

Fibrillation Tendency of Cellulosic Fibers. VII. Combined Effects of Treatments with an Alkali, Crosslinking Agent, and Reactive Dye

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ABSTRACT: Lyocell woven fabrics were treated with crosslinking agents [1,3-dimethyl-4,5-dihydroxyethylene urea (X1) and 1,3-dimethylol-4,5-dihydroxyethylene urea (X2)], binders [polyacrylate (B1), anionic polyacrylate (B2), anionic polyurethane (B3), and aliphatic polyisocyanate (B4)], and polyfunctional reactive dyes in combination with alkali treatments. The effects of the treatment conditions, including the reagent types, the reagent concentrations, the process types (pretreatments or posttreatments with alkali), and the treatment combinations, on the fibrillation tendency and dyeing behavior in treated samples were investigated. Generally, the fibril number decreased with an increasing

concentration of additives and decreased further with reactive dyeing. Fabrics treated with 45 g/L X1 (34 g/kg of fiber) or with 28 g/L X2 (21 g/kg of fiber) and dyed thereafter exhibited no fibrillation. Posttreatments with alkalis caused no changes in either the color shade or fibrillation tendency of crosslinked fabrics, although they enhanced the water retention capacity. The use of mixtures of binders B3 and B4 minimized the fibrillation, but the use of B4 led to changes in the color shade. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 100: 1176–1183, 2006

Key words: dyes/pigments; fibers; polysiloxanes; swelling

INTRODUCTION

Lyocell fibers are spun from solutions of cellulose in tertiary amine *N*-oxide. They are composed of elementary fibrils that consist of cellulose II crystallites separated along the fiber axis by highly oriented amorphous regions.^{1–3} In swollen lyocell fibers, hydrogen-bond cleavage leads to reduced lateral linkage between fibrils, causing fibrillation. The property of fibrillation in lyocell may be exploited to modify the fiber surface to achieve special effects such as peach skin and other soft-touch effects,⁴ but it restricts their wider use in industry.^{5–7} For various reasons, it may be desirable to inhibit the fibrillation tendency of fibers during the manufacturing process, and several such attempts have been reported,^{8,9} but a fundamental solution for this issue has not yet been found. Attempts have also been made to modify the fibrillation tendency in lyocell fibers after spinning, which include enzymatic treatments,^{5,10} crosslinking treatments with polyfunctional reactive dyes, finishing res-

ins, halogenated triazine compounds, and their combinations,^{6,11–19} and other techniques.^{20,21}

Cellulose is frequently treated with aqueous alkali solutions, which are significantly aggressive activating agents and cause changes in the fiber morphology such as the fibril orientation,^{20,22,23} resulting in the improvement of the fiber appearance, chemical accessibility,^{24–26} and mechanical properties.^{20,27–29} In previous studies, we investigated the effects of crosslinking treatment conditions, such as the crosslinker type, its concentration, the temperature, pretreatments and posttreatments with alkali, and the presence of softeners, on the fibrillation tendency and abrasion resistance of lyocell substrates.^{14,21} Treatments with aqueous potassium hydroxide (KOH) solutions both before and after crosslinking accelerated the fibrillation resistance in crosslinked lyocell fabrics. The addition of softeners enhanced the fibrillation resistance of the crosslinked fabric, but not its abrasion resistance. It was suggested that the fibrillation was inhibited not only by the prevention of fibril separation but also by modifications of the fiber surface.

In this work, lyocell woven fabrics were treated with crosslinking agents, binders, reactive dyes, and aqueous alkali solutions. The effects of the reagent type, concentration, alkali treatment, and reactive dyeing on the fibrillation tendency as well as the color shade of the dyed materials were investigated.

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TABLE I
Chemical Reagents and Their Primary Components

Abbreviation	Components		Trade name	Concentration (wt %)
X1	1,3-Dimethyl-4,5-dihydroxyethylene urea	Crosslinker	Fixapret NF	44–46
X2	Modified 1,3-dimethyl-4,5-dihydroxyethylene urea product	Crosslinker	Fixapret ECO	68–71
Cat1	Mixture of inorganic salts	Catalyst	Condensol N	
Cat2	Mixture of metal salts	Catalyst	Condensol FB	
Surf1	Polyacrylate-based product	Wetting agent	Kieralon TX 1563	
Surf2	Polyacrylate-based product	Wetting agent	Kieralon TX 1576	
Surf3		Wetting agent	Sandoflex A	
B1	Polyacrylate dispersion	Binder	Helizarin Binder FWT 2003	
B2	Anionic polyacrylate dispersion	Binder	Perapret HVN	
B3	Anionic polyurethane dispersion	Binder	Perapret PU	
B4	Modified aliphatic polyisocyanate	Hardener	Astacin Haerter CN	

EXPERIMENTAL

Materials

Plain woven lyocell fabrics treated with crosslinkers and binders were supplied by BASF (Ludwigshafen, Germany). The crosslinking agents, catalysts, binders, and wetting agents (the components of which are listed in Table I) were also provided by BASF. Sandoflex A and Leonil SR (133%) were kindly supplied by Clariant (Switzerland). Remazol Golden Yellow RNL (150%) and Remazol Black B were provided by DyStar (Ludwigshafen, Germany). Analytical-grade KOH (>99%) and sodium hydroxide (NaOH; >98%) were purchased from Fluka (Buchs, Switzerland). The sodium silicate was technical-grade.

