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Optimization of parameters for application of sericin on cotton knits

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In this study, a process has been developed for durable coating of sericin on cotton knits. Treated samples are tested for wicking, stiffness, moisture vapour transmission rate and air permeability. Results show that cotton knits become more hydrophilic on application of sericin. Air and water vapour transmission improve, thus making the cotton sample suitable for applications in skin moisturizing and skin healing. These results indicate that sericin can be used to develop a durable finish on cotton for use in medical and sports garments.

Keywords: Air permeability, Cotton, Sericin, Medical garments, Sports garment, Water vapour transmission

1 Introduction

Silk sericin is a water soluble globular protein derived from *Bombyx mori* silk worm[1]. It is made up of amino acids having strong polar side groups such as hydroxyl, carboxyl and amino groups[2]. The total amount of hydroxy amino acids in sericin is 45.8%. There are 42.3% of polar amino acid and 12.2% of non-polar amino acid residues[3]. Sericin, a waste product of the silk industry[4], has been attributed with several unique properties such as biocompatibility, biodegradability[5], nontoxicity, UV resistance, moisture absorption[6], antioxidant[7] and antimicrobial activity[8]. Because of these properties, it has wide applications in industries such as medical, textiles, pharmaceutical and cosmetics[9]. Some researchers have reported studies on applying sericin as a finishing agent on natural and man-made textiles[10]. Kongdee *et al.*[11] modified cotton fabric with silk sericin. They found that the electrical resistivity of the samples decreased and water retention increased, which made the sericin treated fabric more comfortable to wear. In another study[12], crosslinking agents such as glutaraldehyde and dimethyloldihydroxyethylene urea have been used to fix sericin on cotton fibres. Finished fabric showed improved moisture absorption ability with increased durability. Rajendran *et al.*[1] coated sericin on cotton fabric and found these fabrics showed high degree of bactericidal activity against both *E. coli* and *S. aureus.*

This study was undertaken with the objective of developing textiles which can be used bypatients suffering from specific medical conditions such as skin itching, rashes and atopicdermatitis. Atopic dermatitis is a chronic itchy, inflammatory skin disease that is extremelydifficult to treat[13]. Although effective therapeutic agents are available, they may have long-term toxic side effects[14]. Wearing clothes of smooth surface is one of the non-drug based approach for avoiding the disease as it has been suggested that such clothing have high moisture sorption and smoother surface that can be used for patients suffering from such ailments[15]. In this study, sericin was applied to cotton knits and treated fabrics were tested for moisture related properties and comfort characteristics.

2 Materials and Methods

2.1 Materials

Soap alkali degumming liquor was procured from Nath Brothers Exim International Ltd., Noida, U.P, India. The liquor was converted into sericin powder using a laboratory spray dryer LU-227 Advanced (Labultima, India) at inlet temperature of 110°C and the atomization pressure of 3-4 kg/ cm^2. Creamish white sericin powder was obtained with 15.1% nitrogen content and 94.4% protein content. The molecular weight ranged from 60 kDa to 205 kDa with the particle size of 3.6 micron.

Face-to-face cotton knit fabric was procured from the Advantage Organic Natural Technologies Pvt. Ltd., Unit-V, TBIU Synergy-Solarium, IIT Delhi. The specifications of the fabric are 40 wales/inch,

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48 courses/inch and GSM 119. Monosulfonated acid dye, Texacid Sulphone Blue BR was used for staining of cotton. Laboratory grade Alum (KAl $(SO_4)_2$ $12(H_2O)$) (Merck, USA) was used for fixation of sericin on cotton. Deionized water was used for all other purposes.

2.2 Methods

2.2.1 Pretreatment of Fabric

Cotton knit fabric was scoured with 1 gpL Lissapol N for 1 h at boil followed by rinsing and drying. The fabric was then bleached with 15 gpL hydrogen peroxide for 45 min at boil, then rinsed thoroughly and dried.

2.2.2 Application of Sericin on Cotton Knits

Sericin was applied on cotton knits with and without the use of complexing agent. The process parameters, viz. sericin concentration, time and temperature of treatment, were optimized.

