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Comparison of sericin produced through laboratory- and plant-scale extraction

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Abstract

In this study, the structural characteristics of sericin recovered from wastewater released from the silk textile industry (Plant sericin) were comparatively analyzed with those of sericin extracted from a silkworm cocoon produced in a laboratory (Lab sericin). To prepare Plant sericin, ethanol was added to wastewater (i.e., a sericin aqueous solution) after the degumming process to remove nonprotein materials, affording a sericin precipitate. To prepare Lab sericin, nonprotein materials were removed from a silkworm cocoon and sericin was subsequently extracted from the cocoon. Lab sericin and Plant sericin exhibited similar solution viscosities, gel strengths, and crystallinity indices, indicative of the similar molecular weights (MWs) of the two sericin samples. In the case of sericin powder, Plant sericin was more crystalline than Lab sericin due to its treatment with ethanol. The findings of this study revealed that sericin recovered from industrial wastewater can be used equally as its MW is similar to that of sericin obtained through laboratory-scale extraction.

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Introduction

Silk sericin is removed using the degumming process to enhance the luster and touch feeling of silk textiles (Aramwit *et al*., 2012; Blossman-Myer and Burggren, 2010). However, recent studies have revealed the useful properties of sericin, including ultraviolet resistance (Gulrajani *et al*., 2008), antioxidant effect (Suzuki *et al*., 2004; Zhaorigetu *et al*., 2007), antibacterial properties (Jassim and Al-Saree, 2010; Saha *et al*., 2019; Schäfer *et al*., 2023), wound-healing effect (Aramwit *et al*., 2009; Nagai *et al*., 2009), moisturizing effect on skin, and wrinkle reduction (Padamwar *et al*., 2005). Owing to these unique properties of sericin, researchers have investigated the cosmetic and biomedical applications of sericin (Aramwit *et al*., 2012; Nayak *et al*., 2012; Teramoto *et al*., 2008; Zhang, 2002).

Annually, 50,000 t of sericin are discarded globally from the silk textile industry as a result of the degumming process (Aramwit *et al*., 2012; Wang *et al*., 2014; Zhang, 2002), incurring high wastewater treatment costs. In a laboratory, sericin is extracted from a silkworm cocoon to investigate the characteristics of sericin. Considering that silk is an expensive biomaterial, the recovery of sericin from wastewater released by the silk industry can be beneficial for the silk textile industry as well as the applied study on sericin.

Therefore, studies on the recovery of sericin from the degumming solution after the degumming process have been

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reported (Bascou *et al*., 2022; Da *et al*., 2014; Gulrajani *et al*., 2009; Li *et al*., 2015; Wu *et al*., 2007; Yang *et al*., 2013; Zhao *et al*., 2018). However, a comparative study on sericin extracted from a cocoon and that recovered from the silk industry wastewater has not been conducted yet. In particular, it is important to obtain similar molecular weight (MW) of sericin from wastewater to that of sericin from silkworm cocoon in a laboratory. That is, although sericin can be recovered from the wastewater, if its MW is lower than the sericin extracted from silkworm cocoon in the laboratory, the sericin from wastewater cannot replace the sericin from silkworm cocoon.

In this study, the silk textile obtained from the silk textile industry was degummed with hot water at high temperatures and high pressures, and sericin was recovered from wastewater (Plant sericin). Sericin was also extracted from a silkworm cocoon in a laboratory (Lab sericin). The structural characteristics of Plant sericin and Lab sericin were examined to compare their MW. Finally, it was investigated whether sericin recovered from a silk plant can substitute that extracted from a silkworm cocoon in a laboratory.

