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Impact of high temperature and pressure on sericin scouring of muga silk cocoons

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Silk cocoons from *Antheraea assamensis* silkworms have been degummed under high temperature and pressure (autoclave) conditions and the efficiency of degumming is assessed in terms of weight loss, reeling length and fibre quality. The silk cocoon sericin removal percentage by autoclave degumming is found to be maximum (23.67%) as compared to conventional alkali degumming (22.28%). The autoclaved fibre is found easily reelable and produce 280 m of length with 10 numbers of fibre breaks. The morphology of fibres shows smooth surface throughout the length and improved mechanical behavior in terms of tensile properties. The molecular conformation estimated by FTIR and thermal behavior analysis shows unchanged nature of the core fibroin fibre degummed under autoclave conditions. Taken together, these results show the utilization of ecofriendly and less hazardous method of autoclave degumming for production of mechanically strong fibres with uniform surface smoothness.

Keywords: *Antheraea assamensis*, Degumming, Muga silk, Reeling, Tensile properties

The silk cocoon consists of two kinds of structural proteins comprising the inner core crystalline nature fibroin and the outer sticky amorphous sericin. The elongational flow of spinning orients the fibroin chains, and the fibroin (liquid) is converted into partly crystalline, insoluble fibrous filaments (solid)[1]. Sericin acts as a protective shield around the fibroin core that functions to lower the shear stress and absorbs the squeezed water from the stretched fibroin during the course of fibre creation[2]. The whole processing of silk from cocoons to the finished product entails the following steps of reeling, weaving, degumming, dyeing and finishing. To obtain a supreme quality of silk with shiny aspect and elegant drape, sericin is required to be eradicated from the fibroin. Sericin scouring is performed by a thermochemical process known as degumming. During degumming sericin protein is hydrolyzed into its subsequent amino acids and solubilized in the degumming media[3,4]. Industrially sodium carbonate is used as the degumming chemical. During the years, several acidic, alkaline and neutral proteases have been practiced as degumming agents on silk fabric. For uniform degumming and good silk quality, alkaline proteases is preferred more as compared to acidic and neutral proteases[5,6]. However, high cost and low performance in silk handling results in limited use of enzymes in the industrial scale. As far as environment welfare is concerned the consumption of chemicals by most of the previously mentioned methods carries serious pollution to the land and water systems[7].

Indigenous muga silk created by the sericogenous insect *Antheraea assamensis* Helfer is intrinsic to the North Eastern realm of India. Gorgeous and attractive golden color, high mechanical properties and long durability are some of the few distinguishing properties of muga silk. Till date degumming of muga silk is reported to be carried out by using sodium carbonate[8] and a bio surfactant (reetha) as the degumming agent[9]. The present study discusses the effect of degumming muga silk cocoon under autoclave conditions of high temperature and high pressure. Degumming under high temperature and pressure conditions unlike chemical degumming is an economic technique producing no toxic and harmful effect on the fibre as well as on environment.

Experimental

Freshly spun cocoons of *Antheraea assamensis* were collected from rearing house of IASST and sodium carbonate was obtained from Merck Co. Ltd.

Conventional Method

Muga cocoons were degummed by following the standard conventional method of boiling in 0.3% sodium carbonate (Na_2CO_3) solution at 90° C for 30 min. The material- to- liquid ratio used was 1:40.

Autoclave Method

Muga cocoons were degummed in an autoclave at 120° C and pressure 15psi for 20 min. The material- to- liquid ratio used was 1:40.

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All the degumming experiments were performed in triplicate. Degumming loss signifies a quantitative estimation of the degumming efficiency by calculating the weight loss before and after degumming treatment with the help of following equation:

$$D = \frac{W_0 - W_1}{W_0} \times 100$$

where W_o is the weight of the original silk fibre (g); and W_1, the weight of degummed silk fibre (g).

Silk cocoons were wet reeled in water at room temperature. The length of reeled silk and the number of breaks during reeling were analyzed manually. The tensile properties, FTIR spectroscopy, thermogravimetric analysis (TGA), differential scanning calorimetry (DSC) and surface morphology (SEM) were studied using standard protocol. All the data were analysed statistically by one-way analysis of variance (ANOVA). Differences between experimental groups at a level of $p < 0.05$ were considered as statistically significant and those at $p < 0.001$ as highly significant.

