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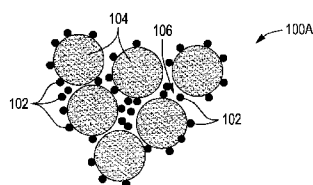


FIG. 1A

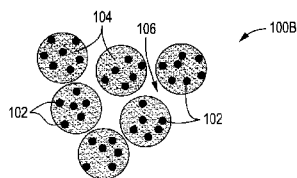


FIG. 1B

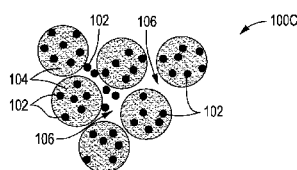


FIG. 1C

(57) Abstract: Composite powders may be formed by blending silicon particles with petroleum pitch under grinding conditions. The composite powders may comprise up to about 50 wt.% silicon particles, based on total mass of the composite powder, and about 15 wt.% to about 95 wt.% petroleum pitch, based on total mass of the composite powder, and, optionally, up to 80 wt.% graphite particles. The silicon particles are dispersed in a matrix comprising the petroleum pitch and the petroleum pitch comprises a plurality of pitch particles. The composite powders may be subsequently converted to a carbon matrix comprising amorphous carbon by heating the petroleum pitch at a temperature of about 700°C to about 1800°C in an environment comprising about 0.1 mol% oxygen or below.

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PITCH-BASED COMPOSITE POWDERS CONTAINING SILICON AND METHODS FOR PRODUCTION AND USE THEREOF

CROSS-REFERENCE TO RELATED APPLICATIONS

5 [0001] The present application claims priority to United States Provisional Application No. 63/508,389 filed June 15, 2023, the disclosure of which is incorporated by reference in its entirety.

FIELD

10 [0002] The present disclosure relates to composite powders and, more particularly, to composite powders containing petroleum pitch or derived from petroleum pitch.

BACKGROUND

[0003] Batteries, including lithium-ion batteries, are commonly used for portable electronic devices, electric vehicles, and general energy storage. Lithium-ion batteries are commonly used in such applications due to their high energy storage density and high power output. Lithium-ion battery cells comprise a cathode and an anode, wherein the cathode is made from a lithium compound (*e.g.*, lithium cobalt oxide, lithium nickel manganese cobalt oxide, or the like) and the anode is conventionally made from a natural or synthetic graphite material. One limitation of lithium-ion batteries is that the graphite material commonly has a rather limited energy storage capacity for electrical charge, with a theoretical gravimetric charge capacity of up to 372 mAh/g in commercial lithium-ion batteries typically being realized. The energy storage capacity may be increased by incorporating an electrochemically active material into the graphite material. However, volume expansion and contraction during charging and discharging cycles may result in mechanical degradation of the electrode, particularly when an electrochemically active material has been added to increase storage capacity.

25 [0004] One precursor of graphite materials suitable for forming an anode in lithium-ion batteries is petroleum pitch. Petroleum pitch is a carbon-rich viscoelastic material originating from petroleum and having properties similar to a thermoplastic polymer. Petroleum pitch can be used as a precursor material for producing an array of consumer and industrial carbon products, such as carbon fiber, graphite, binder pitch, impregnation pitch, and the like. Beyond battery applications, major markets for petroleum pitch and carbon products originating therefrom include, but are not limited to, high-performance and general purpose carbon fibers, refractory materials, carbon/carbon composites, synthetic graphite and graphite parts, binder and impregnated pitch for electrodes, binder and impregnated aluminum production anodes and cathodes, impregnated pitch for steel electric arc furnace electrodes, carbon foam for heat

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transfer applications and sound absorbers, roofing products, lubricants, consumer products (e.g., cosmetics), and the like.

[0005] Limiting the volumetric expansion of electrochemically active materials is the most common approach to minimize the mechanical degradation in anodes. This can be achieved in three ways: (1) coating the electrochemically active material, typically with a carbon-based material, to enhance its mechanical integrity, (2) by mixing the electrochemically active material with graphite while preparing the anode slurry, or (3) using a smaller size active material particle. Relying on one of these approaches doesn't result in a competitive anode with high cycle life and typically, combining all three approaches is commonly practiced for best results. However, the current approaches still cannot push the total amount of electrochemically active material in the anode beyond a resulting gravimetric charge capacity of 500 mAh/g. The limitation is a result of both a poor dispersion of active material particles within the graphite in the slurry and poor mechanical properties of the coating. The latter requires electrochemically active materials to be mixed with up to 70 wt.% of coating/matrix material to make a masterbatch, which is then added to the slurry to make up to 30 wt.% of the anode active material.

SUMMARY

[0006] In various aspects, the present disclosure provides composite powders comprising: up to about 50 wt.% silicon particles, based on total mass of the composite powder; and about 15 wt.% to about 95 wt.% petroleum pitch, and up to about 80 wt.% graphite, based on total mass of the composite powder; wherein the silicon and graphite particles are dispersed in a matrix comprising the petroleum pitch and the petroleum pitch comprises a plurality of pitch particles.

[0007] In some or other various aspects, the present disclosure provides compositions comprising: up to about 60 wt.% silicon particles dispersed in a carbon matrix, based on total mass of the composition; wherein the carbon matrix comprises amorphous carbon.

[0008] In still other various aspects, methods of the present disclosure comprise: forming a blend comprising up to about 50 wt.% silicon particles and about 15 wt.% to about 95 wt.% petroleum pitch, and up to about 80 wt.% graphite, based on a total mass of the blend; and processing the blend under grinding conditions to form a composite powder; wherein the silicon and graphite particles are dispersed in a matrix comprising the petroleum pitch and the petroleum pitch comprises a plurality of pitch particles.

[0009] These and other features and attributes of the disclosed compositions and methods of the present disclosure and their advantageous applications and/or uses will be apparent from the detailed description which follows.

BRIEF DESCRIPTION OF THE DRAWINGS

5 [0010] To assist one of ordinary skill in the relevant art in making and using the subject matter hereof, reference is made to the appended drawings. The following figures are included to illustrate certain aspects of the disclosure, and should not be viewed as exclusive configurations. The subject matter disclosed is capable of considerable modifications, alterations, combinations, and equivalents in form and function, as will occur to those skilled
10 in the art and having the benefit of this disclosure.

[0011] FIG. 1A is a diagram of a composite powder having silicon particles dispersed between pitch particles. FIG. 1B is a diagram of a composite powder having silicon particles within the interior of pitch particles. FIG. 1C is a diagram of a composite powder having silicon particles both within the interior of the pitch particles and dispersed between the pitch particles.

15 [0012] FIGS. 2A-2C are SEM-BSE (scanning electron microscope-backscattered electron) images of illustrative composite powder samples produced according to dry blending procedures of the present disclosure.

[0013] FIG. 3 is a powder XRD (X-ray diffraction) plot of illustrative composite powder samples produced according to dry blending procedures of the present disclosure and
20 carbonized at various temperatures.

DETAILED DESCRIPTION

[0014] The present disclosure relates to composite powders and, more particularly, to composite powders containing petroleum pitch or derived from petroleum pitch.

