Balancing the decomposable behavior and wet tensile mechanical property of cellulose-based wet wipe substrates by the aqueous adhesive

Tongtong Yun [a], Peng Cheng [a], Fang Qian [b], Yi Cheng [a], Jie Lu [a], Yanna Lv [a,*], Haisong Wang [a,*]

[a] School of Light Industry and Chemical Engineering, Dalian Polytechnic University, Dalian 116034, Liaoning, PR China
[b] School of Food Science and Technology, Dalian Polytechnic University, Dalian 116034, Liaoning, PR China

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ABSTRACT

With the current global outbreak of novel coronaviruses, the fabrication of decomposable wet wipe with sufficient wet strength to meet daily use is promising but still challenging, especially when renewable cellulose was employed. In this work, a decomposable cellulose-based wet wipe substrate is demonstrated by introducing a synthetic N-vinyl pyrrolidone-glycidyl methacrylate (NVP-GMA) adhesive on the cellulose surface. Experimental results reveal that the NVP-GMA adhesive not only significantly facilitates the chemical bonding between cellulose fibers in the wet state, but also increase the surface wettability and water retention. The as-fabricated cellulose-based wet wipe substrate displays a superb water retention capacity of 1.9 times, an excellent water absorption capacity (completely wetted with 0° water contact angle), and a perfect wet tensile index of 3.32 N.m. g^{-1}. It is far better than state-of-the-art wet toilet wipe on the market (non-woven). The prepared renewable and degradable cellulose-based substrate with excellent mechanical strength has potential application prospects in diverse commercially available products such as sanitary and medical wet wipes.

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1. Introduction

Nowadays, the wet wipes were widely used for toilets [1], makeup removers [2], biomedical wipes [3], napkins and kitchen cleaning paper, etc. [4–6]. The wet wipe products on the market are mainly made of the non-woven, which have some shortcomings in terms of the comfort, the water absorption ratio and the degradability [7–9]. According to a new market research report by Smithers Pira, "2024: The Future of Global Nonwovens", surgical cavity towels and surgical gowns, including other surgical kit components, account for all medical non-woven markets by value and tonnage 67%. In North America, these disposable nonwoven products account for 90% of all surgical towels and 95% of all surgical gowns. As fear of infectious diseases such as COVID-19 or H1N1 avian flu increases, so too will non-woven masks and disposable disinfection wipes. By 2019, the value of global medical nonwovens will reach 1.28 billion dollars, and will continue to grow at a rate of 4.6% per year, and reach a market size of 1.61 billion dollars by 2024. In the medical market, another growth area for nonwovens is wet wipes. Although the non-woven is softer than the conventional wet wipes, its softness is still insufficient, meanwhile, the

low water retention rate, the strict production conditions, and high cost restrict its further wide application [10,11]. Additionally, most of the wet wipes on the market are made of synthetic chemical fibers, some pulp or the viscose fibers, and the main raw materials for synthetic fibers are polypropylene and polyester. Among these raw materials, synthetic fibers are barely degradable, and the viscose fibers are relatively costly [12].

As comparison, cellulose has been extensively used in various daily applications as fabrics, papers or textile substrates [13–16] thanks to their natural and hierarchical supramolecular architecture. Paper is a sheet substrate of interconnected cellulose fibers interconnected by the inter-fiber hydrogen bonds. Furthermore, the cellulose-based product is a flexible material, which have been widely used for information storage, writing, packaging, or specialized purposes, for instance, medical and sanitary wipes due to their favorable properties such as reactivity, lightness, low-cost, biocompatibility, renewability and environmental friendliness [17–21]. However, the intrinsic hydrophilicity of cellulosic paper (due to the surface hydroxyl) can restrict the scope of its application [22]. In the wet state, interactions between the cellulose fibers, namely H-bonding and van der Waals forces [23] are indeed severely weakened resulting in a dramatic decrease of mechanical strength of the cellulosic paper and in the disintegration of the sheet substrate in spite of fibers entanglement. The softening of the fibers stemming from the water uptake may also have an impact on the transfer of local mechanical stress [24]. Overall, it is particularly important to develop a cellulose-based wet wipe substrate with the sufficient wet

strength for industrial application and excellent decomposable performance in terms of daily wet wipes and medical wipes [12].

The raw materials of the decomposable wet wipes should be degradable, which open a new path to prepare wet wipes with cellulose fibers that having good affinity to skins [25,26]. With the recent emergence of COVID-19 worldwide, the demand for medical wipes with skin affinity has increased rapidly [27,28]. Therefore, based on the dual consideration of economy and environmental protection, the biomass-based (cellulose) wipes material that has a suitable wet mechanical strength (in use) and can be efficiently decomposed after using shows a new application prospect in daily use and medical application. The conventional cellulose wet wipe is generally mixed with chemical wet strength agents (such as polyacrylic ester (PAE) [29,30], urea-formaldehyde resin [31–34], etc.) to treat cellulose and increase its wet strength through formation of chemical bonds between the cellulose fibers after heating and drying [35,36]. A study of PAE adsorption on regenerated cellulose reported a coverage of ~3 mg/m^2 [30]. However, PAE is prepared from organic halide epichlorohydrin, which is hazardous [37,38] and difficult to recycle. The biocompatible and non-toxic approach thus represented the major challenging for suitable wet strength of wet wipe.

Herein, cellulose, as the most abundant natural polymer on earth, provides a sustainable green resource that is renewable, degradable, biocompatible and cost-effective [39]. Additionally, cellulose features a linear molecular structure linked by the β(1-4)-D-glucopyranosyl units [40,41]. The highly ordered assembly of cellulose results in microfibrils and cellulose fibers, which afford excellent mechanical properties. With its unique structural and chemical features, cellulose can be converted into the full biomass-based reinforcement materials by the appropriate modification of surface hydroxyl groups. Based on the extensive research foundations of previous researcher, we noticed that the GMA is an important functional monomer among these functional vinyl monomers because of its reactive epoxy group that can react with carboxyl, hydroxyl, and amine functional groups [42]. The GMA-functionalized polymers are good compatibilizers in immiscible polymer blends and are excellent adhesives [43–45]. Additionally, the NVP is a polar, unconjugated and reactive monomer. The homopolymer of NVP, poly (N-vinyl pyrrolidone) (PNVP) has excellent properties including amphiphilic characteristics, excellent biocompatibility, and it is of interest commercially in the commodity chemical and medical fields [46]. Meanwhile, NVP has been grafted efficiently onto polyolefins in the molten state for good adhesion with polar materials [47]. Most importantly, the NVP monomer can copolymerize with GMA easily [48]. More importantly, the previous researchers thoroughly investigated the FT-IR, ^{1}H NMR spectra and structural characterizations about the NVP-GMA copolymers [49–51], which laid the scientific foundation for our follow-up work.

