CLAIMS

1. A plurality of substantially solid silk fibroin particles comprising silk fibroin fragments, the particles being characterized by at least one of: bulk density, surface area, pore size, pore volume, aspect ratio, and/or Hausner ratio, wherein the substantially solid silk fibroin particles have a bulk density selected from about 0.03 g/ml to about 0.35 g/ml.

2. The plurality of substantially solid silk fibroin particles of claim 1, wherein at least one of the following applies:

i. the substantially solid silk fibroin particles are annealed;
ii. the substantially solid silk fibroin particles are grounded;
iii. the substantially solid silk fibroin particles have a Hausner ratio of between 1.00 and 1.11, between 1.12 and 1.18, between 1.19 and 1.25, between 1.26 and 1.34, or between 1.35 and 1.45;
iv. the substantially solid silk fibroin particles have an average diameter of between about 3 mm and about 10 mm;
v. the substantially solid silk fibroin particles have an aspect ratio between 1 and about 1.45;
vi. the substantially solid silk fibroin particles are substantially spherical;
vii. wherein the substantially solid silk fibroin particles are mesoporous;
viii. wherein the substantially solid silk fibroin particles have a BET (Brunauer–Emmett–Teller) surface area between about 2.50 m2/g and about 6.50 m2/g;
ix. the substantially solid silk fibroin particles have an average pore size between about 25 Å and about 500 Å; or
x. wherein the substantially solid silk fibroin particles comprise a plurality of radially orientated microchannels.

3. The plurality of substantially solid silk fibroin particles of claim 1 or 0, wherein the substantially solid silk fibroin particles further comprise one or more of the following:

i. an emulsifier, a surfactant, a buffering agent, an amino acid, or a sugar;
ii. a polysaccharide, a polysorbate, a glycoside, PBS, arginine, trehalose, glucose, or sucrose;
iii. a surfactant selected from sucrose ester, cetearyl glucoside, caprylyl/capryl glucoside, sucrose laurate, sucrose palmitate, sucrose stearate, sucrose cocoate, sorbitan monostearate, and combinations thereof; or
iv. an additional protein or peptide, a C12-C24 fatty alcohol, a glycolipid, or a lipid.

4. The plurality of substantially solid silk fibroin particles o of claim 1 or 0, wherein one or more of the following applies:

v. the substantially solid silk fibroin particles further comprise a protein, a peptide, a sugar surfactant, a biosurfactant, a lipid, or a combination;
vi. the substantially solid silk fibroin particles further comprise a sugar fatty acid ester, a sugar fatty acid monoester, a sugar fatty diester, a sugar fatty triester, or a sugar fatty and polyester;
vii. the substantially solid silk fibroin particles further comprise a sucrose fatty acid ester, a sorbitan or sorbitol fatty acid ester, an alkyl glucoside, an alkyl polyglucoside, or a combination thereof;
viii. the substantially solid silk fibroin particles further comprise KCl, NaCl, MgCl2, CaCl2, PBS, Tris, Polysorbate 20, Polysorbate 80, Capryl Glucoside, Sucrose, Histidine, Glycine, or Arginine.

5. A silk fibroin nanoclay composite or film, comprising silk fibroin fragments and a clay, wherein the % (w/w) of clay in the composite is from about 1% to about 99%.

6. The nanoclay composite or film of claim 0, wherein the clay is a bentonite clay, and

wherein the concentration of clay in the composite or film is from about 20% (w/w) to about 33% (w/w), from about 33% (w/w) to about 50% (w/w), or from about 50% (w/w) to about 67% (w/w).

7. The nanoclay composite or film of claim 0 or 0, wherein the water vapor permeance (WVP) of the composite is inversely proportional to the concentration of clay in the composite, and

wherein water vapor permeance (WVP, g/m2*Pa*24h) of the composite is from about 0.20 to about 0.30, from about 0.30 to about 0.35, from about 0.35 to about 0.40, from about 0.40 to about 0.45, from about 0.45 to about 0.50, from about 0.50 to about 0.55, from about 0.55 to about 0.60, from about 0.60 to about 0.65, from about 0.65 to about 0.70, from about 0.70 to about 0.75, from about 0.75 to about 0.80, or from about 0.80 to about 0.85.