[0015] It is to be understood that the terms “pitch” and “petroleum pitch” are used
25 interchangeably herein. Moreover, it is to be understood that the “pitch particles” described herein contain petroleum pitch.

[0016] The present disclosure provides ready access to composite powders containing petroleum pitch, silicon, and graphite that may be readily processed into battery anodes and other carbon-based structures following carbonization or graphitization thereof. It is to be
30 appreciated that the oxide forms of silicon can be used as alternatives to pure silicon. The composite powders may contain natural or synthetic graphite and silicon in nanoparticle form, which may afford a number of advantages in battery anodes and other applications. The nanoparticle form of the silicon (silicon nanoparticles) may comprise a previously produced nanoparticle form that is blended with the petroleum pitch, or more advantageously, the

nanoparticle form may be generated *in situ* when blending a silicon source with the petroleum pitch. As used herein, the term “nanoparticle form” refers to any size range below about 1000 nm, preferably below about 500 nm, and more preferably below about 200 nm or below about 100 nm. By incorporating silicon into petroleum pitch to form a composite powder and then carbonizing, the silicon may increase the energy density (*e.g.*, a theoretical gravimetric charge capacity of up to about 3600 mAh/g) to allow for improved performance in battery anodes, for example. Moreover, by incorporating the silicon into the composite powders in a nanoparticle form, a surprising improvement in performance with respect to stability during volume expansion and contraction may be realized. Without being bound by theory or mechanism, the improved performance and mechanical stability during charging and discharging cycles is believed to result from the silicon being more tolerant of recurring expansion and contraction when in nanoparticle form. Ready dispersion of the silicon within the pitch may also play a role in this regard.

[0017] Advantageously, the composite powders of the present disclosure may be prepared under continuous blending and pulverization conditions, either with or without first melting or softening the petroleum pitch. When the petroleum pitch is heated above its softening temperature, the blending process may be referred to as melt blending. Above the softening temperature, the pitch particles become more malleable, thereby allowing silicon particles to become embedded in the outer surface of the pitch particles or even become admixed within the interior of the pitch particles as the pitch particles deform and undergo reshaping. At temperatures where the petroleum pitch remains below its softening temperature, the blending process may be referred to as dry blending. Below the softening temperature, the pitch particles remain hard, and the silicon particles remain external to the pitch particles. In the cases of both melt blending and dry blending, the blending process may be conducted continuously in a screw mill extruder or similar extruder type, and the resulting product may be directly obtained as a composite powder without the need for further grinding. Optionally, the composite powder may be sieved to a desired particle size, if needed. The extruder may promote formation of a decreased particle size for both the petroleum pitch and the silicon particles incorporated therein, while also facilitating intimate blending thereof to form a composite powder. Optionally, the continuous grinding process may occur under dry blending conditions with the extruder being cooled (*e.g.*, between about -10°C to about 5°C) to maintain the petroleum pitch in a hardened state and to limit potential chemical degradation. In the present disclosure, the term “cold blending” will be used to refer to dry blending processes taking place below room temperature (23°C) and below a softening temperature of the components being blended. It is

to be appreciated that the blending of petroleum pitch, silicon, and graphite can be carried out in one step via dry or melt blending, which can enhance the mechanical stability of the final anode composite powder as a result of the good dispersion between the three components. Moreover, blending in a one-step process combines coating both silicon and graphite into one step, which is advantages from a processing perspective. When the composite powders are produced under such dry blending conditions, the intimate blend of petroleum pitch particles and silicon particles may be further heated after dry blending at a temperature below the softening temperature of the petroleum pitch (*e.g.*, about 200°C to about 450°C, or about 200°C to about 300°C, or about 300°C to about 450°C) to set or immobilize the silicon particles upon an outer surface of the pitch particles (*e.g.*, through complete or partial embedment of the silicon particles upon the surface of the pitch particles) while still maintaining the overall composition in a well-dispersed powder form. Since the onset of softening usually occurs some 50°C to 100°C below the actual softening temperature, some embedment of the silicon particles upon the outer surface of the pitch particles may occur within the foregoing temperature ranges. Some crosslinking of the petroleum pitch may also occur under these heating conditions, if the heating is conducted in a low-oxygen environment containing about 1 mol% oxygen up to about 20 mol% oxygen. Heating in an environment having a low-oxygen content (below about 0.1 mol%) may result in minimal or no crosslinking. When the silicon particles are set or immobilized upon the outer surface of the pitch particles, the interior of the pitch particles may continue to remain devoid of silicon particles. Melt blending processes, in contrast, may result in dispersion of at least a portion of the silicon particles throughout an interior of the pitch particles, rather than the silicon particles being localized in the interstitial space between pitch particles. Some silicon particles may still remain external to the pitch particles in a melt blending process, however.

[0018] At least some oxidation of the petroleum pitch and/or the silicon particles may take place under the heating conditions used to set the silicon particles. Partial oxidation of the pitch particles may enhance mechanical integrity of the composite powder through at least partial crosslinking of the petroleum pitch. Additionally, partial oxidation when setting or immobilizing the silicon particles may decrease the time needed to stabilize the petroleum pitch through oxidative crosslinking prior to carbonization of the composite powders. Incidental oxidation of the silicon particles during setting or immobilization thereof is not believed to be problematic, since any silicon oxides that form may be reverted to elemental silicon under the conditions employed for subsequent carbonization or graphitization of the composite powders.

[0019] Composite powders of the present disclosure may comprise silicon or silicon dioxide (SiO₂) particles, preferably silicon or silicon dioxide nanoparticles, and in some case graphite particles blended with petroleum pitch, wherein the silicon or silicon dioxide particles are dispersed in a matrix comprising the petroleum pitch and the petroleum pitch comprises a plurality of pitch particles. Silicon particles and silicon dioxide particles may be used interchangeably when forming the composite powders according to the disclosure herein, since the silicon dioxide may be reverted to elemental silicon under the conditions employed during subsequent carbonization or graphitization of the composite powders. Unless otherwise specified herein, the term “silicon particle” and grammatical variations thereof refers to any particulate material containing primarily elemental silicon or one or more silicon compounds, preferably a silicon oxide such as silicon dioxide. Thus, any description herein referencing silicon or silicon particle(s) is understood to equivalently reference a silicon oxide, such as silicon dioxide. Moreover, use of the term “silicon dioxide” herein is to be understood as being inclusive of other silicon oxides. Other silicon species may similarly be present in the silicon particles as well.

