

Crystallinity of yellow colored silkworm variety cocoons

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Abstract

The structure and properties of silk polymers (fibroin and sericin) can be satisfactorily controlled by choosing a suitable silkworm variety and, hence, this parameter (i.e., silkworm variety) has attracted increasing attention. A previous study reported that the crystallinity of white colored silkworm cocoons depends on the silkworm variety. In the present study, sixteen yellow colored silkworm variety cocoons were produced and their molecular conformation and crystallinity were investigated. The conformation of the silkworm cocoons varied with the silkworm variety. Most cocoons exhibited β -sheet conformation, although random coil and β -sheet conformations co-existed in some cocoons (e.g., 21 and D90). The crystallinity of the silkworm cocoons varied with the silkworm variety and the measurement position of the cocoon (i.e., outer surface or inner surface). However, the difference in the crystallinity indices of the outer and inner surfaces comprising the cocoons varied with the silkworm variety, but was <2% for all cocoons, except for MAL.

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Introduction

Silk is a naturally occurring biomaterial consisting of two polymers: fibroin and sericin. Silk is produced by the silkworm and the *Bombyx mori* silkworm occurs in many different varieties. Also, researchers have bred new silkworm varieties (Kang *et al.*, 2001; Kim *et al.*, 2018).

Recent studies have evaluated the effect of silkworm variety on the structure and properties of silk polymers (fibroin and sericin), and revealed that the structure and properties vary considerably with the silkworm variety. In other words, the structure and properties of silk polymers associated with one variant differ from those corresponding to another variant.

The morphology, molecular weight, and mechanical properties

of regenerated silk fibroin (SF) (Chung *et al.*, 2015a) as well as the structure and properties of sericin (Chung *et al.*, 2015b) vary significantly with the silkworm variety. Moreover, the dope solution viscosity and, in turn, the fiber diameter of electrospun regenerated SF are variety dependent (Park and Um, 2015). However, the post drawing performance of wet-spun SF was found to be insensitive to the silkworm variety, despite the difference in viscosity (Jang and Um, 2015). Lee *et al.* (2017) reported that the color of silkworm cocoons depends on the silkworm variety. Similarly, Kim and Um (2019) reported a variety dependence for the structure and properties of raw sericin on silk. Song *et al.* (2019) reported that the physical properties and chemical structure of silk were strongly dependent on the silkworm variety.

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These studies revealed that the silkworm variety has a strong effect on the structure and properties of the resultant SF and sericin. In other words, the performance of silk polymers can be satisfactorily controlled by choosing a suitable silkworm variety. However, only a few *Bombyx mori* silkworm varieties have been examined in previous studies. Therefore, as a preliminary study, we recently evaluated (via FTIR measurements) the molecular conformation and crystallinity characterizing thirty nine silkworm varieties of white colored silkworm cocoons. The results revealed that the crystallinity of the cocoons varied considerably with the silkworm variety (Park *et al.*, 2019).

The silk textile produced by yellow colored cocoons, which are associated with many *Bombyx mori* silkworm varieties, exhibits a golden luster (Kweon *et al.*, 2012a; 2012b) and, hence, these cocoons have attracted considerable attention recently.

Therefore, in the present study, we produced sixteen different yellow colored silkworm variety cocoons and, as a preliminary investigation, examined the crystallinity of these cocoons via FTIR.

Materials and Methods

Cocoon preparation and silkworm rearing

Sixteen different original *Bombyx mori* silkworm varieties were grown at Kyungpook National University and a total of sixteen silkworm cocoon samples were produced from these sixteen varieties. All varieties of larvae were reared at 25°C on fresh mulberry leaves. Furthermore, pupae and cocoons were kept at 25°C.