Treatments of the lyocell fabric

Crosslinking

Lyocell woven fabric samples were treated by immersion in treatment solutions, the compositions of which are shown in Table II, and by the removal of the excess solution by the passage of the fabrics through squeeze rolls to obtain wet pickups of 75 wt %. The fabric was then dried at 120°C and cured at 175°C (air temperature) for 60 s. Method 1 was a control treatment in which the fabric remained untreated.

Alkali treatments

In the alkali treatments, fabric samples were treated with an aqueous solution of 250 g/L KOH containing a wetting agent [(Surf3); 50 mL/L] at the ambient temperature for 4 h, either before (pretreatment) or after (posttreatment) crosslinking treatments. The liquor pickup was 100 wt %. After successive rinses with water at 90, 70, and 40°C and room temperature, the fabric was neutralized, rinsed with sufficient water, and dried.

Treatments with binders

The fabric was immersed in a solution containing a given amount of a binder [polyacrylate (B1), anionic polyacrylate (B2), anionic polyurethane (B3), and aliphatic polyisocyanate (B4)], 2 g/L wetting agent, and 0.5 g/L acetic acid (60 wt % aqueous). After the excess solution was removed by passage through squeeze rolls, to obtain wet pickups of 75 wt %, the fabric was dried at 120°C and cured at 170°C (air temperature) for 40 s. The treatment conditions are given in Table II.

Dyeing

Fabric samples were dyed with a mixture of dyes in a pad-batch-dyeing method from solutions containing 7.5 g/L Remazol Golden Yellow RNL, 8.0 g/L Remazol Black B and 5.0 g/L Leonil SR with 110 g/L sodium silicate and 25 mL/L NaOH (38°Be). The fabrics were padded through the dye solution at a nip pressure of 2 bar pressure at a speed of 0.5 m/min. The padded samples were rolled around glass rods and wrapped in plastic films to avoid drying, and stored for 24 h at ambient temperature. The samples were then rinsed successively with hot water and cold water, neutralized with acetic acid (0.01 mol/L, pH5.0), and dried at room temperature.

Measurements

The color coordinates the difference between light and dark (L^*), the difference between green and red (a^*), and the difference between yellow and blue (b^*) in the Commission Internationale de l'Eclairage-Lab color space³⁰ of the dyed samples were measured on a Minolta CR-200 chromameter (Innsbruck, Austria) with the illuminant D_{65} and a 10° observer, and the color difference between the samples dyed after the treatments and the samples dyed after no treatment was evaluated in terms of ΔL^* , Δa^* , and Δb^* . Three

TABLE II
Conditions Used in the Treatments of the Lyocell Fabric with Additive Reagents

Method	Chemical (g/L) ^a										60% Acetic acid	Alkali treatment	
	X1	X2	Cat1	Cat2	Surf1	Surf2	B1	B2	B3	B4			
1													
2	5		20		1							0.5	
3	10		20		1							0.5	
4	20		20		1							0.5	
5	40		20		1							0.5	
6	60		20		1							0.5	
7	100		30		1							0.5	
8		3		15	1							0.5	
9		5		15	1							0.5	
10		10		15	1							0.5	
11		20		15	1							0.5	
12		40		15	1							0.5	
13		60		15	1							0.5	
14													
15	15		20									0.5	
16	15		20									0.5	Before ^a
17	15		20									0.5	After ^b
18	60		20									0.5	
19	60		20									0.5	Before ^a
20	60		20									0.5	After ^b
21		5		15								0.5	
22		5		15								0.5	Before ^a
23		5		15								0.5	After ^b
24		20		15								0.5	
25		20		15								0.5	Before ^a
26		20		15								0.5	After ^b
27													
28						2	25					0.5	
29						2		25				0.5	
30						2			25			0.5	
31						2				3		0.5	
32						2	50					0.5	
33						2		50				0.5	
34						2			50			0.5	
35						2				5		0.5	
36						2	25			3		0.5	
37						2		25		3		0.5	
38						2			25	3		0.5	
39						2	50			5		0.5	
40						2		50		5		0.5	
41						2			50	5		0.5	

^a The concentrations of the chemicals are indicated in grams per liter of the products.

