Improving the Antibacterial Activity of Cotton Fabrics Finished with Triclosan by the Use of 1,2,3,4-Butanetetracarboxylic Acid and Citric Acid

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ABSTRACT: For producing antibacterial textiles, the conventional finishing processes have high productivity and low processing costs, but textiles finished in these ways exhibit low durability against laundering. Therefore, cotton fabrics were bleached with hydrogen peroxide, finished with triclosan, and then treated with polycarboxylic acids such as 1,2,3,4-butanetetracarboxylic acid (BTCA) and citric acid (CA) as crosslinking agents to provide durable antibacterial properties. The surface of fibers treated with BTCA had a greater crosslinked area, and the surfaces of fabrics treated with CA were exposed to greater amounts of deformation due to the mechanical and chemical influences after 50 launderings. The bleaching and finishing treatments did not dramatically affect the breaking strength. However, the polycarboxylic acid treatment (both BTCA and CA) alone

INTRODUCTION

Natural fibers such as cotton, viscose, and flax are constantly exposed to microbial attacks and also are more susceptible to attack than synthetics. Therefore, the antimicrobial activity is an important property for some functional fabrics, and triclosan is the most effective finishing agent. The durability of the antimicrobial action against textile aftercare treatments is essential, and a number of approaches have been used for textile materials.^{1–7} To improve the washing durability of antimicrobial finishing, some crosslinking agents are usually used along with the antimicrobial chemicals. Crosslinking agents are usually small molecules containing several functional groups capable of reacting with some active groups in the polymer, such as hydroxyl groups in cellulose. Tra-

showed reductions in the breaking strength when the acid concentration was increased. The polycarboxylic acids were fairly effective against both bacteria, even at lower concentrations, when they were applied to stand-alone cotton fabrics, whereas the antibacterial activity decreased somewhat after the use of polycarboxylic acid and triclosan in the same recipes. Adding polycarboxylic acids to the antibacterial finishing recipes enhanced the durability after 50 launderings, and the durability of the recipes containing BTCA was much higher than that of the recipes containing CA. © 2008 Wiley Periodicals, Inc. J Appl Polym Sci 111: 1344-1352, 2009

Key words: biological applications of polymers; crosslinking; electron microscopy

ditional crosslinking agents used in cellulose are Nmethylol resins and their derivatives, and they have long been used as durable press finishes producing wrinkle-resistant fabrics. However, formaldehydebased finishing agents have caused worldwide concern over their impact on human health and the environment, and some of these are prohibited by some governments. There have been efforts to achieve nonformaldehyde alternatives to replace the traditional N-methylol reagents.^{8–11}

Polycarboxylic acids, particularly 1,2,3,4-butanetetracarboxylic acid (BTCA) and citric acid (CA), are able to form effective crosslinks in cotton fabrics when salts of certain phosphorus-containing acids [sodium hypophosphite (SHP)] are used as catalysts. Polycarboxylic acids have been confirmed as the most promising formaldehyde-free crosslinking agents for cotton cellulose among the various new reagents investigated. It is now known that the crosslinking effect of polycarboxylic acids is maintained by an esterification mechanism, and many studies have shown that cellulose esterification with a polycarboxylic acid proceeds first to form a cyclic anhydride and then to form an ester with the hydroxyl group (-OH) in the cellulose macromolecule. It has also been found that improving the reactivity and number of hydroxyl groups

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				D		the minim	g ileau	nems			
				Chemical concentration (%)						Curing	
Recipe code	Raw	Bleached	BTCA	CA	SHP	Triclosan	pН	Temperature (°C)	Time (min)	Temperature (°C)	Time (min)
0		_	_	_		_		_	_	_	_
1	_		_	—	—	_	_	—	_	—	_
2						45	5.60	150	5	—	
3	_	_	_	—	—	60	5.32	150	5	—	_
4	_	_		_		80	5.24	150	5	_	_
5	_	_	90	—	—	_	2.08	85	5	180	1.5
6	_	_	60	—	—	_	2.10	85	5	180	1.5
7	_	_	30	_		_	2.43	85	5	180	1.5
8	_	_	15	—	—	_	2.58	85	5	180	1.5
9	_	_	15	—	6.78	_	2.63	85	5	180	1.5
10	_	_	10	_	4.52	_	2.63	85	5	180	1.5
11			5		2.26	—	2.98	85	5	180	1.5
12	_	_	5	—	2.26	50	3.05	85	5	150	5
13	_	_	_	90	—	_	2.00	85	5	180	1.5
14				60		—	2.00	85	5	180	1.5
15	_	_	_	30	—	_	2.33	85	5	180	1.5
16	_	_		15		_	2.41	85	5	180	1.5
17	_	_	_	15	7.56	_	2.43	85	5	180	1.5
18	_	_		10	5.04	_	2.43	85	5	180	1.5
19	_	_	_	5	2.52	_	2.75	85	5	180	1.5
20	—	—		5	2.52	50	2.82	85	5	150	5

