CLEAN CLAIM SET

1. A method comprising:

(a) dissolving lignin in a biomass comprising an intertwined structure of lignin, hemicellulose, and cellulose, such that the cellulose is fibrillated;

(b) after (a), *in situ* regenerating the lignin such that the regenerated lignin is deposited on and forms hydrogen bonds between the fibrillated cellulose, so as to form a slurry of lignin-cellulose solids in solution; and

(c) after (b), drying the slurry to form a solid lignocellulosic bioplastic.

2. The method of claim 1, wherein:

(a) comprises subjecting the biomass to a first chemical treatment by immersing the biomass in a first solution with one or more first chemicals, the first chemical treatment being effective to dissolve the lignin and to fibrillate the cellulose into microfibrils, nanofibrils, or both microfibrils and nanofibrils, and

(b) comprises:

(b1) after (a), adding one or more second chemicals to the first solution, such that the lignin is regenerated *in situ* from the one or more first chemicals, so as to form the lignin-cellulose solids within the first solution; and

(b2) after (b1), removing at least the one or more first chemicals from the first solution, so as to form a lignin-cellulose slurry.

3. The method of claim 1, further comprising:

after (b) and prior to (c), depositing the slurry in a mold or cast,

wherein the mold or cast defines a shape of the lignocellulosic bioplastic after (c).

4. The method of claim 1, wherein:

(c) comprises pressing the slurry; or

the method further comprises, after (c), pressing the solid lignocellulosic bioplastic to form a densified bioplastic.

5. The method of claim 1, further comprising:

after (b) and prior to (c), depositing the slurry using a printhead or additive manufacturing nozzle,

wherein locations of the depositing define a shape of the lignocellulosic bioplastic after (c).

6. The method of claim 1, wherein:

(c) comprises freeze drying or critical point drying, and

after (c), the bioplastic is formed as an aerogel.

7. The method of claim 1, wherein:

(c) comprises exchanging a first solvent of the solution with a different second solvent, and

the first solvent comprises water and the second solvent comprises an alcohol.

8. The method of claim 1, further comprising:

prior to (c), adding a polymer or a precursor thereof to the solution,

wherein, after (c), the solid bioplastic is a hybrid structure formed by a combination of lignin-cellulose solids and the polymer, and

the polymer comprises a natural resin or rosin, polymethyl methacrylate (PMMA), polyvinyl alcohol (PVA), polydimethylsiloxane (PDMS), polyethylene terephthalate (PET), polycarbonate (PC), polyethylene glycol (PEO), polyamide (PA), polyethylene terephthalate (PET), polybutylene terephthalate (PBT), polytrimethylene terephthalate (PTT), polyacrylonitrile (PAN), polycaprolactam (Nylon 6), 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), polyglycolide (PGA), poly(methyl methacrylate (PMMA), acrylonitrile butadiene styrene (ABS), polymethysilane (PMS), or any combination of the foregoing.

9. The method of claim 1, wherein after (b) and prior to (c), a content of lignin-cellulose solids in the slurry is in a range of 5 wt% to 20 wt%, inclusive.

10. The method of claim 1, wherein:

the biomass comprises a portion of plant material, and

the plant material comprises wood, bamboo, grass, hemp, or reed.

11. The method of claim 10, wherein: the portion is a mechanically-processed or waste portion of the plant material, and

the waste portion comprises a wood chip, wood powder, sawdust, bagasse, wheat straw, coconut shell, haulm, or corn stalk.

12. The method of claim 1, wherein, after (a):

the lignin in the slurry has β-O-4 ether bonds cleaved as compared to native lignin in the biomass prior to (a);

hydroxyl groups of the lignin are more phenolic than before (a); or

a -COO functional group of the cellulose has a negative charge.

13. The method of claim 1, wherein:

prior to (a), the intertwined structure of the biomass is comprised of microbundles having a cross-sectional dimension of at least 50 μm; and

after (a), the fibrillated cellulose is in a form of microfibrils or nanofibrils having a cross-sectional dimension less than or equal to 300 nm.

14. The method of claim 2, wherein:

the one or more first chemicals comprises an alkali solution,

the alkali solution comprises sodium hydroxide (NaOH), lithium hydroxide (LiOH), potassium hydroxide (KOH), sodium sulfite (Na_2SO_3), sodium sulfate (Na_2SO_4), sodium sulfide (Na_2S), Na_nS wherein n is an integer, urea (CH_4N_2O), sodium bisulfite ($NaHSO_3$), sulfur dioxide (SO_2), anthraquinone ($C_{14}H_8O_2$), ammonia (NH_3), methanol (CH_3OH), or any combination of the foregoing, and

the one or more second chemicals comprises an acid.

15. The method of claim 2, wherein:

The one or more first chemicals comprises an acid solution,

the acid solution comprises formic acid (CH_2O_2), acetic acid (CH_3COOH), methanol (CH_3OH), sodium chlorite ($NaClO_2$), chlorine dioxide (ClO_2), hydrochloric acid (HCl), sulfuric acid (H_2SO_4), or any combination of the foregoing, and

the one or more second chemicals comprises a base.

16. The method of claim 2, wherein the one or more first chemicals comprises an organic solvent, and

the organic solvent comprises formic acid (CH_2O_2), acetic acid (CH_3COOH), lactic acid ($CH_3CH(OH)COOH$), methanol (CH_3OH), ethanol (C_2H_5OH), butanol (C_4H_9OH), valerolactone ($C_5H_8O_2$), acetone (C_3H_6O) or any combination foregoing.

17. The method of claim 2, wherein the one or more first chemicals comprises a solution of choline chloride (ChCl) and oxalic acid ($C_2H_2O_4$).