Application of Sericin without Complexing Agent

Sericin was applied on conditioned cotton knits by exhaust method in high temperature high pressure beaker dyeing machine (R.B. Electronic and Engg. Pvt. Ltd, India). The material-to-liquor ratio was kept as 1:20. The three process factors chosen for this study were sericin concentration, treatment time and treatment temperature and to optimize the conditions of application, the experimental design as explained hereunder was followed.

To optimize the conditions of application of sericin, a 3^3 Box and Behnken Design (BNB) was used. Factors used in the experimental design were sericin concentration, treatment time and treatment temperature. These factor levels were evenly spaced and coded as –1 (low), 0 (central point) and +1 (high) as shown in Table 1. The treated samples were stained with acid dye to estimate the amount of sericin deposited. The higher the colour value, the more is the amount of sericin applied. The selected responses were add-on, colour value (*K/S*) and bending length of treated samples.

Fifteen experiments were carried out on cotton knits with the above mentioned conditions. The treatment conditions are given in Table 2. The analysis of data was carried out using design expert software, version 8. The module applied a quadratic polynomial equation to analyze the relation of each response with the independent variables, as shown below:

$$Y = b_0 + b_1X_1 + b_2X_2 + b_3X_3 + b_4X_1X_2 + b_5X_2X_3 + b_6X_1X_3 + b_7X_1^2 + b_8X_2^2 + b_9X_3^2 + E \quad \ldots (1)$$

where b_0 - b_9 are the regression coefficients; X_1, X_2 and X_3 are the factors studied ; E, an error term, and Y, the measured response. The statistical significance of the model equation and the multiple coefficient of determination (R^2) were also determined. After application, the fabric samples were dried in hot air dryer provided by Advantage Organic Natural Technologies Pvt. Ltd., Unit –V, TBIU Synergy – Solarium, IIT- Delhi.

Application of Sericin with Complexing Agent

Since sericin does not have any affinity for cotton fabric, it cannot be directly attached to cotton fabric. Alum has long been used for its medicinal properties and has been used in dyeing as mordants. Therefore, alum was selected as a complexing agent to attach sericin on cotton surface. Sericin was applied by using the optimized conditions and then post-treated with different concentrations of alum viz. 2, 4, 6 and 8% (owf). The treatment was carried out at 60^0C for 15 min, maintaining M.L.R at 1:20. The fabric was then rinsed with cold water.

2.3 Characterization of Sericin Treated Fabric

All the samples were conditioned for 48 h before testing. Weight add-on %, sericin adsorbed on cotton

Table 1—Variables used in the Box and Behnken design

Variable	Levels		
	Low (-1)	Medium (0)	High (+1)
Sericin concentration, gpL (X_1)	5	15	25
Time, min (X_2)	30	60	90
Temperature, ^{0}C (X_3)	60	70	80

Table 2—Box and Behnken surface response design for application of sericin on cotton knits

Run	Sericin concentration gpL (X_1)	Time min (X_2)	Temperature ^{o}C (X_3)
1	25	60	60
2	15	30	60
3	15	60	60
4	5	60	60
5	5	30	70
6	25	30	70
7	25	90	70
8	5	90	70
9	25	60	80
10	15	90	80
11	5	60	80
12	15	30	80
13	15	60	70
14	15	60	70
15	15	60	70

fabric, durability of finish and testing of physical and comfort characteristics are discussed hereunder.

2.3.1 Weight Add-on %

Add-on % was determined by measuring the difference in the weight of conditioned untreated sample and sericin treated sample, as shown in the following equation:

$$\text{Add-on (\%)} = 100 \times [(W_2 - W_1)/W_1] \quad \ldots (2)$$

where W_1 is the initial weight of the unfinished sample; and W_2, the final weight of the finished sample

2.3.2 Estimation of Absorbed Sericin on Cotton Fabric

Since sericin is composed of amino acids that contain NH_3^+ groups, it can be treated with acid dyes. Acid dye forms electrovalent bonds with amide groups present in sericin. Since cotton does not have any chemical group capable of reacting with anionic acid dyes, the dye taken up by sericin treated fabric can be taken as an indirect measure of the amount of sericin applied on the fabric.

