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The 89th TIWC

Conference Proceedings

Innovation from Fibre to Fashion

2nd ~ 6th November 2014
Wuhan, China
Lessons from Silkworm Cocoons for Future Protective Materials Design
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Abstract. Evolved over millions of years’ natural selection, very thin and lightweight wild silkworm cocoons can protect silkworms from environmental hazards and physical attacks from predators while supporting their metabolic activity. The knowledge of structure-property-function relationship of multi-layered composite silk cocoon shells gives insight into the design of next-generation protection materials. The mechanical and thermal insulation properties of both domestic (Bombyx mori, or B. mori and Samia. cynthia, or S. cynthia) and wild (Antheraea pernyi and Antheraea mylitta, or A. pernyi and A. mylitta) silkworm cocoons were investigated. The research findings are of relevance to the bio-inspired design of new protective materials and structures.

The 180 degree peel tests and needle penetration tests were used for examining the peel resistance and needle penetration resistance of both domestic and wild silkworm cocoon walls. The temperatures inside and outside of the whole silkworm cocoons under warm, cold and windy conditions were monitored for investigating the cocoon’s thermal insulation function. Computational fluid dynamics (CFD) models were created to simulate the heat transfer through the A. pernyi cocoon wall.

The wild cocoons experienced much higher peeling peak loads than the domestic cocoon. This transfers to a maximum work-of-fracture (WOF) of about 1000 J/m2 from the A. pernyi outer layer, which was 10 times of the B. mori cocoon. The A. pernyi wild cocoon exhibited a maximum penetration force (11 N) that is 70 % higher than a woven aramid fabric. Silk sericin is shown to play a critical role in providing needle penetration resistance of the non-woven composite cocoon structure by restricting the relative motion of fibres, which prevents the sharp tip of the needle from pushing aside fibres and penetrating between them. The wild A. pernyi cocoon exhibits superior thermal buffer over the domestic B. mori cocoon. The unique structure of the A. pernyi cocoon wall with mineral crystals deposited on the cocoon outer surface, can prohibit most of the air from flowing inside of the cocoon structure, which shows strong wind resistance under windy conditions.

Keywords: Biological composite, protective materials, mechanical properties, simulation.

1. Introduction

Wild silkworm cocoon is a multilayer composite structure that protects the silkworm from harsh environmental and weather conditions, predator attack and physical damage while supporting silkworm’s metabolic activity. Evolved over many millions of years’ natural selection, the very thin and lightweight cocoon shell performs effective and efficient biological functions to promote the survival chances of silkworm in the wild. In comparison with domestic silkworm such as Bombyx mori (B. mori), wild silkworms need much greater protection from environmental, biotic and physical hazards[1]. For example, some dark cocoons are especially effective in absorbing and trapping heat; some cocoons made for winter tend to be more robust than their summer counterparts; some cocoon species prevent inoculative freezing by isolating the integument form ice crystals on the cocoon surface[2]. A cocoon is a multilayer composite material formed by continuous twin silk filaments bonded by silk sericin. Silk is produced as an aqueous solution in the posterior section of the silkworm’s gland and this solution is extruded through two spinnerets and coated and fastened together with silk sericin into twin filaments during coagulation. A compact cocoon is then formed when a silkworm, along with spinning, wraps the bave around its body through a gyrating motion of its head and cyclically bending and stretching of its body with different shapes in a programmed
manner. Depending upon silkworm species and rearing environment, cocoons vary in weight, thickness, colour and stiffness. Silk fibres have long been used for textiles since more than 4000 years ago. In more recent times, silk-based materials have found growing applications for medical uses such as tissue products, scaffolds and implantable function devices. While the properties of silk fibres and silk proteins have been extensively studied, there are substantial knowledge gaps in understanding how a wild silkworm cocoon provides the essential survival utility for the pupa residing inside.

The mechanical [1, 3] and thermal protection [4, 5] from silkworm cocoons is essential for the survival of silkworms when they enter diapause and metamorphosis. The knowledge of structure-property-function relationship of this important multi-layered biological composite gives insight into new designs of next-generation protection materials and structure. In this work, the mechanical and thermal insulation properties of both domestic (Bombyx mori or B. mori) and wild (Antheraea pernyi and Antheraea mylitta, or A. pernyi and A. mylitta) silkworm cocoons were investigated. The peel tests and needle penetration tests on cocoon walls were conducted for mechanical characterization and the temperature measurements of both the inside and outside of cocoons under windy conditions were conducted for thermal property investigation.

