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STUDIES ON DEGUMMING OF INDIAN BIVOLTINE SILK

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The influence of process conditions on the level and quality of silk degumming has been studied extensively using two factors by the researchers. In this work, the effect of four process conditions viz. time, temperature, soap concentration and soda concentration in degumming process, being adopted in the silk handloom industry, has been studied on Indian bivoltine silk with respect to degumming loss %, whiteness index and yellowness index using Box-Behnken design of experiments. It is found that the temperature and soda concentration significantly influence the three responses studied. Soap concentration is found to influence whiteness index and yellowness index. The optimum process conditions have been set to obtain the desirable degumming loss percentage. (* To whom correspondence should be addressed. Tel: +91 44 27277583, 27230499. Fax: +91 44 27277583 , E Mail: kmakhadir@Yahoo.com)

Key words: Indian bivoltine silk, Degumming, Whiteness Index and Yellowness Index.

INTRODUCTION

Degumming is the process of removing the sericin, a natural gum, in order to dye the silk evenly. The silk fiber is a continuous double filament consists of fibroin surrounded by sericin [1]. The fibroin has highly oriented crystalline domain and it is insoluble. The sericin is amorphous and it is soluble in hot soap and soda solution [2]. The fibroin and sericin contain eighteen kinds of amino acids, which are proteins. These amino acids form into a polypeptide by the aid of a covalent bond. The polypeptides formed together have a higher molecular structure and molecular weight. These high molecules differ from each other depending on their molecular structures even if they are of the same chemical composition [3].

The reason, for sericin to be more soluble in water than fibroin, is because it is not only composed of strong hydrophilic amino acids such as serine, aspartic acid and threonine, but also because most of the molecules are globular with irregular and diffusely formed structure. Whereas the fibroin molecule has a close-knit structure due to a high degree of crystallization [1]. In silk, compound molecules are classified into several groups depending on the composition of atoms with similar characteristics. Thus, sericin has been divided into sericin I, II and III based on the degree of sericin solubility [2].

Sericin is a large complex molecular electrolyte with a base amino radical (NH_2) and an acid radical (CooH) in each molecule. The electric charge in sericin changes, depending on the pH value of the solution. In acidic medium (supply of H^+), the sericin molecule is positively charged and in alkaline medium (supply of OH^-), the sericin molecule is negatively charged. At the iso-electric point of sericin pH 3.8 – 4.2, the electric charge in the sericin molecule is balanced and the effective electric charge become zero, resulting in minimal swelling and solubility of sericin. If the pH is raised, the sericin solubility increases proportionately. Degumming is based on water solubility and higher alkali sensitivity of the sericin as compared with the fibroin at pH value from 9 to 11.5. Strong alkalis as well as long treatment time may however also attack the fibroin [3]. The influence of process parameters on the quality of silk degumming has been studied using two factors in the past [4 & 5].

Handloom industry consumes bulk quantity of Indian silk. The silk is degummed using neutral soap and soda ash (Na_2Co_3) at high temperature for short period. The soap and soda concentration (grams per liter), duration of degumming and its temperature vary considerably from dye house to dye house. In this paper an attempt has been made to standardize the process parameters of degumming for Indian bivoltine silk.

MATERIALS AND METHODS

Material: Commercially available Indian bivoltine mulberry twisted silk [tram] yarn of 19/21 denier two ply with 6-8 twist per inch having uniform characteristic was considered for the study**.**

Experimental Design: The experiments were conducted using Box-Behnken second order composite design for four variables [6]. The variables were selected based on trials. The experimental design with the coded and actual values of the four process variables, soap (neutral) concentration, soda ash (Na_2Co_3) concentration, Time and Temperature of degumming are given below:

Coded Value	***-1***	***0***	***+1***
Soap gpl	1	3	5
Soda gpl	0.5	1.5	2.5
Temperature (°C)	50	75	100
Time (m)	15	30	45

To relate the variables with the response, a multiple regression equation in fitted, representing a quadratic polynomial function.

$$Y = a_0 + a_1 X_1 + a_2 X_2 + a_3 X_3 + a_4 X_4 + a_{11} X_1^2 + a_{22} X_2^2 + a_{33} X_3^2 + a_{44} X_4^2 + a_{12} X_1 X_2 + a_{13} X_1 X_3 + a_{14} X_1 X_4 + a_{23} X_2 X_3 + a_{24} X_2 X_4 + a_{34} X_3 X_4$$

Where X_1 X_2 X_3 and X_4 are process variables, Y is the response from each experiment and $\mathbf{a_i}$ are regression coefficients.

