Water and Oil Resistance Improvement of Paper Coated with Aqueous Mixture of Hydrophilic and Hydrophobic Cross-Linked Copolymers

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An aqueous mixture of hydrophilic and hydrophobic cross-linked copolymers was prepared and coated on the surface of filter paper to improve the water and oil resistances. The mixture was mainly composed of sodium alginate and hydroxyl-terminated polydimethylsiloxane (PDMS)-tetramethoxysilane (TMOS) cross-linked network. The results indicated that the resistances to both water and oil of the resulting paper were significantly enhanced. The water contact angle results demonstrated that the mixture enhanced the hydrophobic properties of the coated paper greatly with water contact angles higher than 110°, and the coated paper showed excellent oil resistance with a Kit number as high as 10.

Keywords: Sodium alginate; PDMS; Mixture; Water resistance; Oil resistance

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INTRODUCTION

Paper products manufactured from cellulose fibers are widely used in the packaging industry. Water and oil/grease resistances are essential properties required for packaging papers. However, cellulose fiber-based paper products are insufficient for high-barrier applications, particularly for moisture barriers and oil/grease resistance because of paper’s hydrophilic nature and the structure. To improve the hydrophobic properties and widen its applications, numerous studies have been conducted that have focused primarily on the hydrophobic modification of cellulose fiber surfaces and/or the preparation of super-hydrophobic papers (Zou *et al.* 2007; Perng and Wang 2012; Gandini and Belgacem 2015). Some other functionalization of cellulose fibers *via* etherification or esterification reactions (Fukuda *et al.* 2013) also have been reported. For oil/grease resistance, the conventional treatments include physical and chemical modifications of fibers or paper surfaces with fluorocarbon chemicals, waxes, and synthesized polymers. Another method is to coat or laminate metal foils or plastic films on paper surfaces to induce grease resistance in the paper products (Yang *et al.* 1999; Vaswani *et al.* 2005; Zou *et al.* 2007; Aulin *et al.* 2008; Fukuda *et al.* 2013). The effects of the type of pulp fibers, refining, and using fillers and binders to optimize greaseproof paper properties have been discussed in detail by Perng and Wang (2012). Recently, an extensive and detailed review on producing of greaseproof paper, including the regulations and approaches, from conventional treatment to newly emerging methods was reported by Hubbe and Pruszynski (2020).

However, most of the above-discussed methods can provide resistance against only one of the two kinds of fluids – grease or water. The only treatment that is widely used to achieve for both hydrophobic and grease resistance properties of paper products involves the use of perfluorinated compounds. Such compounds can inhibit the wetting of fibers by both oil and water because of their low surface energy. However, their negative impacts on the environment and high cost have limited the application of fluorocarbon-based chemicals and waxes. Therefore, it is necessary to develop novel alternatives that can replace these fluoro chemicals and petro-based waxes with environmentally friendly alternatives that are of natural origin, biodegradable, and able to limit water and oil/grease penetration into paper materials.

From the previously reported papers, employing mixed barrier coatings could obtain complex barrier performance of coated paper. For instance, starch-poly(vinyl alcohol) barrier coatings were applied for flexible packaging paper coating, and the oxygen and water vapor transmission rates of starch-ethylene modified poly(vinyl alcohol) barrier coatings coated flexible packaging paper were investigated (Christophliemk *et al*. 2017a). The mixtures based on PVOH and hydrophobically modified starch can provide superior barrier performance for flexible package paper (Christophliemk *et al.* 2017b).