TABLE I Details of the Finishing Treatments

in the macromolecules would also improve their availability for the acid and catalyst. Among the nonformaldehyde-type treatment resins for the esterification crosslinking of cotton fabrics, BTCA and CA are the most favorable compounds. Although BTCA is very effective, it is very costly; consequently, the high cost of BTCA has prevented its commercial applications.^{11–21}

The focus of this work was improving the antibacterial activity of cotton fabrics finished with triclosan through the use of BTCA and CA as crosslinking agents. For this purpose, sample fabrics were bleached with hydrogen peroxide, finished with triclosan, treated with polycarboxylic acids (with and without a catalyst and with and without triclosan), and laundered for 50 cycles. The fabrics were evaluated on the basis of a one-time antibacterial treatment and one use. The antibacterial activities of the fabrics were examined against both *Staphylococcus aureus* and *Escherichia coli*.

EXPERIMENTAL

Materials

In this study, we used plain cotton fabric (110 g/m^2) with 20-s × 20-s ends and picks. BTCA, CA, and SHP (in the form of SHP—NaH₂PO₄) were supplied by Sigma–Aldrich (Steinheim, Germany), and a triclosan derivative (nonionic diphenyl alkane) was supplied by Rudolf-Duraner (Turkey).

Treatments

The pretreatment of fabrics was carried out with hydrogen peroxide (4% owf) and a fluorescent brightening agent (a nonionic stilbene–triazine derivative; 0.2%) at a 1 : 10 material-to-liquor ratio by the conventional exhaust process. The bleaching bath was heated at 90°C and held for 60 min, and this was followed by rinsing and drying.

To acquire the antibacterial activity, the bleached fabrics were finished in different concentrations of triclosan through a conventional pad–dry process. The fabrics were padded through squeeze rollers made by Mathis (Oberhasli, Switzerland) with two dips and two nips at a setting of 3.5 bars to give a wet pickup of 80–85% owf and then dried at 150°C for 5 min in a Mathis curing oven to avoid the yellowing effect of triclosan on the cotton fabrics at higher temperatures.

Bleached fabrics were impregnated in aqueous treatment baths containing BTCA, CA, and SHP, as shown in Table I. Three sets of experiments were performed: one without a catalyst and triclosan, one with a catalyst without triclosan, and one with a catalyst and triclosan. BTCA and CA concentrations were expressed according to the weight of the agent in the water solution, and the solutions with SHP as a catalyst were prepared with a 1 : 1 acid-to-catalyst ratio (w/w), with consideration of conventional durable-press recipes. The fabrics were treated in different concentrations of BTCA and CA and

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padded through squeeze rollers made by Mathis with two dips and two nips at a setting of 3.5 bars to give a wet pickup of 80–85% owf. Polycarboxylic acids form anhydrides at lower temperatures when they are applied to cotton fabrics. However, an increase in temperature increases both the amount of the anhydride and the amount of the ester formed on the cotton (polycarboxylic acids esterify cellulose more effectively at higher temperatures).^{22,23} Therefore, in the studies including polycarboxylic acids as crosslinking agents, the treated fabrics were predried at 85°C for 5 min, and dried samples were cured at 180°C for 1.5 min in a Mathis curing oven, with consideration of conventional durable-press recipes.

Measurements

The surface morphologies of the untreated and treated cotton fabrics were observed with a JEOL JSM 6060 scanning electron microscope (Tokyo, Japan). The standard procedure was followed, in which samples were coated with gold for 150 s before a scanning electron microscopy (SEM) examination.

