

CLAIMS

1. An engineered plant material comprising:
one or more substantially-delignified, cellulose-based fibers chemically-extracted intact from a matrix of lignin and hemicellulose in a piece of natural plant material.
- 5 2. A composite material comprising:
a plurality of substantially-delignified, cellulose-based fibers, each cellulose-based fiber being chemically-extracted intact from a matrix of lignin and hemicellulose in a respective piece of natural plant material; and
10 a polymer infiltrating each cellulose-based fiber.
3. A composite material comprising:
a structural matrix; and
a plurality of substantially-delignified, cellulose-based fibers disposed within the
15 structural matrix, each cellulose-based fiber being chemically-extracted intact from a matrix of lignin and hemicellulose in a respective piece of natural plant material.
4. The material of claim 3, wherein the structural matrix comprises a polymer or concrete.
- 20 5. The material of any one of claims 2 and 4, wherein the polymer comprises epoxy resin, polyvinyl alcohol (PVA), polyethylene glycol (PEO), polyamide (PA), polyethylene terephthalate (PET), polybutylene terephthalate (PBT), polytrimethylene terephthalate (PTT), polyacrylonitrile (PAN), polycaprolactam (PA6), poly(m-phenylene isophthalamide) (PMIA),
25 poly-p-phenylene terephthalamide (PPTA), polyurethane (PU), polycarbonate (PC), polypropylene (PP), high-density polyethylene (HDPE), polystyrene (PS), polycaprolactone (PCL), polybutylene succinate (PBS), polybutylene adipate terephthalate (PBAT), poly(butylene succinate-co-butylene adipate) (PBSA), polyhydroxybutyrate (PHB), poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), poly(glycolic acid) (PGA), polypyrrole (PPy), polythiophene
30 (PTh), polyvinylidene fluoride (PVDF), polyvinyl fluoride (PVF), ethylene vinyl alcohol (EVOH), poly(vinylidene chloride) (PVDC), polyxylylene adipamide (MXD6), polyethylene (PE), polyvinyl chloride (PVC), poly(methyl methacrylate) (PMMA), acrylonitrile butadiene styrene (ABS), polyimide (PI), polyethylenimine (PEI), polylactic acid (PLA), octadecyltrichlorosilane (OTS), polyoctahedral silsesquioxane (POSS), paramethylstyrene

(PMS), polydimethylsiloxane (PDMS), poly(ethylene naphthalate (PEN), a graft copolymer of acrylonitrile-butadiene-styrene-methylmethacrylate (ABSM), dodecyltrimethoxysilane (DTMS), rosin, chitin, chitosan, protain, plant oil, carboxymethyl cellulose, cellulose acetate, starch, agar, alginic acid, or any combination of the foregoing.

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6. The material of claim 4, wherein the concrete comprises Portland cement, agro-concrete, magnesium carbonate (MgCO_3), magnesium hydroxide ($\text{Mg}(\text{OH})_2$), or any combination of the foregoing.

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7. The material of any one of claims 2-3, wherein a content of the plurality of the cellulose-based fibers within the composite material is at least 0.1 wt%.

8. The material of any one of claims 1-3, wherein the natural plant material is a species in the *Poales* order.

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9. The material of any one of claims 1-3, wherein the natural plant material is a species in the *Poaceae* family.

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10. The material of any one of claims 1-3, wherein the natural plant material is a natural grass.

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11. The material of any one of claims 1-3, wherein the natural plant material comprises bamboo, reed, sugarcane, maize, wheat, rice, jute, hemp, kenaf, ramie, banana, corn, wheat, rice, roselle, rattan, sorghum, sisal, palm, henequen, manila, pineapple, curaua, yucca, cabuya, screw pine, abaca, gladiolus, or agave.

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12. The material of any one of claims 1-3, wherein each cellulose-based fiber has a length of at least 5 cm and a maximum cross-sectional dimension in a plane perpendicular to a direction of the length of at least 5 μm .

13. The material of claim 12, wherein the length of each cellulose-based fiber is between 5 cm and 200 cm, inclusive.

14. The material of claim 12, wherein the maximum cross-sectional dimension of each cellulose-based fiber is between 5 μm and 1 mm, inclusive.

15. The material of claim 12, wherein the maximum cross-sectional dimension of each cellulose-based fiber is at least 100 μm .

16. The material of any one of claims 1-3, wherein each cellulose-based fiber has a specific strength of at least 0.5 $\text{GPa} \cdot \text{cm}^3/\text{g}$, a crystallinity of at least 40%, a tensile strength of at least 0.5 GPa, a Young's modulus of at least 20 GPa, a density of at least 0.8 g/cm^3 , or any combination the foregoing.

