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(54) Titre : MATERIAUX COMPOSITES CARBONES A MORPHOLOGIE DE BOULE DE NEIGE
(54) Title: CARBONACEOUS COMPOSITE MATERIALS WITH SNOWBALL-LIKE MORPHOLOGY

Figure 3: Scanning electron microscopy (SEM) images of graphitic carbon powder **6**

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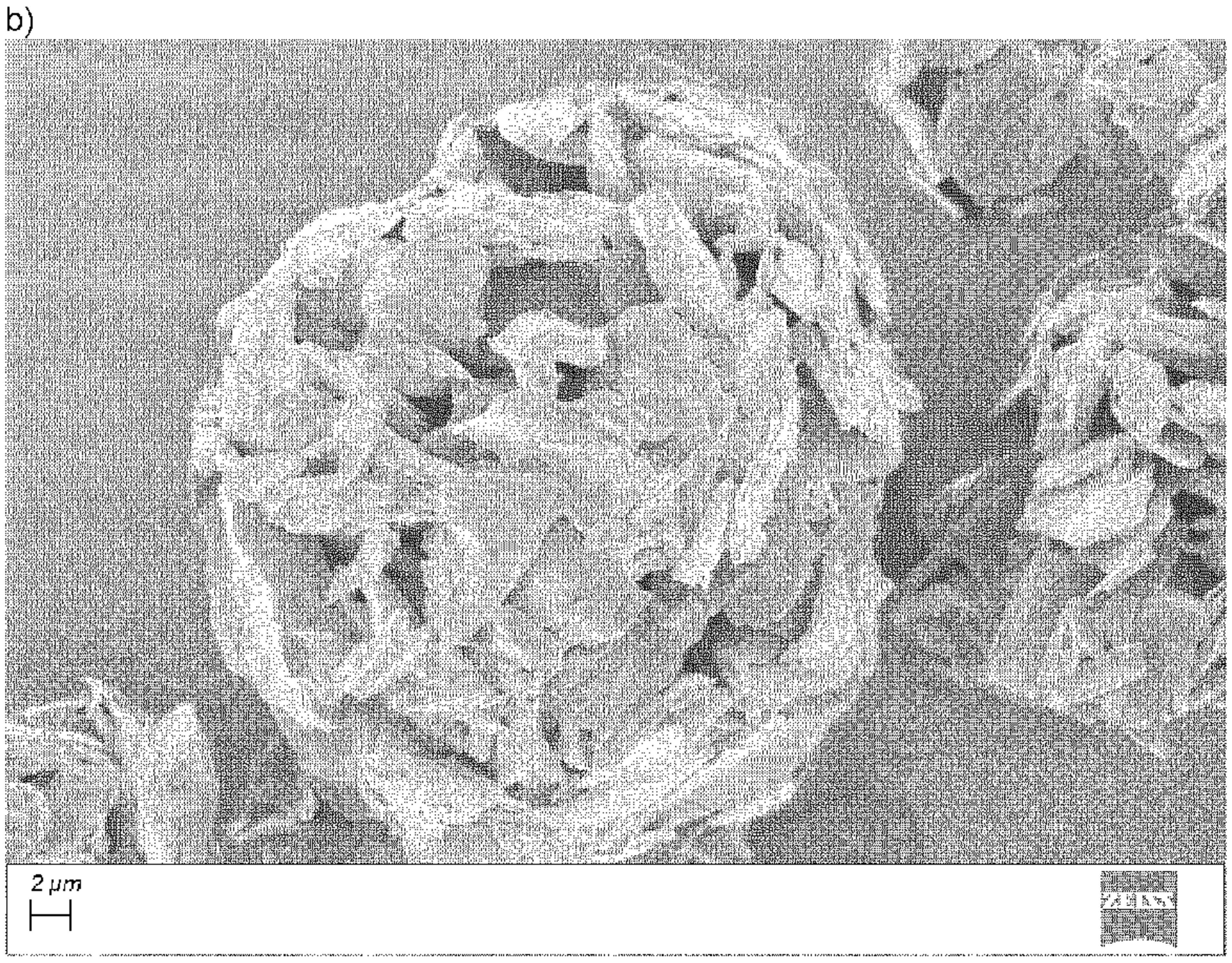

