



Application of Bacterial Nano Cellulose as a Reinforcing Material in The Liner Test Paper

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Abstract

This research is about the application of Bacterial nano cellulose (BNC) as a reinforcing material in the making of liner test paper. BNC was obtained from the fermentation of banana peel extract using *Gluconacetobacter xylinum* bacteria obtained from the making starter of nata de coco. The reason for using banana peel waste is because it's available in large number all across Indonesia. BNC is mixed with secondary fiber as a raw material for making liner test paper. From the experimental handsheets results, strength properties and absorption properties were then tested. Variations in the composition of the use of BNC are 0% (blank), 5%, 10%, 15%, 20%, 25%, 30% of the handsheet dry weight. The BNC is also applied on surface sizing as a substitute for the surface sizing agent. The results of this study indicate that BNC can be used as an alternative raw material on wet end and on surface sizing, because both applications can increase the strength properties of liner test paper, and can reduce the use of chemical additive. The highest increase in strength properties of liner test paper was obtained at the composition of nano cellulose 30% and using surface sizing. Ring crush index is 14.02 Nm / g, concora index is 12.73 Nm / g, bursting index is 3.78 KPa.m² / g, ply bonding is 388.57 J / m². The absorption properties of paper increases but it has a low prosity. The highest cobb size results are obtained at 30% BNC composition, which is 45.30 g / m² without using surface sizing and 41.83 g / m² using surface sizing. The highest porosity value is obtained at 30% BNC composition, which is 158 s / 100cc using surface sizing. This research is expected to be a reference for further research in the field of BNC, as the alternative raw materials besides wood in paper making.

INTRODUCTION

Paper is a product derived from the use of cellulose as a raw material. Paper is widely used from education to packaging industry (Syafii, 2000). The world's production and consumption of paper and cardboard in 2010 reached 399,795,000 and 395,860,000 tonnes, respectively (FAO, 2010). The demand for corrugated cartons for manufactured product packaging tends to increase consistently in line with the production sector, for example the food industry, electrical equipments, commodity products, cosmetics and medicine (Office of Commercial Attache Embassy of the Republic of Indonesia, 2013). The packaging paper

consensus for the past 10 years has grown by an average of 2.3 percent (Kompas, 2018).

As the market develops, there is a high need for packaging paper. The two main ingredients of packaging paper are liner paper as coating and medium paper as its wave component. Liner paper is used to increase the tear resistance in the carton box so that the product inside is protected during the distribution process.

Kraft liner is a paper liner made from a composition of at least 75% virgin pulp (NUKP) and combined with recycle waste paper. The virgin pulp high price makes the paper producers switch to test liner paper. The raw material for liner test paper is 100% recycle waste paper, with certain additives

added to obtain better quality parameters (Media, 2017)

To meet the pulp industry capacity in 2000, 1.2 billion tree trunks were needed with the 166 million tons CO₂ unrelated impact (Aswandi, 2001). Increasing green industry awareness provides direction toward substituting raw materials for paper and cardboard from wood to non-wood (Setiawan, 1999). Paper can be made from all pulp containing cellulose. But until now wood cellulose still dominates as the main material used in the paper making process. Wood cellulose used for making paper is still mixed with other ingredients such as lignin and hemicellulose with a content of 16% and 25% of softwood or needle leaf wood (Sjostrom, 1995). Therefore, it is necessary to separate the cellulose from other ingredients. The separation process can be done in three ways, namely mechanical, chemical, and semi-chemical methods (Sjostrom, 1995).

According to Syamsu et al. (2012), those three ways have several weaknesses like high energy consumption and can cause high environmental pollution. Environmental pollution rises due to the use of hazardous chemicals for the delignification process (dissolution of lignin) and the pulp bleaching process (on certain paper) using bleaching chemicals that can result in environmental pollution (Indonesia, 1976). Other weaknesses are the wood low productivity, long logging time required, and other environmental-related issues. These weaknesses or problems demand an alternative source of cellulose which is expected to replace wood cellulose as the raw material for making paper. One source of alternative cellulose is microbial cellulose (Halib et al., 2012). Microbial cellulose or bacterial cellulose is produced from several types of microorganisms (bacteria) including *Acetobacter* species, such as *A. xylinum*, *A. aceti*, *A. cetianum*, and *A. Pasteuranum*.

