



Production of Materials with High Thermal Insulation from Natural Fibers and Sericin

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ABSTRACT

Silkworm cocoon is a natural biological and composite structure that has evolved over time and has high physical and mechanical properties against stress and acts as insulation against ambient temperature conditions. Understanding the relationships between the two-component structure of silkworm cocoons (sericin and fibroin) inspires the creation of composite structures, including lightweight, high-strength nonwoven biocomposites. In the present study, by analytical-descriptive method, we have tried to use cocoon sericin and introduce some famous and widely used natural fibers in materials science and study their characteristics - because for various reasons such as lightness, lack of pollution and low cost, etc. can be suitable alternative for a replacement of synthetic fibers - suggest the production of non-woven bio-composite materials. Natural fibers such as jute, hemp, flax, etc. with different volume percentages in combination with sericin as a binder, were proposed for this biocomposite and the thermal performance of each of them was compared using Maxwell's theoretical model. All compounds show low thermal conductivity and jute-sericin biocomposite with 70% by volume and 0.061 W/m²-K performance has better performance.

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NOMENCLATURE

V_F	Volume fraction of fiber	Subscripts	
K_c	Thermal conductivity of the composite	E	Young modules (GPa)
K_f	Thermal conductivity of the fibers	TS	Tensile strength (MPa)
K_m	Thermal conductivity of the matrix	Greek Symbols	
		ρ	Density (g/m ³)

INTRODUCTION

The use of architectural composites in comparison with its widespread use in the marine and aerospace industries is in the early stages. This is largely due to the lack of general engineering standards for buildings or materials made of composite materials and the lack of attention of some architects to the use of new materials in design. In terms of stability, structures created with composite materials, to make proper performance, need fewer materials compared to similar structures, which leads to a simultaneous reduction in energy consumption and carbon emissions during the production process. Advances in composite materials science offer new insights into their properties and applications in buildings.

The biggest drawback of synthetic fibre -reinforced composites is the lack of solutions for their recycling except by burying or burning [1]. Research on environmentally friendly composites made from natural fibers is important due to the interest in environmentally friendly materials and concerns about resource depletion, and nonwoven composites are also part of these bio composites. Nonwoven composites formed from fibres that were stacked without special order and texture and bonded together using adhesive (matrix) by chemical, mechanical or thermal methods. An example of this type of composite in nature is the silkworm cocoon. Silk cocoon is a polymer composite of a single string of silk fibres with a length of about 1000-1500 meters, bonded by sericin and has porous microstructures with a specific

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hierarchy and in a multi-layered way. The fibre-forming protein called fibroin has a semi-crystalline structure and about 75% by weight of cocoon and sericin is an amorphous, water-soluble protein polymer that makes up 25% by weight [2, 3]. Each layer of silkworm cocoons has a different mechanical behaviour and ultimately leads to the structure of irregular cocoons that have a good function against moisture, air flow, UV rays and impact and have a certain degree of heat retardant and are able to save heat.

In view of the above, in this research inspired by the unique two-component structure of silkworm cocoons, the process of producing biocomposites made from natural fibers (due to abundance, recyclability, low weight and low thermal conductivity) and sericin) Is proposed and the use of these materials as thermal insulation is examined by examining their thermal performance in Maxwell's theory.

Research questions:

1. How will the silk cocoon and its structure lead to the production of a new bio composite?
2. What features and benefits make it possible to use natural fibers and sericin in composite fabrication?
3. Does natural fiber-sericin bio composite function properly as a thermal insulator?

LITERATURE REVIEW

In the late 19th and early 20th centuries, materials scientists used advanced techniques such as SEM, DMTA, etc. to use silkworm cocoons as models for bio composites. In 2011, Chen [4] studied the engineering aspects of cocoons as a bio-hybrid system by studying a wide range of types of cocoons and proposed a quantitative model that directly linked cocoon structure to its mechanical properties and could be applied to other composite. Blossman-Myer and Burggren [5] introduced the silkworm cocoon as a heterogeneous fibrous structure in diameter and density, and believed that the cocoon, despite its complex structure, maintained its moisture level and oxygen exchange with the outside environment. In a study in 2019, Kwak et al. [6] studied the natural fiber non-woven composite model according to the structure of silkworm cocoons was made using jute and sericin natural fibres of silkworm cocoons and then, the effect of sericin binder content on the physical properties of nonwoven composite was tested. Korjenic et al. [7] studied natural fibre bio composites as thermal insulation in combination with plant facades in construction, and considered the use of these materials to be environmentally friendly and effective in reducing energy and heat. In their research, Khedari et al. [8] also proposed a composite building using a wall-ceiling, consisting of natural fibres, cement and sand, which is very light and has a low conductivity. Understanding bio composites and their properties can be a new path in architectural design, because building

materials as a constructive element affect the shape and performance of buildings, especially in terms of stability and energy, and one of the most important issues studied in the sources that can provide the use of these materials in building, its structure is as a biological composite that is thermal and moisture insulating.