Materials and Methods

Preparation of silk sericin

In this study, laboratory- and plant-scale extraction of sericin was conducted to obtain sericin. For the laboratory-scale extraction, *Bombyx mori* Baekokjam silkworm cocoons provided by the National Institute of Agricultural Science (Wanju, Republic of Korea) were used. Silk contains silk proteins, such as fibroin and sericin, and nonprotein components, such as wax, carbohydrates, organic matter, and pigments (Cao and Zhang, 2016; Wang *et al*., 2012). Initially, the silkworm cocoons were immersed in a 70% (v/v) ethanol aqueous solution at 50 °C for 2 days to remove the nonprotein components. The ratio of the cocoon and ethanol aqueous solution was set to 1:30 (w/v). Then, the silkworm cocoons were washed with purified water and dried at room temperature (Kim and Um, 2019). The purified water was obtained using a water purification system (RO50, Hana Science, Seongnam, Republic of Korea) with a reverse osmosis membrane. The silkworm cocoons were immersed in the purified water and treated at 120 °C for 60 min using an autoclave (JSAC-60, JSR, Gongju, Republic of Korea) and the high-temperature high-pressure method. The ratio of the cocoon and purified water was set to 1:25 (w/v). After hot water treatment, the sericin aqueous solution was filtered using a nonwoven fabric to prepare a 1.0% (w/w) aqueous sericin solution (Park *et al*., 2018), which was termed Lab sericin.

For the plant-scale extraction of sericin, silk textile weaved with silk yarns made from silkworm cocoons, which were produced in the Republic of Korea, were degummed at 120 °C for 60 min using the high-temperature high-pressure method to prepare a 3.95% (w/w) sericin aqueous solution, which was termed Plant sericin.

In the case of Lab sericin, to prepare sericin powder, the sericin aqueous solution was poured into Petri dishes and dried at 60°C in a drying oven (WOF-50, Daihan Scientific, Wonju, Republic of Korea) to obtain solid sericin, which was ground to produce Lab sericin powder.

In the case of Plant sericin, ethanol was added to the sericin aqueous solution (wastewater) to prepare a 70% (v/v) sericin–ethanol aqueous solution. Next, the sericin solution was stored at 50 °C for 2 days to remove nonprotein components (Kim and Um, 2019) and low-MW sericin (Oh *et al*., 2011). Then, the sericin–ethanol aqueous solution was filtered using a polyester nonwoven fabric to obtain precipitated sericin, which was dried at room temperature to obtain solid sericin; this solid was ground to obtain Plant sericin powder.

The Lab sericin and Plant sericin powders were dissolved in 98% formic acid at 55 °C for 30 min to prepare a 0.3% (w/w) sericin–formic acid solution. Further, these solutions were poured into Petri dishes and dried at room temperature to prepare sericin films (Jang and Um, 2017; Park *et al*., 2018).

The Lab sericin and Plant sericin powders were redissolved in purified water at 90 °C for 5 min and filtered using a filter paper (WF1-0900, Whatman, Maidstone, United Kingdom) with a pore size of 11 μm (Lee *et al*., 2023). Finally, a 0.87% sericin aqueous solution was prepared and poured into a specimen cup and stored at 4 °C for 7 days to fabricate sericin gels (Jo *et al*., 2015). The diameter and thickness of the sericin gels were 45 mm and 13 mm, respectively.

Measurement and characterization

The shear viscosity of the 0.3% silk sericin–formic acid solution was measured using a rheometer (MARS III, Thermo Fisher Scientific, Karlsruhe, Germany) at 25 °C with a 60-mm cone and plate geometry and a cone angle of 1° at 25 °C as a function of the shear rate (0.01–100 s^{-1}).

The gel strength of sericin was measured using a rheometer with a 35-mm parallel plate geometry at 25 °C. An axial test was conducted to examine the gelation behavior of sericin. The 35-mm plate of the rheometer compressed the sericin samples at a speed of 0.2 mm/s (Jang and Um, 2017; Park *et al*., 2018). By performing the axial test, the compression strength was estimated, corresponding to the gel strength of the sericin samples.

To examine the effect of temperature on the shear storage modulus (G′) of the sericin gel, G′ was monitored by performing a temperature sweep oscillation test. During the test, the temperature was controlled between 30 °C and 90 °C and the ramp rate was 20 °C/min. Strain and frequency were controlled at 0.01% and 1 Hz, respectively (Jang and Um, 2017).

The molecular conformation and crystallinity of sericin were evaluated through Fourier transform infrared spectroscopy (FTIR; Nicolet 380, Thermo Fisher Scientific, Waltham, MA, USA) in the attenuated total reflection (ATR, Smart iTR ZnSe) mode. The scan range, scan number, and resolution were 4000–650 cm^{-1}, 32, and 8 cm^{-1}, respectively.