^b Alkali treatment with 250 g/L KOH before crosslinking.

^c Alkali treatment with 250 g/L KOH after crosslinking.

individual color measurements were taken on each samples from different areas to obtain representative mean values.

Fabric specimens (0.5 g) were introduced into metal tumblers along with ball bearings and agitated in water at the ambient temperature with a tumbling motion, as described previously.²⁸ Fibers were then extracted from the fabric specimens, and their degree of fibrillation was assessed by the counting of the fibril number (FN_{ball}) on 0.38-mm segments of the fiber specimens with an optical microscope. The fibrillation was measured in five fibers from each sample to obtain mean values.

The water retention value (WRV) in the fibers and fabrics was measured by the centrifugal method, as described previously,²⁸ with two repetitions per sample.

RESULTS AND DISCUSSION

Effects of a triple treatment with crosslinking and dyeing on the fibrillation tendency

Two different crosslinking agents, 1,3-dimethyl-4,5-dihydroxyethylene urea (X1) and 1,3-dimethylol-4,5-dihydroxyethylene urea (X2), were used in the treat-

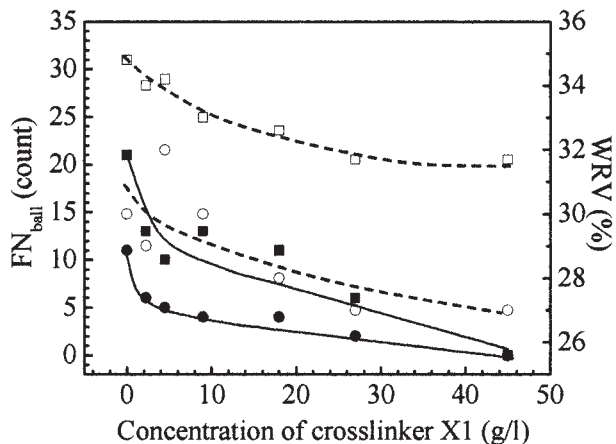


Figure 1 Plots of (■,●) FN_{ball} and (□,○) WRV in lyocell fabrics treated with X1 (■,□) before and (●,○) after dyeing against the active solid concentration of X1. The fabrics were treated according to methods 1–7.

ments. The concentration of these agents was varied, and its effect on FN_{ball} is shown in Figures 1 and 2. The concentration of the crosslinker on the x axis indicates the amount of active solid crosslinker in the aqueous solution. FN_{ball} decreased with increasing crosslinker concentration, and this corresponded to a decrease in WRV. Dyeing with a reactive dye after crosslinking further retarded both the fibrillation and swelling in fibers. In a comparison of crosslinkers X1 and X2, FN_{ball} of the fabrics treated with 5 g/L crosslinker was 4.5 and 1.5 counts for X1 and X2, respectively, whereas in those treated with 45 g/L crosslinker, FN_{ball} was with both X1 and X2. Crosslinker X2 inhibited fibrillation in the lyocell fiber more effectively at a lower concentration level. No fibrillation was observed in fabrics dyed after treatments with 45 g/L X1 or 28 g/L X2.

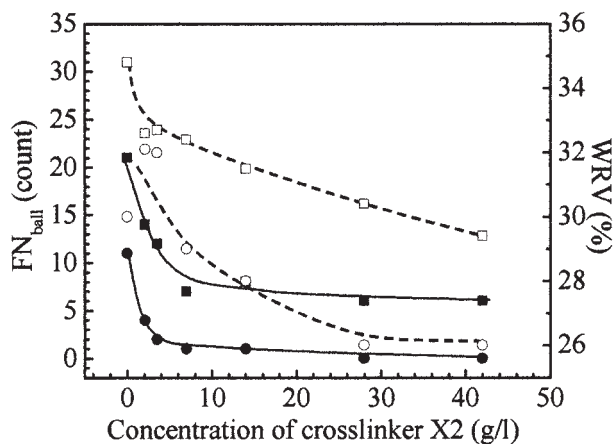


Figure 2 Plots of (■,●) FN_{ball} and (□,○) WRV in lyocell fabrics treated with X2 (■,□) before and (●,○) after dyeing against the active solid concentration of X2. The fabrics were treated according to method 1 and methods 8–13.

