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Physical and Electrical Breakdown Characteristics of Oil-Impregnated Kenaf Paper with the Introduction of External PVA for Transformer Application

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ABSTRACT

This work examines the physical and electrical breakdown characteristics of kenaf paper coated with Polyvinyl Alcohol (PVA) for application in power transformers. The paper was made from kenaf bast fibers using the soda pulping method, whereby the pulps were subjected to 12,000 beating revolutions. PVA with weight percentage concentration up to 6% was introduced to the beaten kenaf through a spin coating approach. The structure of the kenaf paper was examined through Scanning Electron Microscopy (SEM). The physical properties examined were apparent density, Tensile Index (TI), Burst Index (BI), and Tear Index (TeI), while AC breakdown voltage and strength were analyzed for the electrical property. It is found that the beating and external PVA improve the kenaf paper's apparent density, TI, BI, and AC breakdown strength while the TeI decreases.

Keywords: Kenaf paper, physical and electrical properties, PVA coating

INTRODUCTION

Kraft paper has been used as electrical insulation material in power transformers for years due to its high mechanical and electrical strengths (Mathes, 1991). Kraft paper is made from wood fibers, which produce strong paper. Due to recent environmental concerns about extensive deforestation, alternative resources such as

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non-wood fibers are utilized for paper production (Hammett et al., 2001; Kamoga et al., 2013). Fibers such as cotton, manila, hemp, and flax are also utilized to produce papers for electrical insulation (Krause, 2012; Schaible, 1987). Currently, the application of kenaf fibers for electrical insulation is still limited.

Kenaf bast has long fibers that can produce high-strength papers and can be used as electrical insulation in transformers (Umair et al., 2019). Among kraft paper's important characteristics is its ability to retain its strength properties after oil impregnation (Lundgaard et al., 2008). Tensile, burst, and AC breakdown strength characterize kraft paper's mechanical and electrical strengths (Feng et al., 2020; Li et al., 2016). The beating process or chemical treatment can improve a paper's mechanical and electrical properties (Brännvall, 2009; Song et al., 2009). For example, the beating process on pulps improves a paper's mechanical and electrical strengths through fibrillation. On the other hand, chemical treatment can also enhance mechanical and electrical properties by introducing additives (Edeerozey et al., 2007; Wistara & Young, 1999). The most common chemical additives that can enhance a paper's strength are oxidized/natural starches, cationic starches, soluble cellulose derivatives, and synthetic/natural polymers (Auhorn, 2006). Polyvinyl Alcohol (PVA) is a synthetic polymer soluble in water, and it can be used for the chemical treatment of paper. It is biodegradable, non-toxic, and chemically stable. It has high mechanical strength and adhesive properties, which lead to its suitability for a wide range of applications.

The combination of cellulose and PVA can improve the intermolecular and intramolecular hydrogen bonds due to the high concentration of hydroxyl groups, thereby increasing paper strength (Abdulkhani et al., 2013; Moore et al., 2012; Shokrieh et al., 2015; Finch, 2002; Zhang et al., 2019). In addition, the hydrogen bonds can increase the paper's mechanical and electrical strengths (Bao et al., 2011; Medhekar et al., 2010). PVA can be introduced into cellulose internally before the formation of paper or externally after the paper has been produced. The introduction of PVA internally increases paper's mechanical strength by enhancing the hydrogen bonding among fibers. External application of PVA increases the mechanical strength through an extra thin layer of barrier on the surface of a paper. The external PVA can be applied by using the PVA coating process.

PVA coating is the process of external PVA application on the surface of the paper to improve the paper's properties (Schuman et al., 2004). PVA is applied on the paper surface to cement the fibers together and deposit a continuous thin film layer. Electrostatic or non-electrostatic interaction bonds the thin film to the cellulose substrate, which improves a paper's mechanical and electrical properties (Hubbe, 2006). Spin coating is one of the several methods to perform PVA coating. It is a process of depositing coating solution to a horizontal spinning disc through centrifugal force, which results in the removal and evaporation of the solvent and the emergence of a uniform solid film on the surface of the paper (Norrman et al., 2005). PVA can improve the barrier properties of paper, improving the mechanical strength (Schuman et al., 2003, 2004). The PVA coating process is normally used for writing, printing, food, and paper packaging (Schuman et al., 2003). In addition, it can also improve moisture and heat resistance as a result of barrier properties improvement (Edeerozey et al., 2007). The addition of a 10% weight percentage concentration of PVA to cellulose can increase the thermal resistance of paper up to 5% (Nuruddin et al., 2015; Yenidoğan, 2019). Currently, using a polymer such as PVA to improve the mechanical and electrical strengths of kenaf bast fiber-based paper for electrical insulation purposes is still not widely examined.

In this paper, the kenaf paper is developed using the soda pulping technique with 12,000 beating revolutions. The external PVA is applied externally to the kenaf paper through the spin coating approach. The kenaf paper with external PVA weight percentage concentrations of 2%, 4%, and 6% are examined for Tensile Index (TI), Burst Index (BI), and Tear Index (TeI). The oil-impregnated kenaf paper is also prepared to determine AC breakdown voltage and strength.