The crystallinity index was calculated as the intensity ratio of the peaks observed at 1620 and 1643 cm^{-1} in the FTIR spectrum. The crystallinity index was calculated using Eq. (1). FTIR was performed seven times. The mean and standard deviations of the crystallinity index were obtained through the seven FTIR analyses (Choi *et al*., 2020; Kim *et al*., 2022).

$$\text{Crystallinity index (\%)} = \frac{A_{1620cm^{-1}}}{A_{1643cm^{-1}} + A_{1620cm^{-1}}} \times 100 \quad (1)$$

where $A_{1620\ cm^{-1}}$ is the absorbance at 1620 cm^{-1} that corresponds to the β-sheet crystallite (crystalline region) and $A_{1643\ cm^{-1}}$ is the absorbance at 1643 cm^{-1} that corresponds to the random coil conformation (amorphous region).

The silk sericin powders and films were stored under standard conditions (20 °C and 65% relative humidity) for 24 h to determine their moisture regain, which was calculated using Eq. (2) (Park *et al*., 2018). The dry weights of the silk sericin samples were determined using a moisture-balance instrument (XM60, Precisa Gravimetrics, Dietikon, Switzerland).

$$\text{Moisture regain (\%)} = \frac{\text{Initial weight} - \text{Dry weight}}{\text{Dry weight}} \times 100 \quad (2)$$

Differential scanning calorimetry (DSC) curves were recorded on a Thermal Analysis Instrument Q 10 (DS25, TA Instruments, New Castle, USA) in the temperature range of 60 °C–270 °C, scanning rate of 10 °C/min, and nitrogen gas flow of 50 mL/min.

Results and Discussion

The poor mechanical properties of sericin may restrict its use in various fields, including biomedical and cosmetic applications. Therefore, the molecular weight (MW) of sericin has been paid attention to because it strongly affects the crystallinity, hydrophilicity, and mechanical properties of sericin (Park *et al*., 2018).

Previously, the MW of silk proteins was evaluated using sodium dodecyl sulfate–polyacrylamide gel electrophoresis (SDS–PAGE) (Aramwit *et al*., 2010; Um *et al*., 2003). However, extremely broad MW bands were observed when this method was used, making it difficult to precisely estimate the MW of sericin. On the contrary, it was observed that using fast protein liquid chromatography (FPLC) provided a more precise MW distribution of sericin (Park *et al*., 2018; Cho *et al*., 2012; Chung *et al*., 2015; Oh *et al*., 2011). However, it is too delicate to obtain the MW distribution result, making the FPLC measurement a time-consuming process. Previous studies revealed that the solution viscosity, gel strength, and crystallinity index of silk polymers (fibroin and sericin) have a good correlation with their MWs (Cho *et al*., 2012; Chung *et al*., 2015; Park *et al*., 2018). Therefore, in this study, these properties were measured to compare the MWs of Lab sericin and Plant sericin.

Fig. 1 shows the steady-state flow of Lab sericin and Plant sericin to compare their MWs. Two sericin–formic acid solutions exhibited almost the same shear-thinning behavior (Fig. 1A). Notably, the viscosities of the two sericin–formic acid solutions at 1 s^{-1} were very similar (i.e., 8.4 and 8.7 mPa·s), as shown in Fig. 1B, implying that their MW levels are similar.

Hydrogen bonding with an adjacent molecular chain is accelerated by the longer molecular chain as compared to the shorter molecular chain. Therefore, with the increase in the MW of sericin, gelation was accelerated and the gel strength was increased (Jang and Um, 2017; Park *et al*., 2018). Thus, the gel strength of sericin can be another indirect tool to evaluate the MW of sericin. As can be seen in Fig. 2, Lab sericin and Plant sericin exhibited similar gel strength, indicating that their MWs were similar. This result reconfirmed the solution viscosity of sericin shown in Fig. 1.

