Asian Journal of Chemistry; Vol. 23, No. 8 (2011), 3676-3682

ASIAN JOURNAL OF CHEMISTRY

www.asianjournalofchemistry.co.in

Impacts of Ultrasonic Energy on Washability, Sewability and Colour Properties of Materials in Enzymatic Process

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(*Received*: 11 December 2010; *Accepted*: 27 April 2011) AJC-9876

The short fibers on the cellulosic materials are removed by bio-processing and thus smoothing the surface. Within the scope of this study, materials on which cellulase enzyme was applied were washed through the ultrasonic probe method following bio-processing. Comparisons in terms of weight loss, whiteness levels and colour measures and changes of fabrics used in the study were carried out. Washing, rubbing, perspiration and light fastness, pilling and stitching resistance and slipping properties of materials were also observed.

Key Words: Enzymes, Cellulase, Bio-scouring, Antipilling, Ultrasound, Colour differences.

INTRODUCTION

Bio-cleaning process increases the importance of the textile industry in recent times. Cellulosic materials are widely used in applied bio-cleaning enzymes for various purposes. The enzymes involved in such processes are bio-catalysts formed by metabolic products of living organisms obtained from bacterial derivatives[1]. Amylase is used in desizing, protease for protein fibers such as silk and wool, catalase to remove peroxide following bleaching, lactase to remove indigo dye from denim fabrics, peroxidase in oxidation of the covalent-bonded dye contained in reactive dyestuff, lipase as desizing inducer and degreaser in thread spinning where solid and liquid grease is present, pectinase to bio-clean raw cotton or flax, cellulase in enzymatic stone washing and removal of fibril ends from fiber surfaces of jeans and denim fabrics[2,3]. Cellulase enzyme on the surface of fabric made with fibers removed from the bio-processes and to obtain a smoother surface.

Cellulases are highly molecular colloidal proteins obtained from *Aspergillus niger*, *Trichoderma longibrachiatum*, *Fusarium solani* and *Trichoderma viride*. Industrial cellulases, on the other hand, are complex forms, not entirely smooth, of compounds of enzymes such as cellulase and cellobiase. An enzyme-substrate complex is formed in enzyme catalysis (cellulase-cellulose complex). Cellulases are capable of snapping the 1,4-β-glucoside bond of cellulose. Three classes of enzymes display synergetic impact and effect cellulose in a complex way. Endoglucanases affect dissolved and undissolved glucose chains. Exoglucanase pulls apart the glucose unit on the end of cellulose and cellobiosis (glucose dimers) units on the end of the cellobiohydrolase (CBH) cellulose chain. β-glucosidase produces D-glucose from the dimer[4].

Ultrasonic sound waves have frequencies varying between 20 and 20 MHz. The power of ultrasonic energy is released through cavitation. Just as any other sound wave, ultrasonic sound waves are transmitted *via* waves too. Compaction and expansion occurs in the molecular structure of the environment through which these waves pass. When sufficient negative pressure is applied on the liquid, defragmentation is observed and cavitation bubbles are formed. These bobbles collide with each other within the consecutive periods of compaction and release high amounts of energy[5,6]. Use of ultrasonic energy in wet textile processes, provides benefits in terms of saving process time as well as energy and chemical substances used and enhances product quality. As a result of the cavitation that takes place on the solid/liquid interface, mass transfer from liquid to solid increases[7].

Applicability of ultrasonic energy in preparation of desizing baths and emulsion thickening process, desizing, basic processing, bleaching, dyeing, after washing and enzymatic processes is still an ongoing discussion. It is reported that through the use of the ultrasonic method, period of hydrogen peroxide bleaching process is shortened and whiteness level is increased despite low temperatures[8]; sonication provides positive impact on rupture resistance of material as well as

wetting capacity and whiteness level during bio-cleaning of raw cotton using pectinase[9]; enzyme consumption, process time and fiber damage is decreased when combined with the classical method and enzyme efficiency is significantly enhanced in pretreatment of cotton without decreasing fabric resistance[10-13].

EXPERIMENTAL

Thread, woof, warp and basis weight values of the materials used in this study are contained in Table-1.

TABLE-1
MATERIAL PROPERTIES

100 % cotton materials		
	Twill 3/1 Z	Twill 2/1 Z
Fabric densities	270 g/m^2	272 g/m^2
Yarn counts	Weft, Ne 10/2 Warp, Ne 10/2	Weft, Ne 10/2 Warp, Ne 10/2
Weft and warp density of fabric	11 picks (cm)	11 picks (cm)

Method: The enzymatic process, the operating circumstances of which can be found below, was applied prior to, during and after dyeing on fabrics treated with basic and bleaching processes. Each application was repeated thrice so as to ensure the repeatability of the process for all applications.