MATERIALS AND METHODS

Development of Kenaf Paper

The development process of kenaf's paper with the introduction of external PVA is shown in Figure 1. First, kenaf bast fibers were sourced locally, whereby the fiber threads were thoroughly cleaned and screened to remove dust and grime before being cut into 10 cm. In total, 1 kg of raw fibers was prepared through Oven Dry (OD) method. Then, soda pulping was performed whereby the concentration of Sodium Hydroxide (NaOH) was set

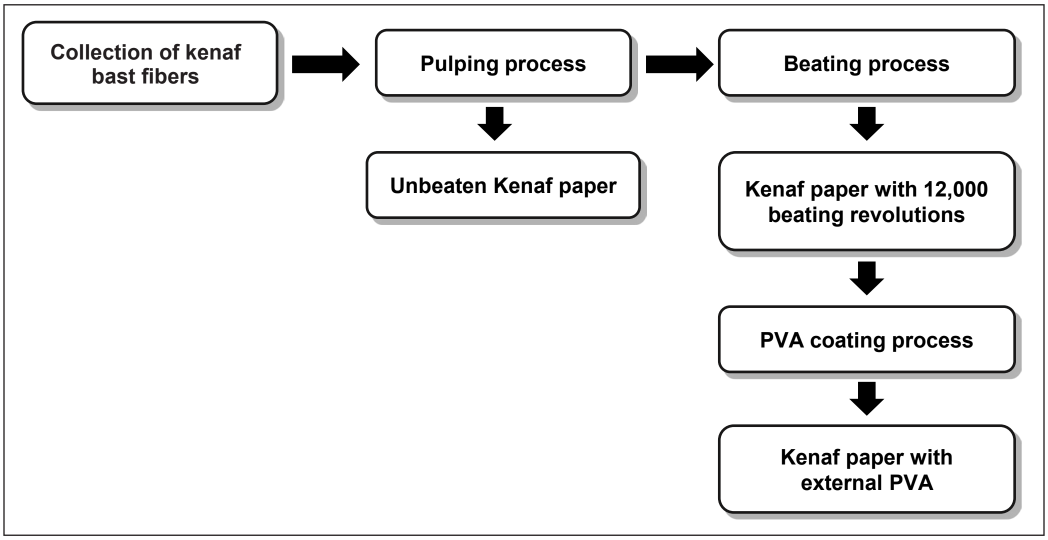


Figure 1. The process to develop Kenaf paper with external PVA

to 14% with the liquor: wood ratio set to 7:1. The liquor is a mixture of NaOH and water. Finally, the raw fibers were cooked in the rotary digester for the soda pulping process. First, the temperature of the rotary digester was set to 35°C with a pressure of 140 psi. The temperature required 90 minutes to reach the maximum set point of 170°C, and it was maintained for 30 minutes during the soda pulping process.

The pulp and paper without the beating process were defined as unbeaten kenaf's pulp and paper. The pulp was subjected to the beating process of up to 12,000 beating revolutions according to the Technical Association of the Pulp and Paper Industry (TAPPI) standard T248. The freeness of the pulp was measured as per TAPPI standard T227. After the beating process, the kenaf paper was formed on the paper machine according to the TAPPI standard T205. The grammage of kenaf paper after the process was 52 g/m^2. Next, the PVA coating was applied on the kenaf paper with 12,000 beating revolutions.

Preparation of PVA Solution

The PVA solution was prepared before the coating process, with weight percentage concentrations set to 2%, 4%, and 6% using Equation 1. The solution was prepared using Fisher Scientific Isotemp heated magnetic stirrer. First, the PVA powder was added to 100 ml of distilled water at a temperature of 90°C using the magnetic stirrer for 2 hours to produce a uniform PVA solution, as seen in Figure 2. Next, the solution was left to cool for 2 hours in a cleanroom at the temperature of 25°C ± 2°C and relative humidity of 25% ± 2%. Next, the PVA solution was applied to the kenaf paper with 12,000 beating revolutions.

$$PVA\ concentration\ (\%) = \frac{Weight\ of\ PVA\ (g)}{Volume\ of\ water\ (ml)} \times 100 \tag{1}$$

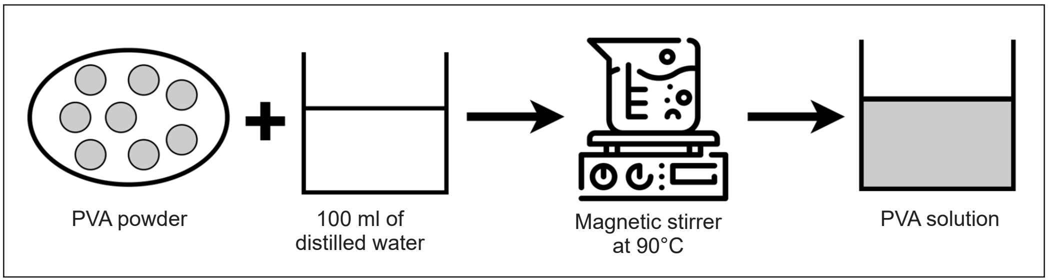


Figure 2. Preparation of PVA solution

PVA Coating Procedure

The spin coating on the kenaf paper was performed by spin coater POLOS SPIN150i as shown in Figure 3. The spin coater consists of a single-phase oil-less piston vacuum pump MAJP-140V(L) with maximum vacuum and power of 60 torrs and 0.41 kW, respectively.