Sericin treated fabrics were stained with 0.5% monosulfonated acid dye, Texacid Sulphone Blue BR at a liquor ratio 1:50, *p*H 5.5, duration 10-15 s and temperature 90^0C. Untreated cotton fabric was taken as a positive control. The stained samples were then washed in water and dried. Colorimetric measurements of the dyed samples were performed on a Color-Eye 7000A computer colour matching system (Gretag Macbeth, USA). Readings were taken in triplicate and the average K/S_{corr} values were recorded according to the following equation:

$$(K/S)_{corr} = (K/S)_{sample} - (K/S)_{blank} \quad \ldots (3)$$

2.3.3 Durability of Finish

Durability of sericin finish applied by using a complexing agent was assessed by washing the dyed sample with 5 gpL of Lissapol N for 15 mins. at 45°C. The samples were washed upto 5 washing cycles. After each wash a sample was taken out, dried and then stained with acid dye followed by the measurement of *K/S* value.

2.3.4 Testing of Physical Characteristics

(i) *Bending Length*

The bending length of finished fabric was measured according to the ASTM D 1388-96. The measurements were carried out on stiffness tester (Paramount, India).

(ii) *Vertical Wicking*

Wicking property of samples was evaluated using the wicking strip test according to standard DIN53924. Specimen of size 200 mm × 25 mm with the length parallel to wale direction was prepared. Each specimen was suspended vertically with its lower end immersed in a reservoir of distilled water to which dilute potassium chromate (0.5%) was added for tracking the movement of water. Potassium chromate was chosen instead of a dye because it is an inorganic salt without having any affinity towards the fibres[16]. The time taken by the sample to reach 5 cm height above the water level in the reservoir was measured with scale.

(iii) *Whiteness Index*

CIE whiteness index of the treated fabrics was measured by Color eye-7000A computer colour matching spectrophotometer (Gretag Macbeth, USA). CIE whiteness index was measured according to ISO 105 - J02:1997 Textiles. Three readings were taken for each sample and the average values are reported.

2.3.5 Testing of Comfort Characteristics

(i) *Air Permeability*

TEXTEST air permeability tester (FX3300) was used to measure the air permeability at 98 Pa air pressure. ASTM D737 test standard was followed. An average of 3 readings for each sample was recorded.

(ii) *Moisture Vapour Transmission Rate*

Moisture vapor transmission (MVTR), also known as water vapor transmission (WVTR), is a measure of the passage of water vapor through a substance. MVTR cell, Modal CS-141 developed by W.R.Grace and Cryovac Division was used to carry out testing. Test standard ISO 9920 was followed and sample size was taken as 10 cm diameter circle.

The cell measures the humidity generated under controlled conditions as a function of time, this is based on the application of the gas permeability equation and the ideal gas law. Moisture vapor transmission was calculated using the equation as shown below:

$$T = (269 \times 10^{-7}) \times (\text{\% change in RH} \times 1440 / \text{Time interval}) (H) \quad \ldots (4)$$

where H is the humidity value; and T, the transmittance of moisture vapour.

3 Results and Discussion

In this study, sericin was applied on cotton knits to enhance its functional properties so as to make it suitable for medical garments. The process parameters for the application of sericin were optimized and the treated samples were characterized for various physical and comfort characteristics.

3.1 Application of Sericin on Cotton Knits without Complexing Agent

In this study, a 3^3 Box-Behnken experimental design was used to optimize the parameters for application of sericin on cotton knits. The experimental results were analyzed by RSM and the results of the experiments are shown in Table 3.

Table 3—Responses obtained on application of sericin on cotton knits

Case	Parameters			Responses		
	Sericin conc., gpL (X_1)	Time min (X_2)	Temp. ^{0}C (X_3)	Add-on % (Y_1)	*K/S* (Y_2)	Bending length cm (Y_3)
1	25	60	60	5.15	1.70	2.4
2	15	30	60	2.6	1.00	1.7
3	15	60	60	3.05	1.09	1.7
4	5	60	60	1.35	1.01	1.3
5	5	30	70	0.98	0.89	1.4
6	25	30	70	5.49	2.29	2.2
7	25	90	70	6.25	2.72	2.3
8	5	90	70	1.61	0.97	1.2
9	25	60	80	5.07	2.46	2.2
10	15	90	80	4.04	1.32	1.7
11	5	60	80	1.21	0.82	1.3
12	15	30	80	4.76	1.18	1.8
13	15	60	70	4.21	1.01	1.7
14	15	60	70	4.01	1.03	1.6
15	15	60	70	3.91	0.99	1.6