In this work, we described a novel N-vinyl pyrrolidone-glycidyl methacrylate (NVP-GMA) adhesive which contributes the mechanical strength enhancement of the cellulose-based material in the wet state. Meanwhile, the chemical synthesis of NVP-GMA copolymers by solution polymerization method using AIBN (same as this work) as free radical initiator has been extensively studied before [49,52,53]. The NVP-GMA adhesive was introduced into the cellulose fibers to prepare the decomposable cellulose-based wet wipe substrate with suitable wet tensile index by a wet forming method. The resulted decomposable cellulose-based wet wipe achieved a superb water retention capacity, excellent water absorption capacity, and an exceptionally suitable wet tensile index of 3.32 N.m.g^{-1} in wet state, substantially superior to the common wet toilet wipe on the market (non-woven). Therefore, the decomposable cellulose-based wet wipe substrate fabricated by introducing the environmentally friendly adhesive into the cellulose synchronously achieved the ideal water retention and suitable mechanical strength, which thereof provided a promising pathway to other related cellulosic enhancement materials.

2. Materials and methods

2.1. Materials

The sulfate softwood pulp and cotton pulp were produced by the Asia Pacific Semper (Shandong) Pulp & Paper Co., Ltd., Shandong, China. The cationic polyacrylamide (CPAM) was purchased from the Sinopharm Chemical Reagent Co., Ltd. The N, N-dimethylformamide (DMF) and the α,α-azobis-(isobutyronitrile) (AIBN) were obtained from the Tianjin Guangfu Technology Development Co., Ltd., Tianjin, China. The N-vinylpyrrolidone (NVP, 99% purity) and Glycidyl methacrylate (GMA, 97% purity) were received from the Shanghai Maclean Biochemical Technology Co., Ltd., Shanghai, China. The commercially available wet toilet wipe product on the market (non-woven, length: 200 mm & width: 140 mm, and the main raw materials are pure water and flushable non-woven fabric, the common sanitary wipes on the market) was obtained from the Jinjiang Hengan Family Life Paper Co., Ltd., Fujian, China. All chemicals were of analytical grade and directly used without further purification.

2.2. Synthesis of the NVP-GMA adhesive

The GMA (1.42 g, 10 mmol) and NVP (10.00 g, 90 mmol) monomers were dissolved in DMF continuous phase (102.82 g) and then stirred at 500 rpm for 30 min. Then 0.057 g AIBN as free radical initiator was added to the reaction system. The reaction mixture was conducted at 70 °C for 10 h under nitrogen atmosphere, and the synthetic reaction scheme was shown in Fig. 1. After the completion of polymerization, the NVP-GMA copolymer as the aqueous adhesive was obtained when the solvent was removed. Finally, the prepared aqueous NVP-GMA adhesive is stored in the refrigerator for further use.

2.3. Preparation of NVP-GMA cellulose-based wet wipes substrate

In this section of the experiment, softwood fiber (88%, mass ratio) and cotton fiber (12%, mass ratio) were used. Based on empirical data, considering the cellulose content of 94% in the cotton fiber (less concomitant content, more hydroxyl groups can be exposed to react with synthetic adhesives), the cotton fiber dosage of the 12% was selected in experiment section [54]. The pulp fibers were refined according to TAPPI standardized method T 248 sp-08 [55], whereby the dry pulp sheet substrate were soaked in deionized water overnight and then loaded in a PFI (Pulp and Fiber Research Institute) refiner (Test Machines Inc.) and exposed to different levels of refining as defined by the number of revolutions. Additionally, the beating degree of softwood pulp and cotton pulp used in this experiment was 40°SR and 45°SR, respectively.

The cellulose sheet substrate was formed using two methods by processing with water, the difference between the two methods is that the NVP-GMA adhesive was added in different ways. Just as shown in the Fig. 2a, the 0–15% of NVP-GMA adhesive (mass fraction relative to dry pulp) was added by surface coating after the wet sheet substrate is dried at 70 °C for 3 min, then the reacted sheet substrate was continuously dried at 100 °C. Additionally, a certain amount of the NVP-GMA adhesive and the refined pulp suspension were mixed and stirred at 70 °C for 3 min, then is directly dried at 100 °C (the Fig. 2b). The cellulose sheet substrate was formed following the TAPPI standardized method T 205 sp-02, whereby the refined pulp is lowered in consistency and then drained under gravity onto a mesh screen. The cellulose-based substrate is subsequently dried by hot pressing (at 100 °C, 0.1 MPa) on a stainless drying room.
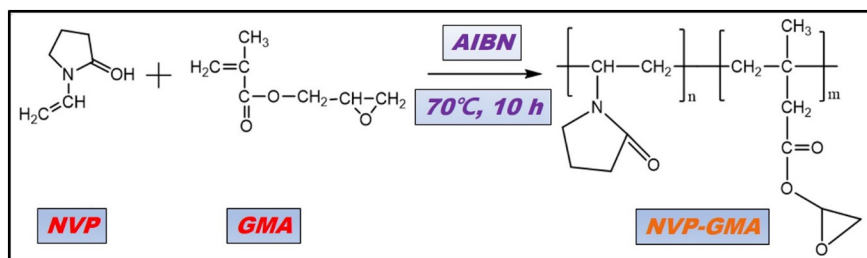
Fig. 1. Chemical reaction schematic of the fabrication process of the NVP-GMA adhesive.