One way to improve both water and oil resistance of paper is to employ a mixture of sodium alginate and organosiloxane materials. These substances are based mainly on renewable and biodegradable materials. Sodium alginate (SA) is a water-soluble linear polysaccharide derived from alginic acid, which is extracted from brown algae (Rezende *et al.* 2009). The interest in alginate is because of its unique colloidal properties, including film-forming, biocompatibility, biodegradability, and bioadhesive properties (Pereira *et al.* 2011). SA is hydrophilic and is a suitable candidate for chemical modification *via* its carboxyl and hydroxyl groups distributed along the backbone (Andersen *et al.* 2012). It has been commonly used to preserve minimally processed food products, such as fruits, vegetables (Raybaudi-Massilia *et al*. 2008), and meat products (Marcos *et al.* 2008; Chidanadaiah and Sanyal 2009; Del Nobile *et al.* 2009; Khanedan *et al.* 2011; Mastromatteo *et al.* 2012). It is attractive for food packaging because of its antimicrobial activity (Kim *et al.* 2010) and ability to protect the product quality and increase the product shelf-life. Since SA has been widely used in food processing and food preservation, it is a definitely potential candidate for coating layer on paper packaging application. Organosiloxane materials have been found throughout the processing of pulp and paper, for example as digester additives in wood chips and cooking processes, as antifoams in pulp washing and papermaking processes, as additives in finishing processes, and as de-inking aids in paper recycling (Andriot *et al.* 2007). Polydimethylsiloxane (PDMS), with its good biocompatibility, low surface tension, mechanical strength, excellent oxygen permeability, high thermal stability, ease of fabrication, *etc*., has good application prospects in the biomedical field, consumer products, and other industrial fields (Fichet *et al.* 2005; Tan *et al.* 2009; Tang *et al.* 2011; Ibrahim *et al.* 2011; Daemi and Barikani 2012).

To enhance both water and oil barrier properties of paper, the hypothesis of this work was that the hydrophobic character and effective barrier for water vapor and oil could be achieved using a mixture (SA-PT) of a relatively hydrophilic copolymer (SA) and a relatively hydrophobic copolymer (PT). In such a system, the PT is a hydrophobic cross-linked network of hydroxyl-terminated PDMS and tetramethoxysilane (TMOS). The schematic illustration of the experimental steps is shown in Fig. 1. This SA-PT mixture may play a significant role in generating both water- and oil-resistant properties, because SA tends to form a high-density film with a high density of hydrogen bonding, which has potential to hinder oil penetration under dry conditions. The PT network tends to migrate to the outer surface during drying, where it would have the potential to act as a protective layer to oppose water penetration. Once the film is dried, the outer part of the film, with its hydrophobic groups of PT oriented at the surface, would tend to keep the rest of the structure from getting wet. Therefore, by mixing the two polymers could lead to enhanced water and oil resistances of the coated paper. The system employs hydrophilic and hydrophobic phases in one coating step to improve barrier properties of paper, which is carried out in a facile way for fabricating nontoxic and environmentally friendly paper-based packaging materials with high barrier properties.

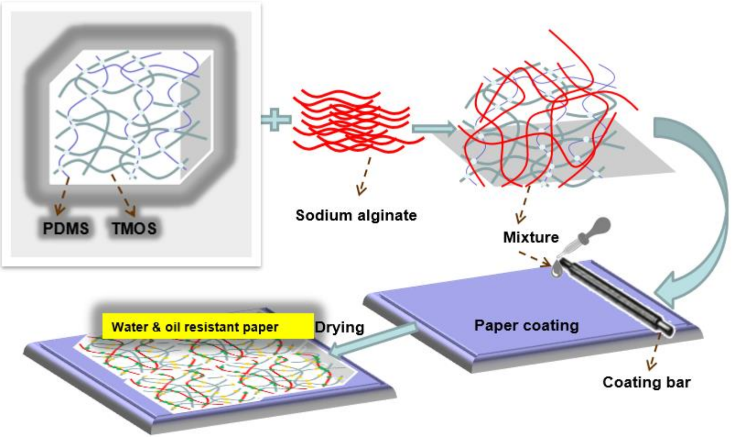


Fig. 1. Schematic illustration of preparation and application of the SA-PT mixture on paper

EXPERIMENTAL

Materials

Hydroxyl-terminated PDMS with an average molecular weight of 4200 was purchased from Alfa Aesar (Shanghai, China), and TMOS (> 99.0%) was purchased from TCI (Tokyo, Japan). Sodium alginate was obtained as a solid powder from Aladdin (Shanghai, China). Dibutyltin dilaurate (DBTL, 95%) was purchased from Macklin (Shanghai, China). The base papers used for coating in this work were fine filter papers and were purchased from Minzheng Filter Paper Company (Fushun, China). Anhydrous ethanol, castor oil, and *n*-heptane were purchased from Fuyu Fine Chemical Co., Ltd. (Tianjin, China). Potassium nitrate (KNO_3) and magnesium nitrate ($Mg(NO_3)_2$, 99%) were purchased from Damao Chemical Company (Tianjin, China). Toluene (99%) was purchased from Zhongshan Chemical Company (Nanjing, China).