[0020] The morphology of the composite powders may differ depending on whether the silicon particles are combined with the petroleum pitch under melt blending conditions, dry blending conditions, or a combination thereof. In dry blending processes, the silicon particles may be dispersed in the composite powder by being located between pitch particles, such as in the interstitial space between the pitch particles. FIG. 1A is a diagram of composite powder **100A** showing silicon particles **102** dispersed between pitch particles **104** within interstitial spaces **106**. Melt blending processes, in contrast, may produce the composite powder with at least a portion of the silicon particles dispersed within the interior of the pitch particles, optionally with some of the silicon particles being exposed to the surface of the pitch particles. FIG. 1B is a diagram of composite powder **100B** showing silicon particles **102** within the interior of pitch particles **104**, in which case interstitial spaces **106** are either unoccupied (as depicted in FIG. 1B) or some silicon particles **102** also reside in interstitial spaces **106** and/or become embedded in the outer surface of pitch particles **104** (FIG. 1C). Optionally, further dry blending of composite powder **100B** with silicon particles **102** may take place to fill at least a portion of interstitial spaces **106** (not shown). FIG. 1C is a diagram of composite powder **100C** showing silicon particles **102** within the interior of pitch particles **104** and also within interstitial spaces **106**, wherein interstitial spaces **106** are filled either during a melt blending process or during a further dry blending process following a melt blending process. While pitch particles **104** and silicon particles **102** are shown in FIGS. 1A-1C as being round and

individually of the same size, it is to be appreciated that the particle shapes may be irregular and a range of particle sizes may be present for both silicon particles **102** and pitch particles **104**.

[0021] More specifically, composite powders of the present disclosure may comprise up to about 50 wt.% silicon particles or up to about 30 wt.% silicon particles, and about 15 wt.% to about 95 wt.% petroleum pitch, and up to about 80 wt.% graphite particles, each based on total mass of the composite powder, and wherein the silicon particles are dispersed in a matrix comprising the petroleum pitch and the petroleum pitch comprises a plurality of pitch particles. In non-limiting examples, the composite powders may contain silicon particles in an amount ranging from about 1 wt.% to about 30 wt.%, or about 5 wt.% to about 30 wt.%, or about 10 wt.% to about 30 wt.%, or about 5 wt.% to about 20 wt.%, or about 5 wt.% to about 15 wt.%, or about 10 wt.% to about 20 wt.%, or about 10 wt.% to about 25 wt.%, or about 9 wt.% to about 15 wt.%, each based on total mass of the composite powders. In some or other non-limiting examples, the composite powders may contain the petroleum pitch in an amount ranging from about 25 wt.% to about 95 wt.%, or about 30 wt.% to about 80 wt.%, or about 40 wt.% to about 75 wt.%, or about 15 wt.% to about 70 wt.%, or about 30 wt.% to about 50 wt.%, or about 50 wt.% to about 70 wt.%, or about 70 wt.% to about 90 wt.%, or about 85 wt.% to about 95 wt.%, each based on total mass of the composite powders. In some or other non-limiting examples, the composite powders may contain the graphite particles in an amount ranging from about 1 wt.% to about 80 wt.%, or about 1 wt.% to about 10 wt.%, or about 10 wt.% to about 75 wt.%, or about 15 wt.% to about 70 wt.%, or about 30 wt.% to about 50 wt.%, or about 50 wt.% to about 70 wt.%, or about 70 wt.% to about 80 wt.%, or about 50 wt.% to about 80 wt.%, each based on total mass of the composite powders.

[0022] The petroleum pitch used in the present disclosure may be obtained from any source or process, provided that the petroleum pitch does not contain components that might be detrimental to an intended application following carbonization of the composite powder. In some examples, at least a majority of the petroleum pitch may comprise a mesophase pitch. Mesophase pitch is an anisotropic pitch that comprises a complex mixture of aromatic molecules that are at least partially ordered and coalesce into a liquid crystalline phase. The crystallinity may enhance mechanical integrity. Moreover, once carbonized, the highly aligned structure may promote enhanced electrical conductivity, such as in battery applications, for example. In non-limiting examples, the petroleum pitch used herein may have a mesophase pitch content of about 50 wt.% or greater, or about 60 wt.% or greater, or about 70 wt.% or greater, or about 80 wt.% or greater, or about 90 wt.% or greater, or about 95 wt.% or greater,

or about 99 wt.% or greater, or about 99.9 wt.% or greater, such as about 50 wt.% to about 90 wt.%, or about 60 wt.% to about 80 wt.%, or about 80 wt.% to about 90 wt.%, or about 85 wt.% to about 99 wt.%, or about 90 wt.% to about 99 wt.%, or about 80 wt.% to about 99.9 wt.%, or about 90 wt.% to about 99.9 wt.%, or about 95 wt.% to about 99.9 wt.%, or even 100 wt.%,
5 each based on a total mass of the petroleum pitch.

[0023] The silicon particles used in the present disclosure may comprise elemental silicon, silicon dioxide, or any combination thereof. The silicon particles used in the present disclosure may be obtained from any source or process, provided that the silicon particles do not contain components that might be detrimental to an intended application. Depending on source and
10 whether elemental silicon or silicon dioxide is present in the silicon particles, an impurity content of the silicon particles may be about 5 wt.% or below, or about 2 wt.% or below, or about 1 wt.% or below, or about 0.5 wt.% or below, or about 0.1 wt.% or below, or about 0.01 wt.% or below, or about 0.001 wt.% or below, or about 0.0001 wt.% or below, or even about 0.00001 wt.% or below. Likewise, depending on whether the silicon particles comprise
15 elemental silicon and/or silicon dioxide and the source of the silicon and/or silicon dioxide, the silicon particles may have a silicon content of about 25 wt.% to about 99.9999 wt.%, or about 40 wt.% to about 99.9999 wt.%, or about 45 wt.% to about 99.9999 wt.%, or about 50 wt.% to about 99.9999 wt.%, or about 90 wt.% to about 99.9999 wt.%, or about 95 wt.% to about 99.9999 wt.%, by mass of the silicon particles. In some examples, if the silicon particles
20 comprise substantially elemental silicon, the mass balance of the silicon particles may comprise silicon dioxide. In embodiments the graphite includes natural and/or synthetic graphite. In embodiments, the graphite includes varying sizes from 1 micron to 200 microns. In embodiments, the graphite includes from 50% to 100% graphitization.

[0024] Once blended with the petroleum pitch, the silicon particles may have any suitable
25 size and any suitable particle size distribution to become dispersed in the resulting carbon matrix following carbonization. For example, the silicon particles blended with the petroleum pitch may have a D50 of about 200 nm or less or about 300 nm or less, and a D90 of about 1000 nm or less or about 1200 nm or less. As used herein, the term “D50” refers to a diameter at which 50% of a sample (on a volume basis unless otherwise specified) is comprised of
30 particles having a diameter less than said diameter. As used herein, the term “D90” refers to a diameter at which 90% of a sample (on a volume basis unless otherwise specified) is comprised of particles having a diameter less than said diameter. Example particle size distributions of the silicon particles may include a D50 of about 10 nm to about 300 nm, or about 50 nm to about 250 nm, or about 100 nm to about 200 nm, or about 1 nm to about 200 nm, and/or a D90 of

about 10 nm to about 1200 nm, or about 10 nm to about 1000 nm, or about 10 nm to about 900 nm, or about 700 nm to about 1100 nm.