2.4. Characterization

2.4.1. FT-IR spectroscopy of the NVP-GMA cellulose-based wet wipe substrate

The chemical composition and connection of samples (the pristine untreated cellulose-based sheet substrate and the as-prepared NVP-GMA cellulose-based wet wipe substrate) was confirmed by Fourier transform infrared (FT-IR) spectroscopy using a Frontier FT-IR spectrometer (PerkinElmer, Limited by Share Ltd., USA) at the wavelength range of 400–4000 cm^{-1} with a resolution of 1 cm^{-1} in the transmission code. Samples were mixed with KBr powders (w/w = 1/100) and pressed into a disk prior to FT-IR measurement.

2.4.2. Wet tensile index of the NVP-GMA cellulose-based wet wipe substrate

Mechanical test of the NVP-GMA cellulose-based wet wipe substrate was carried out after hanging overnight at 23 ± 0.1 °C temperature and $50 \pm 0.2\%$ relative humidity. The thickness (mm) and basic weight (g/m^2) of the samples were measured before the tensile index test. The wet tensile index of the NVP-GMA cellulose-based substrate (machine direction) was determined using the horizontal tensile testing machine (Frank-PTI Company, Germany). The specimens were cut into 15 mm × 150 mm and the span distance was 25 mm. To measure the wet tensile index, the NVP-GMA cellulose-based substrate was dipped in distilled water for 1 h and then immediately subjected to tensile index measurement. Nine samples were measured for each experiment to give an average result.

2.4.3. Surface morphology of the NVP-GMA cellulose-based wet wipe substrate

Morphologies of the NVP-GMA cellulose-based substrate surfaces were investigated under a scanning electron microscope (SEM, JEOL, JSM-7800F) operated at 25 kV. Gold-palladium (12 mm) was coated on the samples prior to the SEM studies.

2.4.4. Determination of water retention of the NVP-GMA cellulose-based wet wipe substrate

In view of the fact that the wet wipes are commonly used in a wet state, and it is usually required to load the raw materials of aloe, spices and the like on the substrate. Therefore, it is extremely important as a cellulose material of the wet wipe substrate, which is hydrophilic or even water-retaining (usually requires excellent water wetting properties). Generally, the surface water contact angle (WCA) be used to evaluate the hydrophilic and hydrophobic ability of the material. In this experiment, water contact angles of the NVP-GMA cellulose-based substrate (10% relative to the dry pulp, internal addition), pristine cellulosic paper substrate, wet toilet wipe on the market (non-woven), and PAE composite cellulosic paper substrate (1.5% loading is relative to the dry pulp) were measured using an optical tensiometer apparatus (Biolin Science Co., Ltd., Finland). The reported contact angles are averages of five measurements, which were carried out on different areas of each sample. The bouncing process of the 5 μL de-ionized water droplets on the sample surface was record via a high-speed camera at 14 frames per second.

Additionally, the liquid content and water retention capacity are also important quality evaluation indexes due to the wet wipe substrate needs to maintain the wet state for a long time. Therefore, liquid content of the wet wipe sheet substrate was measured according to the method GB/T 27728-2011. The different NVP-GMA cellulose-based substrates were cut to a size of 14 mm × 13 mm, immersed in deionized water for 1 h, and then placed in an oven at 85 °C for 4 h. Three samples were measured for each experiment to give an average result. The liquid content is calculated as follow:
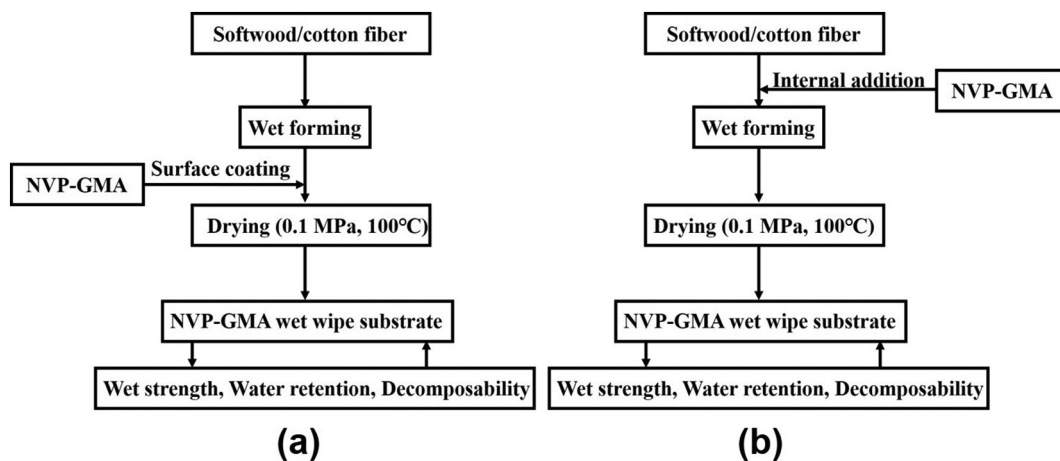

Fig. 2. Preparation methods and characterization flow chart of the decomposable NVP-GMA adhesive cellulose-based wet wipe sheet substrate. a) Surface coating method. b) Internal addition.

$$H = (a-b)/b$$

where: H was the liquid content of NVP-GMA cellulose-based substrate (that is, the ratio of the mass of the water absorbed by the wet wipe cellulose sheet substrate to its own mass, times), a was the mass of the NVP-GMA cellulose-based substrate after being soaked, and the b was the mass of the NVP-GMA cellulose-based substrate after being dried.

2.4.5. Evaluation of the decomposability of the NVP-GMA cellulose-based wet wipe substrate

Considering the convenience during using, it is necessary to investigate the resistance of the prepared NVP-GMA cellulose-based wet wipe substrate to the mechanical force in the wet state (usually affected by the swirling shear force of the water in the toilet during actual use). In order to compare the prepared decomposable properties (post-processing procedures after using) of the NVP-GMA cellulose-based wet wipes substrate with the commercially available similar products. The prepared NVP-GMA cellulose-based wet wipe substrate and the wet toilet wipe on the market (non-woven) were immersed in deionized water for 1 h, and then the surface moisture was absorbed by the absorbent paper, and sealed in the package for 2 h (simulating the wet state of the market wet wipes). After that, the wet toilet wipe and cellulose-based wet wipe substrate were placed in a 1 L beaker containing 500 mL of water and stirred at 1000 r/min with a digital stirrer, and the rotation was stopped every 10 s to observe the decomposition behaviors.

