Nanocellulose and Polysiloxane Coatings for Strength Enhancement and Oil-proof and Hydrophobicity Improvement of Recycled Pulp Sheets

Kai Yi,[a] Shiyu Fu,[a,b,*] Zede Yi,[a] Xuedi Yang,[a] and Xingyu Lan [a]

Recycled fibers are essential raw materials used by the paper industry. However, the strength of the formed paper may fall off excessively after several cycles of reuse of the fibers. Herein, the authors measured the changes in physical properties and fiber size after multiple recycling of fibers and prepared a handsheet from fiber recycled different times. Then the handsheets made with fibers that had been recycled 5 times were double-coated with nanocellulose derivatives to obtain oleophobic and hydrophobic paper. The first layer applied to the paper was cellulose nanofibril (CNF), and the second coating contained polydimethylsiloxane (PDMS) and CNF (CNFmp). The physical properties and barrier performance of coated paper were greatly improved compared to recycling paper. The water contact angle of the coated paper was as high as 139.8° and the $Cobb_{60}$ value was 35.18±2.15 g/m^2. The oil contact angle was 97.1°, and the oil kit number was 12/12.

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Keywords: Recycled fiber; Oleophobic paper; Hydrophobic paper; Polydimethylsiloxane

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INTRODUCTION

Currently, petroleum-based plastic packaging materials are widely used in daily life and industry due to their low cost, simple preparation process, and good mechanical strength (Wang *et al.* 2022). However, the large quantity use of these non-biodegradable products can cause a series of issues, such as environmental pollution, energy crisis, subsequent disposal difficulties, *etc.* (Hossain *et al.* 2021). Therefore, many researchers are trying to explore new biodegradable materials to replace plastics and have made great progress.

Paper has attracted a lot of attention as one of the most used packaging materials, because of its wide range of sources, reusability, and biodegradability (Chen *et al.* 2022). However, a rapid decrease in strength when it contacts water or moisture, and the poor oil-resistant properties limit its application in many fields (Majeed *et al.* 2013). What is worse, the paper's mechanical properties are also greatly reduced after multiple recycles due to the decrease in recycled fiber performance, making it difficult to use the paper without modification. Various approaches have been adopted to enhance the mechanical properties, as well as the water and oil resistance of paper, such as spraying the coating on the paper surface (Lyytikainen *et al.* 2021; Zhou *et al.* 2022), paper sizing (Dang *et al.* 2017), and other surface modification methods.

The decrease in the fiber performance of recycled fibers during reuse is mainly due to fiber hornification (Wang *et al.* 2018). The fibers suffer from hornification after the first drying, and as the number of reuses increases, the degree of fiber hornification is more severe. During the reuse of paper, fiber hornification and the loss of fine fibers can lead to a significant decrease in the physical properties of the paper. Therefore, it is necessary to modify the substandard paper in mechanical performance to meet the requirements of use. It is meaningful to enhance the paper's mechanical properties to give it certain characteristics for use in specific applications. For example, it is promising to modify the paper with oil-proof and hydrophobic properties to make it suitable for packaging applications (Song *et al.* 2020).

As a nanofiber derived from natural plant fiber, cellulose nanofibril (CNF), which traditionally has been called nanofibrillated cellulose, has attracted much attention in many fields because of its robust strength and excellent barrier properties (Tayeb *et al.* 2020). It has a high specific surface area and effective hydrogen bonding between nanofibrils, which is beneficial for paper modification to improve its strength and barrier to air and grease. However, the abundance of hydrogen bonds between fibers also poses a problem of poor hydrophobicity (Zhang *et al.* 2014). Thus, it is feasible to make the material hydrophobized to enable better application.

Polydimethylsiloxane (PDMS) is widely used for the modification of hydrophobic materials due to its inherently good biocompatibility and high hydrophobicity (Jankauskaite *et al.* 2020). Moreover, strong adhesion to a variety of surfaces and inertness to foodstuffs and living tissues make it possible in healthcare applications as well (Phan *et al.* 2014).

In addition to hydrophobic substances, the incorporation of micro/nanoparticles is also helpful in constructing superhydrophobic surfaces. Generally, the preparation of a superhydrophobic surface requires low surface energy material modification and high surface roughness on a very fine scale (Yu *et al.* 2019). Some common particles, such as SiO_2 nanoparticles, ZnO nanoparticles, and nanocellulose microparticles, are often used to spray on the sample surface to increase the roughness and thus obtain superhydrophobic materials. These materials have a wide range of applications in anti-corrosion (Vengatesh and Kulandainathan 2015), anti-icing (Liu *et al.* 2017), self-cleaning, and oil-water separation (Velayi and Norouzbeigi 2018), *etc.*