[0025] In some or other examples, at least a majority of the silicon particles present within the composite powders may have a size ranging from about 25 nm to about 200 nm, or about 50 nm to about 150 nm, or about 50 nm to about 100 nm, or about 50 nm to about 75 nm, or about 75 nm to about 100 nm, or about 75 nm to about 125 nm, or about 125 nm to about 175 nm. It should be noted that silicon particles larger in size than 200 nm may be used to produce the composite powders disclosed herein, since the silicon particles may undergo a reduction in size under the grinding and pulverization conditions used to produce the composite powders according to the further description herein. For example, the silicon particles may have a size up to about 500 nm before processing a blend of the petroleum pitch and the silicon particles under grinding conditions to produce the composite powders disclosed herein.

[0026] The composite powders of the present disclosure may optionally further comprise graphite, which may be introduced when mixing the silicon particles with the petroleum pitch and forming the composite powder, or the graphite may be mixed with the composite powder afterward. In non-limiting examples, the composite powders may comprise graphite in an amount ranging from about 0.1 wt.% to about 85 wt.%, or about 0.1 wt.% to about 60 wt.%, or about 5 wt.% to about 60 wt.%, or about 5 wt.% to about 20 wt.%, or about 20 wt.% to about 40 wt.%, or about 30 wt.% to about 60 wt.%, or about 5 wt.% to about 20 wt.%, or about 20 wt.% to about 40 wt.%, each based on total mass of the composite powder. If present, the graphite may further define the matrix containing the petroleum pitch. That is, the silicon particles (elemental silicon or a silicon oxide) may be dispersed in a matrix comprising the petroleum pitch and graphite, with both the petroleum pitch and the graphite being in the form of particles. Any of natural or synthetic graphite, including synthetic graphite derived from graphite-like materials such as coke, may be used in regard to the foregoing. The graphite may be delivered in any suitable powder form.

[0027] The composite powders of the present disclosure may have a particle size ranging from about 1 μm to about 50 μm , or about 5 μm to about 50 μm , or about 1 μm to about 25 μm , or about 5 μm to about 25 μm , or about 10 μm to about 30 μm . Namely, the composite powders may contain at least petroleum pitch particles within the foregoing size ranges. Silicon particles and graphite particles (if present) may reside within the same size range as the petroleum pitch particles or be smaller. Preferably, the silicon particles may be smaller than the petroleum pitch particles and reside within a nanoparticle size range, as discussed above.

[0028] The above composite powders may serve as a precursor composite for forming carbon composites in which the petroleum pitch is pyrolyzed (carbonized) to form a carbon matrix comprising amorphous carbon and/or graphite. Amorphous carbon may be formed upon exposing the petroleum pitch to a temperature ranging from about 700°C to about 1800°C, or about 900°C to about 1800°C, or about 900°C to about 1500°C, or about 1000°C to about 1500°C, or about 900°C to about 1400°C, in a no-oxygen or very low-oxygen environment (e.g., an oxygen content below about 0.1 mol% or below), preferably in the presence of an inert gas environment. Graphite, in contrast, may be formed upon heating at a higher temperature ranging from about 2000°C to about 3400°C or about 2500°C to about 3400°C, also in a no-oxygen or very low-oxygen environment, preferably in the presence of an inert gas environment. Amorphous carbon may be distinguished from graphite spectroscopically by powder X-ray diffraction, for example, as will be appreciated by one having ordinary skill in the art. Amorphous carbon further lacks long-range molecular order, whereas graphite is a covalent crystal defined by sheets of sp²-hybridized carbon atoms. Upon undergoing carbonization, preferably to amorphous carbon, the resulting compositions may be suitable for forming a battery anode, such as for a lithium-ion battery.

[0029] A small amount of mass loss may occur when carbonizing the composite powders to form the carbon matrix. Without being limited by theory or mechanism, the mass loss is believed to result from various reactions of the petroleum pitch that form gaseous products. Such reactions may include, for instance, dehydrogenation, polymerization with side chain loss and/or hydrogen production, condensation of aromatic rings, and decomposition of oxygen-containing groups. Gaseous products may include, for example, carbon monoxide, carbon dioxide, water vapor, hydrocarbon vapor, methane, and the like. Up to about 20 wt.% of the petroleum pitch may undergo mass loss due to such reactions during carbonization. Preferably, the amount of mass loss is about 10 wt.% or less, or about 5 wt.% or less, or about 2 wt.% or less. With such mass loss occurring during carbonization, the corresponding loading of silicon may increase, such up to about 60 wt.% based on total mass of the resulting carbon matrix.

[0030] Before performing carbonization, the composite powders of the present disclosure may optionally be heated at a temperature below the softening point of the petroleum pitch, preferably in low-oxygen environment (0.1 mol% to 20 mol% oxygen). Heating in this manner may aid in enhancing the mechanical integrity of the composite powder through at least partial crosslinking of the petroleum pitch. Such heating may also aid in setting the silicon particles within the matrix defined by the petroleum pitch (e.g., upon the outer surface of pitch particles), such as by at least partially embedding the silicon particles in an outer surface of petroleum

pitch particles defining the matrix. In non-limiting examples, the heating may take place at a temperature above room temperature and below about 500°C, or below about 400°C, or below about 300°C, or below about 200°C, such as within a range of about 200°C to about 450°C, or about 200°C to about 300°C, or about 200°C to about 250°C, or about 300°C to about 450°C.

5 The actual temperature may be selected in view of the softening temperature of the petroleum pitch. In some examples, the low-oxygen environment may have an oxygen concentration from ranging from about 1 mol% to about 20 mol%, or about 1 mol% to about 15 mol%, or about 1 mol% to about 10 mol%, or about 1 mol% to about 5 mol%. Such heating below the softening temperature of the petroleum pitch in the presence of a low-oxygen environment may result in
10 a crosslinking reaction that may stabilize the petroleum pitch and desirably raise the softening temperature prior to carbonization.

[0031] Following carbonization achieved by heating the petroleum pitch in the composite powders to a suitable temperature, compositions produced from the composite powders may comprise up to about 60 wt.% silicon (elemental silicon or silicon dioxide) particles dispersed
15 in a carbon matrix, wherein the carbon matrix results from carbonization of the pitch. Depending on the carbonization conditions, the carbon matrix may comprise amorphous carbon, graphite, or any combination thereof. Preferably, the carbon matrix may comprise or consist essentially of amorphous carbon. Preferably, the carbon matrix may remain in predominantly particle form, although a small amount of particle consolidation may take place
20 when carbonizing the composite powders.

[0032] In non-limiting examples, the composite powders may be heated at a temperature ranging from about 700°C to about 1800°C, or about 900°C to about 1800°C, or about 900°C to about 1500°C, or about 900°C to about 1400°C, or about 1000°C to about 1500°C. Such temperatures may form a carbon matrix comprising amorphous carbon, which may be
25 accompanied by silicon carbide formation at higher temperatures of about 1500°C or above. In some or other examples, the petroleum pitch may be converted to graphite by heating the composite powders (or a carbon matrix resulting from carbonization of the composite powders) at about 2000°C to about 3500°C, or about 2000°C to about 3400°C, or about 2500°C to about 3400°C, or about 2500°C to about 3000°C, or about 3000°C to about 3400°C, or about 2800°C
30 to about 3200°C. A graphitization catalyst may be present, if desired, to promote graphitization, particularly at lower temperatures within these ranges. Heating to promote graphitization may take place in a no-oxygen or very low-oxygen environment comprising about 0.1 mol% oxygen or below, preferably in the presence of an inert gas.

