METHOD OF PRODUCING BACTERIAL CELLULOSE-BASED BIOLEATHER FROM WASTE VALORIZATION

FIELD OF INVENTION

The present invention relates to a formulation for producing a bacterial cellulose-based leather. The present invention also relates to a bacterial cellulose-based leather and a method of producing thereof.

BACKGROUND

The listing or discussion of a prior-published document in this specification should not necessarily be taken as an acknowledgement that the document is part of the state of the art or is common general knowledge.

Traditionally, leather is made from animal hides which are by-products of the meat industry. However, there are increasing concerns from the public over animal cruelty from common farm practices. Coupled with the large carbon footprint during the rearing of animals and the environmental unfriendly leather making processes, there have been increasing demands for alternative leather. Synthetic leathers are gaining popularity. Two common synthetic leather options are made from polyvinyl chloride (PVC) and polyurethane (PU) but there are also health and environmental concerns associated with their production and use.

There have been promising commercialized vegan leathers made with pineapple agricultural waste and PU, or with cactus and PU, or with grape waste and PU. However, they still present with the same problems with normal PU leather, such as poor performance, low resistance to wear and tear and low resilience with high usage.

The usage of PU/PVC in such alternative leather often serve as a mechanical support. However, the use of petroleum-based products is not environmentally friendly as these parts do not decompose. This effect is multiplied when rapid degradation of such materials results in frequent replacements. This is detrimental when the replacement volume outpaces the biodegradation rate.

Therefore, there is a demand for new, greener, animal-free alternative leathers with improved mechanical properties.

SUMMARY

It has been surprisingly found that some or all of the problems identified above can be solved through the use of a bacterial cellulose in a formulation that can produce an alternative leather made from non-animal sources. This will now be described by reference to the following numbered clauses relating to aspects and embodiments of the current invention.

1. A formulation for producing a bacterial cellulose-based leather comprising:
   (a) bacterial cellulose;
   (b) a gelling agent;
   (c) a plasticizer;
   (d) a crosslinker; and
   (e) a filler.

2. The formulation according to clause 1, wherein the formulation further comprises reinforcement fibres, a preservative and/or an antimicrobial agent.

3. The formulation according to clause 1 or clause 2, wherein the bacterial cellulose is obtained from fermentation using a Symbiotic Culture of Bacteria and Yeast (SCOBY).

4. The formulation according to any of the preceding clauses, wherein the gelling agent is selected from one or more of a group consisting of gelatin, agar, pectin, Xanthan gum, guar gum, an alginate, starch and cornstarch, optionally wherein, the gelling agent is selected from one or more of a group consisting of gelatin, agar and pectin.

5. The formulation according to any of the preceding clauses, wherein the gelling agent is present in the formulation in an amount of from 15 wt% to 50 wt% with respect to the dry mass of the bacterial cellulose, optionally wherein the gelling agent is present in an amount of from 20 wt% to 44 wt% with respect to the dry mass of the bacterial cellulose.

6. The formulation according to any of the preceding clauses, wherein the plasticizer is selected from one or more of a group consisting of glycerol, polyvinyl alcohol and polyvinyl acetate, optionally wherein the plasticizer is glycerol.

7. The formulation according to any of the preceding clauses, wherein the plasticizer is present in the formulation in an amount of from 30 wt% to 55 wt% with respect to the dry mass

of the bacterial cellulose, optionally wherein the plasticizer is present in an amount of from 40 wt% to 50 wt% with respect to the dry mass of the bacterial cellulose.

8. The formulation according to any of the preceding clauses, wherein the crosslinker is selected from one or more of a group consisting of a diacid (such as adipic acid), a diamine (such as ethylenediamine, butanediamine and hexanediamine), glutamic acid, boric acid, and more particularly, citric acid, azelaic acid, a diacrylate, a diisocyanate, a diepoxide, formaldehyde and glutaraldehyde, optionally wherein the crosslinker is citric acid.

9. The formulation according to any of the preceding clauses, wherein the crosslinker is present in the formulation in an amount of from 2 wt% to 20 wt% with respect to the dry mass of the bacterial cellulose, optionally wherein the crosslinker is present in an amount of from 2.5 wt% to 10 wt% with respect to the dry mass of the bacterial cellulose.

10. The formulation according to any of the preceding clauses, wherein the filler is selected from one or more of a group consisting of a polyphenol (such as catechin, caffeic acid and gallic acid) and more particularly, tannic acid, optionally wherein the filler is tannic acid.

11. The formulation according to any of the preceding clauses, wherein the filler is present in the formulation in an amount of from 20 wt% to 65 wt% with respect to the dry mass of the bacterial cellulose, optionally wherein the filler is present in an amount of from 20 wt% to 60 wt% with respect to the dry mass of the bacterial cellulose.

12. The formulation according to any of the preceding clauses, wherein the bacterial cellulose is in the form of a slurry.

13. The formulation according to clause 12, wherein the bacterial cellulose is present in the slurry in an amount of from 2 wt% to 5 wt%, the remaining being water.

14. The formulation according to clause 3, wherein the bacterial cellulose is obtained from the fermentation of a fruit/vegetable juice media with a Brix value of from 7°Bx to 13°Bx.

15. The formulation according to any of clauses 2 to 14, wherein the reinforcement fibres are selected from one or more of a group consisting of fruit fibres and vegetable waste, optionally wherein the reinforcement fibres are processed pineapple leaf fibres.

16. The formulation according to any of clauses 2 to 15, wherein the preservative is selected from one or more of a group consisting of polylysine, sodium benzoate, parabens, natamycin, benzoic acid, sorbic acid and ascorbic acid.

17. The formulation according to any of the preceding clauses, wherein the formulation further comprises ethanol and water.

18. A method of producing a bacterial cellulose-based leather comprising the following steps:

(i) providing a formulation according to any of clauses 1 to 17; and

(ii) pouring the formulation into a mould and casting the formulation at a temperature of from 70°C to 90°C for a period of time to obtain the bacterial cellulose-based leather.

19. The method according to clause 18, wherein the formulation according to any of clauses 1 to 17 is obtained by mixing a bacterial cellulose slurry with a gelling agent, a plasticizer, a crosslinker and a filler, as described in any one of clauses 1 to 17, to provide a mixture and blending the mixture to obtain the formulation, optionally wherein the bacterial cellulose slurry is obtained by providing a bacterial cellulose obtained from fermentation using SCOBY and subjecting the bacterial cellulose to blending, rinsing and straining.

20. The method according to clause 18 or clause 19, further comprising one or both of the following steps:

(ii)(a) degassing the formulation prior to step (ii) of clause 18; and

(ii)(b) subjecting the bacterial cellulose-based leather to a hotpressing step after step (ii) of clause 18.

21. The method according to any of clauses 18 to 20, further comprising:

(iii) subjecting the bacterial cellulose-based leather to a post-treatment step, wherein the post-treatment step comprises one or more of the following:

(iii)(a) applying one or both of a varnish and a waterproofing layer on a surface of the bacterial cellulose-based leather;

(iii)(b) dyeing the bacterial cellulose-based leather; and

(iii)(c) adding a supporting base layer to the bacterial cellulose-based leather, the supporting base layer is selected from the group consisting of polyester, nylon, cotton fabric and pineapple skin textile, optionally wherein the base layer is polyester.