(57) Abrégé/Abstract:

The present disclosure relates to a novel process for preparing isotropic carbonaceous composite particles with favorable crystallographic, morphological & mechanical properties, wherein relatively fine carbonaceous primary particles are coated with a carbonaceous binder precursor material, agglomerated and finally heat-treated at temperatures of between about 1850 and 3500°C to convert the binder precursor material to non-graphitic or graphitic carbon, thereby resulting in stable highly isotropic carbonaceous composite materials wherein the primary particles of the aggregate are held together by the carbonized/graphitized binder. The present disclosure also relates to the isotropic carbonaceous composite particles obtainable by the process described herein. The disclosure further relates to uses of said isotropic carbonaceous composite material in various applications, including as active material in negative electrodes in lithium-ion batteries, and in secondary products containing said isotropic carbonaceous composite material.

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(54) **Title**: CARBONACEOUS COMPOSITE MATERIALS WITH SNOWBALL-LIKE MORPHOLOGY

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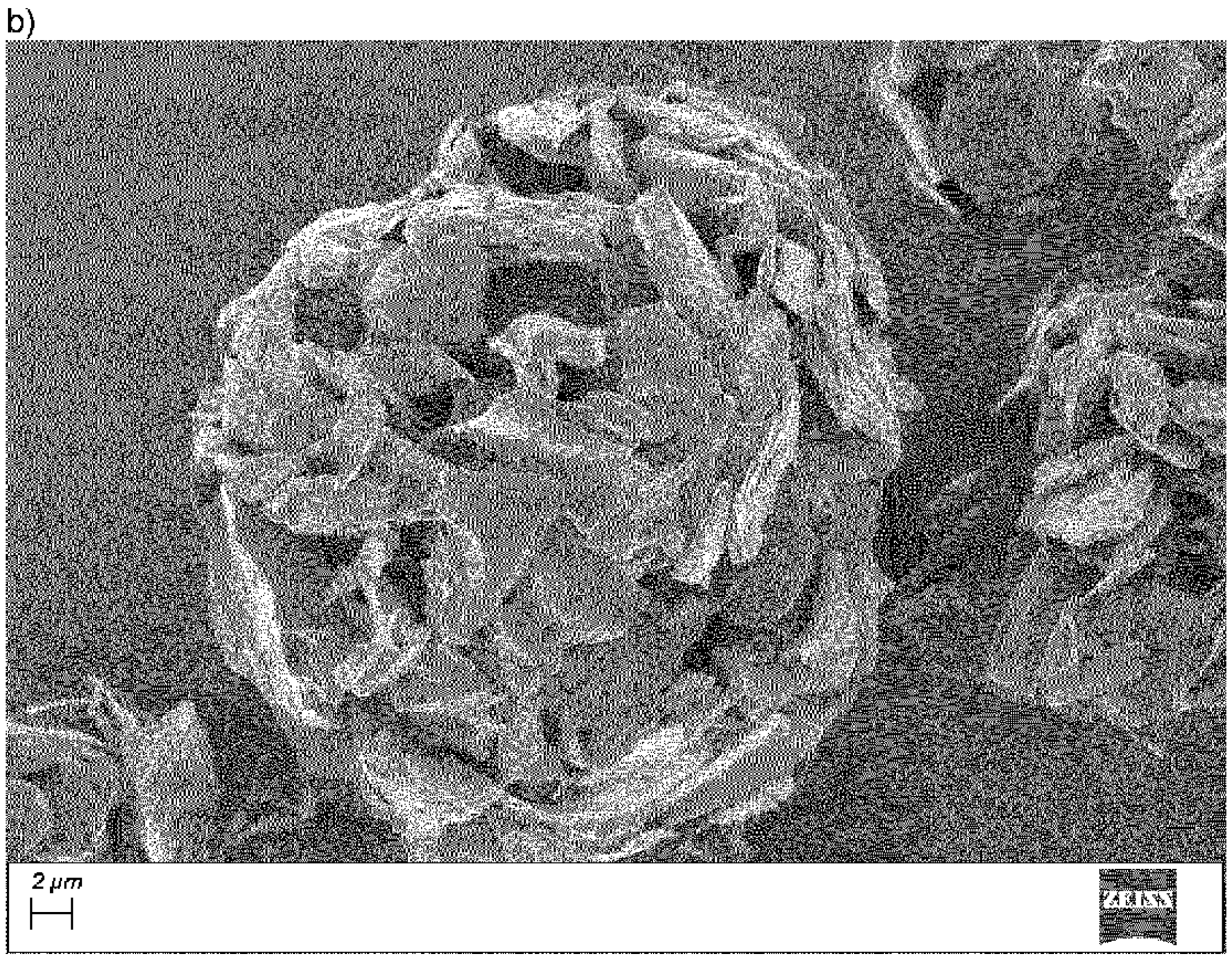