Measurement and characterization

The color and external features of yellow colored silkworm cocoons were photographed using a digital camera (PC1310, Canon, China). The molecular conformation and the crystallinity characterizing different silkworm varieties of these cocoons were evaluated via Fourier transform infrared spectroscopy (FTIR; Nicolet 380, Thermo Fisher Scientific, USA) conducted using the attenuated total reflection (ATR) method. A scan range, scan number, and resolution of 4000 cm⁻¹–650 cm⁻¹, 32, and 8 cm⁻¹, respectively, were employed. Seven samples of each silkworm variety were used for the measurements. The crystallinity index was calculated as the intensity ratio of the peaks occurring at

1260 and 1235 cm⁻¹ in the FTIR spectrum. This index was calculated from (Chung and Um, 2014; Kim and Um, 2014; Park *et al.*, 2019):

$$\text{Crystallinity index (\%)} = \frac{A_{1260\text{cm}^{-1}}}{A_{1235\text{cm}^{-1}}} \times 100$$

$A_{1235\text{cm}^{-1}}$: Absorbance at 1235 cm⁻¹

$A_{1260\text{cm}^{-1}}$: Absorbance at 1260 cm⁻¹



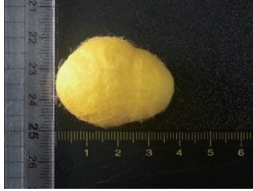


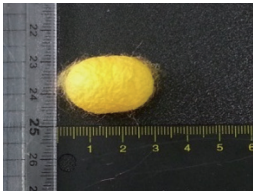


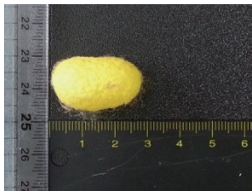


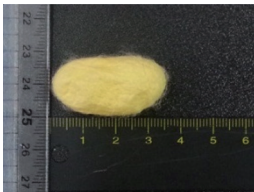




Results and Discussion

Table 1 shows the color and shape of yellow-colored silkworm cocoons with different silkworm varieties. Light yellow cocoons were observed for 1750, BMA, K43, and 7103, whereas intense yellow cocoons were obtained for the other silkworm varieties. Furthermore, the shape and size of the cocoons varied with the silkworm variety.

Previous studies have evaluated the molecular conformation and the crystallinity of silk polymers (Chung and Um, 2014; Kim and Um, 2014; Park and Um, 2016; Park and Um, 2018). Fig. 1 shows representative FTIR spectra of different silkworm cocoon varieties. For most of the cocoons, an IR peak occurred at 1620 cm⁻¹ corresponding to β-sheet conformation (Kim and Um, 2014; Chung and Um, 2014). However, an IR peak at 1620 cm⁻¹ and shoulder at 1645 cm⁻¹ occurred in cocoon 21. Moreover, the peak at 1645 cm⁻¹ arising from D90 is attributed to random coil conformation (Kim and Um, 2014; Chung and Um, 2014). Park *et al.* (2019) reported similar results, i.e., the molecular conformation of the white colored silkworm cocoons varies with the silkworm variety.

The crystallinity of white colored silkworm cocoons of different silkworm varieties was characterized by calculating the crystallinity index from FTIR spectra. A previous study (Park *et al.*, 2019) revealed that the crystallinity of the outer surface associated with a silkworm cocoon differs from that of the inner surface. Therefore, in the present study, FTIR measurements were conducted on the outer surface of silkworm cocoons (see Fig. 2 for the corresponding crystallinities). Among the silkworm varieties, KG2 and C10 exhibited the highest crystallinity (62.0%) and the lowest crystallinity (57.9%), respectively. Fig. 3 shows the crystallinity index of the inner surface comprising yellow colored silkworm cocoons of different silkworm variety. As shown in the figure, KG2 and K43 exhibited the highest

Table 1. Pictures of yellow colored silkworm cocoons associated with different silkworm varieties used in this study.