Methods

Preparation of the coating mixture

In the first step, the PT network was prepared by crosslinking copolymerization of hydroxyl-terminated PDMS with TMOS as a crosslinker in the presence of DBTL as the catalyst. The preparation of the PT network was done in a three-neck reactor equipped with a magnetic stirrer, reflux condenser, and thermometer. Anhydrous ethanol, deionized water, and TMOS were added at a 1:1:2 mole ratio with TMOS:DBTL ratios of 1:0.01, 1:0.02, and 1:0.03, respectively. The hydrolyzed TMOS network was coated on paper, and the water contact angle of coated paper was measured to determine the optimum ratio between TMOS and DBTL (Table 2). Then the PT network was prepared by adding various ratios of PDMS to the previous step. The resulting PT network was coated on paper via a TQC coater. The water contact angle of PT coated paper was measured to determine the optimum ratio between the TMOS and PDMS, and the respective levels of PDMS and TMOS are shown in Table 3, in which the ratios of TMOS, DBTL, deionized water, and ethanol were added based on the previous steps. After that, SA and PT network mixture was prepared by adding the SA solution to the PT network with the same volume ratio of SA to PT and stirred. The schematic illustration of the synthesis route of SA-PT mixture is shown in Fig. 2.

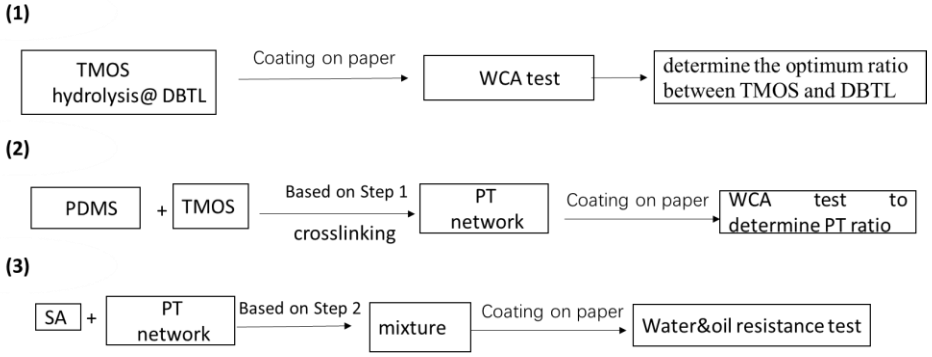


Fig. 2. Schematic illustration of the preparation steps of the SA-PT mixture

Paper surface coating with the SA-PT mixture

The coating process was carried out using a TQC coater (TQC Ltd., Rotterdam, Netherlands) on the filter paper surface with a coating bar at a speed of 5 m/min, which allowed the formation of a uniform layer with a relatively uniform coating thickness. Finally, the samples were dried overnight at 75 °C to 80 °C in an air convection oven.

Characterization

The morphology of the coated and uncoated paper was observed by scanning electron microscopy (SEM) (Quanta 200 SEM, FEI Ltd., Oregon, USA) after being coated with gold using a 10-kV accelerating voltage.

Water contact angle measurements were used to detect the hydrophobic properties of the PT network and the resulting mixture coated paper, which were conducted according to the sessile drop test method using a versatile optical tensiometer (JC2000C1, Shanghai Powereach Co., Ltd., Shanghai, China). Approximately 3 μL of distilled water were dropped on the surface of the samples. The contact angle of each sample was measured at least five times and the average values were calculated. The changes in the contact angle with time were also recorded and calculated.

The oil resistance properties of the mixture coated papers were conducted according to the Kit test method (TAPPI T559 cm-12 (2012)) and oil contact angle test. The oil contact angle test of the coated paper was conducted with the same procedure as the water contact angle test. The Kit method tests the oil resistance of coated paper with different numbered Kit solutions that contain various ratios of castor oil, *n*-heptane, and toluene, which is shown in Table 1. First, a drop of oil was added to the test specimen from a height of 13~~,~~ mm, and the excess test solution was quickly removed with a clean tissue or cotton swatch after 15 s. Immediately, the specimen was examined. If the color of the specimen was unchanged, then the sample met the oil resistance standard. Then, the Kit solution was changed to a higher grade, until the oil penetrated the specimen. The steps were repeated until the highest numbered kit solution that did not result in oil penetration identified. The results were the average of five independent tests.