3.1.1 Effect of Parameters on Add-on %

The effect of sericin concentration (X_1), time (X_2) and temperature (X_3) on add-on % (Y_1) has been studied and the following response surface equation is obtained for the dependent variable:

$$\text{Add-on \% } (Y_1) = 4.04 + 2.10 X_1 + 0.14 X_2 + 0.37 X_3 + 0.03 X_1X_2 + 0.015 X_1X_3 - 0.29 X_2X_3 - 0.44 X_1^2 - 0.022 X_2^2 - 0.41 X_3^2 \quad \ldots (5)$$

P values of sericin concentration, time and temperature of treatment are found to be less than 0.05. From the analysis of measured responses obtained by the Design-Expert software, the quadratic model is found to be statistically significant for the variable add-on %. The regression coefficient R^2 value was estimated to be 0.95. The response surface plots representing the add-on (%) of sericin treated cotton fabric against the application parameters is shown in Fig. 1.

It is observed that % add-on increases significantly with the increase in 140sericin concentration, thus indicating more number of sericin molecules absorbed at higher concentration. However, not much increase in add-on is obtained on increasing the treatment time. In the second plot [Fig. 1 (b)], the add-on % is found to increase on increasing the temperature of treatment from 60^{0}C to 80^{0}C.

3.1.2 Effect of Parameters on Colour Value

The following response surface equation is obtained for the dependent variable Y_2 (*K/S*):

$$K/S\ (Y_2) = 1.59444 - 0.2725X_1 - 0.028413X_2 + 0.036738X_3 + 0.000298X_1X_2 + 0.002352X_1X_3 + 0.0000458X_2X_3 + 0.00528X_1^2 + 0.000198X_2^2 - 0.0004475X_3^2 \quad \ldots (6)$$

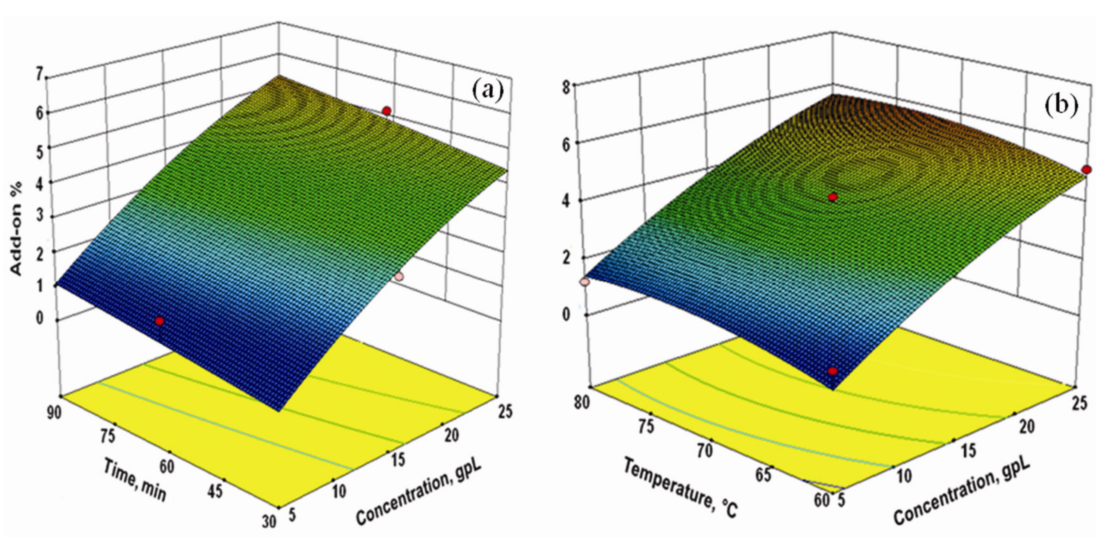

Fig. 1—Response surface plots for add-on % (Y_1) versus (a) time and sericin concentration at 60^{0}C, and (b) sericin concentration and temperature at 60 min

P values of sericin concentration, time and temperature of treatment are found to be less than 0.05. From the analysis of measured responses obtained by the Design-Expert software, the quadratic model is found to be statistically significant for the variable colour value. The regression coefficient R^2 value is estimated to be 0.98. The response surface plots representing the colour value of sericin treated cotton fabric against the application parameters is shown in Fig. 2.