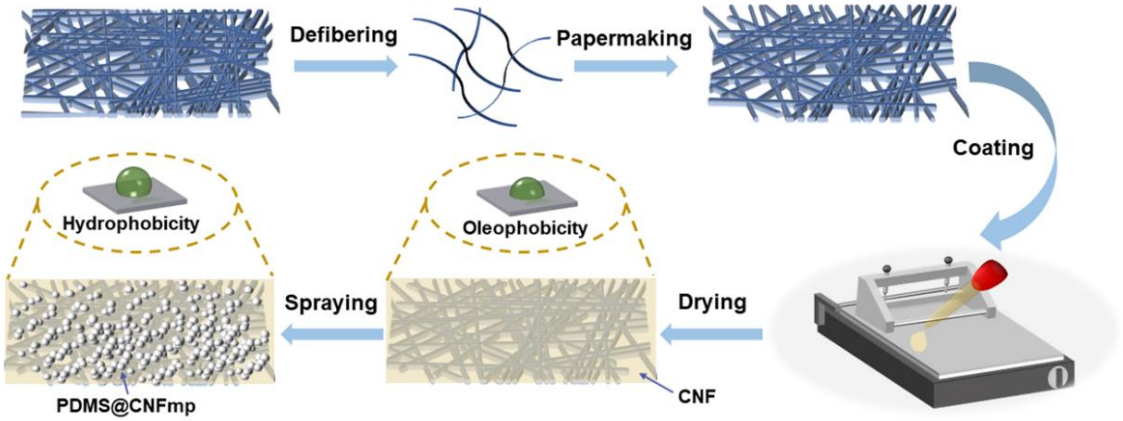

Fig. 1. The preparation process of oil-proof and hydrophobic paper

In this study, the authors investigated the change tendency in mechanical properties of paper reused five times and characteristics of the paper after oil-proof and hydrophobic modification of the reused paper. The preparation process of oil-proof and hydrophobic paper is shown in Fig. 1. More specifically, the performance of paper during the reused process was characterized by bursting strength, tearing strength, tensile strength, zero span tensile strength, and air permeability. The microstructures of uncoated paper and coated paper were analyzed by scanning electron microscopy (SEM). For the oil-proof and hydrophobic modification paper, in addition to mechanical properties characterization, water contact angle, oil contact angle, and oil kit number were also tested. This study provides ideas for the application of recycled fibers in the packaging field.

EXPERIMENTAL

Materials

Bleached pulp was prepared in the lab. Ethyl alcohol (C_2H_5OH, AR, 99%) and sodium bromide (NaBr, AR, 99%) were purchased from Guangzhou Chemical Reagent Factory (Guangzhou, China). Sodium hypochlorite (NaClO, AR, 99%, 7.5 M) and 2, 2, 6, 6-tetramethyl-piperidine-N-oxyl (TEMPO, purity C 99.9%) were obtained from MACKLIN Inc. (Shanghai, China). Sodium hydroxide (NaOH, AR) was obtained from Fuchen Chemical Reagent Co., Ltd. (Tianjin, China). Hydrochloric acid (AR, 36% to approximately 38%) was purchased from Guangzhou Chemical Reagent Factory. The PDMS (Dow Corning Sylgard 184, 1.04 g/mL) and curing agent were supplied by Dow Corning (Dongguan, China). N-heptane (C_7H_{16}, AR, 99%) was supplied by Shanghai Lingfeng Chemical Reagent Co., Ltd. (Shanghai, China). Castor oil was provided by Aladdin Chemical (Shanghai, China). Ethyl acetate (AR, 99%) was supplied by Tianjin Fuyu Fine Chemicals Co., Ltd. (Tianjin, China). In addition, all chemicals were used as received without further purification.

Methods

Preparation of cellulosic pulps

The bleached pulp (200 g dried) was soaked in water (5 L) at room temperature for one night. Next, the soaked pulp board was torn into small pieces. Before loading, 7.7 L water was added into a pulping tank of the Valley beater (a laboratory Hollander-type beater). Then, the beater was started, and the pulp was added into the pulping tank slowly. When the final cellulosic pulps with a beating degree of 38° SR were achieved, the pulps were collected and thickened, and then measured the moisture and saved.

Preparation of CNF

Firstly, TEMPO (0.015 g/g of dry pulp) and sodium bromide (0.1 g/g of dry pulp) was added to the pulp slurry under stirring. Secondly, a certain amount of NaClO (6 mmol effective chlorine per gram of dry pulp) was poured into the mixed slurry and pH was controlled between 9.8 to 10.0 by titrating 0.5 mol/L NaOH solution. Next, the obtained crude product was washed several times by centrifugation at 5500 rpm with distilled water and homogenized 3 to 5 times to obtain the desired CNF and the carboxyl content was 230.4 mmol/kg. Homogenization serves to shorten the length of the fibers after several high stress mechanical shears, eventually reaching the nanoscale. The equipment was a high pressure microfluidization homogenizer purchased from Noozle Fluid Technology

(Shanghai) Co., LTD and the intensity of each pass of treatment was 15,000 PSI. Finally, the concentration of CNF suspension was measured and stored in a refrigerator at 4 °C.

Fabrication of the multifunctional paper

The multifunctional paper was composed of the base layer (original paper) and the mixture layer with PDMS and CNFmp. Firstly, the original paper was prepared with bleached eucalyptus pulp and the paper grammage was 60 g/m^2. The wet paper was dried at 90 °C under negative pressure of -90 kPa for 10 min. Secondly, the prepared paper was broken into fibers and fabricated paper again, with the same grammage. The operation was repeated five times. A portion of each manufactured paper was set aside for performance testing. Subsequently, a layer of CNF suspension with a thickness of 1.5 mm was coated on the surface of the paper prepared five times repeatedly. The squeegee of the coating machine was adjusted to a position 1.5 mm away from the paper surface. Then the coating temperature was set to room temperature and the coating rate to 3 mm/s, and the CNF suspension was added uniformly with a dropper at 16.3 g/m^2 in front of the squeegee. In the end, the sample was dried in an oven at 105 ℃ after coating to get CNF-coated paper. On the one hand, the abundant hydroxyl groups on CNF provided oleophobicity to the sample. On the other hand, the CNF can greatly enhance the mechanical properties of the paper. Finally, a second coating including PDMS and CNFmp was sprayed on the coated paper and cured. The thickness of the second coating was by controlling the volume of PDMS@CNFmp dispersion to be sprayed uniformly over the same area with 0.12 mL/cm^2 of spray solution. The hydrophobicity of the PDMS and the micro/nano structure of the CNFmp endowed excellent hydrophobicity to the paper.

Fiber morphology analysis

Morphologies of fibers with different reuse times were assessed with a FS300 (Metso, Finland) fiber analyzer. More specifically, 30 mg eucalyptus fiber was dispersed in water and a dispersion prepared with a concentration of 30 mg/L. The fiber analyzer was used to characterize the length, width, fiber coarseness, and proportion of fine fibers.

Chemical characterization

Sample composition was analyzed by Fourier transform infrared spectroscopy (FTIR) using a Nicolet iN10 Continuum instrument (Thermo Fisher Scientific, Waltham, MA, USA) with the wavenumber from 4000 to 500 cm^{-1}. X-ray photoelectron spectroscopy (XPS) was conducted to determine the chemical composition of the uncoated and coated paper using a Thermo Fisher instrument (Thermo Scientific K-Alpha). The surface features and morphology of the samples were observed by scanning electron microscopy (SU5000, Hitachi, Tokyo, Japan) with an acceleration voltage of 5 kV. Meanwhile, the elemental distributions and compositions of the coated paper were determined using the X-ray spectroscopy (EDX) mapping with an acceleration voltage of 15 kV.

