SILK SERICIN AND ITS APPLICATION IN FOOD INDUSTRY: AN OVERVIEW

Abstract

Sericin degummed in substantial amounts during the silk reeling process of cocoon boiling may be a valuable source of ingredients for food packaging and storing with immense health-beneficial effects due to high presence of natural antioxidants. Sericin derived from silk cocoon contains a variety of important amino acids, including serine, glycine, and aspartic acid. Similarly, sericin shows hydrophilic properties, which impart beneficial biological biocompatible *aualities* such antibacterial, antioxidant, and antielastase activity. Moreover, sericin when combined with additional biomaterials such chitosan, glycerol, and carbon-based nanoparticles proved to be successful in the production of films, coatings, and packaging of food materials. The present study provides a critical review of the utilization of waste sericin and its various application, particularly in the food industry sector.

Keywords: Coating, Food, Preservation, Sericin.

Authors

Shubhajit Shaw

Department of Sericulture Raiganj University Raiganj, Uttar Dinajpur, India.

Debnirmalya Gangopadhyay

Department of Sericulture Raiganj University Raiganj, Uttar Dinajpur, India. debganguly.rgu@gmail.com

I. INTRODUCTION

Silk fibres which are widely recognized as the queen of textiles play a significant role in the worldwide textile market because of their unique lustre and elegance. Generally, four types of silk namely, mulberry, tasar, muga and eri are exploited for large scale production of silk. Mulberry silk is secreted by the domesticated silkworm, Bombyx mori and contributes around 90% of the total world raw silk production. Tasar silk is secreted by the wild silkworms, Antheraea mylitta, A. pernyi, A. proylei, A. yamamai while muga and eri silks by the silkworms, A. assamensis and Samia ricini and collectively known as non-mulberry silks. The boiling of the silk cocoon facilitates the degumming of sericin, the gluey substance present in the cocoon, thus helping to unwind the silk filament in the silk reeling wheel. Generally, a cocoon consists of 80% fibroin and 20% of sericin content. It is estimated that 50,000 MT of such sericin being generated as silk-industrial waste annually by the global silk industry cause serious environmental pollution and health hazards. (Seo et al., 2023) However, sericin is a highly proteinous substance, therefore, it can be applied to make several highly value-added products, particularly in the food, cosmetic and biomedical industrial sectors. This study provides a critical review of the utilization of waste sericin and its various application, particularly in the food industry sector (Kunz et al., 2016).

II. PROPERTIES OF SERICIN

The properties of sericin may be classified broadly into three distinct types which may include biophysical, biochemical, and biological characteristics (Figure 1). Those properties of sericin are summarized below-

- **1. Biophysical Properties:** The biophysical properties of sericin may further be classified based on water solubility, thermal stability, and UV protection.
 - Water Solubility: Sericin possesses considerable hydrophilicity and water solubility properties, primarily because of high presence of amino acids composition such as glycine (17%), serine (32%), and aspartic acid (16%). Besides, the secondary structure of sericin which consists of a random coils, β sheet, and low content of α helix greatly affected by temperature. For instance, the high temperature of water during the time cocoon boiling results in insoluble development, while the low temperature converts the secondary structure of sericin from random coil into β sheet, thus plays an important role in gel formation for application in various biomaterial sectors (Reddy and Aramwit, 2021).
 - Thermal Stability: The stability of sericin with changes in temperature and time can be evaluated through thermogravimetric studies. A great deal of variation was observed in the structure and properties of silk extracted from different silkworm species. For example, Sahu et al. (2016) who studied the thermal stability of sericin at different temperature regimes using both the mulberry and non-mulberry silks reported that the sericin extracted from non-mulberry silks, particularly from eri silk is having highest thermal stability than those of the sericin extracted from mulberry silk.

