Fibers and Polymers 2019, Vol.20, No.5, 906-912
DOI 10.1007/s12221-019-8865-4

ISSN 1229-9197 (print version)
ISSN 1875-0052 (electronic version)

N-halamine Antibacterial Cellulose Fabrics Functionalized with Copoly(acrylamide-maleic anhydride)

Qunaquan Wang, Xingchi Zhu, Ping Zhu*, and Zhiming Jiang*

Institute of Functional Textiles and Advanced Materials, College of Textile & Clothing, Growing Base for State Key Laboratory of New Fiber Materials and Modern Textile, Collaborative Innovation Center of Marine Biomass Fibers, Materials and Textiles of Shandong Province, Qingdao University, Qingdao 266071, China
(Received September 19, 2018; Revised January 15, 2019; Accepted February 20, 2019)

Abstract: A copolymer of acrylamide and maleic anhydride (PAMA) was prepared and coated onto the surface of cotton fabrics through traditional pad-dry-curing process. After exposing to dilute bleaching solution, the amide groups can be transformed to acyclic N-halamine. The optimum finishing condition was obtained by investigating the effect of PAMA concentration, catalyst concentration and curing temperature on the chlorine content loaded onto cotton fabrics. The chlorinated cotton fabrics coated with PAMA showed good antibacterial properties against *S. aureus* and *E. coli*, and stable washing efficacy against repeat washing. Also, the anti-wrinkle properties and air permeability can be definitely improved after coating with PAMA.

Keywords: Acrylamide, Maleic anhydride, Copolymer, N-halamine, Antibacterial

Introduction

Recently, antimicrobial textile products have become increasingly popular in the market due to consumers' demand for hygienic clothing and activewear [1,2]. There are different classifications of antimicrobial agents applied to textile according to their efficiencies, mechanisms of antimicrobial activity and washing resistance [3]. It has been reported that quaternary ammonium salts [4], N-halamine compounds [5-13], triclosan [14,15] and chitosan [16] have perfect antimicrobial activities in the textiles. Among these different kinds of antimicrobial agents, N-halamine compounds have been extensively studied due to their stabilities, regenerabilities, and efficacies in inactivating bacteria [6].

N-halamines are organic and inorganic compounds containing one or more covalent bond between nitrogen and halogen (N-X) [1]. In deactivating microorganism, the N-halamine bond can be reversibly reacted to N-H. However, the inactive substance can be recharged to N-Cl in a dilute sodium hypochlorite solution. In principle, there are three types of N-halamine structures: imide, amide, and amine [17,18]. According to previous research, the stabilities toward dissociation of the N-Cl moieties are in the order amine > amide > imide halamine. However, N-halamine exhibits better biocidal activity with high dissociation constant.

Acyclic acrylamide containing an amide group is a kind of N-halamine precursor, which can be converted to biocidal N-halamine structures similar to cyclic N-halamine compounds. Acrylamide can be easily incorporated into polymer materials via grafting method with the assistence of initiator such as sodium/potassium persulfate, dicumyl peroxide, and ammonium ceric nitrate. In the previous research, some antibacterial materials were prepared by grafting acrylamide onto the surface of cotton fabrics, kevlar fabrics, polyurethane tube [19-21]. The resultant antibacterial materials showed potent, durable, and rechargeable biocidal activities against gram-negative bacteria, gram-positive bacteria, fungi, and virus, which suggests that acrylamide has great potential in antibacterial materials.

In the past researches, the preparation process of N-halamine antibacterial agent is relatively complicated and longer. And many of N-halamine compounds cannot be dissolved in water and noxious organic solvent should be used to prepare antibacterial textiles. This study is the extending of N-halamine antibacterial textiles and expects to improve washing stability and applies in industrial application. In this study, a water-soluble copolymer of N-halamine precursor was synthesized with acrylamide and maleic anhydride, and applied in antibacterial cellulose fabrics by pad-dry-curing process. The effect of PAMA concentration, SHP concentration and curing temperature was investigated and the optimum finishing condition was obtained. After exposing to dilute sodium hypochlorite solution, the coated fabrics showed good antibacterial properties against *S. aureus* and *E. coli*, and the active chlorine provided perfect washing stability. Besides, the anti-wrinkle properties and air permeability had a little improvement after coating with PAMA.

Experimental

Materials

Desized, scoured and bleached cotton fabric was provided from Weifang Qirong Textile Co., Ltd. (Weifang, China).

