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Dyeability and Colorfastness of Knitted Fabrics with Natural Dye Pinux^{TM} (Part I)

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Abstract

Dyeability and colorfastness of the blended knits of cotton/rayon (40/60; C/R) and wool/tencel (10/90; W/T) are examined using the natural dyestuff (Pinux^{TM}) manufactured from *Pinus radiata* pine bark extract. In addition, pre-treatments (such as bleaching, mercerization and cationization) are performed to improve dyeability and colorfastness. The Pinux^{TM} powder dyestuffs produced by Pinux Co., Ltd. are used as dyestuffs and their properties are examined for dyeing concentration (0.5-2% (owb)), dyeing time (30-120 minutes) and dyeing temperature (30-90^{o}C). Dyeability is evaluated with K/S value at 400nm, which is the maximum absorption wavelength for Pinux^{TM}. The results show the dyeability of W/T sample containing protein fiber with Pinux^{TM} is superior to all cellulose fiber C/R. A concentration of dyestuff greater than 1.5% (owb), dyeing time 120 minutes and dyeing temperature of 90^{o}C are the most optimized conditions. It shows that the dyeability of C/R and W/T samples are high in the condition of an acid-dyeing bath and that dyeability highly declined in alkaline bath due to the instability of the proanthocyanidin pigment. After analyzing the effect of bleaching, mercerizing and cationizing (as pre-treatments on dyeability) it was concluded that the dyeability of the C/R sample was enhanced by mercerization but no significant effect by cationization. However, the simultaneous treatment of cationizing and dyeing resulted in far improved dyeability compared to dyeing after cationizing pre-treatment. As for the W/T sample, the effect of cationization was more prominent than the C/R sample. Colorfastness to color changes in the control W/T sample was higher than that of C/R's level 1-2, and it increased to Level 2 when bleaching pre-treatment was given and when a simultaneous cationizing treatment was adopted to the dyeing process. Colorfastness to light in W/T control sample resulted in Level 3 and further increased to an excellent Level of 4 with bleaching and simultaneous cationizing during dyeing process.

Key words: *Pinus radiata* pine bark, Dyeability, Cotton/Rayon knits, Wool/Tencel knits, Colorfastness

I. Introduction

The natural dye industry has been receiving growing attention from the consumers for its functional characteristics such as soft colors, eco-friendliness and anti-microbial properties. However, the industry has a negative side of difficulty in color extraction and dyeing process, procuring and storing raw dye products, and color reproducibility. Also, except for few rare cases, it cannot be easily industrialized yet due to its low rate for sunlight and colorfastness. In addition to eco-friendly concept, natural dye's medicinal effect and soft natural color play an important role in the aspect of new market development and natural resource utilization. If new natural dyeing method combined with modern advanced technology is developed, it can be anticipated to play a huge role in not only cloth dyeing but in development of natural pigment, perfumes, medicine and environment protection as well (Chang et al., 2000).

There are numerous studies being conducted in Korea

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order to fix problematic factors of natural dye and to help vitalize it. To increase natural dye's safety, studies such as natural dye confrication and standardization (Son et al., 2011), natural dye's urgent problem and prospects using Delphi method (Roh & You, 2006), and more had been published. There have been many reports proposing utilization of natural mordant such as bittern (Yoo et al., 1997), tannin (Kim, 2001), alum, vinegar, bean juice (Park et al., 2007), and chitosan treatment (Hong et al., 2010), or synthetic mordant like Cu, Fe, Al, Mg to improve natural dye's dyeability and colorfastness. Moreover, in order to heighten dyeing affinity of cellulose fabric and natural dyestuff, a method of instituting anion group to cationized cotton fabric to induce chemical bonding with anion of dye is being introduced (Lee et al., 1990; Sung, 2003; Sung et al., 1997).