- UV Protection: The importance of sericin to protect the cells against UV radiation has been widely recognized. Various studies have shown that sericin extracted from the mulberry silk helps to reduce the skin oxidative stress via inhibiting the harmful effect of UVB rays-induced programmed cell death (Dash et al., 2008; Reddy and Aramwit, 2021). The in-vitro studies on the sericin extracted from non-mulberry silk showed significant reduction in UVA and UVB-induced apoptosis as compared to those of the sericin extracted from mulberry silk. Further, a comparative study on sericin extracted from different types of silks showed that mulberry silk though, enhances UVA protection level alone, however, muga silk in addition to UVA protection promotes collagen formation from UVA and UVB exposure. Thus the study suggests that sericin from non-mulberry silks are more photoprotective than those of the sericin from mulberry silk (Kumar et al., 2018).
- **2. Biochemical Properties:** The biochemical properties of sericin may be categorized according to its antioxidant, anti tyrosinase, anti elastase and anti lipid peroxidase properties and summarized below-
 - Antioxidant: The oxidative damage caused by free radicals, scavenging activity of luminol-based free radicals and quantification of free radicals have been measured through various antioxidant assays like DPPH, TRAP and ESR respectively. The sericin extracted from yellowish-green cocoons demonstrated better antioxidant properties than white cocoons of mulberry silk (Takechi et al., 2014). The particle size of sericin extracts vary greatly according to the cocoon colours and strains of silkworm. Several studies indicated that sericin derived from reeling waste products from mulberry and tasar cocoons though retain its antioxidant activities, however, the extraction method largely affects its antioxidant potential (Reddy and Aramwit, 2021). For instance, sericin extracted following the autoclave method show more antioxidant properties than those of the acid extraction method.
 - Anti Tyrosinase: The tyrosinase protein catalyses oxidation process in monophenols, transforming them into diphenols and quinones. Browning is a common side effect of this procedure in fruits and vegetables. Some kind of human ailments like cancer and Parkinson's diseases are affected by tyrosinase (Xing et al., 2016). Sericin has been shown to have anti-tyrosinase activity in several investigations, and the activity varies depending on the strain and extraction technique. A study shows that the method for the extraction of sericin through *Bombyx mori* was compared using various techniques, including urea, temperature, acid, and alkaline chemicals, which contribute to the inhibition of tyrosinase. Sericin's strongest antityrosinase activity was seen when it was extracted with urea. This is due to the abundance of valine and arginine, both of which have a strong affinity for tyrosinase (Reddy and Aramwit, 2021).
 - Anti Elastase: A proteolytic enzyme called elastase works to break down elastin, which causes the skin to lose its elasticity. UV light can stimulate the production of elastase protein. Sericin isolated from cocoons of several breeds of the mulberry silkworm, *B. mori* and tasar silkworm, *Antheraea spp*. have been investigated to study their anti-elastase action. The study found that sericin's antielastase activity may be immensely useful for sun protection of human skin. Thus, it was advocated that

sericin can be a valuable and profitable ingredients for cosmetic industries. More interestingly, sericin isolated from the silk industry waste products also exhibited sufficient anti-elastase action, thus, waste recycling of sericin products from the silk industry can open up new avenues of additional income generation not only to the silk reelers but also to stakeholders associated with cosmetic industries (Reddy and Aramwit, 2021).