***Enzyme process:** For the purpose of enzyme and dyeing processes, a laboratory type HT dyeing machine (Roaches Model- MB) was operated with a scale of 1:10 process solution and the pH value of the process solutions contained in the 2 g/L cellulase enzyme (Megenzyme-6; Meg Kimya Sanayi ve Tic A.S.) was adjusted to 5 using acetic acid. Processes were started with process solutions heated to 45 ºC and upon operating at this temperature for a period of 0.5 h, the temperature was increased to 80 ºC within 10 min and operation was sustained at this increased temperature for a further period of 10 min and the enzyme was deactivated (Fig. 1).

***Dyeing process:** A reactive dye at 4 % of colour strength (Sunfix Blue RSP-4; Birkim) was used for the dyeing process along with 180 g/L of sodium sulphate and 120 g/L sodium carbonate as dyeing auxiliaries based on the temperature-time relation (Fig. 1).

***Washing process:** The washing program specified in Table-2 was employed for washing after dyeing. Sonifier 250 (Branson) ultrasonic probe was used as the source of sonication for the purpose of washing processes using ultrasonic energy. The "output control" of the device was adjusted to "4" and a 1/4 probe tip was used. The "duty cycle" parameter of the device remained at "hold" during dyeing processes and the process was conducted by submerging 1 cm of the probe tip into the process solution. When operation is sustained at over 80 ºC with the ultrasonic probe, sonication power is decreased[14]. Owing to this fact, washing was carried out at room temperature.

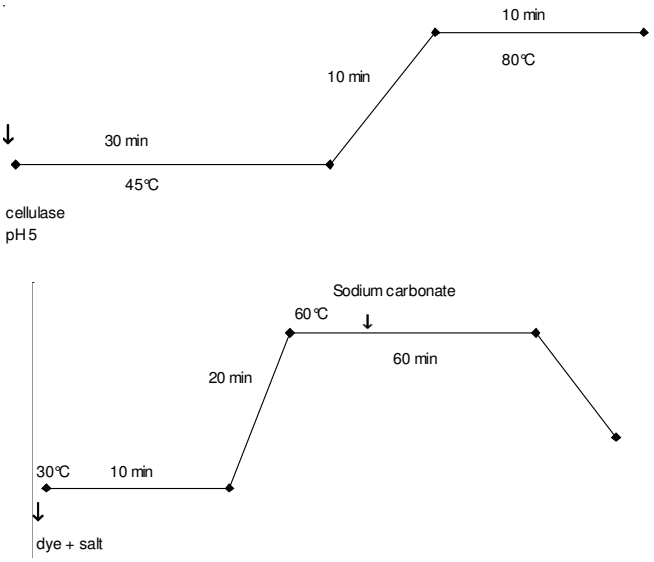


Fig. 1. Temperature-time diagrams for enzyme and dyeing processes

TABLE-2
WASHING CONDITIONS

After-treatment	Conditions
Rinsing with cold water	6 L/5 g material
Neutralization with acetic acid	pH 7
Rinsing with hot water	1000 mL/5 g material
Boil-soaping	95 ºC-2 dk-1000 mL/5 g material
Boil-soaping	95 ºC-2 min-1000 mL/5 g material
Rinsing with cold water	1000 mL/5 g material (flotte
Classical and ultrasonic washing	rate:1/2000)

RESULTS AND DISCUSSION

Basis weight and weight loss ratios: The raw materials and processed fabrics used in the experiments were first conditioned at laboratory and cut in pieces of 100 cm^2 using a fabric cutting apparatus and their mean weight was determined in accordance with TSE 251[15] upon conducting 5 measurements (Table-3).

Weight loss ratio was calculated with the following formula:

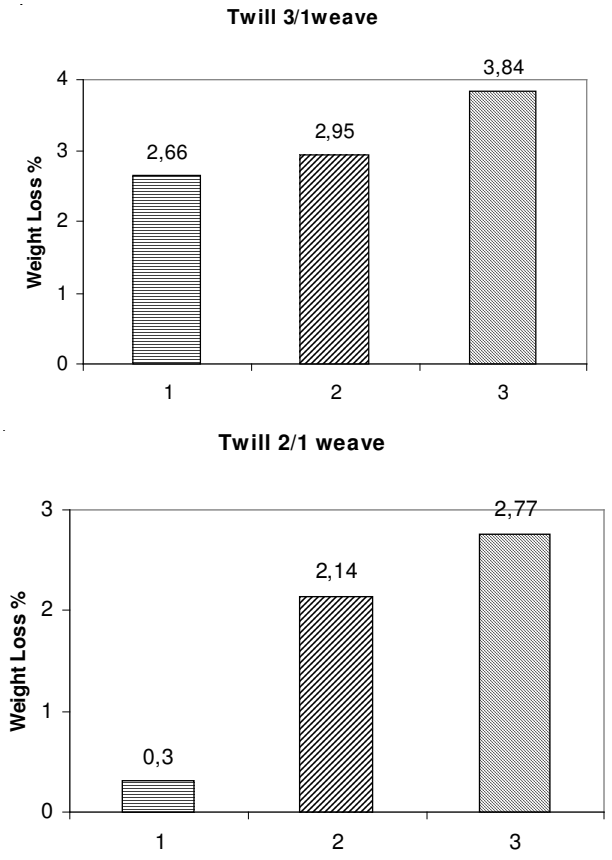
$$\text{Weight loss}\ (\%) = (W1 - W2) \times 100$$

W1: fabric weight before enzymatic process, W2: fabric weight after enzymatic process.

These values also display the hydrolysis degree of the cellulase enzyme and the cellulosic material. Fig. 2 shows the weight loss percentages of fabrics used in the research. Weight loss ratios have been determined by taking into consideration the basis weight of dyed and enzyme-treated fabrics.

TABLE-3
FABRICS WEIGHT OF BEFORE AND AFTER APPLICATIONS

	Fabric densities (g/m^2)						
Type of fabrics	Raw fabrics	Enzyme treatment after bleached	Bleached + enzyme-treatment	Only dyeing	Enzyme-treament + dyeing	Dyeing after enzyme treatment	Enzyme treatment after dyeing
Twill 3/1 Z	270	300	326	338	329	328	325
Twill 2/1 Z	272	299	321	333	323	326	324



1: Enzyme → Dyeing, 2: Enzyme + Dyeing, 3: Dyeing → Enzyme

Fig. 2. Weight loss percentages

An observation of the weight loss of the two different fabrics and the relevant hydrolysis degrees shows that the highest weight loss is realized in the enzymatic process carried out in a separate bath following the dyeing process of the materials.

Whiteness level measurements: Spectrophotometer (Datacolor International) measurements of raw, bleached and enzyme-processed fabrics were taken at an interval of 10 nm for wavelengths varying between 400 and 700 nm. The remission percentages of the fabrics are shown in Table-4. The whiteness levels of raw and bleached fabrics, which were calculated in accordance with the Berger Whiteness Index[16] using the mentioned remission percentages, can be found in Fig. 3. Whiteness level of bleached fabrics are whiter than raw fabric. It is observed that fabrics of twill 3/1 are more white.

Comparison of whiteness values of the fabrics for enzyme application on bleached fabrics shows that the enzymatic process does not affect the whiteness level of fabrics negatively. On the contrary, an increase in whiteness level of all fabrics was observed.

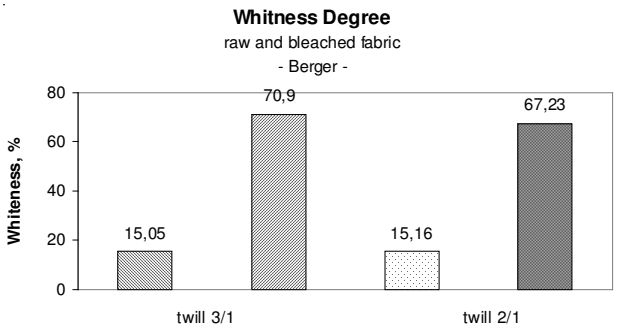
Remission percentages of bleached and enzyme-treated fabrics measured with both the classical and the ultrasonic probe methods are shown in Table-5 and the whiteness levels calculated with the help of remission values in Fig. 4.

Comparison of whiteness values of the fabrics for enzyme application on bleached fabrics shows that the enzymatic process does not affect the whiteness level of fabrics negatively. On the contrary, an increase in whiteness level of all fabrics was observed.