The vacuum pump was used to hold the substrate before the spin coating was performed. The test was performed in a cleanroom at a temperature of 25℃ ± 2℃ and relative humidity of 25% ± 2%. The spin coater's rotation speed and time were set to 2,000 rpm and 60 seconds, respectively. A total of 3 ml of PVA solution was dropped at the center of kenaf paper using a syringe while the spin coater was in operation under the influence of centrifugal forces. The procedure was repeated for all weight percentage concentrations of PVA.

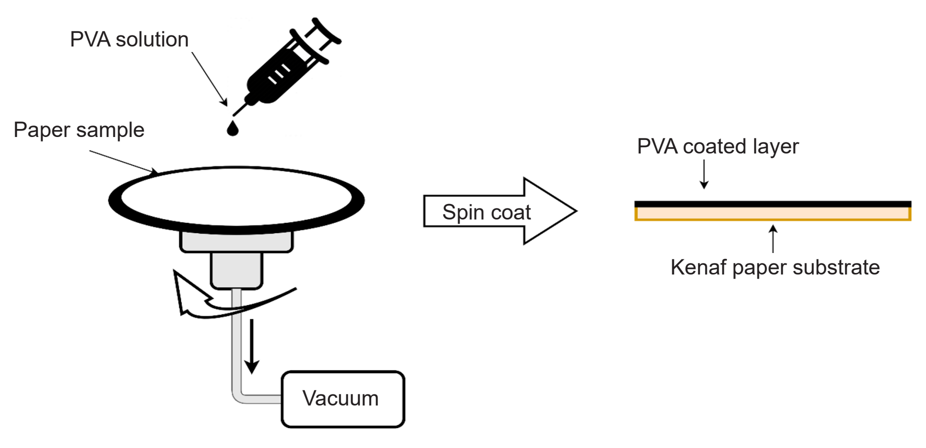


Figure 3. PVA coating process

EXPERIMENTAL SETUP

Physical and Morphological Properties of Paper

The papers were cut into 10 cm × 10 cm squares to determine the grammage of the paper. Equation 2 was used to determine the grammage of the paper based on the mass and area of the paper. The mass of the paper was determined using an OHAUS analytical balance with a sensitivity of ±0.0001g. Equation 3 was used to determine the apparent density of the paper. An L&W micrometer was used to measure the thickness of the paper as per TAPPI standard T411. The coating weight was determined based on differences in grammage between coated and uncoated kenaf papers (Afra et al., 2016; Ni et al., 2021).

$$Grammage\ (g/m^2) = \frac{Mass\ (g)}{Area\ (m^2)} \tag{2}$$

$$Apparent\ density\ (g/cm^3) = \frac{Grammage\ (g/m^2)}{Thickness\ (\mu m)} \tag{3}$$

The surface morphology of kenaf paper with beating revolutions and external PVA was observed using SEM imaging through COXEM EM-30ax. Oil-impregnated papers were cleaned with acetone to remove oil and dried in the air-circulating oven for 24 hours at 105℃ to negate the effect of moisture on the image quality. An ion coater, COXEM SPT-20, was used to prepare the paper to avoid the charging effect on the image.

Tensile, Burst, and Tear Indexes of Paper

The measurement of the tensile strength of kenaf paper was carried out using Buchel B.V horizontal tensile tester as per TAPPI standard T494. The gap between the 2 clamps was adjusted to 100 mm ± 1 mm. The length and width of the paper under test were 150 mm and 15 mm. The crosshead speed of 20 mm/s was used for the tensile strength measurement. The test was performed at a temperature of 23℃ ± 1℃ and relative humidity of 50% ± 2%. The TI was determined based on Equation 4.

$$TI\ (Nm/g) = \frac{Tensile\ strength\ (N/m)}{Grammage\ (g/m^2)} \tag{4}$$

The burst strength measurement of kenaf paper was carried out using a Frank burst machine as per TAPPI standard T403. The paper was cut into length and width of 62 mm × 62 mm. The test was performed at the same temperature and relative humidity conditions as tensile strength. The clamping pressure was set to less than 1,200 kPa. The pressure was subjected to the paper via a rubber diaphragm to a 30.5 mm circular area. The BI was calculated based on Equation 5.

$$BI\ (kPa.m^2/g) = \frac{Burst\ strength\ (kPa)}{Grammage\ (g/m^2)} \tag{5}$$

The tear strength refers to the amount of energy necessary to shred the paper. The tear strength measurement of kenaf paper was performed using an Elmendorf tearing tester based on TAPPI T414. The paper was cut into length and width of 63 mm × 50 mm. The test was performed at the same temperature and relative humidity conditions as tensile strength. Identical clamping pressures of 0.55 MPa were subjected to the clamps that held the paper. A small cut was made at the center of the paper before the tearing force was applied. The TeI was calculated based on Equation 6.

