



Double etherification of corn starch to improve its adhesion to cotton and polyester fibers



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ABSTRACT

In order to investigate the impact of double etherification on the adhesion to fibers and desizability of corn starch, a series of doubly etherified corn starch (DES) with the total degree of substitution (DS) values of 0.014–0.034 were prepared by simultaneous quaternization and sulfonation of acid-converted corn starch (ACS) with *N*-(3-chloro-2-hydroxypropyl) trimethylammonium chloride (CHPTMA) and 3-chloro-2-hydroxy-1-propanesulfonic acid sodium salt (CHPS-Na). The influence of double etherification on the adhesion of starch to cotton and polyester fibers was evaluated. It was found that etherification was able to increase the tensile strengths of starch to the fibers, and the strengths gradually increased as the etherification levels rose. Moreover, etherification was capable of displaying a positive effect on the desizability of starch as determined by measurements of swelling capacity of DES films and the time required to break the films. Etherification also enhanced the paste stability and moisture regain of the films. The DES with a total DS value of 0.034 showed potential for ameliorating the adhesion of starch to cotton and polyester fibers for warp sizing.

1. Introduction

Starch as a polysaccharide consisting of anhydroglucose units (AGU) has been widely used either alone or in blends with other substances as wet-end additives and surface sizing agents in paper making, and as warp sizing agents in textile applications owing to its advantages of renewability, biodegradability, abundant resource, and cheapness [1]. In general, good adhesion to fibers is believed essential for all kinds of starch-based size materials [2]. This is because serviceability of starch relies on its adhesion strength to fibers. The adhesion exhibits the function of increasing the strength of fibrous goods such as papers and sized warps by bonding the fibers in the goods together [3]. Besides, in warp sizing, the adhesion is not only capable of exhibiting the function of diminishing the yarn hairs by adhering the hairs back onto the yarn body, but also capable of strengthening the bonding fastness between yarn body and starch film (covered on the yarn surfaces) [4]. Accordingly, adhesion affects the physical and mechanical properties of sized goods and determines the size pick-up of warps [5]. For this reason, adhesion has received considerable attention, and a large body of work has already been reported [6–8].

Nevertheless, starch shows the feature of brittleness due to

shortcomings like its cyclic structure and numerous hydroxyls. The brittleness results in a low adhesion of starch to fibers. Therefore, a reduced serviceability of starch during sizing use has been displayed owing to its insufficient adhesion to fibers [9]. Consequently, there is an urgent need to overcome the drawbacks (such as insufficient adhesion) of starch so as to enhance its serviceability in applications such as warp sizing and paper-making. Undoubtedly, one alternative for solving the drawback is to introduce a certain number of groups by starch modifications such as cationization and anionization [10–12]. However, anionization introduces negatively charged groups into starch molecules, and the surfaces of cellulose fibers in water also show negative charges [13]. The repulsion induced in this situation will adversely affect the adhesion of starch to fibers. Undoubtedly, cationization can show a positive influence on adhesion by electrostatic attraction at the adhesive layer-fiber interfaces between positive starch and negative fibers, but the attraction will increase the difficulty of removing cationic starch from sized yarns [14]. Moreover, when cationic starch is used together with negative auxiliaries such as lubricants and defoamers, it may result in serious problems such as paste flocculation, etc [15]. Fortunately, cationic and anionic groups can be derivatized onto starch chains by a two-step method [16]. There is no doubt that

Abbreviations: ACS, Acid-converted cornstarch; CHPTMA, *N*-(3-chloro-2-hydroxypropyl) trimethylammonium chloride; CHPS-Na, 3-chloro-2-hydroxy-1-propanesulfonic acid sodium salt; DES, double etherified corn starch; DS, degree of substitution; TMACHP, 3-(trimethylammonium chloride)-2-hydroxypropyl; PSH, 3-propanesulfonic-2-hydroxy; AGU, anhydroglucose units

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amphoteric starch containing both negatively and positively charged groups is likely to minimize the insufficiencies of cationic and anionic starches. For example, 3-(trimethylammonium chloride)-2-hydroxypropyl (TMACHP) and 3-propanesulfonic-2-hydroxy (PSH) groups can be used as cationic and anionic substituents, respectively, for introduction into starch molecules by a one-step method, i.e., double etherification. The two groups are hydrophilic and can enhance the absorption of water into adhesive starch layers (formed from cooked starch paste) between the fibers. In addition, these groups have a large space volume and will generate strong steric restriction. This can obstruct the regular arrangement of starch amylose chains, and partly prevent hydrogen bonding between starch hydroxyls during the formation of starch adhesive layers. The water absorbed [17] and obstruction generated by the hindrance [18] are able to provide plasticization for the layers, thereby reducing internal stresses and their concentration [19]. Accordingly, it is as well to emphasize that an enhanced adhesion of starch to fibers by the introduction of the two groups may be expected. For these reasons, TMACHP and PSH groups were chosen and introduced into corn starch chains via double etherification, i.e., quaternization and sulfonation. The chemical structure of doubly etherified corn starch (DES) is presented in Fig. 1. Currently, there is no literature about the preparation of DES, and the preparation performed by the one-step reaction will shorten the reaction time and lower the cost.