According to Fitriani et al. (2016), bacterial nano cellulose (BNC) is an alternative source of environmentally friendly cellulose obtained from microbial aerobic fermentation of various species of *Acetobacter* (Erythrina, 2011). According to Syamsu et al. (2012), BNC has several advantages such as pure from chemicals (lignin, hemicellulose), high cellulose content, can be produced in a relatively short time, and the cellulose produced are already in sheet form (Suparto et al., 2012).

Cellulose fibers derived from wood must go through a purification process to remove

hemicellulose, lignin, and other extractive substances found in wood. Thus the BNC pulp making process is relatively simple and environmentally friendly. Microbial cellulose that can be harvested after one week of cultivation is more potential than wood cellulose which can only be harvested after 4-6 years (Sijabat et al., 2017).

According to Holmes (2004), bacterial cellulose has the same chemical structure as plant-derived cellulose and is a straight-chain polysaccharide composed of D-glucose molecules via β -1,4 bonds. The negative charge is caused by the same chemical structure constituting the two materials, namely cellulose, albeit of different sizes.

Nano technology is a technology that results from the utilization of molecular properties that are smaller than 100 nanometers (Abdullah, 2009). The BNC has a diameter of about 2-20 nm and a length of 100 - 40,000 nm. The resulting cellulose is stronger, thinner, and lighter compared to cellulose from plants (Stanley et al. 2006). The porosity is also very low with a diameter of 70-80 nm, the degree of crystallinity is quite high at 60-80% and the mechanical strength is large and the modulus of elasticity is high (Jonas & Farah, 1998). According to Mahmudah et al. (2014), making paper by using BNC as an additive material and retention material with the addition of 20% can increase folding resistance up to 500% and as retention material can reduce porosity. BNC that is applied in recycled wood fiber have the strength that can compete with virgin pulp. This research describes the effect of using BNC from banana peel waste as an alternative raw material and as a substitute for using additive materials in the liner test paper making process. Thus it is expected to produce paper of the same quality and productivity yet is more environmentally friendly. According to Sijabat et al. (2017) paper produced with BNC from coconut water performed good physical strength, low porosity value, optimal tensile index was 51.97 at the 30% nanocellulose composition and optimal tear index was 64.64 at the 15% nanocellulose composition.

MATERIALS AND METHODS

Materials and Equipments

The materials used in this study include secondary fiber form Old Corrugated Carton (OCC), BNC, cationic retention aid, cationic starch, and alkyl ketene dimers (AKD) from

Table 1. Raw Material Characteristics.

No.	Parameter	BNC	(OCC) Short Fiber	(OCC) Long Fiber
1.	Fiber Length	26-45 nm	1,006 mm	1,209 mm
2.	Freeness	0 ml CSF (Revolution 1000)	161 ml CSF (Unbeaten)	318 ml CSF (Revolution 1000)
3.	Initial pH	3.90	7.71	7.87
4.	pH washed with NaOH	7.49	-	-
5.	Particle Charge Detector	-157 mV	-185 mV	-173 mV
6.	Charge Demand	0.569 µeq/L	0.421 µeq/L	0.431 µeq/L

Solenis. While the equipments used include beater (PTI), disintegrator (PTI), beaker glass, analytic balance (PTI), vacuum (PTI), filter paper, hot plate (L&W/3-3), dispermat (PTI), handsheet maker (PTI), blotting paper, speed dryer (PTI), turbidity meter (PTI), Canadian Standard freeness (CSF) tester according to TAPPI, pH meter, ring crush tester (L&W/5-2), concora tester (Buchel BV/75-08-01-0002), cobb tester (Workshop/100CMQ), densometer (PTI), internal bonding test (Huygen/1314).