[0033] Dry blending methods for making compositions described herein allow for single-step processing of materials to form precursor composites having a powder form, thereby ensuring appropriate particle sizes as well as an appropriate dispersion of silicon particles therein. Precursor composites obtained by dry blending, preferably cold blending, may combine the plurality of silicon particles, the petroleum pitch, and the optional graphite in a manner to limit unwanted oxidation by keeping the temperature relatively low while still achieving a satisfactory dispersion of the silicon particles. Combining the components of the precursor composites in the foregoing manner may comprise milling, extruding, grinding, the like, or any combination thereof.

[0034] In non-limiting examples, the composite powders disclosed herein may be formed by milling or grinding a suitable petroleum pitch and suitable silicon and graphite particles, preferably during a continuous milling or grinding process, more preferably a milling or grinding process conducted in a screw mill extruder. Without being bound by theory, milling or grinding may suitably combine the silicon and graphite particles and the petroleum pitch into a well-dispersed state and additionally reduce the particle size of the individual components (*e.g.*, the silicon particles, the petroleum pitch, the optional graphite, and the like) within the composite powders. Thus, silicon particles introduced to the screw mill extruder need not necessarily reside within the final size range present in the composite powders. For example, the silicon particles introduced to the screw mill extruder may be up to about 10 microns in size, or up to about 5 microns in size, or up to about 1 micron in size, or up to about 500 nm in size and undergo a reduction in size into the size ranges described above for the composite powder. The screw mill may be any suitable size and configuration for achieving a suitable dispersion of the silicon particles within the petroleum pitch and achieving a desired decrease in size. It is to be appreciated that the composite powders may also be produced in related processes that are non-continuous (batch) processes, such as ball or sand milling.

[0035] Accordingly, methods of the present disclosure may comprise forming a blend comprising up to about 50 wt.% silicon particles and about 15 wt.% to about 95 wt.% petroleum pitch, and optionally about 0.1 wt.% to about 85 wt.% graphite, each based upon a total mass of the blend, and processing the blend under grinding conditions to form a composite powder, as discussed in more detail above. Under the grinding conditions (dry blending conditions) the silicon particles become dispersed in a matrix comprising the petroleum pitch and the petroleum pitch comprises a plurality of pitch particles.

[0036] Preferably, the grinding conditions may comprise use of an extruder, such as a screw mill extruder, such that the composite powder is formed under continuous extrusion

conditions. The extrusion may take place under dry blending conditions below the softening temperature of the petroleum pitch. Preferably, the composite powders may be formed under dry blending conditions to decrease energy usage relative to melt blending processes. In some examples, dry blending in an extruder may take place with cooling of the extruder to a temperature below room temperature, such as at a temperature ranging from about -10°C to about 5°C, or about -10°C to about 0°C, or about -5°C to about 5°C, or about -5°C to about 0°C when processing the blend to form the composite powder. Non-continuous grinding processes such as ball or sand milling may likewise take place at a temperature below room temperature as well. As indicated above, such dry blending processes may result in dispersion of the silicon particles between the pitch particles, such as within the interstitial space between pitch particles.

[0037] Alternately, the composite powders of the present disclosure may be formed under melt blending conditions above the softening temperature of the petroleum pitch. Such melt blending processes may likewise be performed in a screw mill extruder under continuous grinding conditions or batchwise using ball or sand milling above the softening temperature of the petroleum pitch, such that the silicon particles are reduced in size and at least a portion of the silicon particles become incorporated within the petroleum pitch. As the petroleum pitch subsequently cools below the softening temperature and grinding is continued, the petroleum pitch may be ground into a particulate form with all or a majority of the silicon particles dispersed within the interior of the pitch particles. Depending on the melt blending conditions, at least some silicon particles may remain within the interstitial spaces between the pitch particles and/or at least partially embedded within the outer surface of the pitch particles.

[0038] It is to be appreciated that combination grinding processes are also contemplated in the present disclosure. For example, two or more extruders in series may be utilized to achieve a desired particle size or extent of blending and/or two or more extruders in parallel may be utilized to increase throughput. In some or other examples, a ball or jet milling process may follow an extrusion process or vice versa when producing the composite powders described herein. In still other examples, a dry blending process may follow a melt blending process to produce composite powders having silicon particles dispersed both within the interior of the pitch particles and between the pitch particles.

[0039] Embodiments disclosed herein include:

[0040] A. Composite powders. The composite powders comprise: up to about 50 wt.% silicon particles, based on total mass of the composite powder, and about 15 wt.% to about 95 wt.% petroleum pitch, based on total mass of the composite powder; and up to about 80

wt.% graphite particles, based on total mass of the composite powder; wherein the silicon particles are dispersed in a matrix comprising the petroleum pitch and the petroleum pitch comprises a plurality of pitch particles.

5 [0041] B. Compositions obtained from composite powders. The compositions comprise: up to about 60 wt.% silicon particles dispersed in a carbon and graphite matrix, based on total mass of the composition; wherein the carbon matrix comprises amorphous carbon.

[0042] B1. A battery anode comprising the composition of B.

[0043] B2. A lithium-ion battery comprising the battery anode of B1.

10 [0044] C. Methods for making composite powders. The methods comprise: forming a blend comprising up to about 50 wt.% silicon particles and about 15 wt.% to about 95 wt.% petroleum pitch and up to about 80 wt.% graphite particles, based on a total mass of the blend; and processing the blend under grinding conditions to form a composite powder; wherein the silicon particles are dispersed in a matrix comprising the petroleum pitch and the petroleum pitch comprises a plurality of pitch particles.

15 [0045] D. Melt blending methods for making composite powders. The methods comprise: forming a blend comprising up to about 50 wt.% silicon particles and about 15 wt.% to about 95 wt.% petroleum pitch, and up to 80 wt.% graphite particles, based on a total mass of the blend; and processing the blend under grinding conditions above a softening temperature of the petroleum pitch to form a composite powder after cooling and obtaining the composite powder
20 from an outlet of a screw mill extruder; wherein the silicon particles are dispersed in a matrix comprising the petroleum pitch and the petroleum pitch comprises a plurality of pitch particles having the silicon particles dispersed therein. Optionally, at least a portion of the silicon particles may remain external to the pitch particles.

25 [0046] Embodiments A-D may have one or more of the following additional elements in any combination:

[0047] Element 1: wherein the petroleum pitch comprises about 50 wt.% or greater mesophase pitch.

[0048] Element 1A: wherein the petroleum pitch comprises about 80 wt.% or greater mesophase pitch.

30 [0049] Element 2: wherein the composite powder further comprises about 0.1 wt.% to about 85 wt.% graphite, based on total mass of the composite powder; wherein the matrix further comprises the graphite.