*Corresponding author: pzhu99@163.com
*Corresponding author: jzm070315@163.com

Acrylamide and maleic anhydride were purchased from Xiya Reagent (Chengdu, China). Potassium persulfate and sodium hypochlorite were obtained from Tianjin Hengxing Chemical Reagent (Tianjin, China). Sodium hypophosphite and potassium iodide were bought from Tianjin Beilian Fine Chemical Co., Ltd. (Tianjin, China) and Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China), respectively. All reagents were used without further purification.

Copolymerization of PAMA

PAMA copolymer was synthesized in aqueous solution at 75 °C using potassium persulfate as initiator, and synthesis scheme was presented in Figure 1. Briefly, acrylamide (0.2 mol, 14.2 g) and maleic anhydride (0.2 mol, 19.6 g) were placed in three-neck round flask equipped with a circumference condenser, thermometer and nitrogen insert. After completely dissolving in 200 m*l* water, the temperature was increased to 75 °C and then nitrogen was bubbled through the solution for 10 min to remove any dissolved oxygen. After adding potassium persulfate (1 ‰ relative to the solution), the mixture was stirred for 1 h at 75 °C. The resulting copolymer was obtained by evaporating the solvent and a viscous product was received with a yield of 93 %. FTIR-ATR v (cm^{-1}): 3539 (N-H, O-H), 2944 (C-H), 1711 and 1674 (C=O), 1610 (amide II). The stretching vibration of the vinyl bonds of the monomers at around 1640 cm^{-1} disappeared for the copolymers.

Fabrics Treatment

As shown in Figure 2, the cotton fabrics were coated with PAMA through pad-dry-curing process. 12 cm×12 cm of bleached cotton fabrics were soaked in aqueous solutions containing PAMA (2 %, 4 %, 6 %, 8 %. 10 %) and sodium hypophosphite (1 %, 2 %, 3 %, 4 %, 5 %) at a ratio of 1:20 (w/w) for 15 min. Then the fabrics were padded through the nip to reach wet pick-up of 100 %. The samples were treated with two dips and two nips. Next, the fabrics were dried at 100 °C for 5 min, and cured at different temperature (150 °C, 160 °C, 170 °C, 180 °C) for 5 min. Subsequently, the finished samples were rinsed with 0.5 % detergent solution for 10 min, and throughly washed with tap water and dried at 70 °C for 1 h.

Chlorination and Quantitative Titration

To render the cotton fabrics with high antibacterial properties, the coated cellulose fabrics were treated with bleaching solution and the process was shown in Figure 3. Briefly, the commercial sodium hypochlorite solution was diluted with 10 times, and pH value was adjusted to 7 by adding 1 M hydrochloric acid solution. Then the coated cotton fabrics were immersed in the prepared solution for 1 h at room temperature. Subsequently, the treated samples

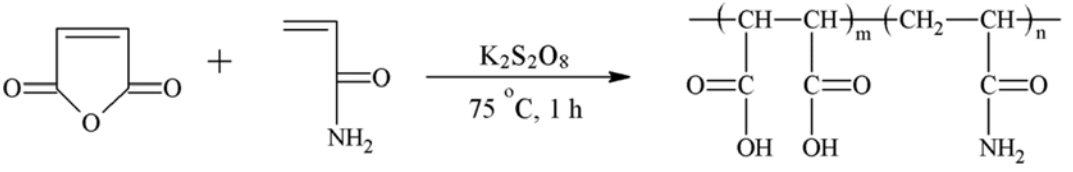

Figure 1. Synthesis scheme of PAMA.

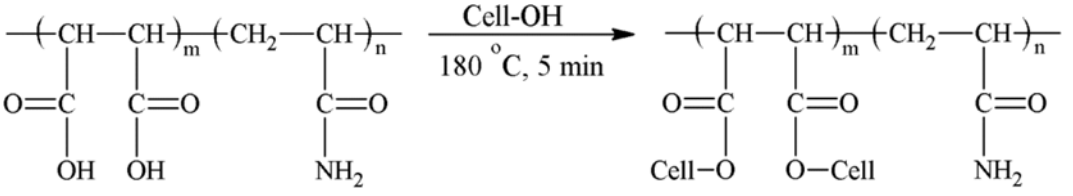

Figure 2. Coating process of PAMA onto cellulose fabrics.