Table 1. Mixtures of Reagents for Preparing the Kit Solutions

Kit No.	Castor Oil (g)	Toluene (mL)	n-Heptane (mL)
1	969.0	0	0
2	872.1	50	50
3	775.2	100	100
4	678.3	150	150
5	581.4	200	200
6	484.5	250	250
7	387.6	300	300
8	290.7	350	350
9	193.8	400	400
10	96.9	450	450
11	0	500	500
12	0	450	550

The water vapor transmission rate (WVTR) of the coated paper was calculated from the amount of water vapor that passed through the test material per unit area (m^2) per unit time (24 h/d) at a certain temperature and relative humidity (RH). In this work, the WVTR was evaluated by the gravimetric method, which was modified according to the standards ASTM E96 (1995) and ASTM D1653-2003(2008). The WVTR determination was performed at 37.8 °C and 90% RH and at 23 °C and 50% RH. The test samples were sealed in test dishes containing saturated KNO_3 or $Mg(NO_3)_2$ solutions, which maintained a RH of 90% or 50%, respectively. Then, the assembled test dishes were placed in a chamber, which was maintained at a constant temperature of 37.8 °C and 23 °C, and 0% RH by anhydrous calcium chloride. The weight changes were obtained by weighing the dishes periodically. The WVTR (g/m^2d) was finally calculated according to the literature (Lu *et al.* 2014).

RESULTS AND DISCUSSION

Determination of the Optimum Reaction Ratio between the PDMS and TMOS

The PT network was prepared *via* the crosslinking reaction with the two materials, PDMS and TMOS, and DBTL was used as a catalyst for the TMOS hydrolysis (Ren *et al.* 2012; Li *et al.* 2013). A condensed polysiloxane network with cross-linking points was formed during this reaction (Mackenzie *et al.* 1992). In an attempt to find an appropriate reaction ratio between the PDMS and TMOS, the reaction conditions and primary results of the barrier properties for the coated paper are listed in Tables 2, 3, and Fig. 3. The filter paper was coated with resulting PDMS/TMOS network and then oven-dried at 35 °C before determining the water contact angle. For this step, anhydrous ethanol, deionized water, and TMOS were added at a 1:1:2 mole ratio with TMOS:DBTL ratios of 1:0.01, 1:0.02, and 1:0.03. The resulting coated papers were labelled DP1, DP2, and DP3, respectively. The various mole ratios of PDMS to TMOS can be seen in Fig. 3. According to the results in Table 2, the coated paper displayed similar water contact angles when the ratio of TMOS to DBTL was 1:0.01 and 1:0.02. However, when the ratio of TMOS to DBTL was 1:0.03, the hydrolyzed TMOS gelled quickly and paper coating failed; therefore, the water contact angle data were not available.

Table 2. Hydrolysis Conditions and Results for the TMOS

Reaction Condition	Water Contact Angle (°)			
TMOS:DBTL	0 s	10 s	20 s	60 s
1:0.01	100	98	96	95
1:0.02	98	97	96	94
1:0.03	NA	NA	NA	NA

Table 3. Trials on Determining PDMS and TMOS Ratios and PT Network Outcomes Description

	Mole ratio		PT network outcome description	
Trial No.	PDMS	TMOS		Water contact angle of coated paper (°)
1	100	0	Transparent oily liquid	NA
2	99	1	Transparent oily liquid	NA
3	95	5	Liquid with solids	NA
4	90	10	Cloudy with granules	97
5	80	20	Cloudy with granules	99
6	70	30	Transparent, sticky	100
7	60	40	Gel-like	99
8	50	50	Gel-like	68
9	40	60	Gel-like	69
10	30	70	Liquid with solid	NA
11	20	80	Liquid with solid	NA
12	10	90	Solid layer &little liquid	NA
13	5	95	Oily layered Liquid	NA
14	1	99	Too much Water	NA
15	0	100	Crystal solids	NA