$$TeI\ (mN.m^2/g) = \frac{Tear\ strength\ (mN)}{Grammage\ (g/m^2)} \tag{6}$$

Preparation of the Oil-Impregnated Paper

The kenaf and kraft papers were impregnated in Mineral Oil (MO) to determine the AC breakdown voltage and strength based on the process, as shown in Figure 4. First, the paper was vacuum drying for 48 hours at 105°C and 0.08 kPa. On the other hand, MO was subjected to air-circulating drying for 48 hours at 85°C. Next, the paper was impregnated with MO in a vacuum oven for 24 hours at 105°C and 0.08 kPa.

Measurement of AC Breakdown Strength of Paper

The measurement of AC breakdown voltage was carried out using BAUR DPA 75 C tester, according to International Electrotechnical Commission (IEC) 60156. The test was performed using spherical electrodes with both sides facing each other. The diameter of the spherical electrode was set to 12.5 mm. The walls of the test cell and electrodes were thoroughly rinsed 3 times using MO. In total, 400 ml of MO was gently filled into the test cell to avoid the formation of bubbles. The distance between electrodes inside the test cell was adjusted based on the thickness of the paper (Figure 5). Since the thickness of 1 layer of kenaf paper was low, the measurement could not be computed. Therefore, 2 and 3 layers of kenaf and kraft papers were used to measure the AC breakdown voltage. The voltage ramping rate was set to 2 kV/s. During the test, the paper was moved to a new position after each breakdown. An average of 20 AC breakdown voltages data was utilized for the study. The AC breakdown strength of the oil-impregnated kenaf paper was calculated based on Equation 7.

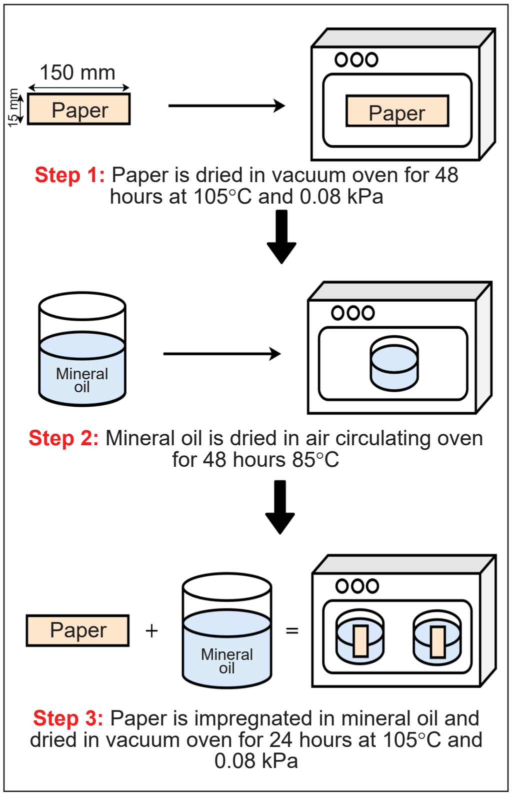


Figure 4. Preparation of oil-impregnated paper

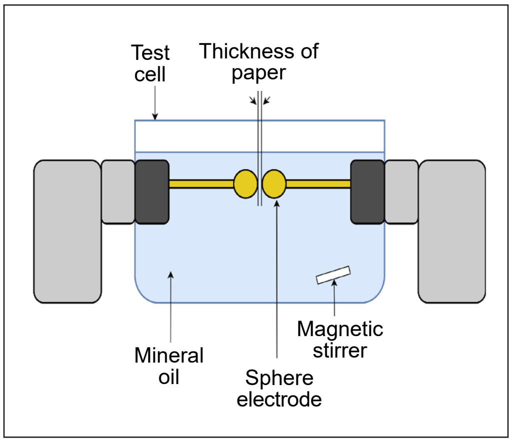


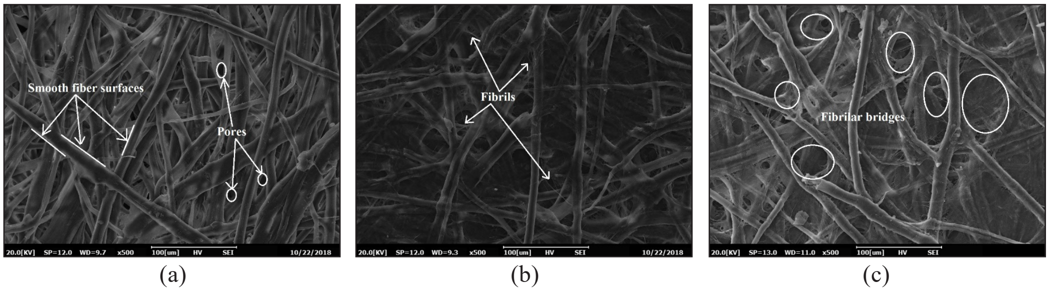
Figure 5. Configuration of AC breakdown measurement

$$AC\ breakdown\ strength\ (kV/mm) = \frac{AC\ breakdown\ voltage\ (kV)}{Thickness\ of\ paper\ (mm)} \quad (7)$$

