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Fabrication of a lithium-ion battery separator from cellulose acetate of empty palm fruit bunches with the addition of PVDF

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ABSTRACT

The separator is a crucial element in lithium-ion batteries that is positioned between the anode and cathode. Its primary function is to prevent direct contact between the electrodes, hence avoiding electrical short circuits. Lithium ion battery separators are typically composed of polyvinylidene fluoride (PVDF) and polyacrylonitrile (PAN) polymers, which possess excellent ionic conductivity and mechanical qualities. Nevertheless, these polymers and materials possess numerous drawbacks, necessitating adjustments for their further development. The objective of this study is to examine the impact of incorporating novel polymers into the constituents of the separator. The alteration utilises a novel polymer called cellulose, specifically the cellulose derivative known as cellulose acetate. The cellulose acetate utilised is derived from the empty fruit bunches of oil palm trees. Cellulose acetate offers several benefits, including affordability, the ability to selectively adsorb substances, solubility in a wide range of solvents (particularly organic solvents), hydrophilicity, and its origin from renewable sources. The separator was fabricated using the reflux process, which involved mixing 5.6 g of PVDF and 0.7 g of $Al(OH)_3$ with varying amounts of CA (0.1 g, 0.2 g, and 0.3 g). The conducted tests include the thickness test, elongation test, and PSA test. According to the test results, the separator is viable and complies with the standards.

Keywords: Cellulose acetate; lithium; PVDF; separator

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INTRODUCTION

Batteries are parts that have a very big role in human needs. Batteries are a source of electrical energy that is highly relied upon to operate electronic equipment that is portable or can be carried anywhere [1]. Lithium-ion batteries are widely used in electronic equipment because of their advantages in the faster self-discharge process and higher energy power density. Another advantage of this lithium-ion battery is that it can last longer [2]. Lithium-ion batteries consist of 3 important components, namely cathode (positive electrode), anode (negative electrode), and separator [2]. Separators have an important role in the process of preventing short circuits at the cathode anode in battery cells. The characteristics of the separator are that it has a small electrical resistance, is an insulator and has high mechanical stability [3].

Lithium-ion battery separators are generally made from polyvinylidene fluoride (PVDF), and polyacrylonitrile (PAN) polymers which have high ion conductivity and good mechanical properties [4]. This polymer still has weaknesses and needs to be developed by making modifications to the separator. Modifications can be made by mixing other polymer materials or new materials to improve the performance weaknesses of the separator [3]. Referring to research conducted by Jiang C et al [5], namely making a separator by mixing PVDF, CA, and $Al(OH)_3$ which produces a separator with an absorption value of 403.9%, a porosity of 68.6% and an ion conductivity of reaching 2.85×10^{-3} S/cm [5]. Data resulting from Jiang *et al.*'s research shows that adding new materials to the separator composition can improve separator performance. The new material that can be used is cellulose.

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Cellulose is a natural polysaccharide polymer with a linear chain consisting of two anhydroglucose units that repeat up to 15,000 units. Cellulose is a very abundant natural resource on earth that can be renewed and is available not only from plants (wood, cotton, wheat, grass, etc.) but even from non-plants such as algae and bacteria [6]. Cellulose has the advantage of being biodegradable, the raw material is abundant and easy to find, having high thermal stability, and high electrochemical stability, and is easily recyclable and renewable [7]. The cellulose derivative product that is widely used in the industrial world is cellulose acetate. Membranes made from cellulose acetate polymer are hydrophilic due to the presence of groups in the polymer chain, cellulose acetate is environmentally friendly and biodegradable. Many cellulose acetate polymers are synthesized, but good cellulose acetate for membrane materials must contain a minimum of 39.5% acetyl [8]. Cellulose acetate is a material that is still not available in Indonesia. This makes cellulose fall into the category of materials that require expensive costs and take a long time to ship.

The advantages of using cellulose acetate are that it can be produced at a low cost, has selective adsorption, can dissolve in most solvents (especially organic solvents), is hydrophilic, and is made from renewable sources. Cellulose acetate is also widely used for the adsorption process or absorption of metal ions or industrial waste because of its good absorption properties [9]. The hydroxyl group in cellulose acetate has been replaced by an acetyl group which is a white solid, non-toxic, tasteless, and odorless. The commercial value of cellulose acetate is quite high because of its advantages. Based on the degree of substitution, cellulose acetate is divided into three, namely cellulose monoacetate, degree of substitution (DS) 0 – 2 with acetyl content < 36.5%. Cellulose monoacetate can be used in making plastics, paints, and lacquers. Cellulose in acetate, degree of substitution (DS) 2.0 – 2.8 with acetyl content 36.5% – 42.2%. Cellulose diacetate can be used in the manufacture of membranes, topographic films, and threads. Cellulose tri acetate, degree of substitution (DS) 2.8 – 3.9 with acetyl content 43.5% – 44.8%. Cellulose triacetate can be used in the manufacture of fabric and wrapping thread [10].