The samples were conditioned for 24 h at 20°C and 65% relative humidity before breaking strength testing. The breaking strength of the fabrics (only weft directions because of the plain fabric structure) was measured with an Instron model 4301 CRE tensile tester (Norwood, MA) on samples 20 mm long at a speed of 100 mm/min according to the standard procedures described in ASTM Method D 1682-64. The testing was carried out at room temperature, and all recorded data were averages of five measurements.

In textile finishing applications, washing durability is one of the most important usage properties. All samples were laundered according to AATCC Test Method 124-2000 (for antimicrobial testing only), and the durability of the antibacterial finishing and crosslinking treatments against repeated home laundering was evaluated. A nonionic detergent (5% owf) was used for each laundering cycle, which lasted about 30 min and consisted of laundering at 60°C, water-rinsing at 30°C, and squeezing. All the treated fabrics were subjected to 10, 25, and 50 consecutive launderings in the presence of a nonionic detergent, and then the antibacterial characteristics were measured.

The antibacterial efficiency of the all fabrics was assessed according to AATCC Test Method 100-1999. Gram-positive *S. aureus* (ATCC 6538) and Gram-negative *E. coli* (ATCC 35218) were used as the test microorganisms because they are the major causes of cross-infection in hospitals. *S. aureus* is the most frequently evaluated species. Among surgical infections, 19% are caused by *S. aureus*, and 11% are caused by *E. coli*.²⁴ The four circular fabric swatches (38 mm in diameter)

were challenged with 1.0 ± 0.1 mL (optical density at the 1.0 McFarland standard) of the bacterial inoculum in a 250-mL container. The inoculum was a nutrient broth culture containing 1.0×10^4 to 10^6 colony forming units (cfu) of the bacteria/mL. Sterilized and distilled water (100 mL) was poured into a vessel and was vigorously shaken and incubated for 24 h at 37°C. After the test and control swatches were in contact with the bacteria for 24 h, the diluted solutions were plated on Muller-Hinton II agar and incubated for 24 h at 37°C. The viable colonies of the bacteria on the agar plate were counted, and the reduction in the number of the bacteria was calculated with the following equation:

Reduction rate $(\%) = [(B - A)/B] \times 100$

where A is the number of bacteria recovered from the inoculated treated test specimen swatches in the jar incubated over the desired contact period (24 h) and B is the number of bacteria recovered from the inoculated treated test specimen swatches in the jar immediately after inoculation (at zero contact time).

During the antibacterial testing of the samples, the control swatches were made of 100% single jersey knitted cotton fabric (13×15 courses/wale and 120 g/m²). All measurements were completed in a microbiology laboratory environment (ca. 24°C and 55% relative humidity) and repeated four times. The results given in the tables are the means of four measurements for each parameter. At the same time, Fourier transform infrared spectroscopy and Commission Internationale de l'Eclairage color coordinates (L^* , a^* , b^* , and ΔE) of all samples were measured, but they were omitted in this study to avoid confusion.

RESULTS AND DISCUSSION

SEM analysis

The bleaching pretreatments of natural fibers remove most types and the greatest amounts of impurities such as natural pigments, protein, pectin, ash, oil, lubricants, and wax that are added during the production process. After the bleaching, cotton fibers are more absorbent or hydrophilic and have whiteness and brightness due to the oxidation of cotton pigments.

The raw and bleached cotton fibers were examined with SEM, and the results are shown in Figure 1. The impurities could be seen around the single cotton fiber. After the bleaching, these components of the outer layer were removed, and imperfections on the surface were more visible. Therefore, raw cotton presented an exactly rough surface (e.g., with a lot of visible grooves), whereas the surface of the bleached cotton was cleaner, and a fibrillar structure could be clearly seen.



Figure 1 SEM photographs of (a) raw cotton and (b) bleached cotton fabrics.

The influence of the crosslinking of BTCA and CA on the cotton surface was investigated with SEM, and the results are shown in Figure 2. It is believed that carboxylic acid groups act as functional groups of the crosslinking reaction in the presence of a catalyst, and the crosslinking reaction for the various acids in the presence of a catalyst may mainly be a condensation reaction that causes the deposition of a crosslinking agent in or on the treated fibers.²⁵ These pictures indicate that crosslinking might play a



Figure 2 SEM photographs of cotton fabrics treated with (a,b) BTCA and (c,d) CA: (a,c) without triclosan and (b,d) with triclosan.