TABLE-4
REMISSION PERCENTAGES OF THE FABRICS

Wavelength (nm)	Remission (%)					
	Twill 3/1			Twill 2/1		
	r	b	be*	r	b	be*
400	45.28	77.79	77.69	44.96	75.35	74.60
420	48.83	80.39	80.43	48.42	78.35	77.39
440	52.22	82.62	82.67	51.77	80.83	79.58
460	55.44	84.49	84.53	54.98	82.9	81.35
480	58.56	86.00	86.06	58.02	84.55	82.76
500	61.52	87.16	87.20	60.95	85.89	83.84
520	64.24	88.00	88.02	63.63	86.88	84.70
540	66.72	88.60	88.60	66.06	87.60	85.35
560	69.04	89.10	89.09	68.35	88.22	85.95
580	71.02	89.56	89.52	70.31	88.74	86.48
600	72.96	89.90	89.86	72.21	89.17	86.88
620	74.72	90.17	90.14	73.95	89.49	87.19
640	76.39	90.45	90.41	75.61	89.80	87.47
660	77.76	90.44	90.39	76.95	89.82	87.46
680	79.29	90.73	90.69	78.48	90.13	87.76
700	80.62	91.21	91.18	79.78	90.54	88.27

r*: raw, b*: bleached, be*: bleached and enzyme-treated.



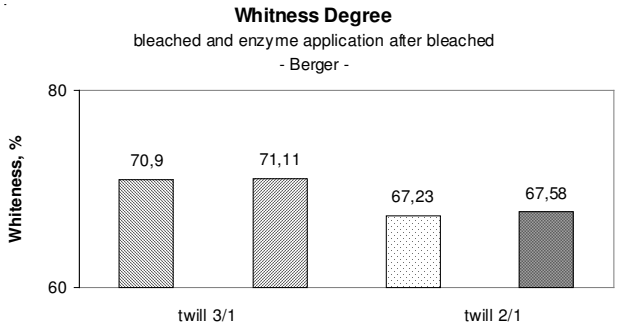


Fig. 3. Fabric of twill 3/1 and twill 2/1 whiteness levels

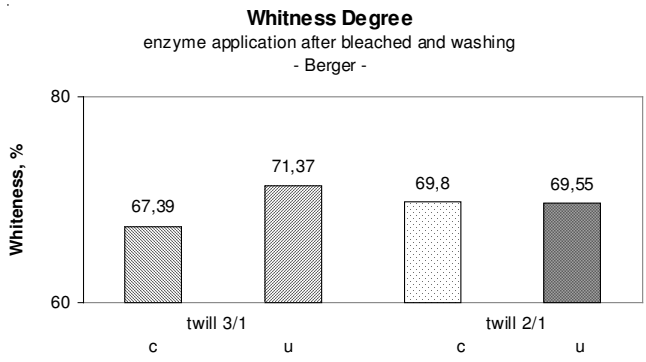


Fig. 4. Whiteness levels of fabrics of twill 3/1 and twill 2/1

TABLE-5
REMISSION PERCENTAGE VALUES OF ENZYME-TREATED FABRICS AFTER CLASSICAL AND ULTRASONIC WASHING

Wavelength (nm)	Remission (%)			
	Twill 3/1		Twill 2/1	
	c*	u*	c*	u*
400	74.99	78.14	77.18	76.87
420	77.83	80.59	80.03	79.44
440	80.05	82.74	82.44	81.59
460	81.88	84.54	84.39	83.32
480	83.35	86.01	85.99	84.73
500	84.51	87.14	87.24	85.85
520	85.40	87.97	88.17	86.72
540	86.12	88.54	88.80	87.38
560	86.73	89.02	89.35	87.97
580	87.30	89.45	89.81	88.48
600	87.74	89.79	90.21	88.89
620	88.07	90.05	90.52	89.20
640	88.37	90.32	90.84	89.48
660	88.37	90.31	90.90	89.49
680	88.68	90.60	91.29	89.81
700	89.19	91.09	91.81	90.30

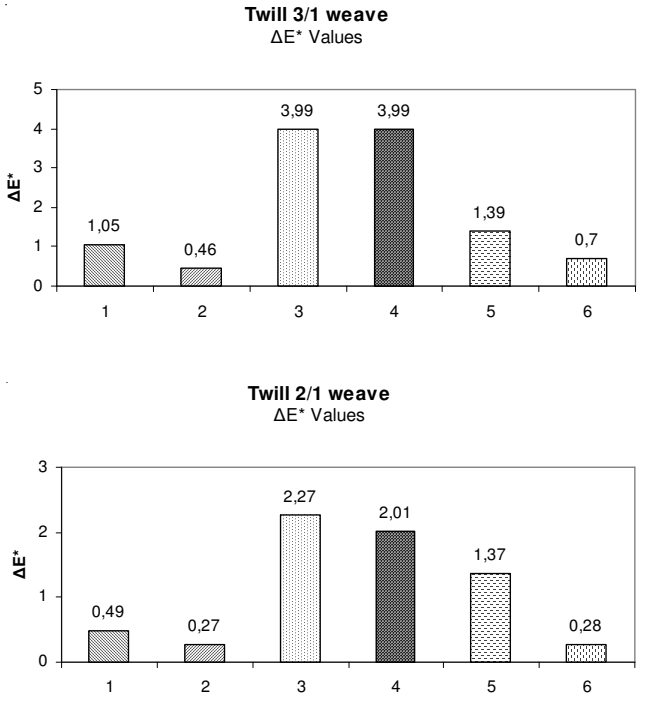
c*: Classical washing, u*: ultrasonic washing.