[0050] Element 2A: wherein the composition further comprises about 0.1 wt.% to about 85 wt.% graphite, based on total mass of the composition; wherein the carbon matrix further comprises the graphite.

5 [0051] Element 3: wherein the silicon particles are dispersed in an interstitial space between the pitch particles.

[0052] Element 4: wherein at least a portion of the silicon particles are embedded in an outer surface of the pitch particles.

[0053] Element 5: wherein at least a majority of the silicon particles are located within an interior of the pitch particles.

10 [0054] Element 6: wherein the pitch particles have a particle size ranging from about 1 μm to about 25 μm .

[0055] Element 6A: wherein the pitch particles have a particle size ranging from about 5 μm to about 25 μm .

15 [0056] Element 7: wherein at least a majority of the silicon particles have a size ranging from about 50 nm to about 200 nm.

[0057] Element 8: wherein the composite powder comprises about 2 wt.% to about 30 wt.% silicon particles.

20 [0058] Element 9: wherein the composition is produced by a process comprising: providing the composite powder of A; and heating the petroleum pitch at a temperature sufficient to form the carbon matrix in an environment comprising about 0.1 mol% oxygen or below, optionally wherein the carbon matrix is in a particle form.

[0059] Element 10: wherein the temperature ranges from about 700°C to about 1800°C.

25 [0060] Element 11: wherein processing the blend under the grinding conditions comprises mixing the blend in a screw mill extruder under dry blending conditions and obtaining the composite powder from an outlet of the screw mill extruder.

[0061] Element 12: wherein the dry blending conditions take place below a softening temperature of the petroleum pitch.

[0062] Element 13: wherein the screw mill extruder is cooled to a temperature of about -10°C to about 5°C when processing the blend.

30 [0063] Element 14: wherein processing the blend under the grinding conditions comprises ball-milling the blend.

[0064] Element 15: wherein the silicon particles have a size up to about 500 nm before processing the blend under the grinding conditions, and a size of the silicon particles after processing the blend under the grinding conditions ranges from about 50 nm to about 200 nm.

[0065] Element 16: wherein the method further comprises heating the composite powder at a temperature ranging from about 200°C to about 450°C.

5 [0066] Element 17: wherein the method further comprises at least partially carbonizing the composite powder at a temperature ranging from about 700°C to about 1800°C in an environment comprising about 0.1 mol% oxygen or below.

[0067] Element 18: wherein processing the blend under the grinding conditions comprises mixing the blend in a screw mill extruder under melt blending conditions above a softening temperature of the petroleum pitch, cooling the blend after mixing, and obtaining the composite powder from an outlet of the screw mill extruder.

10 [0068] By way of non-limiting example, exemplary combinations applicable to A-C include, but are not limited to, 1 and 2 or 2A; 1 and 3; 1, 3, and 4; 1 and 5; 1 and 6; 1 and 7; 1 and 8; 3 and 4; 3 and 6; 3 and 7; 3 and 8; 3, 4, and 6; 3, 4, and 7; 3, 4, and 8; 5 and 6; 5 and 7; 5 and 8; and 7 and 8. With respect to C, any one of 1-8 may be in further combination with any one or more of 9-17. Additional exemplary combinations applicable to C include, but are not
15 limited to, any of the foregoing combinations in further combination with 9; 9 and 10; 11; 11 and 12; 11 and 13; 14; 15; 16; 17; or 18. Still additional exemplary combinations applicable to C include, but are not limited to, 11 and 12; 11 and 13; 11 and 15; 11 and 16; 11 and 17; 14 and 15; 14 and 16; 14 and 17; 15 and 16; 15 and 17; 16 and 17; 15 and 18; 16 and 18; and 17 and 18.

20 [0069] Additional embodiments disclosed herein include those defined by the following non-limiting clauses:

[0070] Clause 1. A composite powder comprising: up to about 50 wt.% silicon particles, based on total mass of the composite powder; and about 15 wt.% to about 95 wt.% petroleum pitch, based on total mass of the composite powder; and up to about 80 wt.% graphite particles,
25 based on total mass of the composite powder; wherein the silicon particles are dispersed in a matrix comprising the petroleum pitch and the petroleum pitch comprises a plurality of pitch particles.

[0071] Clause 2. The composite powder of clause 1, wherein the petroleum pitch comprises about 50 wt.% or greater mesophase pitch.

30 [0072] Clause 3. The composite powder of clause 1 or clause 2, further comprising: about 0.1 wt.% to about 85 wt.% graphite, based on total mass of the composite powder; wherein the matrix further comprises the graphite.

[0073] Clause 4. The composite powder of any one of clauses 1-3, wherein the silicon particles are dispersed in an interstitial space between the pitch particles.

[0074] Clause 5. The composite powder of any one of clauses 1-4, wherein at least a portion of the silicon particles are embedded in an outer surface of the pitch particles.

[0075] Clause 6. The composite powder of any one of clauses 1-3, wherein at least a majority of the silicon particles are located within an interior of the pitch particles.

5 [0076] Clause 7. The composite powder of any one of clauses 1-6, wherein the pitch particles have a particle size ranging from about 1 μm to about 25 μm .

[0077] Clause 8. The composite powder of any one of clauses 1-7, wherein at least a majority of the silicon particles have a size ranging from about 50 nm to about 200 nm.

10 [0078] Clause 9. The composite powder of any one of clauses 1-8, wherein the composite powder comprises about 2 wt.% to about 30 wt.% silicon particles.

[0079] Clause 10. A composition comprising: up to about 60 wt.% silicon particles dispersed in a carbon matrix, based on total mass of the composition; wherein the carbon matrix comprises amorphous carbon.

15 [0080] Clause 11. The composition of clause 10, further comprising: about 0.1 wt.% to about 85 wt.% graphite, based on total mass of the composition; wherein the carbon matrix further comprises the graphite.

[0081] Clause 12. The composition of clause 10 or clause 11, wherein the composition is produced by a process comprising: providing the composite powder of any one of clauses 1-9; and heating the petroleum pitch to a temperature sufficient to form the carbon matrix in an
20 environment comprising about 0.1 mol% oxygen or below.

[0082] Clause 13. The composition of clause 12, wherein the temperature ranges from about 700°C to about 1800°C.

[0083] Clause 14. The composition of any one of clauses 10-13, wherein the carbon matrix is in a particle form.

25 [0084] Clause 15. A battery anode comprising the composition of any one of clauses 10-14.

[0085] Clause 16. A lithium-ion battery comprising the battery anode of clause 15.

[0086] Clause 17. A method comprising: forming a blend comprising up to about 50 wt.% silicon particles and about 15 wt.% to about 95 wt.% petroleum pitch and up to about 80 wt.%
30 graphite particles, based on a total mass of the blend; and processing the blend under grinding conditions to form a composite powder; wherein the silicon particles are dispersed in a matrix comprising the petroleum pitch and the petroleum pitch comprises a plurality of pitch particles.

[0087] Clause 18. The method of clause 17, wherein processing the blend under the grinding conditions comprises mixing the blend in a screw mill extruder under dry blending conditions and obtaining the composite powder from an outlet of the screw mill extruder.