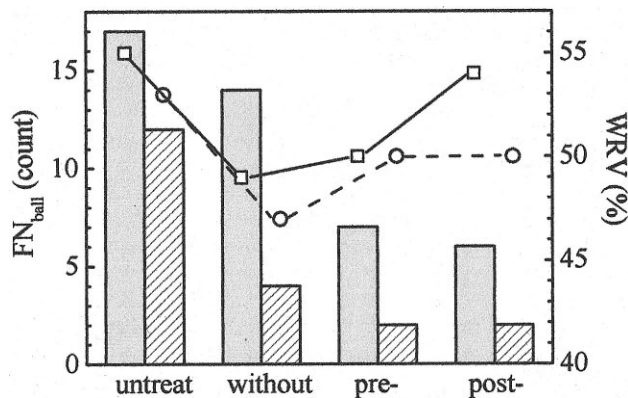


Figure 3 Effect of alkali treatments on (■,▨) FN_{ball} and (□,○) WRV in lyocell fabrics treated with 15 g/L X1 (■,□) before and (▨,○) after dyeing. The abbreviations *without*, *pre-*, and *post-* indicate the crosslinking treatment without alkali and with pretreatments and posttreatments with alkali, whereas *untreat* indicates no treatment. The fabrics were treated according to methods 14–17.

The effects of the treatment combinations on the fibrillation and fiber swelling in the fabrics are shown in Figures 3–6. FN_{ball} for the treated fabrics decreased in the following order: untreated > crosslinked (without alkali) > pre-alkali/crosslinked \approx crosslinked/post-alkali. FN_{ball} in the specimens treated with 15 g/L X1 without alkali and with pretreatment and posttreatment with alkali was 14, 7, and 6 counts, respectively, whereas that of the fabric treated with 60 g/L X1 without alkali and with pretreatment and posttreatment with alkali was 9, 6, and 6 counts, respectively. The fibrillation tendency was influenced by the crosslinker concentration in the crosslinking treatments without alkali, whereas the supplementary al-

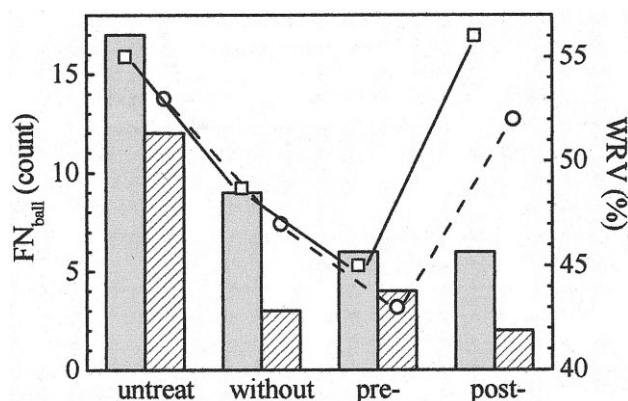


Figure 4 Effect of alkali treatments on (■,▨) FN_{ball} and (□,○) WRV in lyocell fabrics treated with 60 g/L X1 (■,□) before and (▨,○) after dyeing. The abbreviations *without*, *pre-*, and *post-* indicate the crosslinking treatment without alkali and with pretreatments and posttreatments with alkali, whereas *untreat* indicates no treatment. The fabrics were treated according to method 14 and methods 18–20.

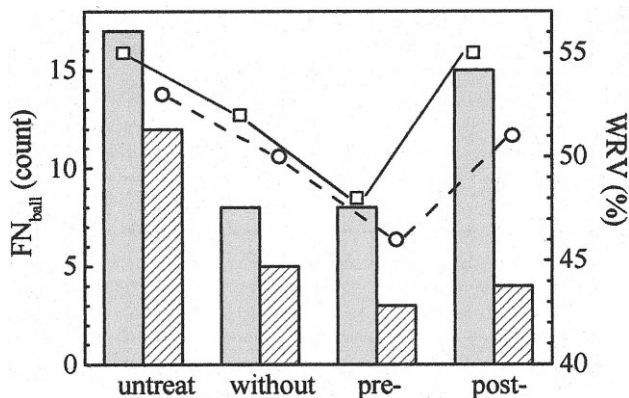


Figure 5 Effect of alkali treatments on (■, ▨) FN_{ball} and (□, ○) WRV in lyocell fabrics treated with 5 g/L X2 (■, □) before and (▨, ○) after dyeing. The abbreviations *without*, *pre-*, and *post-* indicate the crosslinking treatment without alkali and with pretreatments and posttreatments with alkali, whereas *untreat* indicates no treatment. The fabrics were treated according to method 14 and methods 21–23.

alkali treatment enhanced the fibrillation resistance in the fabrics to similar extents, regardless of the crosslinker concentration. This may be due to the reorganization of the fibril structure in aqueous alkali solutions.²⁸ The effect of the alkali treatment type (pretreatment or posttreatment) on the fibrillation tendency was not significant when crosslinker X1 was used in the treatments. In all the crosslinking experiments, including those with alkali treatments, dyeing with reactive dyes resulted in a greater inhibition in fibrillation in comparison with treatments without the dyeing step. The reactive dyeing of the lyocell fabric enhanced the degree of crosslinking among the fibrils, resulting in a stronger interfibril interaction, which