Remission percentages of bleached and enzyme-treated fabrics measured with both the classical and the ultrasonic probe methods are shown in Table-5 and the whiteness levels calculated with the help of remission values in Fig. 4. Comparison of whiteness levels of bleached and enzyme-treated fabrics after washing with classical and ultrasonic probe methods shows that the whiteness level usually increases.

ΔE, ΔL and ΔC values: Colour measurements of the dyed samples were conducted in accordance with the CMC 2:1 CIELab system using Datacolor Spectra Flash 600 plus reflectance spectrophotometry and Datamaster software. For the purpose of the measurements, an 10º observer and a D65 light source were also used. Table-6 shows the standards accepted within the scope of the measurements as well as the ΔE, ΔL and ΔC values of twill 3/1 and twill 2/1 fabrics. The ΔE values of the samples represent the colour difference. If ΔE < 1, the difference between the two colour is little and if ΔE > 1, the difference is substantial. Fig. 5 shows the ΔE values of treated fabrics.

TABLE-6
COLOUR CHANGING VALUES OF FABRICS

	ΔE*	ΔL*	ΔC*
Twill 3/1			
1 Dyeing without enzymes → classical washing	1.05	1.65	0.13
2 Dyeing without enzymes → classical washing	0.46	0.02	0.50
3 Dyeing without enzymes → classical washing	3.99	5.85	0.95
4 Dyeing without enzymes → usp washing	3.99	4.57	1.12
5 Dyeing without enzymes → classical washing	1.39	2.23	0.16
6 Dyeing without enzymes → usp washing	0.70	0.78	0.71
Twill 2/1			
1 Dyeing without enzymes → classical washing	0.49	-0.58	0.10
2 Dyeing without enzymes → classical washing	0.27	-0.20	0.09
3 Dyeing without enzymes → classical washing	2.27	3.38	0.71
4 Dyeing without enzymes → usp washing	2.01	2.81	0.84
5 Dyeing without enzymes → classical washing	1.37	2.23	0.04
6 Dyeing without enzymes → usp washing	0.28	0.33	0.30



1. Std: dyeing without enzymes → classical washing. Example: enzyme → dyeing → classical washing. 2. Std: dyeing without enzymes → usp washing. Example: enzyme → dyeing → usp washing. 3. Std: dyeing without enzymes → classical washing. Example: enzyme application during dyeing → classical washing. 4. Std: dyeing without enzymes → usp washing. Example: enzyme application during dyeing → usp washing. 5. Std: dyeing without enzymes → classical washing. Example: dyeing → enzyme application → classical washing. 6. Std: dyeing without enzymes → usp washing. Example: dyeing → enzyme application → usp washing

Fig. 5. ΔE values of fabrics

For all fabric types, the total colour difference value (ΔE) observed in samples on which enzyme was applied during dyeing was at unacceptable levels in case of both classical and ultrasonic washing.

(–) ΔL value shows that the sample is darker than the standard and (+) ΔL indicates that it is brighter. As can be understood from the values contained in the relevant table, all fabrics were darker than the standard but this difference was even more prominent after washing with the two methods in fabrics which were treated with enzyme during the dyeing process. A (+) ΔC value represents high chroma, *i.e.*, saturation. When ΔC values are observed, saturation turns out to be higher for fabrics dyed and treated with enzyme simultaneously.

Fastness measuring: A fastness test machine (Gyrowash/ James H. Heal Co. Ltd.) was used to test the fastness of dyed and enzyme-treated fabrics in accordance with the ISO 105-C06 standard[17]. Rubbing fastness of dyed samples was tested with a rubbing test device (Crockmeter-James H. Heal 255 A) in accordance with the ISO 105 × 12:2001 standard[18]; perspiration fastness of dyes with a perspiration fastness test device (Perspirometer-James H. Heal 290/1) in accordance with ISO 105-EO4:1994[19] and light fastness with a light fastness test device (James H. Heal) in accordance with TS 1008 EN ISO 105-B02[20]. Table-7 features the washing, rubbing, perspiration and light fastness measurements of twill 3/1 fabric and Table-8 features those of twill 2/1 fabric.