(57) **Abstract**: The present disclosure relates to a novel process for preparing isotropic carbonaceous composite particles with favorable crystallographic, morphological & mechanical properties, wherein relatively fine carbonaceous primary particles are coated with a carbonaceous binder precursor material, agglomerated and finally heat-treated at temperatures of between about 1850 and 3500°C to convert the binder precursor material to non-graphitic or graphitic carbon, thereby resulting in stable highly isotropic carbonaceous composite materials wherein the primary particles of the aggregate are held together by the carbonized/graphitized binder. The present disclosure also relates to the isotropic carbonaceous composite particles obtainable by the process described herein. The disclosure further relates to uses of said isotropic carbonaceous composite material in various applications, including as active material in negative electrodes in lithium-ion batteries, and in secondary products containing said isotropic carbonaceous composite material.

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Claims

1. Carbonaceous composite particles, characterized in that said particles are comprised of a multiplicity of aggregated primary particles, wherein said primary particles are held together by a carbonaceous binder material attached to the surface of the primary particles.

2. The carbonaceous composite particles according to claim 1, wherein said composite particles are further characterized by a pressure stability wherein the BET specific surface area does not increase by more than 3.5 m^2/g, and/or by more than 80% (compared to the BET SSA before pressing) after pressing the composite particles at 15 kN /cm^2 for 10 s.

3. The carbonaceous composite particles according to claim 1 or claim 2, wherein said composite particles are further characterized by a mass loss of non-graphitic carbon according to thermogravimetric analysis of less than 5%, or less than 2%, or less than 1%, or less than 0.5%.

4. The carbonaceous composite particles according to any one of claims 1 to 3, wherein said composite particles are further characterized by having a crystalline surface with a surface crystallinity expressed by an L_a of >4 nm, or >6 nm, or >10 nm, as determined by measuring the I_D/I_G band amplitude ratio via Raman spectroscopy.

5. The carbonaceous composite particles according to any one of claims 1 to 4, characterized by a near-random or random orientation of the primary particles in the aggregated composite particle.

6. The carbonaceous composite particles according to any one of claims 1 to 5, being isotropic in their electrical, mechanical, and/or heat-conductive properties.

7. The carbonaceous composite particles according to any one of claims 1 to 6, wherein the carbonaceous composite particles are further characterized by a ratio of the peak areas of the [004] and [110] reflections of lower than 10, or lower than 8, lower than 6, or lower than 4.

8. The carbonaceous composite particles according to any one of claims 1 to 7, wherein the carbonaceous composite particles are further characterized by

i) a BET specific surface area (BET SSA) of between 0.3 and 20 m^2/g;
ii) a crystallite size L_c of less than 300 nm;
iii) by a xylene density of at least 2.00, or at least 2.10, or at least 2.20 g/cm^3;
iv) an L_c/L_a ratio of at least 1, or at least 2 or at least 3; and/or

v) a spring-back of between 10 and 90 %; and/or

vi) the carbonaceous binder material connecting said primary particles being graphitic, or non-graphitic carbon, or both.

9. The carbonaceous composite particles according to any one of claims 1 to 8, wherein the primary particles are selected from carbonaceous materials such as natural graphite, synthetic graphite, graphene, graphene nanoplatelets, graphene or carbon fibers, fullerenes, nanographite, hard carbon, soft carbon, petroleum- or coal-based coke, graphitized fine coke, char, carbon black, carbon nanotubes (CNT), including single-walled nanotubes (SWNT), multiwalled nanotubes (MWNT), or mixtures of any of the foregoing;
optionally mixed together with non-carbonaceous materials such as silicon, silicon oxide, tin, tin oxide or tin dioxide, aluminum, bismuth, lithium titanate, or mixtures of any of the foregoing non-carbonaceous materials.