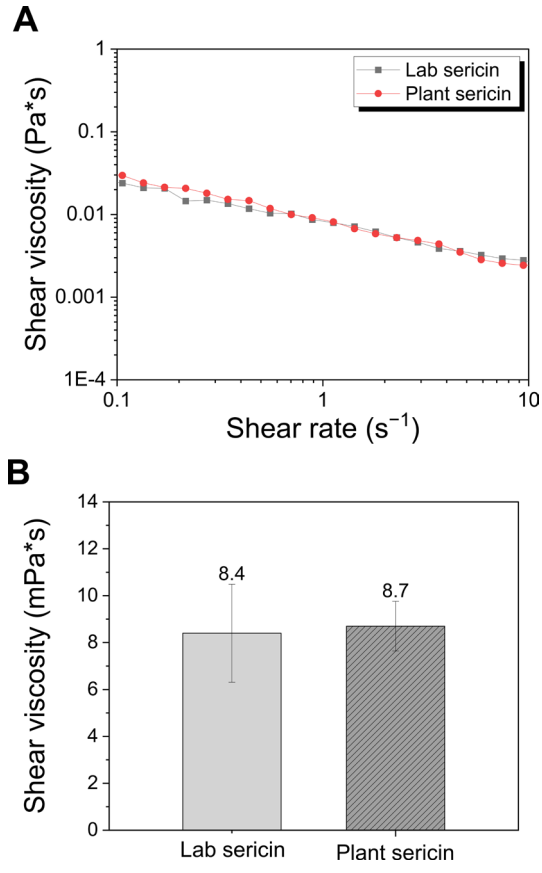

Fig. 1. (A) Steady-state flow and (B) shear viscosity at 1 s^{-1} of 0.3% (w/w) Lab sericin– and Plant sericin–formic acid solutions (n = 3).

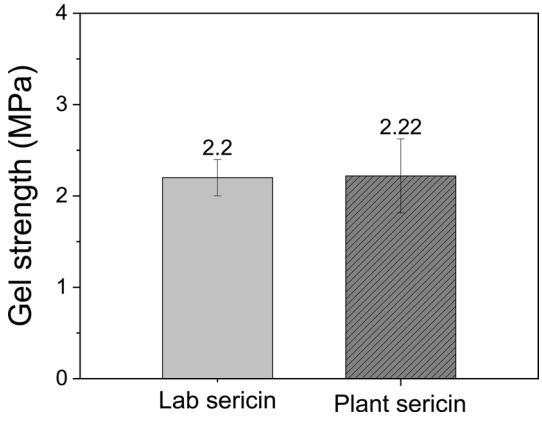

Fig. 2. Gel strength of Lab sericin and Plant sericin (n = 3).

Silk sericin exhibits a thermo-reversible gel–sol transition (Jang and Um, 2017; Jo *et al*., 2015; Kweon *et al*., 2000; Park *et al*., 2018; Zhu *et al*., 1996). The sericin gel was easily transferred into a sol by heating due to the breaking of hydrogen bonds between the sericin molecules. The hydrogen bonds were recovered upon cooling the sericin sol, resulting in the gelation

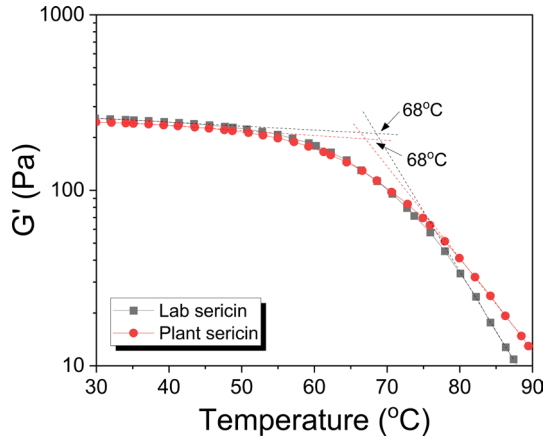

Fig. 3. Temperature sweep test results of the Lab sericin and Plant sericin gels. The sericin concentration was maintained at 0.87% (w/w).

of sericin. This gel–sol transition of sericin was investigated because it provides information regarding the gel structure of the sericin. In particular, Park *et al*. (2018) reported that the gel–sol transition temperature was determined by the MW, and it decreased with a decrease in the MW. Therefore, in this study, the thermal gel–sol transition of the two sericin samples was examined through rheometry. Fig. 3 shows the results.