K/S values are used as an indirect measure to assess the amount of sericin attached onto the fabric surface. It is observed that colour value of sericin treated fabrics increases significantly with the increase in sericin concentration. Thus, results are found correlated with the add-on %. However, both the plots show that only marginal increase in colour value is obtained on increasing the time and temperature of treatment. The results indicate that sericin concentration plays a major role.

3.1.3 Effect of Parameters on Bending Length

The following response surface equation is obtained for the dependent variable Y_3 (bending length):

$$\text{B.L.}(Y_3) = 2.73750 + 0.041250X_1 - 0.000972X_2 - 0.047083X_3 + 0.00025X_1X_2 - 0.0005X_1X_3 - 0.000083X_2X_3 + 0.000916X_1^2 + 0.0000185X_2^2 + 0.000416X_3^2 \quad \ldots (7)$$

From the analysis of measured responses obtained by the Design-Expert software, the quadratic model is found to be statistically significant for the variable bending length. The regression coefficient R^2 value is estimated to be 0.99 and P value is found to be less than 0.05. The response surface plots representing the bending length (cm) of sericin treated cotton fabric against the application parameters is shown in Fig. 3.

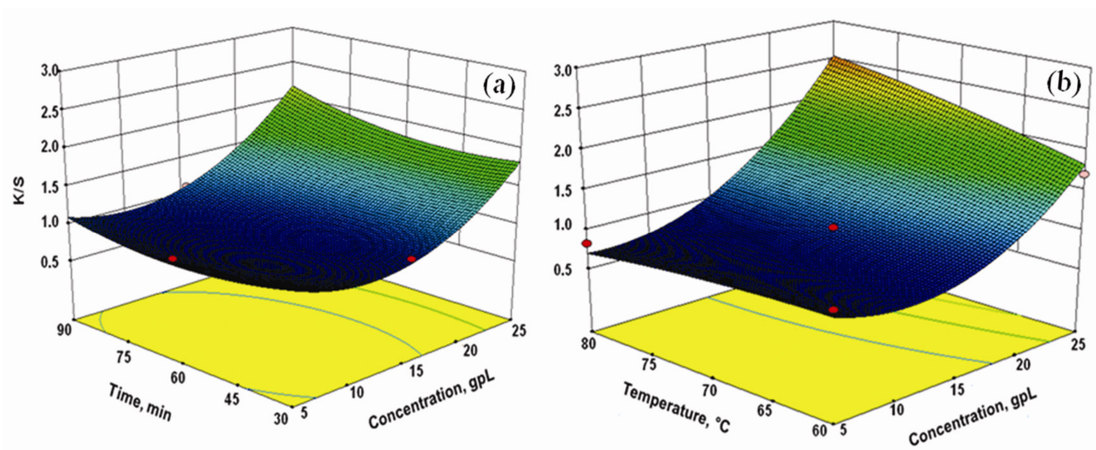

Fig. 2—Response surface plots for colour value (Y_2) versus (a) time and sericin concentration at 60^0C, and (b) sericin concentration and temperature at 60 min

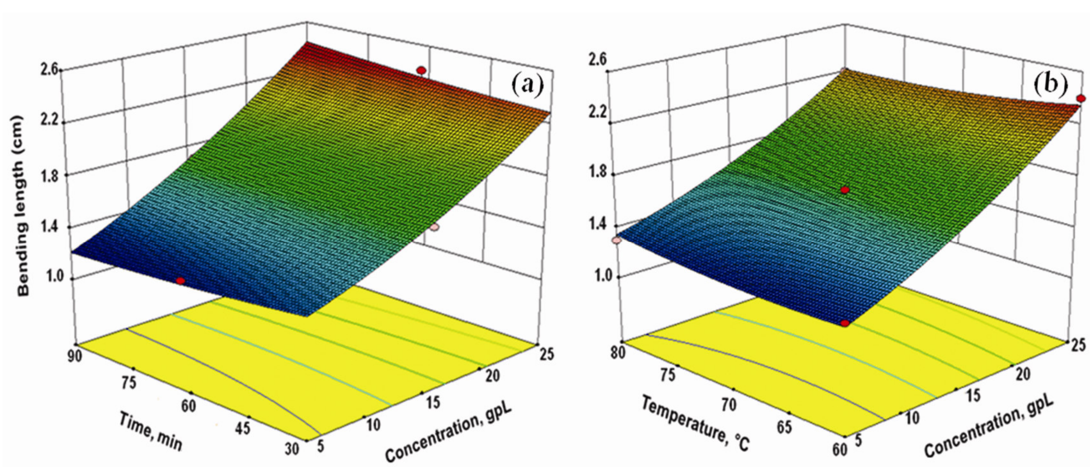

Fig. 3—Response surface plots for bending length (Y_3) versus (a) time and sericin concentration at 60^0C, and (b) sericin concentration and temperature at 60 min

