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Boron nitride enhanced electric insulation paper for extending transformer thermal life☆

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ABSTRACT

Increasing the thermal conductivity of the oil-paper insulation system can significantly extend the thermal life of transformers by enabling effective heat removal, which reduces high temperature-induced insulation degradation. This research shows that the addition of nominally 25 μm diameter boron nitride particles can achieve sufficient increase in through-plane thermal conductivity of insulation paper to an extent that doubles the transformer thermal life. Importantly, the dielectric breakdown strength can also be enhanced by adding boron nitride particles to the developed insulation paper. Moreover, applying lignin containing cellulose microfibrils into the paper can compensate for the paper strength loss due to the disruption of hydrogen bonding by the addition of BN particles. Building on these findings, we outlined a pathway for a boron nitride-based enhanced insulation system with outstanding through-plane thermal conductivity, enhanced dielectric strength, an appropriate dielectric constant and the needed tensile strength. Additionally, thermal aging experiments showed that the proposed material can have a reasonable thermal life under transformer operating conditions. Overall, this research shows that the mix of thermal, mechanical, and dielectric properties can be successfully tuned to achieve a beneficial insulation system which can significantly enhance the transformer life. As the summary, the proposed material has three times better through-plane thermal conductivity (0.76 W/m·K vs. 0.2 W/m·K), 23 % higher dielectric strength (84.6 kV/mm vs. 65.3 kV/mm), 35 % greater tensile strength (71.26 N·m/g vs. 45.91 N·m/g) and 33 % smaller relative permittivity (3.7 vs. 5.5) than the commercial insulation paper with similar thermal degradation rates.

1. Introduction

Paper made of kraft wood pulp fibers has been traditionally used for electric insulation in transformers [1]. Oil-impregnated kraft paper wrapping around the electrical conductors in a transformer provides the needed electrical insulation and heat transfer. While paper is an excellent electrical insulator, it is an excellent thermal insulator as well. The thermal conductivity of typical transformer oil is 0.2 W/m·K, which is only moderately higher than paper. Overall, the conventional paper-oil insulation system presents high thermal resistance to heat removal from the interior of the transformer, particularly in cases with no oil circulation. Transformer overloading in service and the presence of a small quantities of moisture and oxygen [2,3] can increase transformer internal temperature and accelerates the thermal aging of insulation paper [4]. As a result, insulation deterioration is a major cause of transformer failure [5,6]. Transformers are vital to the efficient and reliable delivery of electricity for the electrical grid. There is a growing concern over the age of the transformer infrastructure in the U.S., because the average

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transformer is 30 years old with an estimated transformer life expectancy of 30 years [7].

Research efforts have focused on improving insulation oil thermal conductivity, specifically adding thermally conductive nanoparticles such as boron nitride, ferriferous oxide, zinc oxide, nanodiamond, into the insulation oil to enhance its thermal conductivity and dielectric strength [8–11]. However, the enhancement of thermal conductivity is only in the range between 5 % and 15 %, primarily due to the limited number of nanoparticles that can be added, typically at the level of approximately 0.1 wt% or 0.1 vol%. Since oil ducts can be very narrow in certain regions of a transformer, the nanoparticles in the nanofluid can agglomerate when flowing through those narrow ducts at high particle loadings. As a result, the performance of the nanofluid deteriorates, which makes this approach problematic.

Recent research indicated that improving thermal conductivity of insulation paper to improve heat transfer is promising to potentially double the transformer thermal life [12]. Specifically, improving through-plane thermal conductivity of the insulation paper in the range of 0.2 to 1.0 W/m·K would result in meaningful reductions in internal temperatures and thus increasing lifetime of transformers. Here, we demonstrate that it is possible to significantly improve the through-plane thermal conductivity of kraft insulation paper by novel design of paper using proper constituents. The approach adopted in this study is adding micrometer-sized boron nitride (BN) particles during paper making to increase the effective thermal conductivity of oil-paper combination. Since BN has high intrinsic thermal conductivity and it is a good electric insulator, in concept, BN particles can be utilized to design high thermal conductivity insulation systems for transformers.

A persistent challenge in insulation material design is achieving the desired enhancement in one aspect without simultaneously introducing a significant deleterious change in another [13]. The overall functionality of insulation material rests on a combination of dielectric, mechanical, and thermal properties. These properties are interdependent and affected by the raw material structure, the composition of the raw material matrix and the fabrication method. The key parameters that need to be met to result in an advanced insulation system for future transformers are illustrated in Fig. 1(a). The goal for the novel insulation paper development is to achieve a through-plane thermal conductivity approaching 1 W/m·K while simultaneously improving or with minimal negative impact on its dielectric, mechanical and thermal aging properties.

The concept of using nano- or micron-size particles to enhance the insulation material's performance in one or multiple aspects can be found in the publications dating back to early 2000s [14,15]. Although most early studies focused on improving the dielectric performance of polymer insulation materials, the improvement of thermal conductivity while maintaining electrical properties is still a topic of broad interest today [16–18]. This concept can be also applied to the insulation paper material such as adding nano- or micron-size ceramic particles or nanofibrillated cellulose to enhance the insulation paper performance in certain aspects. It was shown that using Al_2O_3-nanoparticle-modified insulation paper cellulose can improve the insulation paper thermal stability, thus extending the transformer lifetime [19]. Furthermore, adding nano-TiO_2 particles improved the insulation paper dielectric performance with minimal effect on paper tensile strength [20]. It was also reported that the modified insulation paper can have smaller relative permittivity with higher dielectric strength when adding montmorillonite (MMT) [21]. Furthermore, studies demonstrated that adding nanofibrillated cellulose or modifying the cellulose structure can improve the dielectric, mechanical or thermal stability of the insulation paper [22–25]. Unfortunately, all these literature studies either did not show improvement in the thermal conductivity of the proposed material or the resulting thermal conductivity was not reported. Furthermore, performance evaluation was limited to one or two materials properties listed in Fig. 1(a) and the overall performance of the proposed material for power transformer application was not clear. Abundant literature on thermally conductive cellulose material is available. These materials, however, were not designed for power transformer applications [26–28] and thus did not focus on constraints imposed by power transformers in achieving great thermal conductivity. For example, only the performance of virgin or resin-enhanced paper sample was presented with no performance analysis of oil-impregnated paper samples and the suitability of the studied material for the insulation oil environment [26]. Pure cellulose nanofibrils (CNF) as the matrix material can improve paper tensile strength, but the cost is high, and the resulting paper lacks needed flexibility for wrapping conductors in transformers [27]. Fabrication methods such as aerogel or film casting require replacing the existing insulation paper production line/equipment, therefore, not practical due to intensive capital investment and commercial scalability [28]. As the summary, there are still a lot of research interest and exploration space today for the transformer improvement via adding nanoparticles in insulation liquids [29,30] or development of novel papers [31].