The storage moduli (G') of the Lab sericin (255 Pa) and Plant sericin gels (243.5 Pa) were similar at room temperature. This result was consistent with the result of the gel strength shown in Fig. 2. As the temperature increased, the G' values decreased, indicating that the sericin gels were broken and transferred to their sol states at elevated temperatures. The critical transition temperature from a gel to a sol was the same for both sericin gels (i.e., 68 °C), indicating that the two sericin samples exhibited similar MWs. This result reconfirmed the results of the solution viscosity and gel strength shown in Figs. 1 and 2, respectively.

As mentioned above, the crystallinity of sericin exhibited a good correlation with the MW of sericin. Therefore, the FTIR measurement of the Lab sericin and Plant sericin films was performed, and the crystallinity index was calculated from the FTIR spectra. The results were shown in Fig. 4. Both sericin films exhibited IR absorption at 1620 cm^{-1}, corresponding to the β-sheet crystallite (Bae and Um, 2021; Choi *et al*., 2020; Park *et al*., 2019a; Park *et al*., 2019b). This result was attributed to the fact 1) that both sericin films were cast from formic acid and 2) casting with formic acid induced the formation of β-sheet crystallites (Um *et al*., 2003). The two

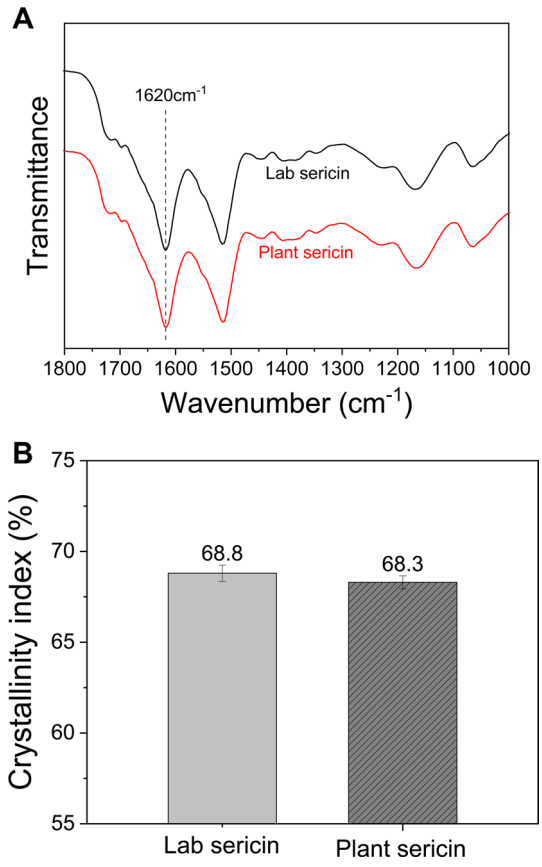

Fig. 4. (A) ATR–FTIR spectra and (B) crystallinity index of a sericin film cast from a 0.3% (w/w) sericin–formic acid solution (n = 7).

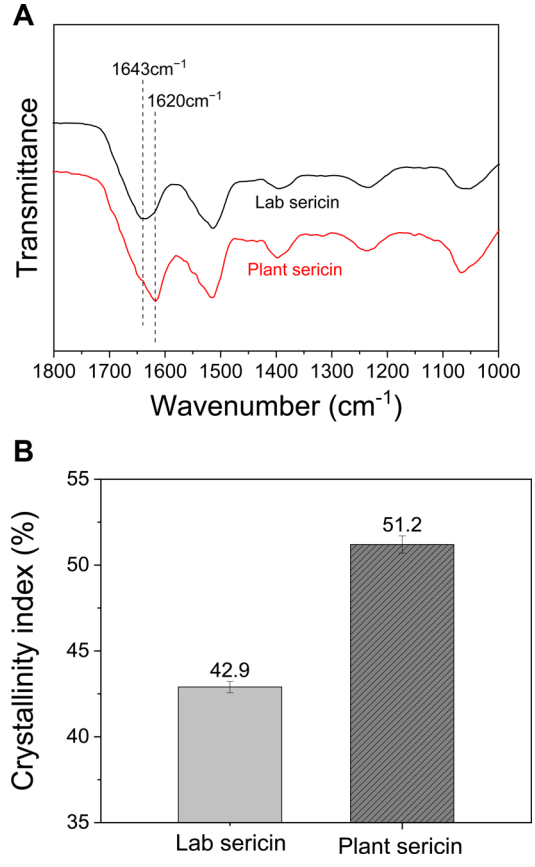

Fig. 5. (A) ATR–FTIR spectra and (B) crystallinity index of the Lab sericin and Plant sericin powders (n = 7).