EXPERIMENTAL RESULTS

Impact of External PVA on Kenaf Fiber

The SEM images of kenaf papers with and without beating revolutions and with external PVA are shown in Figure 6. The kenaf paper, without beating revolutions, shows a clear network of fibers joining together to create a strong bond with each other. The pores in unbeaten kenaf paper are visible, which can promote the flow of MO, as seen in Figure 6(a). There is no fibrillation to the kenaf fibers due to the absence of mechanical and chemical treatments. Fibrillation occurs once the beating to the pulp is applied, as shown in Figure 6(b). Several fibrils on the wall of fiber cells can stimulate bonding and increase the apparent density and mechanical strength. After applying PVA, the fibrillary bridges are improved, and new bonding with fibers and fibrils are formed, which can lead to the increment of the paper strength, as seen in Figure 6(c).



(a) (b) (c)

Figure 6. SEM images of Kenaf paper at a magnification of ×500: (a) Unbeaten Kenaf paper; (b) With 12,000 beating revolutions; (c) With 12,000 beating revolutions and 6% weight percentage concentration of PVA

The Relationship of Coating Weight with PVA Weight Percentage Concentration

The apparent density of kenaf paper increases as the coating weight of PVA increases, as seen in Figure 7. Kenaf paper with 0% PVA weight percentage concentration is referred to as kenaf paper with 12,000 beating revolutions. The coating weight increases linearly at a rate of 0.68 g/m^2 ± 0.02 g/m^2. The grammage increases as the coating weight increases, which leads to the increment of the apparent density

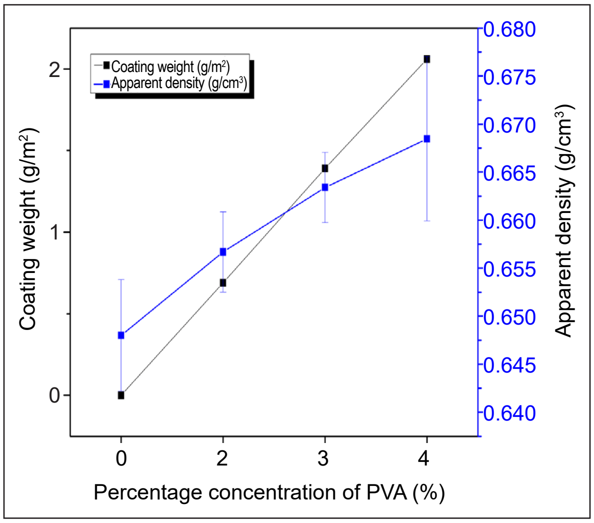


Figure 7. Coating weight and apparent density versus PVA weight percentage concentration

of kenaf paper (Huber et al., 2012). The apparent density of kenaf paper increases by 3% with the introduction of a 6% PVA weight percentage concentration.

Impact of External PVA on Tensile, Burst, and Tear Indexes

The TI of unbeaten kenaf paper is 27.41 Nm/g. It is observed that TI increases almost linearly as the PVA weight percentage concentration increases (Figure 8). The TI of kenaf paper improves by 6%, 8%, and 12% with 2%, 4%, and 6% PVA weight percentage concentrations. The TI of kenaf paper with 6% PVA weight percentage concentration is 81.36 Nm/g, which is 28% lower than the TI of kraft paper. The TI of kenaf paper increases due to the increment of inter-fiber bonding between fibers caused by the presence of the external PVA (Balan et al., 2015).

It is observed that the BI of unbeaten kenaf paper is 1.45 kPa, increasing to 4.59 kPa with 12,000 beating revolutions. The BI of kenaf paper increases as the PVA weight percentage concentration increases, as seen in Figure 9. The BI of kenaf paper increases by 39% with a 2% PVA weight percentage concentration. The increments for BI of kenaf paper with 4% and 6% PVA weight percentage concentrations are quite low. The highest increment for BI of kenaf paper is found at 6% PVA weight percentage concentration, which increases by 51%.

It is observed that the TeI of kenaf paper decreases as the external PVA increases, as shown in Figure 10. The TeI of kenaf paper decreases by 3%, 12%, and 21%, with 2%, 4%, and 6% PVA weight percentage concentration, respectively. Once the inter-fiber bonding exceeds a certain threshold, the TeI of paper normally decreases. It is because high inter-fiber bonding can lead to the brittlement of paper, which causes fracture in a narrow region. As a result, it does not consume high tearing energy, resulting in a lower TeI of paper