Methods

The 110 gsm hand sheet test liner was made by preparing the ingredients first, the beating nata de banana and secondary fiber process were done separately. Then the freeness, charge, pH, and consistency checks were conducted. The hand sheet blank was made by using 100% old corrugated containers with additional cationic starch, Poly Aluminum Chloride (PAC), AKD and cationic retention, and coating the surface sizing with the surface sizing agent.

Whereas the trial hand sheet was made by using variations in the composition of raw materials of old corrugated containers and BNC based on predetermined compositions of 0%, 5%, 10%, 15%, 20%, 25%, and 30% with the addition of cationic starch and AKD only, and coating the surface sizing using BNC as a substitute for the surface sizing agent.

Several wet end stock properties (drainage, turbidity, pH) were then tested along with the tests on several paper properties (ring crush index, concora index, bursting index, ply bonding, porosity, and cobb size). Then an analysis was conducted to compare the test results between blank sample and the trial sample.

RESULTS AND DISCUSSION

The results from the nata de banana and secondary fiber characteristics test are described in Table 1.

Based on table.1, the results from raw materials tests indicate that BNC has the same negative charge as the charge of wood fiber in general. But it has an acidic pH, so it must be washed first with a 1% of natrium hydroxide (NaOH) solution.

Freeness Test Results

Before being used as raw material for the handsheet, the raw material was beaten by using hollander beater, the OCC long fiber has 318 ml CSF freeness after 1000 rpm beating, the secondary fiber of OCC short fiber has 161 ml CSF freeness unbeaten, and BNC has a 0 ml freeness CSF after 1000 rpm revolution. The BNC must be able to be homogeneously mixed with raw materials or other additive materials before it can be added on the surface sizing. Therefore it must be beaten with a hollander beater at a 7000 rpm revolution. Then the freeness value is checked, which is at 0 ml CSF.

According to Sijabat et al. (2017), the absence of water coming out in the BNC freeness test is because the structure of the BNC itself is in the form of gel and very small in size, with a diameter of about 2-20 nm and length of 100 - 40,000 nm. The non-branched glucose chain makes a long fibrillar structure, because of the high number of free hydroxyl groups that produce extensive intra and intermolecular hydrogen bonds between adjacent chains (Brown, 1985). These fibres are very small and made of nano-sized fibers thus making the braid really strong and tight that even water will struggle to penetrate it. Therefore, in freeness testing, water cannot get out of the CSF tester mesh.

Drainage Speed Test Results

Figure 1 shows the slowing down of drainage, that is the water takes longer time to get out of the stock. This happens along with the addition of BNC composition into the stock.

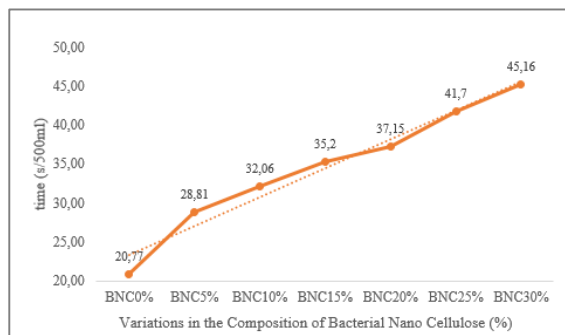


Figure 1. Drainage test result.

This is because BNC is hydrophilic and the fiber structure is in gel form and very small, about 2-20 nm in diameter and 100 - 40,000 nm in length. The non-branched glucose chains make long fibril structures. The high number of free hydroxyl groups produce extensive intra and intermolecular hydrogen bonds between adjacent chains (Brown, 1985). These very small and nano-sized fibers make the braids really strong and tight that even water will struggle to penetrate it. Therefore, the addition of BNC composition into stock must be limited. In this experiment, the maximum BNC composition used was 30% in stock. According to Sijabat et al. (2017), the maximum composition of nanocellulose is 30%, the addition of nanocellulose above 30% can reduce drainage speed and cause problems in the dewatering process.

