

Improved Oil Resistance of Cellulose Packaging Paper by Coating with Natural Polymer Derived Materials

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Abstract: Paper is widely used as food packaging due to its good mechanical strength and degradability. However, it has a relatively strong affinity for water and oil, which limits its application scope. In this work, we prepare two types of coated paper to investigate, the influence the air permeability and polarity on the oil resistance of the coated paper. The results showed that reducing the air permeability improved the grease resistance of the coated paper. High surface energy coatings also showed better oil resistance because of their higher content of polar components that resulted in a higher resistance to grease. The mechanical properties of the paper also improved after applying the coating. These natural derived materials offer an alternative to the fluoride-containing materials currently used in the market to improve the wettability of paper.

Keywords: coated paper · oil resistance · air permeability · polarity

Introduction

As the variety of foods containing grease is abundant, the types of oil-resistant materials used in food packaging has diversified in recent years. Environmental protection and safety issues continue to be important, and as a result, scholars have paid increasing attention to paper packaging materials (Coltelli et al. 2016; Johansson et al. 2012). Paper is a porous structure made from plant fibers that also have capillary phenomena, which result in underdeveloped greaseproof, air permeability, and moisture resistance properties in the paper products. Compounds such as PE (Rhim et al. 2006), EVOH (Zhang et al. 2001), and palm wax (Syahida et al. 2020) are often compounded with cellulose to reduce the porosity and wettability of paper materials and thereby improve their oil resistance. However, these composite papers lose some of their biodegradability and recyclability. Consequently, coating derived from biomass polymers have great research potential in food packaging applications.

Oleophobic materials have contact angle greater than 90°, and often are prepared by physical and chemical methods (Shiraki and Yokoyama 2020). The physical method is inspired by the "lotus leaf effect", and researchers try to recreate a regular, rough structure on the surface of the substrate by adding nano-fillers (Wang et al. 2014), preparing ordered porous structures (Zhang et al. 2015), and using composite paper support materials (Sundar et al. 2020). When an oleophobic materials comes in contact with grease, a layer of air is formed under the oil droplet due to the rough surface

1 structure. The influence of the rough structure on the oleophobicity of the material can
2 be characterized using the dynamic Cassie-Baxter model (Teisala and Butt 2019). The
3 chemical method examines the effect of the surface chemistry on the oil resistance of
4 the material. According to Young's equation, when the solid surface tension is much
5 smaller than the liquid surface tension, it is difficult to wet the surface with the liquid.
6 At present, fluorine-containing oil repellents often are used to reduce the surface energy
7 of the substrate as fluorine-containing compounds repel both grease and water. The
8 principle is that after the lipophilic groups in the fluorine-containing oil repellent
9 combine with the hydroxyl groups in the paper fiber, the surface energy of the thermal
10 trifluoromethyl group is low which protects the paper from being wetted by the liquid.
11 Chemical deposition (Jin et al. 2011), coating (Fukuda et al. 2013), dipping (Zhou et al.
12 2013) and other processes are often used to prepare greaseproof paper. The Owens-
13 Wendt model (Liu et al. 2018) can be used to analyze the effects of the surface energy
14 on the wetting of the paper by grease. Both methods for creating oleophobic surfaces
15 have certain drawbacks. Increasing the surface roughness can weaken the grease
16 wetting on the paper, but is not suitable for paper products that require heat sealing. For
17 example, most materials that contain inclusions cannot be heat sealed because the
18 increased surface roughness reduces the efficiency of the heat seal. Paper products
19 coated with polylactic acid must have a smooth surface to have strong heat seal strength
20 (Rhim and Kim 2009). In chemical methods, fluorine-containing oil repellents easily
21 lower the surface energy, but these fluorides also are easily converted into
22 perfluorooctane sulfonic acid (PFOS) and other harmful substances if they are heated.
23 These harmful substances bioaccumulate in the human body and can trigger multiple
24 chronic diseases (Cui et al. 2009).

25 The oil barrier material is based on forming a surface barrier on the paper to prevent
26 grease penetration. Biomass oil-repellents are gradually being used in food packaging
27 materials, but these coating have high surface energy and strongly adhere to grease. The
28 grease quickly spread over the surface once it comes in contact with the materials.
29 However, if the coating has excellent barrier properties, it can also achieve effective oil
30 resistance. Paper is composed of porous microstructures from the crystalline long-chain
31 polymers in cellulose. The irregular non-crystalline regions continuously destroy the
32 crystalline regions. The fiber pores in the paper are dense and the fiber hydroxyl content
33 is relatively high, consequently, the air permeability of paper is higher than other
34 substrates (Yoo et al. 2012). Air permeability is a non-negligible index that determines
35 the barrier properties of a material. Common biomass oil-repellent materials that reduce
36 air permeability include proteins (Bordenave et al. 2010), chitosans (Kopacic et al. 2018)
37 and alginate (Ham-Pichavant et al. 2005) among others. Although fluorine-containing
38 oil repellents achieve good oleophobic performance, they have little effect on the
39 porosity and air permeability of the substrate (Jiang et al. 2017). There are two ways to
40 improve the barrier properties of paperboard: increasing the degree of the pulp beating
41 or coating the paperboard with materials that act as outstanding barriers. Increasing the
42 degree of pulp beating degree can shorten the fibers and reduce the porosity of the
43 paperboard structure (Goswarni et al. 2008). Alternatively, Hassan (Hassan et al. 2016)
44 used nanocellulose and chitosan nanoparticles to prepare barrier films on paperboard.