Silkworm variety	40V	D90	S14	KG2
				
	21	KG01	GS	GSP
				
	Z1	12	C10	MAL
				
	1750	BMA	K43	7103
				

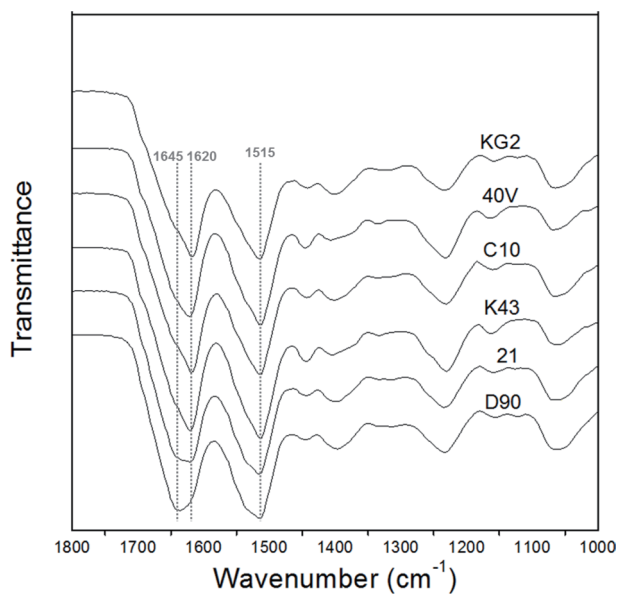


Fig. 1. Representative FTIR spectra obtained for yellow colored silkworm cocoons of different silkworm varieties.

crystallinity (61.8%) and the lowest crystallinity (58.3%), respectively.

Fig. 4 shows the relationship between the crystallinities of the outer and inner surfaces comprising the silkworm cocoon. In the previous study, we classified the different varieties of the white colored silkworm cocoons into three groups, based on the difference in these crystallinities. Group 1 consisted of cocoons with differences of $<2.0\%$; Groups 2 and 3 consisted of cocoons with differences of $>2\%$. However, for Group 2, the crystallinity index of the outside is higher than that of the inside, whereas for Group 3, the index of the inside is higher than that of the outside (Park *et al.*, 2019).

As shown in Fig. 4, most yellow colored silkworm cocoons belong to Group 1, except for MAL, which belongs to Group 2. In the previous study, 20.1% of white colored silkworm cocoons belonged either to Group 2 or Group 3, and in the present study only 6.3% of the yellow colored cocoons belong to these groups.

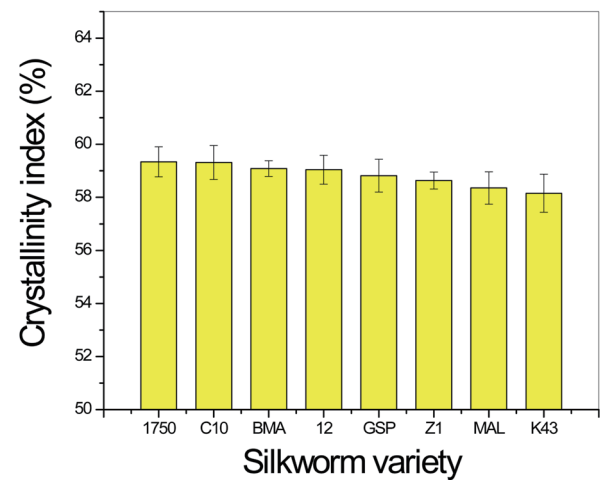
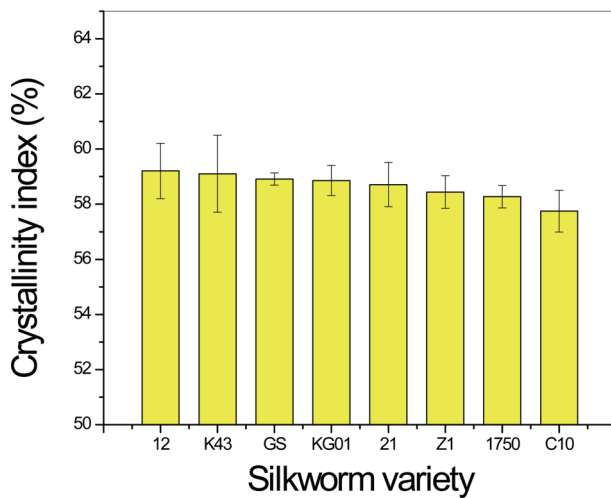
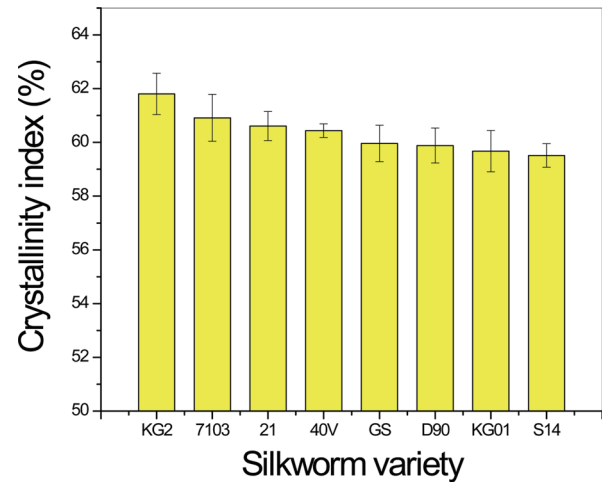
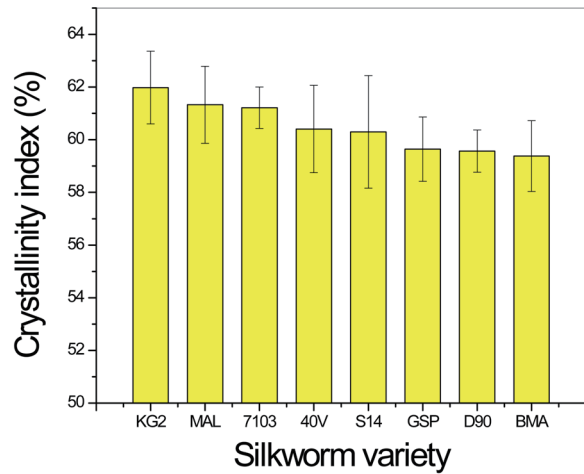