Cellulose acetate can be obtained by extracting it from natural materials, one of which is empty oil palm fruit bunches. Empty palm oil bunches (TKKS) are palm oil mill wastes that are abundant and have not been widely used. Processing 1 ton of palm oil can produce 22% – 23% EFB or 220 – 230 kg EFB [11]. The chemical content and composition contained in empty oil palm fruit bunches is 40% – 43% cellulose, 22% – 25% hemicellulose, and 19% – 21% lignin. The high cellulose content has great potential for making cellulose fibers based on transparent biopolymers [6]. TKKS decomposes more than 3 months naturally because its large size makes it difficult for TKKS to decompose due to its small surface. Reducing the size of EFB will help EFB to decompose more quickly [12].

PVDF made from fluoropolymer has strong piezoelectric and pyroelectric properties, this material is widely used because it has low strength, good response, flexibility, and lightness. PVDF has three molecular structures, namely α phase, β phase, and γ phase. The β phase is most widely used as sensors and actuators because it has the greatest piezoelectric effect. The β phase structure material requires certain fabrication techniques to obtain it by stretching (pulling) at a certain temperature and followed by polarization with high voltage DC electricity [13].

RESEARCH METHODS

The research method used is the reflux method. The tools used in this research include a synthetic distillator, grinding machine, desiccator, digital balance, stirring rod, oven, beaker, measuring cup, Erlenmeyer, magnetic stirrer, filter paper, pH paper, dropper pipette, and thermometer. The main ingredients used in this research were empty palm fruit bunches, NaOH powder, H_2SO_4 72%, NaOCl 1%, distilled water, glacial acetic acid, acetic anhydride, sodium acetate, $Al(OH)_3$, DMAc and PVDF.

Cellulose Extraction

The empty palm oil bunches are separated first, then the empty palm oil bunches are cleaned and dried in the sun until the empty palm fruit bunches are completely dry. Dry empty bunches are ground using a grinding machine until the sample is 60 mesh. A sample of 40 grams of finely ground empty palm oil bunches was put into a synthetic distillator.

2 g of NaOH powder was first dissolved in 100 ml of distilled water and made 2 times. The NaOH liquid is mixed into the distillator and refluxed for 8 hours at 90ºC. The mixture is filtered and tested for lignin by reacting the filtrate and H_2SO_4, if there are still lumps in the filtrate, the residue is dissolved again in NaOH solution and refluxed again until there is no lignin.

Cellulose that no longer contains lignin is refluxed again for one and a half hours with NaOCl solution and 2 g of NaOH are added with a ratio of 100:1 (v/w) at a temperature of 70ºC. Then the mixture was filtered using filter paper, the filtered residue was then soaked in 100 mL of NaOH solution for 30 minutes. The mixture was filtered again and washed using distilled water until the pH was neutral. The residue was dried using an oven at 100ºC for 2 hours. The dried sample is then cooled in a desiccator and then weighed until the mass is constant.

Synthesis of Cellulose Acetate

This stage was carried out by reacting 2 g of alpha-cellulose from empty palm fruit bunches with 25 mL of glacial acetic acid and stirring for 1 hour using a magnetic stirrer at room temperature at continuous speed until the cellulose was activated. Next, the acetylation stage was carried out by adding 3 drops of H_2SO_4, 10 mL of acetic anhydride, and 5 mL of distilled water at a temperature of 40ºC. 2 mL of distilled water and 5 mL of glacial acetic acid were added to the solution and reacted for 30 minutes. 100 mL of distilled water was added to the solution and observed, the precipitate formed was then filtered and dried in an oven at 40ºC. The cellulose acetate that was obtained was then weighed and FTIR functional group identification was carried out.

Making Separators

0.7 g $Al(OH)_3$ was dissolved with 42.3 mL DMAc and stirred for 30 minutes, 5.6 g PVDF and 0.2 g cellulose acetate were added to the mixture and stirred again for 24 hours at 70ºC and ultrasonicated for 30 minutes at 27ºC. The resulting mixture is poured onto the glass substrate and flattened using a casting knife to a thickness of < 1 mm. The polymer film was left at room temperature for 1 minute, then immersed in a coagulation bath filled with a mixture of DMAc and distilled water in a ratio of 1：4 until the polymer film separated from the glass substrate. The polymer film was transferred to a distilled water bath for 48 hours and dried in an oven at 60ºC for 12 hours to remove residual water and solvent.

RESULTS AND DISCUSSION

Separator Thickness

Testing the thickness of the separator was carried out using a screw micrometer, calculating the thickness of the separator using the Tappi test method. The separator thickness value is obtained from the average calculation results carried out at five different points. This measurement is carried out to determine the suitability of the resulting separator for its use. The thickness of the separator also affects the mechanical properties of the separator. The separator thickness test results obtained all meet the thickness standard for application to lithium-ion batteries, namely less than 1 mm. Test result data can be seen in Table 1.