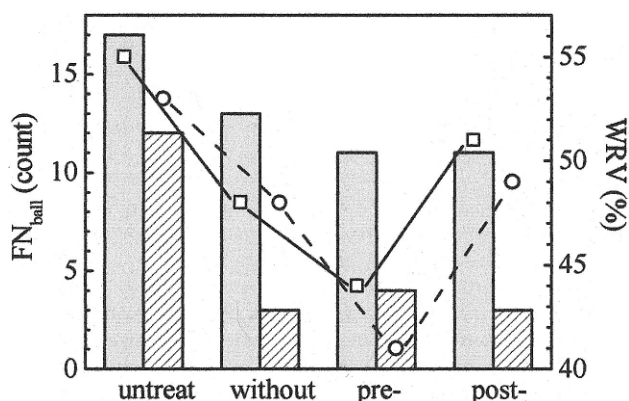


Figure 6 Effect of alkali treatments on (■, ▨) FN_{ball} and (□, ○) WRV of lyocell fabrics treated with 20 g/L X2 (■, □) before and (▨, ○) after dyeing. The abbreviations *without*, *pre-*, and *post-* indicate the crosslinking treatment without alkali and with pretreatments and posttreatments with alkali, whereas *untreat* indicates no treatment. The fabrics were treated according to method 14 and methods 24–26.

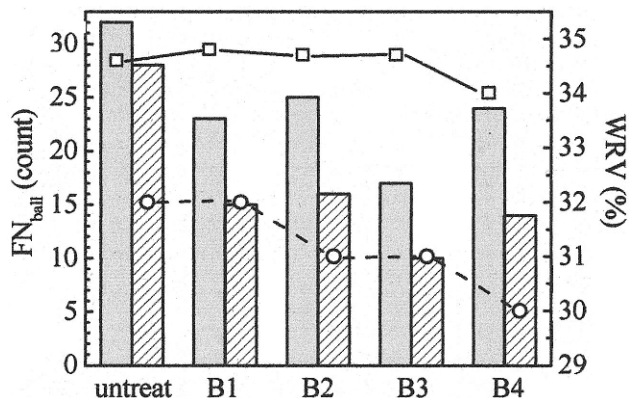


Figure 7 Effect of binders on (■, ▨) FN_{ball} and (□, ○) WRV in lyocell fabrics (■, □) before and (▨, ○) after dyeing. The fabrics were treated with 25 g/L B1–B3 or 3 g/L B4 according to methods 27–31. The abbreviation *untreat* indicates no treatment.

prevented the splitting of fibrils. The degree of swelling in the fabric, as indicated by WRV, among the treated fabrics increased in the order of pre-alkali/crosslinked < crosslinked (without alkali) < untreated and was accompanied by an increase in FN_{ball}. The post-alkali treatment caused a high degree of swelling, although a low fibrillation tendency was retained. This suggests that the post-alkali treatment may change the pore structure in the fibers but not affect the crosslinking between fibrils.

The effect of the alkali treatments on the fibrillation tendency of fabrics treated with crosslinker X2 was not significant (Figs. 5 and 6). FN_{ball} of the fabric treated with 5 g/L X2, followed by a posttreatment with alkali, was higher than that in fabrics crosslinked without an alkali treatment. The chemical crosslinks between X2 and fibrils may be less resistant to alkali than those between X1 and fibrils. FN_{ball} of the fabric treated with 20 g/L X2 decreased from 11–13 to 3–4 counts after dyeing (Fig. 6), whereas that with 60 g/L X1 decreased from 6–9 to 2–4 counts (Fig. 4). The treatment with a reactive dye reduced the degree of fibrillation to similar levels in fabrics treated with the different crosslinking agents, X1 and X2, although their fibrillation tendencies were different before the dyeing step. Further measurements are necessary to study the effect of dyeing on the fibrillation of crosslinked materials.

Effects of double treatments with a binder and dye on the fibrillation tendency

The effects of reactive dyeing on the fibrillation and degree of fiber swelling in lyocell fabrics treated with different types of binders are shown in Figures 7 and 8. The binder B3 retarded fibrillation more markedly than the other binders. FN_{ball} of the fabric treated with

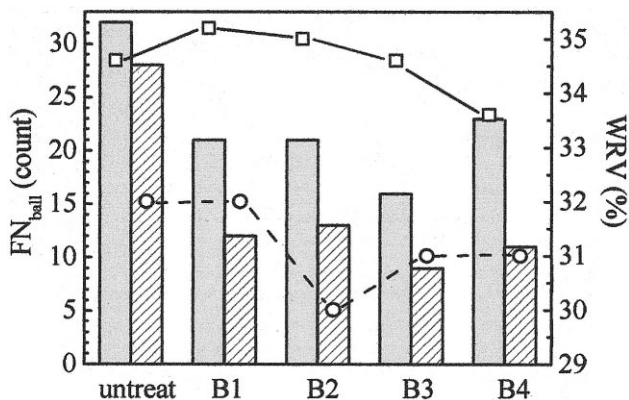


Figure 8 Effect of binders on (■, ▨) FN_{ball} and (□, ○) WRV in lyocell fabrics (■, □) before and (▨, ○) after dyeing. The fabrics were treated with 50 g/L B1–B3 or 5 g/L B4 according to method 27 and methods 32–35. The abbreviation *untreat* indicates no treatment.

