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(54) Title: NON-WOVEN SILK MATERIALS AND METHODS FOR MANUFACTURE THEREOF

(57) Abstract: A method for the manufacture of a silk non-woven silk material (240) is described. The method comprising steaming whole ones of the one or more silk cocoons (230), slitting open the one or more silk cocoons, placing the slit silk cocoons (230) on a heated surface (210, 220) and then compressing the slit silk cocoons on the heated surface at 100-130°C. Applications of the non-woven silk material of this disclosure include for the manufacture of vehicular tyres and panels.

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Description

Title: Non-Woven Silk Materials and Methods for Manufacture Thereof

Cross-Reference to Related Applications

[0001] This application claims priority to and benefit of UK Patent Application No. 1110176.3 filed on 16 June 2011.

Field of the Invention

[0002] The field of the invention relates to a non-woven silk material, a method for manufacture thereof and a use of the non-woven silk materials. Examples of such uses include, but are not limited to, insoles for shoes and vehicular tyres.

Background of the invention

[0003] Silk is a natural protein fibre that has been used for many thousands of years for weaving into textiles. The best-known silk fibre is obtained from cocoons of the larvae of a mulberry silkworm *Bombyx mori* that has have been reared in captivity for many thousands of years. There are other types of silkworms that show potential for commercial use of the silk fibre. These silkworms include silk-producing larvae of the bombycid, saturnid, lasiocampid and thaumatopoid species. The saturnid species of wild silkworms by way of example only include Antheraea pernyi, Antheraea yamamai, Antheraea mylitta, Antheraea assama, Philosamia cynthia ricime and Philosamia cynthia pryeri and are currently used for commercial silk production. Other saturnidae as well as lasiocampid silkworms, for example of the genus Gonometa or Anaphe also have great potential for the commercial production of the silk fibres.

[0004] To date the silk fibres are generally obtained commercially by direct reeling from a cocoon or, less commonly, by carding cocoons. For example, a Chinese utility model No. CN 2811337 Y (assigned to Yu Qingcun) entitled “Silk worm cocoon dressing machine” describes a machine that can be used to unravel the silk fibre from the cocoon of the silk

moths. The machine of the CN '337 utility model is used to unravel so-called "superlong" silk fibres and to provide uniform distribution of the unravelled silk fibres.

[0005] One of the uses described in the past for the silk fibres is the incorporation of the silk fibres into rubber articles, such as vehicular tyres, in order to re-enforce the rubber articles. For example, UK patent application No. GB 178 898 (John Brown) describes a formation of combined rubber and fabric materials that is adapted for strong wear resistance. The GB '898 patent describes a method in which individual ones of the silk fibres are incorporated into a laminate of rubber and the silk thread. The GB '898 patent fails, however, to describe the use of a non-woven silk material.

[0006] Chinese patent application CN 101481850 (University Zhejiang Science and Technology) teaches a method for the manufacture of a non-woven fabric in which oval silk cocoons are cut open along the direction of the major axis and the chysalis is removed from inside of the silk cocoons. The top three to ten layers of the cocoon shell are removed and laid down on a substrate. The removed cocoon layers are humidified and hot and compressed at a temperature of 70 – 100 °C to form the non-woven fabric.

[0007] Japanese patent application JP 05186904 (Ishihara) teaches a method of manufacturing a so-called "plate-like cocoon" that involves the cutting of the cocoon and immersing the opened cocoon in water of 10 – 20 °C for between 5 and 600 seconds. The softened cocoon is then pressed onto a smooth hot metal plate to obtain the plate-like cocoon. A textile can be produced by overlapping the edges of the plate-like cocoons and sewing the overlapping part with a silk yarn or by successively bonding the plate-like cocoons onto a cloth.

Summary of the invention

[0008] This disclosure describes a method for the manufacturing of a non-woven silk material from one or more silk cocoons that comprises the following steps. One or more of whole silk cocoons or pieces of silk cocoons, including individual fibres, cut or chopped or torn from silk cocoons are either taken as a whole and flattened or they split open, for example, by cutting along one side. The compressed or cut silk cocoons are placed on a

heated surface and compressed on the heated surface to bond the cocoons together into a layer of non-woven silk material (in the form of a mat or a moulded article). Spray or steam of water or another solvent can be added before or during the compression heating to better bond by partially melting the sericin gum layers of the cocoon. In one aspect of the disclosure, the heated plate is heated to a temperature of between 100 °C to 130 °C and water is sprayed on to the non-woven silk material before or during the compression pressing, but this is not limiting of the invention.

[0009] In another aspect of the invention it is possible to make a laminate of two or more layers of the non-woven silk materials by compressing two or more layers of the non-woven silk materials in order to obtain the silk laminate. It will be understood that additional materials, by way of example only, steel thread, can be laid between one or more of the two or more layers of non-woven silk material before the compressing.

