Factors to Predict the Fibrillation Tendency of Lyocell Fibers

W. Udomkichdecha[1] and S. Chiarakorn[1]

Lyocell fibers were treated with a cross-linking agent at various levels of concentration to examine the fibrillation tendency. An optimum concentration was achieved to reduce the fibrillation to a minimum level. The influence of physical parameters on the fibrillation index was also studied. These include birefringence, intrinsic viscosity, and relative crystallinity. The effects of each parameter on the fibrillation index are discussed and a simple mathematical model is proposed to represent the correlations among these parameters.

Key words: Lyocell fibers, fibrillation, fibrillation index.

[1] *Department of Materials Science, Faculty of Science, Chulalongkorn University, Bangkok 10330, Thailand.*

ปัจจัยที่มีผลต่อการประเมินแนวโน้มการเกิดขนของเส้นใยไลโอเซล

วีระศักดิ์ อุดมกิจเดชา และ สิริลักษณ์ เจียรากร (2544)
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เส้นใยไลโอเซลที่ทำการทดลองในครั้งนี้ นำไปผ่านสารเชื่อมขวางที่มีความเข้มข้นต่าง ๆ กัน เพื่อศึกษาแนวโน้มของการเกิดขนบนเส้นใย และพบว่าความเข้มข้นของสารเชื่อมขวางดังกล่าวมีระดับที่เหมาะสมจุดหนึ่งที่ทำให้การเกิดขนเกิดได้น้อยที่สุด จากนั้นได้ศึกษาถึงอิทธิพลของปัจจัยทางกายภาพที่มีต่อค่าของการเกิดขน ซึ่งได้แก่ปัจจัยที่เกี่ยวกับค่าดัชนีหักเหภายในโครงสร้างเส้นใย ค่าความข้นหนืด และค่าผลึกสัมพัทธ์ อิทธิพลของปัจจัยแต่ละตัวดังกล่าวได้นำไปวิเคราะห์และนำไปสู่การเสนอความสัมพันธ์ทางคณิตศาสตร์ อย่างง่ายระหว่างปัจจัยเหล่านั้นต่อการเกิดขน

คำสำคัญ เส้นใยไลโอเซล การเกิดขนบนเส้นใย ดัชนีการเกิดขนบนเส้นใย

INTRODUCTION

Lyocell is a new generic name given to a cellulosic fiber which is produced under an environmentaly friendly process by dissolving cellulose in the tertiary amine oxide N-methylmorpholine-N-oxid (NMMO). The fiber is spun into an aqueous spinning bath containing diluted aqueous NMMO. The process is a closed system; solvent and access water evaporation is almost completely recirculated.[1]

Lyocell fiber shows some key advantageous characteristics over other cellulosic fibers; for instance, a high dry and wet tenacity and high wet modulus. However, the fiber also shows an extensive tendency to fibrillate in the wet state.[2] It has been concluded from previous works[3-5] that this is due to the spinning process, which causes the formation of longer and more oriented crystalline regions and smaller but more oriented amorphous regions in the fiber structure. This fibrillar structure is responsible for the high fiber tenacity but low lateral cohesion, especially when subjected to mechanical stress in the swelled state. Even though the fibrillation characteristic has been shown to be advantageous in applications when a peach-skin lappearance of the surface is required, but it has been proven to be disadvantageous for some other applications, such as the launderability of the product and difficulty to control the uniformity of colour uptake during dyeing.

Many possible solutions have been proposed to modify the fibrillation structure and hence the fibrillability of the lyocell fiber. These run from controlling spinning parameters[3] (spinneret size and temperature, draw ratio, etc.) to air gap conditions and aftertreatment of the fiber. Commercially, it was found practical to treat the finished fiber with cross-linking agents or some special polyfunctional reactive dyes to control the degree of fibrillability. Fibrillation measurements are normally presented in terms of fibrillation index, FI, which is the sum of the fibril lengths ($\sum l$) divided by the fiber length (L):

$$FI = \Sigma I/L. \qquad \ldots.(1)$$

The FI value is only a simple measurement assessed through optical micrographs under the restriction of a two-dimensional photograph. In fact, fibrillation of a lyocell fiber occurs all around the fiber skin and thus a more accurate measurement should be carried out to take account of the third dimension. However, there are still some complications in proposing any alternative model for calculating the fibrillation index in a three-dimensional photograph. Therefore, this paper concentrates on predicting the fibrillation ability by some other physical parameters of the fiber. These parameters include birefringence, intrinsic viscosity and relative crystallinity. Finally, a simple mathematical model presenting the relationship between the fibrillation index and those parameters is proposed.