As for the studies researching natural dyeing process using pine tree as a dye material, there is a dyeability research using pine needle extracts. Joen and Park (2009) extracted dye material from pine needles using water and ethanol, and contrasted dyeability and colorfastness with silk, wool, nylon and soybean fiber. As a result, needle extract dye goods' colorfastness for washing and sunlight was generally low, but antibacterial performance and ability to block UV light was excellent (Joen & Park, 2010). Also, Woo and Lee (2011a) compared dyeability and colorfastness of extracts from pine needles using methanol applied to cellulose fiber and protein fiber, and researched dye goods' functionalities such as antibacterial properties, and ability to deodorant and block UV light (Woo & Lee, 2011b).

More than 80% of wood consumption is relied on import in Korea. Import especially *Pinus Radiata* from New Zealand and Australia is being increasingly expanding. Due to a recent decrease in broadleaf tree import because of Oceania timber lumber regulations and price pike closely related to tropical forest protection movement, the import volume of New Zealand and Australian Radiata Pine hardwood which mainly composed of conifers is now exceeding 3 millionm^3 a year. Currently, *Pinus radiata*, which is mainly used for construction, temporary building, civil works, packaging and pulp production, accounts for about 50% of imported hardwood. Considering the bark amount of pine that derive from pulp-paper manufacturing industries and other wood-related manufacturers in the country, the amount of bark being wasted is expected to be some hundred thousand tones (Mun & Hassan, 2002). A part of the wasted bark is being reused as soil amendments or reinforcement materials, but the rest is in the state of being incinerated and buried. Late increased attention to ecological issues have set national research studies to recycle wood bark (Mun & Hassan, 2002; Mun & Ku, 2006). Domestic venture business, Pinux Co., is developing antioxidative products and cosmetics using bark of pine tree. Using anthocyanidin pigment extracted from bark of pine tree, they are producing and selling powdered products as Pinux^{TM}. According to preceding studies with Pinux Co. we have researched Pinux$^{TM'}$ component analysis and colorfastness and dyeability with silk fabrics (Song et al., 2009).

Nowadays, knit, including jersey, has been favored for women's fashion clothing material, and it dominates innerwear and underwear industries. Not only are knit's natural dye products eco-friendly, but they also can be recommended to minimize skin irritation for consumers like children, elders and those with atopic dermatitis who may well be searching for hypoallergenic components. Current Korean natural dye research is mainly on woven fabrics, so it lacks studies on knits (Shin & Yu, 2004; Yu, 2009). According to research results by Yu (2009) about dyeing affinity of cationized cotton fabrics and cotton knit with volcanic ashes, they reported that dyeing affinity was excellent in knit structure due to good surface adsorption property with dyestuff better than in woven structure.

It has been known that among the main components of pine bark, proanthocyanidin (PA) which has antioxidative ability comprises 64% (Ku & Mun, 2007), and the silk cloth dyed with pine bark dyestuff has shown high antimicrobial property of 99% (Song et al., 2009). As a result, natural dye application research using pine bark dyestuff that bears functional pigment and its properties can be anticipated to have a various usage in different areas such as infant clothing, inner wear, bedclothes, and so on

In this research, the dyeability and colorfastness of cellulose fiber mixture knit and protein fiber mixture knit using powdered natural dye from pine bark extracts were experimented. Furthermore, the effect of pre-treatments such as bleaching, mercerizing and cationizing for dyeability and colorfastness improvement was analyzed.

II. Experimental

1. Test Samples and Reagents

1) Test Samples

Cotton/Rayon (C/R) knit and Wool/Tencel (W/T) knit's characteristics that were used in this experiment are as shown in <Table 1>.

2) Dyestuff

Pinux^{TM} powder dyestuff from radiata pine (*Pinus radiata*) was provided by Pinux Co., Ltd. The chemical structure of Pinux^{TM} is as <Fig. 1>. It has been found that the main ingredient of dye is flavonoids series pigment proanthocyanidin whose benzene ring has 2 numbers of hydroxyl groups, and Radiata pine contains 64% of proanthocyanidin which has antioxidant property (Ku & Mun, 2007).