18. The method of claim 2, wherein:

the one or more first chemicals comprises a deep eutectic solvent,

the deep eutectic solvent comprises choline chloride (ChCl), oxalic acid ($C_2H_2O_4$), lactic acid ($CH_3CH(OH)COOH$), glycerol ($C_3H_8O_3$), urea (CH_4N_2O), betaine ($C_5H_{11}NO_2$), zinc chloride ($ZnCl_2$), aluminum chloride ($AlCl_3$), or any combination of the foregoing, and

the one or more second chemicals comprises water.

19. The method of claim 2, wherein:

(a) further comprises maintaining the first solution with the biomass immersed therein at a first elevated temperature for a first time;

(b1) further comprises maintaining the first solution with the one or more second chemicals added thereto at a second elevated temperature for a second time; or

any combination of the foregoing.

20. The method of claim 1, wherein:

after (a), the hemicellulose in the biomass is also dissolved; and

after the *in situ* regenerating of (b), the hemicellulose remains at least partially dissolved.

21. The method of claim 1, wherein:

at least 90% of lignin in the biomass prior to (a) is retained in the slurry after (b);

less than or equal to 10% of hemicellulose in the biomass prior to (a) is retained in the slurry after (b); or

any combination of the foregoing.

22. The method of claim 2, wherein:

the removing of (b2) comprises filtering to separate the one or more first chemicals and at least some of the one or more second chemicals from the lignin-cellulose slurry, and

the method further comprises:

(d1) after (b2), separating the one or more first chemicals from the one or more second chemicals,

wherein:

the separated first chemicals are reused to dissolve lignin in another biomass;

the separated second chemicals are reused in another first solution for in situ regeneration of lignin;

the separating of (d1) comprises filtration, distillation, or both; or

any combination of the foregoing.

23. A bioplastic formed by the method of any one of claims 1-22.

24. A bioplastic comprising:

fibrillated cellulose in a form of microfibrils or nanofibrils having a cross-sectional dimension less than or equal to 300 nm; and

regenerated lignin deposited on and forming hydrogen bonds between the fibrillated cellulose so as to form an interconnected network,

wherein the regenerated lignin and the fibrillated cellulose are derived from a same biomass that had an intertwined structure of native lignin, hemicellulose, and cellulose, and

the regenerated lignin has been chemically modified as compared to the native lignin in the biomass.

25. The bioplastic of claim 24, wherein:

the regenerated lignin has β-O-4 ether bonds cleaved as compared to native lignin in the biomass;

the regenerated lignin has hydroxyl groups that are more phenolic as compared to hydroxyl groups of the native lignin in the biomass; or

-COO functional groups of the fibrillated cellulose have a negative charge.

26. The bioplastic of claim 24, wherein:

the biomass comprises a portion of plant material, and

the plant material comprises wood, bamboo, grass, hemp, or reed.

27. The bioplastic of claim 24, where the bioplastic is substantially devoid of any hemicellulose.

28. The bioplastic of claim 24, wherein the interconnected network forms an aerogel.

29. The bioplastic of claim 24, further comprising a polymer infiltrating or forming a part of the interconnected network, wherein the polymer comprises a natural resin or rosin, polymethyl methacrylate (PMMA), polyvinyl alcohol (PVA), polydimethylsiloxane (PDMS), polyethylene terephthalate (PET), polycarbonate (PC), polyethylene glycol (PEO), polyamide (PA), polyethylene terephthalate (PET), polybutylene terephthalate (PBT), polytrimethylene terephthalate (PTT), polyacrylonitrile (PAN), polycaprolactam (Nylon 6), 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), polyglycolide (PGA), poly(methyl methacrylate (PMMA), acrylonitrile butadiene styrene (ABS), polymethysilane (PMS), or any combination of the foregoing.

30. The bioplastic of claim 24, wherein the bioplastic consists of the fibrillated cellulose and the regenerated lignin.

31. A structure comprising the bioplastic of any of claims 24-30.

32. The structure of claim 31, further comprising:

a coating disposed on one or more exterior surfaces of the bioplastic;

a sub-structure coupled to the bioplastic, the sub-structure having a different material composition from the bioplastic; or

any combination of the foregoing.

33. A slurry comprising:

a solution;

fibrillated cellulose within the solution and in a form of microfibrils or nanofibrils having a cross-sectional dimension less than or equal to 300 nm; and

regenerated lignin within the solution, the regenerated lignin being deposited on and forming hydrogen bonds between the fibrillated cellulose,

wherein the regenerated lignin and the fibrillated cellulose are derived from a same biomass that had an intertwined structure of native lignin, hemicellulose, and cellulose, and

the regenerated lignin has been chemically modified as compared to the native lignin in the biomass.

34. The slurry of claim 33, wherein:

the regenerated lignin has β-O-4 ether bonds cleaved as compared to native lignin in the biomass;

the regenerated lignin has hydroxyl groups that are more phenolic as compared to hydroxyl groups of the native lignin in the biomass;

-COO functional groups of the fibrillated cellulose have a negative charge; or

any combination of the foregoing.

35. The slurry of claim 33, further comprising a polymer or a precursor thereof within the solution, wherein the polymer comprises a natural resin or rosin, polymethyl methacrylate (PMMA), polyvinyl alcohol (PVA), polydimethylsiloxane (PDMS), polyethylene terephthalate (PET), polycarbonate (PC), polyethylene glycol (PEO), polyamide (PA), polyethylene terephthalate (PET), polybutylene terephthalate (PBT), polytrimethylene terephthalate (PTT), polyacrylonitrile (PAN), polycaprolactam (Nylon 6), 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), polyglycolide (PGA), poly(methyl methacrylate (PMMA), acrylonitrile butadiene styrene (ABS), polymethysilane (PMS), or any combination of the foregoing.