In view of the above discussion, the objective of the present study focuses on design novel insulation paper specifically for the transformer application, which requires to meet the design criteria summarized in

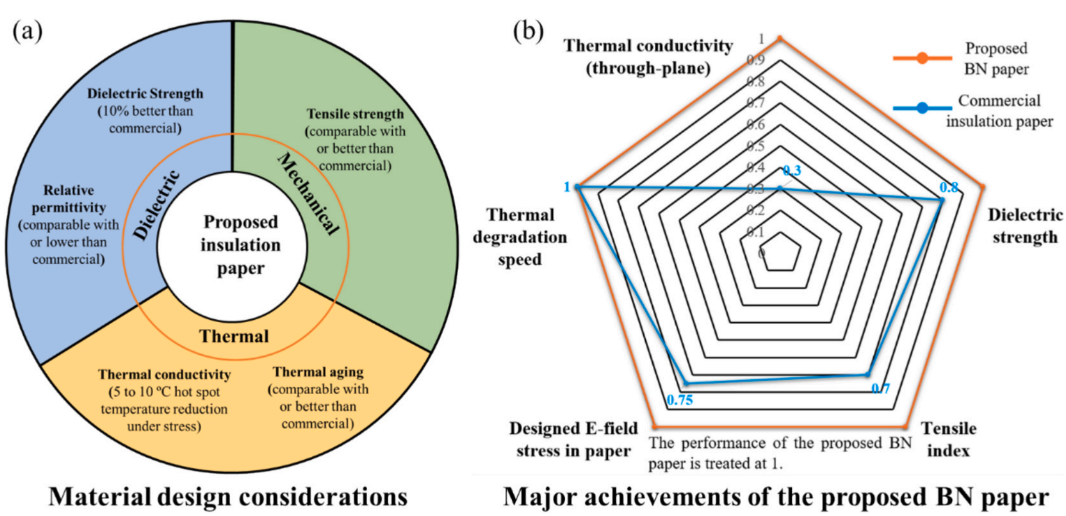

Fig. 1. (a) The design considerations about thermal, dielectric, and mechanical properties of a desired novel insulation paper that has twice the thermal life of current commercial paper for power transformer application (b) The major performance comparisons between our proposed BN paper and the state-of-the-art commercial product, where the performance of our proposed BN paper is treated as 1.

Fig. 1(a). Experiments showed that the designed paper can be produced using a laboratory filtration process that is consistent with procedures adopted in pulp and paper research and adaptable to modern industry papermaking. Our goal is to significantly improve transformer life by improving heat transfer in transformer insulation system and decreasing paper thermal degradation through thermal aging. The major performance comparison between our proposed BN paper and the state-of-the-art commercial product is shown in Fig. 1(b). By using the model established in [12], the hotspot temperature of the transformers can be decreased by approximately 9 °C by using the proposed BN paper and thus, the proposed BN paper is expected to double the transformer thermal life.

2. Materials and methods

2.1. Material preparation and novel paper fabrication

BN particles are available commercially with different morphologies: platelet, turbostratic and agglomerated. Platelets have a deleterious geometry for this application, namely 2D property. The thermal conductivity of platelet BN is greater along the in-plane direction than in the through-plane direction. Therefore, platelet BN paper offers limited advantages for power transformer insulation systems [32]. The agglomerated particles are closer to spheres, but larger than the non-spherical turbostratic BN. Both, however, can provide significant enhancement of the through-plane thermal conductivity. Therefore, turbostratic BN particles and the smallest available agglomerated BN particles were evaluated in our research. Results showed similar overall enhancement in through-plane thermal conductivity. Because the average diameter of turbostratic BN is much smaller (5 μm or 12 μm) than conventional agglomerated BN (25 μm and more), the design trade-off considerations and the optimized fabrication approaches may differ. Here we only present the study using agglomerated BN.

Commercially sourced unbleached, low lignin content kraft softwood (loblolly pine) pulp fibers [33] is used for papermaking. The as-received wood pulp fibers have a length weighted average length of 2.56 mm measured by a Fiber Quality Analyzer (FQA, LDA02, OpTest Equipment Inc., Canada). The chemical composition (Table S1) and morphology (Fig. S1) of the fibers were analyzed. The morphology of the BN particles is typical aggregates (Fig. S2) of diameter 25 μm (D50). The as received kraft pulp fibers were first screened to remove knots and then mechanically refined to Canadian Standard Freeness (CSF) of 120 mL (Fig. S3) which is determined by an ISO standard [34].

The same pulp fibers were used to produce lignin containing cellulose microfibrils (LCMF) to provide additional surface area compared with refined pulp fibers. LCMF was made by fibrillating the refined pulp fibers (CSF 120) suspension at 2 % solids in a stone disk grinder as described previously [35]. LCMF is coarser with less degree of fibrillation than CNF reported in the literature [23–27] and therefore more cost effective. Based on a previous investigation [33,35], four grinding passes at 800 rpm with a disk plate gap setting of −100 μm provided an appropriate degree of fibrillation (Fig. S4). Approximately 15 wt% of LCMF was incorporated into the proposed BN paper by substituting the same amount of CSF 120 wood pulp fibers, and thus to maintain the BN paper mechanical strength and improve BN retention. A control hand sheet made of 100 % CSF 120 kraft wood fibers was also produced for comparison with the novel BN paper. A commercial insulation paper of 125 g/m^2 (gsm) (LK-125, calendered, non-thermally upgraded kraft paper, density 1 g/cm^3) from Weidmann (Weidmann Electrical Technology AG, Rapperswil, Switzerland) was also used as a control.

Agglomerated commercial BN particles from Momentive Technology (Part No. PTX25, Momentive Technology, Strongsville, USA) with a nominal diameter of 25 μm, were used in this study. The loadings of BN in weight percentages (wt%) of lignocellulosic materials (= Fibers + LCMF) were 50 wt%, 70 wt% and 100 wt%. BN retention rates were determined to be 83.4 %, 85.5 % and 85.9 % at these loadings to result in actual loadings of 41.7 %, 59.9 %, 85.9 %, respectively, based on a standard ash test method [36]. Laboratory BN handsheets (papers) were fabricated through filtration method following the TAPPI standard method T205 [37] with slight modifications as shown in Fig. 2. Some steps used in our fabrication method are also reported in [38,39]. Since there are no chemical interactions between BN particle and wood fibers, no chemical structure changes and no new material production are expected in the proposed BN paper. As a matter of fact, adding BN particles can disrupt hydrogen bonding in the fiber network by blocking bonding sites and thus LCMF is added to compensate for the reduction of hydrogen bonding.

Oil-impregnation of the resultant BN and control papers was conducted using the following process. The insulation oil (Cargill FR3 fluid, natural ester transformer oil) was dried at 115 ℃ under vacuum for 18 h. The insulation paper was then dried at 105 ℃ for 3 h. The dried oil and paper were combined in a vacuum environment. Air in the insulation paper was expelled in this step as could be observed from the visible air bubbles in the oil. After keeping vacuum for several minutes, the pressure inside the oven was reset to atmospheric pressure and followed by a second vacuum cycle. Repeating the procedure 3 to 4 times can minimize the air in the paper until no air bubbles were observed in the insulation oil. Finally, the oil-impregnated paper was kept at 105 ℃ under vacuum for 12 h, to further minimize the effects of air and moisture on the insulation system. More details and pictures of the oil-impregnation procedure can be found in Figs. S5 and S6 of the Supplementary Information (SI).