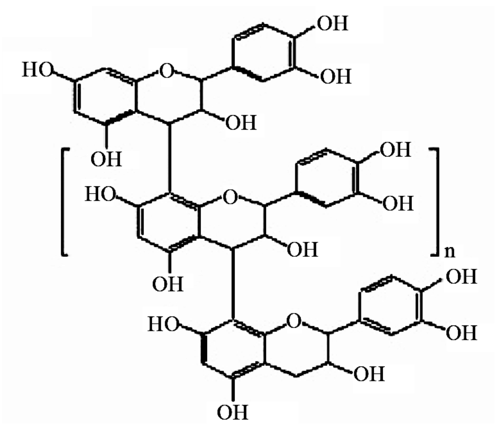

Fig. 1. Chemical structure of proanthocyanidin (PA) isolated from *Pinus radiata* bark.

2. Methods

1) Dyeing

Various dyeing processes were performed by making dye bath ratio 1:50 with dye concentrations 0.5, 1.0, 2% (owb) under temperature of 30°C, 50°C, 70°C, and 90°C for the duration of 30 min, 60 min, 90 min and 120 min respectively. Also, dyeabilty was analyzed by changing pH (5, 7, 9, 11, and 13) of dye bath using acetic acid and sodium hydroxide.

2) Pre-Treatment

For the purpose of pre-treatment effect on dyeability examination, beaching, mercerizing and cationizing treatments were given to sample cloths either individually or stage wise.

(1) Bleaching

Fabric bleaching was done with $H_2O_2$15% (owf), NaOH 3% (owf), Hydrogen peroxide stabilizer 3% (owf) under bath ratio 1:50, 60°C temperature for the duration of 30minutes followed by washing and drying.

(2) Mercerizing

Mercerizing was done only on C/R sample using NaOH 20% (owb) solution under ambient temperature for 5 minutes followed by neutralizing with diluted acetic acid and washing with water.

(3) Cationizing

Cationizing was done on the sample with 20% (owf) cationizing agent, NaOH amount equivalent to 30% of cationizing agent (Snogen CAT-800 Snogen Corp), 2% (owf) refining-permeating agent (Snogen GS-35, Snogen Corp), and 2% (owf) hydrogen peroxide stabilizer (Snobil DS, Snogen Corp) with dye bath ratio 1:50 under temperature 80°C for 30 minutes. After the treatment, sample was washed and then dried (Kim & Kim, 1996; Lee et al., 1990).

3) Dyeability Measurement

Using color meter (JS-555), Pinux^{TM} pine bark dye's

Table 1. Characteristics of C/R and W/T sample

Sample	Fiber Content	weave	Density (inch)		Weight (g/m²)	Thickness (mm)
			Wale	Course		
C/R	Cotton/Rayon (40/60)	Plain stitch	45	56	28.05	0.340
W/T	Wool/Tencel (10/90)	Plain stitch	33	44	25.10	0.330

dyeabiliy was measured with K/S value with 400nm as reference wavelength which is the maximum absorption wavelength of the subject. Surface color was also compared by measuring L*, a*, b* , H, and V/C.

4) Colorfastness Measurement

Colorfastness to washing was measured as per KS K ISO 105-C-01:2007, colorfastness to light was measured using XENON-ARC-LAMP as per KS K ISO 105-B02:2010 for 20 hours, color fastness to perspiration was measured with acidic solution as well as alkaline solution as per KS K ISO 105-W04L2010, and colorfastness to rubbing was measured as per KS K 0650:2006.

III. Results and Discussion

1. Dyeability

1) Dyeability by Dye Concentration

K/S value of each sample cloth depending on dye concentration is shown in <Fig. 2>. With increase in dye concentration, K/S values were increased a bit, but with dye concentration greater than 1.5%, K/S value increase almost ceased. On the contrary, W/T sample showed good dyeability even under low dye concentration of 0.5%, but with an increase in dye concentration K/S values were abruptly increased from 5.35 with 0.5% dye concentration to 11.92 with 2% dye concentration showing a high dyeing affinity. The result suggests that dyeing on C/R sample simply lets the dye penetrate into pore inside the fabric to make coloring done via dyeing mechanism of cellulose and hydrogen bonding. Whereas in W/T sample, the mechanism of coloring power of wool fabric have affected largely on dyeing affinity. In other words, dyeing mechanism by which cation amine group, which is dyeing site of wool fabric and is anion component in dye, form salt linkage (Song & Baik, 2002) and tencel, which is mixed in large in sample, has characteristics of water penetrating inside the fabric as compared to cotton, and thus hydrogen bonding or van der waals forces were generated more easily under high dye concentration attributing to high dyeing affinity.