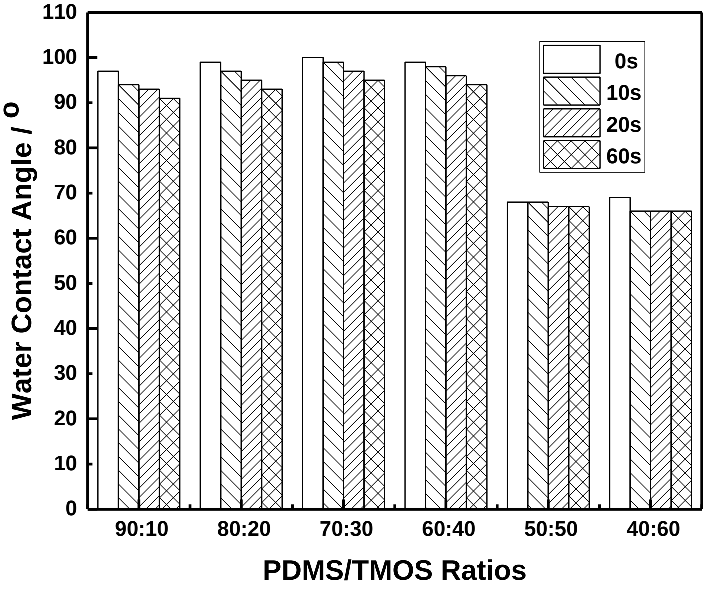


Fig. 3. Water contact angles of the PT network-coated paper

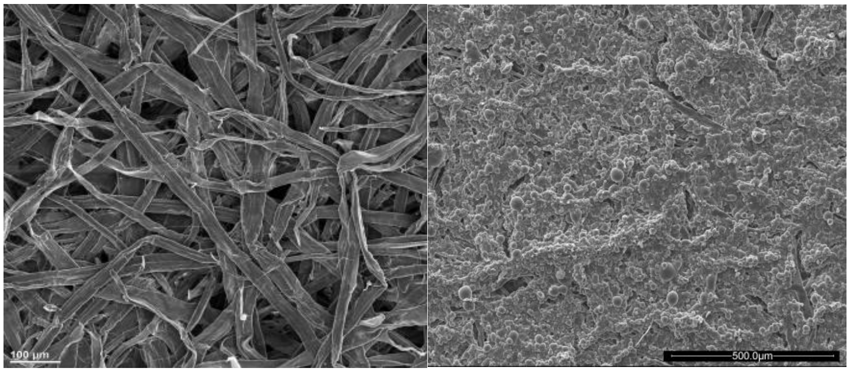
Based on the resulting appearances of PT network with different ratios of PDMS and TMOS, as well as the water contact angle results (Fig. 3 and Table 3), it was concluded that a hydrophobic property of the coated paper was obtained, and the water contact angles decreased slightly within 60 s when the mole ratios of PDMS to TMOS were higher than 50:50. When the ratios were getting higher, the water contact angle values reached approximately 100°. For comparison, PDMS to TMOS mole ratios of 80:20, 70:30, and 60:40 were used for the next steps.

Properties Study of the Coated Paper

A set of mixtures was prepared based on different experimental variables, as is shown in Table 4. The mole ratios of PDMS to TMOS were kept at 80:20, 70:30, and 60:40. The mole ratio of TMOS to DBTL was 1:0.01 and the SA solution concentration was 2%.

Surface morphology of coated paper via SEM analysis

The morphologies of the paper and coated paper were analyzed by SEM, as shown in Fig. 4. Compared to the uncoated samples with clear texture of fibers (Fig. 4a), the surface of the coated paper (Fig. 4b) was covered by the mixture with obvious granules.



(a) **(b)**

Fig. 4. SEM images for the uncoated (a) and coated paper samples(b) with a coating weight of approximately 10 g/m^2

Water contact angles of the SA-PT mixture coated paper

The surface hydrophobic properties of the coated paper were examined by water contact angle measurement. The water contact angle and its change with time (60-s period) were also recorded, and the results are shown in Fig. 5. The water contact angle of the control sample was measured around 45°, and it quickly changed to zero because the water drop was absorbed by the filter paper. In comparison, the coated paper showed a higher water contact angle in the range of 100° to 120°, which indicated good hydrophobic properties. The water contact angle values in Fig. 5 decreased slightly within the 60-s testing period, which meant that the coating surfaces were water resistant and there was less water absorbed by the testing samples. During the coating process, the SA would tend to form a dense film with a high density of hydrogen bonding, which would have potential to block gases under dry conditions. The hydrophobic PT system would tend to migrate to the outer surface during drying, where it may have the potential to act as a protective layer. when the film is dried, the outer part of the film, with the hydrophobic groups oriented at the surface, would tend to keep the rest of the structure from getting wet. That is why, as reported, the PDMS was considered to be an attractive material because of its water repellency, strong resistance, and low-density surface properties (Jin *et al.* 2010).