2.2. Characterization of insulation dielectric properties

Insulation paper needs sufficient dielectric strength to withstand the voltage stress among different parts inside transformers, such as high voltage windings, low voltage windings and magnetic cores. For a given voltage rating, focusing on insulation perspective, the greater the dielectric strength of the insulation paper, the thinner the sheet needs to be applied, which means less thermal resistance and better thermal transfer. Moreover, the greater the initial dielectric strength, the longer the electrical life of the insulation paper can be, because the dielectric strength also decreases with paper thermal degradation. Thus, the dielectric strength of an insulation paper is important and must be evaluated. Greater dielectric strength can provide more flexibility in transformer designs and help achieve a good electric life.

The experimental setup used to measure the dielectric strength of the samples is shown in Fig. 3. A 60 Hz power supply provides the test voltage with a rate-of-rise of 750 V/s. The high voltage passive probe is used for voltage measurement and an oscilloscope records the breakdown voltage profile. All voltages are rms values.

The parallel plate electrodes were used to apply the electric field (E-field) stress to the test samples. Both electrodes were cylindrical. The dimensions of the top (HV) electrode were 25 mm in diameter and 25 mm in height, with the bottom (GND) electrode75 mm in diameter and 35 mm in height. The corners of both electrodes were rounded with a 3 mm radius. The top and bottom surfaces of the electrodes were flat and had parallelism within 5 μm. Slots and tapped holes were designed and fabricated on the electrodes for the electric connections. Three measurement slots were added on the high voltage electrode, separated by 120 degrees, in order to measure the interelectrode space after the insulation paper coupon was inserted. The dimensions, pictures and design details about the electrodes system can be found in the SI (Fig. S7).

Measurement procedures follow IEC and ASTM standards [40,41]. Based on the breakdown voltage and the thickness of each sample, the breakdown strength was calculated. According to the IEC standard [42], the expression for the cumulative breakdown probability density function of the two-parameter Weibull distribution is shown in (1):

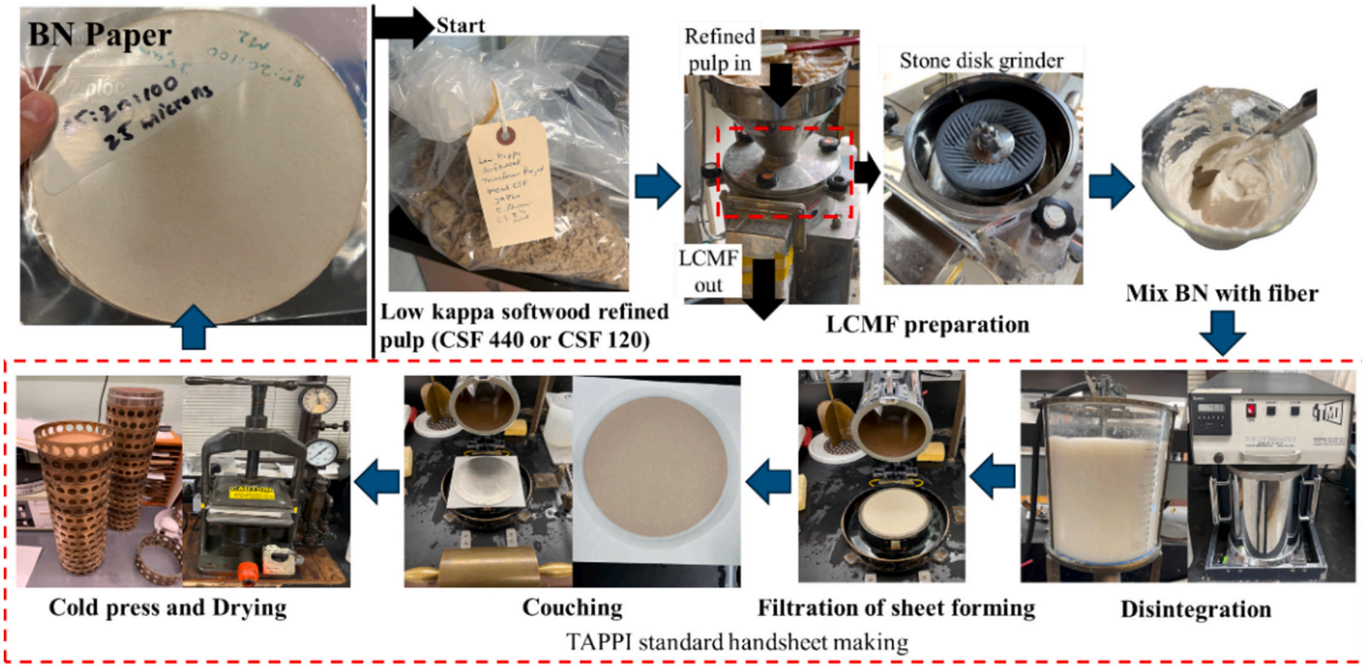

Fig. 2. Flow diagram shows the fabrication of boron nitride enhanced insulation paper.

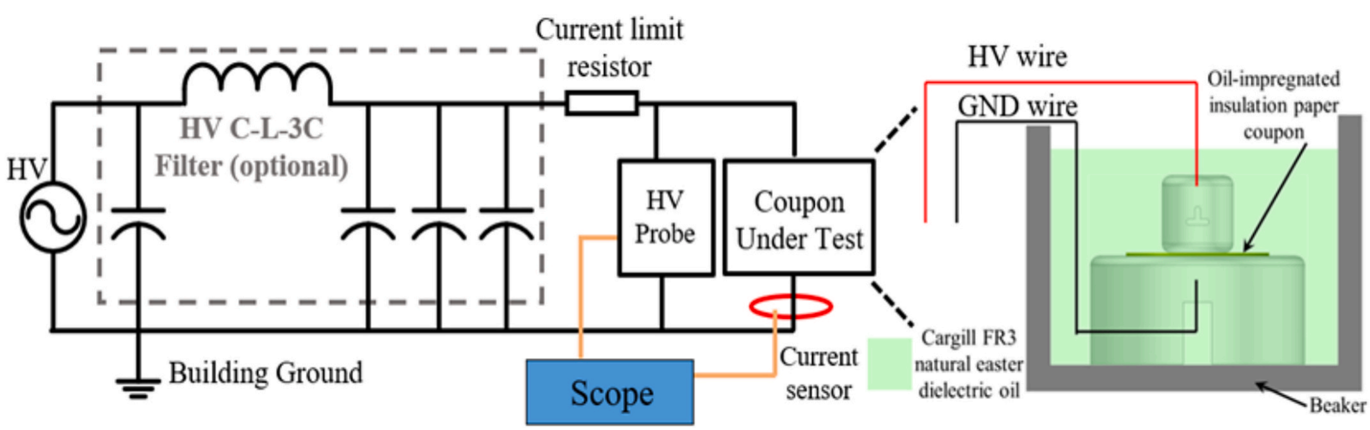

Fig. 3. Short-time breakdown test setup diagram and side view of the parallel plate electrodes system for the proposed BN paper dielectric strength evaluation.

$$F(t;\alpha;\beta) = 1 - exp\left\{-(t/\alpha)^{\beta}\right\} \quad (1)$$

Where t is the measured breakdown strength; $F(t)$ is the probability of failure at the breakdown strength less than or equal to t; α is the scale factor and is positive; β is the shape factor and is positive. For a two-parameter Weibull distribution, the scale factor α represents the breakdown strength at which the failure probability is 0.632. The shape factor β is a measure of the breakdown strength variation. The higher the value of β, the less the variation in the experimental data.

