CLAIMS

1. A composite comprising a first polymeric macromolecular species or polymer and a second polymeric macromolecular species or polymer, wherein one or more of the following applies:
    i. a portion of the first polymeric macromolecular species or polymer and a portion of the second polymeric macromolecular species or polymer are physically and/or chemically entangled;
    ii. a portion of the first polymeric macromolecular species or polymer are physically and/or chemically crosslinked;
    iii. a portion of the second polymeric macromolecular species or polymer are physically and/or chemically crosslinked;
    iv. a portion of the first polymeric macromolecular species or polymer are chemically and/or physically integrated into a portion of the second polymeric macromolecular species or polymer;
    v. a portion of the first polymeric macromolecular species or polymer and a portion of the second polymeric macromolecular species or polymer are not separable;
    vi. a portion of the first polymeric macromolecular species or polymer and/or a portion of the second polymeric macromolecular species or polymer are cross-linked;
    vii. a portion of the first polymeric macromolecular species or polymer and/or a portion of the second polymeric macromolecular species or polymer are partially organized and/or crystallized;
    viii. a portion of the first polymeric macromolecular species or polymer and a portion of the second polymeric macromolecular species or polymer cannot be delaminated; and/or
    ix. a portion of the first polymeric macromolecular species or polymer and a portion of the second polymeric macromolecular species or polymer are self-assembled.

2. The composite of claim 1, wherein a portion of the first polymeric macromolecular species or polymer in the composite has a second structure different than a first structure of the first polymeric macromolecular species or polymer, and/or a portion of the second polymeric macromolecular species or polymer in the composite has a second structure different than a first structure of the second polymeric macromolecular species or polymer.

3. The composite of claim 1 or 2, wherein the first polymeric macromolecular species or polymer comprises a protein component, and

wherein the protein component comprises one or more of silk fibroin proteins or fragments, collagen, elastin, gelatin, corn zein, wheat gluten, pectin, chitin, casein, and/or whey.

4. The composite of claim 1 or 2, wherein the first polymeric macromolecular species or polymer comprises a biodegradable polymer, and

wherein the first polymeric macromolecular species or polymer comprises one or more of a polyurethane component.

5. The composite of claim 1 or 2, wherein the first polymeric macromolecular species or polymer comprises a poly lactic acid (PLA) component, a poly(lactic-*co*-glycolic acid) (PLGA) component, or both.

6. The composite of any one of claims 1 to 5, wherein the second polymeric macromolecular species or polymer comprises a cellulose and/or cellulose derivative component.

7. The composite of claim 6, wherein the cellulose derivative is selected from methyl cellulose, ethyl cellulose, ethyl methyl cellulose, hydroxyethyl methyl cellulose, hydroxypropyl methyl cellulose, ethyl hydroxyethyl cellulose, cellulose triacetate, cellulose propionate, cellulose nitrate, cellulose sulfate, carboxymethyl cellulose, hydroxyethyl cellulose, hydroxypropyl cellulose, cellulose acetate, cellulose acetate propionate, cellulose acetate butyrate, and microcrystalline cellulose.

8. The composite of claim 7, wherein the ethyl cellulose has at least one of the following properties:

an ethoxyl content from 45.0% to 49.5%, from 45.0% to 46.0%, from 45.0% to 47.0%, from 47.0% to 48.0%, or from 48.0% to 49.5%; and/or

a degree of substitution of 0.5 to 1, 1 to 1.5, 1.5 to 2, 2 to 2.5, or 2.5 to 3.

9. The composite of any one of claims 6 to 8, wherein a second structure of the cellulose derivative comprises a degree of crystallinity of between about 5% and less than about 100%.

10. The composite of any one of claims 1 to 9, wherein the w/w ratio between the first polymeric macromolecular species or polymer and the second polymeric macromolecular species polymer in the composite is between about 1:100 and about 100:1.

11. The composite of any one of claims 1 to 10, wherein the first polymeric macromolecular species or polymer is distributed isotropically or anisotropically over a cross section of the composite.

12. The composite of claim 11, wherein a concentration of the first polymeric macromolecular species or polymer closer to a first surface of the composite is higher than a concentration of the first polymeric macromolecular species or polymer closer to a second surface of the composite.

13. The composite of any one of claims 1 to 12, wherein the second polymeric macromolecular species or polymer is distributed isotropically or anisotropically over a cross section of the composite.