While enzyme applications had a negative impact on washing and rubbing fastness of dyes, they did not negatively

TABLE-7
COLOUR FASTNESS VALUES OF TWILL 3/1 FABRIC

	Washing		Rubbing		Perspiration		Light
	Change in colour	Fading	Dry	Wet	Acid	Base	
Only dyeing	5	4-5	5	3-4	5	5	4-5
Dyeing + enzyme	4	4	4-5	3-4	5	5	4
Dyeing → enzyme	4	4	4-5	4	5	5	4-5
Enzyme → dyeing	4	4	4	3-4	5	5	4-5

TABLE-8
COLOUR FASTNESS VALUES OF TWILL 2/1 FABRIC

	Washing		Rubbing		Perspiration		Light
	Change in colour	Fading	Dry	Wet	Acid	Base	
Only dyeing	5	4-5	5	3-4	5	5	4-5
Dyeing + enzyme	4	4	4-5	3-4	5	5	4
Dyeing → enzyme	4	4	4-5	4	5	5	4-5
Enzyme → dyeing	4	4	4	3-4	5	5	4-5

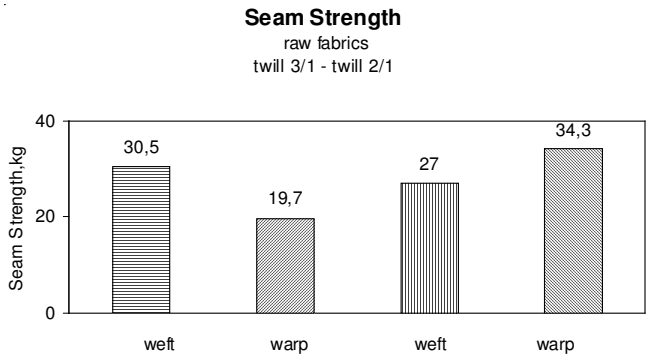
affect perspiration fastness. Light fastness was also negatively affected in fabrics simultaneously dyed and treated with enzyme.

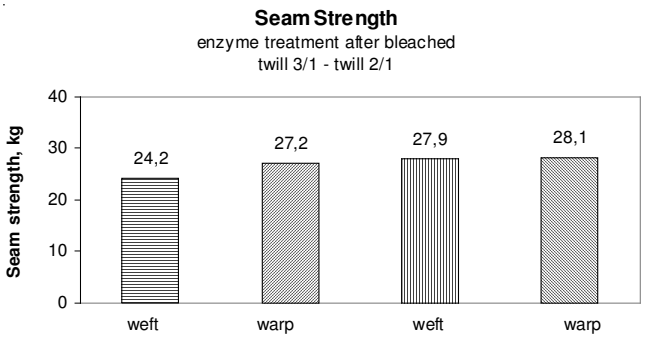
Pilling measurements: A pilling test device (Martindale SDL 2000) was used to measure the pilling levels of fabrics in accordance with TS EN ISO 12945-2[21].

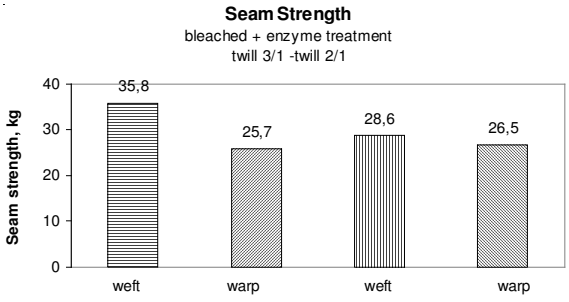
Pilling values of fabrics at cycles of 125, 500 and 2000 rpm/min, respectively can be seen in Table-9. An observation of pilling levels of fabrics shows that ultrasonic washing had a positive impact on pilling especially at cycles of 500 and 2000 rpm/min.