Figure 3 SEM photographs of laundered cotton fabrics finished with triclosan and treated with (a) BTCA and (b) CA.

important role in coating the surface of cotton fibers because of chemical bonding. The surface of the fiber treated with BTCA had a greater crosslinked area and higher deposition of the crosslinking agent, whereas fibers treated with CA had a small crosslinked area and no deposition of the crosslinking agent. The better crosslinking of the fabrics treated with BTCA was caused by the presence of the four carboxylic acid groups in BTCA, which could produce the hydroxyl ion. These results agree with the fact that a crosslinking agent with more carboxylic acid groups is more greatly deposited in the treated fabrics. However, the influence of crosslinking on the cotton fibers could not be clearly seen after both BTCA and CA were treated with triclosan for antibacterial finishing.

To evaluate the durability against repeated home launderings, cotton fabrics were treated with BTCA and CA as crosslinking agents in the presence of triclosan. The treated fabrics were laundered for 50 cycles, and the results are shown in Figure 3. We expected the crosslinking agents with more carboxylic acid groups to have higher washing durability in the finished fabrics. The results indicated that the surfaces and crosslinking of cotton fabrics treated with CA were exposed to greater amounts of deformation because of mechanical and chemical influences after 50 launderings.

Breaking strength analysis

The pretreatment of raw cotton fabrics was carried out with hydrogen peroxide, and the results for the breaking strength are given in Table II. The breaking strength of the raw fabrics decreased 11.93% (from 24.57 to 21.64%) after the bleaching. We know that the greatest problems occurring during bleaching with peroxide are radical reactions of the bleaching compounds with the fiber. Radical reactions can occur that affect the cellulose backbone; these reactions can lead to a decrease in the degree of polymerization (DP) and eventually to a drop in the breaking strength.²⁶ Afterwards, the bleached fabrics were finished with triclosan at different concentrations. The breaking strength of the finished fabrics decreased 14.14 and 13.77% when the triclosan concentration was increased (60 and 80 g/L). It could be concluded that the bleaching and finishing treatments did not dramatically affect the breaking strength of the fabrics.

The influence of the addition of BTCA and CA on the breaking strength of the fabrics is shown in Tables III and IV. It is well known that cellulose molecules can be crosslinked with polycarboxylic acids containing three or more carboxylic acid groups. Crosslinks between cellulose molecules can impart an elastic property. However, the mechanical strength of crosslinked cotton can be adversely affected by a polycarboxylic acid treatment, and this can be attributed to irreversible acid degradation and reversible cellulose crosslinking. The breaking strength decreased dramatically, depending on the increasing acid concentration after the polycarboxylic acid treatment (both BTCA and CA) alone. This could be associated with lower acidity (the stronger the acid was, the greater the damage was).²⁷ The

TABLE II Breaking Strength of the Fabrics Finished with Triclosan (Only Weft Directions)

		Recipe code								
	0	1	2	3	4					
Breaking strength (MPa)	24.57	21.64	20.16	18.58	18.66					

Breaking Strength	Breaking Strength of the Fabrics Treated with BTCA (Only Weft Directions)													
	Recipe code													
	0	1	5	6	7	8	9	10	11					
Breaking strength (MPa)	24.57	21.64	10.25	10.80	11.18	11.52	11.67	16.62	20.14					

TABLE III

addition of SHP as a catalyst contributed to enhancing the strength retention of the crosslinked cotton at lower concentrations of both BTCA and CA.

Cotton fabrics were treated with polycarboxylic acids such as BTCA and CA as crosslinking agents in the presence of triclosan. The results for the treated fabrics are shown in Table V. In this situation (12 and 20), the treated fabrics showed better results in strength retention than those treated with the polycarboxylic acids alone. This could be associated with a higher pH of the finishing solution.

Antibacterial activity analysis

B

First, the pretreatment of raw cotton fabrics was carried out with hydrogen peroxide, and the results for the antibacterial activity are given in Table VI. Hydrogen peroxide is widely used as a bleaching agent in textile treatments, and it came into use around 1878. It is also a well-known biocide for disinfection, sterilization, and antisepsis in medical applications. It demonstrates broad-spectrum efficacy against viruses, bacteria, yeasts, and bacterial spores. The results indicated that the bleaching treatment was more effective against S. aureus than E. coli, as expected. Here, hydrogen peroxide acted as an oxidant by producing hydroxyl free radicals (•OH), which attacked essential cell components, including lipids, proteins, and DNA.²⁸ For each bacterial species, the antibacterial activity of the raw fabric was rather low, but the antibacterial activity was significantly enhanced (from 10 to 77.60% against S. aureus and from 1 to 42.86% against E. coli) after the bleaching. However, this increment was not suitable for microbial protection. It is generally agreed that reduction rates of at least 99% are needed to retard the exponential growth of most microorganisms.