[0010] The non-woven silk material manufactured by the method of this disclosure can be used, for example, for the manufacture of a component for a vehicle, such as but not limited to a vehicular tyre, fender panels, or doors, and also for in-soles of a shoe, but are not limited to such applications.

Description of the Figures

[0011] Fig. 1 shows a flow diagram of a method for the manufacture of a non-woven silk material according to one aspect of the invention.

[0012] Fig. 2 shows an overview diagram of an apparatus for the manufacture of the invention.

[0013] Fig. 3 shows a photograph of an example of a non-woven silk material

[0014] Fig. 4 shows a photograph of one product produced from the non-woven silk material of the invention.

Detailed description of the invention

[0015] The invention will now be described on the basis of the drawings. It will be understood that the embodiments and aspects of the invention described herein are only examples and do not limit the protective scope of the claims in any way. The invention is defined by the claims and their equivalents. It will be understood that features of one aspect or embodiment of the invention can be combined with a feature of a different aspect or aspects and/or embodiments of the invention. The invention allows for the manufacture of silk materials without previously boiling the silk cocoons to allow unravelling of silk fibres. It will be understood that the teachings of the disclosure enable an ecologically sustainable method, as the method for the manufacture of a non-woven silk material reduces the use of water.

[0016] When distinguishing between cocoons then the term "mulberry silkworm" is used in this disclosure for the larvae of the Lepidopteran Bombyx mori and the term "wild silkworm" is used for the larvae of moths of the families Bombycoidea (other than B. mori), Saturniidae, Lasiocanpidae. Mimallonidae, Geometridae, Rhopalocera and Noctuoidea which produce large larvae cocoons.

[0017] Fig. 1 shows an overview of a method for the manufacture of a non-woven silk material according to one aspect of the invention. Cocoons in this aspect of the invention are cocoons of the mulberry silkworm. It will be appreciated that the cocoons may also be from wild silkworms and in either case the larva will either have emerged or been killed by the methods known to a person skilled in the art. In a first step 110, the cocoon is either compressed as a unit if there is no larva or cut open by slitting along one or both sides. The larva is then removed from the cocoon through the slit. The slit cocoon can be placed in steam from a steam generator in step 120 that heats up the cocoon to soften sericin gum in the cocoon. It would also be possible to use an iron to heat up the cocoon to soften the sericin gum. In step 130, the cocoon is placed in a heated press together with other ones of the silk cocoons and compressed to produce a layer of the non-woven silk material. The heated press may also include a mould to form the non-woven silk material. The heated press may also include a device to enable steam to be introduced between plates of the

heated press. The layer of the non-woven silk material can be used to produce textiles and laminates in step 140.

[0018] Fig. 2 shows an example of the heated press 200 in one aspect of the invention. The heated press consists of a top plate 210 and a bottom plate 220. The top plate 210 and the bottom plate 220 are initially placed apart from each other and the one or more cocoons 230 are placed between the top plate 210 and the bottom plate 220. The top plate 210 and the bottom plate 220 are generally made of aluminium, but this is not limiting of the invention and other metals, such as various types of steel, or indeed temperature tolerant polymers may be considered. The heated press may also include a device to enable steam to be introduced between the top plate 210 and the bottom plate 220 of the heated press 200.

[0019] The one or more cocoons 230 are used in their whole form, except that, as noted above, any larvae are removed from the cocoon. The whole cocoon is in itself a complex composite structure with a range of important properties, as discussed in Chen et al., "Silk Cocoon: Multilayer Structure and Mechanical Properties", Acta Biomaterialia, 8 (7) 2620-2627 and also in Chen et al. "Morphology and Structure of Silkworm Cocoons, Materials, Science and Engineering" C -Mmaterials for Biological Applications, 32 (4) 772-778, "a Non-Woven Composite Model based on Silkworm Cocoon," Journal of Materials, Science and Engineering, 4 (9), 28-33 and "Silkworm Cocoons expire Models for Random Fibre and Particular Composites", Physical Review E 82 (4), 041911-041917. The use of the whole cocoon 230 will therefore retain all of these properties, rather than destroy one or more of the properties. This will therefore enable extra strength and toughness to be given to the non-woven silk material made according to the teachings of this disclosure. The more sericinaceous outer layers of the whole cocoon are known to have good adhesion properties, which can be used to fix the cocoon in place. Furthermore using the whole of the cocoon adds more bulk to the material.

[0020] Once the cocoons 230 are placed on the top side of the bottom plate 220 the top plate 210 and/or the bottom plate 220 are heated to a temperature of approximately 100 to 130 °C. A light spray or mist of water or another solvent can be applied via a spritzer before heating. The bottom side of the top plate 210 and the top side of the bottom plate

220 are than moved towards each other as in step 130, thus flattening or compressing the cocoons 230 into a layer of non-woven silk material 240. An example of the non-woven silk material 240 is shown in Fig. 3.