Fig. 2. Crystallinity index of outer surface comprising yellow colored silkworm cocoons of different silkworm varieties.

Fig. 3. Crystallinity index of inner surface comprising yellow colored silkworm cocoons of different silkworm varieties.

Because the number of silkworm variety cocoon sample is too small (i.e. sixteen) in this study, it is difficult to conclude the difference of crystallinity between white colored and yellow colored silk cocoons. It is thought that more silkworm variety cocoons should be surveyed in the future to examine the difference of crystallinity between them clearly.

Conclusions

The color, shape, molecular conformation, and crystallinity of sixteen silkworm cocoon varieties were examined in the present study. The color, shape, and size of the cocoons varied with the silkworm variety. Similarly, as in the case of white colored silkworm cocoons, the crystallinity index of yellow colored silkworm cocoons varied with the silkworm variety.

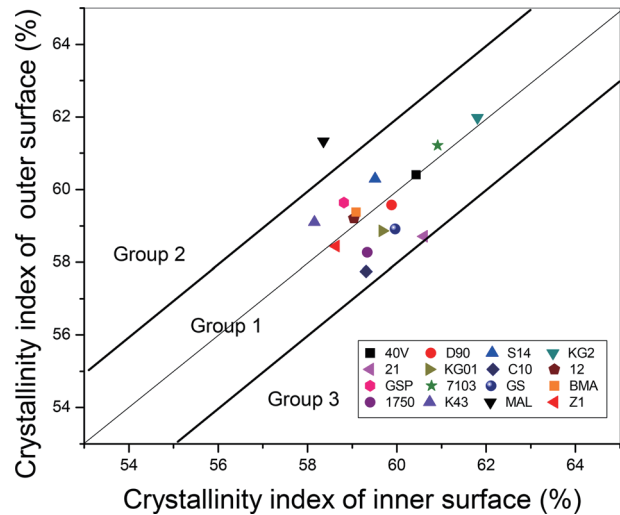


Fig. 4. Relationship between the crystallinity indices of the outer and inner surfaces comprising yellow colored silkworm cocoons of different silkworm varieties.

The crystallinity index associated with the outer surface of the cocoons differed from that of the inner surface. In addition, the magnitude of the difference between the crystallinity indices varied with the silkworm variety. Compared with those obtained for the white colored silkworm cocoons, the differences were quite low. The properties of silk materials depend strongly on the crystallinity of the material (Lee *et al.*, 2016), which varies with the silkworm variety. Owing to the dependence of the crystallinity on the variety, the variety may represent a viable option for controlling the performance of silk materials. Therefore, the effect of silkworm variety on the structure and properties warrants extensive investigation.

Acknowledgments

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