10. The carbonaceous composite particles according to any one of claims 1 to 9, wherein the primary particles are selected from a single material, optionally selected from a single carbonaceous material.

11. The carbonaceous composite particles according to any one of claims 1 to 10, wherein the primary particles are selected from at least 2, 3, 4, or at least 5 different materials.

12. The carbonaceous composite particles according to any one of claims 1 to 11, wherein the carbonaceous binder material is the same for all primary particles in the composite particle.

13. The carbonaceous composite particles according to any one of claims 1 to 12, wherein the carbonaceous binder material attached to the surface of said primary particles is graphitic carbon.

14. The carbonaceous composite particles according to any one of claims 1 to 13, wherein the composite particles are graphitic composite particles characterized by an interlayer distance c/2 of 0.337 nm or less (“graphitic composite particles”).

15. The carbonaceous composite particles according to any one of claims 1 to 12, wherein the carbonaceous binder material attached to the surface of said primary particles is non-graphitic carbon.

16. The carbonaceous composite particles according to any one of claims 1 to 12 and 15, wherein the composite particles are non-graphitic composite particles characterized by an interlayer distance c/2 of at least 0.338 nm, or at least 0.340 nm (“non-graphitic composite particles”).

17. The carbonaceous composite particles according to any one of claims 1 to 16, wherein the average length of the major axis of the primary particles as observed by scanning electron microscopy (SEM) is between 1 and 15 µm, or between 1 and 10 µm, or between 1 and 7 µm.

18. The carbonaceous composite particles according to any one of claims 1 to 17, wherein the carbonaceous binder material is different for at least a portion of the primary particles in the composite particle, optionally wherein at least 2, 3, 4, 5, or more different carbonaceous binder materials are present in a composite particle.

19. The carbonaceous composite particles according to claim 18, wherein the multiple carbonaceous binder materials are obtained by different coating methods and/or by employing different carbon precursors.

20. The carbonaceous composite particles according to any one of claims 1 to 19, wherein the carbonaceous binder material attached to the surface of the primary particles is obtainable by a method selected from

i) mixing of primary carbonaceous particles with a carbonaceous binder precursor material and subsequent carbonization of said carbonaceous binder precursor material;

ii) mixing of primary carbonaceous particles with a carbonaceous binder precursor material and subsequent high temperature treatment (>1850°C) of said carbonaceous binder precursor material;

iii) by melting the carbon precursor onto the primary particles;

iv) pitch-coating,

v) pyrolysis, and

vi) evaporation.

21. The carbonaceous composite particles according to any one of claims 1 to 20, further characterized by a particle size distribution (PSD) of the composite particles having a D_{90} value ranging from 5 to 70 µm, and/or a D_{50} value ranging from 2 to 30 µm, and/or a D_{10} value ranging from 0.5 to 20 µm.

22. The carbonaceous composite particles according to any one of claims 1 to 21, wherein the carbonaceous composite particles are further characterized by a non-graphitic carbon coating on the surface of the composite particles.

23. The carbonaceous composite particles according to any one of claims 1 to 22, characterized by further comprising an additive selected from the group consisting of carbon black, colloidal graphite, graphene, graphene nanoplatelets, graphene or carbon fibers, fullerenes, nanographite, char, carbon nanotubes (CNT), including single-walled nanotubes (SWNT), multiwalled nanotubes (MWNT), or mixtures of any of the foregoing, metals/metalloids such as silicon, aluminum, tin, silver, copper, nickel, antimony, germanium, metal / metalloid oxides such as TiO_2, lithium titanate, silicon oxide, or tin oxide, chalcogenides, or metal alloys, optionally wherein the metals/metalloids are selected from silicon, aluminum, or tin, or alloys comprising said metals.

24. The carbonaceous composite particles according to any one of claims 1 to 23, having a polycyclic aromatic hydrocarbon (PAH) concentration of less than 200 mg/kg, less than 150 mg/kg, less than 30 mg/kg, less than 10 mg/kg, less than 5 mg/kg, less than 2 mg/kg, less than 1 mg/kg, or less than 0.5 mg/kg.