2. Materials and methods

2.1 Materials

B. mori and S. cynthia cocoons were purchased from silk rearing houses in Northeast India; A. pernyi cocoons were collected from Northeast China and A. mylitta cocoons were collected from Central India. They were received as stifled cocoons, commonly used prior to reeling silk filament for textile applications. The aramid fabric used for needle penetration tests was from a TurtleSkin Insider protective glove (Warwick Mills, USA) and has a plain weave texture (240 ends/10 cm warp, 480 picks/10 cm weft).

2.2 Methods

The 180 degree peel tests, modified from the ASTM standard test D 1876-08 for peel resistance of adhesives, were used for testing the peel resistance of cocoon walls. A loading rate of 2 mm/min was applied to delaminate cocoon samples with the dimension of 20 mm × 5 mm. The cocoon specimens were peeled artificially for a length of 5 mm before they were pulled apart by the tester. Three specimens for each type of cocoons were peeled into multiple layers. Needle penetration tests were performed on a 5967 Materials Testing System (Instron Corporation, USA) equipped with a 100 N load cell. A lab-designed test set-up consisted of a base holding the test specimen in position and a puncture probe to which the needle was installed for applying a compression force to the specimen. A tension force of 2.3 N was applied to the specimen by a spring between the two mounting blocks. A loading rate of 10 mm/min was used to test samples with the dimension of 25 mm × 8 mm. Motion image capture was undertaken using a QC3247 USB camera (Digitech Industries, Hong Kong). The morphologies of the mechanically failed cocoons and the fracture surfaces from failed specimens were investigated by a scanning electron microscopy (SEM) (Supra 55VP).

To simulate natural windy conditions, an artificial air flow field was created using an electric fan (DC12V, 0.2A) and a connected duct (a plastic tube with the same diameter as the fan). The cocoons were translocated from ambient environment to the middle of the duct through a hole at the bottom, while this ventilation system was situated in an oven (Binder) with isothermal setting of 45 °C. The electric fan was driven by a power source (Powertech™ MP 3081 DC power supply, 0–30V, 0–3A) and the wind velocity was adjusted by changing the voltage output. Two needle-type temperature probes (ICT SFM) were used to measure the temperature both inside and outside the cocoons.
3. Results and discussion

3.1 Peel resistance of cocoon walls

Fig. 1 shows the comparison between the B. mori and the A. pernyi and A. mylitta cocoon walls, on their peeling test performance. For the B. mori cocoon, the peeling loads were less than 1 N and the load values were comparable for three peeled layers. For both A. pernyi and A. mylitta cocoons, the peeling curves became more rugged with much higher loads (the maximum load values reached 4 – 5 N); the outer layers, i.e. Layers 1 and 1' (In Fig. 1b, layers were numbered from the outer to inner layers) were more difficult to separate. This further indicates that delamination resistance of different B. mori layers was similar from outer to inner cocoon surfaces; however, the delamination resistance of A. pernyi and A. mylitta cocoon layers was stronger in the outer layers. It indicates that the outer layers of these two types of wild cocoons are tougher than the inner layers, which results from the more stringent requirement in the wild to protect the pupa against physical attack from natural predators and harsh environments.

During peeling, the wild cocoons experienced much higher peak loads than the domestic ones. The average peeling load ($P_{average}$) was 0.34 N for the B. mori cocoon, but was 2.5 N and 2.4 N for the A. pernyi and A. mylitta cocoons, respectively. This transfers to a maximum work-of-fracture (WOF) of about 1000 J/m$^2$ for the A. pernyi outer layer and 980 J/m$^2$ for the A. mylitta outer layer, which was almost 10 times of the B. mori cocoon. The cubic crystals deposited on the outer surface/layers were not shown to improve interlaminar fracture toughness, but exhibited extremely high hardness. In contrast to the B. mori and A. mylitta inner layers which had hardness less than 25 MPa and the A. pernyi inner layer which exhibited hardness of 125 MPa, the hardness of the calcium oxalate crystals was around 2 GPa. This extremely high hardness explains why these crystals remained intact after compression tests on those wild cocoon walls.