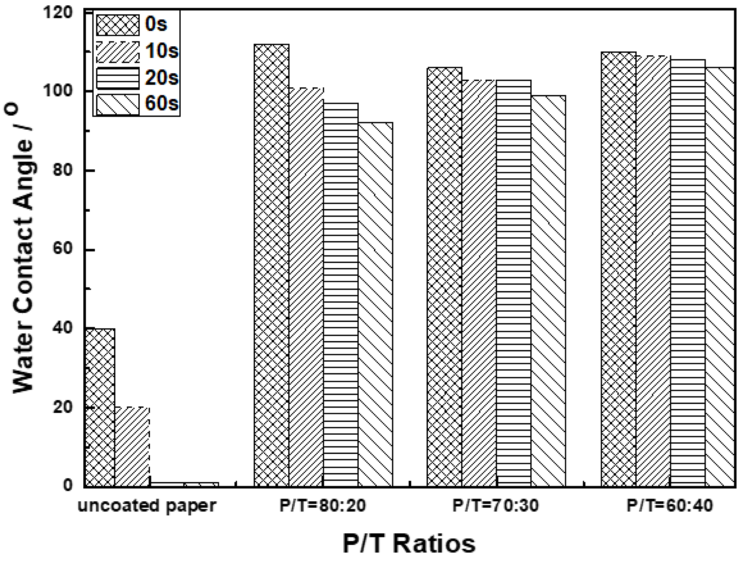


Fig. 5. Water contact angles of the SA-PT mixture coated paper

Oil contact angles of the SA-PT mixture coated paper

The oil contact angle results of the SA-PT mixture coated paper are shown in Fig. 6. For all types of SA-PT mixture coated paper, it was found that the oil contact angle values were around 40 to 80°. As explained in Fig. 5, the outer surface of the coating layer was the PT network hydrophobic layer, and it was reasonable that the coated surface was lipophilic.

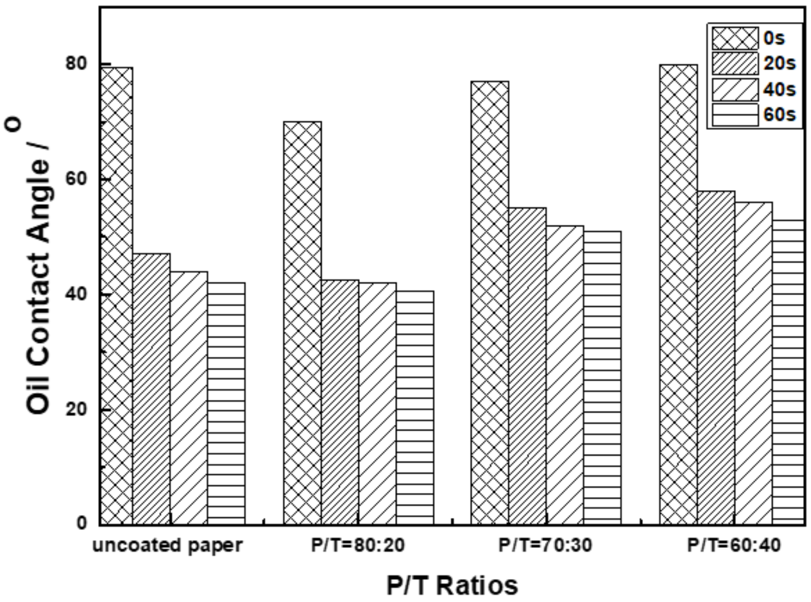


Fig. 6. Oil contact angles of the SA-PT mixture coated paper

WVTR and Kit No. of the coated paper

The WVTR values and the Kit levels of the uncoated paper and paper coated with SA, PT system, and SA-PT mixture are shown in Table 4.