- Anti-Lipid-Peroxidase: Kato et al. studied the sericin protein for its anti-lipid peroxidation action for the first time in 1998. Sericin extracted by heating from mulberry silk cocoon prevents the lipid peroxidation process. Moreover, sericin isolated from non-mulberry silk cocoons also found to decrease the lipid peroxidation. The response against lipid peroxidation though vary greatly ranging from 75 to 90% based on concentration of the sericin, however, the extraction procedures including autoclaving and alkali do not affect the anti-lipid peroxidation activity (Kumar and Mandal, 2019). The sericin from the silk cocoons not only found to have anti-lipid peroxidation activities but also the sericin obtained from the silk industrial waste products showed significant amount of anti-lipid peroxidation potential (Jena et al., 2018).
- **3. Biological Activities:** The biological activities of sericin may be categorized according to its antibacterial, anti-inflammatory and collagen formation and summarized below-
 - Antibacterial Activity: The antibacterial responses of sericin obtained from both mulberry and non-mulberry silk cocoons against different bacteria like E. coli, S. aureus, and P. aeruginosa have been investigated (Aramwit et al., 2020). It has been found that the extraction procedure greatly affect the quality of sericin and its antibacterial potentiality. Pure sericin has antibiotic-like action against S. aureus despite having a significantly low response against P. aeruginosa and E. coli. The sericin extracted from autoclaving method shows little effect only in S. aureus. Sericin has been shown to reduce biofilm development; however, the technique of extraction influences its efficacy. In terms of inhibitory and disruption effects, ureaextracted sericin had the greatest potential anti-biofilm action for Streptococcus mutants. Heat and acid-extracted sericins inhibited biofilm development in a dosedependent manner, but alkaline-extracted sericin had no inhibitory effects on the bacterial biofilm. Sericin's changed structure may sabotage the cell wall of bacteria, resulting in the weakening of the membrane and cell apoptosis. The type of cocoons and technique of processing are important variables in sericin selection. The most significant attribute may not be the antibacterial activity, but it exhibits a wide range of bioactivity qualities for medicinal purposes. Combining sericin with additional antibacterial compounds can boost its response and increase the quality of biomaterials. Sericin-derived biomaterials have been coupled with biopolymers, and chemical agents, to improve their antibacterial and biological capabilities.
 - Anti-Inflammatory Properties: Several investigations reported that sericin of mulberry silk cocoons has anti-inflammatory properties. Aramwit et al. (2013) found that sericin primarily increased the response of pro-inflammatory cytokines such as TNF-α (tumour necrosis factor- α) and IL-1 (interleukin-1), however, after seven days of long-term medication, the inflammation did not worsen. Sericin produced

inflammation at the initial phase of therapy in this trial, but it did not speed up the course of wound inflammation. Sericin's anti-inflammatory effect might be employed for a variety of applications, including wound healing, nano-micelles for tumour therapy, and drug delivery nanoparticles (Reddy and Aramwit, 2021).

• Collagen Formation: Sericin induces fibroblast cell proliferation as well as collagen synthesis. Sericin obtained using four different extraction procedures including heat, urea, acid, and alkali promoted collagen synthesis at different concentrations (Aramwit et al., 2013). The heat extraction approach showed the most significant response to collagen production. Sericin's amino acid content influences collagen synthesis's efficacy. The amino acid contents of several silkworm strains were discovered. Sericin protein has a high concentration of methionine and cysteine residues, which increases collagen formation.

III.SERICIN AND ITS APPLICATION IN FOOD-SECTOR INDUSTRIES

It is estimated that more than 0.8 billion people globally are suffering from malnutrition, and roughly 40% of newly harvested and domestically produced goods end up wasted. In this direction, if the shelf life of food can be enhanced by even just one week, it would be a significant benefit to the agricultural industries. This would also have a positive impact on reducing the amount of waste in the world, especially as 30% of food purchased in stores goes uneaten (Seo et al., 2023). Plastic pollution is a growing concern, and researchers have identified natural and biodegradable food packaging solutions as an alternative to synthetic polymers. Synthetic polymers are harmful to the environment as they are nonrenewable and non-biodegradable. The use of biodegradable polymers can help mitigate these concerns (Low et al., 2022). Biopolymers are a favourable option as they are not only biodegradable but also readily available, renewable, nontoxic, and can be derived from numerous natural origins. However, scientists are conducting research on different kinds of biopolymers to characterize, and understand their features, and create food packaging materials from natural nutrients to substitute synthetic or artificial polymers. The trend of research on application of sericin for food preservation using the keywords "sericin", "food", "coating" and "preservation" from SCOPUS database during the year 2007 to 2023 is presented in Figure 2. The figure depicts that research on sericin as food coating materials has gained wide popularity among the global scientific community. This section describes application of sericin, a naturally protein based products in the food industry as an important alternative of food coating material to enhance shelf-life of the products.