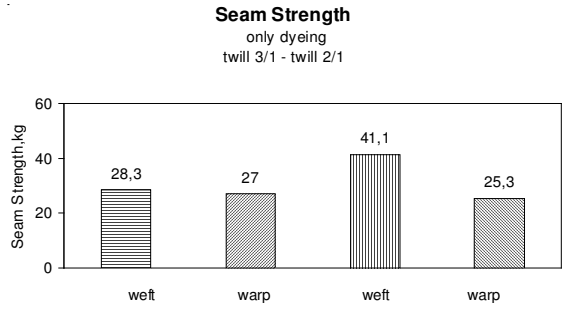
Seam slippage and strength: The materials were condition at 20 ± 2 ºC-65 ± 2 % subjected to seam slippage and seam strength tests using a resistance measurement device (Instron 4411-100 ± 10 mm/min) based on the ISO/DIS 13936-1:1998 standard[22] to find out the relevant slippage and strength values of the fabrics used in the research (Fig. 6-Table-10).

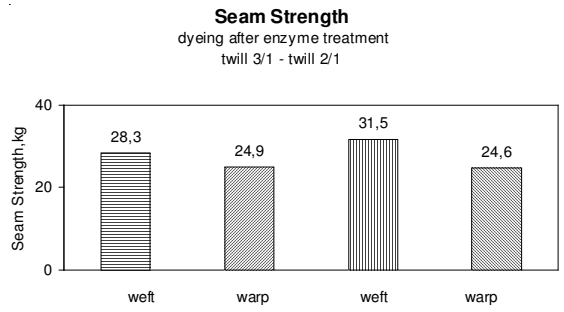
Fig. 7 is a graphical illustration of the compared seam strength values of the fabrics used in this study. In the case of











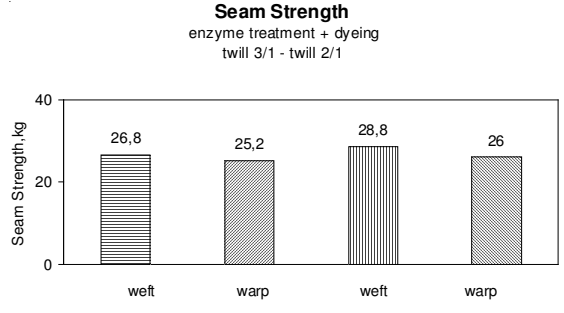


TABLE-9
PILLING VALUES OF FABRICS

	Twill 3/1						Twill 2/1					
	125 rpm/min		500 rpm/min		2000 rpm/min		125 rpm/min		500 rpm/min		2000 rpm/min	
	c*	u*	c*	u*	c*	u*	c*	u*	c*	u*	c*	u*
Bleached	5	5	4	5	1-2	2	5	5	4-5	5	2-3	3
Only dyeing	5	5	5	5	4	5	5	5	5	5	5	5
Dyeing + enzyme	5	5	5	5	4	5	5	5	5	5	5	5
Dyeing → enzyme	5	5	5	5	4	5	5	5	5	5	5	5
Enzyme → dyeing	5	5	5	5	4	5	5	5	5	5	5	5

c*: Classical washing, u*: ultrasonic washing.

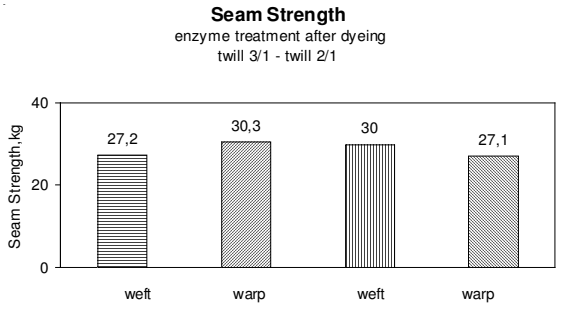
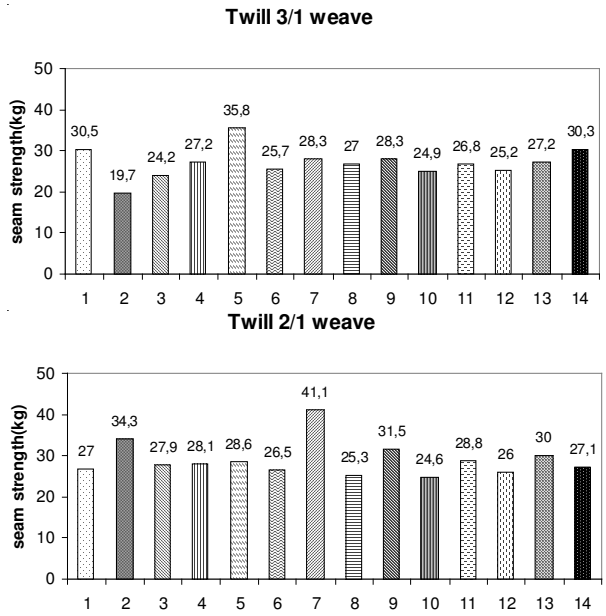