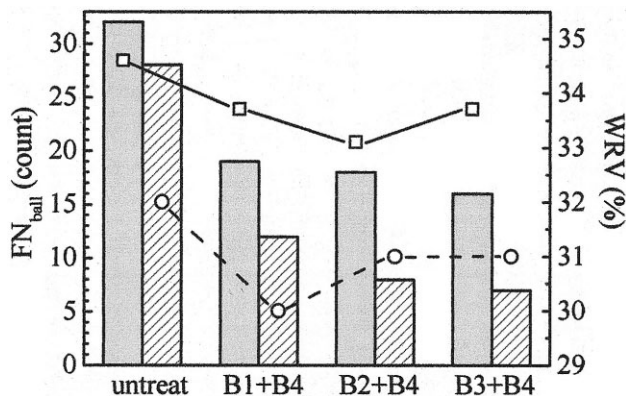


Figure 10 Effect of a mixture of binders on (■, ▨) FN_{ball} and (□, ○) WRV in lyocell fabrics (■, □) before and (▨, ○) after dyeing. The fabrics were treated with 50 g/L B1–B3 and 5 g/L B4 according to method 27 and methods 39–41. The abbreviation *untreat* indicates no treatment.

25 g/L B3 was 17 counts, whereas that with 50 g/L B3 was 16 counts; this indicated that the effect of the concentration was not meaningful under the experimental conditions used in this work. The reactive dyeing further reduced both the fibrillation tendency and extent of swelling in the fabrics treated with binders.

FN_{ball} of the fabric treated with mixtures of two binders is exhibited in Figures 9 and 10. FN_{ball} of the fabric treated with 25 g/L B2 decreased from 25 to 17 counts when 3 g/L B4 was added, whereas FN_{ball} did not change with the addition of B4 in treatments with 25 g/L B1 (23 counts). This means that the addition of B4 provided extra fibrillation resistance to the lyocell fabric in the presence of B2. The dyeing treatment lowered the degree of fibrillation in all fabrics treated with binder mixtures.

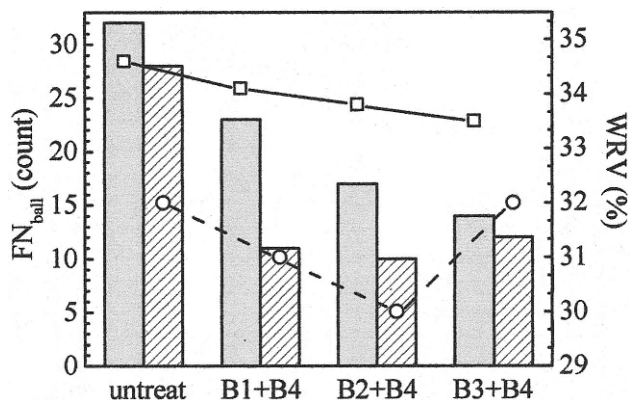


Figure 9 Effect of a mixture of binders on (■, ▨) FN_{ball} and (□, ○) WRV in lyocell fabrics (■, □) before and (▨, ○) after dyeing. The fabrics were treated with 25 g/L B1–B3 and 3 g/L B4 according to method 27 and methods 36–38. The abbreviation *untreat* indicates no treatment.

Effects of treatments with a crosslinking agent, binders, and alkali on the color shade

The color shade in the fabric samples treated with crosslinking agents, binders, and alkali was measured with a chromameter. The colorimetric data are given in Figures 11–13. The effects of the two different crosslinking agents and their concentrations on the color shade are shown in Figure 11. With increasing concentrations of X1 and X2, ΔL^* and Δb^* increased and Δa^* decreased; this indicated a lighter color shade and a change in the color shade. The decrease in the dye uptake may have been caused by an increased degree of crosslinker binding with the hydroxyl groups of cellulose, resulting in a decreased availability of active sites for the reactive dye. ΔL^* for the fabric treated with X2 was higher than that for the fabric treated with X1. The fact that the WRV of the fabric treated with X2 was lower than that in the fabric

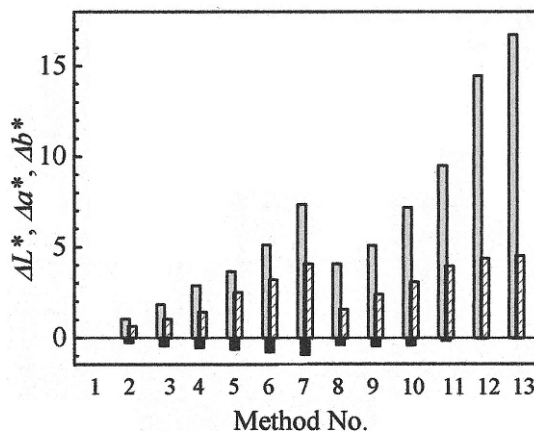


Figure 11 Effect of crosslinking treatments on color coordinates (■) ΔL^* , (▨) Δa^* , and (▩) Δb^* of lyocell fabrics dyed with reactive dyes.