Food-Coating and Food-Packaging Material: Recent studies on sericin as food coating material demonstrated its high prospects to improve the food quality via enhancing shelf life of the product while proved to be environmental friendly due to its natural origin (Seo et al., 2023). polarity of sericin determines binding ability The its hydrophobicity/hydrophilicity in films. A sericin layer with greater moisture content is created when higher-polarity sericin is used (Meerasri et al., 2022). Therefore, the buildup of sericin hydrolysate to sericin film decreases both moisture content and molecular weight and enhances water-vapour permeability, antioxidant activity, polyphenols, and alkaloids production, as well as overall phenolic content, due to strong acid hydrolysis (Meerasri et al., 2022). The physical characteristics of sericin film are enhanced by reinforcing it with nanocelluloses (bamboo-derived cellulose nano-fibrils) and it shows significant antioxidant properties (Kwak et al., 2018). Another researcher discovered that utilising a sericin-based edible covering material that includes aloe-vera, glycerol, and chitosan increases tomato shelf life at 25°C and 70% RH. In post-harvest situations, it is possible to preserve the same amount of fruits and avoid ageing when compared to uncoated fruits. Moreover, the ATR-FTIR study demonstrates that the coating substance has no effect on the fruit's structure (Tarangini et al., 2022). It also increases the elasticity of the film by working with water molecules synergistically.

The influence of sericin-based edible coatings on the quality and storage life of tomatoes held for 40 days at a temperature of 25°C and 70% RH have been evaluated (Tarangini et al., 2022). The results showed that using this coating substance reduces the weight and firmness of the tomatoes, as shown in Figure 3 A-C. Furthermore, as storage time increased, the titratable acid level increased, while the total antioxidant content, phenol content, soluble solid content, lycopene, and pH concentration kept being lower relative to untreated tomatoes. Oh et al. (2021) carried out another study that indicated the feasibility of using sericin in the food packaging industry. The inherent deficiencies of the sericin film, such as mechanical characteristics and waterproofing, had been substantially enhanced due to the chemical cross linking responses depending on the Maillard reaction developed. Thus, the sericin coating developed using glucose found to reduce food oxidation. Another study by Me et al. (2022) demonstrated that a mixture of silk, chitosan, and carbon dots when utilized for food storage, it prevents nutrient loss and retain the significant quality of the fruits. A film was formed via the combination of sericin with agarose, to increase its mechanical characteristics. The Ag/ZnO nanoparticles and polydopamine were coated with sericin/agarose flim and showed high adhesive properties without surface characteristics (Li et al., 2020). As a result, the coating developed displayed exceptional antibacterial activity response against gram^{+ve} (E. coli) and gram^{-ve} (S. aureus). Linking with Ag/ZnO increases the mechanical characteristics and exceptional antibacterial activity. Polymeric films with similar qualities might be utilized to cover different surfaces to kill bacteria (Seo et al., 2023).

IV. LIMITATIONS AND PROSPECTS OF USING SILK SERICIN

Sericin has some drawbacks that make it inappropriate for application in food-related sectors in the future. This also applies to its application in food packaging, which is hampered by poor mechanical qualities caused by sericin self-aggregation, resulting in low mechanical strength. Furthermore, due to its hydrophilic nature, it is unsuitable for usage in a water-environment, which is a big limitation that prevents it from being used in a range of sectors. Furthermore, the powder sericin's limited solubility in a solvent and the terrible scent of silkworm insects are barriers to its widespread utilisation in nutritious, effective foods and supplements. (Table 1).

V. CONCLUSIONS AND FUTURE PROSPECTS

Sericin is a protein generated spontaneously by the *B. mori* silkworm insect. This glycoprotein is water-soluble that makes up around 25-30% of a silk cocoon. Sericin's characteristics vary depending on the technique of extraction, and it has a wide (10-400 kDa) molecular weight range. Sericin comprises both amino acids, hydrophobic and hydrophilic, which confer antibacterial, antioxidant, anti-tyrosinase, anti-inflammatory, and anticancer characteristics. Sericin when coupled with other biomaterials, has a wide range of uses,

notably in the food sector. According to studies, sericin protein has few side effects and allergies, making it a promising functional protein for further investigation in a variety of fields.