22. The method according to any one of clause 19 and clauses 20 and 21, as dependent upon clause 19, wherein one or both of the following apply:

(a) the fermentation step in clause 19 comprises fermenting a fruit/vegetable juice media with a Brix value of from 7°Bx to 13°Bx using SCOBY; and

(b) the period of time in clause 18(ii) is from 18 to 24 hours.

23. The method according to any one of clause 19 and clauses 20 to 22, as dependent upon clause 19, wherein the blending step comprises blending the bacterial cellulose with a 5 vol% to 10 vol% bleach.

24. The method according to any one of clause 19 and clauses 20 to 23, as dependent upon clause 19, wherein the gelling agent is agar, optionally wherein the gelling agent is agar predissolved in water.

25. The method according to any one of clause 19 and clauses 20 to 24, as dependent upon clause 19, wherein the blending step comprises blending the mixture with a wet miller.

26. The method according to any one of clause 20 and clauses 21 to 25, as dependent upon clause 20, wherein step (ii)(a) comprises degassing the formulation in a vacuum oven for about 5 mins.

27. The method according to any one of clause 20 and clauses 21 to 26, as dependent upon clause 20, wherein step (ii)(b) comprises subjecting the bacterial cellulose-based leather to a hotpressing step at a temperature of about 80°C and at a pressure of about 5 bar for about 5 mins.

28. A bacterial cellulose-based leather comprising:

(a) bacterial cellulose;

(b) from 15 wt% to 50 wt% gelling agent;

(c) from 30 wt% to 55 wt% plasticizer;

(d) from 2 wt% to 20 wt% crosslinker; and

(e) from 20 wt% to 65 wt% filler,

wherein the wt% of each of (b) to (e) is with respect to the dry mass of the bacterial cellulose, and

wherein one or both of the following apply:

(i) the bacterial-cellulose leather has a Young's Modulus of from 10 MPa to 300 MPa; and

(ii) the bacterial-cellulose leather has an Ultimate Tensile Strength of from 5 MPa to 35 MPa.

29. The bacterial cellulose-based leather according to clause 28, wherein the bacterial cellulose-based leather comprises:

(a) bacterial cellulose;

(b) from 20 wt% to 44 wt% gelling agent;

(c) from 40 wt% to 50 wt% plasticizer;

(d) from 2.5 wt% to 10 wt% crosslinker; and

(e) from 20 wt% to 60 wt% filler.

wherein the wt% of each of (b) to (e) is with respect to the dry mass of the bacterial cellulose.

30. The bacterial cellulose-based leather according to clause 28 or clause 29, wherein the gelling agent is selected from one or more of a group consisting of gelatin, agar, pectin, Xanthan gum, guar gum, an alginate, starch and cornstarch, optionally wherein, the gelling agent is selected from one or more of a group consisting of gelatin, agar and pectin.

31. The bacterial cellulose-based leather according to any of clauses 28 to 30, wherein the plasticizer is selected from one or more of a group consisting of glycerol, polyvinyl alcohol and polyvinyl acetate, optionally wherein the plasticizer is glycerol.

32. The bacterial cellulose-based leather according to any of clauses 28 to 31, wherein the crosslinker is selected from one or more of a group consisting of a diacid (such as adipic acid), a diamine (such as ethylenediamine, butanediamine and hexanediamine), glutamic acid, boric acid, and more particularly, citric acid, azelaic acid, a diacrylate, a diisocyanate, a diepoxide, formaldehyde and glutaraldehyde, optionally wherein the crosslinker is citric acid.

33. The bacterial cellulose-based leather according to any of clauses 28 to 32, wherein the filler is selected from one or more of a group consisting of a polyphenol (such as catechin, caffeic acid and gallic acid) and more particularly, tannic acid, optionally wherein the filler is tannic acid.

34. The bacterial cellulose-based leather according to any of clauses 28 to 33, wherein the bacterial cellulose-based leather further comprises reinforcement fibres, a preservative and/or an antimicrobial agent.

35. The bacterial cellulose-based leather according to any of clauses 28 to 34, wherein the bacterial cellulose-based leather further comprises one or more of the following:
(a) a varnish;
(b) a waterproofing layer;
(c) a dye; and
(d) a supporting base layer to the bacterial cellulose-based leather, the supporting base layer is selected from the group consisting of polyester, nylon, cotton fabric and pineapple skin textile, optionally wherein the base layer is polyester.

BRIEF DESCRIPTION OF DRAWINGS

FIG. 1 includes photographs of A) fruit juice media; B) bacterial cellulose grown from mixed fruit juice; C) blended bacterial cellulose slurry according to Example 1 of the present disclosure.

FIG. 2 includes photographs of the casted bioleather using A) a formulation with no ethanol; B) a formulation with ethanol according to Example 2 of the present disclosure.

FIG. 3A-D include photographs of finished leather pieces that were post treated by dyeing (leather dyes) and varnish (resolene) according to Example 3 of the present disclosure. FIG. 3E is a photograph of finished leather pieces that have good sewability which were be made into various leather wallet and keychain prototypes.

FIG. 4. includes photographs of the bioleather (A) that had undergone degassing, (B) that did not undergo degassing and (C) that did not undergo degassing before hotpress (left) and after hotpress (right) according to Example 4 of the present disclosure.

DESCRIPTION

It has been surprisingly found that bacterial cellulose obtained by a suitable method (e.g. fermentation using a Symbiotic Culture of Bacteria and Yeast (SCOBY)) can be used to synthesize an alternative leather with comparable mechanical properties and a similar look and feel to animal leather. Specifically, the method of producing the bacterial cellulose-based leather according to the present invention enables the production of a leather-like material with consistent quality, controllable thickness and tuneable properties. Advantageously, the method does not involve the use of conventional polyurethane which is associated with poor mechanical performance and non-biodegradability.

Thus, in a first aspect of the invention, there is provided a formulation for producing a bacterial cellulose-based leather comprising:

(a) bacterial cellulose;

(b) a gelling agent;

(c) a plasticizer;

(d) a crosslinker; and

(e) a filler.

In embodiments herein, the word "comprising" may be interpreted as requiring the features mentioned, but not limiting the presence of other features. Alternatively, the word "comprising" may also relate to the situation where only the components/features listed are intended to be present (e.g. the word "comprising" may be replaced by the phrases "consists of" or "consists essentially of"). It is explicitly contemplated that both the broader and narrower interpretations can be applied to all aspects and embodiments of the present invention. In other words, the word "comprising" and synonyms thereof may be replaced by the phrase "consisting of" or the phrase "consists essentially of" or synonyms thereof and *vice versa*.

The phrase, "consists essentially of" and its pseudonyms may be interpreted herein to refer to a material where minor impurities may be present. For example, the material may be greater than or equal to 90% pure, such as greater than 95% pure, such as greater than 97% pure, such as greater than 99% pure, such as greater than 99.9% pure, such as greater than 99.99% pure, such as greater than 99.999% pure, such as 100% pure.