Second, the bleached fabrics were finished with triclosan at different concentrations. Triclosan is one of the most widely used biocides with broad-spectrum antibacterial properties, but it also has some antifungal and antiviral properties (minimum inhibitory concentrations ranging from 0.1 to 33 mg/mL) in the bisphenol group, which has hydroxyl-halogenated derivates of two phenolic groups connected by various bridges, and exhibits particular activity against Gram-positive bacteria. It works with the concept of controlled release and provides a killing field or zone of inhibition. It is active at very low concentrations and acts mainly by inhibiting fatty acid biosynthesis through the blocking of lipid biosynthesis (e.g., phospholipid, lipopolysaccharide, and lipoprotein synthesis by specific inhibition of the enzyme enoyl-acyl carrier protein reductase).²⁸⁻³⁰ According to antibacterial tests, the fabrics finished with triclosan exhibited better activity and were more effective against Gram-positive bacteria than Gramnegative bacteria. The antibacterial activity of the fabrics was enhanced further by increasing triclosan concentrations, especially against E. coli.

Earlier studies have shown that antimicrobial cellulosic fabrics can be developed with BTCA and CA along with subsequent oxygen bleaching. Therefore, the antibacterial activity test was primarily performed to determine the effectiveness of BTCA and CA on the cotton fabrics, and the results are shown in Table VII. If we consider cotton fabrics treated with BTCA without a catalyst (5-8), all BTCA concentrations are fairly effective against both Grampositive and Gram-negative bacteria. We have known that the generation of microorganisms is affected by many factors such as the temperature, pH, oxygen, salt concentration, and nutrients. Bacteria need a physiological pH inside their cells, and

TABLE IV Breaking Strength of the Fabrics Treated with CA (Only Weft Directions)

					Recipe code	e			
	0	1	13	14	15	16	17	18	19
reaking strength (MPa)	24.57	21.64	9.75	10.69	13.26	14.88	14.66	15.97	18.65

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Breaking Stree	Breaking Strength of the Modified Recipes (Only Weft Directions)													
	Recipe code													
	0	1	2	3	4	11	12	19	20					
Breaking strength (MPa)	24.57	21.64	20.16	18.58	18.66	20.14	20.88	18.65	19.46					

TABLE V

there are also no certain pH levels for their maximum growth. Most bacteria grow best in the pH range of about 6 to 8 and will not grow at pH levels below 4.6; in general, bacteria survive at an alkaline pH better than at an acidic pH.³¹ Consequently, lower pH levels of BTCA at different concentrations caused antibacterial activity; it was especially more effective against S. aureus. We found that the antibacterial activity was increased when SHP as a catalyst (9) was used at lower concentrations of BTCA against E. coli, considering the same concentrations of BTCA without SHP (8).

When similar experiments were performed with CA, it was seen that CA was more effective against both bacteria (Table VIII). However, a reduction of the concentration was accompanied by a decrease in the antibacterial properties; especially against *E. coli*. Here, increased antibacterial activity was measured after the use of SHP as a catalyst at the same concentration of CA against E. coli, as we found for BTCA (16 and 17). In addition, at lower concentrations, CA had greater antibacterial activity than BTCA against E. coli. This could be associated with differences between BTCA and CA with respect to dissociation, functionality, solubility, location, acidity, and number of crosslinks. Because CA has lower acidity, it can have better antibacterial properties and can kill the bacteria at lower concentrations. However, in CA treatments, noticeable yellowing of the crosslinked cotton has been previously reported. Therefore, BTCA would be a better choice in imparting durable antibacterial activity to cotton without significantly impacting the mechanical strength or whiteness via a single application.^{27,32,33} Considering the results of cotton fabrics treated with BTCA and CA, we found that both BTCA and CA have remarkable antibacterial activity against both bacteria.