Bending length is also taken as an indirect measure of the amount of sericin attached on the surface. It is observed that the bending length increases slightly as the concentration is increased from 5 gpL to 15 gpL and thereafter the increase is sharp. However, time and temperature of treatment do not play much role. Since stiffening of the fabric is an undesirable property, 15 gpL sericin concnetration is considered to be optimum.

On the basis of the measured responses, optimum conditions for application of sericin on cotton are found to be 15 gpL sericin concentration, 60 min treatment time and 60^0C temperature. These conditions are used for preparing the subsequent samples.

3.3 Application of Sericin on Cotton Knits with Complexing Agent

Once the optimum parameters for sericin application are identified, the next set of experiments has been carried out to optimize the concentration of alum. Fabrics are treated with 15 gpL sericin and then post treated with different concentrations of alum (2 - 8 % owf). The samples are then washed, dried, and conditioned. *K/S* values of sericin treated fabrics stained with acid dye are given in Fig. 4.

K/S of sericin treated cotton fabric increases by 36% as compared to untreated cotton. This can be attributed to the binding of acid dyes with the amide groups available on cotton due to the application of sericin. The colour value increases from almost 43% to 252% as the concentration of alum is increased from 2 % to 6 %. Since acid dye forms electrovalent bonds with sericin, the significant increase in the dye uptake might be due to the presence of sericin - alum complex which is formed due to the post-treatment with alum. The increase in dye uptake is only marginal when the alum concentration is further increased to 8 %. Therefore, 6 % (owf) alum concentration is considered to be optimum.

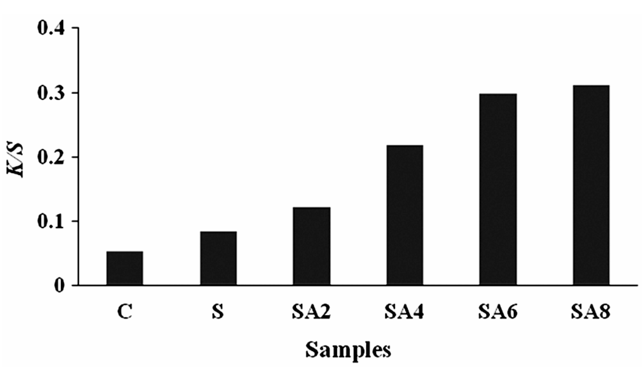

Fig. 4—Colour value of sericin treated cotton knits post-treated with different concentrations of alum [C – control, S – 15 gpL sericin, SA2 – 15gpL sericin + 2% owf alum, SA4 –15gpL sericin + 4% owf alum, SA6 – 15gpL sericin + 6% owf alum and SA8 – 15gpL sericin + 8% owf alum]

3.4 Durability of Finish

Samples treated with 15 gpL sericin and post-treated with 6% (owf) alum are tested for durability of treatment to washing. *K/S* value of samples was recorded and results are shown in Table 4.

It is observed that sericin treated cotton knit lost almost 22% colour on washing as compared to untreated cotton, which shows 63% reduction in colour. Interestingly, the sample post-treated with alum results in ~ 13% loss in colour after 5 washes. This may be because the ionizable groups on the side chains of various amino acid residues of sericin (such as aspartic and glutamic acids) bind with the metal ions, resulting in a complex formation thus making the finish insoluble (Fig. 5). Since colour value is an indirect measure of the amount of sericin present on the substrate, it can be inferred that the use of alum as a complexing agent increases the durability of the finish.

3.5 Characteristics of Sericin Treated Samples

Cotton knit samples have been prepared with the optimized parameters and characterized for various physical and comfort properties. The results are shown in Table 5. Fabric wicking is a function of the capillary movement of water through the fabric. Capillary action occurs when the adhesive intermolecular forces between the liquid and the substrate are stronger than the cohesive intermolecular forces within the liquid[19]. Table 5 shows that the wicking in fabric improves by almost 19 % after application of sericin. However, post-treatment with alum does not result in further increase in wicking.