8. A stabilized silk fibroin solution comprising silk fibroin fragments and a stabilizer, wherein:

the solution has a z-average value lower than a substantially similar silk fibroin solution comprising silk fibroin fragments but excluding the stabilizer; and/or

the solution has a z-average plateau value lower than a substantially similar silk fibroin solution comprising silk fibroin fragments but excluding the stabilizer.

9. The stabilized silk fibroin solution of claim 0, wherein the z-average is measured after a period of time after the silk fibroin fragments and the stabilizer are co-formulated, wherein the period time ranges from 1 minute to 10 minutes, from 1 minute to 20 minutes, from 1 minute to 30 minutes, from 1 minute to 40 minutes, from 1 minute to 50 minutes, from 1 minute to 60 minutes, from 1 minute to 70 minutes, from 1 minute to 80 minutes, from 1

hour to 250 hours, from 1 hour to 350 hours, from 1 hour to 450 hours, from 1 hour to 550 hours, from 1 hour to 650 hours, or from 1 hour to 1000 hours.

10. The stabilized silk fibroin solution of claim 0 or 0, wherein one or more of the following applies:

ix. the stabilizer is an emulsifier, a surfactant, a buffering agent, an amino acid, or a sugar;

x. the stabilizer is a polysaccharide, a polysorbate, a glycoside, PBS, arginine, trehalose, glucose, or sucrose;

xi. the stabilizer is a surfactant selected from sucrose ester, cetearyl glucoside, caprylyl/capryl glucoside, sucrose laurate, sucrose palmitate, sucrose stearate, sucrose cocoate, sorbitan monostearate, and combinations thereof;

xii. the stabilizer is an additional protein or peptide, a C12-C24 fatty alcohol, a glycolipid, or a lipid;

xiii. the stabilizer is a protein, a peptide, a sugar surfactant, a biosurfactant, a lipid, or a combination;

xiv. the stabilizer is a sugar fatty acid ester, a sugar fatty acid monoester, a sugar fatty diester, a sugar fatty triester, or a sugar fatty and polyester;

xv. the stabilizer is a sucrose fatty acid ester, a sorbitan or sorbitol fatty acid ester, an alkyl glucoside, an alkyl polyglucoside, or a combination thereof; or

xvi. the stabilizer is KCl, NaCl, MgCl2, CaCl2, PBS, Tris, Polysorbate 20, Polysorbate 80, Capryl Glucoside, Sucrose, Histidine, Glycine, or Arginine.

11. The stabilized silk fibroin solution of any one of claims 0 to xvi, wherein the solution is sprayable.

12. A liquid in air suspension comprising a plurality of droplets comprising the stabilized silk fibroin solution of claim 0, wherein the droplets are sufficiently stable after being sprayed, for a period of time necessary to reach a surface.

13. A plurality of drops or droplets comprising the stabilized silk fibroin solution of any one of claims 0 to xvi, wherein the droplets or drops are sufficiently stable after being formed, for a period of time necessary to reach a surface.

14. The plurality of substantially solid silk fibroin particles of any one of claims 1 to 0, the silk fibroin nanoclay composite or film of any one of claims 0 to 0, the stabilized silk fibroin solution of any one of claims 0 to 0, the liquid in air suspension of claim 0, or the plurality of drops or droplets of claim 0, wherein the silk fibroin fragments have a weight average molecular weight selected from between about 1 kDa and about 5 kDa, from between about 5 kDa and about 10 kDa, from between about 6 kDa and about 17 kDa, from between about 10 kDa and about 15 kDa, from between about 14 kDa and about 30 kDa, from between about 15 kDa and about 20 kDa, from between about 17 kDa and about 39 kDa, from between about 20 kDa and about 25 kDa, from between about 25 kDa and about 30 kDa, from between about 30 kDa and about 35 kDa, from between about 35 kDa and about 40 kDa, from between about 39 kDa and about 54 kDa, from between about 39 kDa and about 80 kDa, from between about 40 kDa and about 45 kDa, from between about 45 kDa and about 50 kDa, from between about 50 kDa and about 55 kDa, from between about 55 kDa and about 60 kDa, from between about 60 kDa and about 100 kDa, from between about 80 kDa and about 144 kDa, from between about 144 kDa and about 250 kDa, or from between about 250 kDa and about 350 kDa, and a polydispersity from 1 to about 5.