EXPERIMENTAL

The lyocell fibers chosen for this experiment were Tencel staple fibers from a local authorized agent, J.P. Bosco Co., Ltd. Different levels of fibrillation were controlled by treating the fiber with a cross-linking agent, GP®, supplied by the same company. This cross-linking agent is a polyfunctional, colorless reactive dye containing at least two amine groups. The concentrations of GP® stock solution were

[Tencel® is the registered trademark of Courtaulds Fibers for lyocell.]

varied at 2%, 4%, 6% and 8% by volume as recommended by the supplier. The finishing solutions were prepared in a flask, and 12% g/l of sodium sulfate and 12% g/l of sodium carbonate were added into 14 ml of distilled water to improve the fiber swelling properties. The sample of 2 grams of fibers was put in the flask, followed by the procedure shown in the flow chart in Figure 1.

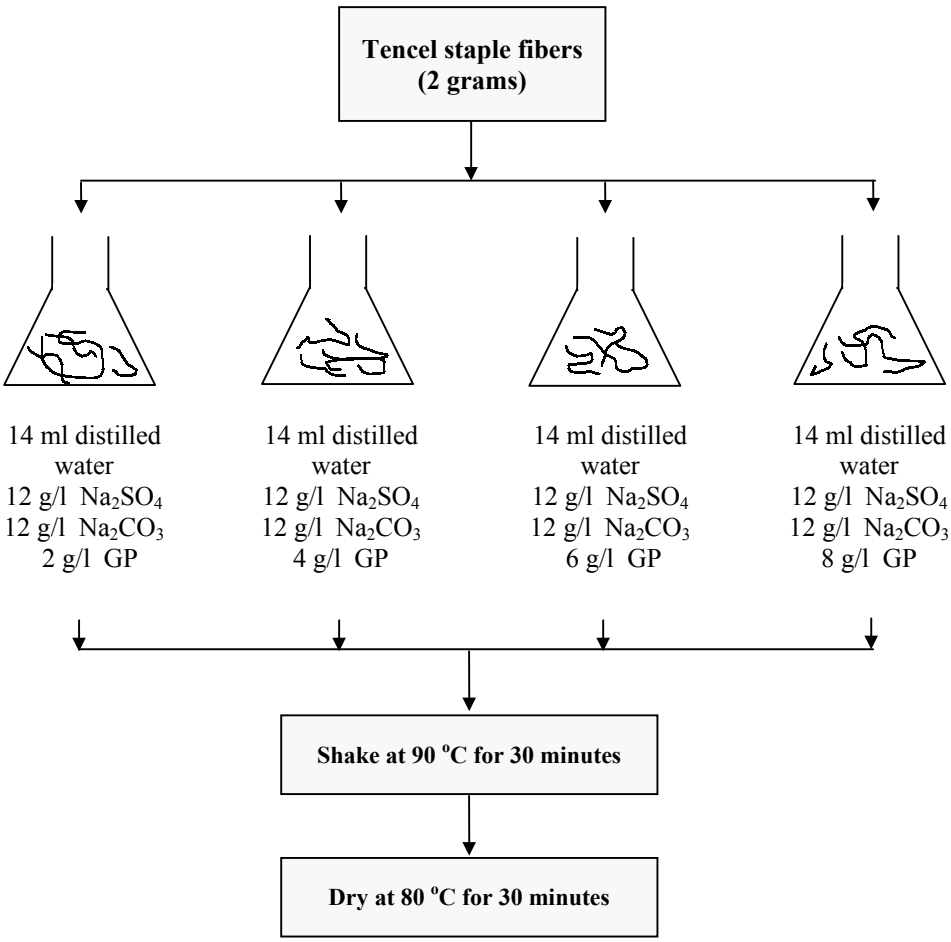
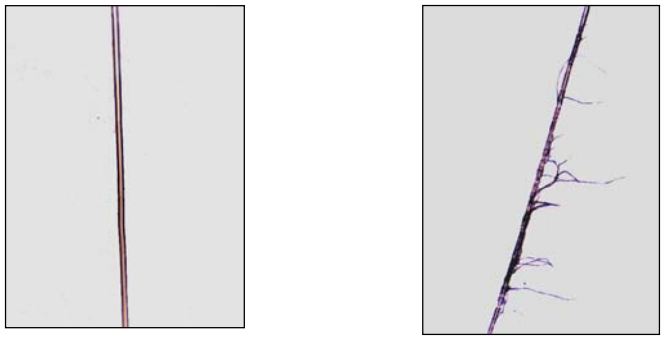


Figure 1. Flow chart of defibrillation finishing.