3. Results and discussion

3.1. Effect of adhesives on wet strength of NVP-GMA cellulose-based wipe substrate

The effect of NVP-GMA adhesive on wet tensile index of cellulose-based wet wipe substrate was shown in Fig. 3, the wet tensile index of the NVP-GMA cellulose-based wet wipe substrate increased from 1.11 N.m.g^{-1} (pristine cellulose sheet substrate) to 1.59 N.m.g^{-1} after adding NVP-GMA adhesive to cellulose-based wet wipe substrate (15% loading, relative to dry pulp, drying at 100 °C for 5 min), which achieved an increment of 46.8%. The wet strength of the cellulose-

based wet wipe substrate increased due to the ring opening reaction of epoxy group in the NVP-GMA adhesive at 100 °C, which leads to an intermolecular crosslinking reaction with the hydroxyl groups of cellulose. Furthermore, the formation of an ether bond between the cellulose and the adhesive made the bonding between the fibers more closely [24]. Moreover, the presence of the ether bond maintained the wet strength of the cellulose-based substrate when the cellulose was swollen and the hydrogen bond was destroyed, since the formed ether bond is a covalent bond [56]. Therefore, the wet strength of the obtained NVP-GMA cellulose-based wet wipe substrate was greatly improved compared to the pristine cellulose sheet substrate. Conversely, the wet strength achieved a minor growth after the internal coating as compared to the surface coating when the NVP-GMA adhesive loading was low (less than 5%). It was due to the significant loss of the internally added adhesive during the wet forming process.

The internal addition of 10% adhesive was desired for the following reasons. Firstly, the improvement in the wet strength of the obtained NVP-GMA cellulose-based wet wipe substrate tends to be gentle when the dosage of adhesive changed from 10% to 15%. The enhancement of wet strength could be contributed by the NVP-GMA adhesive and the hydroxyl groups on the cellulose crosslink into a network that achieved the moisturizing effect [57]. Secondly, the radical polymerization reaction may be saturated, and the excess adhesive may not be effectively combined to function as a humidifying agent when the dosage of NVP-GMA adhesive was higher than 10% both for surface coating and internal addition. Considering both the process convenience and the preparation cost, the optimal dosage of NVP-GMA adhesive was chosen to be 10% for the subsequent experiments.

3.2. Effect of drying time on wet strength of the NVP-GMA cellulose-based wet wipe substrate

The drying time in this experiment is a variable worth exploring due to its significant effect of temperature on free radical polymerization reaction has been widely confirmed [58–61]. The experimental conditions were optimized by comparing the wet strength of different drying times with a widely used wet toilet wipe product on the market. The wet tensile index of different materials under different drying times was shown in Fig. 4. Wet tensile index of the NVP-GMA cellulose-based wet wipe substrate was improved gradually when the drying time was prolonged
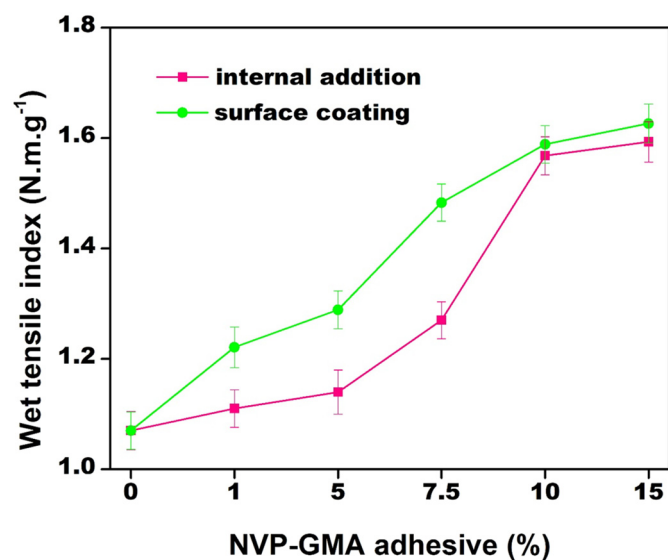

Fig. 3. Effect of NVP-GMA on wet tensile index of the composites. The NVP-GMA adhesive was composited with cellulose fiber by internal and surface coating, the amount of adhesive is relative to dry pulp and all samples were dried at 100 °C for 5 min.
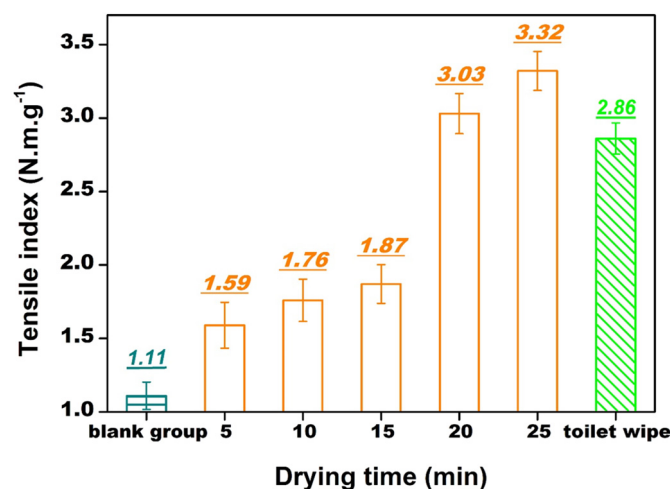
Fig. 4. Effect of drying time on wet tensile index of NVP-GMA cellulose-based wet wipe substrates. Blank group is the pristine cellulose sheet substrate, the NVP-GMA cellulose-based wet wipe substrates were prepared by internal addition (10% NVP-GMA loading, relative to the dry pulp, all samples were dried at 100 °C for different time), the wet toilet wipe product on the market (non-woven, length: 200 mm & width: 140 mm) was obtained from the Jinjiang Hengan Family Life Paper Co., Ltd., Fujian, China.

at 100 °C, jumped from 1.59 N.m.g^{-1} at 5 min to 3.32 N.m.g^{-1} at 25 min in the increments of 108.8%. Long drying time is required because the ring opening reaction of the epoxy group occurred slowly. Meanwhile, the epoxy group gained sufficient energy to undergo ring-opening crosslinking when the drying time was prolonged, which shortened the distance between cellulose fibers in sheet substrate and thus greatly increased the wet strength of the prepared NVP-GMA cellulose-based wet wipe substrate. Excitingly, the wet tensile index of the NVP-GMA cellulose-based wet wipe substrate that was dried at 100 °C for 20 min reached up to the 3.03 N.m.g^{-1} when 10% of the adhesive was added internally, which has exceeded the wet tensile index of wet toilet wipe (2.86 N.m.g^{-1}) on the market.