Silk cocoon

The silk cocoon has a layered structure, without texture and is made of two components of protein, sericin and silk fibres (fibroin). Sericin makes up one-fourth of the total weight of the cocoon, and its amount decreases from the outer layers to the inner layers, eventually leading to the formation of a highly interconnected network of fibres [9-12]. Figure 1 demonstrates part of the layered structure of a silkworm cocoon.

The porous structure of the cocoon and its aerodynamic properties affect the air flow rate. The air velocity is much slower in the smaller cracks in the cocoon wall, and near the surface of the silk fibres, due to the adhesion of sericin. In addition, a number of airflow lines in the cocoon wall are terminated due to the loss of kinetic energy due to friction between the silk fibres and the air and air turbulence through the cocoon. Silkworm cocoons are resistant to temperature changes in the environment and temperature fluctuations inside the cocoon are low if the temperature in the environment changes. The silk cocoon provides a UV protective layer for the worm pupa, which sericin is responsible for absorbing [9-12].

Sericin also has important biological properties such as corrosion resistance, antimicrobial activity, easy absorption of moisture, etc., and is known as a compatible and biodegradable substance. In the textile industry, sericin is a byproduct of silk fiber production and is always looking for new applications of this protein in many research projects. Due to its amorphous nature, it dissolves easily in water at 60°C and returns to a gel state upon cooling, providing significant commercial, economic, and environmental benefits if recycled [3, 13, 14]. Sericin plays a natural binding role in the biocomposite structure of the silk cocoon and plays a significant role in its structure, so it can be used as a suitable alternative to synthetic binders in the production of natural fiber composites.

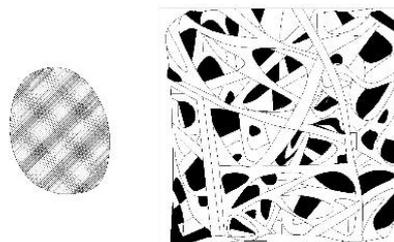


Figure 1. Illustration part of the porous and non-woven structure of a silk cocoon

According to research, formaldehyde-based synthetic binders have important disadvantages such as water absorption, which can greatly reduce the life of the composite material, and more energy is consumed due to the temperature of the process required for the polymerization of the synthetic binder [15-17]. In addition, formaldehyde-based chemical binders as toxic substances can cause side effects [15]. The use of sericin as an adhesive and fiber binder in bio composites will reduce production costs, reduce pollution and energy consumption.

Natural fibers

Natural fibers are materials derived from nature, their use has expanded over the years. The use of natural fibers has long been used in the manufacture of building materials and today, with increasing environmental concerns, has been used in many studies in the preparation of bio composites. Natural fibers have several advantages that have led to interest in using them as an alternative to synthetic fibers, which have been used in industry for a long time. For this purpose, in this study, natural fibers in the two-component structure of bio composite have been used.

The natural fibers/synthetic fibers have been used as substitutes/ reinforces in composites since 1950. Some of the characteristics of natural fibers are: low density, lightness, cost-effectiveness, renewability and recyclability, mechanical properties and optimum strength, suitable thermal properties, easy access to it in

many regions and countries and compared to fibers synthetic non-carcinogens reduce carbon emissions and are naturally biodegradable [18].

To identify and select natural fibers, it is necessary to first examine them based on their design needs and compare their strengths and weaknesses. Table 1 summarized some natural fibers with emphasis on their properties that are required in the development of lightweight materials for construction and non-construction applications.

According to Table 1 and the comparison of natural fibers mentioned in it, it can be seen that jute fibers usually resist rot and heat well, have a lower thermal conductivity than other fibers and have a high tensile strength after flax and hemp fibers. Flax fibers have high tensile strength and low moisture absorption. Coir fibers are less dense than all other fibers and can stay afloat for a long time. All of these fibers are used today as fillers or reinforcements for composite materials.

MATERIAL AND METHODS

Non-woven natural fiber composite model

In this research, a model of a non-woven composite using natural fibers as fiber and sericin binder as a matrix is proposed, the structure of which will be described based on a leading process.