14. The composite of claim 1, wherein a concentration of the second polymeric macromolecular species or polymer closer to a second surface of the composite is higher than a concentration of the second polymeric macromolecular species or polymer closer to a first surface of the composite, and/or

wherein the second polymeric macromolecular species or polymer is substantially undetectable at a first surface of the composite substrate-coating interface.

15. The composite of any one of claims 1 to 14, wherein a first surface and/or a second surface of the composite is adhesive.

16. The composite of any one of claims 1 to 13, wherein the composite has an increased water resistance compared to one of: i) a non-composite material comprising the first polymeric macromolecular species or polymer, but excluding the second polymeric macromolecular species or polymer, ii) a non-composite material comprising the second polymeric macromolecular species or polymer, but excluding the first polymeric macromolecular species or polymer, or iii) a non-composite material comprising the first polymeric macromolecular species or polymer and the second polymeric macromolecular species or polymer, wherein the polymeric macromolecular species or polymers are not physically and/or chemically molecularly entangled.

17. The composite of any one of claims 1 to 13, wherein the composite has an increased water vapor permeability compared to one of: i) a non-composite material comprising the first polymeric macromolecular species or polymer, but excluding the second polymeric macromolecular species or polymer, ii) a non-composite material comprising the second polymeric macromolecular species or polymer, but excluding the first polymeric macromolecular species or polymer, or iii) a non-composite material comprising the first polymeric macromolecular species or polymer and the second polymeric macromolecular species or polymer, wherein the polymeric macromolecular species or polymers are not physically and/or chemically molecularly entangled.

18. An article comprising a substrate and a coating, the coating comprising the composite of any one of claims 1 to 17, wherein one or more of the following applies:.

i) the substrate comprises an irregular surface;

ii) the coating has a thickness between about 10 μm and about 1000 μm;

iii) the amount of coating on the substrate is between about 0.01 g/ft^2 and about 25 g/ft^2;

iv) the amount of first polymeric macromolecular species or polymer in the coating on the substrate is between about 0.001 g/ft^2 and about 20 g/ft^2;

v) the amount of second polymeric macromolecular species or polymer in the coating on the substrate is between about 0.001 g/ft^2 and about 15 g/ft^2;

vi) the substrate comprises a substantially flexible material; vii) the substrate comprises a leather material or a textile material;

viii) the substrate comprises one or more of collagen, cellulose, and/or lignin.

19. A method of coating a substrate, the method comprising applying to a surface of the substrate a first composition comprising a first polymeric macromolecular species or polymer, and a second composition comprising a second polymeric macromolecular species or polymer.

20. The method of claim 19, wherein the first composition comprises an unstructured first polymeric macromolecular species or polymer, or a first structure of the first polymeric macromolecular species or polymer.

21. The method of claim 19 or 20, wherein the first polymeric macromolecular species or polymer comprises a protein component, preferably

wherein the protein component comprises one or more of silk fibroin proteins or fragments, collagen, elastin, gelatin, corn zein, wheat gluten, pectin, chitin, casein, and/or whey.

22. The method of claim 19 or 20, wherein the first polymeric macromolecular species or polymer comprises a biodegradable polymer, preferably

wherein the first polymeric macromolecular species or polymer comprises one or more of a polyurethane component.

23. The method of any one of claims 19 to 22, wherein the first polymeric macromolecular species or polymer comprises a poly lactic acid (PLA) component, a poly(lactic-*co*-glycolic acid) (PLGA) component, or both.

24. The method of any one of claims 19 to 23, wherein the second composition comprises an unstructured second polymeric macromolecular species or polymer, or a first structure of the second polymeric macromolecular species or polymer, preferably

wherein the second polymeric macromolecular species or polymer comprises a cellulose and/or cellulose derivative component.

25. The method of claim 24, wherein the cellulose derivative is selected from methyl cellulose, ethyl cellulose, ethyl methyl cellulose, hydroxyethyl methyl cellulose, hydroxypropyl methyl cellulose, ethyl hydroxyethyl cellulose, cellulose triacetate, cellulose propionate, cellulose nitrate, cellulose sulfate, carboxymethyl cellulose, hydroxyethyl cellulose, hydroxypropyl cellulose, cellulose acetate, cellulose acetate propionate, cellulose acetate butyrate, and microcrystalline cellulose.