The estimated regression equation was evaluated in terms of Fisher's test (F) values, multiple correlation co-efficient (R^2), summary of model fit and lack of fit test. If the R^2 valve lies between 0.75 to 0.99, the fitted regression equation is considered to be a good fit of the models.

The twisted silk tram weighing approximately 30 g each was considered for the 27 experiments carried out as per Box-Behnken model. The pre wet twisted silk was degummed according to the factors considered in the experiment and experiments were carried out randomly as per the run order of the model. After the experiment, the silk samples were washed in cold water, dried, conditioned and then, the properties viz. degumming loss %, whiteness Index (Hunter) and yellowness index (ASTM) were tested [7 & 8].

RESULTS AND DISCUSSION

Degumming loss %: The analysis of variance, for degumming loss % (Table 1) suggests significant regression, that is, atleast one of the factors in the regression equation makes a significant impact on the mean response. Further linear order of the regression model is significant but the square (quadratic) and interaction order of the regression model are not significant. The lack of fit, the variation due to model inadequacy is not significant, which implies that there is no evidence that the model does not adequately explain the variation in the response. High R^2 value suggests that 93% of the variation in the observed response is explained by the regression model. The regression coefficient, in coded units and its statistical inference along with regression coefficient, in un-coded units are given in Table 2. It is evident from the Table 2 that the p value of two linear effects (Temperature and Soda concentration) are less than 0.05 (α value) which suggest that there is a significant linear effect of temperature and / or soda concentration on the degumming loss percentage.

Contour plots are useful to establish desirable response values and operating conditions. Figure 1 shows how the degumming loss % relates to temperature and soda concentration at constant level of time with low, medium and high soap concentration based on the model equation.

Table 1 Analysis of Variance for Degumming loss, Whiteness index and Yellowness index

Source	Degree of freedom	Degumming loss		Whiteness Index		Yellowness index	
		F – Value	P - Value	F – Value	P - Value	F – Value	P - Value
Regression	14	11.46	0.000	15.63	0.000	8.21	0.000
Linear	4	37.46	0.000	48.02	0.000	26.87	0.000
Square	4	1.18	0.367	5.58	0.009	0.88	0.502
Interaction	6	0.98	0.480	0.74	0.626	0.65	0.692
Residual Error	12						
Lack-of-Fit	10	16.01	0.060	13.42	0.071	1.32	0.505
Pure Error	2			15.63	0.000		
Total	26						

Table 2 Regression coefficients for degumming loss, Whiteness index and Yellowness index