The amount of sericin, depending on the amount in the silkworm cocoon, can be used in the production of natural fibrous bio composites up to 30% by volume, and the rest

Table 1. Comparison of properties of natural fibers with each other as fibers used in non-woven fiber composite

Fibre	ρ (g/m ³)	E (GPa)	TS (MPa)	Moisture absorption (%)	Elongation at break point (Longitudinal strain)	Thermal conductivity (W/m ² -k)*	Medium thickness of fibers (μ mm)	Ref.
Jute	1.3-1.4	10-30	393-800	12-13	1.5-1.8	0.0580	94.2	[7, 18-23]
Flax	1.4-1.5	27.6-46.9	350-1500	7	1.2-3.2	0.0650	111.1	[18-22, 24]
Hemp	1.48	17-70	550-900	8	1.6	0.0620	155.2	[7, 19, 22]
Sisal	1.3-1.5	9-38	300-855	10-11	2-7	NA**	50-200	[18, 19, 21-25]
Coir	1.1	3-6	95-100	8	47	NA	NA	[18, 19, 21-24, 26]
Cotton	1.5-1.6	5.5-13	287-597	NA	3-10	0.0815	NA	[20-22, 27]

* At a temperature of 23 ° C and a relative humidity of 50%

** Not available

should be made of natural fibres. The production process of the proposed materials, which must be done in the workshop and is possible in a prefabricated way, begins after selecting the natural fibers and preparing them and cutting those fibers to lengths of 1 cm. The natural fibers are poured into a container and then sericin solution is added. The solution should then be stirred gently to achieve a homogeneous dispersion of the fibers in the solution. The prepared material is then poured into a metal mold according to the desired standard sizes. To remove moisture, create compression between the fibers and produce the final product, the mixture should be placed inside a hot press machine at a temperature of 120°C for ten minutes and the composite panel should be produced under high pressure and temperature.

Figure 2 demonstrates the method of producing non-woven composite from natural fibers and sericin.

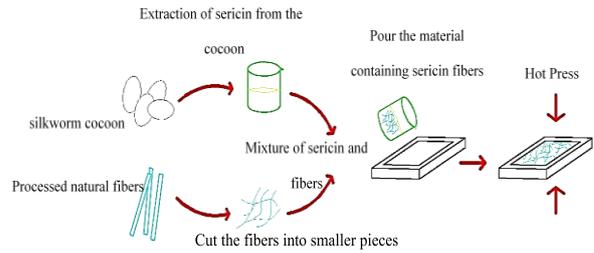


Figure 2. Production method of non-woven composite of natural fibres and sericin

Natural fiber- sericin bio composite insulation performance

Layering and non-weaving, bonding of fibers through sericin leads to a porous structure in these composite materials. For this reason, it is expected to act as a buffer and insulation against heat. The use of thermal insulation materials is done in order to reduce heat transfer. The thermal insulation performance of homogeneous materials is usually evaluated through thermal conductivity and heat transfer. The coefficient of thermal conductivity, the steady state, defines the amount of heat flow that passes through the surface unit of a homogeneous material with a thickness of 1 m and creates a temperature difference of 1 K on that surface and is expressed in W/m²-K.

The thermal conductivity of fiber nonwoven bio composite as the proposed insulation materials was calculated approximately based on Maxwell's theoretical model as stated in Equation (1) and the proposed thickness was considered to be 5 cm.

$$K_c = \frac{K_m(K_f + 2K_m - 2V_f(K_m - K_f))}{K_f + 2K_m + V_f(K_m - K_f)} \quad (1)$$

In this regard, V_F is the volume fraction of fiber. K_c, K_f and K_m are the thermal conductivity of the composite, the fibers and the matrix, respectively. Based on the researches, it was observed that in measuring the thermal conductivity of fiber composite, the experimental results and the results of Maxwell's theoretical model in high fiber content with error rate of 4.8 are close to each other and the calculated [28].

Table 2 shows the calculated values of thermal conductivity of nonwoven natural fiber composite-sericin based on Maxwell's theoretical equation.

By calculating the thermal conductivity of materials and in comparison with hemp and flax, coir and cotton fibers in combination with sericin, jute-sericin bio comp-

Table 2. Comparison of thermal insulation materials in terms of thermal conductivity