15. The plurality of substantially solid silk fibroin particles, the silk fibroin nanoclay composite or film, the stabilized silk fibroin solution, the liquid in air suspension, or the plurality of drops or droplets, of claim 0, further comprising about 0.001% (w/w) to about 10% (w/w) sericin relative to the silk fibroin fragments.

16. The plurality of substantially solid silk fibroin particles, the silk fibroin nanoclay composite or film, the stabilized silk fibroin solution, the liquid in air suspension, or the plurality of drops or droplets, of any one of claims 0 to 0, wherein the silk fibroin fragments do not spontaneously or gradually gelate and do not visibly change in color or turbidity when in an aqueous solution for at least 10 days prior to being formulated into the substantially solid silk fibroin particles, the silk fibroin nanoclay composite or film, or the stabilized silk fibroin solution.

17. The plurality of substantially solid silk fibroin particles, the silk fibroin nanoclay composite or film, the stabilized silk fibroin solution, the liquid in air suspension, or the plurality of drops or droplets, of claim 0, wherein the fibroin is a fibroin heavy chain, a fibroin light chain, or a fibrohexamerin wherein:

one or more fibroin heavy chain protein fragments comprise at least one amino acid modification at an amino acid position corresponding to an amino acid residue in SEQ ID NO: 49 selected from Q58E, M64X, N68D, N70D, N77D, M80X, N93D, M103X, Q125E, N132D, Q139E, Q275E, N4191D, Q5216E, and N5262D;

one or more fibroin light chain protein fragments comprise at least one amino acid modification at an amino acid position corresponding to an amino acid residue in SEQ ID NO: 50 selected from N23D, Q24E, N28D, M69X, N105D, N108D, N118D, N136D, N138D, Q149E, N186D, N200D, Q202E, N204D, N240D, N248D, and Q255E; and

one or more fibrohexamerin protein fragments comprise at least one amino acid modification at an amino acid position corresponding to an amino acid residue in SEQ ID NO: 51 selected from Q62E, N93D, M120X, N149D, N172D, N174D, and N202D,

wherein X is methionine oxide.

18. The plurality of substantially solid silk fibroin particles, the silk fibroin nanoclay composite or film, the stabilized silk fibroin solution, the liquid in air suspension, or the

plurality of drops or droplets, of claim 0, wherein each modification, substitution, and/or replacement is independently ranging between about 1% to about 99% in the silk fibroin fragments portion of the substantially solid silk fibroin particles, the silk fibroin nanoclay composite or film, or the stabilized silk fibroin solution composition.

19. The plurality of substantially solid silk fibroin particles, the silk fibroin nanoclay composite or film, the stabilized silk fibroin solution, the liquid in air suspension, or the plurality of drops or droplets, of claim 0, wherein a % modification, substitution, and/or replacement is defined as (number of peptide or protein fragments comprising a modification, substitution, and/or replacement at a specific position, divided by the total number of peptide or protein fragments which include the specific position, whether comprising a modification, substitution, and/or replacement, or not) x 100.

20. The plurality of substantially solid silk fibroin particles of any one of claims 1 to 0, the silk fibroin nanoclay composite or film of any one of claims 5 to 7, the stabilized silk fibroin solution of any one of claims 0 to 0, the liquid in air suspension of claim 0, or the plurality of drops or droplets of claim 0, wherein the silk fibroin fragments are included in one or more fractions, each fraction independently comprising a plurality of fibroin heavy chain fragments, a plurality of fibroin light chain fragments, and/or a plurality of fibrohexamerin (p25) fragments, wherein:

one or more fibroin heavy chain protein fragments comprise at least one amino acid modification at an amino acid position corresponding to an amino acid residue in SEQ ID NO: 49 selected from Q58E, M64X, N68D, N70D, N77D, M80X, N93D, M103X, Q125E, N132D, Q139E, Q275E, N4191D, Q5216E, and N5262D;