17. The material of any one of claims 1-3, wherein each cellulose-based fiber has a cellulose content of at least 70 wt%, a hemicellulose content less than or equal to 10 wt%, a lignin content less than or equal to 10 wt%, or any combination of the foregoing.

18. The material of any one of claims 1-3, wherein each cellulose-based fiber has a moisture content less than or equal to 10 wt%.

19. The material of any one of claims 1-3, wherein provided on or within each cellulose-base fiber is a conductive additive, a magnetic additive, a piezoelectric material, a stimuli-responsive material, a catalytic material, or any combination of the foregoing.

20. A method comprising:

(a) subjecting a piece of natural plant material to one or more chemical treatments so as to remove substantially all lignin therefrom; and
(b) after (a), extracting from the chemically-treated piece a plurality of substantially-delignified, cellulose-based fibers separated from each other.

21. The method of claim 20, wherein (b) comprises agitating the chemically-treated piece in solution.

22. The method of claim 20, wherein at least one of the one or more chemical treatments comprises partial or full immersion in one or more chemical solutions.

23. The method of claim 22, wherein the one or more chemical solutions comprise an alkaline solution.

24. The method of claim 22, wherein the one or more chemical solutions comprise
5 sodium hydroxide (NaOH), lithium hydroxide (LiOH), potassium hydroxide (KOH), sodium sulfite (Na₂SO₃), sodium sulfide (Na₂S), Na_nS wherein n is an integer, urea (CH₄N₂O), sodium bisulfite (NaHSO₃), sulfur dioxide (SO₂), anthraquinone (C₁₄H₈O₂), methanol (CH₃OH), ethanol (C₂H₅OH), butanol (C₄H₉OH), formic acid (CH₂O₂), hydrogen peroxide (H₂O₂), acetic acid (CH₃COOH), butyric acid (C₄H₈O₂), peroxyformic acid (CH₂O₃), peroxyacetic acid (C₂H₄O₃),
10 ammonia (NH₃), tosylic acid (p-TsOH), sodium hypochlorite (NaClO), sodium chlorite (NaClO₂), chlorine dioxide (ClO₂), chlorine (Cl₂), or any combination of the foregoing.

25. The method of claim 20, wherein after (b) the plurality of cellulose-based fibers remain attached to a base portion of the natural plant material, and the method further comprises
15 severing the cellulose-based fibers from the base portion.

26. The method of claim 20, further comprising:

(c) after (b), drying the cellulose-based fibers such that evaporation of water from each cellulose-based fiber causes densification thereof.

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27. The method of claim 26, wherein the drying of (c) comprises exposing to an air or gas flow, exposing to a stagnant volume of air or gas, exposing to vacuum, exposing to room temperature, heating to a temperature above room temperature, or any combination thereof.

28. The method of claim 26, wherein, after (c), each cellulose-based fiber has a moisture content of 10 wt% or less.

29. The method of claim 26, wherein a volume of each cellulose-based fiber after (c) is at least 10% less than a volume of each cellulose-based fiber before (c).

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30. The method of claim 20, wherein (a) comprises:

(a1) immersing at least a first portion of the piece of natural plant material within a first solution to remove some of the lignin in the immersed first portion; and

(a2) after (a1), immersing at least the first portion within a second solution to remove remaining lignin in the immersed first portion.

31. The method of claim 30, wherein, after (a1) and prior to (a2), at least the first
5 portion is washed with water to remove remnants of the first solution.

32. The method of claim 30, wherein the first solution comprises peroxyformic acid
(CH₂O₃).

10 33. The method of any one of claim 30 and claim 32, wherein the second solution
comprises sodium hydroxide (NaOH) or potassium hydroxide (KOH).

34. The method of claim 30, wherein the first solution comprises a mixture of
potassium hydroxide (KOH) and sodium sulfite (Na₂SO₃), a mixture of potassium hydroxide
15 (KOH) and sodium sulfide (Na₂S), a mixture of sodium hydroxide (NaOH) and sodium sulfite
(Na₂SO₃), or a mixture of sodium hydroxide (NaOH) and sodium sulfide (Na₂S).

35. The method of any one of claims 30, 32, and 34, wherein the second solution
comprises a mixture of hydrogen peroxide (H₂O₂) and acetic acid (C₂H₄O₂), or a mixture of
20 hydrogen peroxide (H₂O₂) and formic acid (CH₂O₂).

36. The method of claim 20, further comprising infiltrating a polymer into each
cellulose-based fiber to form a composite material.