Natural fibers-sericin biocomposite	Thermal conductivity (W/m ² -K) calculated based on Maxwell's theoretical equation		
	Natural fiber biocomposite with a volume percentage of 90	Natural fiber biocomposite with a volume percentage of 80	Natural fiber biocomposite with a volume percentage of 70
Jute - Sericin Biocomposite	0.069	0.065	0.061
Flax - Sericin Biocomposite	0.072	0.071	0.066
Hemp - Sericin Biocomposite	0.074	0.072	0.065
Coir - Sericin Biocomposite	0.075	0.072	0.068
Cotton - Sericin Biocomposite	0.086	0.085	0.082

osite has the lowest amount of thermal conductivity and can have a more desirable performance as insulation. Also, with increasing sericin in the bio composite structure and filling the pores between the fibers, the porosity decreases and the thermal conductivity increases. It is important to note that by reducing the amount of sericin in the bio composite structure of natural fibers, the bond between the fibers is reduced and the strength of bio composite materials is reduced. After jute fibers in the bio composite composition, hemp, flax and coir, respectively, and then cotton are acceptable for insulation performance. All the mentioned natural fibers are environmentally friendly and can be degraded and returned to nature. In the meantime, jute is a natural plant fiber and polymer that can be planted and harvested annually in hot and humid climates, including Iran, and even in low-fertility fields that are no longer suitable for other cereal crops. In addition, jute can be easily grown without pesticides and chemical fertilizers and is also known as golden fiber because of its benefits. Because

jute has universal consumption, high production potential and various uses, it has the second rank among plant fibers after flax. The binder of this bio composite, namely silk sericin, is also completely natural, and the recycling and exploitation of sericin from the silk yarn and fabric industry will bring economic and environmental benefits. In the process of silk production, a significant amount of sericin is separated from the silkworm cocoon and today, with extensive research on the properties of sericin and its application in many sciences, including cosmetics, health, therapy, materials production, etc., its use is welcomed by scientists and Engineers in many of these areas. Non-woven bio composite of natural fibers-sericin, is a lightweight material, resistant to moisture and heat and has good strength against incoming loads. It is a component of green materials and is economically suitable for production and use. Nonwoven bio composites due to their suitable mechanical and thermal behavior, can be used in various applications in construction. Further studies on these nonwoven biocomposites, due to their importance in materials science, conducting experiments related to them and their production, will be an important step in achieving environmental sustainability and reducing energy consumption.

CONCLUSION

By introducing suitable materials from renewable natural raw materials as natural fibers and using sericin silkworm cocoons, non-woven biocomposite production was achieved. This biocomposite is recyclable due to natural raw materials and is considered environmentally friendly. Its production process requires low energy and can be prefabricated with standard control. The porous structure of fiber-sericin biocomposite, due to its non-woven nature, can provide thermal insulation function. This theory, by calculating the thermal conductivity of these materials, was theoretically tested using Maxwell's theoretical model. The thermal conductivity of some natural fibers such as jute, hemp, coir, flax and cotton with different volume percentages (70-80 and 90) was calculated in the proposed model of nonwoven fiber biocomposite. Among the introduced natural fibers, jute fibers in combination with sericin pass less heat and jute-sericin biocomposite with a volume percentage of 70 will have a thermal conductivity of 0.061 (W/m²-K). Also, according to the obtained results, with increasing sericin in the biocomposite structure and filling the pores between the fibers, the porosity decreases and the thermal conductivity of natural fiber biocomposite increases.

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**Persian Abstract****چکیده**

پایله کرم ابریشم یک ساختار بیولوژیکی و کامپوزیتی طبیعی است که در طول زمان تکامل یافته و دارای خواص فیزیکی و مکانیکی بالا در برابر تنش می باشد و به عنوان عایق در برابر شرایط دمایی محیط عمل می کند. درک روابط ساختار دوجزئی پایله کرم ابریشم (سریسین و فیبروئین) الیام بخش ایجاد ساختارهای کامپوزیتی، از جمله کامپوزیت های زیستی نفاخته کم وزن و با استحکام بالا است. در پژوهش حاضر، با روش تحلیلی- توصیفی، سعی شده است تا با استفاده از سریسین پایله و معرفی برخی الیاف طبیعی مشهور و پر کاربرد در علم مواد و بررسی مشخصات آن ها- که به دلایل مختلف از جمله سبکی، عدم آلودگی، فراوانی و هزینه کم و غیره می تواند جایگزین مناسبی برای الیاف مصنوعی باشند- تولید مواد کامپوزیتی زیستی نفاخته پیشنهاد شود. الیاف طبیعی از جمله جوت، کنف، کتان و غیره با درصد های حجمی متفاوت در ترکیب با سریسین به عنوان اتصال دهنده، برای این بیوکامپوزیت پیشنهاد و عملکرد حرارتی هر یک از آن ها با استفاده از مدل نظری ماکسول مورد مقایسه و بررسی قرار گرفت. تمامی ترکیب ها ضریب هدایت حرارتی پایینی را نشان می دهند و بیوکامپوزیت جوت- سریسین با درصد حجمی ۷۰ و ضریب هدایتی $0.061 \text{ (W/m}^2\text{-K)}$ عملکرد بهتری را دارد.