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Enhancement of mechanical and electrical performances of insulating presspaper by introduction of nanocellulose

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ABSTRACT

To enhance the insulating performance of presspaper used in power transformers, unmodified nanofibrillated cellulose (UNFC), cationic nanofibrillated cellulose (CNFC), anionic nanofibrillated cellulose (ANFC) and cellulose nanocrystals (CNC) were investigated as the potential nano-additives. Concentration of the nano-additives was 10 wt%. Functional groups and crystallinities of the obtained samples were characterized by Fourier transform infrared spectroscopy (FTIR) and X-ray diffraction (XRD). Tensile strength, DC volume resistivity, dielectric response and breakdown behavior of the nanomodified and conventional presspaper were analyzed. Results show that nanofibrillated cellulose (NFC) can remarkably enhance tensile strength of insulating presspaper. Compared to ANFC, CNFC has a greater improvement which is mainly ascribed to the electrostatic attraction between the positively charged NFC and negatively charged softwood fiber. In addition to mechanical strength, introduction of nanocellulose can also obviously improve both AC and DC breakdown performances. The reason can be attributed to the reduction of partial discharges inside the composites due to the presence of nanocellulose in the original air voids. Moreover, and importantly, the negative effect of nanocellulose on DC volume resistivity and dielectric response can be neglected. On the basis of the overall performance, we conclude that NFCs are promising nano-additives for insulating presspaper, especially for CNFC.

1. Introduction

Insulating presspaper with a density higher than 0.9 g/cm^3 has been widely used in power transformers for more than 70 years [1]. As presspaper is a porous material having a lot of air voids that will sharply decrease the dielectric breakdown strength, insulating presspaper is commonly used in combination with mineral oil [2]. The main purpose of presspaper is to resist the flow of electric current between conductors. Besides that, it also need to provide some mechanical support. Although presspaper has relative good properties, insulation breakdown is still the major factor that causes the electrical failures of power transformers [3]. With the development of DC ultra-high voltage power transmission, especially the ±1100 kV transmission system under construction in China, reliability of oil-impregnated presspaper insulation becomes a big challenge [4]. Insulating presspaper with better performance is in great demand. However, compared to the intense investigations on the modifications of mineral oil [5–8], only fewer studies focus on the improvement of insulating presspaper [1,9].

Among the drawbacks of insulting presspaper, hygroscopicity is the first unavoidable disadvantage. Moisture in presspaper can not only decrease the short-term properties but also accelerate aging [10,11]. Graft polymerization, polymer deposition and alkylation can be used to reduce the hygroscopicity of insulating presspaper [1]. Oommen and Andrady pointed out that the grafted cellulose had the best performance [12]. For instance, conventional presspaper absorbed 6.5% moisture at 50% relative humidity and room temperature, whereas the grafted presspaper only picked up 2.5% moisture. The mismatch of relative permittivity between insulating presspaper and mineral oil is another shortcoming [13]. Higher relative permittivity of cellulose fiber can lead to the AC electric field distortion. By blending synthetic fiber with natural cellulose fiber, we can obtain insulating presspaper with a lower relative permittivity. Kamata developed a new kind of pressboard composed of polymethylpentene (PMP) fiber and cellulose fiber [14]. The permittivity of the composite was 3.5, which was lower than 4.7 for the ordinary pressboard. Unfortunately, there is little

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research on the improvement of the crucial properties of insulating presspaper, namely the tensile strength and dielectric breakdown behavior. Moreover, although chemical modification and introduction of synthetic fiber can improve certain properties of insulating presspaper, they are not environmentally-friendly and may deteriorate other properties. Therefore, new approaches that can achieve the overall enhancement or an improved trade-off are still in great need.

Nanotechnology is probably a promising solution. Applying nanotechnology to dielectric material results in a new term, nanodielectric, which means a well dispersed homogeneous blends composed of conventional dielectric material and filler particles with a few to a few tens of nanometers in size [15]. The theoretical paper published by Lewis in 1994 was regarded as the beginning of nanodielectric [15,16]. He further pointed out that interfaces were the most important feature for dielectrics at nanometric level [17,18]. The first experimental validation was accomplished by Nelson and his colleagues in 2002 [15]. They prepared the nanodielectric composed of epoxy resin and TiO_2 nanoparticles. Compared to epoxy modified by TiO_2 microparticles, Nelson et al. found that the nanometric fillers could mitigate the interfacial polarization and reduce the accumulation of space charges. That encouraging result made the research of nanodielectric become a hot topic [19]. Subsequent investigations reported the improvements of insulating properties of polymers (such as polyethylene, polypropylene, rubber) by adding SiO_2, Al_2O_3, MgO, ZnO nanoparticles, nanoclays or carbon nanotubes [20–22]. Ruijin Liao et al. investigated the electrical properties of insulating paper modified by nanometric hollow SiO_2 [23]. They found that AC breakdown field of the oil/nanomodified paper composite increased from 26.4 kV to 30.5 kV due to the low relative permittivity of hollow SiO_2 particles. Further studies suggested that the addition of nano-TiO2 [24] or nano-montmorillonite (nano-MMT) [25] to paper could also improve AC breakdown strength. Unfortunately, tensile strength of the modified paper decreased, and DC breakdown strength that was especially important for the converter transformers was not evaluated.

