal and forestry wastes. The waste can be reduced to smaller components in the form of nano cellulose. The top down nano cellulose matrix destruction method can be described as shown in the figure 1 below.

Lignocellulose is a carbohydrate complex polymer containing polysaccharides which are constructed from monomer units (xylose and glucose) and lignin. Lignocellulose is a reactive intermediate fraction that contains the composition of glucose, cellulose, hemicellulose and lignin for the production of other derivative products [19]. The structure of the cellulose polymer itself consists of crystalline and amorphous units which can be depolymerized by amorphous parts and obtained by crystalline units in nano meter size or often called nano crystalline cellulose through the process of hydrolysis using acids.
Figure 2. Depolimerisation of cellulose to nanocellulose [19]

Cellulose production technology is top down, from several sources of raw materials will produce several byproducts namely hemicellulose and lignin. The proportion of cellulose, hemicellulose and lignin content from several sources of raw materials is quite varied. Based on the results of Lee et al. [19] from several sources of raw materials obtained cellulose content: 45% corn cobs, 30% wheat straw, 33-40% straw barley, corn stover 39-42%, 25-30% Nut Shells, 41% empty fruit bunch, switch grass 45%, Hardwood stems 40-55%, softwood stems 45-50%, leaves 15-20%, waste paper from chemical pulps 60-70%, organic compound from waste water solid 8-15%.

The cellulose composition was obtained from the total concentration of cellulose, lignin and hemicellulose. As a result of the study by Lee et al. [19] which states that for corn cobs obtained cellulose concentration of 45%, hemicellulose of 35% and lignin of 15%.

4. Nano Cellulose Characteristics

The morphological structure of cellulose can be described as a fiber structure which is divided into three categories, namely fibrils, microfibrils and microfibril bonds. More specifically the size of cellulose lengths is hundreds of nano meters, diamaterally 1.5 to 3.5 nm, 10-30 nm and about 100 nm. Cellulose also shows different characteristics in solution conditions depending on the degree of substitution, solvent, concentration and distribution of the length of the chain [20].

Other characteristics of nano cellulose are nano structures that can be analyzed using SEM (Scanning Electron Microscope) and TEM (Transmission Electron Microscope). In the results of SEM profile analysis can only be known from the profile of the surface appearance, while with the Tem tool can be known the profile of the structure on the inside. Elazzouzi-Hafraoui et al.[21] conducted a TEM analysis of several sources of raw materials and obtained results as shown in the figure below.

Figure 3. The results of TEM analysis of nano cristaline cellulose are some fiber sources (a) cotton (b) avicel (c-e) tunicate [21]
One other characteristic that must be considered related to nano cellulose is the ability to form glassy phases and the dispersed phase. Furthermore Nondernstrom et al. [22] describes the phase of change in the diagram as shown below.

![Diagram of glassy and dispersed state](image)

Figure 4. Glassy and dispersed state [22]

Nano cellulose suspensions have rigid VASs (volume-spanning arrested state) characteristics at very low volume fractions. In transition conditions can be done by increasing the particle concentration or decreasing inter particle repulsion. In some researchers it can be done by influencing the particle ratio and surface charge density to classify VASs. Based on this classification can be used to predict the condition of nano cellulose under glass and gel conditions. The diagram can also be used to predict the transition phase and its reversibility capabilities. Gel conditions that are stiff or glass and their reversibility can be predicted from these conditions.

5. Uses of NanoCellulose

Some nano cellulose can be used for several applications according to the characteristics of the desired functional properties in the final product. Several opportunities for nano hydrocolloid applications as shown in the table 3 below.

| No | Application | Sources               |
|----|-------------|-----------------------|
| 1  | Nano Film   | [23,24]               |
| 2  | Packaging   | [25,26]               |
| 3  | Absorber Logam Berat | [27]          |
| 4  | Food Additive | [18]               |
| 5  | Kesehatan  | [28,29]               |
| 6  | Cosmetic    | [30]                 |

Several studies on the implementation of nano fiber cellulose (CNF), Cellulose NanoCrystal (CNC) and Bacterial nano celulosa (BNC) are widely applied for fat replacer [31], stabilizer [32], thickening agent [18], water binding properties are similar as probiotic components [33]. The formation of crum bread which is added with nano cellulose from the fermentation results in thick gluten filaments so that it can trap larger gases, and produce a softer and more porous texture compared to controls [18].