2) Dyeability by Dyeing Temperature

<Fig. 3> shows samples' dyeing results from different temperature settings of 30°C, 50°C, 70°C and 90°C with same bath ratio of 1:50, dye concentration of 1.5% and duration of 90min to analyze dyeability by dyeing temperature. As for the C/R sample, the K/S value increased a little as the temperature rose, but there was no significant difference noted above 50°C. On the other hand, the W/T sample's K/S value increased tremendously up to 70°C, and in temperature above that level the increase level has slowed down. This result was similar to that of preceding research on dyeability test with silk fabric using Pinux^{TM} (Song et al., 2009). It may be due to loosened molecular structure inside the fabric with increased temperature to make molecular movement activated which enables faster dye diffusion speed inside the fabric (Cho, 1997), and salt linkage between amine group and dye could be formed more easily. Also, dyeability may be improved with easiness of dye solution penetrating into tencel fabric.

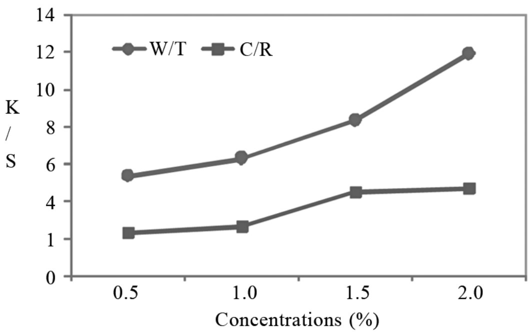

Fig 2. Effect of dyeing concentration on dye uptake (90°C, 60 min).

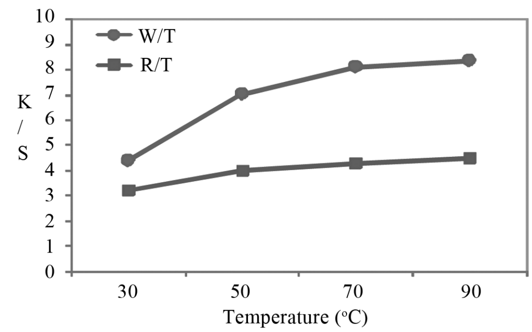

Fig. 3. Effect of dyeing temperature on dye uptake (1.5% owb, 60 min).

3) Dyeability by Dyeing Duration

<Fig 4> shows samples' dyeability results from different time duration of 30 min, 60 min, 90 min and 120 min with same bath ratio of 1:50, dye concentration of 1/5% and temperature of 90°C to experiment if there is a dyeability difference depending on dyeing duration. For both samples, dyeing affinity has improved as the time duration increased. For W/T sample, K/S value was increased with dyeing duration of 60 min and slowed down after 60 min has elapsed. Generally with an increase in dyeing time, it shows saturation (Jo & Lee, 2004). In C/R sample dyeability also showed similar curve as in W/T followed by steady dyeability rate when the duration became longer than 90 min.

4) Dyeability by pH of Dye Bath

<Fig 5> shows samples' dye results under various pH dye bath conditions but the same 1% (owb), 90°C, and 60 min duration dyeing condition. Both samples resulted in good dyeability under acidic environment of pH 5. As the pH level increased, the dyeability decreased significantly; with pH of 13, both C/R and W/T samples' K/S values showed low dyeability of less than 1.0. It could be a result of instability of Pinux™ dye's color component, proanthocyanidin (PA), caused by alkaline bath. That is, color became unstable due to C-ring opening in PA caused by alkali (Ku & Mun, 2007). Yellow series color in neutral dye bath was changed to pink series color in alkali dye bath. The color C/R: 6.8 YR, W/T: 8.75YR appeared as C/R: 2.42YR, W/T: 2.14YR in alkali dye bath. Therefore it would better to carry out Pinux™ dyeing in somewhere between weak acid to neutral dye baths.