After the finishing process, the dried samples were put into another flask containing 50 ml of 12 g/l sodium carbonate and shaken at 120 revolutions per minute at ambient temperature for 30 minutes. Then they were rinsed and dried at 80°C for another 30 minutes. The fibers with induced fibrillation were then divided into 4 portions for evaluation of the fibrillation index, birefringence, intrinsic viscosity and relative crystallinity. The fibrillation index was calculated from the optical micrographs taken from the treated fibers. Some of these, for different values of fibrillation indices, are shown in Figure 2.



FI = 0 **FI = 0.45**

Figure 2. Photomicrographs of sampled fibers from optical microscopy (x100) for FI = O and FI = 0.45.

Birefringence examinations[6] were by microscopy techniques with the aid of an adjustable retardation compensator. The standard test method ASTM D1795-96 was performed to determine the intrinsic viscosity of the fibers.

The relative crystallinity of the fibers was obtained by measuring the peak heights of X-ray diffractograms at diffraction angles 2θ of 19.8 and 22.0. These show the intensities of amorphous and crystalline peaks, respectively. The relative crystallinity, %C, was then calculated by equation (2):

$$\%C = \frac{I_C}{(I_C + I_A)} \times 100, \quad \ldots(2)$$

where I_C is the intensity of the crystalline peak while I_A is the intensity of the amorphous peak.

Results from the experiments were analyzed by standard statistical analysis. Linear regression analysis and analysis of variance of multiple regression by the forward method were the two major statistical tools used in this study.

RESULTS AND DISCUSSION

In order to vary the fibrillation level of the Tencel fibers, we performed fiber finishing with a cross-linking agent, GP®. Table 1 shows the results of applying different cross-linking agent concentrations to the tested fibers. The non-finished fiber (0% GP®) shows the highest tendency to fibrillate after subjection to the wet abrasion process. These values (fibrillation indices) decrease with increasing concentration of GP® as shown in Figure 3. This is due to the effect of the cross-linking reaction between the cross-linking agent and the cellulosic fibers. However, too much concentration of the cross-linking agent may lead to the precipitation of GP® from the finishing solution before reacting with cellulose chains. Thus, the efficiency of GP® to defibrillate decreases and hence the fibrillation index increases after passing the optimum concentration which is 6% GP®. This result confirms the recommendation of the supplier that the most effective concentration of GP® to control the Tencel fibrillation is 6% g/L of GP®.

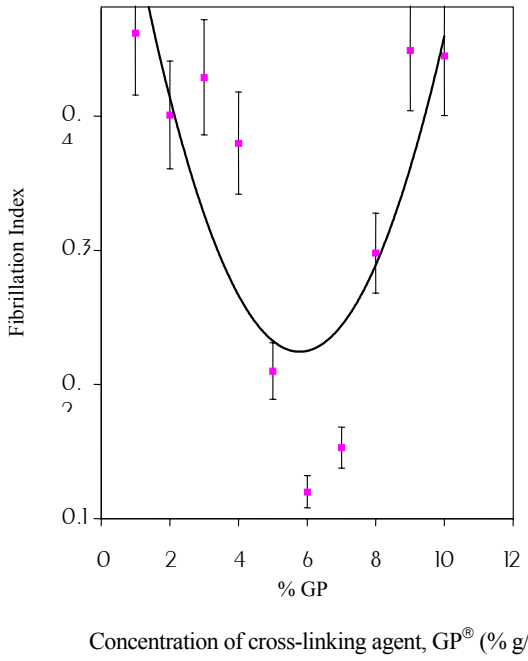


Figure 3. The effect of the cross-linking agent, GP®, on the fibrillation index of Tencel fibers.

In this study, fibrillation of the tested lyocell fibers was controlled by post-treatment with the cross-linking agent, GP®. At each application level of GP®, the resulting fibers were examined to obtain their fibrillation index, birefringence, intrinsic viscosity and relative crystallinity. All the data are presented together in Table 1.