1 The addition of the nanochitosan particles reduced the air permeability of the film, and
2 at the same time, enhanced the antibacterial and oil resistance of the film. Aulin (Aulin
3 et al. 2010) used carboxymethyl nanocellulose to prepare coated paper. The air
4 permeability of the coated paper was significantly reduced, and the resistance to castor
5 oil and turpentine also greatly improved. Li (Li and Rabnawaz 2019) coated the surface
6 of paper with chitosan, and good coverage reduced the porosity and improved the oil
7 resistance of the paper.

8 Surface chemistry suggests that coatings with a lower surface energy are not easily
9 wetted. Polar components are an important component of the total surface energy, and
10 polar groups repel grease. For example, Tuominen (Noeske et al. 2004) attached LDPE
11 and PP to the surface of paper, and then plasma and corona treated the surface. The air
12 permeability of the resulting paper changed, the coating surface energy and the content
13 polar content increased, and the oil resistance also improved. Ovaska (Ovaska et al.
14 2017) performed a corona treatment on hydroxypropyl starch coated paper. After a
15 negative corona treatment, the polar content and oil resistance of the coated paper
16 decreased. Fazeli (Fazeli et al. 2019) found that although the polarity decreased after
17 corona treatment, the adhesion between the coating and the substrate increased.

18 Two kinds of carboxymethyl chitosan (CMCS) based biodegradable coated papers
19 were prepared in this study, with a special focus on changing the oil resistance of the
20 coated paper. We mainly studied the oil resistance of two kinds of coated paper prepared
21 by either adding sodium carboxymethyl cellulose (CMC) or sodium alginate (SA). In
22 particular, we explored the influence of the air permeability and surface polarity on the
23 oil resistance of the coated paper. The main purpose of this work was to offer more
24 experimental data to better understand the factors that affect oil resistance in paper and
25 lay the foundation for future investigations.

26 **Experimental**

27 **Materials**

28 Carboxymethyl Chitosan (CMCS, degree of substitution $\geq 80\%$) with a viscosity of
29 10~80mPa·s, sodium alginate (SA, $(C_6H_7NaO_6)_n$) and sodium carboxymethyl cellulose
30 (CMC) with a viscosity of 300~800mPa·s were purchased from Sinopharm Chemical
31 Reagent Co., Ltd. White cardboard (quantity of 280g/m²) was provided by Shanghai
32 Young Sun Printing Co., Ltd. All the chemicals were chemical grade.

33 **Preparation of greaseproof paper**

34 Deionized water containing 1.5% (wt) CMC, 1.5% (wt) SA and 5% (wt) CMCS, was
35 stirred continuously for 2 hours at 50°C. CMCS and CMC were mixed at a 1:1 ratio,
36 and CMCS and SA were mixed at different ratios (CMCS:SA=1:0, 8:2, 6:4, 5:5, 4:6,
37 2:8, 0:1) to prepare the mixed solutions (Model of ZY-TB-B, Shandong Zhongyi
38 Instrument Co., Ltd.). Grease-proof paper was prepared by coating the cardboard with
39 the two mixed liquids using a coating machine. The coated papers were dried at 70°C
40 for 180 seconds, and then placed in a controlled environment with a relative humidity
41 of 50% and a temperature of 25°C for 24 hours.

1 Oil resistance tests

2 The standard TAPPI T559cm-12 often used in the paper industry was used to test the
3 oil resistance of coated paper. A total of 12 test solutions containing different
4 proportions of castor oil, toluene and n-heptane were prepared, thereby, dividing the
5 greaseproof paper into 12 grades. The liquid was released from a height of about 13mm
6 above the paper, and then quickly cleaned from the paper after 15 seconds to determine
7 the level of the oil resistance. Each coated paper was tested 5 times. According to the
8 standard test method, an oil proof value of more than 5 met the defined requirements
9 for oil-proof paper.

10 Contact angle measurements

11 A contact angle surface analyzer (Model JC2000D1, from Shanghai Zhongchen
12 digital technic apparatus Co.,Ltd, China) was used to measure the water contact angle
13 of the coated paper. Distilled water droplets with a volume of 5 μ l were placed on the
14 samples, and each sample was tested ten times to determine the average value of the
15 contact angle. The wetting of papers coated paper with the CMCS/CMC solutions with
16 oil resistance values of 8 from the above tests was measured.

17 Air permeability measurement

18 According to GB/T 458 2008, the papers was cut into 60mm \times 100mm samples, and
19 the air permeability value was measured using a paper and cardboard air permeability
20 analyzer (Model J-TQY10, Sichuan Changjiang Papermaking Instrument Co., Ltd.)
21 after treating the paper in a constant temperature and humidity box.