As used herein, the singular forms "a," "an," and "the" include plural referents unless the context clearly dictates otherwise. Thus, for example, reference to "a compound" includes mixtures of two or more such compounds, reference to "a composition" includes mixtures of two or more such compositions, and the like.

When used herein, the term "bacterial cellulose" refers to cellulose synthesized and isolated from any suitable bacterial source. For example, the bacterial cellulose may be obtained from a bacterium from the genera *Komagataeibacter, Acetobacter, Sarcina ventriculi* and *Agrobacterium*. In certain embodiments, the bacterial cellulose may be obtained from fermentation using a Symbiotic Culture of Bacteria and Yeast (SCOBY). In more particular embodiments, the bacterial cellulose may be obtained from the fermentation of a fruit/vegetable juice media with a Brix value of from 7 to 13°Bx using SCOBY. The term "Brix value" refers to the amount of dissolved sugar in a liquid solution and may be determined using any suitable method to do so.

Details of how to harvest bacterial cellulose from the fermentation broth are supplied in the example section below, though any suitable method may be used to achieve this.

In certain embodiments, the bacterial cellulose harvested may be cut into smaller pieces and further blended together with water, to form a slurry prior to mixing with other components of the formulation. In some embodiments, the bacterial cellulose may be present in the slurry in an amount of from 1 wt% to 10 wt%, such as from 2 wt% to 5 wt%, the remaining being water.

When used herein, the term "Symbiotic Culture of Bacteria and Yeast (SCOBY)" refers to a symbiotic fermentation culture consisting of a lactic acid bacteria (LAB), an acetic acid bacteria (AAB), and a yeast, which may arise in the preparation of sour foods and beverages such as kombucha.

When used herein, the term "gelling agent" refers to any suitable gel-forming agent which increases the viscosity of the formulation without substantially changing its other properties. In certain embodiments, the gelling agent may be selected from one or more of a group consisting of gelatin, agar, pectin, Xanthan gum, guar gum, an alginate, starch and cornstarch. In certain exemplified embodiments, the gelling agent may be selected from one or more of a group consisting of gelatin, agar and pectin.

The gelling agent may be present in any suitable amount in the formulation. In certain embodiments, the gelling agent may be present in the formulation in an amount of from 15 wt% to 50 wt% with respect to the dry mass of the bacterial cellulose. In certain exemplified embodiments, the gelling agent may be present in an amount of from 20 wt% to 44 wt%, such as about 20 wt%, about 40 wt% or about 44 wt%, with respect to the dry mass of the bacterial cellulose.

The term "about" as used herein can allow for a degree of variability in a value or range, for example, within 10%, within 5%, within 1%, within 0.5%, within 0.1%, within 0.05%, within 0.01%, within 0.005%, or within 0.001% of a stated value or of a stated limit of a range, and includes the exact stated value or range.

When used herein, the term "plasticizer" refers to any suitable substance that is added to make the bacterial cellulose-based leather softer and more flexible and/or to increase its plasticity. In certain embodiments, the plasticizer may be glycerol, polyvinyl alcohol and polyvinyl acetate. In certain exemplified embodiments, the plasticizer may be glycerol.

The plasticizer may be present in any suitable amount in the formulation. In certain embodiments, the plasticizer may be present in the formulation in an amount of from 30 wt% to 55 wt% with respect to the dry mass of the bacterial cellulose. In certain exemplified embodiments, the plasticizer may be present in an amount of from 40 wt% to 50 wt%, such as about 40 wt% or about 50 wt%, with respect to the dry mass of the bacterial cellulose.

When used herein, the term "crosslinker" refers to any suitable molecule that contains two or more reactive groups capable of chemically attaching to specific functional groups in the bacterial cellulose and/or other components in the formulation. For example, the crosslinker may have two or more carboxyl groups or amine groups (or a combination of both) to react with the hydroxyl groups in the bacterial cellulose to form ester or amide linkages respectively. In another example, the crosslinker may have two or more carboxyl groups to react with a hydroxyl group in the bacterial cellulose on one end and an amine group in another component in the formulation (e.g., gelatin) on the other end, thereby attaching the components in the formulation together. In certain embodiments, the crosslinker may be selected from a group consisting of a diacid (such as adipic acid), a diamine (such as ethylenediamine, butanediamine and hexanediamine), glutamic acid, boric acid, and more particularly, citric acid, azelaic acid, a diacrylate, a diisocyanate, a diepoxide, formaldehyde and glutaraldehyde. In certain exemplified embodiments, the crosslinker may be citric acid.

The crosslinker may be present in any suitable amount in the formulation. In certain embodiments, the crosslinker may be present in the formulation in an amount of from 2 wt% to 20 wt% with respect to the dry mass of the bacterial cellulose. In certain exemplified embodiments, the crosslinker may be present in an amount of from 2.5 wt% to 10 wt%, such as about 2.5 wt%, about 3 wt%, about 5 wt% or about 10 wt%, with respect to the dry mass of the bacterial cellulose.

When used herein, the term "filler" refers to any suitable substance that may be added to improve specific mechanical properties (e.g., increasing tensile strength and reducing brittleness) of the bacterial cellulose-based leather depending on the requirements of the final product. In certain embodiments, the filler may be selected from one or more of a group consisting of a polyphenol (such as catechin, caffeic acid and gallic acid) and more particularly, tannic acid. In certain exemplified embodiments, the filler may be tannic acid.

The filler may be present in any suitable amounts in the formulation. In certain embodiments, the filler may be present in the formulation in an amount of from 20 wt% to 65 wt% with respect

to the dry mass of the bacterial cellulose. In certain exemplified embodiments, the filler may be present in an amount of from 20 wt% to 60 wt%, such as about 20 wt%, about 22.5 wt%, about 30 wt%, about 40 wt% or about 60 wt%, with respect to the dry mass of the bacterial cellulose.

In certain embodiments, the formulation may further comprise reinforcement fibres, a preservative and/or an antimicrobial agent.

When used herein, the phrase "reinforcement fibres" refers to any suitable material (typically macro-sized, preferably in the range of from 500μm to 2mm compared to fillers which are typically molecules or nano-sized) that may be added to reinforce/strengthen the structural properties of the bacterial-cellulose based leather. In some embodiments of the invention that may be mentioned herein, the reinforcement fibres may be fruit fibres and vegetable waste, including processed pineapple leaf fibres.

When used herein, the term "preservative" refers to any suitable substance that may be added to prevent decay/decomposition of the bacterial cellulose-based leather. In some embodiments of the invention that may be mentioned herein, the preservative may be polylysine, sodium benzoate, parabens, natamycin, benzoic acid, sorbic acid and ascorbic acid. It will be appreciated that any suitable preservative that can be incorporated into the formulation and the bacterial cellulose-based leather may be used herein.

When used herein, the phrase "antimicrobial agent" refers to any suitable substance that kills or inhibits the growth of micro-organisms such as bacteria, fungi and algae.