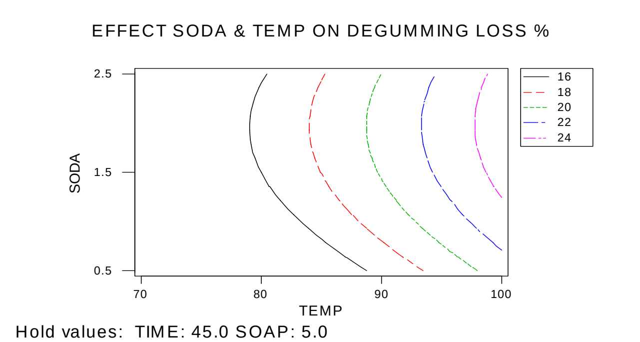
Term	Coefficient of Degumming loss		Coefficient of Whiteness Index		Coefficient of Yellowness index	
	Coded	Un coded	Coded	Un coded	Coded	Un coded
Constant	12.857**	-38.614	63.303**	81.340	8.038**	17.242
Temperature	11.062**	0.287	9.731**	-0.921	-3.652**	0.033
Time	1.62	0.266	0.938	0.041	-0.305	-0.108
Soap	1.634	6.205	3.552**	-0.348	-1.752**	-1.005
Soda	2.599**	8.052	2.107**	-4.506	-1.05**	0.742
Temp*Temp	1.063	0.002	5.227**	0.008	-0.648	-0.001
Time*Time	-1.149	-0.005	0.99	0.004	0.374	0.002
Soap*Soap	-1.583	-0.396	1.189	0.297	-0.705	-0.176
Soda*Soda	-1.868	-1.868	0.458	0.458	-0.13	-0.130
Temp*Time	-0.667	-0.002	-1.391	-0.004	0.089	0.000
Temp*Soap	-0.937	-0.019	0.985	0.020	0.297	0.006
Temp*Soda	0.17	0.007	1.791	0.072	-0.812	-0.032
Time*Soap	0.697	0.023	-0.354	-0.012	-0.039	-0.001
Time*Soda	2.125	0.142	0.717	0.048	-0.263	-0.018
Soap*Soda	-3.073	-1.536	-1.044	-0.522	1.04	0.520

**** - p value is less than 0.05 (α value)**

Table 3 Comparison of actual results of degumming experiment with calculated values

Particulars	Parameters		
	Degumming loss (%)	Whiteness Index (Hunter)	Yellowness Index (ASTM E313)
Regression Value (calculated)	24.01	78.12	4.21
Mean	23.97	76.15	5.938
Standard Deviation	0.275	0.969	0.785
Standard Error	0.159	0.560	0.453
CI Lower	23.285	73.738	3.988
Upper	24.650	78.554	7.887
t- Value	-0.27	-3.53	3.81
p- Value	0.814	0.072	0.062

It is evident from the Figure 1 that the desirable degumming loss % (24% maximum level of sericin content in bivoltine silk) can be achieved at boil (100°C) with soda concentration of 1.25 gpl, 1.35 gpl and 1.8 gpl at soap concentration of 5 gpl, 3 gpl and 1 gpl respectively, when the degumming is carried out for 45 minutes. Higher concentration of soap results in swelling and emulsifying of the sericin, which easily hydrolyzed at polypeptide chain ends with little soda concentration.



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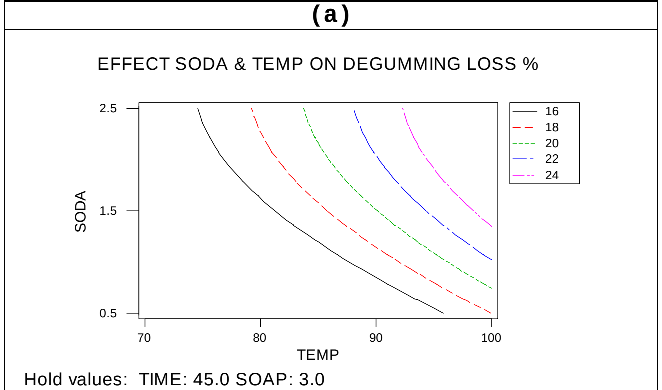

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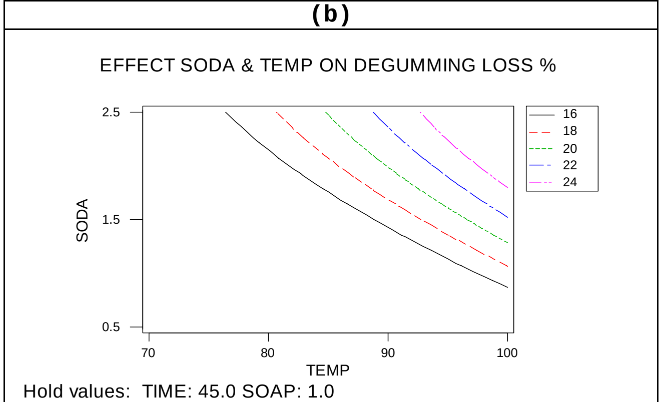