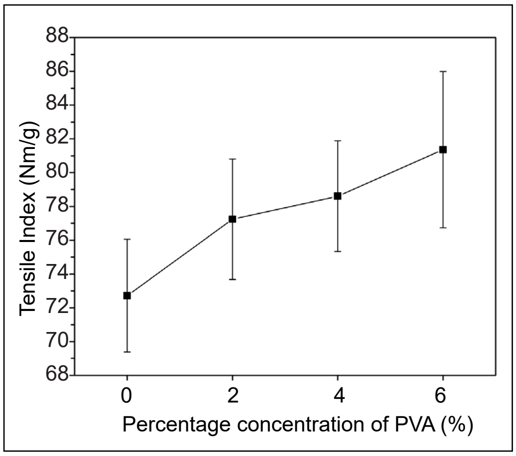


Figure 8. Tensile Index versus PVA weight percentage concentration

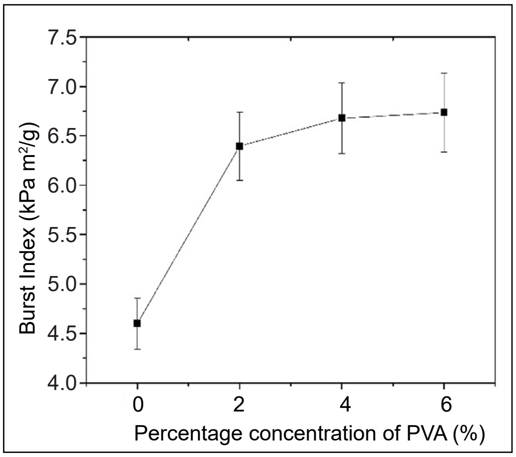


Figure 9. Burst Index versus PVA weight percentage concentration

(Karlsson, 2010). The phenomenon can also lead to the partial dissolution of cellulosic material and affects the TeI of kenaf paper.

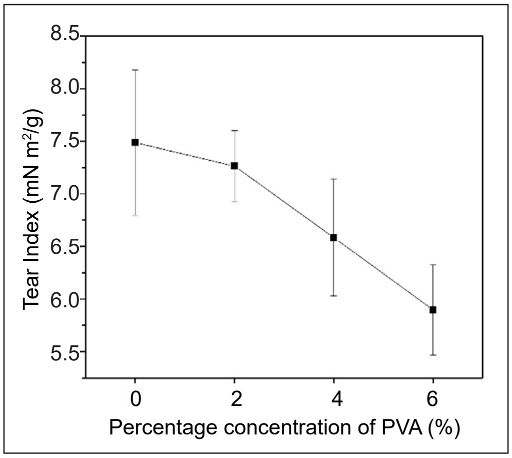


Figure 10. Tear Index versus PVA weight percentage concentration

Impact of External PVA on AC Breakdown Strength

The AC breakdown voltage of MO-impregnated kenaf paper in the presence of external PVA is shown in Figure 11. The unbeaten kenaf paper has a lower AC breakdown voltage than the kenaf paper, with 12,000 beating revolutions for 2 and 3 layers. It is found that the AC breakdown voltage of both types of paper increases as the layer number increases. The pattern is expected since the gap distance increases with the increment of paper thickness, which results in the increment of AC breakdown voltage. The AC breakdown voltage for the 2 layers of kenaf paper remains constant with the increment of PVA weight percentage from 2% to 4%. However, it increases with the increment of the PVA weight percentage to 6%. For 3 layers of kenaf paper, the AC breakdown voltage steadily increases with the external PVA. The highest increment of kenaf paper AC breakdown voltage is observed at 6% PVA weight percentage concentration, which increases by 44% and 72% for 2 and 3 layers, respectively.

The AC breakdown strength of MO-impregnated kenaf paper in the presence of external PVA is shown in Figure 12. The AC breakdown strength is the lowest for unbeaten kenaf paper, and it increases as the external PVA increases. The highest increment of AC

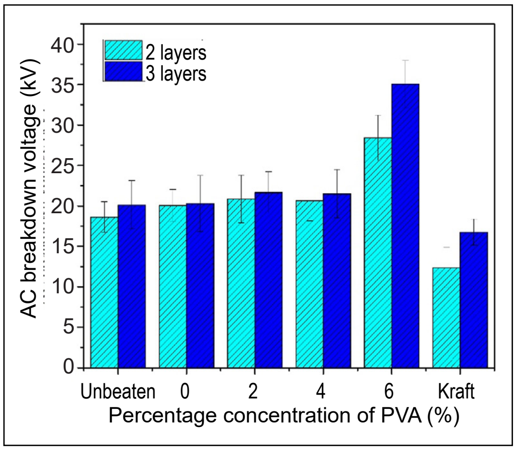


Figure 11. AC breakdown voltage of 2 and 3 layers of Kenaf paper with external PVA