22 SEM analysis

23 The morphology of the coated paper was investigated by a SU1510 (Hitachi Ltd.,
24 Japan) scanning electron microscope (SEM) operating at 10kv high pressure to
25 determine the uniformity and coverage of the coating.

26 Surface roughness measurement

27 The surface roughness was measured using an MFP-D (RETC, America) atomic
28 force microscope (AFM) on samples that were cut into 2 \times 2cm sections. The
29 measurement area was 0.55 \times 0.44cm². Intuitively, the roughness of the coating was
30 reflected in the distance between the peaks and valleys in the images. The average
31 roughness (Ra) was calculated from the images, and each sample was measured ten
32 times to obtain the average.

33 Mechanical property measurements

34 According to the GB/T 22898-2008, the paper sample was cut into strips with a size
35 of 15 \times 100mm², and the tensile strength and elongation at break of the paper were
36 measured using a universal electronic material testing machine (LRX Plus, LLOYD,
37 UK). Each sample was tested three times and the average value was taken.

38 **Results and discussion**

1 Air permeability and oil resistance of the CMCS/CMC coated paper

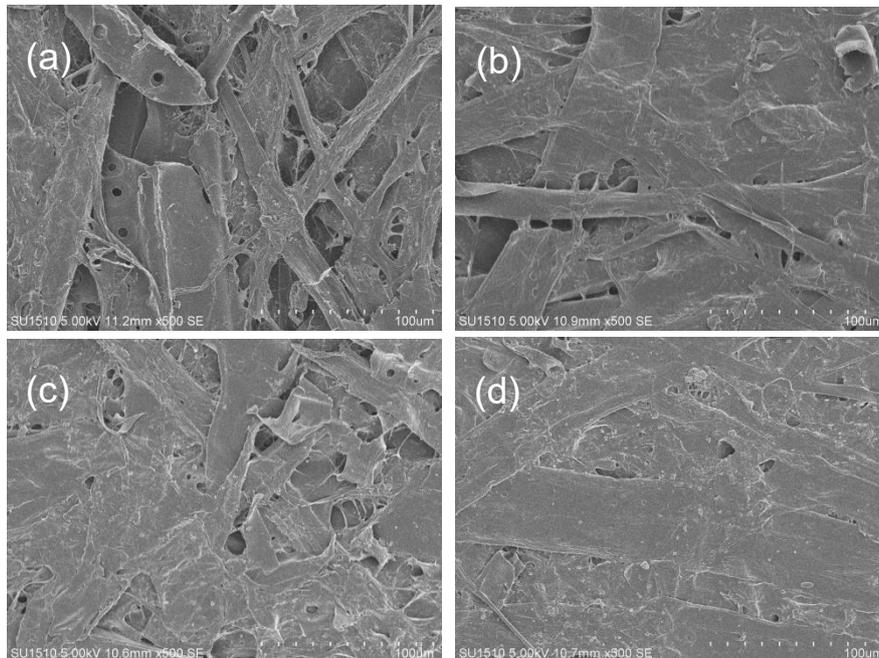
2 Table 1 Air permeability and oil resistance of CMCS/CMC coated paper

Coated weight (g/m ²)	0	1.02	2.04	3.06	4.08	5.1	6.12
Air permeability (mL/min) ^a	0.020±0.011	0.016±0.006	0.010±0.009	0.005±0	0.003±0	0.003±0	0.003±0
Kit no.	0	5	6	7	8	8	9

3 ^aMean±standard deviation

4 Table 1 shows the influence of the weight of the CMCS/CMC coating on the air
 5 permeability and oil resistance of the coated paper. Although the surface smoothness
 6 and barrier properties of cardboard were improved, the air permeability was still high.
 7 The selective gas permeability of the CMCS was outstanding, but strongly inhibited to
 8 O₂ and CO₂ permeation. As the coating weight increased from 1.02 g/m² to 4.08 g/m²,
 9 the sample kit value gradually increased from 5 to 8, but the air permeability gradually
 10 decreased. The barrier properties of the composite film formed by the CMCS/CMC
 11 coatings to gas and grease varied with the coating weight. An increase in coating weight
 12 improved the relatively high air permeability of the base material, which was also seen
 13 in work by Park (Park et al. 2000). Experimental results by Ham-Pichavant (Ham-
 14 Pichavant et al. 2005) also showed that the oil resistance positively correlated with air
 15 permeability, and the kit no. did not change significantly with increasing coating weight.
 16 The air permeability reached the limit when the coating amount was over 4.08 g/m².
 17 After coating, the air permeability significantly reduced, and the oil resistance level of
 18 the paper greatly improved to meet the requirements of packaging paper.