Based on the experimental result, at the trial 6 drainage stock headbox is very slow at 45.16 s/500 ml, because the BNC composition used is at 30%.

Basis Weight Test Results

Figure 2 shows that the basis weight value is decreasing from the hand sheet blanko to the hand sheet trial 6. The paper basis weight value depends on the paper sheet weight. The paper sheet weight is influenced by the amount and type of pulp given. The decrease in weight base value is caused by the increasing BNC composition used, from the hand sheet blanko to the hand sheet trial 6.

In general, cellulose consists of α cellulose and β cellulose. Wood cellulose and bacterial cellulose consist of both cellulose, only with different compositions. In wood cellulose, the

cellulose α content is higher, which is around 70% and the remaining 30% is β cellulose. Whereas in bacterial cellulose, the β cellulose content is higher at 60%. The density of cellulose α is bigger than that of cellulose β , hence the density of bacterial cellulose is smaller than that of wood cellulose (Sugiyama et al., 1991). This bacterial cellulose has a diameter of about 2-20 nm and a length of 100 - 40,000 nm. The cellulose produced is stronger, thinner, and lighter than the cellulose that comes from plants (Stanley et al., 2006). The porosity is also very low with a diameter of 70-80nm, the degree of crystallinity is quite high, namely 60-80% and has large mechanical strength and high modulus of elasticity (Jonas & Farah, 1998). Therefore it can make the difference in fiber weight between bacterial cellulose and wood cellulose.

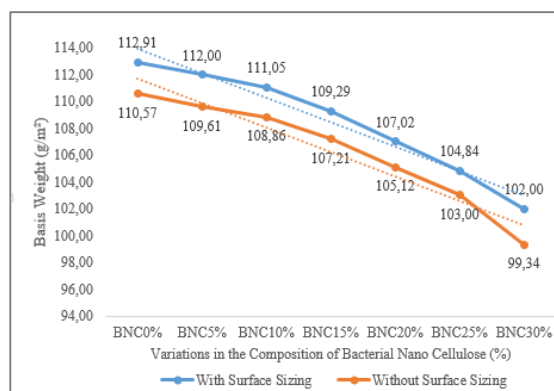


Figure 2. Basis weight test result.

Ring Crush and Concora Tests Result

Figures 3 and 4 show that the ring crush index and concora index are increasing, from the hand sheet blanko to the hand sheet trial 6, both in the experiment without surface sizing and in the experiment with surface sizing coating. The increase in the paper ring crush index and concora index values is caused by the increasing BNC composition used from the hand sheet blanko (0% BNC) to the hand sheet trial 6 (30% BNC).

According to Wiley (2014), Nanocellulose has a low density (1.6 g.cm⁻³), and the surface of the -OH group is more reactive than cellulose in general. This is because the high number of free hydroxyl groups produce extensive intra and intermolecular hydrogen bonds between adjacent chains (Brown, 1985). Therefore the nano-sized BNC has a vast surface area, with many highly reactive OH groups that can bind easily and establish very strong fiber bonds. The porosity is also very low at 70-80nm in diameter, the

crystallinity degree is quite high at 60-80% and the mechanical strength is high and the elasticity modulus is high (Jonas & Farah, 1998).

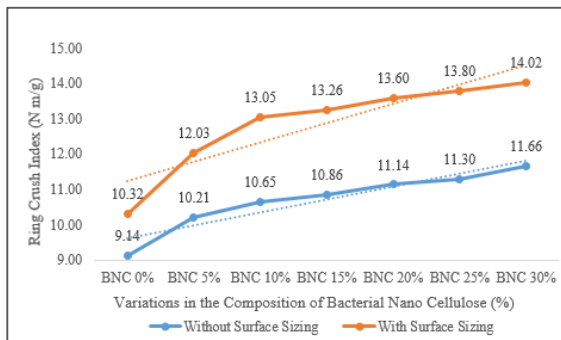


Figure 3. The ring crush index result.