Table 1. Thickness test results.

CA Variations (g)	Separator thickness value (mm) 1	2	3	4	5	Average (mm)
0.1	0.12	0.11	0.80	0.80	0.12	0.390
0.2	0.15	0.10	0.15	0.10	0.15	0.130
0.3	0.20	0.20	0.19	0.20	0.20	0.198

The standard used is SNI 2-1707

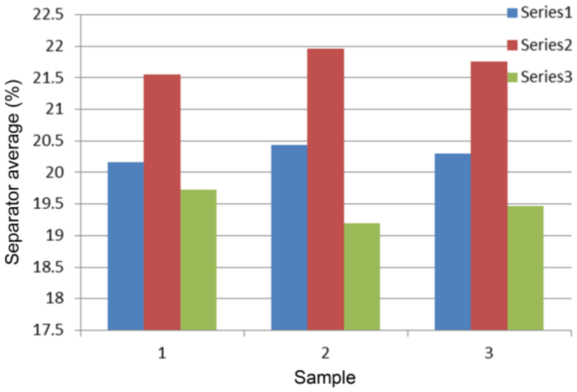

Figure 1. Separator thickness graph.

Separator Elongation

Elongation is the percentage change in film length which is calculated when the film is pulled until it breaks. The analysis results show that the average value of elongation of the separator is the highest in the 0.2 g CA addition variation, namely 21.755%, while in the 0.1 g CA variation, it is 20.305% and the 0.3 g CA variation is 19.465%. The average value of the separator extension can be seen in Table 2.

Table 2. Elongation test results.

CA sample (g)	Elongation (%) 1	2	Average (%)
0.1	20.17	20.44	20.305
0.2	21.55	21.96	21.755
0.3	19.73	19.20	19.465

The standard used is SNI 7818:2014

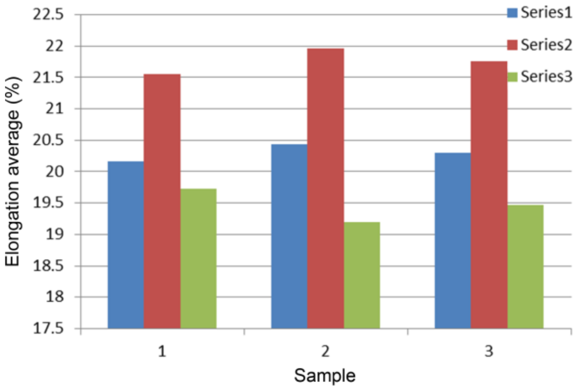

Figure 2. Graph of elongation results.

PSA Testing

Particle Size testing of CA empty oil palm bunches was carried out using HORIBA L300 Particle Size Analysis. The results of this test can be seen in Figure 3. The test results show that the smallest particle is 0.115 µm and the largest particle is 592.387 µm with an average diameter of 73.1870 µm.

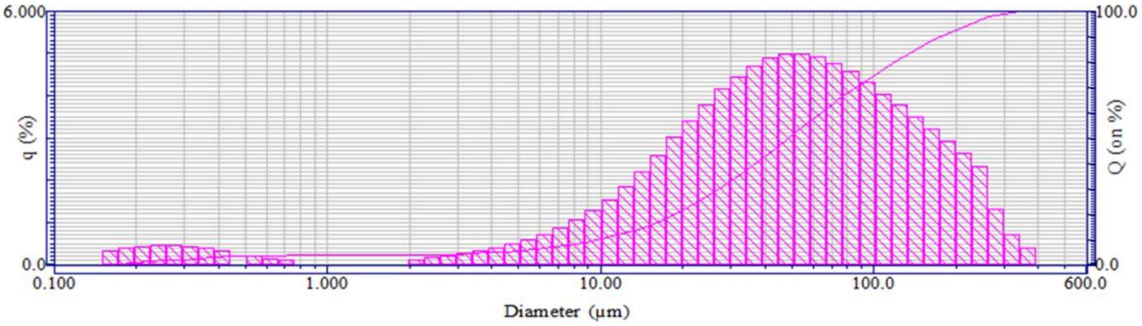

Figure 3. Graph of PSA results.

CONCLUSION

Based on the research that has been carried out, it can be concluded that the use of cellulose acetate from empty oil palm fruit bunches can be used as an alternative in making lithium-ion battery separators. The composition of the type of material the battery separator is made from can affect the results of the separator produced. The separator that has the best elongation properties is produced from a variation of PVDF 5.6 g, $Al(OH)_3$ 0.7 g with CA 0.2 g. The resulting separator is suitable for application to lithium-ion batteries because it meets separator standards.

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