19 Surface micromorphology



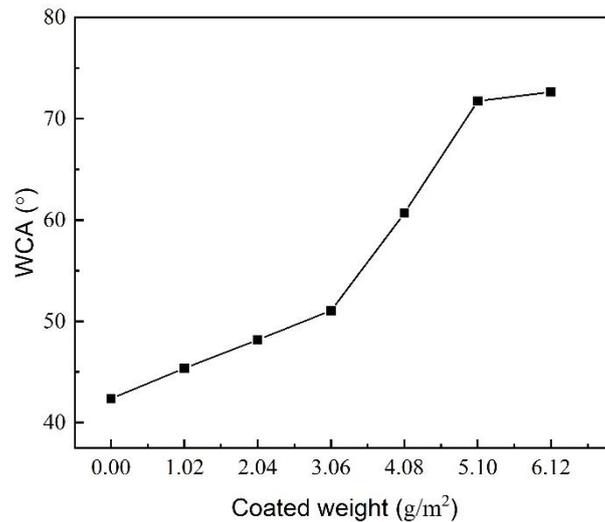
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21

22 Figure 1 SEM image of **a** uncoated paper, **b** coated CMCS/CMC with 1.02 g/m², **c** coated with
 23 3.06 g/m², **d** coated with 5.1 g/m²

24 In order to observe the coating coverage of the base paper, SEM image of the

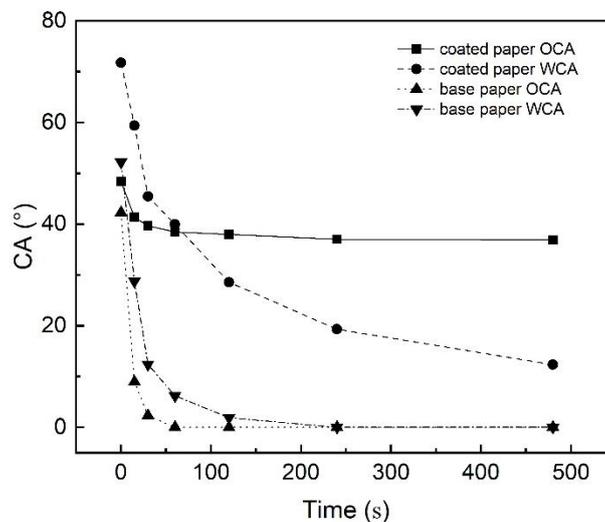
1 paperboard surface were analyzed. It can be vividly seen from the images that the
 2 degrees of coverage were uneven. While the paperboard with a coating weight of 1.02
 3 g/m^2 was covered by the coating, some uncoated fibers were still visible. When the
 4 coating weight reached 3.06 g/m^2 , most of the fibers were covered, but the coating was
 5 not uniform. Increasing the coating weight to 5.1 g/m^2 , the fibers and pores on the
 6 surface were fully covered, and the surface was flatter compared with base paper.
 7 Wettability of the coated paper surface



8
 9

Figure 2 WCA of CMCS/CMC coated paperboard

10 As seen in Figure 2, the anti-wetting performance of the paper was enhanced after
 11 coating with a CMCS/CMC mixture. When the coating weight reached 5.1 g/m^2 , the
 12 water contact angle (WCA) increased by nearly 30° . The formation of the composite
 13 film reduced the direct contact area between the paper fibers and water, and weakened
 14 the combined effects that lead to attraction between the fiber and water of the fiber's
 15 attraction to water. The water contact angle (CA) increased slowly with further
 16 increases in the coating weight, because the surface was already completely covered by
 17 the composite film, and the additional coating did not further affect the hydrophobicity.

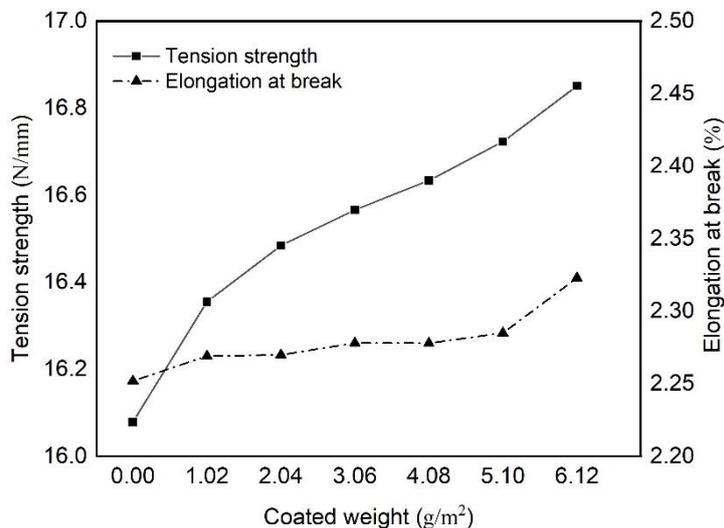


18
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Figure 3 Wetting properties of CMCS/CMC coated paper and base paper