However, little is known about the influence of double etherification on adhesion to fibers because neither investigation on the impact of etherification upon adhesion has been conducted nor was there evidence that revealed that adhesion could be improved by means of etherification. Therefore, the objectives of this work were to determine if etherification could ameliorate adhesion, and to analyze the influence of the total degree of substitution (DS) on adhesion since the usability of a modified starch depends on the amount of the groups introduced, i.e., DS [20]. In addition, due to the importance of desizability in warp sizing, the effect of etherification on desizability should also be estimated. Measurements of adhesion and desizability were performed not only on DES, but also on acid-converted starch (ACS). Such an experimental arrangement enabled us to determine the influence of the groups themselves from the experimental data.

2. Materials and methods

2.1. Materials and reagents

Food grade corn starch with 11.7% moisture and a viscosity of 53 mPa·s, was used as a raw material in this research and purchased from

Shandong Hengren Industry & Trade Co. Ltd. (Shandong, P. R. China). Industrial *N*-(3-chloro-2-hydroxypropyl) trimethylammonium chloride (CHPTMA, 60% aqueous solution) was procured from Aladdin Industrial Corporation (Shanghai, P. R. China) and used without further treatment. 3-chloro-2-hydroxy-1-propanesulfonic acid sodium salt (CHPS-Na) was obtained from the Jiaying Sicheng Chemical Co. Ltd. (Zhejiang, P. R. China). All other chemicals, such as sodium hydroxide, anhydrous sodium sulfate, hydrochloric acid, methanol, and ethanol, were analytical grade and obtained from Sinopharm Chemical Reagent Co. Ltd. (Shanghai, P. R. China).

Pure cotton roving (460 tex, twist factor: 112) and polyester roving (396 tex, twist factor: 49.8) employed for adhesion measurements, were kindly supplied by Talak Cotton Factory (Jiangsu, P. R. China) and Yizheng Chemical Fiber Co. Ltd. (Jiangsu, P. R. China), respectively. The length of the fibers in both rovings was 29 and 38 mm, respectively. And the fineness was 1.72 and 1.73 dtex, respectively.

2.2. Double etherification of corn starch

Before the double etherification process, the starch was suspended in a methanol-distilled water mixture in a 1000 mL beaker and continuously stirred at 40 °C for 1 h to remove its protein components [21]. Then, the starch was acid-converted with hydrochloric acid so as to reduce apparent viscosity [22] via hydrolysis of glucosidal bonds between AGU in starch chains.

ACS was reacted with CHPTMA and CHPS-Na simultaneously in aqueous medium for preparing the DES. Briefly, a 40% (w/w) starch aqueous dispersion was prepared by dispersing 324 g of dried ACS in distilled water containing 32.4 g of Na₂SO₄. Na₂SO₄ was used to prevent the swelling of starch granules. The dispersion was transferred into a 1000 mL four-necked round-bottom flask that was immersed in a water bath. Thereafter, a 3% (w/w) NaOH solution was added into the dispersion to adjust its pH to 10–11. Then, the dispersion was constantly stirred and heated to 45 °C in the water bath. The mixture of CHPTMA, CHPS-Na and NaOH solution (the total mole quality of CHPTMA and CHPS-Na was equal with NaOH) was added immediately into the dispersion. Subsequently, the pH of the dispersion was adjusted to 11–12.5 using 3% NaOH solution, and the reaction was carried out at 45 °C for 8 h under mechanical agitation. Finally, the rough product was neutralized to pH = 6.5–7 using a 2 M HCl standard solution, and then filtered and washed with an ethanol/distilled water mixture for at least 3 times. The resulting DES was dried in an oven at 45 °C for 12 h, pulverized, and sieved with a 100-mesh sieve.

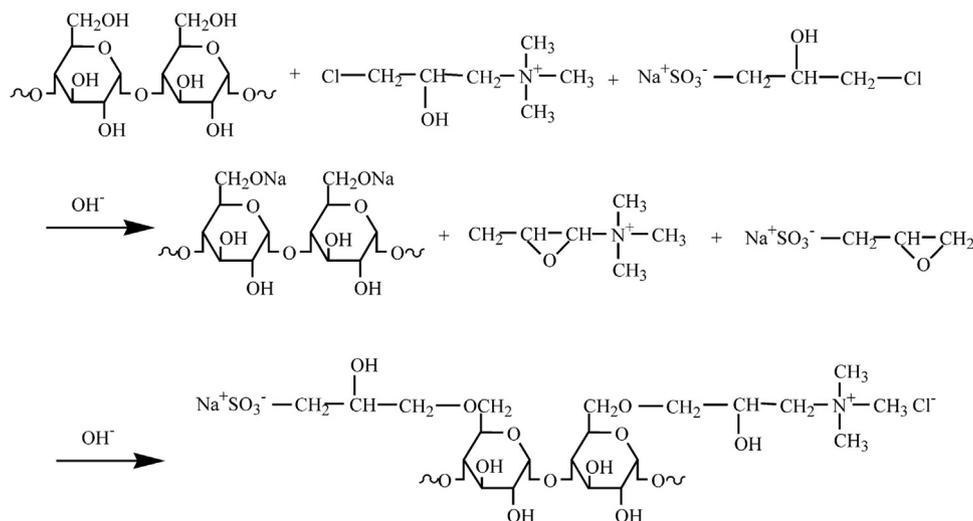


Fig. 1. Double etherification of starch for the preparation of DES.

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