The fracture surfaces of different cocoon types showed different levels of silk fibre damage after the peel tests. For the B. mori cocoon, slight fibre damage can be observed at the intersections where B. mori silk fibres meet. However, the fracture surfaces of A. pernyi and A. mylitta cocoons showed more severe fibre damage such as fibre splitting. Many silk fibre fibrils were found on the peeled surface, indicating the stronger interlaminar bonding and fibre/matrix adhesion.

![Load displacement curves for peel tests on the a) domestic B. mori cocoon walls and on the b) wild A. pernyi and A. mylitta cocoon walls](image)

3.2 Needle penetration resistance of cocoon walls

The needle penetration loading curves for the cocoon walls B. mori, S. cynthia, A. pernyi and the synthetic aramid protective glove fabric are shown in Fig. 2a. Against the 21G hypodermic needle (Fig. 2b), the A. pernyi cocoon wall showed the highest needle penetration resistance among all tested pieces with a maximum needle penetration load of 11.8 N, followed by the aramid fabric of 7.1 N and the much lower loads between 2 to 5 N for the B. mori and S. cynthia with larger displacements at maximum load than the A.
**pernyi** cocoon and aramid fabric. During penetration of the *A. pernyi*, the resistance of the cocoon structure caused the needle shaft to bend elastically (Fig. 2b), which was not observed in other materials measured. Three main distinctive load peaks (1, 2 and 3) were observed during penetration by the three-facet needle to every sample type. Peak 1 coincides with the needle tip penetration, exposing the tip to the underside of the sample; peak 2 coincides with the end of the cutting induced by the upper part of the beveled edge, reaching the underside of the sample; and peak 3 represents the ejection of the bevel heal from the cutting hole. After that the needle can slide easily into the hole that has been made, with a decrease in the applied force. The failed specimens were examined under SEM. In contrast to the cleanly and sharply-cut fibres from the low resistance specimens, the *A. pernyi* ones showed apparent fibre kinking near the broken ends (at the needle insertion side) and a large amount of fibre debris following the needle perforation, indicating significant cutting in the puncture process.

The role of sericin on the penetration resistance was also studied. The removal of 30% of the sericin matrix resulted in a 28% decrease in needle resistance. With further removal of sericin matrix, the cocoon wall started to lose its integrity and the fibres became loosely packed; as a result, the loading curves became smoother and no longer presented the typical main peaks. The needle easily penetrated the degummed specimens through many thin individual debonded layers after sericin removal. Hence, the sericin plays a critical role of bonding the silk fibres together, restricting the relative motion of fibres, which causes the needle to cut the fibres to enable penetration as opposed to just pushing them aside and penetrating between them.

![Graph showing load-displacement curves](attachment:load_displacement_curve.png)

**Fig. 2.** a. Load-displacement curves of the needle penetration tests on the cocoon walls and the aramid fabric; 1, 2 and 3 on the loading curve of *A. pernyi* correspond with the moments when video images were photographed in Fig 2b.

### 3.3 Thermal insulation of cocoons under windy conditions

![Graph showing static pressure changes](attachment:static_pressure_changes.png)

**Fig. 3.** Static pressure changes with time at specific locations on the *A. pernyi* cocoon outer layers (a1, a2 and a3) and inner layers (b1, b2 and b3), under warm conditions at the wind velocity of 0.7 m/s [5].
A CFD model was built to simulate the heat transfer process through the wild *A. pernyi* cocoon wall in the flow field. Three positions on the inner and outer surfaces were defined. A1 is in the centre of the outer surface and a2 and a3 are 200 µm away from the centre; similarly, b1 is located in the centre of the inner surface and b2 and b3 are 250 µm from the centre. It can be seen that the static pressures at positions b1, b2, and b3 are nearly the same under all conditions; the static pressure from the model with crystals fluctuates, while the static pressure from the model without crystals keeps steady. In the model without crystals, the static pressure on the outer surface of the cocoon decreases along the positive direction of x-axis, which is due to energy loss during flowing caused by the friction between the boundary of the cocoon wall and the fluid (air). The presence of the crystals disrupts the external flow and creates uneven pressures along the surface of the cocoon, which would give rise to transient lateral (as opposed radial) flow patterns. This also contributes to minimizing penetration of outside air into the inside of the cocoon.

4. References