1 In order to further explore the dynamic anti-wetting properties of the coated and base
 2 papers, the changes in the water and oil CA were observed over time to better reflect
 3 the barrier effects of coated paper. The trend in the dynamic changes of the CA of the
 4 coated papers with different coating amounts was similar, therefore, the coated paper
 5 with a coating weight of 5.1 g/m² was studied further. It could be seen from Figure 3
 6 that the oil contact angle (OCA) of the base paper quickly dropped to 0° within 60
 7 second, and the wetting effect was stronger. However, the CA of the coated paper
 8 decreased rapidly within first 15 second, and then only gradually decreased with
 9 increasing time. The OCA stabilized around 40°, suggesting that the grease-wetting
 10 resistance of the coated paper was improved greatly, and the coating maintained good
 11 oil-repellent performance for a long time. The changes in the water contact angle (WCA)
 12 of the base paper and coated paper followed the same trends over time. After 240
 13 seconds, the WCA of the base paper reached 0°, and the base paper was completely
 14 wetted. The anti-wetting behavior was enhanced after coating the base paper. At 0
 15 second, the WCA of the base paper increased by 19.6°, but then decreased over time.
 16 The reason was that the composite film formed by the CMCS/CMC coating prohibited
 17 direct contact between the water molecules and fibers. However, the film also contained
 18 abundant hydrophilic hydroxyl and carboxymethyl groups, and hydrophilicity of the
 19 film was also comparatively strong. Over time, the water molecules gradually
 20 penetrated into the spaces between the fibers and were finally completely absorbed.
 21 Therefore, the CMCS/CMC coating not only imparted excellent oil resistance, but also
 22 prolonged the water wetting time. These results were consistent with the experimental
 23 results of Arancibia (Arancibia et al. 2016).

24 Mechanical properties of the CMCS/CMC coated paper



25 Figure 4 Mechanical properties of CMCS/CMC coated paper

26 In practical applications, processed paper must have sufficient strength to protect the
 27 contents. Therefore, changes in the strength of the paper after coating will affect its
 28 application range. Figure 4 shows the effect of the CMCS/CMC coating weight on the
 29 mechanical properties of paper. Mechanical strength is an important indicator of the
 30 paper packaging materials. Sufficient mechanical strength can reduce damage to the
 31

1 contents caused by external impact. As shown in figure 4, the tensile strength (TS) and
 2 elongation at break (E) of coated paper increased with increasing of coating weight.
 3 When the coating weight reached 6.12 g/m², the TS was maximum and E was much
 4 larger than the base paper. The TS of the coated paper was affected by the fiber structure
 5 and the external environment. The length of the fiber and the bonding force between
 6 the fibers had a greater impact on the TS. The coating solution possessed a certain
 7 viscosity. After the paper surface was coated with CMCS/CMC solution, the molecules
 8 penetrated the surface and blended with the fibers. After drying, the fiber surfaces and
 9 solute molecules were tightly bound, therefore, the surface strength and TS of the paper
 10 was enhanced. However, the coating liquid did not penetrate into all of the gaps of the
 11 paper, so only the extensibility of the surface layer was improved and the resulting E
 12 increased slowly.

13 Surface energy and oil resistance of CMCS/SA coated paper

14 The surface tension is the sum of the interaction forces between the molecules on the
 15 surface. The surface tension concept better explains the wetting phenomena in papers.
 16 The polar components of the solid surface layer have a certain degree of repellency to
 17 grease, and the wetting performance of paper is influenced by the surface tension. In
 18 order to explore the effect of surface tension changes in the wetting behavior of the
 19 paper, different amounts of SA were added into the CMCS coating solution. Young's
 20 equation (1) describes the relationship between the surface tension and the measured
 21 CA, but does not enough to reflect the influence of each component to the CA. Owens
 22 and Wendt proposed a new surface tension model based on formula (2) and (3),
 23 where γ_l represents the surface tension of the liquid, γ_s represented the surface tension
 24 of the solid, γ_s^d represented the dispersion force component of the surface tension, γ_s^h
 25 represents the pole component of the surface tension, and θ is the measured angle
 26 between solid and liquid. However, to use this model, the contact must be measured
 27 with two liquids with opposite polarities. Water and diiodomethane were selected as the
 28 probe liquids, and combined with equation (4) to calculate the dispersion force and the
 29 polar components. Equation (5) allows for the calculation of the contributions of the
 30 polar component on the surface tension.

31 Table 2 The polar component (γ_l) and dispersion force component (γ_l) of the surface energy
 32 (γ_l) in the test liquid

Liquid	γ_l^d (mN/m)	γ_l^h (mN/m)	γ_l (mN/m)
Water	21.8	51.0	72.8
Diiodomethane	48.5	2.3	50.8

33
$$\gamma_s = \gamma_{sl} + \gamma_l \cos \theta \quad (1)$$

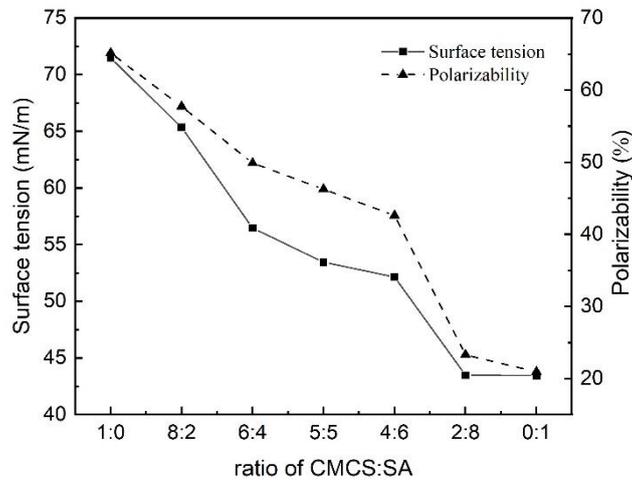
34
$$\gamma_l = \gamma_l^d + \gamma_l^h \quad (2)$$