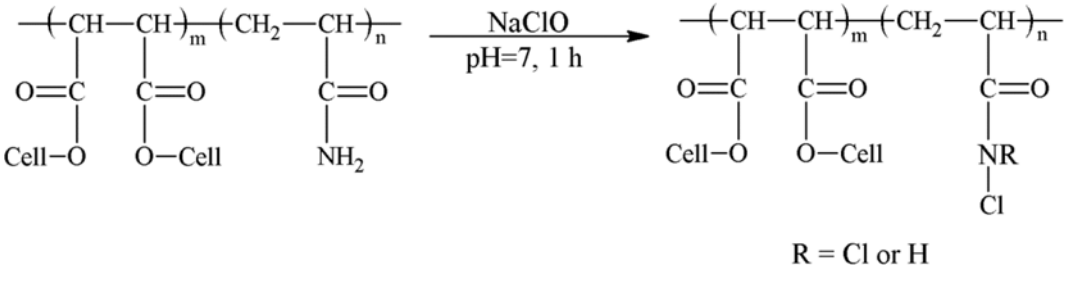

Figure 3. Chlorination scheme of PAMA-coated fabrics.

were washed throughly with tap water and dried at 45 °C for 1 h.

The oxidative chlorine content on the cotton fabrics can be determined by iodometric/thiosulfate titration method. Detailedly, about 0.2 g above coated cotton fabrics was dispersed in 20 m*l* deionized water, followed by adding about 0.2 g of potassium iodide and ten drops of 0.5 % starch solution. Then the mixture was titrated with 0.001N sodium thiosulfate until the solution turned to colorless. The content of oxidative chlorine can be calculated according to following formula:

$$Cl^{+}\% = \frac{35.45 \times N \times V}{2 \times W} \times 100$$

where $Cl^{+}\%$ is the content of oxidative chlorine on the samples, N and V are the normality (equiv./L) and volume (L) of sodium thiosulfate, respectively, and W is the weight of the cotton samples (g).

Characterization Techniques

Fourier transform infrared (FTIR) were collected on a Nicolet iS 50 FTIR spectrometer (Thermo Fisher Scientific, USA) using the ATR method in the range of 500-4000 cm^{-1} to analyze the untreated and treated samples.

The surface morphologies of the untreated and the PAMA coated cotton fabrics were observed using a TESCAN VEGA3 scanning electron microscope (TESCAN, Czechoslovakia) with magnification of 1000× and 2000× (imaging beam voltage: 30 kV).

Antibacterial test was measured against *Staphylococcus aureus* and *Escherichia coli* O157:H7 according to GB/T 20944.3-2008. Two layers of nutrient medium were poured on glass plate. After 15 m*l* of sterile agar medium was coagulated on the bottom, 5 m*l* of bacterial agar was poured on the top and solidified for a few minutes. Then the unchlorinated and chlorinated cotton fabric was cut into a round specimen with a diameter of 25 mm and placed on the middle of the plate with sterile nipper. After the glass plate was cultivated in incubator for 24 h, the diameter of the inhibition zone was determined.

For washing durability testing, the PAMA coated cotton fabrics were chlorinated and the initial oxidative chlorine content was measured by iodometric/thiosulfate titration method according to the literature [22]. The chlorinated samples were soaked in 0.5 % detergent solution, stirred for 15 min. Then the samples were washed several times with tap water and dried at 45 °C for 1 h. The above washing process was repeated for 5 times. After each washing cycle, the sample was used to measure active chlorine content.

Wrinkle recovery angles (WRA) of control and PAMA coated cotton fabrics were determined according to GB/T 3819-1997. The samples were tested in multiplicate for each sample and the results were averaged. The testing was carried out at ambient temperature. The recovery angles were measured in both warp and weft directions, and the sum of these angles was reported. The air permeability was determined on a Fully Automatic Permeability Instrument (YG461E-III, Ningbo Textile Instrument Factory) under air pressure of 100 Pa. The handle of cotton sample including surface and bending properties was detected on a KES-FB-AUTO-A automatic test machine (KESKATO, Japan).

Results and Discussion

Preparation of Antibacterial Cotton Fabrics

The polymer of N-halamine precursor (PAMA) containing polycarboxylic acid, which can be bound onto cellulose fibers with the assist of sodium hypophosphite (SHP) through pad-dry-curing process, was synthesized by co-polymer of acrylamide and maleic anhydride. After exposing to dilute sodium hypochlorite solution, the oxidative chlorine content on the cotton fabrics can be determined by iodometric/thiosulfate titration method. In this study, the finishing parameters have a great effect on the chlorine loading of samples, and the optimum finishing conditions are obtained by measuring the influence of PAMA concentration, SHP concentration and curing temperature on the chlorine content.