In some embodiments of the invention that may be mentioned herein, the formulation may further comprise ethanol and water. Advantageously, the addition of liquids (e.g., water and ethanol) increases the efficiency of the blending process of the formulation and ensures thorough mixing of the components in the formulation. Furthermore, the addition of ethanol reduces the surface tension of the formulation and minimises the formation of bubbles during the blending process. Consequently, the casted product (i.e., BC-based leather) would have fewer bubbles and at the same time feels thicker and similar to conventional leather.

In a second aspect of the invention, there is provided a method of producing a bacterial cellulose-based leather comprising the steps of: (i) providing a formulation according to the first aspect of the invention; and (ii) pouring the formulation into a mould and casting the

formulation at a temperature of from 70°C to 90°C for a period of time to obtain the bacterial cellulose-based leather.

In certain embodiments, the formulation may be obtained by mixing a bacterial cellulose slurry with a gelling agent, a plasticizer, a crosslinker and a filler, as described above, to provide a mixture and blending the mixture to obtain the formulation. In more particular embodiments, the bacterial cellulose slurry may be obtained by providing a bacterial cellulose obtained from fermentation using SCOBY and subjecting the bacterial cellulose to blending, rinsing and straining to obtain the bacterial cellulose slurry. In even more particular embodiments, the bacterial cellulose may be obtained from fermenting a fruit/vegetable juice media with a Brix value of from 7°Bx to 13°Bx using SCOBY.

When used herein, the term "casting" refers to pouring the formulation into a mould and allowing the formulation to solidify for a period of time.

The period of time for step (ii) to be conducted may be determined by the skilled person based on their knowledge of the field and whether the casting process has been deemed completed or not. In certain embodiments, the period of time may be 18 hours to 24 hours, such as about 18 hours or about 24 hours.

In certain embodiments, the blending step may comprise blending the bacterial cellulose with a 5 vol% to 10 vol% bleach to obtain the bacterial cellulose slurry. In certain embodiments, the blending step may comprise blending the mixture with a wet miller to obtain the formulation. In certain embodiments, the gelling agent may be agar that is predissolved in water.

In certain embodiments, the method may further comprise the steps of (ii)(a) degassing the formulation prior to step (ii); and (ii)(b) subjecting the bacterial cellulose-based leather to a hotpressing step after step (ii).

When used herein, the term "degassing" refers to any suitable method to remove dissolved gases from the formulation that were introduced during, for example, mixing. In some embodiments, the degassing step may comprise degassing the formulation in a vacuum oven for about 5 mins. Advantageously, the degassing step minimizes shrinkage of the BC-based leather during casting and enhances the mechanical properties of the BC-based leather (for example the ultimate tensile strength (UTS)). Furthermore, as mentioned, the degassing step removes bubbles that may be formed during the blending process and consequently, the BC-

based leather would have fewer bubbles and at the same time feels thicker and similar to conventional leather.

When used herein, the phrase "hotpressing step" refers to the application of heat and pressure to compact and increase the density of the bacterial cellulose-based leather. In certain embodiments, the hotpressing step may comprise subjecting the bacterial cellulose-based leather to a temperature of about 80°C and at a pressure of about 5 bar (about 500 kPa) for about 5 mins. It will be appreciated that the exact time and pressure will vary depending on the desired thickness of the resulting leather, as well as the desired properties. Advantageously, the hotpressing step may create a more compact and uniform BC-based leather product with similar mechanical properties (e.g., Young's Modulus and ultimate tensile strength (UTS)) to animal leather.

In certain embodiments, the method may further comprise a step (iii), which subjects the bacterial cellulose-based leather to a post-treatment step, wherein the post-treatment step may comprise: (iii)(a) applying one or both of a varnish and a waterproofing layer on a surface of the bacterial cellulose-based leather; (iii)(b) dyeing the bacterial cellulose-based leather; and (iii)(c) adding a supporting base layer to the bacterial cellulose-based leather, the supporting base layer may be selected from the group consisting of polyester, nylon, cotton fabric and pineapple skin textile, optionally wherein the base layer is polyester.

In a third aspect of the invention, there is provided a bacterial cellulose-based leather comprising: (a) bacterial cellulose; (b) from 15 to 50 wt% gelling agent; (c) from 30 to 55 wt% plasticizer; (d) from 2 to 20 wt% crosslinker; and (e) from 20 to 65 wt% filler, wherein the wt% of each of (b) to (e) is with respect to the dry mass of the bacterial cellulose.

In certain embodiments, the bacterial cellulose-based leather may comprise: (a) bacterial cellulose; (b) from 20 wt% to 44 wt% gelling agent; (c) from 40 wt% to 50 wt% plasticizer; (d) from 2.5 wt% to 10 wt% crosslinker; and (e) from 20 wt% to 60 wt% filler, wherein the wt% of each of (b) to (e) is with respect to the dry mass of the bacterial cellulose.

The components of the bacterial cellulose-based leather are identical to those described in relation to the formulation in the first aspect of invention hereinbefore. As such, discussion of the components of the bacterial cellulose-based leather to this aspect of invention is omitted for the sake of brevity.

In certain embodiments, the bacterial-cellulose leather may have a Young's Modulus of from 10 MPa to 300 MPa; and/or the bacterial-cellulose leather may have an Ultimate Tensile Strength of from 5 MPa to 20 MPa. As will be appreciated, the components in the formulation may be adjusted to modify the mechanical properties of the BC-based leather depending on the requirements of the material (e.g., stretchability and touch and feel of the material)

In certain embodiments, the bacterial cellulose-based leather may further comprise reinforcement fibres, a preservative and/or an antimicrobial agent.

In certain embodiments, the bacterial cellulose-based leather may further comprise (a) a varnish; (b) a waterproofing layer; (c) a dye; and (d) a supporting base layer to the bacterial cellulose-based leather, the supporting base layer may be polyester, nylon, cotton fabric and pineapple skin textile.

Further aspects and embodiments of the invention will now be discussed by reference to the following non-limiting examples.

Examples

Example 1: Fermentation/production of bacteria cellulose (BC) slurry

Symbiotic Cultures of Bacteria and Yeast (SCOBYs) were kindly donated/collected from the public or local kombucha breweries. The SCOBYs were placed in a 5 L jar and 500 ml of boiled and subsequently cooled sugared tea (2 tea bags with 70 g/L sugar) was added into the jar. Fermentation was allowed to take place for about 5 days to obtain a starter culture. The starter culture was topped up with 500 ml of the sugared tea every 5 days.

Fruit/vegetable juices (for example, pineapple juice, papaya juice and orange juice or combinations thereof) were extracted via a juicer. The Brix level of the juice was tested and the fruit/vegetable juice media was diluted with water to around 7-8 Brix (FIG. 1A). Two methods of sterilization can be used: 1) autoclaving the fruit juice at 120°C for 20min or 2) heating the fruit juice to a boil and allowing it to cool to room temperature. The SCOBY inoculum from the starter culture was then added to the diluted and sterilized juice and allowed to ferment for 10-14 days. A thick bacterial cellulose (BC) mat was observed from the fermentation (FIG. 1B).