35
$$\gamma_s = \gamma_s^d + \gamma_s^h \quad (3)$$

36
$$1 + \cos \theta = 2\sqrt{\gamma_s^d} \left(\frac{\sqrt{\gamma_l^d}}{\gamma_l} \right) + 2\sqrt{\gamma_s^h} \left(\frac{\sqrt{\gamma_l^h}}{\gamma_l} \right) \quad (4)$$

1
2

$$X = \frac{\gamma_s^h}{\gamma_s} \times 100\% \quad (5)$$



3

Figure 5 Surface tension and polarizability of the CMCS/SA coated paper

4

5 As revealed in Figure 5, different ratios of CMCS and SA led to different polar
6 component contents in coating. The surface tension and polarizability of the coating
7 decreased with the decreasing CMCS content. The addition of SA not only reduced the
8 surface tension, but also improved the gas barrier properties of the coating. For example,
9 after the solution was coated with a CMCS:SA ratio of 5:5, the surface tension was
10 lower, the kit value was up to 7, and the air permeability reached the measurement limit
11 of the equipment.

12 It can be seen from Young's equation that substances with a lower solid surface
13 tension are less wettable. According to Table 3, as the ratio of the solution changed from
14 1:0 to 8:2, the surface tension reduced to 65.34 N/km, the air permeability reduced, and
15 the oil resistance level rose to 8. The surface tension of the CMCS/SA coating with a
16 ratio of 8:2 was much higher than the coating with a ratio of 2:8, but the oil repellency
17 performance was significantly declined. Most greases have zero dipole moment and
18 small electronic activity, so they appear non-polar. According to the principle of
19 "similar polarity", the polar components in the coating were more resistant to the test
20 solution. Therefore, for high surface tension coatings with higher polarizability, the
21 polar components had a greater impact on grease the barrier properties.

22 The oil resistance of the coating with an 8:2 ratio was better than that of 1:0; however,
23 it was not clear if the decrease in air permeability or greater polar content played a more
24 critical role in determining the oil resistance. Therefore, paper with a coating weight of
25 1.02 g/m² and CMCS/SA coatings with a ratio of 8:2 and 0:1 were prepared to further
26 explore the effects of high-polarity components on the anti-oil effects under this test
27 condition. The results are shown in Table 4. Compared to the coated paper with a
28 coating weight of 3.06 g/m² and a ratio of 1:0, the coated paper with a coating weight
29 of 1.02 g/m² and a ratio of 8:2 had a higher air permeability, but the kit value was also
30 high. Meanwhile, the coating with a ratio of 6:4 in Table 3 and with a ratio of 0:1 in
31 Table 4 had the same air permeability, but the oil resistance performance was
32 significantly different. Combining results in Figure 5 verified that the higher polar

group content in the coating resulted in higher surface tension and the better oil resistant coating.

Table 3 Air permeability and oil resistance of CMCS/SA coated paper with a coating weight of 3.06 g/m²

CMCS:SA	1:0	8:2	6:4	5:5	4:6	2:8	0:1
Air permeability (μm/Pa•s)	0.010	0.007	0.004	0.003	0.003	0.003	0.003
Kit no.	5	8	7	7	6	4	3

Table 4 Air permeability and oil resistance of CMCS/SA coated paper with a coating weight of 1.02 g/m²

CMCS:SA	8:2	0:1
Air permeability (μm/Pa•s)	0.013	0.004
Kit no.	6	2

Surface micromorphology of CMCS/SA coated paper

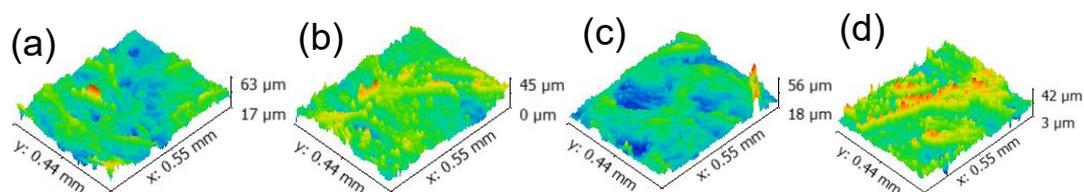
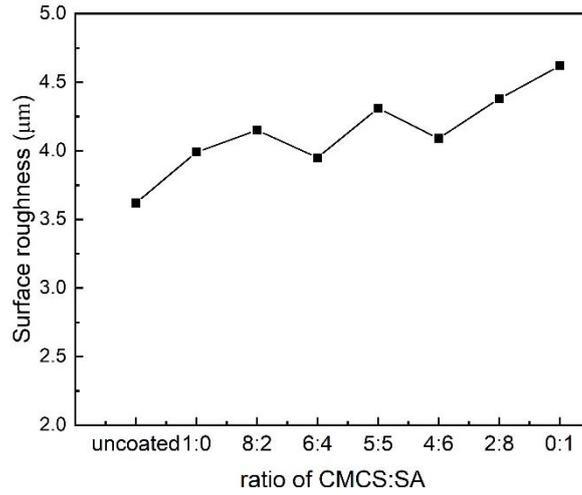