Physical property analysis

Before the tests, the samples were placed under an environment of 23 ± 1 ℃ and 50 ± 2% relative humidity (RH) for 48 h. For the tensile strength test, the paper samples were cut into long strips of 15 × 150 mm^2 size and were measured using an L&W CE062 (L&W, Stockholm, Sweden) strength tester. Air permeability was measured using an L&W 166 air permeance tester (Lorentzen & Wettre, Stockholm, Sweden) according to Chinese national standard GB/T 458 (2008) and the effective test area of the sample was 50 cm^2. Bursting strength was investigated using an L&W 180CE bursting strength tester (L&W, Stockholm, Sweden) according to GB/T 454 (2020). Tearing strength was tested using an L&W 009 tearing tester (L&W, Stockholm, Sweden) according to GB/T 455 (2002), and the measured sample size was 50 × 60 mm^2. Zero span tensile strength was determined using a z-span 2400 instrument (Fabco-Air, Inc., Cleveland, OH, USA) according to GB/T 26460 (2011) and the sample size was 65 × 180 mm^2. More than five specimens were tested, and the average value was recorded.

Micromorphology and roughness analysis

The micromorphology of original paper, recycled five times paper, CNFmp, coated CNF, and CNFmp@PDMS paper was observed by using a scanning electron microscope (SEM; SU5000 Japan) with an acceleration voltage of 3 kV during operation. The EDS mapping of coated CNF and CNFmp@PDMS paper was characterized with an acceleration voltage of 15 kV. The roughness of samples was measured by a L&W CE165 surface roughness tester (L&W, Stockholm, Sweden) with 1 MPa collet pressure and measured more than 5 groups of data to take the average.

Oil resistance analysis

The oil kit number of the paper was determined according to the standard TAPPI T559 cm-12 (2002). Twelve mixtures of n-heptane, toluene, and castor oil with different surface tensions were prepared in different volume ratios. Droplets with different permeability were dropped on the surface of the sample to be tested at a height of 25.4 mm (1 inch) and allowed to be absorbed for 15 s (Long *et al.* 2015). The droplet on the paper's surface was wiped with a clean tissue without wetting the sample, the oil kit number of the sample depended on the surface tension of the droplets. Especially, the test was conducted from the high oil kit rating to the low in turn.

Hydrophobicity and contact angle (CA) analysis

To confirm the hydrophobicity of the modified paper, several different liquids including water, juice, cola, 0.1M HCl, 0.1M NaCl, and 0.1M NaOH were dropped on the paper surface. $Cobb_{60}$ value (g/m^2) was also used to evaluate the water resistance of paper before and after coating. In detail, water absorption of paper was measured by using the Cobb tester (LB-K100, Shenzhen, China) with a test area of 100 cm^2 according to GB/T10739. Besides, a contact angle measurement instrument (Model: ZJ-7000, (Shenzhen Zhijia Instrument Equipment Co., Ltd., Shenzhen, China) was conducted to determine the oil contact angle (OCA) and water contact angle (WCA) of the samples. Especially, castor oil was used to analyze the oleophobicity and water was used to characterize the hydrophobicity. Approximately 5 μL of liquid was dropped on the samples' surface to measure the contact angle.

RESULTS AND DISCUSSION

Effect of Cycle Times on Fiber Size

To investigate the fiber changes after papermaking several times, the fiber was analyzed in dimensions. The results of fiber weighted length, width, coarseness, and weighted ratio of fine fiber length are shown in Fig. 2. It could be seen in Fig. 2a that the fiber weighted length of the original paper could reach 1.285 mm. However, the fiber weighted length of the paper was only 1.199 mm after five times of recycling. As for fiber width, it decreased from 30.7 μm to 27.7 μm as the number of recycled times increased. The fiber coarseness decreased from 0.1979 mg/m to 0.1237 mg/m and the weighted ratio of fine fiber length decreased from 59.82% to 39%. This may be due to the decrease in fiber length and coarseness as the number of fibers recovered increases and the fibers were continuously cut during the dredging process. During the paper forming process, small fibers were lost during water filtration, so the weighted percentage of fine fiber length decreases.

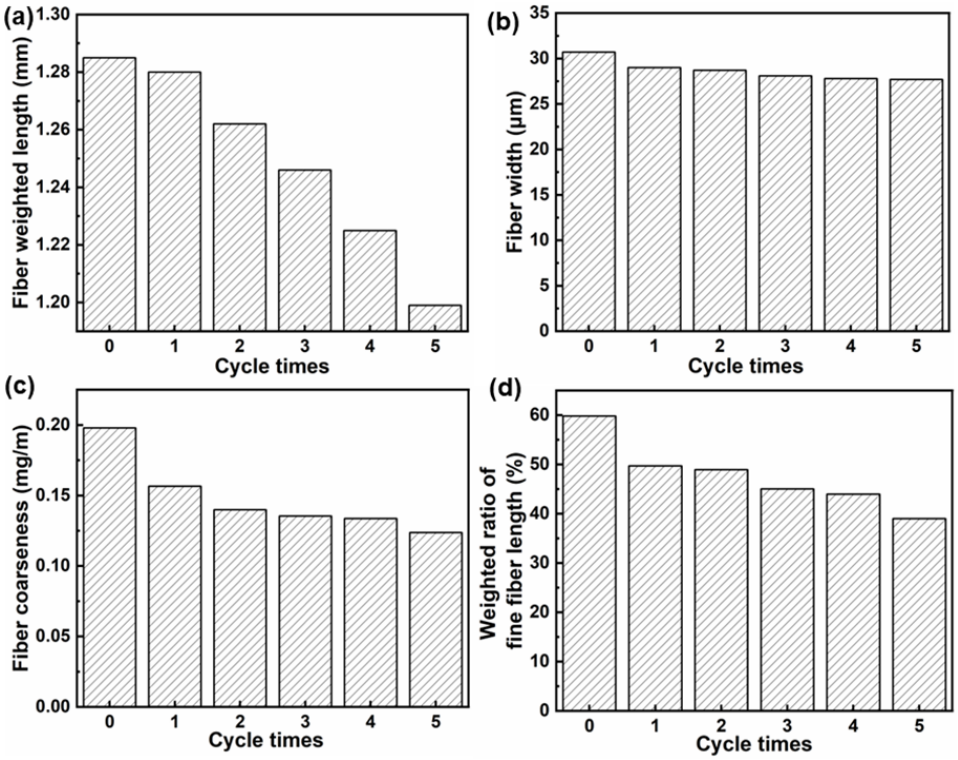

Fig. 2. Fiber analysis of raw pulp and pulp with different repetitions of papermaking, fiber weighted length (a), fiber width (b), fiber coarseness (c), weighted ratio of fine fiber length (d)