The effect of concentration of PAMA on the chlorine loading was studied and the result was showed in Figure 4. In this procedure, the cotton fabrics was treated with 2-10 % of PAMA under curing temperature of 150 °C and no catalyst (SHP) was used. It can be seen that the chlorine content raised from 0.03 % to 0.1 % with the increase of PAMA concentration from 2 % to 10 %. This indicates that higher PAMA concentration can favor the reaction between PAMA and cellulose fibers because more reaction chance might happen with the increase of PAMA concentration. In the previous work, it has been indicated that 0.1 % oxidative chlorine content is efficient for rapid inactivation of bacteria [17]. Considering the economic cost and maximum chlorine

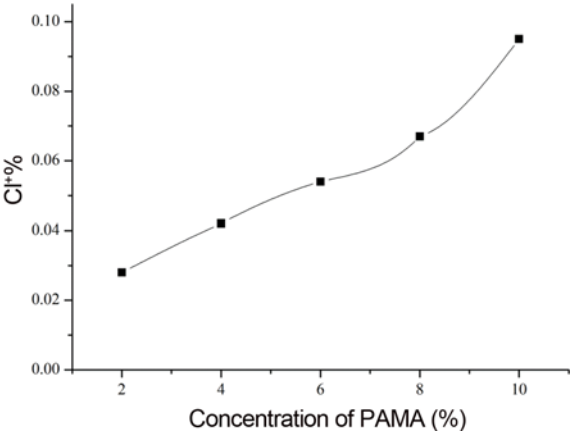

Figure 4. Effect of PAMA concentration on the chlorine content in the finishing process (SHP: 0 %, curing temperature: 160 °C).

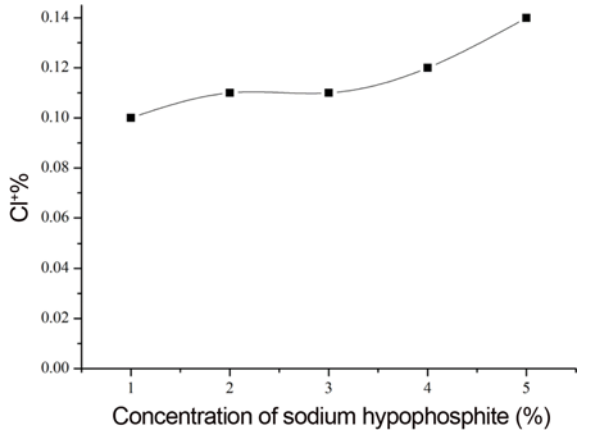

Figure 5. Effect of SHP concentration on the chlorine content in the finishing process (PAMA: 10 %, curing temperature: 160 °C).

loading, there is no need sequentially increasing the PAMA concentration, and 10 % of PAMA was used in the following treatment.

Polycarboxylic acid, such as 1,2,3,4-butancetetracarboxylic acid and maleic acid, can be bound to cellulose fibers with the assist of sodium hypophosphite by formation of cyclic anhydride, which can react with hydroxyl group in the cellulose to form cellulose esterification [23,24]. The reaction between PAMA and cellulose could be greatly influenced by the amount of sodium hypophosphite, thus deciding the chlorine content deposited on the cotton fabrics. The effect of sodium hypophosphite on the chlorine content was measured and the result was presented in Figure 5. With the increasing amount of sodium hypophosphite from 1 % to 5 %, the chlorine content gradually increased. When the sodium hypophosphite reached to 5 % with 10 % PAMA under 160 °C, the chlorine loading was 0.14 %, which is enough for repaid inactivation of bacteria.

The effects of curing temperature on the chlorine content were studied, and the result was presented in Figure 6. As

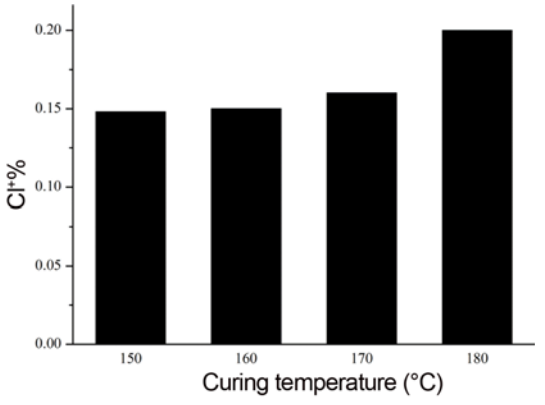

Figure 6. Effect of curing temperature on the chlorine content in the finishing process (PAMA: 10 %, SHP: 5 %).