Figure 6 AFM image of CMCS/SA coated paper (a) with 1:0, (b) with 6:4, (c) with 5:5, and (d) with 0:1

The coating weight of the four samples was the same, and the solution coverage on the paper was ideal, so there was little difference in the microscopic state seen with the electron microscope. In order to further analyze the differences in the surface layers of the different coated papers, AFM was selected to observe the 3D morphology of the surface layers. The surface roughness is shown in figure 6 (a-d) after coating the papers with different proportions of the solutions. It was vividly found that the wave distance was within 80 μm. However, the surface roughness of the coated papers changed with increasing dosage of SA in the coating. The reason might be based on the following aspects: the fibers were relatively large, the surface of the cardboard was not smooth surface, and the roughness presently differently. After coating, film formed on the fibers had excellent replicability and was better than the original roughness of the cardboard surface. The solute molecules penetrated some gaps between the fibers and adhered to the fibers and surface. After drying, they had different effects on the surface roughness. As seen in Figure 7 the roughness of the coated paper did not show regular changes with the coating composition.



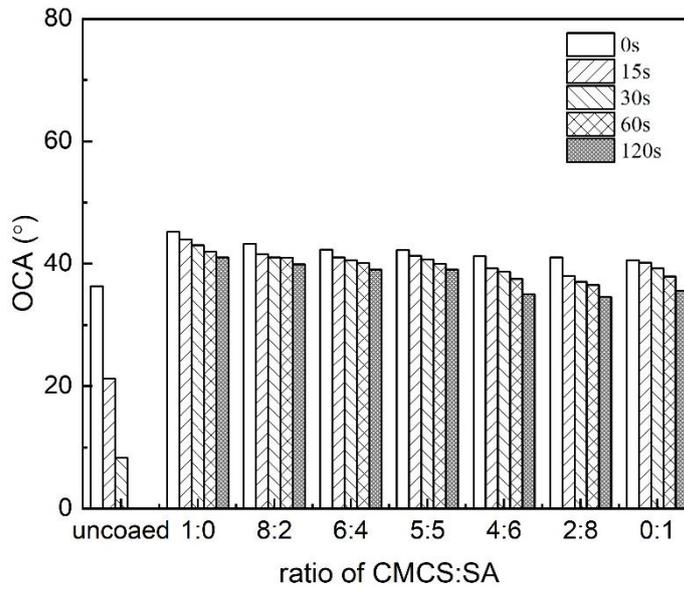
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Figure 7 Surface roughness of CMCS/SA coated paper

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Surface wettability of CMCS/SA coated paper