Applications of nano cellulose for the world of wide-ranging quite health include wounhealing, bone restructuring, drug delivery and other applications. NCC can also be used as a conditioner for cosmetic products. The accuracy of the use of surfactants as NCC emulsifier materials
for personal care or cosmetics products is important to be applied, so the product becomes stable during application and storage [30].

6. Conclusion

The source of cellulose is quite abundant in the world and in Indonesia in general. The potential of these raw material sources can be further developed through production technology both top down and bottom up. The top down method of nano cellulose production can be done using several sources of fiber from agricultural and forestry waste. While bottom up can be developed using liquid waste from processing agricultural products. Characterization of nano cellulose products can be done by using a PSA measuring instrument to ensure the size in the nano meter range. While other characterizations can also affect physical characteristics in glass and gel phase formation. The implementation of cellulose nano products is quite a lot, including for film products, packaging, absorber, food, health and cosmetics. The possibility of further development of nano cellulose products is still very wide open.

References

[1] Herawati H. 2018. Potensi Hidrokoloid Sebagai Bahan Tambahan Pada Produk Pangan dan Non Pangan Bermutu. Jurnal Litbang Pertanian Vol 37 No. 1: 17-25. DOI: 10.21082/jp3.v37n1.2018.p17-25.
[2] Kroon-Batenburg LMJ, Kroon J. 1997. The crystal and molecular structures of cellulose I and II. *Glycoconjugate J* 14:677-690.
[3] Saxena IM, Brown RM. 2005. Cellulose biosynthesis: current views and evolving concepts. *Annals of Bot* 96:9–21.
[4] Milala MA, Shugaba A, Gidado A, Ene AC and Wafar JA. 2005. Studies on the Use of Agricultural Wastes for Cellulase Enzyme Production by *Aspegillus niger*. *Res J of Agric and Bio Sci* 1(4):325-328.
[5] Nilsson C. 2017. Preparation and characterization of nanocellulose from wheat bran. Thesis Lund University.
[6] Boufi S and Chaker A. 2016. Easy production of cellulose nanofibrils from corn stalk by aconventional high speed blender. *Industrial Crops and Products*: 1-9. http://dx.doi.org/10.1016/j.indcrop.2016.05.030.
[7] Oliveira FB, Bras J, Pimenta MTB, Curvelo AAS, Belgacem MN. 2016. Production of cellulose nanocrystals from sugarcane bagasse fibersand pith. *Industrial Crops and Products*. 1-10. http://dx.doi.org/10.1016/j.indcrop.2016.04.064.
[8] Garcia A, Gandini A, Labidi J, Belgacem N, Bras J. 2016. Industrial and crop wastes: A new source for nanocellulose biorefinery. *Industrial Crops and Products* 93: 26–38.
[9] Qiao C, Chen G, Zhang J, Yao J. 2016. Structure and rheological properties of cellulose nanocrystals Suspension. *Food Hydrocolloids* 55: 19-25.
[10] Anwar B. 2015. Isolation of Cellulose Nanocrystals from Bacterial Cellulose Produced From Pineapple Peel Waste Juice as Culture Medium. *Procedia Chemistry* 16: 279-284.
[11] Majesty J, Argo BD, Nugroho WA. 2015. Pengaruh Penambahan Sukrosa dan Lama Fermentasi Terhadap Kadar Serat Nata Dari Sari Nanas (Nata de Pina). *Jurnal Keteknianan Pertanian Tropis and Biosistem* Vol. 3 No. 1: 80-85.
[12] Sutiyani S, Wignyato, Sukardi. 2002. Pemanfaatan Limbah Cair (Whey ) Industri Tahu Menjadi Nata De Soya Dan Kecap Berdasarkan Perbandingan Nilai Ekonomi Produksi. J. Tek. Pert. Vol 4(1): 70-83.
[13] Arifiani N, Sani TA, Utami AS. 2015. Peningkatan kualitas nata de cane dari limbah nira tebu metode *Budchips* dengan penambahan ekstrak tauge sebagai sumber nitrogen. *Bioteknologi* 12 (2): 29-33.
[14] Naufalin R. dan Wibowo C. 2004. Pemanfaatan Hasil Samping Tepung Tapioka Untuk Pembuatan Nata de Cassava: Kajian Penambahan Sukroza dan Ekstrak Kecambah. Jurnal Teknol. Dan Industri Pangan. Vol XV(2): 153-158.

[15] Nuraini H dan Sari ER. 2016. Identifikasi Mutu Nata Kulit Buah Naga (Hylocereus undatus) Dengan Variasi Konsentrasi Sukroza. AGRITEPA, Vol. II, No.2, Januari – Juni 2016: 2407 – 1315.