### 3.3. Surface morphology of the NVP-GMA cellulose-based wet wipe substrate

In order to further explore the wet strength enhancement mechanism of the NVP-GMA cellulose-based wet wipe substrate, the blank group (the same cellulose fiber material, without NVP-GMA adhesive, drying for 25 min) and the optimal NVP-GMA cellulose-based wet wipe substrate were chosen to investigate the surface morphology using a scanning electron microscope. Observed distribution of the loaded NVP-GMA adhesive on the surface of the fiber or the possible connection state with the plant cellulose fiber was shown in Fig. 5. The bonding between the fibers in pristine cellulose sheet substrate is the simple contact connection and winding, and no chemical agents play a role in connecting fibers (Fig. 5a). Furthermore, the hydrogen bonds between cellulose fibers can be easily interrupted by immersing the wet wipe substrate into water. Therefore, the chemically cross-linking was conversed into the physically cross-linking in the form of water bridges, resulting in a significant decrease in binding force [62,63].

However, the different situation can be observed in the NVP-GMA cellulose-based wet wipe substrate (Fig. 5c). The connection between the fibers in NVP-GMA wet wipe substrate and PAE wet strength cellulose sheet substrate (Fig. 5b) are more compact than the connection between the fibers in the pristine cellulose sheet substrate. Meanwhile, some gelatinous substance adhered on the cellulose fiber surface to generate the more chemical link points are produced between the prepared adhesive and cellulose surface thus pulled and fixed the distance

between the fibers, indicating that the NVP-GMA adhesive was successfully embedded on the surface between the fibers. Additionally, the spatial cross-linking network caused by the ether bond formed by the ring-opening reaction made the fiber more tightly connected. The formed ether bond as a covalent chemical bond that is insensitive to water even if the water molecules enter the fibers [64,65]. Therefore, the ether bond formed between the NVP-GMA adhesive and the hydroxyl group on the fibers surface can still maintained the stability and wet strength of the wet wipe substrate even if the hydrogen bond was destroyed, and thus the wet strength of the NVP-GMA cellulose-based wet wipe substrate be greatly improved. Besides, to exemplify the viability and superiority of the synthetic adhesive, SEM image of the wet toilet wipe on the market (non-woven) was investigated after hanging overnight in a constant temperature and humidity laboratory (temperature: 23 ± 0.1 °C, relative humidity: 50 ± 0.2%) to remove excess moisture. The wet toilet wipe on the market (non-woven) was made of non-degradable chemical synthetic fibers, which showed the smooth cylindrical surface (Fig. 5d) and excellent mechanical properties due to the inherent excellent mechanical properties of chemically synthesized fibers [66,67].

3.4. Hydrophilicity and water retention capacity of the NVP-GMA cellulose-based wet wipe substrate

In order to further explore the effect of the NVP-GMA adhesive on the hydrophilicity and water retention of wet wipe substrate, the water contact angle of pristine cellulose sheet substrate, NVP-GMA cellulose-based wet wipe substrate and PAE enhancement paper were investigated as shown in Fig. 5. The pristine cellulose sheet substrate was wettable with the contact angle of 16° (Fig. 5a), while the NVP-GMA wet wipe substrate was completely wettable due to its contact angle picture could not be taken by the optical contact angle measuring instrument (Fig. 5c, water droplets instantly soaked the paper without angle formed, indicating the contact angle did not exist). Therefore, the NVP-GMA cellulose-based wet wipe substrate was more hydrophilic than the pristine cellulose sheet substrate. The superior wettability can be attributed to the high polarity and hydrophilicity of the NVP, and thus the load of NVP-GMA adhesive could significantly increase the hydrophilicity of the carrier [68]. Furthermore, the carbonyl group of the NVP and the ether bond formed by the ring-opening and subsequent
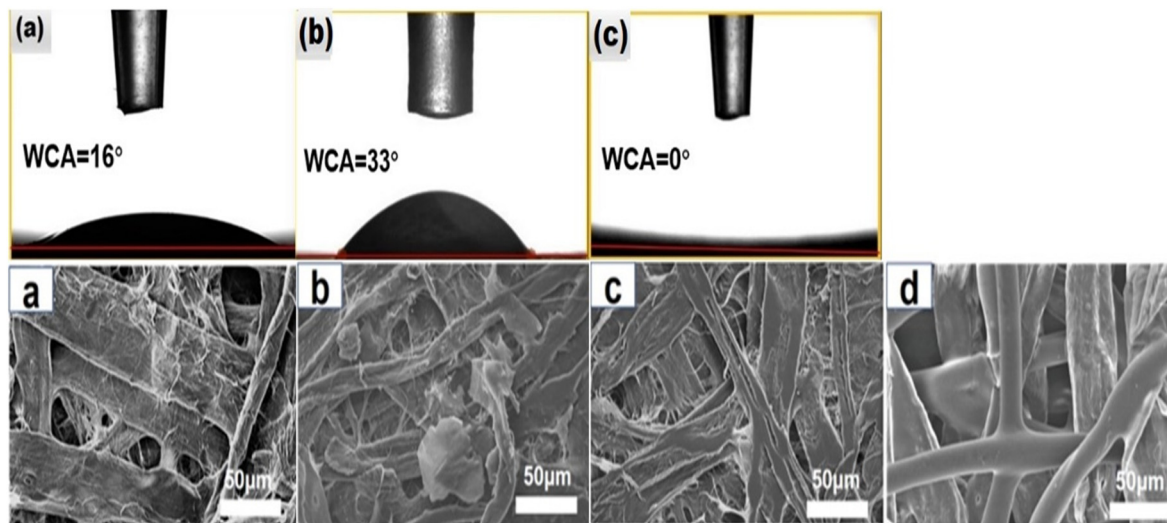
Fig. 5. Water contact angle pictures and SEM images of different wet wipe substrate surface. a) The pristine cellulose sheet substrate (88% softwood fiber, 40°SR, 12% cotton fiber, 45°SR, drying at 100 °C for 25 min). b) Traditional wet strength agent enhancement paper (1.5% PAE loading is relative to dry pulp). c) The NVP-GMA cellulose-based wet wipe substrate (10% loading is relative to dry pulp, internal addition, drying at 100 °C for 25 min). d) The wet toilet wipe on the market (non-woven, length: 200 mm & width: 140 mm) was obtained from the Jinjiang Hengan Family Life Paper Co., Ltd., Fujian, China.