Chemical Analysis of Coated and Uncoated Paper

Both FTIR and XPS were used to analyze differences in structural properties and elemental composition of the uncoated paper and coated paper. As shown in Fig. 3a, new peaks appeared in the curve representing the coated paper. The peak at 1257 cm^{-1} corresponds to the -Si-CH_3 stretching vibration and the peak at 796 cm^{-1} in the spectrum belongs to the Si-O-Si bending vibration (Li and Rabnawaz 2019). These peaks demonstrate the presence of PDMS on the paper surface. Moreover, in the XPS spectrum, the curve representing the coated paper also showed two new peaks. One peak was at 153.4

eV (Si 2s) and another was at 102.3 eV (Si 2p), which further showed that PDMS successfully stayed on the coated paper (Wang *et al.* 2021).

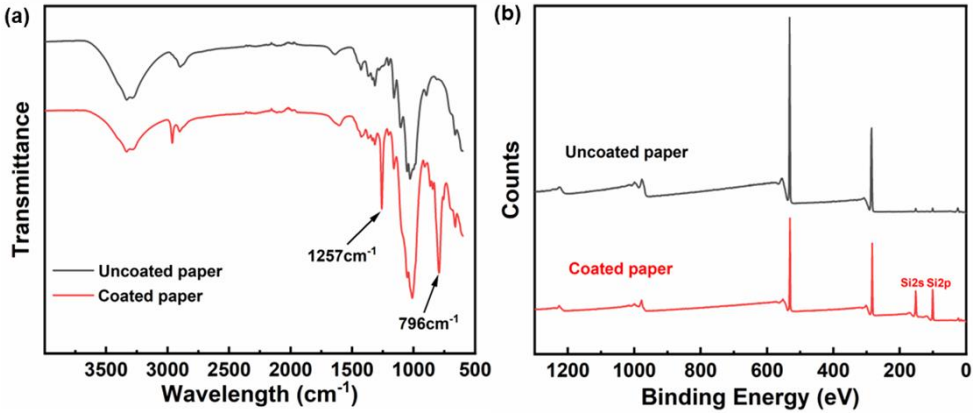

Fig. 3. FTIR and XPS of coated and uncoated paper, FTIR (a), and XPS (b)

Physical Properties of Coated and Uncoated Paper

The mechanical properties of paper are critical to the daily use of packaging materials. The excellent physical strength of the packaging material allows it to withstand greater external pressure and extend its service life (Xie *et al.* 2021). Therefore, the physical performances of the original paper, recycled paper, and coated paper were investigated. The tensile strength of coated and uncoated paper is shown in Fig. 4, the tensile strength of the paper decreased remarkably as the number of recycled times increased. The tensile strength of the original paper was 2.456 kN/m, and it decreased to the minimum of 1.058 kN/m. This is due to the large loss of fine fibers and the macrofibers were broken during the papermaking process, so the inter-fiber bonding force was greatly reduced. Fortunately, the mechanical strength of coated paper was greatly improved and exceeded the original paper, reaching a surprising tensile strength of 2.564 kN/m. Clearly, the physical strength of the paper was greatly enhanced by the strong bonding of hydrogen bonds and the filling of the pore structure between the fibers after the coating of nanocellulose. At the same time, the zero-span tensile strength had the same trend, decreasing from 95.76 N/cm to 89.33 N/cm for uncoated paper, but increasing 12.5% to 107.75 N/cm for coated paper compared to the original paper (Fig. 5). After the paper was coated with CNF, the bond between the fibers became stronger, resulting in increased fiber strength and therefore greater zero-span tensile strength.

In addition to tensile strength, breaking strength and tearing strength are also commonly used to evaluate the mechanical properties of paper. As shown in Fig. 6, the bursting strength of the original paper reached 142 KPa, after 5 times of recycling. The bursting strength of the paper was only 72.8 KPa, which was only about half of the original paper. Although the performance of the recycled paper was greatly reduced, the paper was modified by coating with CNF and CNFmp@PDMS to achieve a bursting strength of 120.8 KPa. The breaking strength of the paper after coating was 66% higher than that before coating. However, due to the repeated cutting and drying of the fiber, the great loss of hemicellulose decreases the contribution of hemicellulose to bonding. Besides, a large loss of fine fibers makes the pores between the fibers larger, the breaking strength of the paper after coating fails to reach the strength of the base paper. As for tearing strength, the value was reduced from 538 mN to 363 mN for the uncoated paper. This corresponds to a

reduction by up to 32.5%. The tearing strength of coated paper was as high as 869.6 mN, which was 1.6 times of original paper (Fig. 7). After the paper was coated with nanocellulose, the pore structure between the fibers was filled, and the hydrogen bonding between the fibers was more firm, so it can effectively improve the tearing strength of the paper.