[0021] The heating of the top plate 210 and/or the bottom plate 220 allows sericin within the cocoons 230 to flow. It will be noted, however, that the sericin is not heated to more 135 °C, as this will start to degrade the non-woven silk material.

[0022] It has been found that the use of temperature between approximately 100 – 130 °C enables the water vapour in gaseous form (steam) to be effective in softening sericin in the whole cocoon 230 in order to get the sericin to flow and to rebond with the compressed cocoon. It is known that the glass transition of the sericin in dry form and unwetted will be about 155 – 180 °C. The glass temperature reduces with water content and therefore some water vapour is useful. The selection of a temperature range between 100 °C and 130 °C appears to be effective.

[0023] The top plate 210 and the bottom plate 220 are pulled apart from each other and the layer of the non-woven silk material 240 of the cocoons 230 can than be removed. The non-woven silk material 240 is sprayed with water in step 40 to cool it down. In an alternative method of cooling, the non-woven silk material 240 is conveyed onto a cold plate or merely placed in air.

[0024] It will be appreciated that several layers of the non-woven silk material 240 can be laminated together by using the press 200 and laying several layers of the non-woven silk material 240 on top of each other and then compressing. It is also possible to insert other materials, for example agents to enhance bonding, metals or other textiles, between the several layers of the non-woven silk material 240. It will also be possible to infuse the material with chemicals, such as additives. One non-limiting example is a neem-tree extract having anti-microbial properties.

[0025] The step 40 of spraying the non-woven silk material 240 could be replaced by spraying with another solvent, in particular a polar solvent.

[0026] The non-woven silk material 240 of the disclosure can be used in numerous applications. For example, it is possible to use the non-woven silk material to produce an insole for a shoe or an inner liner of a bicycle or other vehicular tyre or molded kneepads or lining for brassieres. For example, it is also possible to create large sheets into mats for exercise or mattresses. The material can also be used for panels requiring a great deal of mechanical stability, such as those used on vehicle fenders and doors.

[0027] These uses are merely exemplary and other uses can be contemplated which take advantage of the properties.

Reference Numerals

200	Heated Press
210	Top Plate
220	Bottom Plate
230	Cocoons
240	Non-woven silk material

Claims

1. A method for the manufacture of a non-woven silk material (240) from one of more silk cocoons (230) comprising:
- compressing the whole of one or more silk cocoons (230) on a heated plate (210, 220).

2. The method of claim 1, further comprising the heating of the heated plate (210, 220) to a temperature between 100°C to 130 °C.

3. The method of claim 1 or 2 wherein the heated plate (210, 220) is in the form of a shaped mould.

4. The method of any of the above claims, further comprising :
- placing two or more layers of the non-woven silk material (240) on top of each other and on a heated surface; and
- compressing the two or more layers of the non-woven silk material (240) to obtain a silk laminate.

5. The method of any one of the above claims, further comprising placing a layer of a further material between at least two layers of the non-woven silk material (240) prior to the compression.

6. The method of any one of the above claims, further comprising applying at least one of a light spray or mist water or a solvent to the silk cocoons (230) prior to compression.

7. The method of any one of the above claims, further comprising slitting open the one or more silk cocoons (230) prior to compression.

8. The method of any one of the above claims, further comprising application of steam during the compression.

9. Use of a non-woven silk material (240) manufactured by the method of any one of claims 1 to 8 for at least one of a part of a tyre, a felted slipper or a shoe.

10. At least part of a component for a vehicle comprising the non-woven silk material (240) manufactured according to any one of claims 1 to 8.

11. An insole or sole for a shoe or the whole of a felted slipper or shoe comprising the non-woven silk material (240) manufactured according to any one of claims 1 to 8.

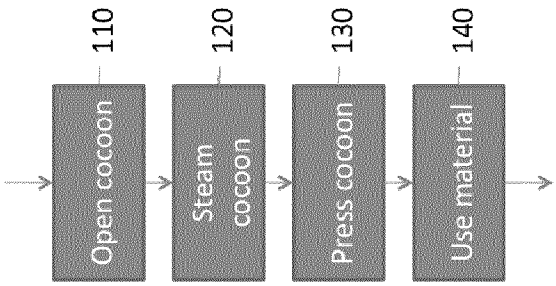

110
120
130
140
Open cocoon
Steam cocoon
Press cocoon
Use material

Fig. 1

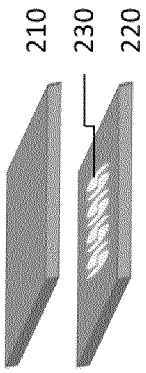


Fig. 2



Fig. 3



Fig. 4