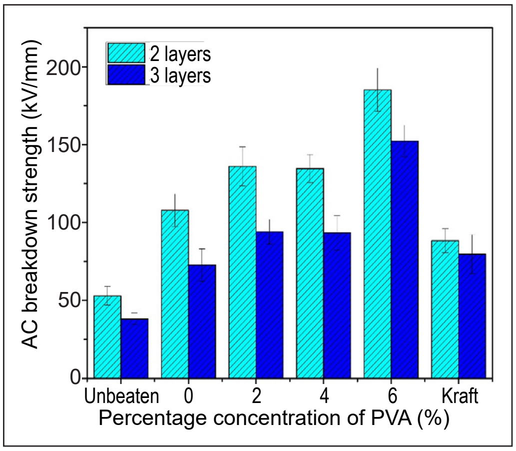


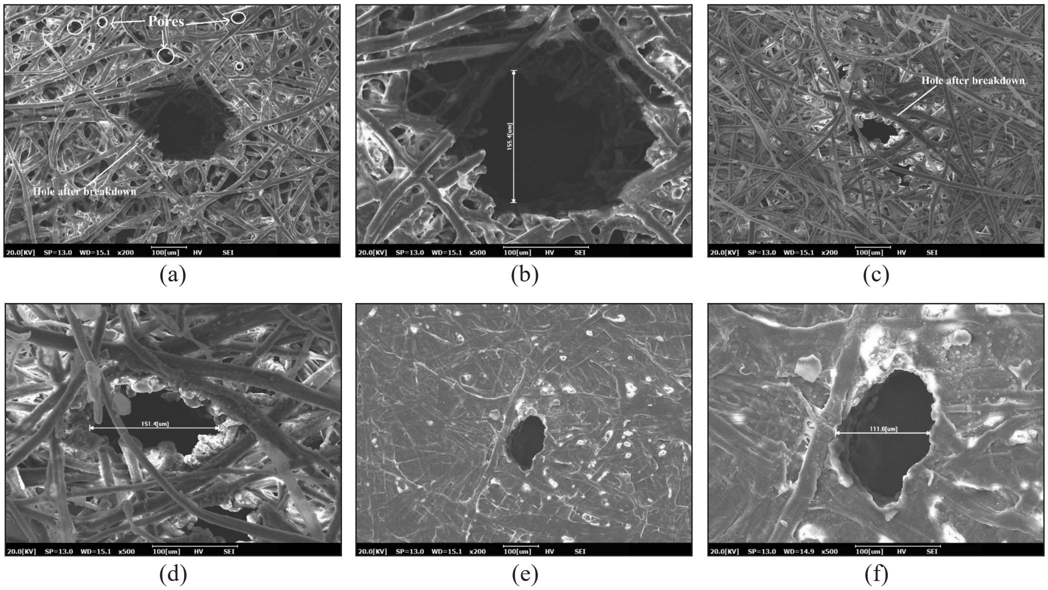
Figure 12. AC breakdown strength of 2 and 3 layers of Kenaf paper with external PVA

breakdown strength is observed at 6% PVA weight percentage concentration, with 74% and 108% increments for 2 and 3 layers of kenaf paper. The AC breakdown strength of kenaf paper in the presence of external PVA is higher than kraft paper.

Effect of Electrical Stress on Kenaf Fiber

The effect of electrical stress on the fiber networks of oil-impregnated kenaf and kraft paper after the AC breakdown voltages can be observed using SEM imaging, as shown in Figure 13. The clear hole is produced through the fiber networks after the AC breakdown test due to the vaporization of fibers under electrical stress.

The AC breakdown strength improves with the decrement of pore size inside the kenaf fiber network (Kamata et al., 1990; Liu et al., 2016). The diameters of the hole caused by electrical breakdown are less than 200 µm for both kenaf and kraft papers. The images obtained by SEM exhibit differences at the edges of the holes for kenaf and kraft papers. The edges of the holes are smooth in kraft paper, whereas it is uneven in kenaf paper with 12,000 beating revolutions and 6% PVA weight percentage concentration. The diameters of the hole after breakdowns in kenaf papers are between 151.1 µm and 155.7 µm, while the diameter of the hole in kraft paper is around 111 µm. There is no clear conclusion that can be carried out to compare the relationship between the diameter of the hole and the AC breakdown strength of kenaf and kraft papers.



(a) (b) (c)

(d) (e) (f)

Figure 13. SEM image of oil-impregnated Kenaf paper after the AC breakdown strength measurement: (a) With 12,000 beating revolutions at a magnification of ×200; (b) With 12,000 beating revolutions at a magnification of ×500; (c) With 12,000 beating revolutions and 6% PVA weight percentage concentration at a magnification of ×200; (d) With 12,000 beating revolutions and 6% PVA weight percentage concentration at a magnification of ×500; (e) Kraft paper at a magnification of ×200; (f) Kraft paper at a magnification of ×500.

DISCUSSION

A paper is a dielectric material where an electric field may be maintained with either zero or near-zero power dissipation, whereby only a few electrons can move on the surface (McShane et al., 2003; Qu et al., 2020). AC breakdown strength of paper is an intrinsic property of dielectric material, independent of the electrode configuration where the electric stress is applied (Amin et al., 2018; Ramli et al., 2014; Zhou & Chen, 2017). The minimum voltage at which the dielectric material fails is known as AC breakdown voltage (Baur et al., 2017; Elanseralathan et al., 2000).