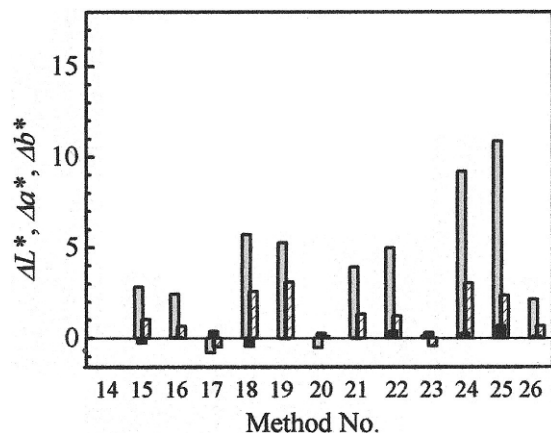


Figure 12 Effect of crosslinking and alkali treatments on color coordinates (□) ΔL^* , (■) Δa^* , and (▨) Δb^* of lyocell fabrics dyed with reactive dyes.

treated with X1 (Figs. 1 and 2) suggests that the higher uptake of X2 induced lower dye uptake.

The influence of the pretreatments and posttreatments with alkali in combination with crosslinking with X1 and X2 on the color shade is shown in Figure 12. The difference in the color coordinates between methods 14 (untreated lyocell fabric) and 17, 20, 23, and 26 (lyocell fabrics treated with an alkali solution after crosslinking) was quite small; there was an increase in the color depth, ΔL^* . On the contrary, the color difference between the fabrics treated with a crosslinking agent alone and those crosslinked after alkali pretreatments was significantly large. The results implied that the double treatment of crosslinking and alkali posttreatment did not affect dyeing, whereas the crosslinking treatment alone and the double treatment of crosslinking with an alkali pretreatment influenced dyeing. The alkali posttreatment might have changed the pore structure of the fiber, thereby increasing dye accessibility,³¹ whereas the crosslinking treatment covered the pores enlarged with the alkali pretreatment. This assumption was supported by the fact that the WRV in the fabrics treated with the crosslinking/alkali posttreatment was higher than that of the fabrics treated with the alkali pretreatment/crosslinking, as shown in Figures 3–6. In light of the low fibrillation tendency in samples 23 and 26, as shown in Figures 5 and 6, the retardation of the fibrillation was still evident even after the alkali treatment. The color difference in the fabrics treated with X2 was larger than that in fabrics treated with X1, although a lower concentration of X2 was used in the crosslinking treatment.

The results of the color measurement of the lyocell fabrics treated with one or two binders are shown in Figure 13. The addition of B4 in the binder treatments increased the color difference, regardless of the concentration of the binders. On the contrary, ΔL^* , Δa^* ,

and Δb^* in samples 28–30 and 32–34 were nearly zero, and this indicated no significant color change by the treatment with binders B1–B3.

CONCLUSIONS

The fibrillation tendency of lyocell fabrics treated with various crosslinking agents, binders, and reactive dyes was investigated. The influence of the treatment conditions on the color shade in the fabrics was also studied. FN_{ball} decreased with an increasing concentration of crosslinking agents, resulting in decreased WRV and a change in the color shade. The degree of fibrillation was reduced more distinctly by a treatment with lower concentrations of X2. The supplemental alkali treatments enhanced the fibrillation resistance of crosslinked lyocell fabrics, except in the fabrics treated with 5 g/L X2; however, the influence of the process type (pretreatment or posttreatment) was not significant. Contrarily, the degree of fiber swelling was markedly influenced by the process type in the alkali treatments, and the posttreatment with alkali heightened fiber swelling in the lyocell fabric. The alkali pretreatment process also caused significant changes in the color shade. The supplemental alkali posttreatment did not change the color shade in comparison with the untreated fabric, whereas a lighter shade was obtained with the alkali pretreatment. The treatments with binders, especially B3, retarded fibrillation in the fabric. The use of a mixture of 25 g/L B3 and 3 g/L B4 resulted in the lowest fibrillation tendency among the binder combinations used in this study, but the addition of B4 caused a significant change in the color shade.