crosslinking reaction of the epoxy group, both are of hydrophilic feature. These structures introduced more hydrogen bonds with water molecules, thereby improving the hydrophilicity of the NVP-GMA adhesive wet wipe substrate.

The wet wipes are used in a wet state. Even if exposed to the air, it is required to keep a certain amount of water for cleaning and other purposes. We thus measured the liquid content of the prepared NVP-GMA cellulose-based wet wipe substrate at high temperature for a specified period of time according to the method in GB/T 27728-2011. The liquid content of NVP-GMA cellulose-based wet wipe substrate is H = $(2.0 + 2.0 + 1.8) / 3 = 1.9$ times, which satisfies the requirement of liquid content in GB/T 27728-2011 (H > 1.7). Therefore, by adding synthetic NVP-GMA adhesive, the prepared wet wipe substrate satisfied the combined effect of hydrophilic, water retention, and suitable wet strength.

3.5. Decomposability of the NVP-GMA cellulose-based wet wipe substrate

Conventional wet wipes are non-dispersible and non-degradable due to the addition of chemical fibers or nonwoven fabrics [7,69,70]. By introducing the synthetic NVP-GMA adhesive, the resultant NVP-GMA cellulose-based wet wipe substrate is successfully made of the pure natural cellulose fibers, and thus the final degradation can be achieved. The prepared NVP-GMA cellulose-based wet wipe substrate and wet toilet wipe on the market (non-woven) were taken to evaluate the decomposability, the disintegration of the wet wipe substrate observed during the stirring process was used as an evaluation standard, meanwhile, the deformation processes and results are shown in Fig. 6 (dynamic details in Videos S1a and S1b). The prepared NVP-GMA cellulose-based wet wipe substrate started to break down in 10 s, and ruptured to 4 fragments at 20 s. Further disintegration of the wet wipe substrate formed the cracked debris in 30–60 s (Video S1a). Conversely, the wet toilet wipe (non-woven) remained intact in 0–60 s without any breakage (Video S1b). Therefore, the prepared NVP-GMA cellulose-based wet wipe substrate achieved obvious decomposability compared with the wet toilet wipe (non-woven).

3.6. Mechanism comparison between NVP-GMA adhesive and PAE wet strength agent

The proposed synthesis procedure of the NVP-GMA aqueous adhesive [51] was shown in Fig. 1. In order to fully investigate how the NVP-GMA adhesive to crosslink with cellulose fiber and to compare the difference between the synthetic green adhesive and traditional PAE wet strength. Fig. 7 shows the Fourier transform infrared (FT-IR) spectrum of the pristine untreated cellulose-based sheet substrate and prepared NVP-GMA cellulose-based wet wipe substrate. In spectrum a (dark line, the pristine untreated cellulose-based sheet substrate), the bands located at 3445, 2911 cm^{-1} could be assigned to -OH stretching and CH$_2$ groups, respectively. Markedly, the band at 1637 cm^{-1} (spectrum a & b) is due to the bending mode of the absorbed water. Additionally, for pristine cellulose, the ether bond has a wide and complex absorption band at 1200–950 cm^{-1}, but for the NVP-GMA cellulose-based wet wipe substrate (spectrum b, red line), a sharper peak (with higher peak intensity) at 1078 cm^{-1} corresponds to the ether bond: (1) ether bond of cellulose itself and (2) the ether bond formed by the reaction between NVP-GMA and the cellulose surface hydroxyl group. Meanwhile, compared with the untreated pristine cellulose-based sheet substrate, new band emerged at 1728 cm^{-1} in NVP-GMA cellulose-based wet wipe substrate (spectrum b), which was attributed to the stretching vibration of the C=O groups introduced by the reaction of NVP-GMA and cellulose. Consequently, as the reaction mechanism illustration shown in Fig. 8, the wet strength enhancement of cellulose is mainly due to the ether bond formed by the ring opening reaction of NVP-GMA adhesive and the two different hydrogen bonds (including the hydrogen bond formed between the hydroxyl groups on the

surface of the cellulose and the hydrogen bond formed by the hydroxyl group on the cellulose surface with the carbonyl group on the NVP-GMA adhesive) to form a crosslinked network, which made the bond between the cellulose fibers more compact. Conversely, compared with the prepared aqueous adhesive, the cationic azetidinium groups and protonated secondary amine groups promote PAE adsorption onto cellulose fiber surface [30,71,72]. The reactive azetidinium forms covalent grafts with carboxyl groups on fiber, and crosslinks within and between PAE chains during heating and drying [29]. The result is a crosslinked polymer networks between contacting fiber surfaces. Cured PAE network are somewhat hydrophobic showing little tendency to swell or hydrolyze in water [35]. This insensitivity to water maintains fiber-fiber joint strength when cellulose sheet substrate is exposed to water, which is highly consistent with our water contact angle results illustrated in Fig. 5b, c (the contact angle of PAE enhancement cellulose sheet substrate is 33° compared to the completely wettable NVP-GMA cellulose-based wet wipe substrate). Furthermore, the PAE preparation process is toxic compared to synthetic NVP-GMA adhesive in this study [37]. Therefore, the prepared NVP-GMA adhesive wet wipe substrate not only obtained excellent hydrophilic property and water retention capacity, but also met the criteria for daily use such as decomposability and degradability.