5) Pre-treatment Effect on Dyeability

<Table 2> shows resulting colors and K/S values of samples that received pre-treatments of bleaching, mercerizing, and cationizing. Munsell values of C/R sample dyed with Pinux™ without pre-treatments was 8.26Y, and this color change almost did not take place after

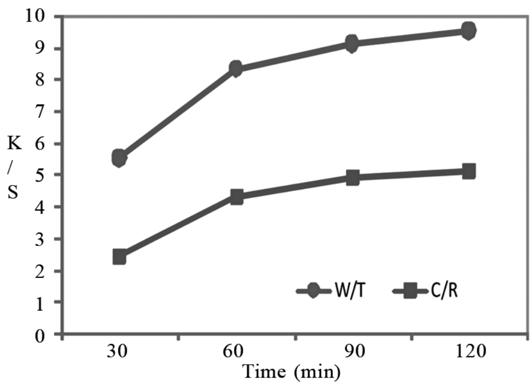

Fig. 4. Effect of dyeing time on dye uptake (1.5% owb, 90°C).

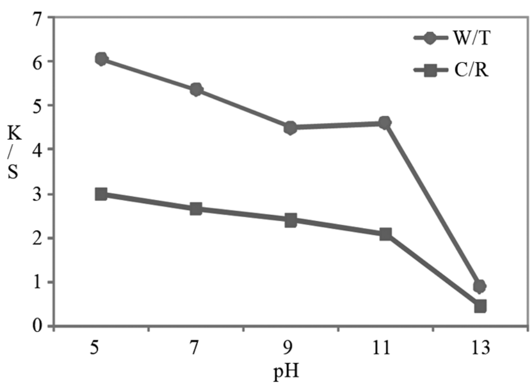

Fig. 5. Effect of pH on dye uptake (1% owb, 90°C, 60 min).

Table 2. L*, a*, b*, H, V/ C, and K/S values of C/R and W/T sample dyed with Pinux™ after various pre-treatment

Sample	L*		a*		b*		H		V/C		K/S	
	C/R	W/T	C/R	W/T	C/R	W/T	C/R	W/T	C/R	W/T	C/R	W/T
Control	56.02	46.99	8.10	16.45	11.7	20.73	8.28Y	5.39Y	5.64	4.35	4.25	8.24
B*	61.52	48.71	10.87	14.57	19.83	17.09	9.13Y	6.03Y	5.98	4.23	3.54	8.12
M**	53.06	-	13.78	-	23.23	-	7.28Y	-	5.15	-	7.99	-
C***	57.15	42.05	8.97	11.66	22.18	19.25	9.84YR	7.68YR	5.55	4.08	4.78	9.98
C+D****	52.58	40.24	12.17	10.56	22.47	19.24	7.95Y	7.21YR	5.38	4.02	5.93	10.68

*Bleaching, **Mercerizing, ***Cationizing, ****Cationizing & Dyeing bleaching with a value of 9.13Y. After mercerizing C/R, the color changed into brown series (7.28Y), but it became 9.84YR with cationizing treatment. In W/T sample, control showed 8.28Y, bleaching pre-treatment 6.03Y, and cationizing pre-treatment 9.84YR respectively.

Dyeability improvement was observed in K/S values from C/R samples in the order of bleaching (3.54)< control (4.25)<cationizing (4.78)<<mercerizing (7.99). Cationization effect was not noticeable, whereas, mercerizing effect had a great impact on improving dyeability. This may be due to swelling of cellulose fabric which consequently relaxed fabric structure that maximized fabric's amorphous region from mercerization (Lim et al., 1997). It has been expected that dyeability would significantly improve by cationizing pre-treatment inducing cation into cellulose fabric to make it chemically bonded with dye's anion (Shin & Yu, 2004), but the effect could not be achieved in this experiment with Pinux^{TM} dye. On the contrary, when cationization and dyeing took place at the same time, the dyeability had much better outcome than with pre-treatment. It was assumed that cation group could be bonded with dye more efficiently while dyeing in one bath at the same time cationizing took place to introduce cation. As for the W/T sample, K/S values showed larger cation effect than C/R as following: bleaching (8.12), control (8.24) <cationizing (9.98).