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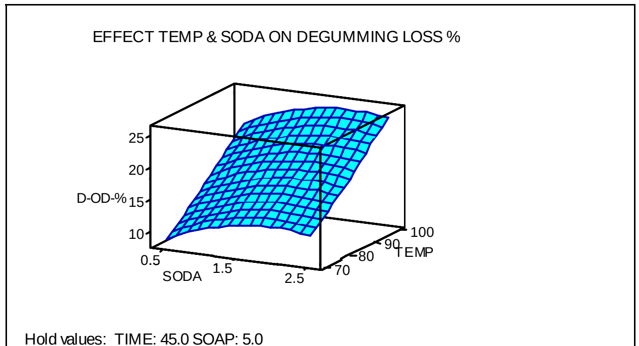
Figure 1 Contour plots – Effect of temperature and soda concentration on the degumming loss %

The surface plot, a three-dimensional wire-frame, represents the functional relationship between the response and experimental factors. Figure 2 shows how the degumming loss % relates to temperature and soda concentration at constant level of time with low, medium and high soap concentration. It is evident from the figure 2 that the degumming loss % increases rapidly if you increase the temperature while holding the soda concentration constant. The slope of the linear effect decreases, when you decrease the soap concentration.



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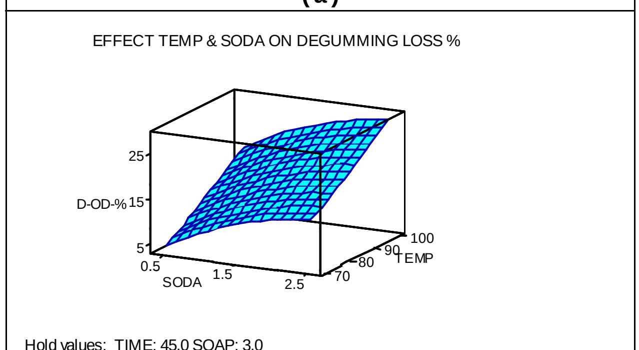

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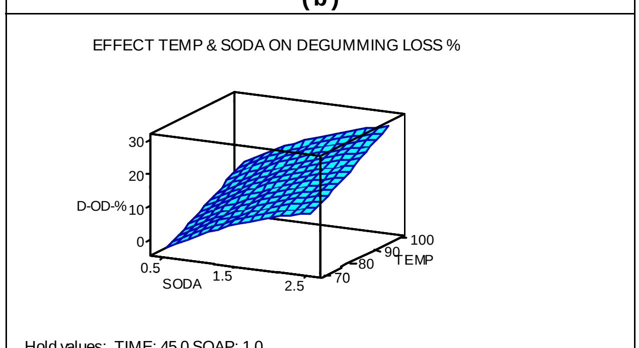

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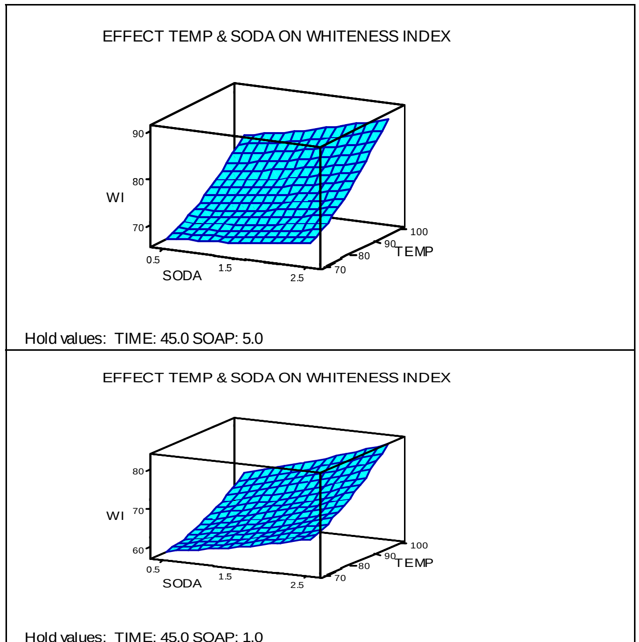
Figure 2 Surface plot- Effect of temperature and soda concentration on the degumming loss %

The degumming loss % increases gradually if you increase soda concentration up to a certain level and then decreases, while holding the temperature constant. The threshold point changes according to the temperature level. The slope of the linear effect, rate of change in degumming loss, increases when you decrease the soap concentration.