one or more fibroin light chain protein fragments comprise at least one amino acid modification at an amino acid position corresponding to an amino acid residue in SEQ ID NO: 50 selected from N23D, Q24E, N28D, M69X, N105D, N108D, N118D, N136D, N138D, Q149E, N186D, N200D, Q202E, N204D, N240D, N248D, and Q255E; and

one or more fibrohexamerin protein fragments comprise at least one amino acid modification at an amino acid position corresponding to an amino acid residue in SEQ ID NO: 51 selected from Q62E, N93D, M120X, N149D, N172D, N174D, and N202D,

wherein X is methionine oxide.

21. The plurality of substantially solid silk fibroin particles, the silk fibroin nanoclay composite or film, the stabilized silk fibroin solution, the liquid in air suspension, or the plurality of drops or droplets, of claim 0, wherein the silk fibroin fragments have a weight average molecular weight (Mw) selected from between about 1 kDa and about 20 kDa, from between about 20 kDa and about 40 kDa, from between about 40 kDa and about 60 kDa, from between about 60 kDa and about 80 kDa, from between about 80 kDa and about 100 kDa, from between about 100 kDa and about 120 kDa, from between about 120 kDa and about 140 kDa, or from between about 140 kDa and about 160 kDa, from between about 160 kDa and about 180 kDa, from between about 180 kDa and about 200 kDa, or from between about 200 kDa and about 250 kDa, and a polydispersity between 1 and about 1.7.

22. The plurality of substantially solid silk fibroin particles, the silk fibroin nanoclay composite or film, the stabilized silk fibroin solution, the liquid in air suspension, or the plurality of drops or droplets, of claim 0 or 0, wherein each modification, substitution, and/or replacement is independently ranging in the composition between about 1% to about 99%.

23. The plurality of substantially solid silk fibroin particles, the silk fibroin nanoclay composite or film, the stabilized silk fibroin solution, the liquid in air suspension, or the plurality of drops or droplets, of claim 0, wherein the % modification, substitution, and/or replacement is defined as (number of peptide or protein fragments comprising a modification, substitution, and/or replacement at a specific position, divided by the total number of peptide or protein fragments which include the specific position, whether comprising a modification, substitution, and/or replacement, or not) x 100.

24. The plurality of substantially solid silk fibroin particles, the silk fibroin nanoclay composite or film, the stabilized silk fibroin solution, the liquid in air suspension, or the plurality of drops or droplets, of any one of claims 0 to 0, wherein a molecular weight is determined by MALS.

25. A pouch or a laundry pod comprising the plurality of substantially solid silk fibroin particles of any one of claims 1 to 0.

26. A method of making the plurality of substantially solid silk fibroin particles of any one of claims 1 to 0 or 0-0, the method comprising dripping a solution comprising a plurality of the silk fibroin fragments into liquid nitrogen.

27. The method of claim 0, further comprising a lyophilization step.

28. The method of claim 0 or 0, wherein the concentration of silk fibroin fragments in the solution is from about 3% (w/w) to about 50% (w/w).

29. The method of any one of claims 0 to 0, wherein the silk fibroin fragments comprise one or more of a molecular weight, polydispersity, and/or a modification, substitution, and/or replacement at a specific amino acid position, as defined in claim 0 or 0.

30. The method of any one of claims 0 to 0, wherein the solution is stabilized as defined in any one of claims 0 to 0.

31. The plurality of substantially solid silk fibroin particles of any one of claims 1 to 0, wherein the particles have a reconstitution yield in DI water of more than 90%.

32. The method of claim 0, or the plurality of substantially solid silk fibroin particles of claim 0, wherein a reconstitution rate of at least 90% is preserved after a stability testing comprising simulated ageing of a plurality of substantially solid silk fibroin particles of any one of claims 1 to 0, the ageing comprising storage a temperature between about 40 °C and about 60 °C, for a period of time ranging from about 400 days to about 650 days.

33. The method of claim 0, wherein the simulated age ranges from about 4 years to about 15 years.