For example, if α is equal to 100 kV/mm for a given material, then this material has a breakdown chance of 63.2 % when the applied E-field stress is 100 kV/mm. For research, the scale factors can be used to compare different materials at the same test condition. However, transformers are not designed based on the average breakdown strength. Rather, a safety margin is selected, typically based on the 1 % probability of failure [43]. Therefore, the breakdown strength at 1 % cumulative probability is important and also provided in this study.

The relative permittivity (ε_r) values of the oil-impregnated insulation paper sample were measured by placing the paper sample between two parallel-plate electrodes (cylindric shape). The capacitance of the paper sample was then measured by using an LCR meter (Hioki IM3536, Hioki, Nagano, Japan). Since the system can be treated as a parallel-plate capacitor, the relative permittivity of the medium (the paper sample) between the metal plates can be estimated by using equation (2) when the capacitance, electrode size and the interelectrode spacing are known.

$$\varepsilon_r = Cd/\varepsilon_0\pi r^2 \quad (2)$$

Where C is the measured capacitance; r is the radius of the top electrode; d is the interelectrode spacing after the test coupon is applied; ε_r is the relative permittivity of the test coupon; ε_0 is permittivity of free space and its value is 8.8542 F/m.

2.3. Paper through-plane thermal conductivity measurements

A commercial measurement system (Hot Disk TPS2200, Hot Disk, Gothenburg, Sweden) is used to measure the through-plane thermal conductivity of paper samples via a hot-disk method, as shown in Fig. 4.

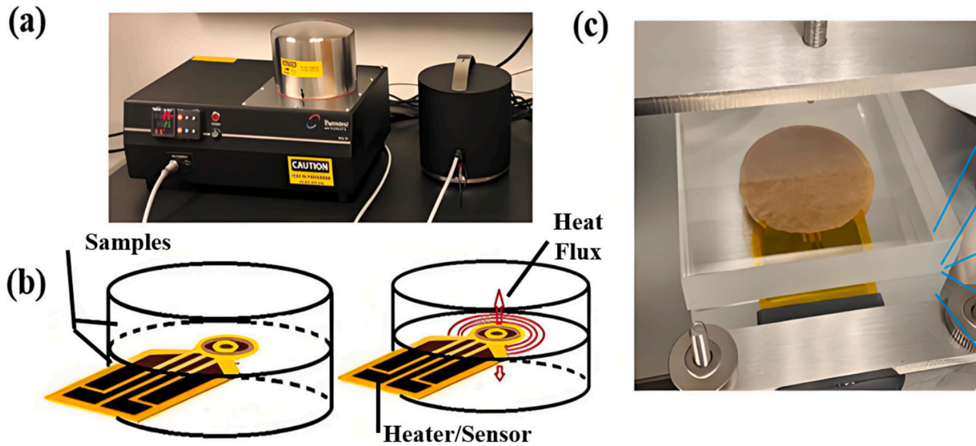

Fig. 4. (a) Photo of thermal conductivity measurement system (b) The schematic diagram of the thermal conductivity tester and (c) Photo of the specimen stage illustrating the test unit.

2.4. Characterization of paper mechanical strength

Paper tensile strength is the most relevant mechanical property of transformer insulation papers for wrapping metallic components. The tensile strength of the papers was measured according to standard methods [44,45]. Paper samples were cut into strips 15 mm wide and 80 mm long (the span length is 50 mm with 15 mm for each grip). Twelve specimens were prepared for each paper sample to obtain statistical significance as papers are highly anisotropic in terms of formation and strength. Tensile tests were conducted on the universal testing system Instron 3367 (Instron, Norwood, USA). Because paper is highly compressible and nonuniform due to anisotropic formation, accurate thickness measurements are difficult to obtain. Consequently, tensile index is commonly used rather than tensile stress to compare the tensile strength among different samples.

The tensile index (TI) is the failure tensile stress divided by the density of the paper. Mathematical manipulation can yield Eq. (3). By using TI instead of tensile stress, the difficulties in accurately measuring thickness of paper can be avoided.

$$Tensile\,Index\,(N \bullet m/g) = \frac{F}{b \bullet G} \tag{3}$$

where F is the force in Newtons obtained at the point of failure, b is the strip width in meters. G is the paper grammage in g/m^2.

2.5. Paper thermal aging test

The procedure adopted for thermal aging evaluation, including the apparatus, test procedure, and data analysis, meets the requirements of existing standards [46].

A 3D drawing of the thermal aging tank with its major body dimensions can be found in Fig. 5. The aging tank included a stainless-steel base with the cylindrical body soldered on it and four tapped mounting holes on the corners, a stainless-steel lid with four mounting holes on the corners, two high-temperature valves and one pressure release device, four bolts and one high-temperature polymer washer (Part. No. Bluegard 3000, Garlock Sealing Technologies, Palmyra, USA). The cylindric container had an inner diameter of 44.45 mm and height of 111.76 mm.

Based on the standard [46], tensile strength of the paper samples after thermal aging test should be measured to provide information for lifetime estimation. To match our tensile strength test procedure as described earlier, paper samples with 15 mm width and 80 mm length were prepared and loaded into the thermal aging tanks for the tensile strength evaluation. More than 20 paper samples were loaded into each test vessel, to result in a total volume of the paper samples at

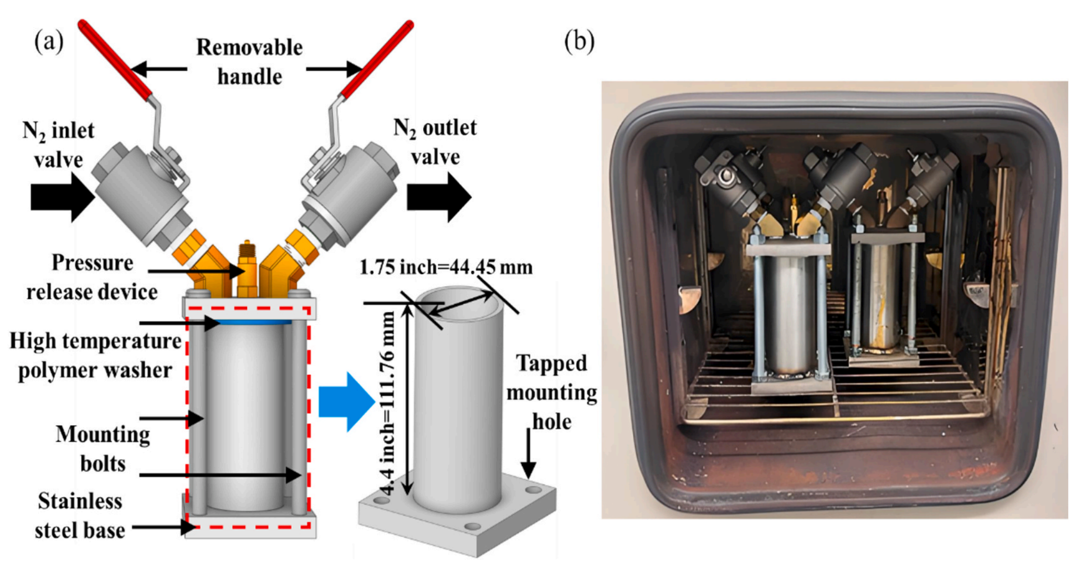

Fig. 5. (a) The 3D drawing of the thermal aging tank with its major body dimensions (b) Multiple sealed tanks in the oven to complete a set of data for the thermal aging test.