FTIR spectra were similar.

The crystallinity indices of the two sericin films (Fig. 4B) were also extremely similar, confirming the result of the FTIR measurement (Fig. 4A). Considering that the crystallinity exhibited a good relation with the MW of sericin, the result of crystallinity reconfirmed that the MWs of two sericin films were extremely similar.

Based on the results of the solution viscosity, gel strength, and crystallinity index of Lab sericin and Plant sericin, it can be concluded that their MWs were found to be similar.

Fig. 5 showed the ATR–FTIR measurement results and the crystallinity index of Lab sericin and Plant sericin powders. Lab sericin exhibited an IR absorption peak at 1643 cm^{-1}, corresponding to the random coil conformation, while Plant sericin exhibited an IR peak at 1620 cm^{-1}, corresponding to the β-sheet crystallites (Bae and Um, 2021; Choi *et al*., 2020; Park *et al*., 2019a; Park *et al*., 2019b). The higher crystallinity index of Plant sericin (51.2%) compared to Lab sericin (42.9%) confirmed the different FTIR spectra of the two sericin powders. Notably, the FTIR results of the sericin films and powders were different, possibly due to the differences in the 1) preparation methods of the film and powder and 2) preparation conditions of Lab sericin and Plant sericin. The Lab sericin powder was obtained by drying a sericin aqueous solution. Therefore, it was not crystalline; hence, it exhibited a random coil conformation. On the contrary, the Plant sericin powder was produced by drying a sericin–ethanol aqueous solution because ethanol was added into the sericin aqueous solution to remove the nonprotein components. Because sericin was crystallized using ethanol, the dried Plant sericin powder exhibited β-sheet crystallites (Fig. 5).

The similar crystallinities of the Lab sericin and Plant sericin films were attributed to the dissolution and crystallization in formic acid, i.e., although the Lab sericin and Plant sericin powders exhibited different crystallinities, the β-sheet crystallites of sericin powders were disrupted during dissolution in formic acid. As a result, all sericin molecules exhibited a random coil conformation in formic acid. Finally, the sericin molecules

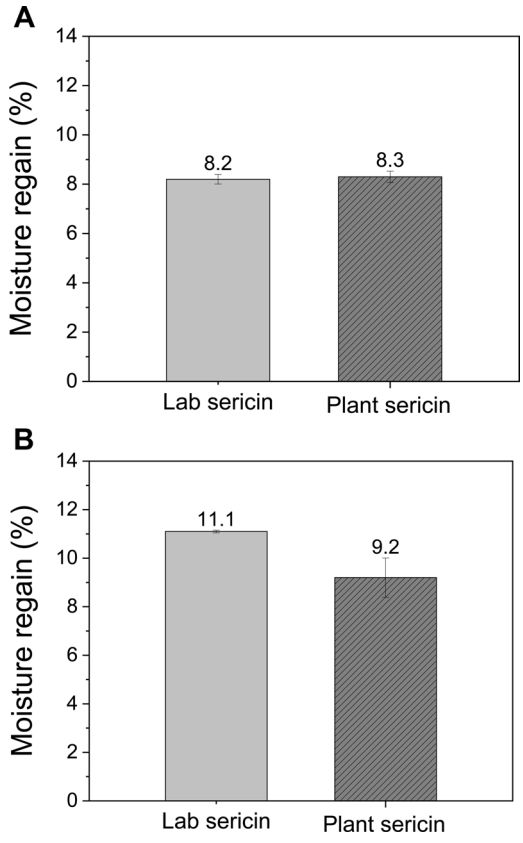

Fig. 6. Moisture regain of the (A) Lab sericin and Plant sericin films cast from a 0.3% (w/w) sericin–formic acid solution and (B) Lab sericin and Plant sericin powders (n = 3).