The BC mats/pieces were harvested and cut into smaller pieces for sterilization. 500 g of the chopped BC mats/pieces were added to a blender jar, together with 40 g of baking soda and 1.2L of water. The mixture was blended at 3000 rpm for 30 seconds to obtain a slurry-like consistency (FIG. 1C). 150 ml of 10% bleach was added and the mixture was further blended until the colour changed to creamy/pure white and allowed to rest at room temperature for 30 mins. The slurry was then poured into a filter pail and allowed to drip dry for 2 hours. 1 L of water was added to the rinse the slurry. The rinsing process was repeated 5 times. At the last rinse, the slurry was allowed to drip dry overnight and the slurry was collected.

The water content of the BC slurry was determined by drying an initial weight (for e.g. 10 g) of the wet slurry in a 60 °C oven for 4 hours and then measuring the dried weight. The dried weight is then expressed as a percentage of the initial weight (dried weight/initial weight x 100%). The bacterial cellulose may be present in the BC slurry in an amount of from 2 wt% to 5 wt%, the remaining being water.

Example 2: Production of bioleather

Example 2.1: Production of gelatin bioleather

The BC slurry was mixed with additives such as gelatin/pudding powder with gelatin, glycerol, citric acid and/or processed reinforcement fibres together with an ethanol/water mixture and blended into a homogenous paste using a wet miller. Various formulations are shown in Table 1. The additives are chosen for these reasons: gelatin as a gelling material, glycerol as a plasticizer, citric acid as a cross linker and fillers for reinforcement.

As bubbles are produced during the mechanical blending process through the introduction of air, ethanol was mixed with water to help reduce surface tension of the mixture and minimise the formation of bubbles. If only pure water was used during the blending process, the casted product would have entrapped bubbles (FIG. 2A). With the addition of ethanol with water and coupled with degassing, the casted product would have fewer bubbles and at the same time feels thicker and similar to leather. (FIG. 2B).

The mixture comprising the BC slurry and the additives was degassed under lower pressure at room temperature for about 5 min or until the bubbling settles, and then poured into a flat mould for casting. Degassing is important to remove any entrapped air bubbles within the blended BC slurry formulation. The gelatin bioleather formulation was casted in an oven at 70 °C for 18 to 24 hours depending on the amount of liquid used in blending process. More

liquid will require a longer drying time. The casted gelatin bioleather piece was allowed to cool to room temperature.

Table 1: Gelatin bioleather formulations

Sample	**Conditions**	**Young's Modulus (MPa)**	**Ultimate tensile strength (MPa)**	**Strain at break (%)**
Gelatin bioleather, 40 wt% gelatin, 2.5 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid wrt dried BC	Direct addition gelatin, Degas, hotpress	32.17 ± 12.82	8.76 ± 2.85	32.03 ± 1.97
Gelatin bioleather, 40 wt% gelatin, 5 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid wrt dried BC	Direct addition gelatin, Degas, hotpress	26.43 ± 7.18	5.45 ± 0.72	31.57 ± 4.90
Gelatin bioleather, 40 wt% gelatin, 10 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid wrt dried BC	Direct addition gelatin, Degas, hotpress	15.95 ± 6.21	5.32 ± 1.96	37.04 ± 5.98

- Gelatin Bioleather Formulation Example 1:

225 g of wet BC slurry (11 g dried BC), 10.5 g pudding powder (~4.4 g gelatin and 6.1 g sugar), 5.25 g glycerol, 0.3 g or 0.6 g or 1.1 g citric acid and 4.5 g tannic acid were directly added and mixed together with 225 ml ethanol and 300 ml water. The mixture was then blended using a wet miller at 3000 rpm for 1.5 mins and then degassed in a vacuum oven at room temperature for 5 minutes. It was then poured into a 20 x 20 cm silicon tray or plastic container, spread evenly and casted in the oven at 70 °C for 18 hours. The casted gelatin bioleather piece was allowed to cool to room temperature.

- Gelatin Bioleather Formulation Example 2:

600 g of wet BC slurry (28 g dried BC), 28 g pudding powder (~11.7 g gelatin and 16.3 g sugar), 14 g glycerol, 0.8 g citric acid, 12 g tannic acid and 1.33 g carbon black were directly added and mixed together with 600 ml ethanol and 800 ml water. The mixture was then

blended using a wet miller at 3000 rpm for 1.5 mins and then degassed in a vacuum oven at room temperature for 5 minutes. It was then poured into a 30 x 40 cm (length x width) plastic container, spread evenly and casted in the oven at 70 °C for 24 hours. The casted gelatin bioleather piece was allowed to cool to room temperature.

Example 2.2: Production of agar and pectin bioleather

The BC slurry was added with additives such as agar or pectin or combination of both agar and pectin, glycerol, citric acid and/or processed reinforcement fibres together with water and blended into homogenous paste using a wet miller. Various formulations are shown in Table 2 and 3. The additives are chosen for these reasons: Agar, pectin as gelling material, glycerol as plasticizer, citric acid for as cross linker and fillers for reinforcement. Agar is specifically predissolved in water at 80-90°C before pouring into the wet milling chamber.

Reinforcement fibres such as pineapple leaf fibre (PALF) and agriculture waste is processed mechanically via freeze milling and then added into the bioleather formulation. Specifically, 2-3 g of dried PALF was placed into the grinding vial and inserted into the freezer mill containing liquid nitrogen. The parameters were set as follows: precool (2 min) followed by 3 cycles of run time (2 min) and cool time (2 min) at a rate of 10 counts per min. The milled PALF was then collected.

The mixture comprising the BC slurry and additives was degassed under lower pressure at room temperature for about 5 min or until the bubbling settles, and then poured into a flat mould for casting. The agar/pectin bioleather formulation was casted in the oven at 90 °C for 24 hours. The casted agar/pectin bioleather piece was allowed to cool to room temperature.

Table 2: Agar bioleather formulations

Sample	**Conditions**	**Young's Modulus (MPa)**	**Ultimate tensile strength (MPa)**	**Strain at break (%)**
Agar bioleather, 20 wt% Agar, 2.5 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid wrt BC wrt dried BC	Predissolved Agar, Degas, hotpress	132.45 ± 2.42	13.94 ± 1.07	21.63 ± 1.32

Agar bioleather, 20 wt% Agar, 2.5 wt% citric acid, 48 wt% glycerol, 60 wt% tannic acid wrt dried BC	Predissolved Agar, Degas, hotpress	50.59 ± 8.77	9.21 ± 0.26	33.99 ± 1.70
Agar bioleather, 20 wt% Agar, 2.5 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid, 9 wt% defribrillated PALF wrt dried BC	Predissolved Agar, Degas, hotpress	131.81 ± 16.22	14.60 ± 0.31	19.07 ± 0.04

- Agar Bioleather Formulation Example 1:

225 g of wet BC slurry (11 g dried BC), 2.2 g predissolved agar, 5.25 g glycerol, 0.3 g citric acid, 4.5 g or 6 g tannic acid and/or 1 g processed pineapple leaf fibers (PALF) were predissolved with 525 ml water heated to 80 °C. The mixture was then blended using a wet miller at 3000 rpm for 1.5 mins and then degassed in a vacuum oven at room temperature for 5 minutes. It was then poured into a 20 x 20 cm (length x width) silicon tray or plastic container, spread evenly and casted in the oven at 90 °C for 24 hours. The casted agar bioleather piece was allowed to cool to room temperature.