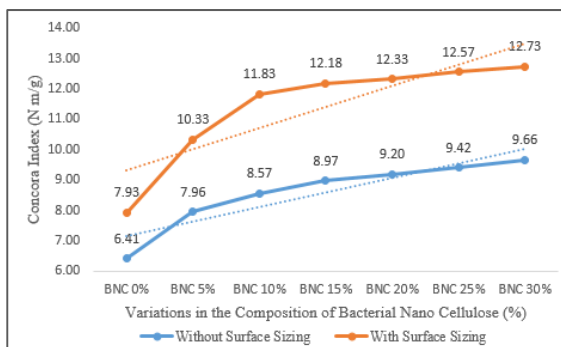


Figure 4. The concora test index.

Therefore, the ring crush index and concora trial index values obtained are higher, compared to ring crush index and concora index blanko. Thus the use of BNC in trial proved to increase the handsheet strength properties, compared to the use of additives (PAC and cationic retention) in blanko.

Surface sizing is the application of adhesive material (size) on the surface of the paper that has been formed. The main objectives of surface sizing are to obtain paper that is resistant to liquid penetration, to increase the paper surface resistance to chemical absorption, and to increase the physical characteristics of the paper such as its resistance from cracking, cracking, pulling, and folding. Thus the resulting ring crush index and concora index with surface sizing coating are better compared to those without surface sizing coating both in blanko using surface sizing agent as well in trial using BNC to replace the surface sizing agent.

The increasing ring crush index and concora index are also influenced by coat weight of the sizing. The heavier the coat weight will increase the ring crush index and concora index, and vice versa.

Bursting Test and Ply Bonding Results

Figures 5 and 6 show that the bursting index and ply bonding values are increasing, from the hand sheet blanko to the hand sheet trial 6, both in the experiment without the surface sizing coating, and in the experiment with surface sizing coating.

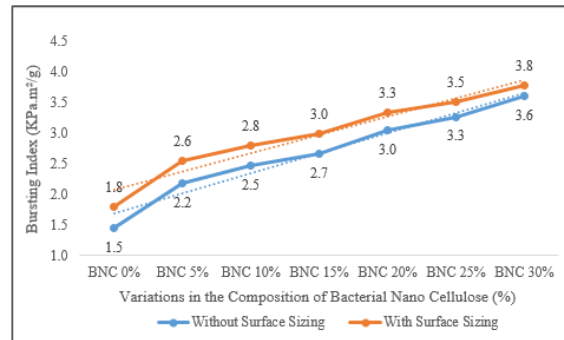


Figure 5. Bursting index test result.

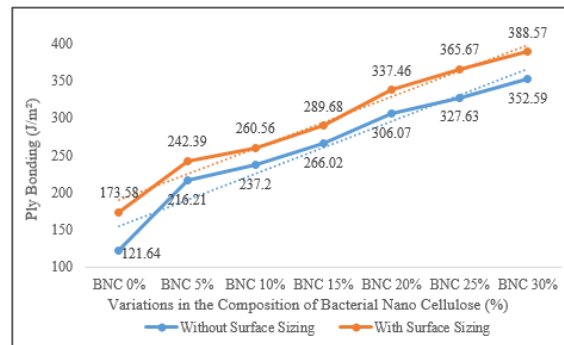


Figure 6. Ply bonding test result.

The increase in the paper bursting index and ply index values is caused by the increasing BNC composition used from the hand sheet blanko (0% BNC) to the hand sheet trial 6 (30% BNC).