26. The method of claim 25, wherein the ethyl cellulose has at least one of the following properties:

an ethoxyl content from 45.0% to 49.5%, from 45.0% to 46.0%, from 45.0% to 47.0%, from 47.0% to 48.0%, or from 48.0% to 49.5%; and/or

a degree of substitution of 0.5 to 1, 1 to 1.5, 1.5 to 2, 2 to 2.5, or 2.5 to 3.

27. The method of any one of claims 24 to 26, wherein the cellulose derivative comprises a first structure of the cellulose derivative having a degree of crystallinity lower than a second structure of the cellulose derivative comprising a degree of crystallinity of between about 5% and less than about 100%.

28. The method of any one of claims 19 to 27, wherein the second composition comprising a second polymeric macromolecular species or polymer further comprises a solvent component.

29. The method of claim 27 or 28, wherein the solvent component comprises one or more of an alcohol, an ether, a ketone, an aldehyde, and/or a ketal,

wherein the solvent component is from about 75% w/w to about 99% w/w of the composition, from about 80% w/w to about 98% w/w of the composition, from about 85% w/w to about 97.5% w/w of the composition, or from about 85% w/w to about 95% w/w of the composition, preferably

wherein the solvent component comprises one or more of methanol, ethanol, n-propanol, 2-propanol, n-butanol, 2-butanol, pentanol, hexanol, acetone, butanone, methoxypropanol, di-isopropylidene glycerol, 2,2-dimethyl-4-hydroxymethyl-1,3-dioxolane, 2,2-dimethyl-1,3-dioxolane-4-methanol, or any combination thereof.

30. The method of any one of claims 19 to 29, wherein a first composition comprising a first polymeric macromolecular species or polymer further comprises one or more of a polyethylene glycol (PEG) component, a polypropylene glycol (PPG) component, a polyether component, a fatty acid or fatty acid derived amide, and/or a monoglyceride, diglyceride, a triglyceride, a triethylene glycol monomethyl ether component, a diethylene glycol butyl ether component, a diethylene glycol ethyl ether component, a dimethyl tetradecanedioate component, an erucamide component, a glyceryl stearate component, an isocyanate component, a polyol component, a blocked isocyanate component, a blocked polyol component, a partially polymerized, partially crosslinked, a partially cured polyurethane component, and/or a polyurethane prepolymer component.

31. The method of any one of claims 19 to 30, wherein a surface of the substrate is coated first with the first composition comprising a first polymeric macromolecular species or polymer, and the coated with the second composition comprising a second polymeric macromolecular species or polymer.

32. The method of claim 31, further comprising a drying or partial drying step between the two coating steps.

33. The method of claim 31 or 32, wherein the first composition comprising a first polymeric macromolecular species or polymer is only partially polymerized, partially dried, and/or partially cured before the second composition comprising a second polymeric macromolecular species or polymer is applied, and/or

wherein the second composition comprising a second polymeric macromolecular species or polymer is applied at a temperature above a glass transition temperature (T_g) of the first polymeric macromolecular species or polymer or is applied at a temperature above a glass transition temperature (T_g) of the second polymeric macromolecular species or polymer.

34. The method of any one of claims 19 to 33, wherein the first composition comprising a first polymeric macromolecular species or polymer is applied one or more times at a rate from about 0.5 mL/ft^2 to about 5 mL/ft^2, and/or

wherein the second composition comprising a second polymeric macromolecular species or polymer is applied one or more times at a rate from about 0.5 mL/ft^2 to about 5 mL/ft^2.

35. An article comprising a substrate and a coating, the article made by a method of any one of claims 19 to 34.

36. The article of claim 35, wherein the first polymeric macromolecular species or polymer is distributed isotropically or anisotropically over a cross section of the coating from a substrate-coating interface to an external surface of the coating.

37. The article of claim 35, wherein a concentration of the first polymeric macromolecular species or polymer closer to a substrate-coating interface is higher than a concentration of the first macromolecular species or polymer closer to an external surface of the coating, and / or

wherein the first polymeric macromolecular species or polymer is substantially undetectable at an external surface of the coating.

38. The article of any one of claims 35 to 37, wherein the second polymeric macromolecular species or polymer is distributed isotropically or anisotropically over a cross section of the coating from a substrate-coating interface to an external surface of the coating.

39. The article of any one of claims 35 to 37, wherein a concentration of the second polymeric macromolecular species or polymer closer to a substrate-coating interface is lower than a concentration of the second polymeric macromolecular species or polymer closer to an external surface of the coating, and/ or wherein the second polymeric macromolecular species or polymer is substantially undetectable at a substrate-coating interface.