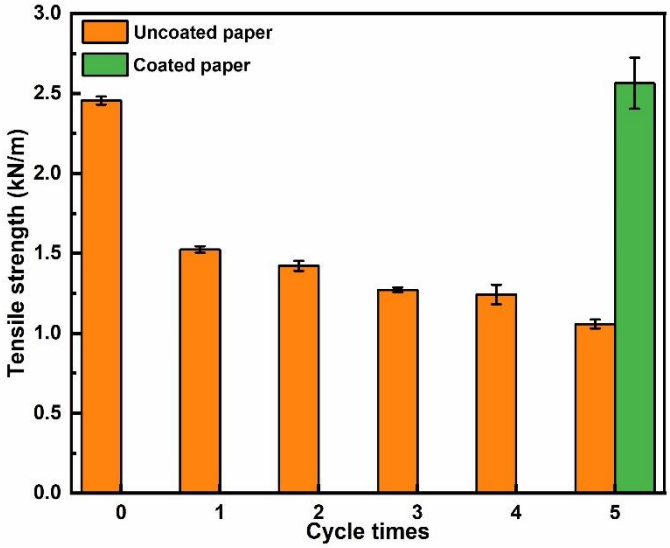

Fig. 4. Tensile strength of uncoated paper and coated paper

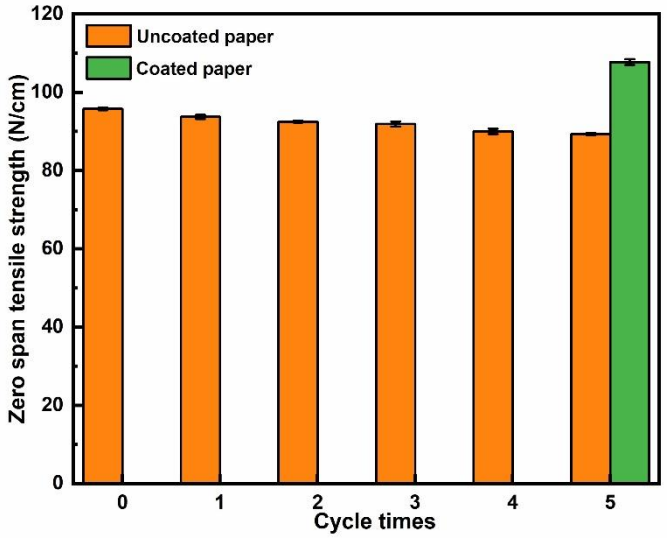

Fig. 5. Zero span tensile strength of uncoated paper and coated paper

The barrier properties of paper also play an important role in the actual use of packaging. Paper is a porous material, so it does not have good barrier properties. Thus, water, oil, gas, and other liquids can easily permeate and diffuse into the matrix (Jing *et al*. 2021). As shown in Fig. 8, the air permeability of the original paper was 773.6 mL/min. The loss of fine fibers made the air permeability of the paper increase greatly, reaching 7446 mL/min, which is almost 10 times of the original paper.