This is because the high number of free hydroxyl groups produce extensive intra and intermolecular hydrogen bonds between adjacent chains (Brown, 1985). Microbial cellulose has several advantages including high degree of crystallinity, has a density between 300 and 900 kg m⁻³ and is elastic (Krystynowicz & Bielecki, 2001). The use of BNC can also fill the space between fibers. This advantage causes the bursting index and ply bonding values to increase.

The highest bursting index and ply bonding value are in the trial handsheet 6, with the BNC composition at 30%. This shows that using BNC can increase internal strength between fiber layers. Thus using BNC in the trial proved to increase the strength properties handsheet, compared to using

additives (PAC and cationic retention) in the blanko.

The values of bursting index and ply bonding with surface sizing coating are better, compared to those without surface sizing coating, both in the blanko which uses surface sizing agent and in the trial which uses BNC to replace the surface sizing agent.

The increase in bursting index and ply bonding is also affected by coat weight of the sizing. The heavier the coat weight will increase the ring crush index and concora index, and vice versa.

Cobb Size Test Results

Figure 7 shows that the cobb size value is increasing, from the hand sheet blanko to the hand sheet trial 6, both in the experiment without the surface sizing coating, and in the experiment with surface sizing coating. The increase in the paper cobb size value is caused by the increasing BNC composition used from the hand sheet blanko (0% BNC) to the hand sheet trial 6 (30% BNC).

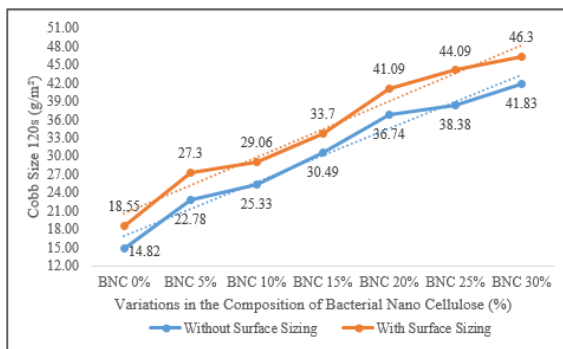


Figure 7. Cobb size test result.

According to Windarti & Siahaan (2008), one of the BNC unique characteristics is that its cellulose membrane is very strong and can bind water to more than 100 times of its own weight, thus forming a hydrogel. The ability of cellulose to bind large amounts of water is because the number of -OH groups possessed by cellulose, causes cellulose to be hydrophilic. The cellulose ability to bind large amounts of water is due to the numerous -OH groups possessed by cellulose, thus making it to be hydrophilic.

As the BNC composition increases in the handsheet trial, it will increase the cobb size value. Because the higher the BNC, the more water can be binded. Whereas using internal sizing (AKD) as penetration barrier in the trial is less effective because the AKD dosing point is carried out on

stock with low consistency. Thus BNC does not bind with AKD, because it is already bound to water.

The cobb size result with surface sizing coating decreases compared to that of without surface sizing, both in the blanko, using surface sizing agent and in the trial using BNC to replace surface sizing agent, because the function of adhesive material (sizing) is to make the paper more water resistant, to increase paper strength and its folding power (Fitriani et al., 2016). In the handsheet trial with surface sizing coating, using BNC to replace the surface sizing agent can keep the liquid from penetrating because the starch added also functions as sizing agent that can block access to free OH groups in BNC (Syamsu et al., 2012).

Also the BNC composition used on surface sizing is less than in raw materials, namely 5.56% of surface sizing solution.

Figure 8 shows that the porosity value is increasing from the hand sheet blanko to the hand sheet trial 6, both in the experiment without the surface sizing coating, and in the experiment with surface sizing coating. The increase in the paper porosity value is caused by the increasing BNC composition used from the hand sheet blanko (0% BNC) to the hand sheet trial 6 (30% BNC).