Results and Discussion

Degumming loss per cent represents an outstanding quality of degumming efficiency with high percentage of degumming loss (23.67) for autoclave degumming and 22.28 for sodium carbonate degumming. The one-way ANOVA analysis shows that the results are highly significant at p value <0.001. Owing to its random structures, sericin of muga silk fibre can be completely hydrolyzed into its subsequent amino acids[10,11] under high temperature and pressure without further deterioration to the fibre content. Sodium carbonate degumming targets the non-covalent bonds of silk fibroin imposing derogatory effects[12,13] on silk fibre and rendering it mechanically weak.

Degumming of silk under high temperature and pressure or boiling in sodium carbonate solution results in yield of continuous fibre by effective removal of the adhesive gum from the silk fibre. The total length of reeled silk is obtained to be 280 m in autoclave degumming and 235 m in sodium carbonate degumming. The number of breaks during reeling are found to be more (30) in case of sodium carbonate degumming as compared to that in case of autoclave degumming (10 only). This might be because of the fact that sodium carbonate attacks the non-covalent bonds of silk fibre, which, in turn, affects its mechanical strength and fibre thickness[13], resulting in fibre breakage.

Tensile strength, elongation, Young's modulus and toughness of sodium carbonate degummed fibre are found to be 4.01±0.22 g/den, 30.85±0.47 %, 70.39±2.71 g/den and 0.719±0.18 g/den respectively. Degumming under high temperature and pressure conditions certainly augmented the tensile behavior of the silk cocoon fibre as compared to conventional degumming, having tensile strength 4.80±0.30 g/den, elongation 33.53±0.52%, Young's modulus 85.30±3.09g/den and toughness 0.994±0.56 g/den. After sodium carbonate treatment the tensile strength of the fibre decreases, indicating partial harmful damages of cross-sectional area and microstructure of core fibroin[14]. Autoclave degumming sequestered sericin protein from silk cocoon fibres without disturbing the chemical bonds and hence the mechanical strength of fibroin filaments.

The FTIR spectrographs of degummed muga silk fibres are shown in Fig. 1(A). The amide I peak of

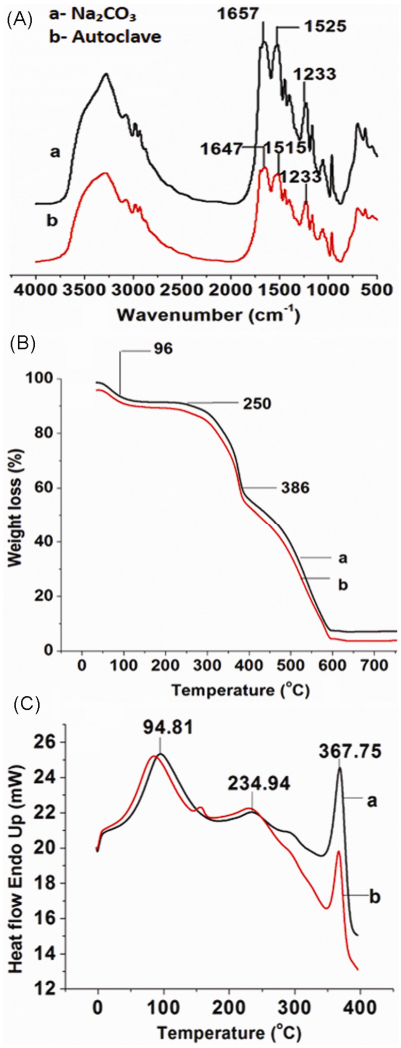

Fig. 1—(A) FTIR spectroscopy, (B) thermo gravimetric analysis, and (C) differential scanning calorimetry of degummed muga silk fibres

sodium carbonate and autoclave treated muga fibre at 1657cm^{-1}and 1647 cm^{-1} respectively can be attributed to α helix/random coil configuration of silk protein[15,16]. The amide I vibration arise entirely due to C=O stretching vibration (about 80%) with little contribution from the out of phase CN stretching vibration, the CCN deformation and the NH in-plane bend. The amide II band implies mainly on in-plane NH bending (40 – 60% of potential energy) and from the CN stretching vibration (18- 40%)[17]. Amide II band shown at 1525 cm^{-1} for sodium carbonate and 1515 cm^{-1} for autoclave degummed fibre is significant of β sheet configuration structure[18]. Similarly, amide III mode is the in-phase combination of the NH bending and the CN stretching vibration with minimum amount of contribution from the CO in-plane bending and the CC stretching vibration[19]. The amide III band at 1233 cm^{-1} for both degummed fibres can be ascertained due to oriented β sheet structure of silk fibre[18]. It is seen that degumming by both the said methods does not alter the intrinsic molecular conformation of muga cocoons.