approximately $6cm^3$. The volume ratio between oil and insulation paper was fixed at approximately 16.3 based on the standard. Thus, the oil level inside each vessel was more than 7 cm, and the paper samples were completely submerged in the insulation oil. Pure copper chips were added to each vessel with the ratio of oil weight to copper area at 90 g/cm^2. After loading the paper samples, copper chips and insulation oil, the test vessels were sealed with the headspace inside each tank filled by dry pure nitrogen. The test vessels were then placed in an oven at 150 °C as shown in Fig. 5(b). The relief valve on the lid can keep the headspace pressure inside the vessel at approximately 1.7 atm regardless of the test temperature and the initial oil level. Time-dependent aging information was obtained by moving one vessel out of the oven every three weeks to terminate the testing of one sample set. A total of three vessels were used to complete the thermal degradation test for one type of paper samples and the entire experiment lasted 9 weeks or 1512 h.

2.6. *SEM analysis of paper physical structure*

The morphology of the paper was characterized using a TESCAN XEIA SEM (from TESCAN GROUP, Kohoutovice, Czech Republic) microscope. The freeze-dried sample was mounted on conductive carbon tape and sputter-coated with gold to enhance conductivity.

3. Results and discussion

3.1. *Novel BN paper physical structure*

The visual appearance of the novel BN paper is shown in Fig. 6(a) in comparison with the control kraft paper sample in Fig. 6(b) without BN and LCMF. The laboratory handsheet paper samples were round with a diameter of 16 cm. The BN paper had a whitish color due to the addition of BN particles while the control paper shows typical brownish color of unbleached kraft papers. SEM imaging revealed the presence of BN particles on the BN paper surface embedded in the fiber/fibril networks in Fig. 6(c). BN distribution within the BN paper can be observed in SEM image of the paper cross-section in Fig. 6(d). BN regions can be found from the BN paper top surface to its bottom in a layered structure separated by lignocellulosic fibers as insulation binders. Kraft pulping process, along with extensive mechanical fiber refining (CSF120), collapsed the lumens of softwood fibers as shown by the SEM image in Fig. 6(d). However, voids were present as shown by the black areas in the SEM image illustrating the BN paper remains a porous composite material. These voids can be filled with insulation oil, which increases the dielectric breakdown strength and through-plane thermal conductivity of the BN paper-based insulation system.

3.2. *BN paper through-plane thermal conductivity*

The oil-impregnated lab control paper made of only the CSF 120 refined kraft pulp fibers without BN had a through-plane thermal

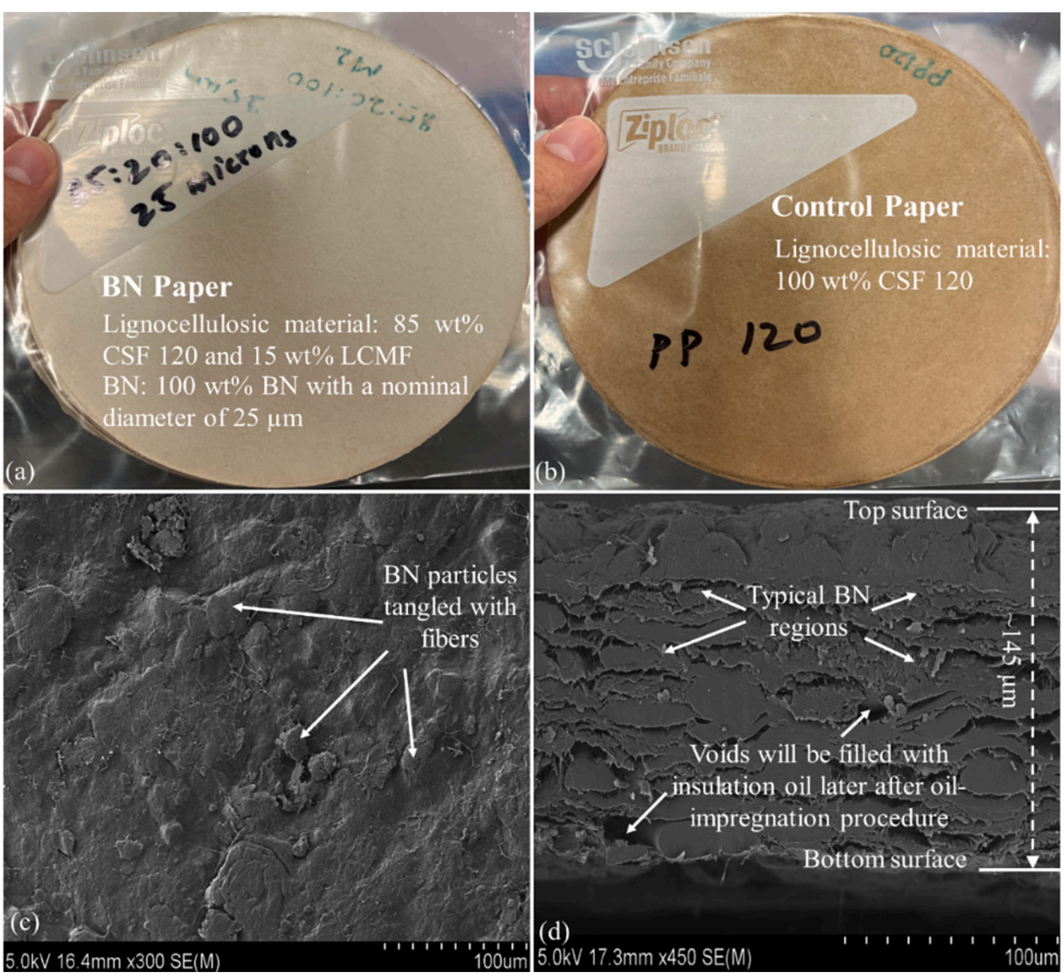

Fig. 6. (a) Photo of the control paper (b) Photo of the BN paper (c) SEM image showing the BN paper surface morphology and (d) SEM image showing the cross-sectional BN paper structure.

conductivity of 0.1 W/m·K as shown in Fig. 7(a). The oil-impregnated commercial paper as another control as a thermally enhanced commercial product had a through-plane thermal conductivity of 0.2 W/m·K. This commercial paper had a grammage of 125 g/m^2 and typical thickness of 127 μm. The lab control paper had a grammage of 60 g/m^2 based on the mass of fibers with a much thinner thickness of 80 μm. Therefore, the commercial paper had a typical density of 1 g/cm^3, while the density of the lab control paper was only 0.75 g/cm^3. So, the greater though-plane thermal conductivity of the commercial paper was a result of higher density.