Table 1: Prospects and Limitations of Sericin-based Food-Packaging Materials

Prospects and Major Findings	Limitation Addressed	Reference
The shelf life of tomatoes extended significantly by applying sericin-based edible coating supplemented with glycerol, aloe vera, and chitosan compared to uncoated ones.	Low hydrophobicity of sericin in a water-based environment.	Tarangini et al. (2022)
The shelf life of food is extended by the combination of glucose and sericin		Oh et al. (2021)
The physical properties of sericin film can also be improved by reinforcing it with nano celluloses (cellulose nanofibrils derived from bamboo).	Sericin is prone towards self- aggregation due to poor mechanical properties.	Kwak et al. (2018)
The mixture of the sericin/agarose film and Ag/ZnO increases the mechanical characteristics and exceptional antibacterial activity.		Li et al. (2018)
A mixture of silk, chitosan, and carbon dots is utilized for storing food, it appears to prevent nutrient loss and retain the higher quality of the litchi fruit.	The particle size of extracted sericin is large in size and uneven in nature.	Me et al. (2022)

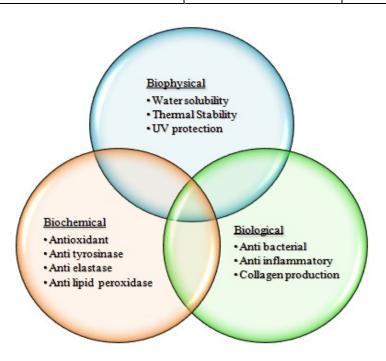


Figure 1: Properties of Sericin

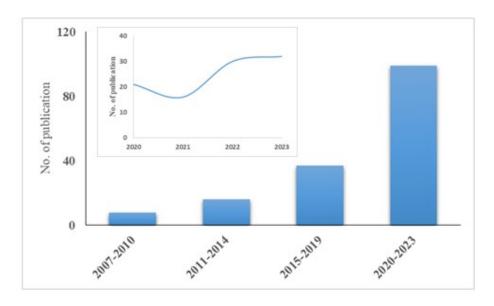


Figure 2: Trend of Research on Application of Sericin for Food Preservation (Inset Figure Shows No. Of Publication 2020-23)

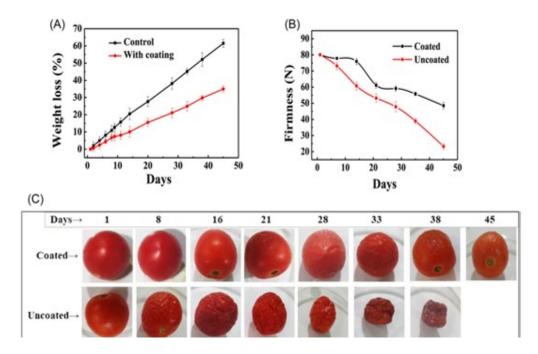


Figure 3:

(A) Weight Loss Effect of a Sericin-Based Edible Coating
(B) Firmness of a Sericin-based Edible Coating After 45 Days of Storage at 25°C
(C) Images of Tomatoes with and without the Edible Sericin Covering Source: Tarangini Et Al. (2022) IIP Series, Volume 3, Book 15, Part 6, Chapter 9 SILK SERICIN AND ITS APPLICATION IN FOOD INDUSTRY: AN OVERVIEW