25. The carbonaceous composite particles according to any one of claims 1 to 24, wherein the particles are further characterized by a rate capability 2C/0.2C of at least 97%, or at least 98%, or at least 99%, or at least 99.5% when present as an active material in a negative electrode of a Lithium-ion battery.

26. Carbonaceous composite particles as defined in any one of claims 1 to 25, obtainable by a process as defined in any one of claims 30 to 56.

27. Composition comprising the carbonaceous composite particles as defined in any one of claims 1 to 26.

28. The composition of claim 27, mixed together with one or more other type of carbonaceous composite particles as defined herein.

29. The composition of claim 27 or claim 28, mixed together with other unmodified or modified carbonaceous particles.

30. Process for preparing carbonaceous composite particles, comprising

(a) attaching a carbonaceous binder precursor material to the surface of carbonaceous particles, optionally in the presence of a solvent, thereby forming a coating of the carbonaceous particles by the carbonaceous binder precursor material;

(b) in case a solvent was used in step (a), drying the material obtained from step a;

(c) during or after step (a) or step (b), causing agglomeration of the coated primary carbonaceous particles.

31. The process of claim 30, wherein the process further includes subjecting the agglomerated primary particles from step (c) to a heat treatment of between 300 to 3500°C, preferably wherein the heat treatment step d) is a high temperature treatment between about 1850 and 3500°C.

32. The process according to claim 30 or claim 31, wherein the carbonaceous composite particles are comprised of a multiplicity of aggregated primary particles, wherein said primary particles are held together by a carbonaceous binder material attached to the surface of the primary particles; preferably wherein the carbonaceous composite particles obtained from the process are as defined in any one of claims 1 to 25.

33. The process of any one of claims 30 to 32, wherein the carbonaceous binder precursor material is not or does not include ammonium lignosulfonate.

34. The process of any one of claims 30 to 33, wherein the carbonaceous binder precursor material is not or does not include coal tar, tar pitch, and petroleum pitch.

35. The process according to any one of claims 30 to 34, wherein the carbonaceous particles employed in step (a) are selected from the group consisting of natural graphite, synthetic graphite, graphene, graphene nanoplatelets, graphene or carbon fibers, fullerenes, nanographite, hard carbon, soft carbon, petroleum- or coal-based coke, graphitized fine coke, char, carbon black, carbon nanotubes (CNT), including single-walled nanotubes (SWNT), multiwalled nanotubes (MWNT), carbon nanofibers (CNF) or mixtures thereof; or from non-carbonaceous materials such as silicon, silicon oxide, tin, tin oxide or tin dioxide, aluminum, bismuth, lithium titanate, or mixtures of any of the foregoing.

36. The process according to any one of claims 30 to 35, wherein the carbonaceous particles employed in step (a) are non-graphitic particles, optionally selected from the group consisting of hard carbon, soft carbon, petroleum- or coal-based coke, graphitized fine coke, char, carbon black and mixtures thereof; or wherein the carbonaceous particles to be coated

are selected from fine coke and carbon black;
optionally mixed together with graphitic particles.

37. The process according to any one of claims 30 to 36, wherein the particle size distribution of said carbonaceous particles employed in step (a) is characterized by a D_{90} of $<$ 35 µm, and/or a D_{50} of $<$ about 20 µm, optionally by a D_{90} of $<$ 25 µm, and/or a D_{50} of $<$ about 15 µm.

38. The process according to any one of claims 30 to 37, wherein the carbonaceous particles to be coated in step (a) have
(i) a sphericity Q3 [S=0.8] of equal or more than about 22 % ; and/or
(ii) a Scott density of >0.2 g/cm^3.