5 [0088] Clause 19. The method of clause 18, wherein the dry blending conditions take place below a softening temperature of the petroleum pitch.

[0089] Clause 20. The method of clause 18 or clause 19, wherein the screw mill extruder is cooled to a temperature of about -10°C to about 5°C when processing the blend.

[0090] Clause 21. The method of clause 17, wherein processing the blend under the grinding conditions comprises ball-milling the blend.

10 [0091] Clause 22. The method of clause 17, wherein processing the blend under the grinding conditions comprises mixing the blend in a screw mill extruder under melt blending conditions above a softening temperature of the petroleum pitch, cooling the blend after mixing, and obtaining the composite powder from an outlet of the screw mill extruder.

15 [0092] Clause 23. The method of any one of clauses 16-22, wherein the petroleum pitch comprises about 50 wt.% or greater mesophase pitch.

[0093] Clause 24. The method of any one of clauses 16-23, wherein the blend further comprises about 0.1 wt.% to about 85 wt.% graphite, based on a total mass of the blend; wherein the matrix further comprises the graphite.

20 [0094] Clause 25. The method of any one of clauses 16-24, wherein the pitch particles have a particle size ranging from about 1 µm to about 25 µm.

[0095] Clause 26. The method of any one of clauses 16-25, wherein the silicon particles have a size up to about 500 nm before processing the blend under the grinding conditions, and a size of the silicon particles after processing the blend under the grinding conditions ranges from about 50 nm to about 200 nm.

25 [0096] Clause 27. The method of any one of clauses 16-26, wherein the composite powder comprises about 2 wt.% to about 30 wt.% silicon particles.

[0097] Clause 28. The method of any one of clauses 16-27, further comprising: heating the composite powder at a temperature ranging from about 200°C to about 450°C in an environment containing about 0.1 mol% to about 20 mol% oxygen.

30 [0098] Clause 29. The method of any one of clauses 16-28, further comprising: at least partially carbonizing the composite powder at a temperature ranging from about 700°C to about 1800°C in an environment comprising about 0.1 mol% oxygen or below.

[0099] To facilitate a better understanding of the embodiments of the present disclosure, the following examples of preferred or representative embodiments are given. In no way should the following examples be read to limit, or to define, the scope of the invention.

EXAMPLES

- 5 **[0100]** Composite powder samples were prepared by blending neat petroleum pitch, silicon or silicon dioxide (SiO₂) particles having a diameter of approximately 500 nm, and optionally graphite under dry blending or melt blending conditions in a screw mill extruder. Compositional details for samples E1-E6 are provided in Table 1 below. Dry blending (samples E1-E3) was conducted at an extruder rotation rate of 300 rpm with the extruder cooled in the
10 range of -5°C to 5°C. Melt blending (samples E4-E9) was conducted above 280°C at the same rotation rate. After obtaining the resulting composite powders, the samples were placed inside a furnace and further heated below the softening point of the petroleum pitch at a temperature between 200°C and 300°C to set the silicon particles within the pitch matrix and at least partially react oxygen with the petroleum pitch.
- 15 **[0101]** Comparative samples lacking petroleum pitch (samples C1 and C2) were ground to fine powders in a ball and/or jet mill to achieve a D50 similar to the experimental composite powders. Composite powders containing silicon were ball/jet milled with graphite at appropriate ratios to make comparative samples of graphite-pitch-silicon (samples E4 and E5). Composite powders containing silicon were added with graphite in the anode slurry to make
20 comparative samples of graphite-pitch-silicon (E10 and E11).

Table 1

Sample ID	Petroleum Pitch (wt.%)	Si (wt.%)	SiO₂ (wt.%)	Graphite (wt.%)
E1	90	10	0	0
E2	80	20	0	0
E3	70	30	0	0
E4	18	0	2	80
E5	27	3	0	70
E6	87	13	0	0
E7	97	3	0	0
E8	40	0	10	50
E9	85	15	0	0
E10	21	4	0	75

Sample ID	Petroleum Pitch (wt.%)	Si (wt.%)	SiO₂ (wt.%)	Graphite (wt.%)
E11	14	2	0	84
C1	0	0	0	100
C2	0	3	0	97

[0102] Cross-sectional SEM-BSE (scanning electron microscope-backscattered electron) images of Samples E1-E3 are shown in FIGS. 2A-2C. A uniform dispersion of silicon in the resulting composite powder was achieved at loadings up to 30% by weight. Bright spots in the SEM-BSE images are Si particles.

[0103] Carbonization of the composite powders was then conducted at temperatures of 900°C, 1100°C, 1200°C, and 1500°C. The extent of silicon carbide formation was followed using powder X-ray diffraction (XRD), as shown in FIG. 3. The patterns and peak positions of the samples heated at 900°C and 1200°C suggest the continued presence of elemental silicon.

At 1500°C, the 26° peak became more prominent, which is indicative of carbon being formed from pitch, and the new peaks at 36°, 41.5°, 60°, 72° are believed to be indicative of silicon carbide formation.

[0104] Selected samples carbonized up to 1100°C were formed into half-cell coin batteries, with the carbonized sample forming the anode and the counter electrode comprising lithium.

The batteries were activated at a charging rate of 0.1 C for up to five cycles. The discharging capacity and initial coulombic efficiency (ICE) are shown in Table 2 below.

Table 2

Sample ID	1st discharging capacity (mAh/g)	ICE (%)
E1	750	80
E2	1050	83
E3	1350	84
C1	348	96
C2	435	81

Sample ID	1 st discharging capacity (mAh/g)	ICE (%)
E4	395	81
E5	428	82
E6	626	77
E7	353	79
E8	523	83
E9	484	89
E10	422	93
E11	400	84

[0105] As shown, the samples formed from silicon-containing composite powders exhibited enhanced battery capacity beyond what is theoretically predicted for pure carbon-based active materials and demonstrated in the comparative samples. The initial columbic efficiency (ICE) of both the experimental and comparative samples were all relatively high and remained above 75%.

[0106] All documents described herein are incorporated by reference herein for purposes of all jurisdictions where such practice is allowed, including any priority documents and/or testing procedures to the extent they are not inconsistent with this text. As is apparent from the foregoing general description and the specific embodiments, while forms of the disclosure have been illustrated and described, various modifications can be made without departing from the spirit and scope of the disclosure. Accordingly, it is not intended that the disclosure be limited thereby. For example, the compositions described herein may be free of any component, or composition not expressly recited or disclosed herein. Any method may lack any step not recited or disclosed herein. Likewise, the term “comprising” is considered synonymous with the term “including.” Whenever a method, composition, element or group of elements is preceded with the transitional phrase “comprising,” it is understood that we also contemplate the same composition or group of elements with transitional phrases “consisting essentially

of,” “consisting of,” “selected from the group of consisting of,” or “is” preceding the recitation of the composition, element, or elements and vice versa.