REFERENCES

- [1] Aramwit, P., Napavichayanum, S., Pienpinijtham, P., Rasmi, Y. and Bang, N. "Antibiofilm activity and cytotoxicity of silk sericin against Streptococcus mutans bacteria in biofilm: An in vitro study." *Journal of Wound Care* 29, no. Sup4 (2020): S25-S35.
- [2] Aramwit, P., Towiwat, P., and Srichana, T. "Anti-inflammatory potential of silk sericin." *Natural Product Communications* 8, no. 4 (2013): 1934578X1300800424.
- [3] Dash, R., Acharya, C., Bindu, P.C., and Kundu, S.C. "Antioxidant potential of silk protein sericin against hydrogen peroxide-induced oxidative stress in skin fibroblasts." *BMB reports* 41, no. 3 (2008): 236-241.
- [4] Jena, K.; Pandey, J.P.; Kumari, R.; Sinha, A.K.; Gupta, V.P.; Singh, G.P. "Tasar silk fiber waste sericin: new source for anti-elastase, anti-tyrosinase and anti-oxidant compounds." *International journal of biological macromolecules* 114 (2018): 1102-1108.
- [5] Kumar, J.P. and Mandal, B.B. "The inhibitory effect of silk sericin against ultraviolet-induced melanogenesis and its potential use in cosmeceutics as an anti-hyperpigmentation compound." *Photochemical & Photobiological Sciences* 18 (2019): 2497-2508.
- [6] Kumar, J.P., Alam, S., Jain, A.K., Ansari, K.M. and Mandal, B.B. "Protective activity of silk sericin against UV radiation-induced skin damage by downregulating oxidative stress." *ACS Applied Bio Materials* 1, no. 6 (2018): 2120-2132.
- [7] Kunz, R.I.; Brancalhao, R.M.; Ribeiro, L.F.; Natali, M.R. "Silkworm sericin: properties and biomedical applications." *BioMed research international* 2016 (2016): 8175701
- [8] Kwak, H.W., Lee, H., Lee, M.E. and Jin, H.J. "Facile and green fabrication of silk sericin films reinforced with bamboo-derived cellulose nanofibrils." *Journal of Cleaner Production* 200 (2018): 1034-1042.
- [9] Li, W., Huang, Z., Cai, R., Yang, W., He, H. and Wang, Y. "Rational design of Ag/ZnO hybrid nanoparticles on sericin/agarose composite film for enhanced antimicrobial applications." *International Journal of Molecular Sciences* 22, no. 1 (2020): 105.
- [10] Low, J.T., Yusoff, N.I.S.M., Othman, N., Wong, T.W. and Wahit, M.U. "Silk fibroin-based films in food packaging applications: A review." *Comprehensive Reviews in Food Science and Food Safety* 21, no. 3 (2022): 2253-2273.
- [11] Meerasri, J., Chollakup, R. and Sothornvit, R."Factors affecting sericin hydrolysis and application of sericin hydrolysate in sericin films." *RSC advances* 12, no. 44 (2022): 28441-28450.
- [12] Mei, S., Fu, B., Su, X., Chen, H., Lin, H., Zheng, Z., Dai, C. and Yang, D.P. "Developing silk sericin-based and carbon dots reinforced bio-nanocomposite films and potential application to litchi fruit." *LWT* 164 (2022): 113630.
- [13] Oh, S., Park, J., Nam, J., Hyun, Y., Jin, H.J. and Kwak, H.W. "Antioxidant and UV-blocking glucose-crosslinked sericin films with enhanced structural integrity." *Reactive and Functional Polymers* 165 (2021): 104942.
- [14] Reddy, N., and Aramwit, P. Sustainable uses of byproducts from silk processing. John Wiley & Sons, 2021
- [15] Sahu, N., Pal, S., Sapru, S., Kundu, J., Talukdar, S., Singh, N.I., Yao, J. and Kundu, S.C. "Non-mulberry and mulberry silk protein sericins as potential media supplement for animal cell culture." *BioMed Research International* 2016 (2016): 1-13.
- [16] Seo, S. J., Das, G., Shin, H. S., & Patra, J. K. "Silk Sericin Protein Materials: Characteristics and Applications in Food-Sector Industries." *International Journal of Molecular Sciences* 24, no. 5 (2023): 4951.
- [17] Sothornvit, R., Chollakup, R. and Suwanruji, P. "Extracted sericin from silk waste for film formation." *Songklanakarin Journal of Science & Technology* 32, no. 1 (2010).
- [18] Takechi, T., Wada, R., Fukuda, T., Harada, K. and Takamura, H. "Antioxidant activities of two sericin proteins extracted from cocoon of silkworm (Bombyx mori) measured by DPPH, chemiluminescence, ORAC and ESR methods." *Biomedical Reports* 2, no. 3 (2014): 364-369.
- [19] Tarangini, K., Kavi, P. and Jagajjanani Rao, K. "Application of sericin-based edible coating material for postharvest shelf-life extension and preservation of tomatoes." *eFood* 3, no. 5 (2022): e36.
- [20] Xing, R., Wang, F., Dong, L., Zheng, A.P., Wang, L., Su, W.J. and Lin, T. "Inhibitory effects of Na7PMo11CuO40 on mushroom tyrosinase and melanin formation and its antimicrobial activities." Food Chemistry 197 (2016): 205-211.
- [21] Yun, H., Kim, M.K., Kwak, H.W., Lee, J.Y., Kim, M.H. and Lee, K.H. "The role of glycerol and water in flexible silk sericin film." *International journal of biological macromolecules* 82 (2016): 945-951.