Fig. 6. Seam strength values

TABLE-10
FABRIC SEAM SLIPPAGE VALUES

	Twill 3/1		Twill 2/1	
Seam slippage 6 mm	we*	wa*	we*	wa*
Raw	10.6 ss	8.0 ss	16.6 ss	16.7 ss
Bleached	–	–	–	–
Bleached → enzyme	25.2 ss	–	–	–
Only dyeing	–	–	25.9 ss	–
Enzyme → dyeing	–	–	–	–
Enzyme + dyeing	–	–	–	–
Dyeing → enzyme	–	–	–	–

ss*: Seam slippage, we*: weft wa*: warp.



1: Raw-weft, 2: raw-warp, 3: bleached-weft, 4: bleached-warp, 5: bleaching → enzyme-weft, 6: bleaching → enzyme-warp, 7: dyeing-weft, 8: dyeing-warp, 9: enzyme → dyeing-weft, 10: enzyme dyeing-warp, 11: enzyme + dyeing-weft, 12: enzyme + dyeing-warp, 13: dyeing → enzyme-weft, 14: dyeing → enzyme-warp

Fig. 7. Seam strength graphics

samples treated with enzyme during the dyeing process, seam strength tended to decrease very little at the weft and warp directions. Seam slippage usually occurred in raw fabrics.

Conclusion

More weight loss is observed in both fabrics during the enzymatic process carried out in separate baths following the dyeing process. The rips-woven fabric also suffered more weight loss during the enzymatic process carried out in separate baths as the linking points in rips-woven fabric are less in number when compared to twill 3/1 fabric.

When the whiteness levels of bleached fabrics are compared to those of raw fabrics, the highest increase in whiteness level was observed in twill 3/1 fabric with a ratio of 2/1 due to the fact that seam slippage was less and the bleaching agent penetrated into the fabric easily.

When the whiteness levels of fabrics are compared in case of enzyme treatment of bleached fabrics, it is understood that the enzymatic process does not have a negative impact on whiteness levels of fabrics and in addition, an increase in whiteness level was observed for all fabrics. Twill 3/1 fabric displays better whiteness level than twill 2/1 fabric due to its less number of linking points.

Comparison of whiteness levels of bleached and enzyme-treated fabrics after washing with both classical and ultrasonic probe methods shows that the ultrasonic method usually led to increases in whiteness level.

When the dyeing process takes place after enzyme treatment, significant colour changes are observed when compared to cases where there is no enzyme treatment. However, change in colour decreases significantly when the ultrasonic washing method is employed at the end of the application. The colours obtained with both types of fabric after washing with the ultrasonic method are very close to those obtained in case of application without enzymes. Use of the ultrasonic method minimizes change in colour.

Carrying out the enzyme application at the same time with the dyeing process has a significantly higher level of impact on colour for both types of fabric. When compared to application without enzymes, the colours obtained after simultaneous application are brighter. This might originate from the enzyme decreasing colour efficiency or the dye being disrupted by the enzyme. Washing with the ultrasonic methods does not change the result in any way.

When enzyme treatment takes place after the dyeing process, colour change does not turn out to be significantly higher than the case with application without enzymes.

However, the resulting colour difference is still a bit higher when compared to the samples subjected to enzyme treatment before the dyeing process.

The samples obtained upon washing with the ultrasonic method had colours closer to the those obtained after application without enzymes than the samples obtained after classical washing.

In general, the ultrasonic washing process notably decreases colour change and provides colours close to those obtained after application without enzymes regardless whether the enzyme treatment takes place before or after the dyeing process. The ultrasonic washing method provides more positive results in terms of changes in colour than the classical washing method. On the other hand, experimental results show that carrying out the enzyme treatment at the same time with the dyeing process excessively decreases colour efficiency and provides negative results.

Whereas enzyme treatment affected washing and rubbing fastnesses of dyed samples negatively, it did not have any negative impact on the perspiration fastness value. Also, the enzyme negatively affected light fastness in fabrics where the dyeing process and enzyme treatment took place at the same time. As for the pilling levels of fabrics, ultrasonic washing positively affected pilling levels of bleached fabrics especially at 500 and 2000 rpm/min.

Compared to other methods, in general, very little decrease in seam strength at weft and warp directions was observed in samples where enzyme treatment took place during the dyeing process. The problem of seam slippage was usually encountered in fabrics other than raw fabrics.

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