- Agar Bioleather Formulation Example 2:

300 g of wet BC slurry (14.5 g dried BC), 7 g predissolved agar, 7 g glycerol, 0.4 g citric acid, 6 g tannic acid were predissolved with 700 ml water heated to 90 °C. The mixture was then blended using a wet miller at 3000 rpm for 1.5 mins and then degassed in a vacuum oven at room temperature for 5 minutes. It was then poured into a 29 x 20 cm (length x width) plastic container, spread evenly and casted in the oven at 90 °C for 24 hours. The casted agar bioleather piece was allowed to cool to room temperature.

Table 3: Pectin bioleather formulation

Sample	**Conditions**	**Young's Modulus (MPa)**	**Ultimate tensile strength (MPa)**	**Strain at break (%)**

Pectin bioleather 44 wt% low MW Pectin, 50 wt% glycerol, 3 wt% citric acid, 22.5 wt% tannic acid, wrt dried BC	Predissolved Pectin, Degas, hotpress	271.19 ±8.53	33.75 ± 2.42	16.30 ± 2.28

- Pectin Bioleather Formulation Example:

100 g of wet BC slurry (5 g dried BC), 2.2 g predissolved low molecular weight pectin, 2.5 g glycerol, 0.15 g citric acid, 1.125 g tannic acid were directly added and mixed together with 100 ml water. The mixture was then blended using a wet miller at 3000 rpm for 1.5 mins and then degassed in a vacuum oven at room temperature for 5 minutes. It was then poured into a 9 x 9 cm silicon tray, spread evenly and casted in the oven at 90 °C for 24 hours. The casted pectin bioleather piece was allowed to cool to room temperature.

Example 3: Post Processing of bioleather

Commercial resolene was applied using a dishwashing sponge onto the surface of the bioleather. The coating was allowed to dry completely for 1 to 2 hours. The coated bioleather was then placed between stainless steel plates together with a film releasing layer. They are then subjected to a heat press at 70°C (for gelatin bioleather pieces) or 90°C (for agar/pectin bioleather pieces) for 5 min. The hotpressing step helps in achieving better finishing. The sandwiched plates were allowed to cool to room temperature before removing the finished bioleather piece.

FIG. 3A-D includes photographs of finished leather pieces that were post treated by dyeing (leather dyes) and varnish (resolene). FIG. 3E is a photograph of finished leather pieces that have good sewability which were made into various leather wallet and keychain prototypes.

Example 4: Investigating the effects of changing the concentration of tannic acid and citric acid, hotpressing and degassing

Tannins or tannic acid is commonly used in tanning of leather, acting as filler and imparting colour. The addition of tannic acid gave the material a more leather-like appearance and feel. It also made the material less stiff (lower Young Modulus); increasing tannic acid content resulted in lower Young Modulus. However, tannic acid also reduced the tensile strength and strain at break. Overall, the mechanical properties of the bioleather were comparable to that of animal leather but at the lower limit (Table 4).

The citric acid content in the bioleather formulation was varied (2.5, 5 and 10%) and the bioleather was subjected to hotpressing (80°C, 5 bar, 5 mins) to investigate the impact of citric acid and hotpressing on the mechanical properties of the bioleather (Table 4). 5% citric acid content resulted in the lowest Young Modulus but had comparable ultimate tensile strength (UTS) and strain at break to 10% citric acid.

Table 4. Mechanical properties of bioleather with varying tannic acid and citric acid content

Sample	**Conditions**	**Young's Modulus (MPa)**	**Ultimate tensile strength (MPa)**	**Strain at break (%)**
Gelatin bioleather, 40 wt% gelatin, 2.5 wt% citric acid, 48 wt% glycerol, 0 wt% tannic acid wrt BC	Direct addition gelatin, Degas, no hotpress	46.02 ± 16.17	12.09 ± 2.67	36.51 ± 14.98
Gelatin bioleather, 40 wt% gelatin, 2.5 wt% citric acid, 48 wt% glycerol, 20 wt% tannic acid wrt BC	Direct addition gelatin, Degas, no hotpress	45.06 ± 13.32	10.40 ± 4.21	26.11 ± 1.21
Gelatin bioleather, 40 wt% gelatin, 2.5 wt% citric acid, 48 wt% glycerol, 30 wt% tannic acid wrt BC	Direct addition gelatin, Degas, no hotpress	41.16 ± 13.43	7.51 ± 1.97	23.12 ± 5.25
Gelatin bioleather, 40 wt% gelatin, 2.5 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid wrt BC	Direct addition gelatin, Degas, no hotpress	32.62 ± 8.10	7.30 ± 1.66	26.61 ± 2.37
Gelatin bioleather, 40 wt% gelatin, 2.5 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid wrt dried BC	Direct addition gelatin, Degas, hotpress	32.17 ± 12.82	8.76 ± 2.85	32.03 ± 1.97
Gelatin bioleather, 40 wt% gelatin, 5 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid wrt dried BC	Direct addition gelatin, Degas, hotpress	26.43 ± 7.18	5.45 ± 0.72	31.57 ± 4.90

Gelatin bioleather, 40 wt% gelatin, 10 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid wrt dried BC	Direct addition gelatin, Degas, hotpress	15.95 ± 6.21	5.32 ± 1.96	37.04 ± 5.98
Animal leather [1,2,3,4]		30-100	6.6 to 72	22.5 to 53.4

[1] Meyer, M., Dietrich, S., Schulz, H. & Mondschein, A. Comparison of the Technical Performance of Leather, Artificial Leather, and Trendy Alternatives. Coatings 11, 226 (2021).

[2] Ali, F., Kamal, M. & Islam, S. Comparative Study on Physical Properties of Different Types of Leather in Bangladesh. Mahbub Kamal J. Eng. Res. Appl. 10, 55 63 (2020).

[3] Dietrich, S. et al. Simulation Of Leather Visco-Elastic Behavior Based On Collagen Fiber- Bundle Properties And A Meso-Structure Network Model. Materials. 14, 1 19 (2021).

[4] Yim, S. M., Song, J. E. & Kim, H. R. Production And Characterization Of Bacterial Cellulose Fabrics By Nitrogen Sources Of Tea And Carbon Sources Of Sugar. Process Biochem. 59, 26 36 (2017).

Degassing is an important step as samples that were not subjected to degassing suffered from shrinkage, were thicker and had uneven surfaces (FIG. 4). Mechanical properties were also affected with decrease in UTS for samples that did not undergo degassing (Table 5).

Table 5. Mechanical properties of bioleather with varying citric acid content without degassing.