Table 1. Optimum finishing condition of cotton fabrics with PAMA

Condition	PAMA concentration	SHP concentration	Curing temperature	Chlorine content
Result	10 %	5 %	180 °C	0.20 %

shown in Figure 6, higher curing temperature leads to higher chlorine content with the same curing time. This suggests that the curing temperature, which can promote the formation of ester bond between cellulose and PAMA, is a very important factor affecting the chlorine content on the fabrics [25]. When the curing temperature increased to 180 °C, 0.2 % of chlorine content on the cotton fabrics was obtained, which can provide efficient biocidal properties against bacteria.

After studying the effect of PAMA concentration, SHP concentration and curing temperature on the chlorine loading in the cotton fabrics, the optimum finishing conditions of PAMA on the cotton fabrics were achieved, shown in Table 1. When the cotton fabrics were treated with 10 % PAMA and 5 % SHP at 180 °C curing temperature for 5 min, the chlorine loading can reach at 0.2 %, which can provide cotton fabrics with efficient antibacterial efficacy.

Characterization of Untreated and Treated Cotton Fabrics

After treated with PAMA, the functional group on the cotton fabrics will be changed, which can be determined by FTIR spectra. As shown in Figure 7, the FTIR spectra of untreated, PAMA treated cotton fabrics before and after chlorination were presented. Compared with untreated cotton fabrics (Figure 7(A)), two new peaks at 1576 cm^{-1} and 1719 cm^{-1} were detected on the PAMA treated samples (Figure 7(B)), which correspond to the amino group and

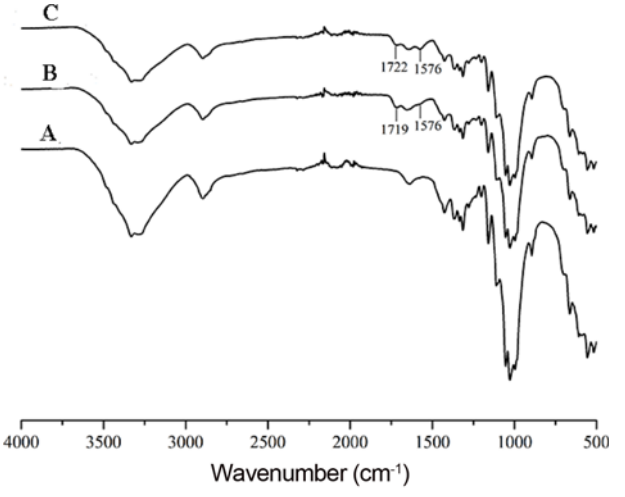

Figure 7. FT-IR spectra of (A) control cotton fabrics, (B) unchlorinated PAMA-coated cotton fabrics, and (C) chlorinated PAMA-coated cotton fabrics.

carbonyl group in the PAMA. This indicates that PAMA was successfully coated on the cotton fabrics. After chlorination, the absorption peak of carbonyl group shifted to 1722 cm^{-1}, which might be caused by the electron-withdrawing effect of oxidative chlorine [26-28].

To further provide the evidence that these PAMA was attached onto cotton fibers successfully, the surface difference of cotton fibers before and after treatment with PAMA can be measured by SEM and the results were shown in Figure 8. Obviously, the cotton fibers after treating with PAMA become more rough (Figure 8(B) and (B_1)), and the surface of the uncoated cotton fibers was relatively smooth (Figure 8(A) and (A_1)), which suggests that PAMA was successfully attached onto the cotton fabrics in the other aspect.

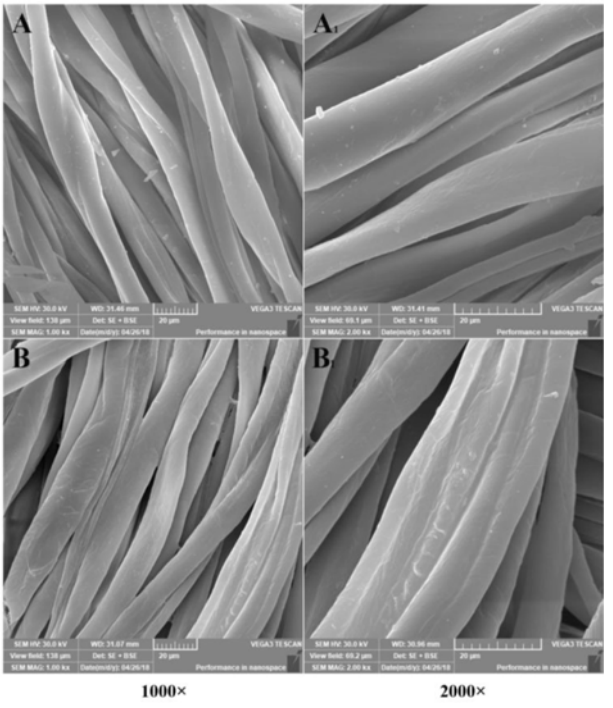

Figure 8. SEM micrographs of (A, 1000× and A_1, 2000×) control cotton fabrics and (B, 1000× and B_1, 2000×) PAMA-coated cotton fabrics.