Under the BN loadings of 50 wt%, 70 wt%, and 100 wt% in weight percentages (wt%) of lignocellulosic materials (= Fibers + LCMF), the lab BN paper had a grammage of 85 g/m^2, 96 g/m^2 and 112 g/m^2 (count the mass of fibers including LCMF and BN) with an average of BN paper thickness of 108 μm, 121 μm and 145 μm, respectively.

Adding agglomerated BN particles significantly increased the resultant paper through-plane thermal conductivity to reach 0.76 W/m·K after oil-impregnation at BN loading above 70 wt% as shown in Fig. 7(a). Significant through-plane thermal conductivity enhancement was also achieved even at BN loading of 50 wt%. As BN loading increased to 70 wt%, however, the through-plane thermal conductivity was plateaued. Further increase in BN loading did not result in significant increase of through-plane thermal conductivity. Nevertheless, a more consistent thermal conductivity enhancement was achieved at BN loading of 100 wt% as evidenced by the smaller standard deviation than those at lower BN loadings.

3.3. BN paper dielectric properties

The relative permittivity of the constituent materials in transformers is a design consideration. The combinations of the applied voltage, the geometry, and the material relative permittivity determine the electric

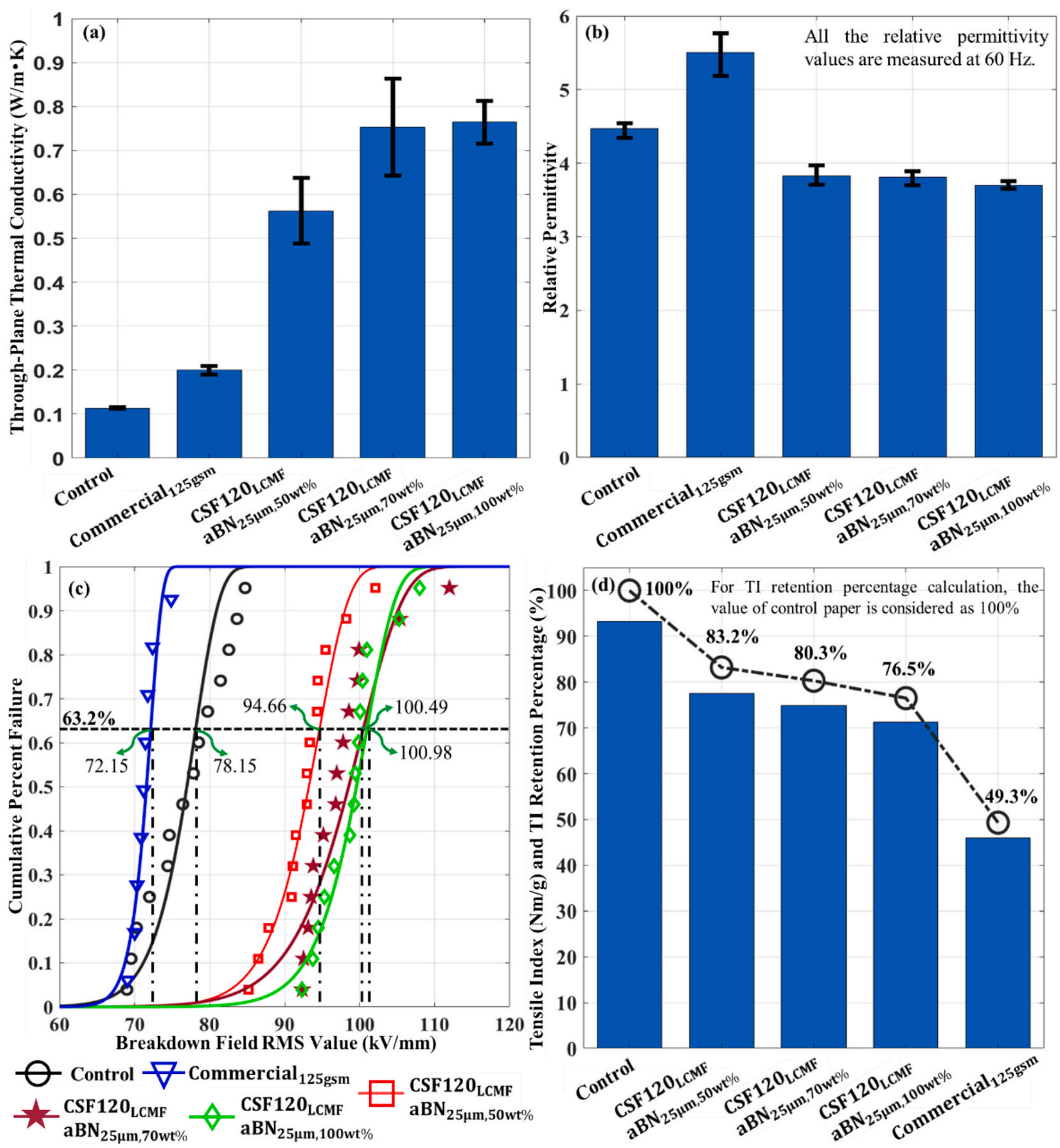

Fig. 7. (a) Thermal conductivities of BN-based papers and papers without BN (commercial control paper and lab control paper) (b) Relative permittivity measurements for oil-impregnated insulation paper samples with or without agglomerated BN (c) Breakdown strength evaluation for the control, commercial and BN paper samples and (d) Tensile strength characterization of insulation paper samples with or without BN. Note: The subscript text label after the letters "aBN" describes the BN type, with "25 μm" indicating the nominal diameter of the BN particles. The wt% indicates the quantity of BN introduced as a weight percentage of all cellulosic materials (fibers + LCMF). "Control" is lab control paper of 60 g/m^2 (gsm). "Commercial$_{125gsm}$" is Weidmann LK125 insulation paper with 125 g/m^2 (gsm). All the BN samples contain 15 wt% LCMF to balance the tensile strength reduction after BN is added. 15 wt% of LCMF is applied to substitute the same amount of CSF 120 fibers, which helps keep the fiber quantity the same among all the test samples.

field (E-field) distribution in the transformer. All the relative permittivity values reported in this article were measured under sinusodial excitation at 60 Hz, which is the line frequency in the U.S. The oil-impregnated novel BN paper samples had the relative permittivity values ranged between 3.7 and 4 as shown in Fig. 7(b), lower than those of the commercial and lab control samples. The relative permittivity values for cellulose, BN, and natural ester oil are approximately 6.1 [47], 3.9 [48] and 3 [49], respectively, suggesting lower relative permittivity of the novel BN paper was due to the addition of BN. As a result, the smaller relative permittivity for insulation paper shifts more electric stress to the insulation paper instead of the weaker insulation oil. Fortunately, the BN paper has an adequate breakdown strength margin, as shown in Table 1 and to be discussed later, to withstand the increasing share of electric stress due to its lower relative permittivity induced the changes in the E-field distribution. The dielectric strength result suggests that it is not likely necessary to change present transformer designs, by using the proposed BN insulation paper to replace the existing commercial insulation paper.