Table 1. Fibrillation index, birefringence, intrinsic viscosity and % relative crystallinity* of Tencel® fiber treated with 0%, 2%, 4%, 6% and 8% GP®.

% GP	FI	BF	η	C (%)
0	0.45	0.0430	3.016	66.43
2	0.40	0.0432	3.284	53.20
4	0.38	0.0421	3.779	46.56
6	0.12	0.0410	7.953	23.74
8	0.30	0.0426	3.300	42.84

* FI: Fibrillation Index BF: Birefringence
[η]: Intrinsic viscosity C: Relative crystallinity

As mentioned before, the fibrillation can be reduced to an optimum level at a 6% concentration of GP®. It is also clearly seen the changes of all other parameters correspond to the variation of the fibrillation index. The interesting question arises as to whether there is any significant correlations between fibrillation index and the other parameters. If so, can we propose an empirical model to predict the fibrillation tendency from any of those parameters in order to avoid measuring the fibrillation index.

Increasing the amount of GP® induced more cross-linking between the cellulose chains in the amorphous region and also increased the alignment of lateral bonds in the direction across the fiber axis. This result was confirmed by the peak height of the amorphous region, which increased correspondingly. Therefore, the value of relative crystallinity, which was calculated in terms of I_C over the sum of I_C and I_A, decreased from 66.43% down to 23.74%. Meanwhile, the lateral bonding orientation also affected the velocity of light through that direction. This, finally, brought down the fiber birefingence (the difference between the refractive index along and across the fiber axis) in a similar trend.

Table 1 also suggests some possible relationships between the physical properties and the fibrillation index of treated fibers. When the fibrillation index decreases, both birefringence and relative crystallinity decrease. These results agree very well with the explanation that fibrillation of the fiber is due to the formation of more oriented crystalline regions. On the other hand, increasing the amount of GP® application leads to a decreasing fibrillation index but increasing intrinsic viscosity, which is due to the high cross-linking along the side chains of polymer molecules in the amorphous region.

Table 2. Pearson's correlation coefficients of all parameters.

		FI	BF	C	η
Fl	Pearson Correlation	1.000	.744*	.861**	-.853*
	Sig. (2-tailed)	.	.047	.009	.027
	N	10	10	10	10
BF	Pearson Correlation	.774*	1.000	.883*	-.823*
	Sig. (2-tailed)	.047	.	.047	.025
	N	10	10	10	10
C	Pearson Correlation	.861**	.883*	1.000	-.862
	Sig. (2-tailed)	.009	.047	.	.060
	N	10	10	10	10
η	Pearson Correlation	-.853*	-.823*	.862	1.000
	Sig. (2-tailed)	.027	.025	.060	.
	N	10	10	10	10

* Correlation is significant at the 0.05 level (2-tailed).
** Correlation is significant at the 0.01 level (2-tailed).

In order to examine the possibility of significant correlations among these properties, Pearson's correlation coefficients of all pairs of parameters were evaluated based upon 2-tailed hypotheses. The results are summarized in Table 2, showing that significant correlations occur among these parameters. More precisely, it can be observed that the fibrillation index is affected positively by the relative crystallinity and birefringence while affected negatively by the viscosity. The correlation coefficients appeared to be 0.861, 0.774 and 0.853, respectively. The result was then verified again utilizing the analysis of variance of multiple regression by the forward method. Setting a 90% confidence level, it was found that only the intrinsic viscosity and relative crystallinity affected the fibrillation index significantly. The proposed relationship is then giron by equation 3.

$$FI = 1.32-(1.82 \times 10^{-2})\,\eta + (5.894 \times 10^{-3})C \qquad \ldots.(3)$$

CONCLUSIONS

Fibrillation of lyocell fibers can be reduced effectively by post-treatment of the fibers with a commercial cross-linking agent. The optimum concentration of this cross-linking agent was found to be important. A short range near 6% concentration is the most effective to reduce the fibrillation to a minimum; otherwise the fiber will fibrillate increasingly.

This paper also proposed a simple mathematical model to predict the fibrillation index from the values of intrinsic viscosity and relative crystallinity. The model also implies a positive correlation between the relative crystallinity and the fibrillation index, while the intrinsic viscosity shows the opposite correlation.

Further studies on the three-dimensional measurement of the fibrillation index should be conducted, and then how those parameters affect this new, measured fibrillation index should be explored.

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