2. Colorfastness

1) Colorfastness to Washing

<Table 3> shows C/R sample's colorfastness to washing. Control sample's colorfastness to washing showed high level of 4-5 in stain for cotton and rayon fabrics, while colorfastness to color changes showed low level of 1. Though colorfastness to color changes by both mercerizing and cationizing pre-treatments was not improved, the sample became level 1-2 by bleaching. And by adding cationizing pre-treatment and dyeing together, colorfastness to color changes improved to level 2. Same as above, dyeing along with cationizing agent was effective in colorfastness rather than dyeing after pre-treating.

W/T sample's colorfastness to washing was a little higher than that of C/R. Colorfastness to color changes in control group was level 1-2, and this was improved to level 2 by bleaching and simultaneously cationizing and dyeing. Colorfastness to stain was high to the level of 4-5 in both control group and treated group. Colorfastness to washing in both C/R and W/T samples with bleaching pre-treatment suggests that residual pigments remained in samples might had been removed effectively by bleaching. Also, it is assumed that dyeability and colorfastness improvement when cationizing pre-treatment and dyeing took place simultaneously might have attributed to increasing efficiency of bonding between introduced cation and dye.

2) Colorfastness to Light

<Table 4> shows the result of colorfastness to light in control group and pre-treated C/R and W/T samples. Colorfastness in C/R control group showed level 1-2, whereas bleached sample and sample that was cationized and dyed altogether improved to level 2. However, with mercerizing, although dyeability was significantly improved, no improvement on colorfastness to washing and to light was noted. Colorfastness to light in control W/T sample was excellent compared to C/R sample and showed level 3. When it was simultaneously treated

Table 3. Colorfastness to washing

Sample	C/R			W/T		
	Color change	Stain		Color change	Stain	
		Cotton	Rayon		Rayon	Wool
Control	1	4-5	4-5	1-2	4-5	4-5
B*	1-2	4-5	4-5	2	4-5	4-5
M**	1	4-5	4-5	-	-	-
C***	1	4-5	4-5	1-2	4-5	4-5
C+D****	2	4-5	4-5	2	4-5	4-5

*Bleaching, **Mercerizing, ***Cationizing, ****Cationizing & Dyeing

Table 4. Colorfastness to light

Sample	Color change	
	C/R	W/T
Control	1-2	3
B*	2	3-4
M**	1-2	-
C***	1-2	3
C + D****	2	4

*Bleaching **Mercerizing, ***Cationizing, ****Cationizing & Dyeing

with bleaching and cationizing, it further improved to level 4.

3) Colorfastness to Perspiration

<Table 5> shows colorfastness to perspiration in C/R and W/T samples after pre-treatment. First, colorfastness to color changes in acidic sweat solution in control C/R sample showed level 2-3, and the level improved a little by receiving all three pre-treatments, bleaching, mercerizing, cationizing and when cationizing and dyeing were received at the same time. However, colorfastness in alkaline sweat solution had a slight improvement in colorfastness to stain but no pre-treatment effect was noted for colorfastness to color changes.

As for the W/T sample, colorfastness to perspiration in acidic sweat solution was observed to be the same as C/R sample in all pre-treatment blocks, i.e., bleaching, cationizing after bleaching, cationizing, and cationizing along with dyeing. Alkali sweat solution also showed a slight increase in colorfastness to sweat in all pre-treatment blocks. As explained above, pre-treatment effect was greater in colorfastness to perspiration as compared with other colorfastness characters.