The dielectric breakdown strength values of the paper samples were determined by fitting the failure probability data to the Weibull distribution function as shown in Fig. 7(c). The resultant fitting parameters are listed in Table 1. All lab-made handsheet paper samples in general had a lower shape factor β than the commercial paper sample, indicating that the commercial paper had better morphological consistency. However, all the BN paper samples had a greater scale factor α, i.e., the paper dielectric breakdown strength, than paper samples without BN. As a result, the novel BN paper samples had a greater dielectric breakdown strength 78 kV/mm than the 65 kV/mm for the commercial paper even setting a safety margin at failure probability of 1 %. Overall, the data suggests that the BN particles can provide an additional safety factor from the dielectric perspective.

In the hybrid system of insulation oil, BN and cellulose, typically the dielectric constant of insulation oil is the lowest, and the value of the cellulose is the highest. However, for dielectric strength, BN is usually the highest while insulation oil is the lowest. Under 60 Hz AC excitation, the E-field stress is higher in the oil regions due to its lower dielectric constant, which leads to the failure of insulation oil first as a result of its lowest dielectric strength. After the failure of insulation oil regions, the insulation oil can no longer hold the electrical stress and thus E-field redistribution is required along the remaining components of cellulose and BN. Since BN has a lower dielectric constant but higher dielectric strength than cellulose, BN particles can take more electric stress and hold the stress before the completed breakdown path can be identified along the insulation oil and cellulose regions. This is one of the major reasons that the BN additions can improve the dielectric strength of the hybrid BN paper system.

At 25 °C and under 60 Hz AC excitation and the E-field stress comparable to the operation condition (∼50 kV/mm), the dielectric loss of our oil-impregnated BN paper had the range of 1 % to 2.5 %. Based on the industry standard, the dry insulation paper should have a dielectric loss below 0.5 %. The measured dielectric loss of the proposed BN paper

Table 1
The scale factors and shape factors from the two-parameter Weibull distribution fitting of the experimental dielectric strength data of five different paper samples.

Paper type	Shape factor β	Scale factor α (kV/mm)	Dielectric strength at 1 % cumulative failure rate (kV/mm)
Control paper (no BN)	24.01	78.15	64.6
Commercial 125 gsm (no BN)	46.21	72.15	65.3
$CSF120_{LCMF}aBN_{25\mu m,50wt\%}$	23.91	94.66	78.1
$CSF120_{LCMF}aBN_{25\mu m,70wt\%}$	18.61	100.49	78.5
$CSF120_{LCMF}aBN_{25\mu m,100wt\%}$	26.05	100.98	84.6

was higher than the standard, perhaps due to the BN particles and the natural ester oil applied in this study. Since the dielectric loss usually is much smaller than winding and magnetic loss in power transformers, the dielectric loss increase for the proposed BN paper should be acceptable and should not significantly influence the entire loss of power transformers. However, this parameter and its possible influence on the overall loss of transformers need future study.

3.4. BN paper tensile strength

Transformer insulation paper must have sufficient tensile strength for wrapping the conductor and sustaining the electromagnetic force introduced by the transformer in both normal operations and fault conditions. Tensile strength is also correlated with thermal aging in insulation papers [50]. This is because paper made of long fibers with a greater cellulose degree of polymerization (DP) tend to have a greater tensile strength and simultaneously have a longer life under thermal-aging-induced cellulose depolymerization. In addition to fiber length, a dominant contributor to paper tensile strength is hydrogen bonding between adjacent cellulosic fibers [51–53]. The addition of BN is expected to disrupt this hydrogen bonding by decreasing the bonding area, leading to decreasing tensile strength. LCMF with greater surface areas than refined fibers were therefore added to substitute portion of fibers to increase hydrogen bonding in the paper composite to enhance the tensile strength [53].

Fourdrinier paper manufacturing is a continuous process using a moving wire, which results in an anisotropic fiber orientation distribution, i.e., in the machine direction (wire moving direction) [54]. Consequently, the tensile strength of commercial papers is orientation dependent. As an industry practice, commercial paper tensile strength is characterized by the machine direction (MD) and cross-machine direction (CMD). The difference in the MD and CMD direction tensile strength depends on the extent of alignment of the lignocellulosic fibers.

However, the laboratory batch filtration papermaking process does not introduce this anisotropy in the fiber orientation distribution. As a result, the tensile strength of a lab-made handsheet is orientation independent. To compare tensile strength of lab-made handsheets with commercial paper samples, the equivalent tensile strength of commercial papers based on the geometric mean [55] of the tensile strength in the MD and CMD as shown in Eq. (4), can be used.

$$EquavlentTensileIndexorStength = \sqrt{(TIorTSinMD) \times (TIorTSinCMD)} \tag{4}$$

Increasing BN loading decreased paper TI as shown in Fig. 7(d) due to the physical disruption (blocking) of hydrogen bonding by the BN particles. With the addition of LCMF, the hydrogen bonding sites in the composite matrix increased which improved the tensile strength of the resultant paper. At 100 wt% BN loading with ∼ 15 wt% LCMF substitution, the BN paper retained approximately 76 % of the TI of the control sample (without BN), which is still approximately 55 % greater than that of the commercial paper. The results provided confidence that adequate tensile strength can be maintained with the addition of the agglomerated BN and LCMF. Overall, the results shown in Fig. 7 suggest that it is feasible to add BN to paper to achieve the desired amount of increase in the paper through-plane thermal conductivity without adverse effects on the resultant BN paper dielectric and mechanical properties. This conclusion assumes that today's specification of tensile index for commercial insulation papers is sufficient for the immediate future. The data suggests that the change in tensile strength is not likely to be a concern in transitioning to novel BN insulation paper with greater through-plane thermal conductivity.

3.5. Thermal aging and life evaluation

Thermal aging test lasted nine weeks at 150 ℃, higher than the expected operation temperature of 100 ℃ [12]. The test setup and procedures were consistent with a widely used IEEE standard [46] as described earlier. Two types of paper samples were made of kraft softwood pulp fibers refined to CSF 440 and CSF 120, respectively. The other two types of paper contained 15 wt% LCMF as substitution of refined fibers to maintain the same total amount of lignocellulosic material. The tensile strength of paper samples was measured for fresh paper samples as well as samples aged for three, six, and nine weeks. The time-dependent tensile strength retention values of the four types of paper are shown in Fig. 8, and the results indicate that neither fiber refining nor the addition of LCMF affected the thermal degradation rate.