4

5

Figure 8 OCA of CMCS/SA coated paper

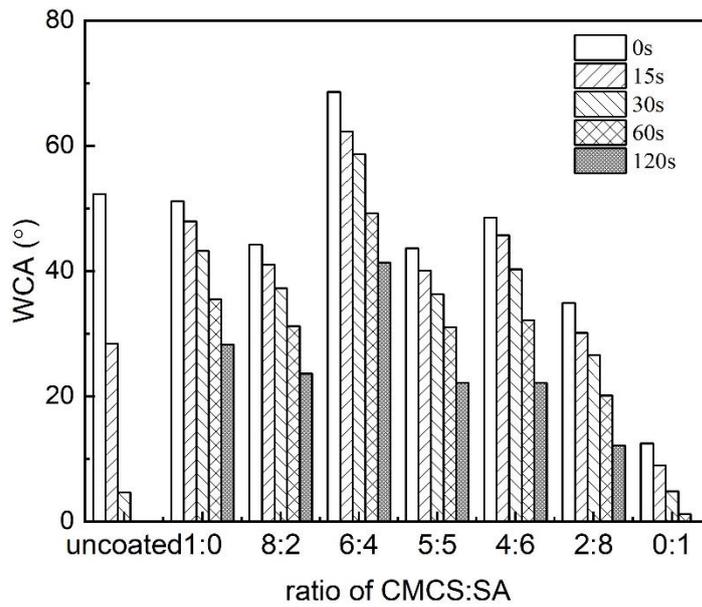


Figure 9 WCA of CMCS/SA coated paper

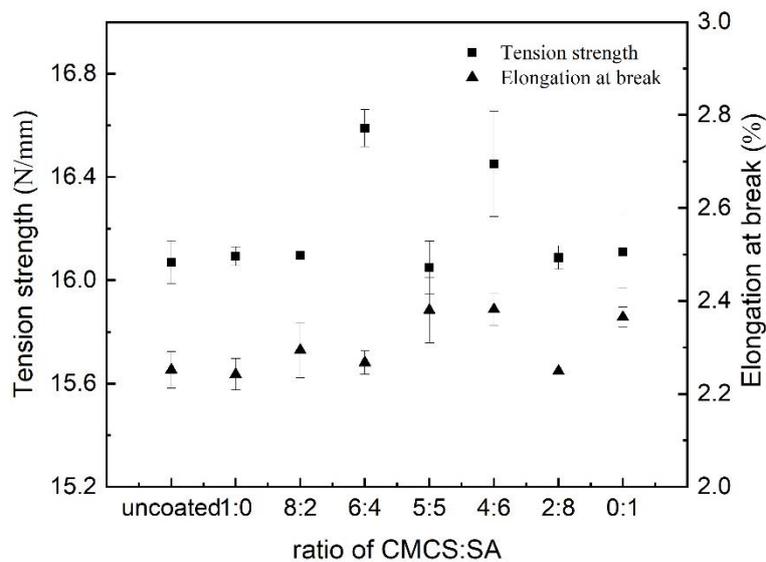
The dynamic changes of the OCA within a specified time reflect the oil resistance of the coated paper. It can be seen (Figure8) that the OCA of the various coated papers increased compared to the base paper. After 60 seconds, the base paper was completely wetted by grease, and the contact angle became 0°. Meanwhile the contact angle of the coated paper only decreased by 5° after 120 seconds, indicating that the coating formed a good barrier to grease. Within the same time, as the CMCS content in the coating decreased, the reduction of the OCA gradually increased, indicating that the oil resistance was better for the given period time.

The Figure 9 shows that the water resistance of the coated paper varied irregularly with the SA content in the coating, but no matter what kind of coating was used, the water-blocking performance of the paper improved. Although the WCA of the coated paper with a ratio of 0:1 was much smaller than the base paper, the water blocking time was prolonged, indicating that the water resistance of the paper improved regardless of the coating composition.

Due to the differences in the barrier properties, surface energies, and polar contents of the different coatings, the coated papers exhibited differences in their water and oil resistance. However, the changes in OCA and WCA reflected in the above figure are not consistent with the changes in barrier properties shown in Table 3. The impact of the barrier properties to grease and water was not a direct factor. After analyzing Figure 5, it was found that the surface tension and polarization rate of the coating gradually decreased. Most grease molecules are non-polar, while water molecules are polar due to their uneven charge distribution. The coatings presented here repelled grease but were hydrophilic. The barrier properties of the coated paper to grease first increased and then decreased, indicating that the surface tension and polarization mainly impacted the grease resistance of the coating. The roughness of the different coated papers was different (Figure7). Previous research suggested (Taguet et al. 2014) that more complex surface microstructures with WCAs over 90° was easier to wet, but materials with

1 WCAs less than 90° presented opposite phenomenon. The trends in moisture resistance
2 of the coatings was similar to the trends in surface roughness. The surface roughness of
3 the coated paper with a ratio of 0:1 was relatively high. Although the surface tension
4 was small, it can be concluded from Figure 9 that the surface roughness had a greater
5 effect on the water wettability than the surface tension, which was similar to
6 conclusions reached by Azimi (Azimi and He 2020). Therefore, the surface roughness,
7 surface tension, and polarization of the coating together affected the water and oil
8 resistance of the coated paper.

9 Mechanical strength of the CMCS/SA coated papers



10

11 Figure 10 TS and E of CMCS/SA coated paper

12

13 The mechanical properties of coated paper were improved compared to the base
14 paper (Figure 10). The viscosity of the solution was relatively high, and the bonding
15 strength between the fibers was enhanced after coating penetrated into the pores. Both
16 the CMCS and SA composition were experimental variables. The different CMCS and
17 SA proportions in the coating, but E was relatively stable. When the composition ratio
18 was 6:4, the TS reached a maximum value and the E was also better. Therefore, the
various coatings improved the TS, but had little effect on the E of the paper.

19 Conclusion

20 Coating biomass compounds onto base paper has great development potential and
21 application value in food packaging. This method abandons the application of
22 traditional fluorine-containing materials, yet still meets the oil resistance level required
23 for food packaging. The lower air permeability of the coating had a significant effect
24 on the oil resistance of the coated paper, and the influence of the surface tension and
25 polar content of the coating on the oil resistance of the coated paper could not be ignored.
26 For hydrophilic materials, reducing the surface roughness was conducive to improving
27 the hydrophobicity of the material. The mechanical properties of the coated papers were
28 improved to a certain extent. Research and development into new coatings, preparation

1 processes for creating coated paper and the reduction in coating costs still need to be
2 studied further. Biomass materials also have good prospects in antibacterial resistance,
3 and could be used to develop multifunctional environmentally friendly food packaging.

4 **Author Contribution**

5 F.W. and L.W. conceived the idea and supervised the research. F.W., X.Z. and F.M.
6 contributed to the material preparation and characterization. F.W., F.M. and L.W.
7 conducted the property measurements. F.W. and L.W. contributed to the writing of the
8 manuscript. All authors reviewed and commented on manuscript.

9 **Notes**

10 The authors declare no conflicts of interest.

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