3.7. Evaluation of cellulose-based composites toxicity as wet wipe substrate

The main components of the composite material are: cellulose (green and non-toxic), NVP/GMA (unreacted part), and the generated NVP-GMA adhesive. Firstly, there have been studies using NVP as a monomer for the synthesis of highly hydrophilic colored contact lenses due to its non-toxic and excellent affinity with human tissues. Secondly, the GMA can also be used as a raw material for medical selective filtration membranes, anti-coagulation agents, dental materials, non-soluble adsorbents, etc. Therefore, the GMA can be used to prepare medical wipes substrate due to its relatively stable biocompatibility and non-toxicity in the material [52].

Finally, in view of the synthetic NVP-GMA adhesive, there are already researchers reposted the hydrophilic porous magnetic poly (GMA-MBAA-NVP) composite microspheres containing oxirane groups as an efficient carrier for immobilizing penicillin G acylase [53]. Tyagi et al. synthesized GMA-co-NVP copolymers with various GMA:NVP ratios and dip coated it onto polypropylene (PP) strips [49]. All the polymer coated PP strips were evaluated for their application in dot-ELISA (enzyme-linked immunosorbent assay) using a model system, showing that the best results were obtained with GMA-NVP copolymer of 78:22 mol% ratio of GMA and NVP respectively, coated onto the PP strips. The developed system is highly specific, showing no non-specific reactions either in model system or with blood samples. Moreover, these polymer coated strips have a strong potential to be used as matrix in dot-ELISA for detection of various diseases to be performed in the field conditions and in minimally equipped laboratories. Interestingly, the above-mentioned practical applications of the NVP-GMA are based on enzyme carriers or immunoadsorption assays, which has a very high requirements for the toxicity and biocompatibility of composite materials. Therefore, the safety and toxicity of the prepared cellulose-based wet wipe substrate basically meets the production requirements.

3.8. Industrialization prospect of the NVP-GMA cellulose-based wet wipe substrate

The prepared cellulose-based wet wipe substrate as the main material of commercial wipes, which is usually add a certain amount of the RO pure water, aloe essence, and antibacterial agent to finally package and sale. In view of the comprehensive consideration of environmental and economic benefits, we will discuss the feasibility of its commercial application in two aspects.
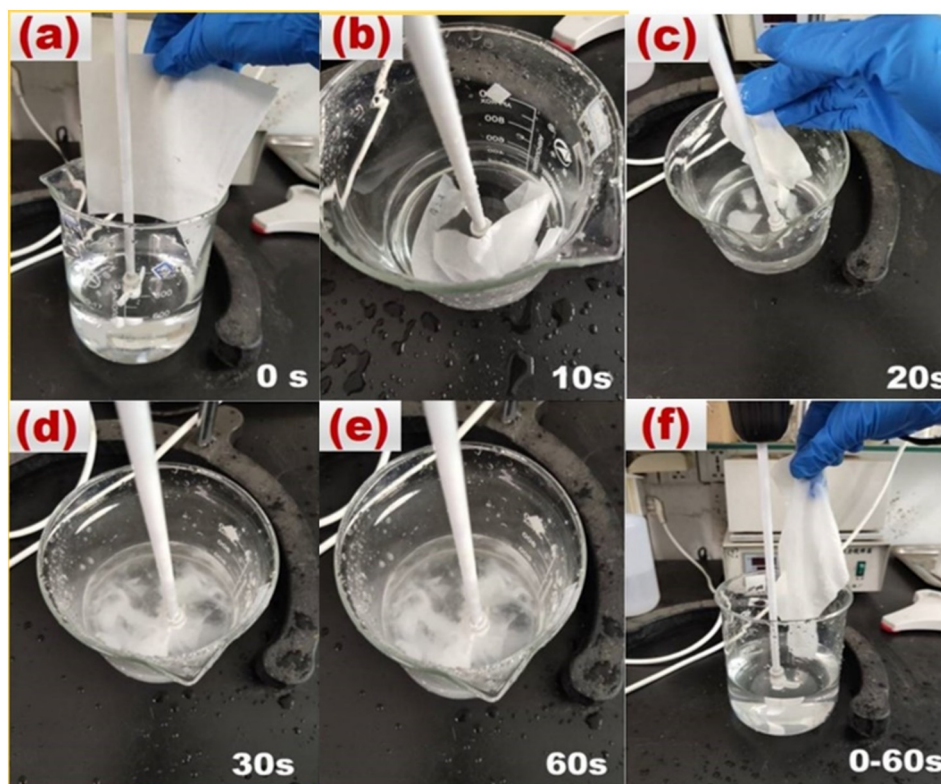
Fig. 6. Photographs of different cellulose wet wipe sheet substrates which were soaked in distilled water then mechanically stirred. a–e) The prepared NVP-GMA cellulose-based wet wipe substrate stirred for 60 s. f) The commercial wet toilet wipes on market (non-woven) stirred for 0–60 s, respectively.

With regard to economic benefits, the difference between our cellulose-based wet wipe substrate and other commercially available wet wipes products is the raw materials of wet wipe substrate due to the economic cost of the additives in various wet wipe is basically equivalent. Non-woven textiles mainly comes from the base material of many sanitary products such as the wet wipes and sanitary towels [73]. A recent study identified the material composition of "baby/wet wipes" as white micro-polyethylene terephthalate (PET) fibers used in the manufacture of products labelled as flushable and other components such as high-density polyethylene (HDPE) and polyethylene/vinyl acetate (PEVA/EVA) [74]. Meanwhile, compared with our cellulose-based wet wipe substrate, the commercially available non-flushable wipes are manufactured from PET, polypropylene (PP), or a combination of PET and cellulose. Moreover, 50% of brands labelled flushable are comprised of a mixture of PET and cellulose and the remainder of cellulose alone. Cellulose-based materials being economical, because the cellulose is one of the most abundantly present natural, and renewable biomacromolecule polymers on earth. In contrast, the strength of chemical synthetic fibers such as PET, PP, etc. satisfies the requirements as a wet wipe substrate, but their preparation cost is much higher than cellulose-based materials. Therefore, the prepared cellulose-based materials as the commercially available wet wipe substrate are more economical when the wet strength and water retention capacity are equivalent.