During the accelerated thermal aging test, the insulation oil inside the aging tank will contain some acid and moisture, which mimic the conditions inside the transformers after years of operation. As a result, cellulose depolymerization is unavoidable by acid hydrolysis. It is well known that paper tensile strength depends on two factors: fiber hydrogen bonding and fiber length [53]. Thus, thermal aging decreases paper tensile strength because cellulose depolymerization shortens fibers. On one hand, adding LCMF can increase hydrogen bonding due to the greater LCMF surface area (or available bonded area), on the other hand, LCMF has a lower cellulose DP than normal refined fibers which may lead to a higher thermal degradation rate. Quantitative prediction of the LCMF compensation on tensile strength and the possible negative effect of low LCMF DP on paper thermal degradation is difficult, because it depends on a lot of factors, such as the loading percentage of LCMF, the surface area or size of LCMF, the DP or fibril length of LCMF, and the type of insulation oil in use etc. Therefore, for the proposed BN paper, experimental exploration is selected to check the possible influence. Based on our test data obtained, a LCMF loading of 15 wt% did not have significant negative effects on paper thermal degradation under the test conditions.

For reference, the commercial insulation paper of 125 g/m^2 (gsm) from Weidmann (non-thermally upgraded kraft paper, density 1 g/cm^3) had approximately 68 % tensile strength retention after thermal aging test for 3 weeks under the same test conditions. The lab control paper (CSF 440 without LCMF and BN) had a comparable thermal degradation rate to the commercial insulation paper. As we reported earlier, adding BN increased BN paper through-plane thermal conductivity to 0.76 W/m·K. It is estimated that the hotspot temperature decreased by 9 °C when using the present BN paper to replace the commercial material based on model predictions [12]. Therefore, we expect that the lifetime of this novel BN insulation paper can be twice as long as that of the commercial paper. As a result, the novel BN paper can double the power transformer lifetime because the insulation paper failure is identified as the major cause of transformer failures.

4. Conclusions

This investigation designed and constructed a novel insulation paper by incorporating highly thermal conductive material, agglomerated BN particles, into kraft softwood fibers with the substitution of a portion (15 wt%) of wood fibers with microfibrils (LCMF). The LCMF was made from the same kraft softwood fibers but with greater surface area to compensate for the losses of hydrogen bonding due to the addition of BN particles. The addition of BN particles at 70 wt% of the kraft fibers increased the resultant BN paper through-plane thermal conductivity from 0.1 W/m·K to 0.76 W/m·K with oil-impregnation. The substitution of kraft fibers with LCMF was able to retain 80 % of the tensile strength of the control paper (without BN and LCMF). The novel BN paper with additives (BN and LCMF) also decreased the relative permittivity of the resultant BN paper compared to the commercial insulation paper or control paper without BN. The decrease is not significant but helpful, as a greater share of electrical stress is sustained by the novel BN paper that has adequate dielectric breakdown strength to accommodate such an increase in electrical stress. Overall, this implies that the present design of insulation paper fits the need of existing transformers. Thermal aging results indicated that highly refined fibers at CSF 120 and microfibrils (LCMF) did not significantly affect the thermal degradation rate of the resultant paper.

To summarize, the present novel BN enhanced insulation paper yielded three times better through-plane thermal conductivity, 23 % higher dielectric strength and approximately 35 % higher tensile strength than the commercial insulation paper with similar thermal degradation rates. This combination of factors enables lower temperature transformer operation, while minimizing the need for transformer design changes. Also, the proposed BN paper has approximately 33 % smaller relative permittivity than the commercial insulation paper, further reducing the E-field stress in the insulation oil ducts of the power transformers. The E-field stress reduction in power transformers is a desired feature, because insulation oil ducts are usually the weak insulation parts for this application and can introduce partial discharge events, which can significantly threaten the safe operation and also shorten the transformer life.

The proposed BN paper fabrication method is conventional and can fit the current commercial paper production line with slight modifications, which is actually very important for commercialization because

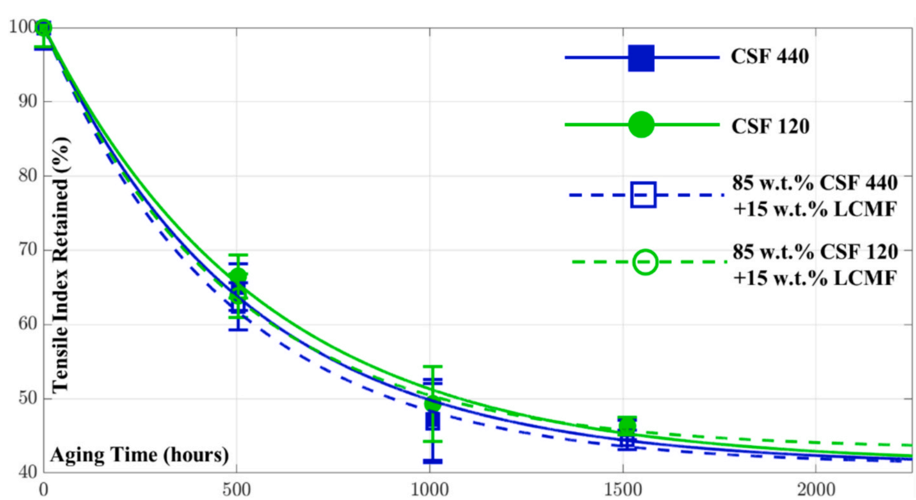

Fig. 8. Tensile index retained versus the thermal aging time for two paper samples containing 100% pure lignocellulose pulp and two paper samples containing 15 wt% of LCMF. The square and circle symbols correspond to the matrix CSF 440 and CSF 120 fibers, respectively. Note: 15 wt% LCMF is applied to substitute the same amount of CSF 440 or CSF 120 fibers, which helps keep the total amount of fibers the same among all the test samples.

developing any novel insulation paper has to rely on existing paper-making capital infrastructure and process due to the great cost (billions of US dollars) needed for capital to establish a completely new paper-making infrastructure. As a result, our proposed BN paper not only has overall desired features for power transformer applications as mentioned above, but also compatible with the state-of-the-art commercial paper production practice.

Based on all these advancements, the proposed BN insulation paper offers a promising pathway to double the life of power transformers.

CRediT authorship contribution statement

Yue Xu: Writing – review & editing, Writing – original draft, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Tejaswi Soori:** Writing – review & editing, Writing – original draft, Validation, Data curation. **Qian Zhang:** Writing – review & editing, Writing – original draft, Validation, Investigation, Formal analysis, Data curation. **J.Y. Zhu:** Writing – review & editing, Writing – original draft, Methodology, Investigation, Conceptualization. **J. Keith Nelson:** Writing – review & editing, Writing – original draft, Investigation, Conceptualization. **Liangbing Hu:** Writing – review & editing, Writing – original draft, Methodology, Investigation, Conceptualization. **Vaibhav Bahadur:** Writing – review & editing, Writing – original draft, Methodology, Conceptualization. **Robert Hebner:** Writing – review & editing, Writing – original draft, Supervision, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Robert Hebner reports financial support was provided by Advanced Research Projects Agency-Energy. Robert Hebner reports a relationship with Advanced Research Projects Agency-Energy that includes: funding grants. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability.

Data will be made available on request.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.matdes.2025.114112.

Data availability

Data will be made available on request.

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