Sample	**Conditions**	**Young's Modulus (MPa)**	**Ultimate tensile strength (MPa)**	**Strain at break (%)**
Gelatin bioleather, 40 wt% gelatin, 2.5 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid	Direct addition gelatin, No degas, hotpress	24.17 ± 10.72	3.81 ± 2.56	46.68 ± 29.05
Gelatin bioleather, 40 wt% gelatin, 5 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid	Direct addition gelatin, No degas, hotpress	22.84 ± 6.80	4.69 ± 1.86	25.36 ± 5.58
Gelatin bioleather, 40 wt% gelatin, 10 wt% citric	Direct addition gelatin, No degas, hotpress	13.40 ± 5.43	3.41 ± 1.19	29.55 ± 1.23

acid, 48 wt% glycerol, 40 wt% tannic acid				

The hotpressing step creates a more compact and uniform bioleather product with similar Young's Modulus and ultimate tensile strength (UTS) to animal leather. An additional supporting base layer such as polyester (iron-on) can be used in conjunction with the bioleather to further improve the mechanical strength of the final product. The UTS for the bioleather with 5% citric acid content improved from 5.45 MPa (without iron-on polyester) to 15.68 MPa (with iron-on polyester).

Example 5: Summary

Table 6 summarises the different bioleather formulations and their mechanical properties.

Table 6: Different bioleather formulations and their mechanical properties

Sample	**Conditions**	**Young's Modulus (MPa)**	**Ultimate tensile strength (MPa)**	**Strain at break (%)**
Gelatin bioleather, 40 wt% gelatin, 2.5 wt% citric acid, 48 wt% glycerol, 0 wt% tannic acid wrt BC	Direct addition gelatin, degas, no hotpress	46.02 ± 16.17	12.09 ± 2.67	36.51 ± 14.98
Gelatin bioleather, 40 wt% gelatin, 2.5 wt% citric acid, 48 wt% glycerol, 20 wt% tannic acid wrt BC	Direct addition gelatin, degas, no hotpress	45.06 ± 13.32	10.40 ± 4.21	26.11 ± 1.21
Gelatin bioleather, 40 wt% gelatin, 2.5 wt% citric acid, 48 wt% glycerol, 30 wt% tannic acid wrt BC	Direct addition gelatin, degas, no hotpress	41.16 ± 13.43	7.51 ± 1.97	23.12 ± 5.25
Gelatin bioleather, 40 wt% gelatin, 2.5 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid wrt BC	Direct addition gelatin, degas, no hotpress	32.62 ± 8.10	7.30 ± 1.66	26.61 ± 2.37

Gelatin bioleather, 40 wt% gelatin, 2.5 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid wrt dried BC	Direct addition gelatin, degas, hotpress	32.17 ± 12.82	8.76 ± 2.85	32.03 ± 1.97
Gelatin bioleather, 40 wt% gelatin, 5 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid wrt dried BC	Direct addition gelatin, degas, hotpress	26.43 ± 7.18	5.45 ± 0.72	31.57 ± 4.90
Gelatin bioleather, 40 wt% gelatin, 10 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid wrt dried BC	Direct addition gelatin, degas, hotpress	15.95 ± 6.21	5.32 ± 1.96	37.04 ± 5.98
Agar bioleather, 20 wt% Agar, 2.5 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid wrt BC wrt dried BC	Predissolved agar, degas, hotpress	132.45 ± 2.42	13.94 ± 1.07	21.63 ± 1.32
Agar bioleather, 20 wt% Agar, 2.5 wt% citric acid, 48 wt% glycerol, 60 wt% tannic acid wrt dried BC	Predissolved agar, degas, hotpress	50.59 ± 8.77	9.21 ± 0.26	33.99 ± 1.70
Agar bioleather, 20 wt% Agar, 2.5 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid wrt dried BC	Direct addition agar, degas, hotpress	71.61	8.42	14.18
Agar bioleather, 40 wt% Agar, 2.5 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid wrt dried BC	Direct addition agar, degas, hotpress	38.18	7.26	19.09
Agar bioleather, 20 wt% Agar, 2.5 wt% citric acid, 48 wt% glycerol, 40 wt% tannic acid, 9 wt% defribrillated PALF wrt dried BC	Predissolved agar, degas, hotpress	131.81 ± 16.22	14.60 ± 0.31	19.07 ± 0.04
Pectin bioleather	Predissolved Pectin, Degas,	271.19 ±8.53	33.75 ± 2.42	16.30 ± 2.28

44 wt% low MW Pectin, 50 wt% glycerol, 3 wt% citric acid, 22.5 wt% tannic acid, wrt dried BC	hotpress			
Animal leather [1,2,3,4]		30-100	6.6 to 72	22.5 to 53.4

CLAIMS

1. A formulation for producing a bacterial cellulose-based leather comprising:
   (a) bacterial cellulose;
   (b) a gelling agent;
   (c) a plasticizer;
   (d) a crosslinker; and
   (e) a filler.

2. The formulation according to claim 1, wherein the formulation further comprises reinforcement fibres, a preservative and/or an antimicrobial agent.

3. The formulation according to claim 1 or claim 2, wherein the bacterial cellulose is obtained from fermentation using a Symbiotic Culture of Bacteria and Yeast (SCOBY).

4. The formulation according to any of the preceding claims, wherein the gelling agent is selected from one or more of a group consisting of gelatin, agar, pectin, Xanthan gum, guar gum, an alginate, starch and cornstarch, optionally wherein, the gelling agent is selected from one or more of a group consisting of gelatin, agar and pectin.

5. The formulation according to any of the preceding claims, wherein the gelling agent is present in the formulation in an amount of from 15 wt% to 50 wt% with respect to the dry mass of the bacterial cellulose, optionally wherein the gelling agent is present in an amount of from 20 wt% to 44 wt% with respect to the dry mass of the bacterial cellulose.

6. The formulation according to any of the preceding claims, wherein the plasticizer is selected from one or more of a group consisting of glycerol, polyvinyl alcohol and polyvinyl acetate, optionally wherein the plasticizer is glycerol.

7. The formulation according to any of the preceding claims, wherein the plasticizer is present in the formulation in an amount of from 30 wt% to 55 wt% with respect to the dry mass of the bacterial cellulose, optionally wherein the plasticizer is present in an amount of from 40 wt% to 50 wt% with respect to the dry mass of the bacterial cellulose.

8. The formulation according to any of the preceding claims, wherein the crosslinker is selected from one or more of a group consisting of a diacid (such as adipic acid), a diamine (such as ethylenediamine, butanediamine and hexanediamine), glutamic acid, boric acid, and

more particularly, citric acid, azelaic acid, a diacrylate, a diisocyanate, a diepoxide, formaldehyde and glutaraldehyde, optionally wherein the crosslinker is citric acid.

9. The formulation according to any of the preceding claims, wherein the crosslinker is present in the formulation in an amount of from 2 wt% to 20 wt% with respect to the dry mass of the bacterial cellulose, optionally wherein the crosslinker is present in an amount of from 2.5 wt% to 10 wt% with respect to the dry mass of the bacterial cellulose.

10. The formulation according to any of the preceding claims, wherein the filler is selected from one or more of a group consisting of a polyphenol (such as catechin, caffeic acid and gallic acid) and more particularly, tannic acid, optionally wherein the filler is tannic acid.