Antibacterial Properties

The antibacterial properties of unchlorinated and chlorinated PAMA treated cotton fabrics were determined by inhibitation zone against *S. aureus* and *E. coli* O157:H7 according to GB/T 20944.3-2008, and the results were presented in Figure 9. Obviously, the unchlorinated sample did not show any inhibition zones for both *S. aureus* and *E. coli* (Figure 9(A)). However, in the case of the chlorinated PAMA treated cotton fabrics containing 0.2 % of oxidative chlorine, a clear inhibition zone (2-3 mm) could be detected against both *S. aureus* and *E. coli* (Figure 9(B)). These findings indicated that at least some of the biocidal agents like oxidative chlorine diffused away from the chlorinated PAMA treated cotton fabrics to kill the bacteria during the antimicrobial tests. According to the previous studies on the N-halamines, it was believed that the oxidative chlorine generated from the dissociation of N-halamines were most likely the effective biocidal agents that provided the potent antimicrobial activity [29].

Washing Stability

For functional textiles, the washing durability and stability of functional moieties are important for their practical application. For N-halamine antibacterial textiles, the washing

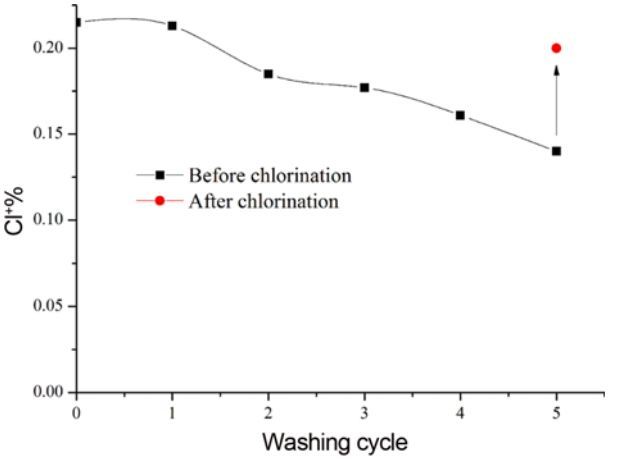

Figure 10. Washing durability of chlorinated cotton fabrics coated with PAMA.

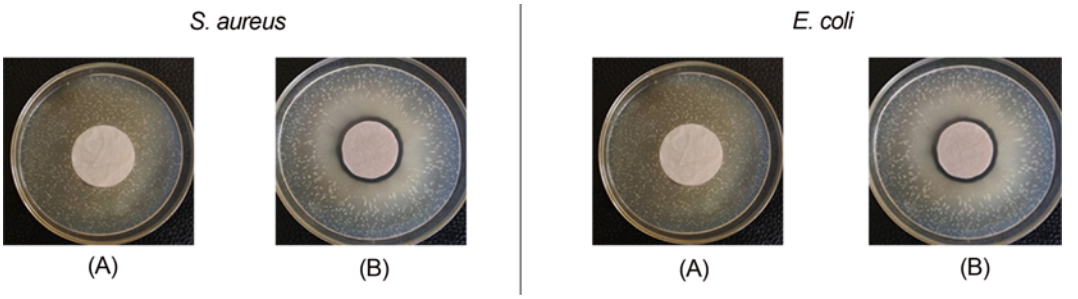

Figure 9. Antibacterial properties of (A) unchlorinated and (B) chlorinated PAMA-coated fabrics against *S. aureus* and *E. coli*.

stability can be tested by measuring the chlorine content on the textiles. The decrease of chlorine content on the textiles can be caused by decomposition of N-Cl to N-H and destruction of covalent bonds between N-halamine moieties and textiles [22]. In this study, the washing durability was simulated and the results were presented in Figure 10. The chlorine loading decreased about 30 % from 0.21 % to 0.14 % after 5 washing cycles. However, after exposing to dilute sodium hypochlorite, the chlorine content can be recovered to the original level, which means that the reduction of reactive chlorine is mainly caused by decomposition of N-Cl groups and covalent bonds between PAMA and cotton fibers are very stable and resistant to washing.