[16] Fifendy M, Putri DH, Maria SS. 2011. Pengaruh Penambahan Tauge Sebagai Sumber Nitrogen Terhadap Mutu Nata De Kakao. Jurnal Sainstek Vol. III No.2: 165-170.

[17] Ramana K.V. and Singh L. 2000. Effect of various carbon and nitrogen sources on ce llulose synthesis by Acetobacter xylinum. World J. Microbiol. Biotechnol. 16 (3). 245–248.

[18] Corral ML, Cerrutti P, Vaquez A, Califano A. 2016. Bacterial nanocellulose as a potential additive for wheat bread. Food Hydrocolloids. Food Hydrocolloids 67: 189-196. 10.1016/j.foodhyd.2016.11.037.

[19] Lee HV, Hamid BA, Zain SK.  2014. Conversion of Lignocellulosic Biomass to Nanocellulose: Structure and Chemical Process. The Scientific World Journal. 1-20.

[20] Klemm D, Heublein B, Fink HP, Bohn A. 2005. Cellulose: Fascinating biopolymer and sustainable raw material. Angewandte Chemie-International Edition. 44:3358-93.

[21] Elazzouzi-Hafraoui S, Nishiyama Y, Putaux J, Heux L, Dubreuil F, Rochas C. 2008. The shape and size distribution of crystalline nanoparticles prepared by acid hydrolysis of native cellulose. Biomacromolecules. 9:57-65.

[22] Nordenstrom M, Fall A, Nystrom G, Wagberg L. 2017. Formation of Colloidal Nanocellulose Glasses and Gels. Langmuir, 33 (38): 9772–9780.

[23] Rodriguez FJ, Torres A, Penaloza A, Sepulveda H, Gallotto MJ, Bruna AJ. 2013. Development of an antimicrobial material based on a nanocomposite cellulose acetate film for active food packaging. Food Additives & Contaminants: Vol. 31, No. 3, 342–353.

[24] Hossain ABMS, Uddin MM, Fawzi M, Veetil VN. 2018. Nano-cellulose biopolymer based nano-biofilm biomaterial using plantbiomass:Aninnovative plant biomaterial dataset. Data in Brief: 1245–1252.

[25] Li F, Mascheroni E, PIERGIOVANNI L. 2015. The Potential of NanoCellulose in the Packaging Field: A Review. Packag. Technol. Sci: 28: 475–508.

[26] Azeredo HMC, Rosa MF, Mattoso LH. 2016. Nanocellulose in bio-based food packaging applications. Industrial Crops and Products. 1-8. http://dx.doi.org/10.1016/j.indcrop.2016.03.013.

[27] Mahmoud ME, Abdou AEH, Sobhy ME, Fekry NA. 2017. Solid-solid crosslinking of carboxymethyl cellulose nanolayer on titanium oxide nanoparticles as a novel biocomposite for efficient removal of toxic heavy metals from water. International Journal of Biological Macromolecules. http://dx.doi.org/doi:10.1016/j.ijbiomac.2017.07.156.

[28] Howard, RL, Abotsi E, Jansen van Rensburg EL, Howard S. 2003. Lignocellulose biotechnology: issue of bioconversion and enzyme production. African J of Biotechnol 2 (12):602-612.

[29] Treessupharat W, Rojanapanthu P, Siangsanh C, Manuspiya H, Ummartyotin S. 2017. Synthesis and characterization of bacterial cellulose and gelatin-based hydrogel composites for drug-delivery systems. Biotechnology Reports 15 :84–91.

[30] Lima MMD, Borsali R. 2004. Rodlike cellulose microcrystals: Structure, properties, and applications. Macromolecular Rapid Communications. 25:771-87.

[31] Golchoobi L, Alimi M, Shokoohi S, Yousefi H. 2016. Interaction between nano fibrillated cellulose with guar gum and carboxy methyl cellulose in low-fat mayonnaise. J Text Stud 47: 403-412.

[32] Capron I, Rojas OJ, Bordes R. 2017. Behavior of nanocelluloses at interfaces. Current Opinion in Colloid & Interface Science 29: 83-95.
[33] Nahr FK, Mokarram RR, Hejazi MA, Ghanbarzadeh B, Khiyabani MS, et al. 2015. Optimization of the nanocellulose based cryoprotective medium to enhance the viability of freeze dried Lactobacillus plantarum using response surface methodology. LWT-Food Science and Technology 64(1): 326-332.