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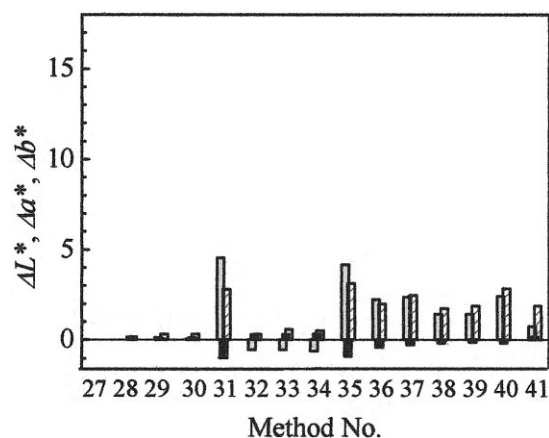


Figure 13 Effect of a treatment with binders on color coordinates (□) ΔL^* , (■) Δa^* , and (▨) Δb^* of lyocell fabrics dyed with reactive dyes.

References

1. Lenz, J.; Schurz, J.; Wrentschur, E. *J Appl Polym Sci* 1988, 35, 1987.
2. Lenz, J.; Schurz, J.; Wrentschur, E. *Holzforschung* 1988, 42, 117.
3. Lenz, J.; Schurz, J. *Cellulose Chem Technol* 1990, 24, 3.
4. Kobayashi, N. *Sen-I Gakkaishi* 1992, 48, 584.
5. Carrilo, F.; Colom, X.; Valldeperas, J.; Evans, D.; Huson, M.; Church, J. *Text Res J* 2003, 73, 1024.
6. Fang, K.; Hao, L.; Hu, X.; Shao, H. *Text Res J* 2003, 73, 1013.
7. Zhang, J.; Shi, M.; Zhu, H.; Lai, K. *Chem Fibers Int* 1999, 49, 494.
8. Mortimer, S. A.; Peguy, A. A. *J Appl Polym Sci* 1996, 60, 1747.
9. Mortimer, S. A.; Peguy, A. A. *J Appl Polym Sci* 1996, 60, 305.
10. Shin, Y.; Son, K.; Yoo, D. *J Appl Polym Sci* 2000, 76, 1644.
11. Udomkitchdecha, W.; Chiarakorn, S.; Potiyaraj, P. *Text Res J* 2002, 72, 939.
12. Lewis, D. M. *Journal of The Society of Dyers and Colourists* 1993, 109, 357.
13. Nechwatal, A.; Nicolai, M.; Mieck, K.-P. *Text Chem Colorists* 1996, 28, 24.
14. Zhang, W.; Okubayashi, S.; Bechtold, T. *J Appl Polym Sci*, accepted.
15. Zhang, W.; Okubayashi, S.; Bechtold, T. *J Appl Polym Sci*, submitted.
16. Rohrer, C.; Retzl, P.; Firgo, H. *Lenzinger Ber* 2001, 80, 75.
17. Collier, B. J. *Text Chem Colorist Am Dyestuff Rep* 1999, 1, 40.
18. Eibl, M. *Lenzinger Ber* 1997, 76, 112.
19. Mieck, K.-P.; Nicolai, M.; Nechwatal, A. *Melliand Int* 1997, 1, 34.
20. Kasahara, K.; Sasaki, H.; Donkai, N.; Yoshihara, T.; Takagishi, T. *Cellulose* 2001, 8, 23.
21. Okubayashi, S.; Bechtold, T. *J Appl Polym Sci* 2005, 12, 463.
22. Dinand, E.; Vignon, M.; Chanzy, H.; Heux, L. *Cellulose* 2002, 9, 7.
23. Sunol, J. J.; Saurina, J.; Carrillo, F.; Colom, X. *J Therm Anal Calorim* 2003, 72, 753.
24. Tanczos, I.; Borsa, J.; Sajo, I.; Laszlo, K.; Juhasz, Z. A.; Toth, T. *Macromol Chem Phys* 2000, 201, 2550.
25. Colom, X.; Carrillo, F. *Eur Polym J* 2002, 38, 2225.
26. Ribitsch, V.; Stana-Kleinschek, K.; Kreze, T.; Strnad, S. *Macromol Mater Eng* 2001, 286, 645.
27. Chae, D. W.; Choi, K. R.; Kim, B. C.; Oh, Y. S. *Text Res J* 2003, 73, 541.
28. Zhang, W.; Okubayashi, S.; Bechtold, T. *Carbohydr Polym* 2005, 59, 173.
29. Zhang, W.; Okubayashi, S.; Bechtold, T. *Carbohydr Polym* 2005, 61, 427.
30. Oakes, J.; Dixon, S. *Coloration Technol* 2003, 119, 315.
31. Kasahara, K.; Sasaki, H.; Donkai, N. *Text Res J* 2004, 74, 509.