4) Colorfastness to Rubbing

<Table 6> shows C/R and W/T samples' results for colorfastness to rubbing. For C/R sample, colorfastness to dry rubbing was excellent with level 4-5 in control group, but there was no improvement in pre-treatment

Table 6. Colorfastness to rubbing

Sample	C/R		W/T	
	Dry	Wet	Dry	Wet
Control	4-5	3-4	4-5	4
B*	4-5	3-4	4-5	4
M**	4-5	3-4	-	-
C***	4-5	3-4	4-5	4
C+D*****	4-5	3-4	4-5	4

*Beaching, **Mercerizing, ***Cationizing,****Cationizing & Dyeing

Table 5. Colorfastness to perspiration

C/R						
Sample	Acidic			Alkalic		
	Color change	Stain		Color change	Stain	
		Cotton	Rayon		Cotton	Rayon
Control	2-3	3-4	3-4	2-3	3	3-4
B*	3	3-4	3-4	2-3	3	3-4
M**	3	3-4	3-4	2-3	3	3-4
C***	3	4	3-4	2-3	3-4	3
C+D****	3	3-4	3-4	2-3	3-4	3

W/T						
Sample	Acidic			Alkalic		
	Color change	Stain		Color change	Stain	
		Rayon	Wool		Rayon	Wool
Control	3	4	3-4	3	3	3-4
B*	3-4	3-4	3-4	3-4	3	3-4
M**	-	-	-	-	-	-
C***	3-4	4	3-4	3-4	3-4	3-4
C+D****	3-4	3-4	3-4	3-4	3-4	3-4

*Beaching, **Mercerizing, ***Cationizing, ****Cationizing & Dyeing group. For wet rubbing, both control and pre-treated groups showed level 3-4.

Colorfastness to rubbing in W/T sample was level 4-5 in dry rubbing and level 4 in wet rubbing. Pre-treatment effect on colorfastness to rubbing was not prominent in W/T sample.

IV. Conclusions

Research study was conducted to find out dyeability and effect of pre-treatment beaching, mercerizing, and cationizing on C/R, W/T knits using natural dye Pinux™ extracted from *Pinus radiata* pine bark. The results are as below.

Dyeability of Pinux™ resulted in far more excellent performance in protein series mixed spinning W/T samples than cellulose series C/R sample. The optimum dyeing condition for C/R sample was dye concentration of 1.5% (owb), temp 50-70°C and duration of 60 min, and for W/T sample was dye concentration of 2% (owb), temp 70-90°C and duration of 60 min. Both samples showed high dyeability in weak - neutral acid dye bath, and dyeability significantly decreased under alkali dye bath due to dye's instability.

After analyzing the effect of bleaching, mercerizing and cationizing pre-treatments on dyeability, it was concluded that the C/R sample's dyeability was greatly enhanced by mercerization but no significant effect by cationization. However, simultaneous treatment of cationizing and dyeing resulted in far improved dyeability compared to dyeing after cationizing pre-treatment. This is thought to be due to more efficiently bonded cationization and dye from simultaneous cationizing and dyeing being done in one bath process. As for the W/T sample, the effect of cationization was more prominent than C/R sample.

Colorfastness to color changes for washing in C/R sample showed as low as level 1. Thought colorfastness to color changes by mercerizing and cationizing was not improved, when bleaching treatment was given its value increased to level 1-2 and simultaneous treatment of both cationizing and dyeing helped improving level to reach 2. Colorfastness to color changes in control W/T sample was higher than that of C/R's level 1-2, and it was increased to level 2 when bleaching pre-treatment was given and when simultaneous cationizing treatment was adopted to the dyeing process. However, mercerizing did not attribute to colorfastness to washing and to light despite how dyeability has improved. Colorfastness to light in W/T control sample resulted in level 3 and this level was further increased to an excellent level of 4 with bleaching and simultaneous cationizing during dyeing process.

Colorfastness to color changes in acidic sweat solution of control C/R showed level 2-3 and this level increased a little in all the treatment block of bleaching, mercerizing, cationizing and simultaneous cationizing with dyeing. In W/T sample, with acidic sweat solution, colorfastness to sweat was also improved a little in all treatment block.

C/R and W/T both showed a great level of 4-5 on colorfastness to rubbing, but there was not significant improvement with pre-treatment. Same result was found that pre-treatment had no effect on dyeability for colorfastness to wet rubbing.

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