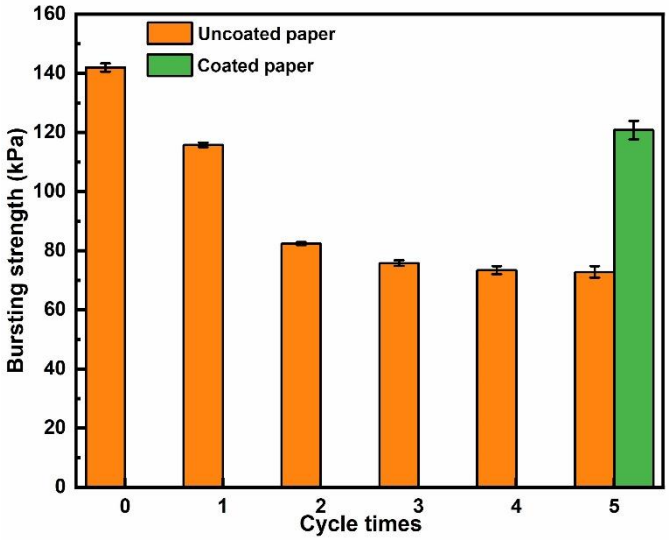

Fig. 6. Bursting strength of uncoated paper and coated paper

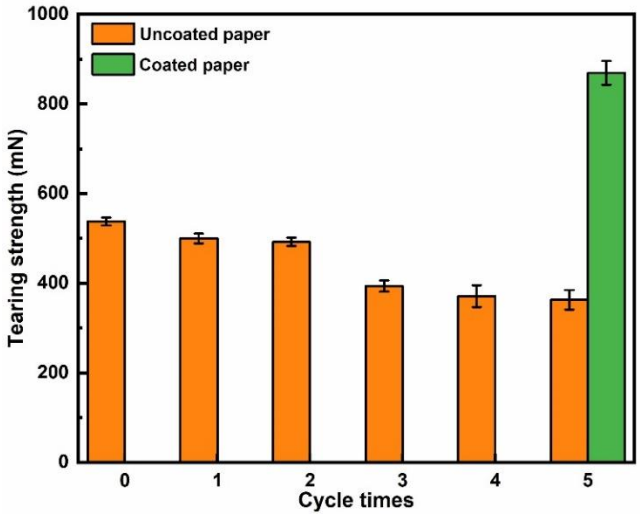

Fig. 7. Tearing strength of uncoated paper and coated paper

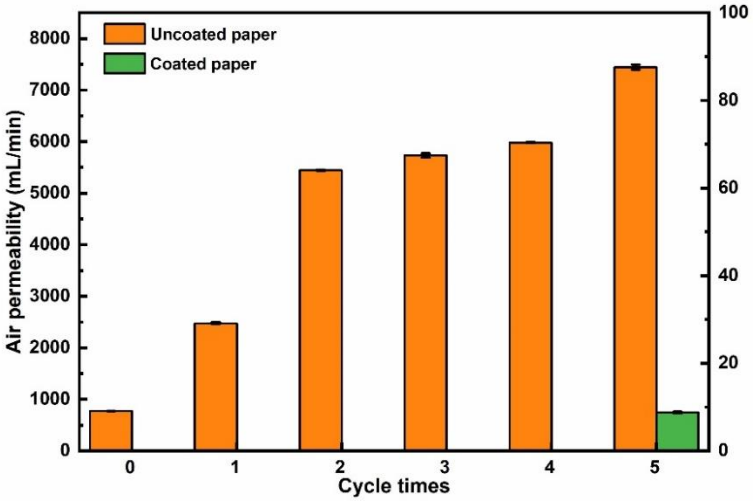

Fig. 8. Air permeability of uncoated paper and coated paper

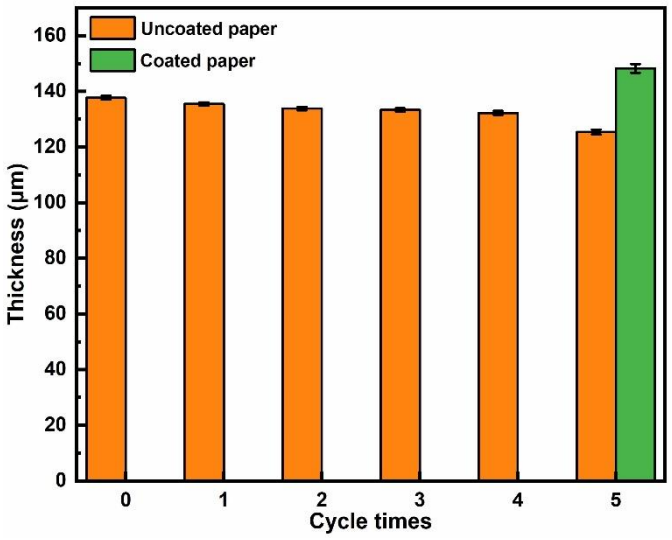

Fig. 9. Thickness of uncoated paper and coated paper

The gas barrier performance of coated paper was greatly improved because nanocellulose can effectively fill the pores between the fibers and make the paper smooth (Aulin *et al*. 2013). Therefore, the coated paper's air permeability was only 8.784 mL/min. The thickness of uncoated paper and coated paper is shown in Fig. 9, the thickness of the original paper was 137.75 μm, and it reduced to 125.42 μm after recycling 5 times. After coating, the thickness of the paper increased to 148.26 μm, indicating that the total thickness of the double coatings was 22.84 μm.

Morphology Observation

To confirm the difference in fibers and observe the morphology of CNFmp and coated paper, SEM characterization was performed. As can be seen in Fig. 10a-b, the paper surface was distributed with many fine fibers, covering the surface of the coarse fibers, which can fill the pores between the fibers. However, many fine fibers were lost, and thick fibers were exposed according to the paper surface morphology after repeated papermaking 5 times. In the process of water filtration, fine fibers are easily lost through the copper mesh, and only a large number of macrofibers and a little of fine fibers are retained. Therefore, the pores between the fibers greatly increased (Fig. 10c-d).

The micromorphology of CNFmp (Fig. 11a-b) shows many grooves and rough structures, and the size of CNFmp was only several microns, which is good for spraying and dispersing on the paper surface. It is obvious that the fibers of the paper were covered with microparticles after spraying (Fig. 11c), and the roughness of the paper was greatly increased. Surface roughness test results (Table 1) showed significant changes in paper before and after coating, the surface roughness of uncoated paper was 4.40 μm, the CNF-coated paper was 3.52 μm and 5.01 μm for double-coated paper. In addition, the SEM magnification (Fig. 11d) shows that the microparticles were wrapped with a thin film, which was the protective layer of PDMS after curing and effectively improved the hydrophobicity of the sample. To determine the elemental composition of the coated paper surface, the EDS was measured, and the results are listed in Fig. 12. The atomic content of C, O, and Si was 51.34%, 43.52%, and 5.14%, respectively.

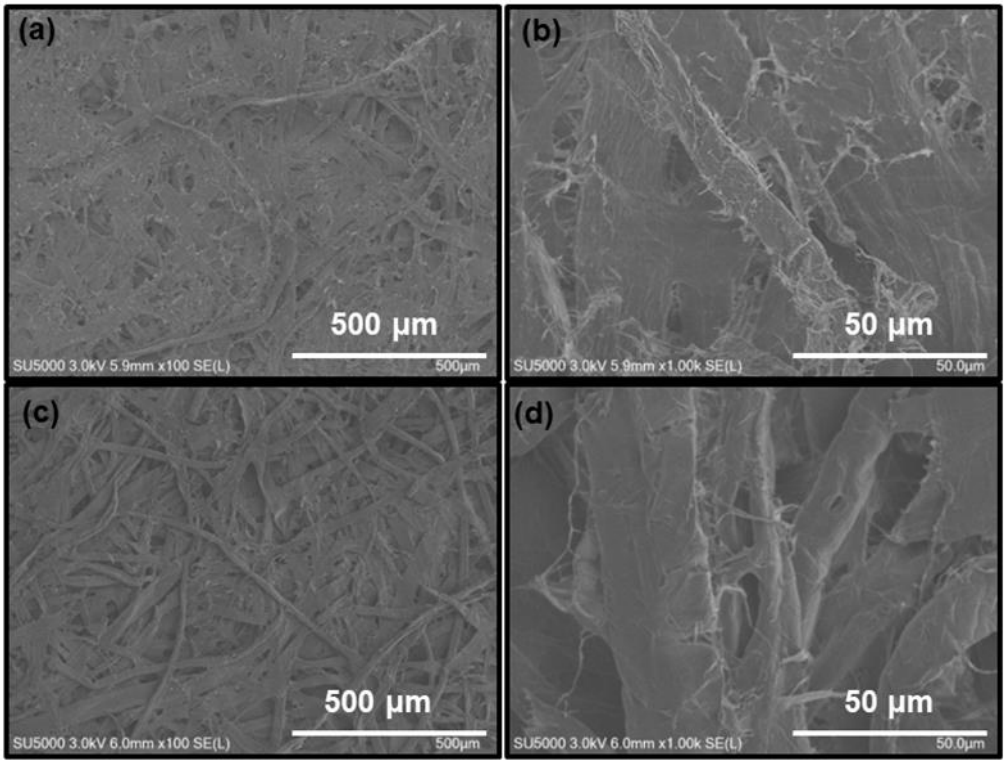

Fig. 10. SEM of original paper and repeated five times paper: original paper (a), enlarge of a (b), repeat five times paper (c), and enlarge of c (d)