39. The process according to any one of claims 30 to 38, wherein the carbonaceous binder precursor material to be used in step (a) is selected from the group consisting of polymers such as a lignin-based polymer, a polystyrene or derivative thereof, styrene-butadiene, melted phenol resin, polyvinylalcohol, polyfurfuryl alcohol, furfural, polyurethane, polystyrene-acrylate, polyacrylate, polymethylmethacrylate, polymethacrylonitrile, polyoxymethylene, poly(methyl atropate), polyisobutene, polyethyleneoxide, polypropyleneoxide, polyethylene, polypropylene, polymethylacrylate, polybutadiene, polyisoprene, polyacrylonitrile, polyaniline, tannic acid, starch, gum arabic, maltodextrin, formaldehyde phenol resins, formaldehyde tetrahydrofuran resins, nitrile butyl rubber, sucrose, glucose, or other sugars, polyethyl ether ketone, polyphenylene sulfide, polyvinyl chloride, carboxymethylcellulose, methyl cellulose, gelatins, polyvinyl pyrrolidone, polylactic acid, latexes thereof, a hydrocarbon gas such as methane, ethane, ethylene, propane, propene, acetylene, butane, benzene, toluene, xylene, or an alcohol such as ethanol, propanol, isopropanol, mixed with an inert carrier gas, and combinations thereof.

40. The process according to any one of claims 30 to 39, wherein the attachment of the carbonaceous binder precursor material to the surface of the carbonaceous particles is achieved by a method selected from the group consisting of
i) mixing to form a dispersion, optionally in the presence of a solvent and subsequent drying;
ii) melting the carbonaceous binder precursor onto the primary particles;
(iii) pyrolysis;
(iv) pitch-coating; and
(v) evaporation.

41. The process according to any one of claims 30 to 40, wherein the carbonaceous particles and the carbonaceous binder precursor material is dispersed in the presence of a solvent.

42. The process according to claim 41, wherein the solvent is a polar solvent, optionally wherein the solvent is selected from water, methanol, ethanol, propanol, isopropanol, acetone, or mixtures thereof.

43. The process according to any one of claims 30 to 42, wherein the carbonaceous binder precursor material is melted on the carbonaceous particles.

44. The process according to any one of claims 30 to 43, wherein further additives are added during step (a), optionally wherein said additives are selected form the group consisting of citric acid, ammonia, acetic acid, formic acid, malic acid, stearic acid, and combinations thereof.

45. The process according to any one of claims 30 to 44, wherein the agglomeration of step (c), and, optionally steps (a) and (b), is achieved by spray-drying a dispersion comprising the primary carbonaceous particles and the carbonaceous binder precursor material.

46. The process according to any one of claims 30 to 44, wherein the drying of step (b) and the agglomeration of step (c) is achieved by vacuum-drying a dispersion comprising the primary carbonaceous particles and the carbonaceous binder precursor material obtained from step (a) in a heatable vacuum reactor.

47. The process according to any one of claims 30 to 44, wherein the drying of step (b) and the agglomeration of step (c) is achieved by freeze-drying a dispersion comprising the primary carbonaceous particles and the carbonaceous binder precursor material obtained from step (a) in a stirred freeze dryer.

48. The process according to any one of claims 30 to 44, wherein the drying of step (b) and the agglomeration of step (c) is achieved by flash-drying a dispersion comprising the primary carbonaceous particles and the carbonaceous binder precursor material obtained from step (a) in a flash dryer.

49. The process according to any one of claims 30 to 44, wherein the drying of step (b) and the agglomeration of step (c) is achieved by drying a fluidized dispersion comprising the

primary carbonaceous particles and the carbonaceous binder precursor material obtained from step (a) in a fluidized bed dryer, optionally in combination with a spray system.

50. The process according to any one of claims 30 to 44, wherein the drying of step (b) and the agglomeration of step (c) is achieved by disc drying a dispersion comprising the primary carbonaceous particles and the carbonaceous binder precursor material obtained from step (a) in a disc dryer.

51. The process according to any one of claims 30 to 44, wherein the drying of step (b) and the agglomeration of step (c) is achieved by paddle drying a dispersion comprising the primary carbonaceous particles and the carbonaceous binder precursor material obtained from step (a) in a paddle dryer.

52. The process according to any one of claims 30 to 44, wherein said carbonaceous binder precursor material attached to the surface of the agglomerated particles from step (c) is carbonized by a thermal decomposition under vacuum or an inert atmosphere, optionally under a nitrogen or argon atmosphere, at temperatures ranging from 400°C to 3500°C; optionally wherein said carbonization of the binder precursor material is carried out in a separate step prior to step (d).