25 37. The method of claim 20, further comprising embedding each cellulose-based
fiber within a structural matrix to form a composite material.

38. The method of claim 37, wherein the structural matrix comprises a polymer or
concrete.

30 39. The method of any one of claims 36 and 38, wherein the polymer comprises
epoxy resin, polyvinyl alcohol (PVA), polyethylene glycol (PEO), polyamide (PA),
polyethylene terephthalate (PET), polybutylene terephthalate (PBT), polytrimethylene
terephthalate (PTT), polyacrylonitrile (PAN), polycaprolactam (PA6), poly(m-phenylene

isophthalamide) (PMIA), poly-p-phenylene terephthalamide (PPTA), polyurethane (PU), polycarbonate (PC), polypropylene (PP), high-density polyethylene (HDPE), polystyrene (PS), polycaprolactone (PCL), polybutylene succinate (PBS), polybutylene adipate terephthalate (PBAT), poly(butylene succinate-co-butylene adipate) (PBSA), polyhydroxybutyrate (PHB),
 5 poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV), poly(glycolic acid) (PGA), polypyrrole (PPy), polythiophene (PTh), polyvinylidene fluoride (PVDF), polyvinyl fluoride (PVF), ethylene vinyl alcohol (EVOH), poly(vinylidene chloride) (PVDC), polyxylylene adipamide (MXD6), polyethylene (PE), polyvinyl chloride (PVC), poly(methyl methacrylate) (PMMA), acrylonitrile butadiene styrene (ABS), polyimide (PI), polyethylenimine (PEI), polylactic acid
 10 (PLA), octadecyltrichlorosilane (OTS), polyoctahedral silsesquioxane (POSS), paramethylstyrene (PMS), polydimethylsiloxane (PDMS), poly(ethylene naphthalate (PEN), a graft copolymer of acrylonitrile-butadiene-styrene-methylmethacrylate (ABSM), dodecyltrimethoxysilane (DTMS), rosin, chitin, chitosan, protain, plant oil, carboxymethyl cellulose, cellulose acetate, starch, agar, alginic acid, or any combination of the foregoing.

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40. The method of claim 38, wherein the concrete comprises Portland cement, agro-concrete, magnesium carbonate (MgCO_3), magnesium hydroxide (Mg(OH)_2), or any combination of the foregoing.

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41. The method of any one of claims 36-37, wherein a content of the plurality of the cellulose-based fibers within the composite material is at least 0.1 wt%.

42. The method of claim 20, wherein the natural plant material is a species in the *Poales* order.

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43. The method of claim 20, wherein the natural plant material is a species in the *Poaceae* family.

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44. The method of claim 20, wherein the natural plant material is a natural grass.

45. The method of claim 20, wherein the natural plant material comprises bamboo, reed, sugarcane, maize, wheat, rice, jute, hemp, kenaf, ramie, banana, corn, wheat, rice, roselle, rattan, sorghum, sisal, palm, henequen, manila, pineapple, curaua, yucca, cabuya, screw pine, abaca, gladiolus, or agave.

46. The method of claim 26, wherein, after (c), each cellulose-based fiber has a length of at least 5 cm and a maximum cross-sectional dimension in a plane perpendicular to a direction of the length of at least 5 μm .

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47. The method of claim 46, wherein the length of each cellulose-based fiber after (b) is between 5 cm and 200 cm, inclusive.

48. The method of claim 46, wherein the maximum cross-sectional dimension of each cellulose-based fiber after (b) is between 5 μm and 1 mm, inclusive.

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49. The method of claim 46, wherein the maximum cross-sectional dimension of each cellulose-based fiber after (b) is at least 100 μm .

50. The method of claim 26, wherein, after (c), each cellulose-based fiber has a specific strength of at least 0.5 $\text{GPa}\cdot\text{cm}^3/\text{g}$, a crystallinity of at least 40%, a tensile strength of at least 0.5 GPa, a Young's modulus of at least 20 GPa, a density of at least 0.8 g/cm^3 , or any combination the foregoing.

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51. The method of claim 26, wherein, after (c), each cellulose-based fiber has a cellulose content of at least 70 wt%, a hemicellulose content less than or equal to 10 wt%, a lignin content less than or equal to 10 wt%, or any combination of the foregoing.

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52. The method of claim 20, further comprising providing on or within each cellulose-base fiber a conductive additive, a magnetic additive, a piezoelectric material, a stimuli-responsive material, a catalytic material, or any combination of the foregoing.

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53. An engineered plant material formed by the method of any one of claims 20-52.

54. A composite material formed by the method of any one of claims 20-52.

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