11. The formulation according to any of the preceding claims, wherein the filler is present in the formulation in an amount of from 20 wt% to 65 wt% with respect to the dry mass of the bacterial cellulose, optionally wherein the filler is present in an amount of from 20 wt% to 60 wt% with respect to the dry mass of the bacterial cellulose.

12. The formulation according to any of the preceding claims, wherein the bacterial cellulose is in the form of a slurry.

13. The formulation according to claim 12, wherein the bacterial cellulose is present in the slurry in an amount of from 2 wt% to 5 wt%, the remaining being water.

14. The formulation according to claim 3, wherein the bacterial cellulose is obtained from the fermentation of a fruit/vegetable juice media with a Brix value of from 7°Bx to 13°Bx.

15. The formulation according to any of claims 2 to 14, wherein the reinforcement fibres are selected from one or more of a group consisting of fruit fibres and vegetable waste, optionally wherein the reinforcement fibres are processed pineapple leaf fibres.

16. The formulation according to any of claims 2 to 15, wherein the preservative is selected from one or more of a group consisting of polylysine, sodium benzoate, parabens, natamycin, benzoic acid, sorbic acid and ascorbic acid.

17. The formulation according to any of the preceding claims, wherein the formulation further comprises ethanol and water.

18. A method of producing a bacterial cellulose-based leather comprising the following steps:

(i) providing a formulation according to any of claims 1 to 17; and

(ii) pouring the formulation into a mould and casting the formulation at a temperature of from 70°C to 90°C for a period of time to obtain the bacterial cellulose-based leather.

19. The method according to claim 18, wherein the formulation according to any of claims 1 to 17 is obtained by mixing a bacterial cellulose slurry with a gelling agent, a plasticizer, a crosslinker and a filler, as described in any one of claims 1 to 17, to provide a mixture and blending the mixture to obtain the formulation, optionally wherein the bacterial cellulose slurry is obtained by providing a bacterial cellulose obtained from fermentation using SCOBY and subjecting the bacterial cellulose to blending, rinsing and straining.

20. The method according to claim 18 or claim 19, further comprising one or both of the following steps:

(ii)(a) degassing the formulation prior to step (ii) of claim 18; and

(ii)(b) subjecting the bacterial cellulose-based leather to a hotpressing step after step (ii) of claim 18.

21. The method according to any of claims 18 to 20, further comprising:

(iii) subjecting the bacterial cellulose-based leather to a post-treatment step, wherein the post-treatment step comprises one or more of the following:

(iii)(a) applying one or both of a varnish and a waterproofing layer on a surface of the bacterial cellulose-based leather;

(iii)(b) dyeing the bacterial cellulose-based leather; and

(iii)(c) adding a supporting base layer to the bacterial cellulose-based leather, the supporting base layer is selected from the group consisting of polyester, nylon, cotton fabric and pineapple skin textile, optionally wherein the base layer is polyester.

22. The method according to any one of claim 19 and claims 20 and 21, as dependent upon claim 19, wherein one or both of the following apply:

(a) the fermentation step in claim 19 comprises fermenting a fruit/vegetable juice media with a Brix value of from 7°Bx to 13°Bx using SCOBY; and

(b) the period of time in claim 18(ii) is from 18 to 24 hours.

23. The method according to any one of claim 19 and claims 20 to 22, as dependent upon claim 19, wherein the blending step comprises blending the bacterial cellulose with a 5 vol% to 10 vol% bleach.

24. The method according to any one of claim 19 and claims 20 to 23, as dependent upon claim 19, wherein the gelling agent is agar, optionally wherein the gelling agent is agar predissolved in water.

25. The method according to any one of claim 19 and claims 20 to 24, as dependent upon claim 19, wherein the blending step comprises blending the mixture with a wet miller.

26. The method according to any one of claim 20 and claims 21 to 25, as dependent upon claim 20, wherein step (ii)(a) comprises degassing the formulation in a vacuum oven for about 5 mins.

27. The method according to any one of claim 20 and claims 21 to 26, as dependent upon claim 20, wherein step (ii)(b) comprises subjecting the bacterial cellulose-based leather to a hotpressing step at a temperature of about 80°C and at a pressure of about 5 bar for about 5 mins.

28. A bacterial cellulose-based leather comprising:

(a) bacterial cellulose;

(b) from 15 wt% to 50 wt% gelling agent;

(c) from 30 wt% to 55 wt% plasticizer;

(d) from 2 wt% to 20 wt% crosslinker; and

(e) from 20 wt% to 65 wt% filler,

wherein the wt% of each of (b) to (e) is with respect to the dry mass of the bacterial cellulose, and

wherein one or both of the following apply:

(i) the bacterial-cellulose leather has a Young's Modulus of from 10 MPa to 300 MPa; and

(ii) the bacterial-cellulose leather has an Ultimate Tensile Strength of from 5 MPa to 35 MPa.

29. The bacterial cellulose-based leather according to claim 28, wherein the bacterial cellulose-based leather comprises:

(a) bacterial cellulose;

(b) from 20 wt% to 44 wt% gelling agent;

(c) from 40 wt% to 50 wt% plasticizer;
(d) from 2.5 wt% to 10 wt% crosslinker; and
(e) from 20 wt% to 60 wt% filler.
wherein the wt% of each of (b) to (e) is with respect to the dry mass of the bacterial cellulose.

30. The bacterial cellulose-based leather according to claim 28 or claim 29, wherein the gelling agent is selected from one or more of a group consisting of gelatin, agar, pectin, Xanthan gum, guar gum, an alginate, starch and cornstarch, optionally wherein, the gelling agent is selected from one or more of a group consisting of gelatin, agar and pectin.

31. The bacterial cellulose-based leather according to any of claims 28 to 30, wherein the plasticizer is selected from one or more of a group consisting of glycerol, polyvinyl alcohol and polyvinyl acetate, optionally wherein the plasticizer is glycerol.

32. The bacterial cellulose-based leather according to any of claims 28 to 31, wherein the crosslinker is selected from one or more of a group consisting of a diacid (such as adipic acid), a diamine (such as ethylenediamine, butanediamine and hexanediamine), glutamic acid, boric acid, and more particularly, citric acid, azelaic acid, a diacrylate, a diisocyanate, a diepoxide, formaldehyde and glutaraldehyde, optionally wherein the crosslinker is citric acid.

33. The bacterial cellulose-based leather according to any of claims 28 to 32, wherein the filler is selected from one or more of a group consisting of a polyphenol (such as catechin, caffeic acid and gallic acid) and more particularly, tannic acid, optionally wherein the filler is tannic acid.

34. The bacterial cellulose-based leather according to any of claims 28 to 33, wherein the bacterial cellulose-based leather further comprises reinforcement fibres, a preservative and/or an antimicrobial agent.

35. The bacterial cellulose-based leather according to any of claims 28 to 34, wherein the bacterial cellulose-based leather further comprises one or more of the following:
(a) a varnish;
(b) a waterproofing layer;
(c) a dye; and

(d) a supporting base layer to the bacterial cellulose-based leather, the supporting base layer is selected from the group consisting of polyester, nylon, cotton fabric and pineapple skin textile, optionally wherein the base layer is polyester.