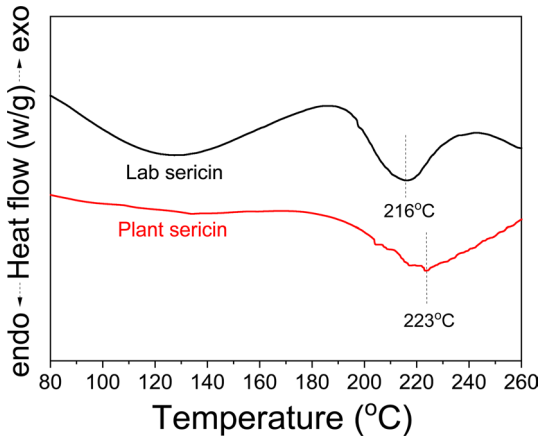

Fig. 7. DSC thermograms of the Lab sericin and Plant sericin powders.

became crystalline due to casting with formic acid. Hence, Lab sericin and Plant sericin films exhibited almost the same crystallinity (Fig. 4).

Water absorption ability is one of the key properties of biomedical and cosmetic materials, considering that these materials are used in the wet state (Bae *et al*., 2022). Fig. 6A shows the moisture regain of sericin films. The Lab sericin (8.2%) and Plant sericin (8.3%) films exhibited similar moisture regain values. This result can be easily understood by considering that the crystallinity of the sericin film strongly affected the moisture regain (Park *et al*., 2018) and that the crystallinity indices of the Lab sericin and Plant sericin films were similar (Fig. 4). On the contrary, the moisture regain of Lab sericin (11.1%) was greater than that of Plant sericin (9.2%) owing to their different crystallinities, i.e., the crystallinity of Plant sericin was greater than that of Lab sericin (Fig. 5B). Owing to the higher amounts of the crystalline regions in Plant sericin than in Lab sericin, the access of water was restricted considerably more in Plant sericin. In addition, the higher moisture regain values of sericin powders (Fig. 6B) than those of the sericin films (Fig. 6A) were attributed to the lower crystallinities of sericin powders (Fig. 5B) than those of the sericin films (Fig. 4B). This result reconfirmed the reverse relation between the crystallinity and moisture regain of sericin.

Fig. 7 showed the DSC thermograms of the Lab sericin and Plant sericin powders. The Lab sericin powder exhibited a broad endothermic peak at 120 °C–130 °C, corresponding to the vaporization of water, and an endothermic peak at 216 °C, corresponding to the thermal decomposition of sericin (Lee *et al*., 2018). The Plant sericin powder exhibited an endothermic peak at 223 °C, corresponding to the thermal decomposition of sericin. The presence of the endothermic peak at 120 °C–130 °C for the Lab sericin powder and the absence of the peak at 120 °C–130 °C for the Plant sericin powder were related to their crystallinity, i.e., Lab sericin was mainly composed of amorphous regions (Fig. 5); therefore, water was absorbed easily on the Lab sericin powder, resulting in the endothermic peak of water vaporization. However, the Plant sericin powder was crystalline (Fig. 5); hence, the Plant-sericin powder absorbed less water, resulting in the absence of the peak of water vaporization in the DSC curves.

The higher thermal decomposition temperature of Plant sericin (223 °C) compared to Lab sericin (216 °C) was also attributed to the higher crystallinity of the Plant sericin powder than that of the Lab sericin powder. The thermal decomposition of silk polymers (fibroin and sericin) can be delayed by increasing crystallinity (Jo *et al*., 2015).

Conclusions

In this study, two sericin samples (i.e., Lab sericin and Plant sericin, respectively) were prepared using different scales and preparation methods, and their structural characteristics were examined. In addition, their solution viscosity, gel strength, and film crystallinity were found to be quite similar, indicative of their similar MWs. In the case of sericin powder, Lab sericin exhibited an amorphous region, while Plant sericin exhibited more crystalline characteristics due to its treatment with ethanol. The results revealed that sericin recovered from the wastewater released from the silk industry could replace sericin with a similar MW extracted in a laboratory. These findings can be used to industrialize sericin on a larger scale with a lower production price for bio-related fields, including cosmetic and biomedical applications.

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