Whiteness of the fabric is found to reduce slightly from 71 to 69 with the application of sericin. Adsorption of sericin by the cotton results in the yellowing of the fabric since sericin used in this study has a light yellow colour. Whiteness is found to

Table 4—Durability of sericin treated fabrics in terms of colour value

Sample	Original *K/S*	Retained *K/S*	% R
C	0.054	0.02	63
S	0.085	0.066	22.5
SA6	0.299	0.259	13.4

C – Control, S – Sericin treated, SA6 – Sericin + post treatment with 6% owf alum.

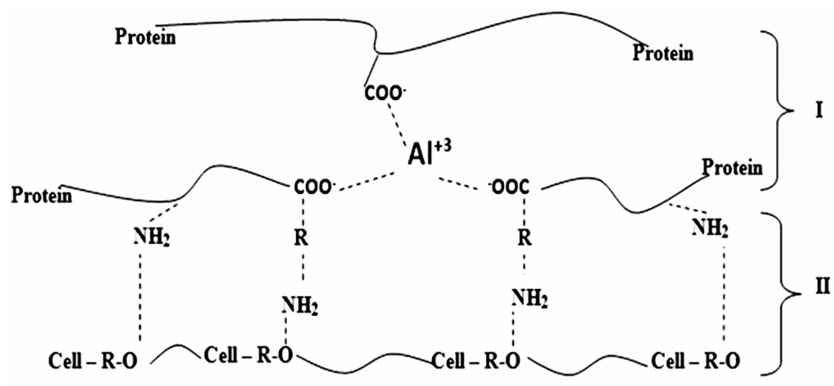

Fig. 5—(1) Binding formation between Al3+ ions and carboxylic acid in sericin molecular chain and (II) Sericin molecules adsorbed on the cotton fabric by H-bond and Vander Waal's force[17, 18]

Table 5—Characteristics of sericin finished cotton knits

Sample	Physical properties		Comfort properties	
	Wicking cm	Whiteness index-CIE	Air permeability $cm^3/cm^2/s$	Moisture vapour transmission, %
UT	19.4	71.3	53.8	58.6
S	24	69	72.6	61.2
SA6	25	64	75.1	71.3

UT —Untreated, S – Sericin treated, and SA6 – Fabric treated with 15gpL sericin and fixed with 6% alum.

reduce further on application of alum. Where whiteness is a consideration, sericin can be bleached before use to take care of this matter.

Textile fabrics have an inherent rough structure. Therefore, it is assumed that a smoother fabric surface having good water transport properties and high breathability which can have therapeutic effect on patients suffering from skin ailments. To study the effect of sericin coating on the comfort properties, treated fabric was tested for air permeability and water vapour permeability. Air permeability of sericin treated cotton knit is found to be 26 % higher than that of untreated cotton. This can be due to the coating of sericin which glued the yarns together, thus making the yarns compacter. As the yarn compactness increased, the pore size between the yarns increased, thus increasing the air permeability. However, not much increase in air permeability is observed when sericin is fixed with alum.

Sericin treated fabrics show almost 18 % increase in moisture vapour transmission rate as compared to control fabric. Sorption – desorption is an important phenomena of moisture vapour transmission which is responsible for maintaining the microclimate during the transient condition. The hygroscopic fibre material absorbs moisture from the humid air close to it and release this absorbed moisture in dry air. The transmission of moisture vapour in case of hygroscopic material is higher. Since, sericin is composed of 80 % amino acids that contain hydrophilic groups such as serine, aspartate and glycine, it can absorb moisture very well and transmit this moisture to the surrounding atmosphere. In this way, it may be concluded that the moisture vapour transmission of the sericin finished fabric is higher than the unfinished fabric.

4 Conclusion

Results of this study show that durable sericin based finish can be applied on cotton knits using alum as complexing agent. Sericin concentration of 15gpL with 6 % owf of alum is optimized for application of sericin. Air permeability and moisture absorption properties are found to improve considerably after treatment with sericin. It is concluded that sericin can be used to enhance the properties of cotton, thus making it suitable for medical garments.

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