WRA and Air Permeability Testing

The performance of WRA and air permeability for untreated cotton fabrics, PAMA-treated cotton fabrics before and after chlorination was investigated, and the results were presented in Table 2. It shows that WRA of cotton fabric had a small improvement after treating, increasing from 135 ^{o} to 173 ^{o}. The improvement of WRA might be owing to the formation of ester bonds between the hydroxyl groups of cotton fabric and the carboxyl groups in PAMA. In addition, the WRA of PAMA-treated cotton fabrics after chlorination did not show big change compared with unchlorinated PAMA-treated samples. The air permeability of cotton fabrics after treated with PAMA has a little improvement, increasing from 231.78 mm/s to 265.72 mm/s, which indicated that the PAMA-coated cotton fabric possessed bigger interspaces between fibers enhancing the penetrability of air comparing with control samples [30]. The reason might be that the acidity of finishing solution and high curing temperature caused some damage in the cotton fiber and formed holes on the fiber [31]. The air permeability of chlorinated PAMA-coated cotton fabrics was 240.61 mm/s, which is lower than unchlorinated samples but better than control cotton fabrics.

Table 2. Wrinkle recovery angle of cotton samples

Sample	WRA (^{o})			Air permeability (mm/s)
	Warp	Weft	Warp+weft	
Control cotton	73	62	135	231.78
Cotton-PAMA	93	80	173	265.72
Cotton-PAMA-Cl	92	85	177	240.61

KES Surface and Bending Properties

The hand feel and softness properties of control cotton, PMAM treated samples before and after chlorination were detected by measuring surface and bending properties with a KES test machine. In this test, MIU (coefficient of friction), MMD (variation of the MIU), SMD (geometrical roughness), B (bending rigidity), 2HB (bending hysteresis) in the warp and weft direction were determined, and the results were presented in Table 3. Compared with control samples, PAMA-treated cotton fabrics showed higher MIU, MMD and SMD values, which indicates that PAMA loaded on cotton fibers increases the surface roughness and decreases the leveling, leading to bad handle for cotton samples [32]. The bending rigidity and bending hysteresis after treatment was also increasing because of cross-linking formed between PAMA and cellulose fibers, resulting in higher stiffness than control cotton. After chlorination, the values of surface and bending properties had a little decrease and the hand feel can be slightly improved in the bleaching solution, compared with unchlorinated cotton fabrics.

Conclusion

An N-halamine precursor polymer (PAMA) was synthesized by copolymer of acrylamide and maleic anhydride, and coated onto cotton fabrics to prepare antibacterial material by traditional pad-dry-curing method. The effect of PAMA concentration, SHP concentration and curing temperature was studied and 0.20 % of active chlorine content was obtained under the optimum finishing condition: 10 % PAMA, 5 % SHP and 180 ^{o}C. After chlorination, the antibacterial properties were measured by inhibitation zone, and 2-3 mm of inhibitation zone can be observed. The coated cotton fabrics showed good washing stabilities and 70 % of active chlorine can be remained after 5 washing cycles. Besides, almost all of active chlorine can be recharged after exposing to dilute sodium hypochlorite solution again. The anti-wrinkle property and air permeability was also investigated and PAMA coating had a little improvement for the wrinkle recovery angle. However, compared with control samples, the PAMA-coated fabrics showed higher surface roughness and stiffness, which had a little negative effect on the hand feel of cotton fabrics.

Table 3. KES surface and bending properties of cotton samples

Sample	MIU (-)		MMD (-)		SMD (μm)		B (gf · cm^2/cm)		2HB (gf·cm/cm)	
	Warp	Weft	Warp	Weft	Warp	Weft	Warp	Weft	Warp	Weft
Control cotton	0.173	0.178	0.0143	0.0142	3.250	4.43	0.0454	0.0781	0.0483	0.0950
Cotton-PAMA	0.192	0.172	0.0188	0.0183	3.695	3.14	0.3663	0.6174	0.2051	0.8591
Cotton-PAMA-Cl	0.153	0.148	0.0162	0.0147	2.882	3.01	0.3040	0.4374	0.1521	0.2314