To both enhance the antibacterial activity and increase the durability against repeated home launderings of cotton fabrics finished with triclosan, we modified recipes with BTCA or CA (Table IX). The antibacterial activity decreased somewhat after the use of a polycarboxylic acid and triclosan in the same recipes (12 and 20) with single triclosan applications (2-4). Because that state caused a change in the pH level of the solution from basic to acidic (i.e., it was affected by the mechanism of triclosan inhibi-

tion), this phenomenon might play a important role in reducing the antibacterial activity.

Durability against repeated launderings

One of the methods for producing antibacterial textiles is adding antibacterial agents to textiles through conventional finishing processes. The advantages of such an application are high productivity and relatively low processing costs. However, many textiles finished in this way exhibit low durability against repeated home laundering. The performance and washing durability of the antibacterial functions depend on the amount of the agents imparted to the textiles and interactions between the agents and the fibers. Therefore, BTCA and CA as crosslinking agents were added to the antibacterial finishing solutions. To evaluate the durability against repeated home laundering, the samples were laundered for 50 cycles, and the results are shown in Table X. We found that repeated home laundering improved the antibacterial activity of the raw and bleached fabrics, and the activity of fabrics treated with BTCA or CA in the same solution of triclosan showed the biggest value after laundering. In addition, the durability against laundering of recipes containing BTCA was much higher than that of recipes containing CA; greater activity was especially seen against bacteria versus Gram-negative Gram-positive bacteria. Also, higher values of BTCA were expected because it has four carboxyl groups and can crosslink three cellulosic hydroxyls. Consequently,

TABLE VI Antibacterial Activity of the Fabrics Finished with Triclosan

	Recipe code								
Bacterial reduction (%)	0	1	2	3	4				
<i>S. aureus</i> (ATCC 6538)	10.00	77.60	98.20	100	100				
E. coli (ATCC 35218)	1.00	42.86	88.20	94.76	96.40				

Antibac	Antibacterial Activity of the Fabrics Treated with BTCA													
	Recipe code													
Bacterial reduction (%)	0	1	5	6	7	8	9	10	11					
S. aureus (ATCC 6538)	10	77.60	100	100	100	100	100	100	100					
E. coli (ATCC 35218)	1	42.86	100	100	99.90	77.69	92	85.54	78.50					

TABLE VII

TABLE VIII Antibacterial Activity of the Fabrics Treated with CA

Bacterial	Recipe code												
(%)	0	1	13	14	15	16	17	18	19				
<i>S. aureus</i> (ATCC 6538)	10	77.60	100	100	100	100	100	100	100				
E. coli (ATCC 35218)	1	42.86	100	100	99.92	84.62	99.23	98.08	85.06				

TABLE IX Antibacterial Activity of the Modified Recipes

Bacterial	Recipe code												
(%)	0	1	2	3	4	11	12	19	20				
S. aureus (ATCC 6538)	10	77.60	98.20	100	100	100	100	100	100				
E. coli (ATCC 35218)	1	42.86	88.20	94.76	96.40	78.50	88.24	85.06	90.20				

adding polycarboxylic acids to the antibacterial finishing recipes enhanced the durability of the antibacterial activity against laundering.

TABLE X Antibacterial Activity After 50 Launderings

Bacterial		Recipe code									
(%)	0	1	3	11	12	19	20				
<i>S. aureus</i> (ATCC 6538)	46.24	58.74	76.52	78.96	90.87	80.12	83.70				
E. coli (ATCC 35218)	24.61	37.65	52.38	60.26	74.76	53.70	59.52				

CONCLUSIONS

For producing antibacterial textiles, the pad-drycure technique as a conventional finishing process has high productivity and low processing costs, but textiles finished in this way exhibit low durability against repeated home laundering. Therefore, BTCA and CA as crosslinking agents were added to the antibacterial finishing solutions. We found that the polycarboxylic acids were fairly effective against both bacteria, even at lower concentrations, when they were applied to stand-alone cotton fabrics, whereas the antibacterial activity decreased somewhat after the use of polycarboxylic acid and triclosan in the same recipes. Adding polycarboxylic acids to the antibacterial finishing recipes enhanced than that of the recipes containing CA. Finally, the use of polycarboxylic acids in the antibacterial finishing recipes enhanced the durability against laundering.

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1352

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