The beating process initiates the fibrillation process and decreases the number of pores. The application of external PVA further decreases pore number, as shown in Figure 6. The porosity and pore size distribution can affect the apparent density of a paper (Gao et al., 2015). It is found that the increment of PVA coating weight increases the apparent density of paper (Figure 7). The increment of apparent density could be due to the reduction of pores in the paper. The AC breakdown behavior is also affected by the fibers' properties and the paper's porous structure (Mo et al., 2019). It is found that the AC breakdown strength of kenaf paper has a positive correlation with fibrillation in fibers since the AC breakdown strength increases with beating and external PVA (Figure 12). The fibrillation process increases the fibrils on the surface of the fibers and reduces the pores in the paper (Wai et al., 1985). The dry-strength additive in the paper structure improves the strength properties through the improvement of the inter-fiber bonding (Balan et al., 2015). The improvement of TI due to external PVA could be caused to the migration of PVA into the internal structure of the paper (Balan et al., 2015). This phenomenon can reduce the number of pores and increase the AC breakdown strength of paper. The reduction of inter-fiber bonding caused by repeated wetting and drying during the coating process is one of the factors that affect the mechanical strength of paper (Balan et al., 2015). Therefore, it is important to control these parameters to further improve the strength properties of kenaf paper with external PVA.

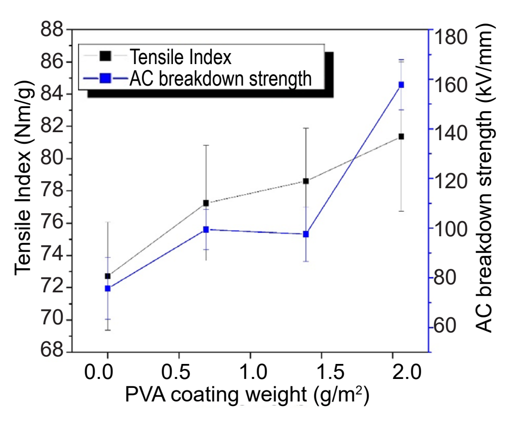


Figure 14. The relationship between AC breakdown strength and TI with PVA coating weight.

The AC breakdown strength for 2 layers of kenaf paper increases by 74%, resulting in an increment of 12% in TI, as shown in Figure 14. The apparent density of kenaf paper is found to increase to 0.67 g/cm^3 with a 6% PVA weight percentage concentration. The AC breakdown strength is 191.41 kV/mm for 2 layers of kenaf paper with a 6% PVA weight percentage concentration. The increment of apparent density also leads to the increment of the BI of the kenaf paper (Umair et al., 2019). Since cellulose and PVA both contain hydroxyl groups, it increases hydrogen bonding, which leads to the increment of the BI of kenaf paper.

The physio-mechanical and electrical breakdown properties of kenaf paper with internal and external PVA are summarized in Table 1. External PVA has a better impact on the physio-mechanical properties of kenaf paper than internal PVA, even at low percentage concentrations. The apparent density of kenaf paper with 6% external PVA is 28% higher than the 12% internal PVA, but it is still 9% lower than kraft paper. On the other hand, the TI and BI of kenaf paper with external PVA are 10% and 31% higher than internal PVA. Kenaf paper's AC breakdown voltage and strength with external PVA is 50% higher than internal PVA.

Table 1
Comparison of physical and electrical breakdown characteristics of kenaf paper with internal and external PVA

Property	Kenaf paper				Kraft paper
	Unbeaten	With 12,000 beating revolutions	With 12% internal PVA (Umair et al., 2020)	With 6% external PVA	
Apparent density (g/cm^3)	0.35	0.648	0.522	0.67	**0.74**
Tensile Index (Nm/g)	27.41	72.72	74.1	81.36	**113**
Burst Index (kPa.m^2/g)	1.45	4.47	5.12	6.73	**-**
Tear Index (mN.m^2/g)	16.52	7.49	6.67	5.89	**-**
AC breakdown voltage (kV)	21.045	21.11	26.54	36.30	**17.605**
AC breakdown strength (kV/mm)	39.88	75.67	89.37	157.76	**83.83**

It is well known that adding enhancement materials can improve paper's mechanical and electrical strengths (Umair et al., 2020). In this study, PVA is one of the enhancement materials that can enhance kenaf paper's physiomechanical and electrical breakdown characteristics for application as electrical insulation in transformers. In comparison to internal PVA, external PVA has positive impacts on the apparent density, TI, BI, and AC breakdown strength of kenaf paper.

CONCLUSION

The external PVA increases kenaf paper's TI, BI, and AC breakdown strength. The TI and BI of kenaf paper increase by 165% and 208% with 12,000 beating revolutions, and these parameters further increase by 12% and 51% with 6% PVA weight percentage concentration. On the other hand, the TeI of kenaf paper decreases by 55% with 12,000 beating revolutions and decreases by 21% with a 6% PVA weight percentage concentration. Furthermore, the kenaf paper has 102% and 90% AC breakdown strengths increments with

12,000 beating revolutions for 2 and 3 layers of kenaf paper. Further increments of 74% and 108% are found with 6% PVA weight percentage concentration for the same number of layers. The external PVA improves kenaf paper's mechanical and AC breakdown strength properties, possibly due to the reduction of the number of pores and increment of the inter-fiber bonding among fibers.

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