It is always desirable to have controlled degumming process, as the fibroin, the core of silk, is also susceptible to degradation due to hydrolysis of peptide linkage. The rate of degumming is rapid at high soap concentration with respect to temperature. Hence, 3 gpl soap concentration shall have a moderate rate of degumming with respect to temperature. The rate of degumming (Figure 2 b) is slow at high soda concentration with respect to temperature and the rate of degumming increases while soda concentration decreases. Hence, around 1.5 gpl soda concentration, precisely 1.35 gpl, shall have moderate rate of degumming with respect to temperature. Thus, the degumming process can be carried out at 100°C for 45 minutes with the degumming recipe of 3 gpl soap and 1.35 gpl soda concentration to get the desirable degumming loss of 24% in Indian bivoltine silk.

Whiteness Index: The ANOVA for whiteness index (Table 1) suggests significant regression. Further linear and squared (quadratic) orders of the regression model are significant but the interaction is not significant. The regression coefficient, in coded units, and its statistical inferences, along with regression coefficient, in un-coded units, is given in Table 2. It is evident from the table 2 that the p values of three linear effects (Temperature, Soap concentration and Soda concentration) are less than 0.05 (α value), which suggest that there is significant linear effect of temperature and / or soap concentration and /or soda concentration on the whiteness index. The p value of only one squared effect (Temperature X Temperature) is less than 0.05 (α value), which suggests that there is significant quadratic effect of temperature on the whiteness index. The surface plot (Figure 3 a) shows how the whiteness index relates to temperature and soda concentration at constant level of time with low and high soap concentration. The Figure 3 b shows the effect of Temperature and Soap concentration and Figure 3 c shows the effect of Soap concentration and Soda concentration. It is evident from the Figure 3 a that the whiteness index increases rapidly if you increase the temperature while holding the soda concentration constant. The slope of the linear effect, rate of change in Whiteness Index, decreases if you decrease the soap concentration.



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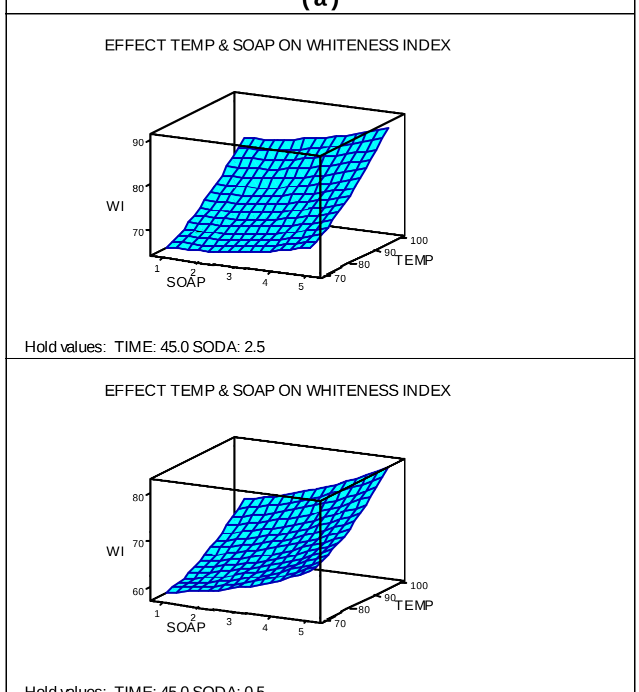