Nanocellulose including nanofibrillated cellulose (NFC) and cellulose nanocrystals (CNC) is a novel nanomaterial [26–29]. Compared to the inorganic nanoparticles (such as SiO_2, Al_2O_3, TiO2) mentioned above, nanocellulose has the following advantages. First, because nanocellulose is organic, surface modification is then not necessary [28]. Therefore, preparation of nanocellulose modified presspaper will be simpler. Second, nanocellulose is more compatible with cellulose insulation. For commercial insulating presspaper, no micro- or nanofillers are used during the manufacture process because the fillers may lead to some unexpected impact on the short-term or long-term insulation performance [24]. Considering that nanocellulose is prepared from natural cellulose fiber, it probably has a smaller effect and will be more easily accepted by the traditional power industry. Moreover, nanocellulose is environment-friendly and has a low cost [29]. Enhancement of mechanical strength of ordinary paper (not for electrical insulation) by adding NFC has been reported by many studies [30–34]. Houssine Sehaqui et al. studied the effect of NFC content on tensile strength of paper [30]. They found that the strength increased from 98 MPa to 160 MPa with an addition of 10 wt% NFC. Further research validated the reinforcement effect for high-density paper (>1 g/cm^3) [32]. As cellulose fiber has negative surface charges, cationic nanofibrillated cellulose (CNFC) and anionic nanofibrillated cellulose (ANFC) were explored by Sedat Ondaral et al. to enhance the dry strength of paper [35]. They reported that paper had a lower tensile index when only CNFC was added. Besides mechanical property, R. Hollertz et al. investigated the dielectric response of paper made of NFC [36]. They found that relative permittivity and dielectric loss of NFC paper are higher than those of kraft paper without NFC. The reason was largely attributed to the higher density of the NFC paper. On the basis of the available references, nanocellulose undoubtedly has a high possibility of improving the overall insulating performance of presspaper.

To understand the effect of morphology and surface charge property of nanocellulose and to determine a more suitable nano-additive, insulating presspaper modified by CNFC, ANFC, UNFC or CNC as well as the conventional presspaper without nanocellulose were prepared in this study. The content of the nano-additives was 10 wt%. Functional groups and crystallinities of the obtained samples were characterized by Fourier transform infrared spectroscopy (FTIR) and X-ray diffraction (XRD). To achieve a comprehensive understanding of the effect of nanocellulose, insulating performance including tensile strength, DC volume resistivity, dielectric response and breakdown behavior of prepared presspaper samples were measured and analyzed.

2. Experimental

2.1. Materials

The CNFC, UNFC, ANFC and CNC were provided by Tianjin University of Science and Technology (Tianjin, China). UNFC was obtained by a high pressure homogenizer and no chemical reagents were used. CNC was prepared based on an acid hydrolysis process. Surface charge densities of CNFC and ANFC were 0.69 meq/g and −1.5 meq/g, respectively. The unbleached sulphate softwood pulp for the manufacture of insulating presspaper was Prince George supplied by Canfor Corporation (British Columbia, Canada).

2.2. Preparation of conventional and nanomodified presspaper

Preparation of reference presspaper sample: First, the dry kraft pulp board was soaked in pure water (about 3 μS/cm) for 12 h. Then, the soaked pulp was dispersed and refined by a Valley beater to about 35 Schopper-Riegler freeness (SR). After that, the 0.5 wt% pulp suspension was transferred to a sheet former to make handsheet. Lastly, the wet handsheet was hot-pressed at 115 °C under 450 N/cm^2 for 10 min.

Preparation of presspaper modified by nanocellulose: The samples were prepared according to a procedure reported by Houssine Sehaqui. Prior to the sheet forming, 0.5 wt% nanocellulose (UNFC, CNFC, ANFC, or CNC) suspension was added to the 0.5 wt% wood fiber slurry to obtain a mixture containing 10 wt% nanocellulose. Then, the blend was stirred for 5 min. After that, the following sheet-forming and hot-pressing processes were the same as the preparation of the reference presspaper. Fig. 1 presents the detailed procedure for the preparation of unmodified and nanomodified presspaper handsheets.

The presspaper samples prepared in this study had a thickness of about 400 μm and a grammage in the range from 370 g/m^2 to 420 g/m^2. Compositions and apparent densities (measured according to IEC 60641-2) of the samples are shown in Table 1. Prior to the following experiments, the obtained presspaper handsheets were kept in sealed bags to prevent contamination.

2.3. FTIR and XRD

FTIR spectra of the reference and nanomodified presspaper were obtained by a Bruker vertex 70v vacuum spectrometer in reflection mode. Measurement range was from 4000 cm^{-1} to 600 cm^{-1}, and the resolution was 2 cm^{-1}. XRD measurements were performed by a Rigaku RINT 2000 wide angle goniometer operating