53. The process according to any one of claims 30 to 44, wherein said heat treatment of step (d) is carried out at temperatures and for a sufficient time to convert any non-graphitic carbon in the intermediary agglomerated particles obtained from step c to graphitic carbon.

54. The process according to any one of claims 30 to 53, wherein prior to the carbonization step (d), the coated agglomerated carbonaceous particles are subjected to a pre-treatment performed under vacuum, air, nitrogen, argon or CO_2 atmosphere at temperatures of below 1100°C, or below 700°C.

55. The process according to any one of claims 30 to 54, wherein the particles obtained from step (d) are subjected to an additional heat treatment in a gas atmosphere such as nitrogen, argon, mixtures of nitrogen with hydrocarbons like acetylene, propane or methane, or with oxidative gases such as air, steam, or CO_2 to adjust the morphology and surface chemistry of the carbonaceous composite particles, optionally wherein said heat treatment is carried out at a temperature ranging from 300°C to 1500°C.

56. The process according to claim 55, wherein the additional heat treatment is performed by contacting the particles with an oxidant either in a gaseous / solid phase process with air, carbon dioxide, water vapor, oxygen, ozone, or any combination thereof, or,

alternatively, in a liquid / solid phase process with aqueous hydrogen peroxide or other oxidants present in said liquid phase.

57. Use of the carbonaceous composite particles as defined in any one of claims 1 to 26 or the composition of any one of claims 27 to 29 for preparing a negative electrode material for a lithium-ion battery.

58. A negative electrode of a lithium-ion battery comprising the carbonaceous composite particles as defined in any one of claims 1 to 26 or the composition of any one of claims 27 to 29 as an active material in the negative electrode of the battery.

59. An energy storage device comprising the carbonaceous composite particles as defined in any one of claims 1 to 26 or the composition of any one of claims 27 to 29.

60. A carbon brush comprising the carbonaceous composite particles as defined in any one of claims 1 to 26 or the composition of any one of claims 27 to 29.

61. A polymer composite material comprising the carbonaceous composite particles as defined in any one of claims 1 to 26 or the composition of any one of claims 27 to 29.

62. A lithium-ion battery comprising the carbonaceous composite particles as defined in any one of claims 1 to 26 or the composition of any one of claims 27 to 29 as an active material in the negative electrode of the battery.

63. The lithium-ion battery according to claim 62, wherein the battery is characterized by a rate capability 2C/0.2C of at least 97%, or at least 98%, or at least 99%, or at least 99.5%.

64. The lithium-ion battery according to claim 62 or claim 63, wherein the lithium-ion battery has a first rate of charging and/or discharging greater than a second rate of charge and/or discharging of a lithium-ion battery devoid of the carbonaceous composite particles as defined in any one of claims 1 to 26 or the composition of any one of claims 27 to 29.

65. An electric vehicle, hybrid electric vehicle, or plug-in hybrid electric vehicle comprising the lithium-ion battery according to any one of claims 62 to 64.

66. The electric vehicle, hybrid electric vehicle, or plug-in hybrid electric vehicle of claim 65, wherein the carbonaceous composite particles comprise a graphitic material.

67. The electric vehicle, hybrid electric vehicle, or plug-in hybrid electric vehicle of claim 65, wherein the carbonaceous composite particles comprise a non-graphitic material.

68. A sodium ion battery comprising the carbonaceous composite particles as defined in any one of claims 1 to 26 or the composition of any one of claims 27 to 29.

69. A carbon-based coating exhibiting isotropic electric, mechanical or heat-conducting properties, wherein said coating comprises the carbonaceous composite particles as defined in any one of claims 1 to 26 or the composition of any one of claims 27 to 29.

70. Use of the carbon-based coating according to claim 69 as a coating of a current collector in batteries.

71. A dispersion comprising the carbonaceous composite particles as defined in any one of claims 1 to 26 or the composition of any one of claims 27 to 29.

72. The dispersion according to claim 71, wherein the dispersion is in the form of a slurry, optionally wherein the solvent is selected from water or N-methyl-2-pyrrolidone (NMP).

73. A method for making a building block of a negative electrode, employing the carbonaceous composite particles as defined in any one of claims 1 to 26 or the composition of any one of claims 27 to 29.