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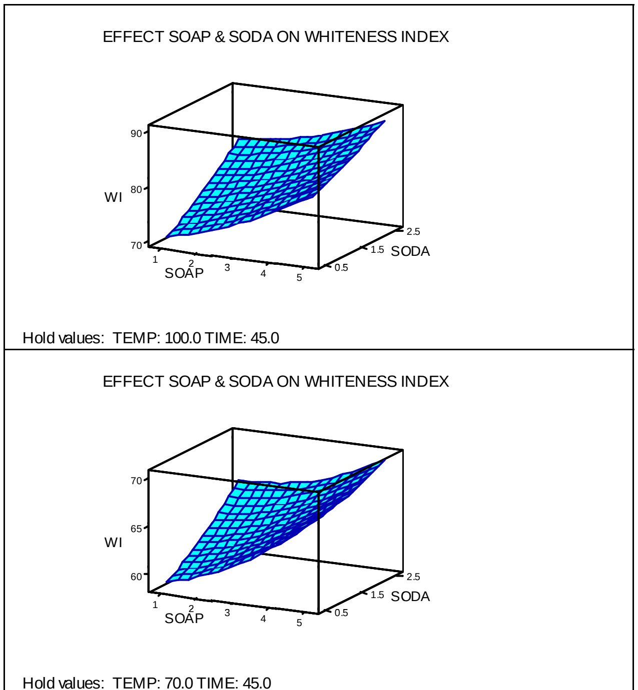

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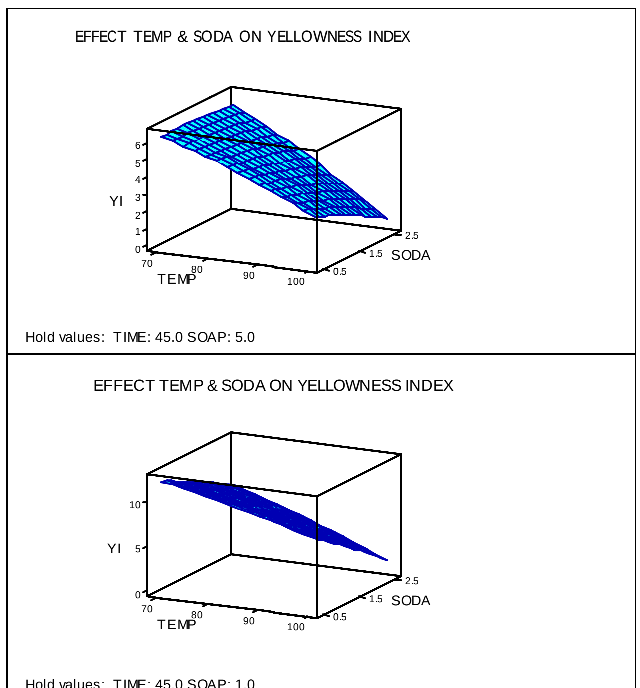
Figure 3 Surface plot - Effect of temperature, soap and soda concentration on Whiteness index

The whiteness index increases gradually if you increase the soda concentration, while holding the temperature constant. The slope of the linear effect, rate of change in whiteness index, increases when you decrease the soap concentration. Similar trend is observed (Figure 3 b) in the case of temperature and soap. As shown in Figure 3 c, the whiteness index increases gradually, if you increase the soda concentration while holding the soap concentration constant. The slope, the rate of change, decreases at high soap concentration and the said effect is severe at lower temperatures.

Yellowness Index: The ANOVA for yellow index (Table 1) suggests significant regression. Further, the linear orders of the regression model, is significant. The regression coefficient, in coded units, and its statistical inferences along with the regression equation, in uncoded units, is given in Table 2. It is evident from the table 2 that the p values three linear effects (Temperature, Soap and Soda concentration) are less than 0.05 (α value), which suggests that there is significant linear effect of temperature and / or soap concentration and / or soda concentration on the yellowness index.

The surface plot (Figure 4 a) shows how the yellowness index relates to temperature and soda concentration at constant level of time with low and high soap concentration. The Figure 4 b shows the effect of Temperature and Soap concentration and Figure 4 c shows the effect of Soap concentration and Soda concentration. It is evident from the Figure 4 a that the yellowness index decreases rapidly if you increase the temperature while holding the soda concentration constant. The slope of the linear effect, rate of change in yellowness Index, decreases if you decrease the soap concentration. The yellowness index decreases gradually if you increase the soda concentration, while holding the temperature constant. The slope of the linear effect, rate of change in yellowness index, increases when you decrease the soap concentration. Similar trend is observed (Figure 4 b) in the case of temperature and soap. As shown in Figure 4 c, the yellowness index decreases, if you increase the soda concentration while holding the soap concentration constant. The slope, the rate of change, decreases at high soap concentration and the said effect is severe at lower temperatures.