Considering environmental benefits, lignocellulosic materials being green, sustainable, and biodegradable [75,76]. This renewable material also facilitates its applicability as an environmentally friendly wet wipe substrate for achieving the large-scale commercial production, increasingly investigated both in research and industry [77–79]. Besides, the ubiquity of microplastic fibers degraded from widely flushed personal care textile products (wet wipes and sanitary towels) were comprised of PP and polystyrene [73], which confirms that wet wipes (non-cellulose substrate) and sanitary towels flushed down to toilets are an underestimated source of white microplastic fibers in the environment.

Therefore, compared with the chemical synthetic fibers-based wet wipe substrate, the biodegradable cellulose-based wet wipe substrate has more long-term environmental benefits in commercial applications. In summary, the cellulose-based wet wipe substrate prepared in this work has a foreseeable possibility of commercial application.
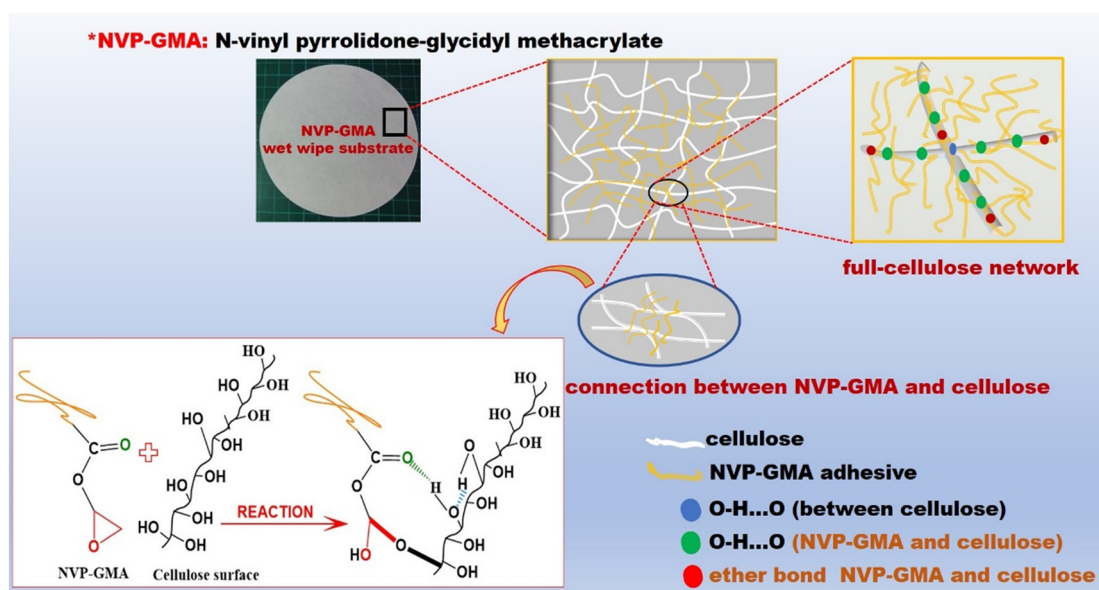
4. Conclusion

In this study, the decomposable wet wipe substrate with suitable wet strength was successfully prepared with the renewable, degradable
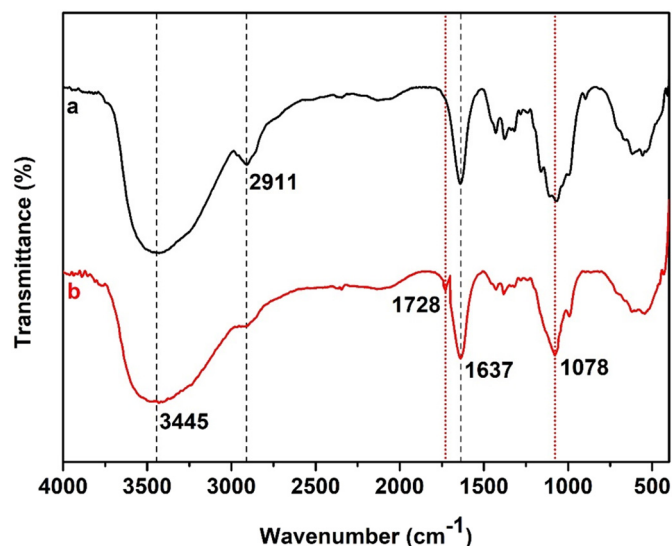
Fig. 7. FT-IR spectra of the different substrate samples. a) The pristine untreated cellulose-based sheet substrate. b) The as-prepared NVP-GMA cellulose-based wet wipe substrate.

Fig. 8. Enhancement mechanism illustration of the NVP-GMA adhesive for reinforcing wet strength of cellulose-based materials. The formation process of ether by ring-opening reaction of NVP-GMA adhesive with the hydroxyl groups on the cellulose surface.

cellulose fiber by introducing the synthetic NVP-GMA aqueous adhesive. The result revealed that the as-fabricated NVP-GMA cellulose-based wet wipe substrate exhibited excellent hydrophilic properties, good water retention capacity, and suitable wet tensile index. Interestingly, their mechanical properties could be controlled effectively by adjusting the dosage of NVP-GMA adhesive. For the first time, the ideal wet wipe substrate has been prepared by blending green cellulose polymer with synthetic NVP-GMA adhesive in wet state, and the controllable interaction between cellulose and adhesive existed in the composite cellulose-based wet wipe substrate. Meanwhile, the prepared NVP-GMA cellulose-based wet wipe substrate could be completely broken at 30 s under 1000 r/min when it was soaked into 500 mL distilled water compared to the intact wet toilet wipe after the same treatment. More encouragingly, the prepared NVP-GMA adhesive wet wipe substrate achieved higher water retention (1.9 times) and wet tensile index (3.32 N.m.g^{-1}) than the commercial wet toilet wipe (2.86 N.m. g^{-1}). The NVP-GMA adhesive wet wipe substrate was safe, regenerated, as well as degradable and simultaneously balanced the decomposable behavior and suitable wet tensile mechanical strength, which thus provided a new pathway for producing renewable cellulose-based wet wipe substrate. Therefore, the novel green NVP-GMA adhesive will find wide applications in the fields of hygiene, medical care, and other regenerated cellulose-based functional materials.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

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