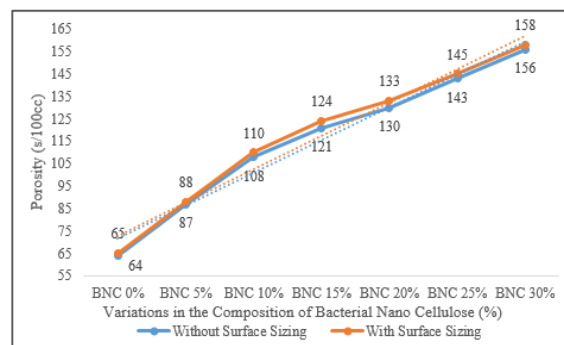


Figure 8. Porosity test result.

According to Mahmudah et al. (2014), paper making can use BNC as an additive and retention material because it's a large molecule with 4000-6000 in polymerization degree. The non-branched glucose chain makes a long fibrillar structure, because of the high number of free hydroxyl groups that produce extensive intra and intermolecular hydrogen bonds between adjacent chains (Brown, 1985). These fibres are very small and made of nano-sized fibers thus making the braid really strong and tight that even water will struggle to penetrate it. According to Sijabat et al. (2017), the addition of nanocellulose can reduce porosity

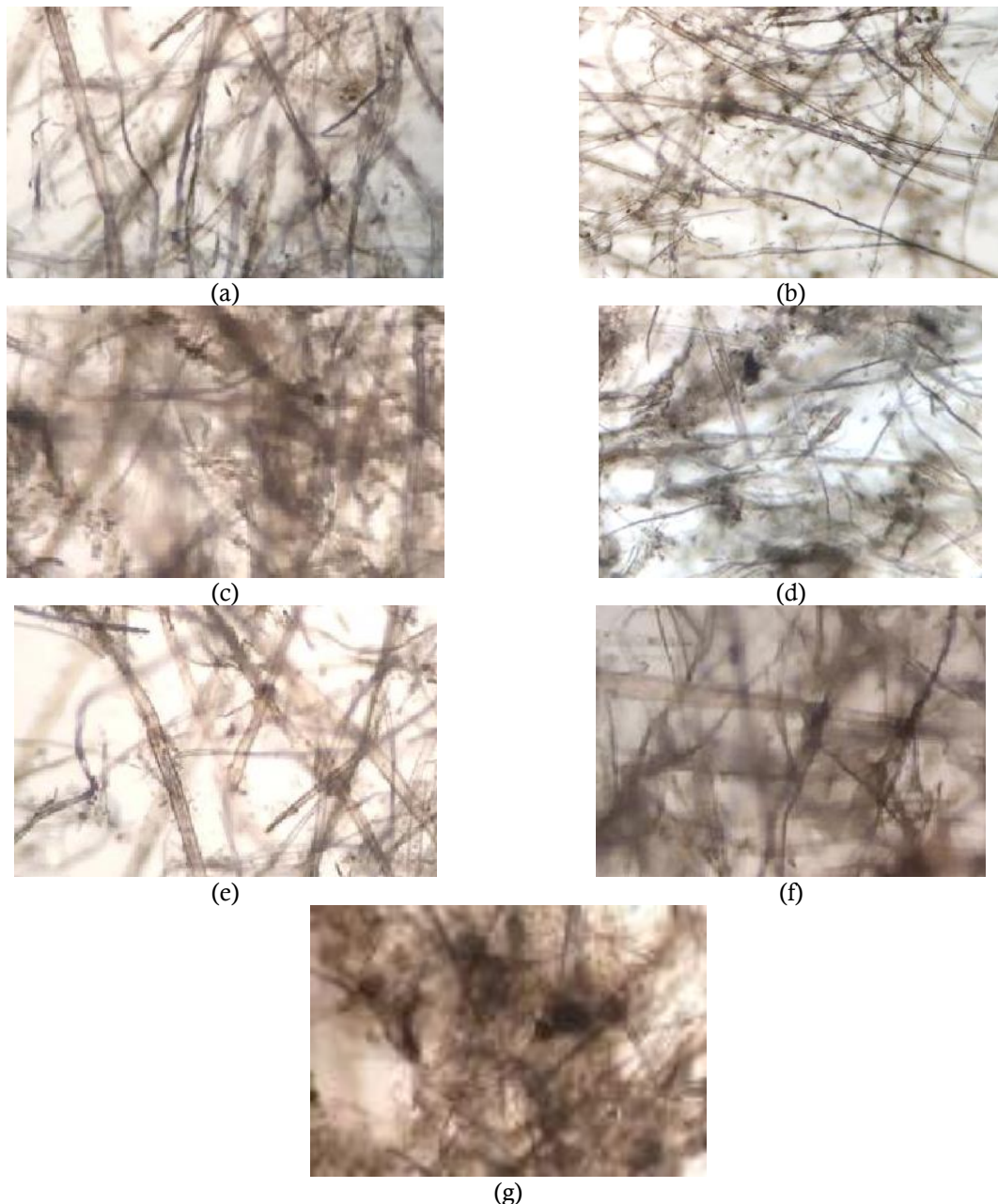


Figure 9. Microscope Test Results on Stock BNC (a) 5% (b) 10% (c) 15% (d) 20% (e) 25% (f) 30% and (g) 35% at 300X Magnification

because nanocellulose, whose size is in nanoscale, will fill the pore gap between wood fibers, so that the pore gap will be getting smaller or even closed-up until there is no more gap. In the experiment where the BNC composition used is increasing, there is an increase in the porosity value from the hand sheet blanko (0% BNC) to the hand sheet trial 6 (30% BNC).

The porosity result with surface sizing coating increases compared to that of without surface sizing, both in the blanko, using surface sizing agent and in the trial using BNC to replace surface sizing agent, because one of the surface

sizing functions is to smooth the paper surface that the air will struggle to penetrate through the paper sheet. Because according to Syamsu et al. (2012), tapioca functions as a binder between fibers and increases the number of bonds among fibers and reduces the number of pores.

Microscope Test Results

Microscope testing is conducted at 300X magnification on the stock before the hand sheet was made and the results can be seen in Figure 9.

Based on the microscope results on stock blanko (0% BNC) to stock trial 6 (BNC 30%) above,

there is a difference in the structure of each stock variation. The more visible morphology is fiber from pulp secondary fiber (wood fiber) pulp, whereas the BNC morphology is not visible in stock. This is because the BNC size is far smaller than the size of the wood fiber itself.