The TGA study [Fig. 1(B)] shows initial mass loss step in the fibres at around 96°C, which is due to evaporation of moisture. The second event of weight loss arises in the range 250 – 386°C. This is evident owing to the final degradation of highly oriented β sheet crystalline structure of silk[20].

The DSC scrutiny reveals first endothermic peak at around 94°C, which ascertains the loss of water bound to the amorphous region of silk through hydrogen bonding [Fig. 1(C)]. The major endothermic peaks at 234°C and 366°C above the glass transition temperature T_g (190-200°C) signifies phase transition from amorphous to crystalline one[21,22] and finally complete degradation of stable β sheet orientation[23,24]. There is not much apparent difference in thermal behavior of the considered silk fibres.

The surface micrographs of the degummed muga silk cocoon fibres are illustrated in Fig. 2. Surface architecture of the sodium carbonate degummed fibre shows removal of sericin (about 80%), while still leaving certain amount of sericin patches on its surface topology. However, the topography of autoclave degummed fibre shows perfect degumming with sericin molecules breaking away from the fibroin filaments. Surface of autoclave degummed fibre is very smooth, uniform and clean with no sign of damage or destruction to the silk fibroin, showing only longitudinal striations characteristic of fibrillar structure of the truly degummed silk fibres[25].

Degumming of muga silk cocoons under high temperature and pressure conditions shows a good measure of degumming loss (23.67 %), against sodium carbonate degumming (22.28 %). The quality of fibre remains strong with improved shininess after autoclave degumming, which is typical characteristic of muga silk. Autoclave degumming thus have produced an excellent quality of muga silk in terms of sericin loss, surface smoothness and fibre strength in contrast to conventional degumming. Therefore, the present findings propose the use of autoclave cocoon degumming on industrial scale basis as the process is cost-effective and chemical free leaving no hazardous effect on the fibre as well as on the environment.

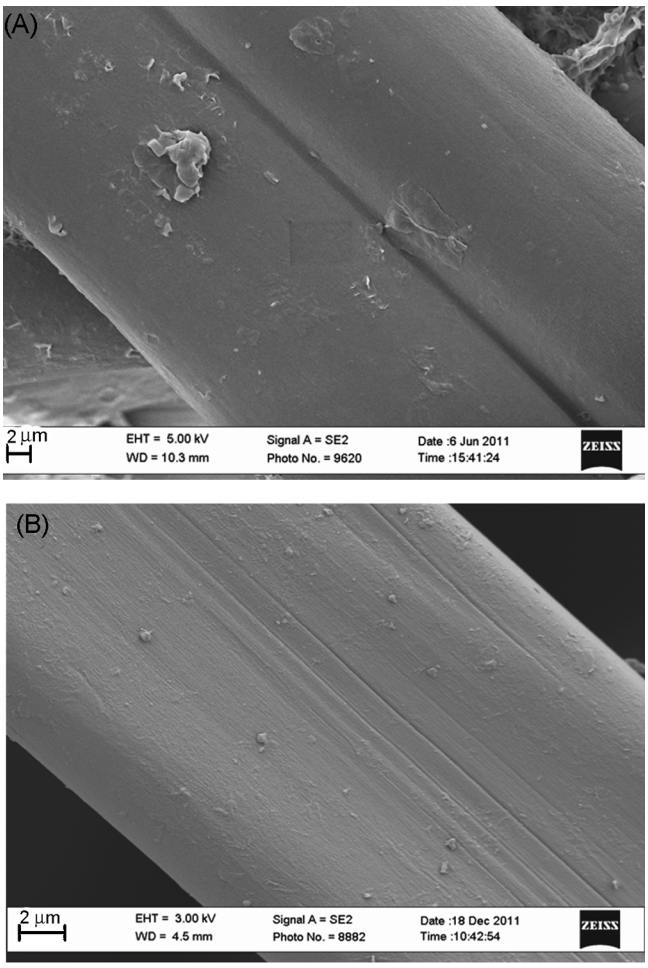

Fig. 2—SEM micrographs of (A) sodium carbonate, and (B) autoclave degummed muga silk fibre

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