References

1. B. Simoncic and B. Tomsic, *Text. Res. J.*, **80**, 1721 (2010).
2. G. Sun, X. Xu, J. Bickett, and J. Williams, *Ind. Eng. Chem. Res.*, **40**, 1016 (2014).
3. T. Ramachandran, K. Rajendrakumar, and R. Rajendran, *J. Inst. Eng.*, **84**, 42 (2004).
4. S. Chen, S. Chen, S. Jiang, M. Xiong, J. Luo, J. Tang, and Z. Ge, *ACS Appl. Mater. Inter.*, **3**, 1154 (2011).
5. X. Ren, K. Hasanb, S. D. Worley, R. M. Broughton, and T. S. Huang, *Carbohydr. Polym.*, **75**, 683 (2009).
6. X. Ren, A. Akin, K. Hasanb, S. D. Worley, R. M. Broughton, and T. S. Huang, *Carbohydr. Polym.*, **78**, 220 (2009).
7. L. Qian and G. Sun, *J. Appl. Polym. Sci.*, **91**, 2588 (2010).
8. X. Ren, L. Kou, H. B. Kocer, C. Zhu, S. D. Worley, R. M. Broughton, and T. S. Huang, *Colloid Surface A*, **317**, 711 (2008).
9. X. Ren, L. Kou, J. Liang, S. D. Worley, Y. M. Tzou, and T. S. Huang, *Cellulose*, **15**, 593 (2008).
10. X. Ren, L. Kou, H. B. Kocer, S. D. Worley, R. M. Broughton, Y. M. Tzou, and T. S. Huang, *J. Biomed. Mater. B*, **89B**, 475 (2010).
11. L. Jie, Y. Chen, X. Ren, W. Rong, K. Barnes, S. D. Worley, R. M. Broughton, U. Cho, H. Kocer, and T. S. Huang, *Ind. Eng. Chem. Res.*, **46**, 6425 (2007).
12. L. Kou, J. Liang, X. Ren, H. B. Kocer, S. D. Worley, Y. M. Tzou, and T. S. Huang, *Ind. Eng. Chem. Res.*, **48**, 6521 (2009).
13. L. Kou, J. Liang, X. Ren, H. B. Kocer, S. D. Worley, R. M. Broughton, and T. S. Huang, *Colloid Surface A*, **345**, 88 (2009).
14. M. Orhan, D. Kut, and C. Gunesoglu, *J. Appl. Polym. Sci.*, **111**, 1344 (2010).
15. A. J. Kugel, S. M. Ebert, S. J. Stafslien, I. Hevus, A. Kohut, A. Voronov, and B. J. Chisholm, *React. Funct. Polym.*, **72**, 69 (2012).
16. S. H. Lee, M. J. Kim, and H. Park, *J. Appl. Polym. Sci.*, **117**, 623 (2010).
17. J. Lee, R. M. Broughton, A. Akdag, S. D. Worley, and T. S. Huang, *Fiber. Polym.*, **8**, 148 (2007).
18. A. Akdag, S. Okur, M. L. Mckee, and S. D. Worley, *J. Chem. Theory Comput.*, **2**, 879 (2006).
19. S. Liu and G. Sun, *Ind. Eng. Chem. Res.*, **48**, 103 (2017).
20. J. Luo and Y. Sun, *Ind. Eng. Chem. Res.*, **47**, 5291 (2008).
21. J. Luo, N. Porteous, and Y. Sun, *ACS Appl. Mater. Inter.*, **3**, 2895 (2011).
22. Z. Kang, B. Zhang, Y. Jiao, Y. Xu, Q. He, and J. Liang, *Cellulose*, **20**, 885 (2013).
23. C. Q. Yang, D. Chen, J. Guan, and Q. He, *Ind. Eng. Chem. Res.*, **49**, 8325 (2010).
24. C. Q. Yang, Q. He, and B. Voncina, *Ind. Eng. Chem. Res.*, **50**, 5889 (2011).
25. R. Li, M. Sun, Z. Jiang, X. Ren, and T. S. Huang, *Fiber. Polym.*, **15**, 234 (2014).
26. Z. Jiang, K. Ma, J. Du, R. Li, X. Ren, and T. S. Huang, *Appl. Surf. Sci.*, **288**, 518 (2014).
27. I. Cerkez, H. B. Kocer, S. Worley, R. Broughton, and T. Huang, *React. Funct. Polym.*, **72**, 673 (2012).
28. J. Li, Y. Liu, Z. Jiang, K. Ma, X. Ren, and T. S. Huang, *Ind. Eng. Chem. Res.*, **53**, 13058 (2014).
29. Z. Cao and Y. Sun, *J. Biomed. Mater. Res. A*, **85A**, 99 (2010).
30. Z. Li, H. Wang, J. Du, J. Fu, and W. Wang, *Fiber. Polym.*, **19**, 1228 (2018).
31. Y. M. Deng, S. F. Wang, and S. J. Wang, *Fiber. Polym.*, **17**, 1384 (2016).
32. C. K. Chan, X. Y. Jiang, K. L. Liew, L. K. Chan, W. K. Wong, and M. P. Lau, *J. Mater Process. Tech.*, **174**, 183 (2006).