Table 4. WVTR Test and Kit Test of the SA-PT Mixture Coated Paper

Samples	P/T ratios	Coating weight (g/m^2)	WVTR values ((g/m^{2}d))		Kit No.
			50%RH,23 ^{o}C	90%RH,37.8 ^{o}C	
Uncoated paper		0	848.1	3180.8	0
SA coated paper		17.5	766.6	3106.1	8
PT system coated paper	P/T = 90:10	13.4	703.2	2878.9	1
	P/T = 80:20	11.1	680.3	2874.7	1
	P/T =70:30	15.3	670.9	2958.8	1
	P/T =60:40	12.9	801.1	2811.9	1
SA-PT mixture coated paper	P/T = 90:10	12.1	635.6	2291.9	10
	P/T = 80:20	13.2	548.1	2627.1	10
	P/T =70:30	13.4	610.5	2793.5	10
	P/T =60:40	15.1	467.7	1806.1	10

From Table 4, for the SA coated paper, the WVTR values decreased slightly (9.6%) compared to the uncoated paper, while the Kit No. was up to 8. This is due to the hydrophilic nature of sodium alginate and its good film-forming property. The paper surface was sealed with a layer of dense film formed by sodium alginate, so it was difficult for the oil to penetrate through the paper so that the coated paper showed high oil resistant property. Comparably, for the WVTR test, the hydrophilicity of sodium alginate film can only slightly delay but cannot completely block the transmission of water vapor through the paper, especially when the SA film was thoroughly surrounded in the water vapor at high relative humidity (RH 50% or 90%). Because SA is hydrophilic and it possesses considerable water absorbing and retaining properties, it is widely used for water absorption or superabsorption applications (Berger *et al.* 1953; Mallepally *et al.* 2013). Further explanations in the literature mentioned that moisture has a plasticizing or swelling effect on hydrophilic polymers that results in increased permeability (Ashley 1985). Therefore, under extreme testing conditions, the water absorbed by the SA tended to swell the coating layer and further damage it, so the water vapor transmitted from the crack and induced a higher WVTR. Hence, from the tests conducted at 37.8 °C and 90% RH, the WVTR of the coated paper only decreased by 20% to 30%.

For the PT network coated samples, the WVTR decreased around 20% at 23 °C and 50% RH, 11.6% at 37.8 °C and 90% RH, respectively. The Kit level was only around level 1. The results indicated that the hydrophobic PT network had the potential to act as a protective layer and it was effective on hindering the transmission of water vapor, but not for the oil transmission.

It is noticeable that the SA-PT mixture coated paper showed much better water vapor barrier properties and oil resistant compared with other samples. For the samples tested at 37.8 °C and 90% RH, the WVTR values decreased by 43.2% compared with that of the uncoated paper; for the samples tested at 23 °C and 50% RH, the WVTR values decreased by approximately 44.8% compared with that of the uncoated paper. It is significant that the Kit No. of the mixture coated paper reached as high as 10. This indicated that the oil resistance properties of the SA-PT mixture coated paper have been in the satisfying oil resistance range required for regular food packaging paper and the resulting SA-PT mixtures are suitable for the preparation of oil-resistant paper. As discussed above, the PT system and the SA formed a synergistic effect, in the way that the hydrophobic nature of PT system can delay the wetting of the sample and the presence of SA in the mixture can form a dense layer to obstruct the transmission of the water vapor and oil through the sample.

CONCLUSIONS

1. A new coating mixture involving hydrophilic sodium alginate (SA) and hydrophobic cross-linked network of hydroxyl-terminated PDMS and tetramethoxysilane (PT) was prepared, and the resulting mixture was successfully applied as a paper surface treatment to simultaneously enhance the water and oil barrier properties of paper substrates. The results showed that the SA-PT mixture enhanced the water repellency of the coated papers, which was revealed by the water contact angle and water vapor transmission rate (WVTR) measurements.
2. The measured water contact angle was higher than 110°, and it decreased slightly within the 60-s testing period, which revealed a relatively steady hydrophobic property. The WVTR values of the coated paper were reduced by different degrees with respect to the different testing conditions.
3. The SA-PT mixture also played positive roles in increasing the oil resistance of the coated paper with the highest Kit number of 10.
4. It is worth mentioning that the present work offers a good method for the preparation of hydrophilic-hydrophobic coating mixtures for use in the papermaking industry, such as for producing paper-based packaging materials with water- and oil-resistance.

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