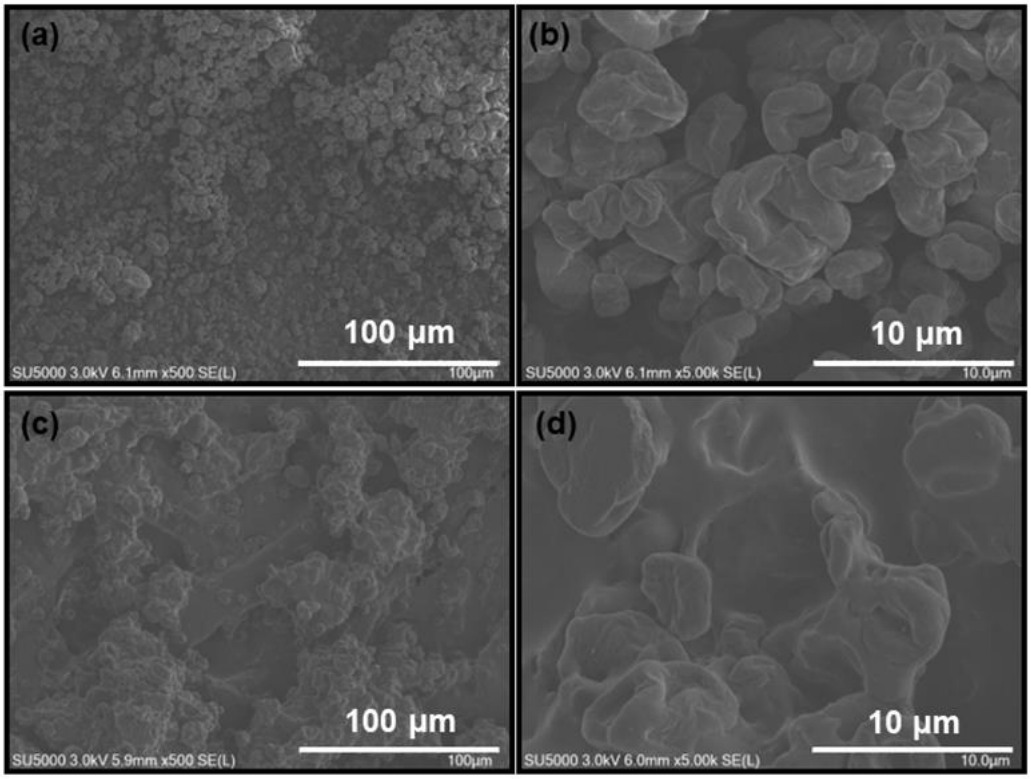

Fig. 11. SEM of CNFmp and coated paper: CNFmp (a), enlarge of a (b), coated paper (c), and enlarge of c (d)

Table 1. Surface Roughness of Three Paper Specimens

Samples	Uncoated paper	CNF-coated paper	Double-coated paper
Surface roughness (μm)	4.40	3.52	5.01

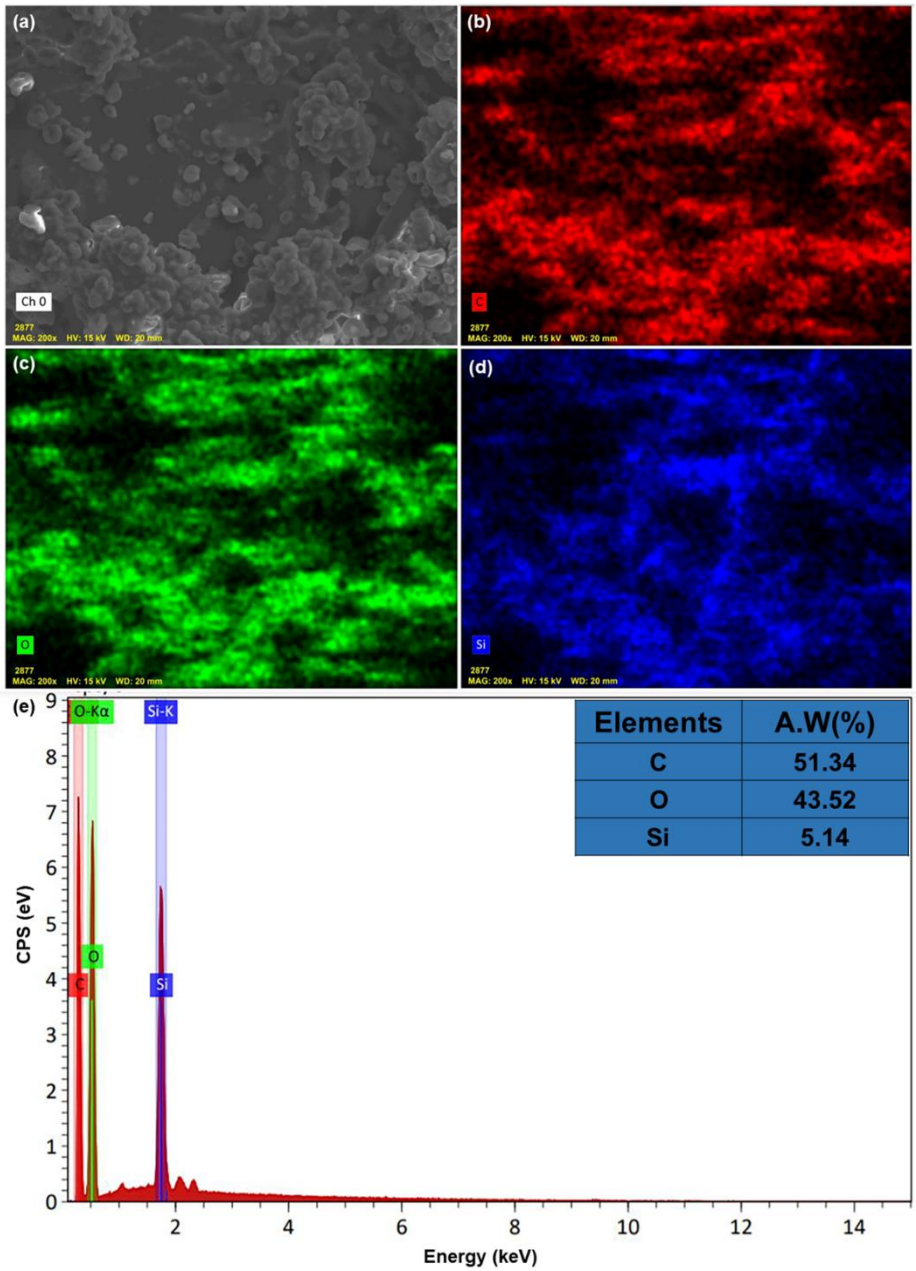

Fig. 12. Mapping of coated paper: surface morphology (a), C element (b), O element (c), Si element (d), and atoms mass (e)