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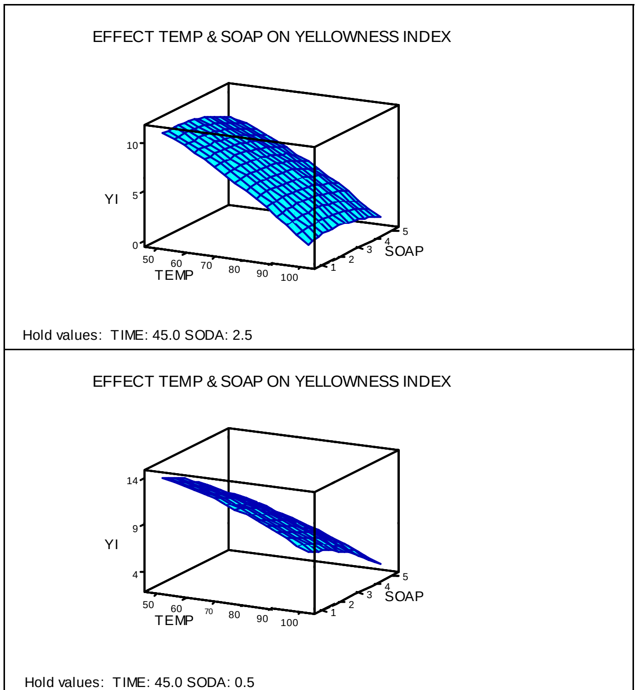

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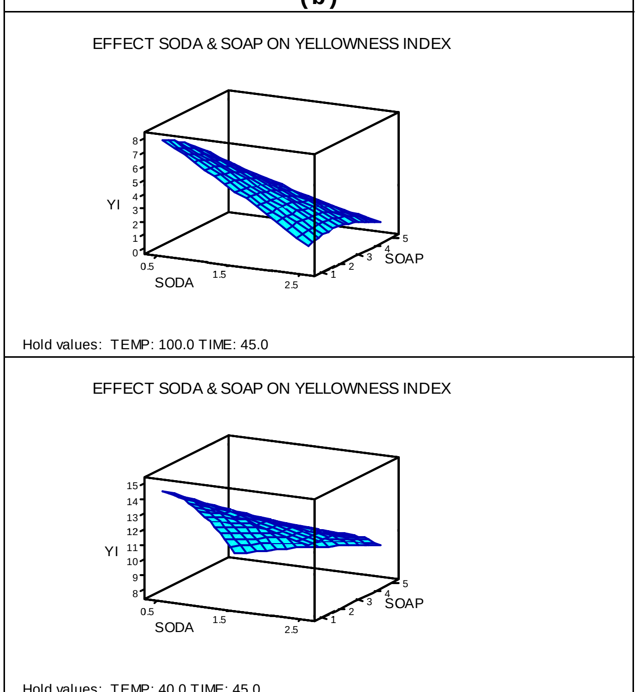

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Figure 4 Surface plot - Effect of temperature, soap and soda concentration on Yellowness index

Three degumming trials, using the standard condition set for Indian bivoltine raw silk (100 °C temperature, 45 minutes time duration, 1.35 gpl soda concentration and 3 gpl soap concentration), were taken to compare the actual and the calculated values of degumming loss %, whiteness index and yellowness index. It is evident from the Table 3 that there is no significant difference in the tested parameters. The average of the three trial experiment values, tenacity and elongation of Indian bivoltine degummed silk, are found to be 4 grams per denier and 16 % respectively.

CONCLUSION

The Indian bivoltine silk can be degummed to the desired extent of 24 % weight loss, when the process of degumming is carried out at 100 °C for 45 minutes with 3 gpl soap and 1.35 gpl soda concentration at a liquor ratio of 1:30. The associated whiteness index and yellowness index are 78 and 4 respectively.

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