But as the BNC composition used is added, the stock looks denser and clots (flocculation). This is because the non-branched glucose chain makes a long fibrillar structure, because of the high number of free hydroxyl groups that produce extensive intra and intermolecular hydrogen bonds between adjacent chains (Brown, 1985).

CONCLUSION

The conclusions drawn from this study are BNC can be used as the environmentally friendly alternative source of cellulose in making liner test paper because it does not require a delignification process which is generally found in the process of making paper from wood cellulose. There is an increase in strength properties handsheet trial using bacterial nano cellulose without additional PAC and cationic retention compared to the strength properties handsheet blanko with additional PAC and cationic retention. Therefore BNC can be used as a raw material and can reduce the use of chemical additive. BNC can be applied to surface sizing solutions to further improve paper strength properties as the substitute for surface sizing agent. The quality of paper produced by mixing recycle waste paper and BNC as raw material for liner test paper as a whole increases the strength properties of the test liner trial paper. The use of BNC has an impact on decreasing basis weight handsheet, but paper strength properties such as ring crush index, concora index, bursting index, and ply bonding have increased along with the increase in BNC composition in, as well as in the use of BNC in surface sizing solution. The porosity and cobb size values increase with increasing BNC composition in the handsheet. Thus the paper absorption properties is considered not good, because the trial result underwent an increase from the blanko. The BNC composition of 30% (trial 6) achieved the highest value for paper strength properties, both with he composition of BNC 30% (trial 6), is the highest value for paper strength properties, both without surface sizing coating or with surface sizing coating.

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