Oil-proof and Hydrophobicity Analysis

Figure 13 shows the oil resistance and hydrophobic properties of uncoated and coated paper. For uncoated paper, the liquid with oil-proof grade 6 easily penetrated the sample and left visible wetting marks on the backside, indicating that the original paper was not oleophobic (Fig. 13a-b). However, the coated paper was able to resist the invasion of the liquid with oil-proof grade 12, indicating that the oil-proof performance of the

modified paper was greatly improved, and the oil-proof grade was up to 12/12 (Fig. 13c-d). This is because the surface of the CNF-coated paper became flat and smooth, and the pores between the fibers were filled. Moreover, the large number of hydroxyl groups on the surface of the nanocellulose helped to provide hydrophilicity, which enabled the oleophobic effect to be achieved (Sheng *et al.* 2021).

The resistance of coated paper to various liquids is shown in Fig. 13e. As can be seen in the figure, the juice, water, cola, and other liquids maintained their spherical shape and did not wet the paper, indicating that the modified material had good hydrophobic properties. Besides, the results of the water adsorption test showed that the double-coated paper had a waterproof effect, and its $Cobb_{60}$ value was 35.18±2.15 g/m^2, which was much lower than the uncoated paper. The reason for the relatively high Cobb value of the double-coated paper may be the rough structure of the surface and the hydrophilic nature of nanocellulose used in mixture with PDMS. The WCA and OCA test results (Fig. 13f) show that the WCA of the sample was as high as 139.8° and the OCA was 97.1°, further verifying the oil resistance and excellent hydrophobicity of the product. The good hydrophobicity of the paper was due to the low surface energy of PDMS, and the micro/nano rough structure provided by CNFmp, which together facilitated the hydrophobic modification of the material. Therefore, the coated paper has promising applications in packaging materials.

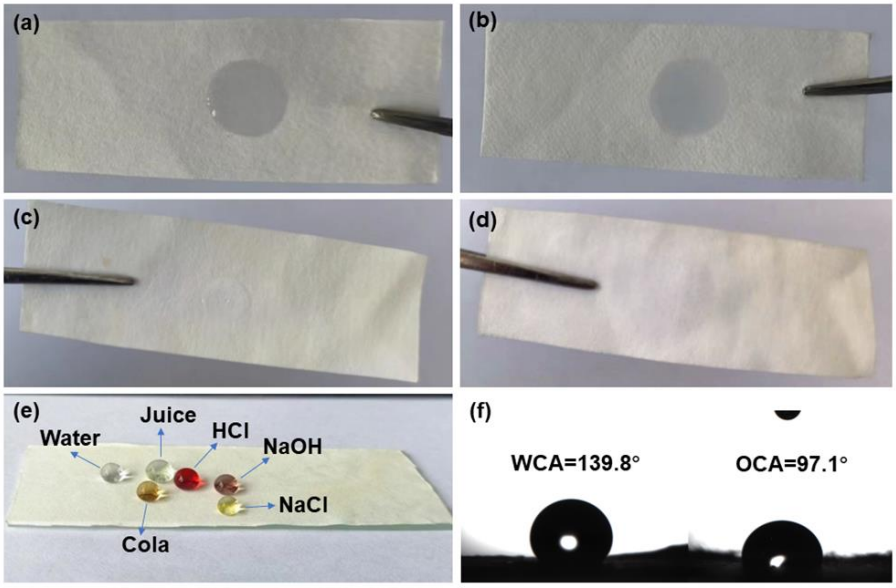

Fig. 13. Oil resistance and hydrophobicity of uncoated paper and coated paper: uncoated paper drops oil proof grade 6 liquid front view (a), uncoated paper drops oil proof grade 6 liquid back view (b), coated paper drops oil proof grade 12 liquid front view (c), coated paper drops oil proof grade 12 liquid back view (d), different liquids dropped on coated paper (e), WCA and OCA of coated paper (f)

Table 2. $Cobb_{60}$ Value of Three Paper Specimens

Samples	Uncoated paper	CNF-coated paper	Double-coated paper
$Cobb_{60}$ value (g/m^2)	81.17±2.75	98.97±5.63	35.18±2.15

CONCLUSIONS

1. After being recycled five times, the mechanical properties of the paper were greatly reduced. Tensile strength decreased from 2.46 kN/m for the original paper to 1.06 kN/m, zero span tensile strength from 95.8 N/cm to 89.3 N/cm, breaking strength dropped from 142 KPa to 72.8 KPa, tearing strength from 538 mN to 363 mN, and air permeability increased remarkably from 774 mL/min to 7450 mL/ min.
2. The physical properties of the paper recycled 5 times were much improved after coating with CNF and spraying PDMS@CNFmp. Tensile strength was up to 2.56 kN/m, zero span tensile strength was improved to 107.8 N/cm, bursting strength was increased to 121 KPa, tearing strength increased to 870 mN, and air permeability was reduced to 8.78 mL/min.
3. Coated paper exhibited excellent hydrophobicity and oil repellency. The measured water contact angle was up to 140°, the $Cobb_{60}$ value was 35.18±2.15 g/m^2 and the paper was able to resist various liquid droplets. The oil contact angle was 97.1°, and the oil kit number was 12/12.
4. This work elucidates the changes in mechanical properties and fibers of paper during recycling and provides new strategies for recycled fiber utilization, such as the production of paper-based packaging materials that were both oil-resistant and hydrophobic.

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