[0107] Unless otherwise indicated, all numbers expressing quantities of ingredients, properties such as molecular weight, reaction conditions, and so forth used in the present specification and associated claims are to be understood as being modified in all instances by the term “about.” Accordingly, unless indicated to the contrary, the numerical parameters set forth in the following specification and attached claims are approximations that may vary depending upon the desired properties sought to be obtained by one or more embodiments described herein. At the very least, and not as an attempt to limit the application of the doctrine of equivalents to the scope of the claim, each numerical parameter should at least be construed in light of the number of reported significant digits and by applying ordinary rounding techniques.

[0108] Whenever a numerical range with a lower limit and an upper limit is disclosed, any number and any included range falling within the range is specifically disclosed. In particular, every range of values (of the form, “from about a to about b,” or, equivalently, “from approximately a to b,” or, equivalently, “from approximately a-b”) disclosed herein is to be understood to set forth every number and range encompassed within the broader range of values. Also, the terms in the claims have their plain, ordinary meaning unless otherwise explicitly and clearly defined by the patentee. Moreover, the indefinite articles “a” or “an,” as used in the claims, are defined herein to mean one or more than one of the element that it introduces.

[0109] One or more illustrative embodiments are presented herein. Not all features of a physical implementation are described or shown in this application for the sake of clarity. It is understood that in the development of a physical embodiment of the present disclosure, numerous implementation-specific decisions must be made to achieve the developer's goals, such as compliance with system-related, business-related, government-related and other constraints, which vary by implementation and from time to time. While a developer's efforts might be time-consuming, such efforts would be, nevertheless, a routine undertaking for one of ordinary skill in the art and having benefit of this disclosure.

[0110] Therefore, the present disclosure is well adapted to attain the ends and advantages mentioned as well as those that are inherent therein. The particular embodiments disclosed above are illustrative only, as the present disclosure may be modified and practiced in different but equivalent manners apparent to one having ordinary skill in the art and having the benefit of the teachings herein. Furthermore, no limitations are intended to the details of construction

or design herein shown, other than as described in the claims below. It is therefore evident that the particular illustrative embodiments disclosed above may be altered, combined, or modified and all such variations are considered within the scope and spirit of the present disclosure. The embodiments illustratively disclosed herein suitably may be practiced in the absence of any
5 element that is not specifically disclosed herein and/or any optional element disclosed herein.

CLAIMS

What is claimed is:

1. A composite powder comprising:
up to about 50 wt.% silicon particles, based on total mass of the composite powder; and
about 15 wt.% to about 95 wt.% petroleum pitch, based on total mass of the composite powder,
and, optionally, up to about 80 wt.% graphite particles, based on total mass of the composite
powder, wherein the silicon particles are dispersed in a matrix comprising the petroleum pitch
and the petroleum pitch comprises a plurality of pitch particles.
2. The composite powder of claim 1, wherein the petroleum pitch comprises about 50
wt.% or greater mesophase pitch.
3. The composite powder of claim 1, further comprising: about 0.1 wt.% to about 85 wt.%
graphite, based on total mass of the composite powder, wherein the matrix further comprises
the graphite.
4. The composite powder of claim 1, wherein the silicon particles are dispersed in an
interstitial space between the pitch particles.
5. The composite powder of claim 4, wherein at least a portion of the silicon particles are
embedded in an outer surface of the pitch particles.
6. The composite powder of claim 1, wherein at least a majority of the silicon particles
are located within an interior of the pitch particles.
7. The composite powder of claim 1, wherein the pitch particles have a particle size
ranging from about 1 μm to about 25 μm .
8. The composite powder of claim 1, wherein at least a majority of the silicon particles
have a size ranging from about 50 nm to about 200 nm.
9. The composite powder of claim 1, wherein the composite powder comprises about 2
wt.% to about 30 wt.% silicon particles.

10. A composition comprising:
up to about 60 wt.% silicon particles dispersed in a carbon and graphite matrix, based on total mass of the composition, wherein the carbon matrix comprises amorphous carbon with a degree of graphitization up to 60%.
11. The composition of claim 10, further comprising:
about 0.1 wt.% to about 85 wt.% graphite, based on total mass of the composition;
wherein the carbon matrix further comprises the graphite.
12. The composition of claim 10, wherein the composition is produced by a process comprising:
providing the composite powder comprising up to about 50 wt.% silicon particles, based on total mass of the composite powder; and about 15 wt.% to about 95 wt.% petroleum pitch, based on total mass of the composite powder, and, optionally, up to about 80 wt.% graphite particles, based on total mass of the composite powder, wherein the silicon particles are dispersed in a matrix comprising the petroleum pitch and the petroleum pitch comprises a plurality of pitch particles; and
heating the composite powder to a temperature sufficient to form the carbon matrix in an environment comprising about 0.1 mol% oxygen or below.
13. The composition of claim 12, wherein the temperature ranges from about 700°C to about 1800°C.
14. The composition of claim 10, wherein the carbon matrix is in a particle form.
15. A battery electrode comprising the composition of claim 10.
16. A lithium-ion battery comprising the battery electrode of claim 15.
17. A method comprising:
forming a blend comprising up to about 50 wt.% silicon particles and about 15 wt.% to about 95 wt.% petroleum pitch, and optionally up to 80 wt.% graphite particles, based on a total mass of the blend; and

reducing an average particle size of the blend to form a composite powder, wherein the silicon particles are dispersed in a matrix comprising the petroleum pitch and the petroleum pitch comprises a plurality of pitch particles.

18. The method of claim 17, wherein the blend further comprises a graphitization catalyst.

19. The method of claim 17, wherein reducing an average particle size of the blend comprises mixing the blend in a screw mill extruder under dry blending conditions and obtaining the composite powder from an outlet of the screw mill extruder.

20. The method of claim 18, wherein the dry blending conditions take place below a softening temperature of the petroleum pitch.

21. The method of claim 18, wherein a screw mill extruder is cooled to a temperature of about -10°C to about 5°C when processing the blend.

22. The method of claim 17, wherein reducing an average particle size of the blend comprises ball or jet-milling the blend.

23. The method of claim 17, wherein reducing an average particle size of the blend comprises mixing the blend in a screw mill extruder under melt blending conditions above a softening temperature of the petroleum pitch, cooling the blend after mixing, and obtaining the composite powder from an outlet of the screw mill extruder.

24. The method of claim 17, wherein the petroleum pitch comprises about 50 wt.% or greater mesophase pitch.

25. The method of claim 17, wherein the blend further comprises about 0.1 wt.% to about 85 wt.% graphite, based on a total mass of the blend;
wherein the matrix further comprises the graphite.

26. The method of claim 17, wherein the pitch particles have a particle size ranging from about 1 μm to about 25 μm .

27. The method of claim 17, wherein the silicon particles have a size up to about 500 nm before reducing an average particle size of the blend, and a size of the silicon particles after reducing an average particle size of the blend ranges from about 50 nm to about 200 nm.

28. The method of claim 17, wherein the composite powder comprises about 2 wt.% to about 30 wt.% silicon particles.

29. The method of claim 17, further comprising:
heating the composite powder at a temperature ranging from about 200°C to about 450°C in an environment containing about 0.1 mol% to about 20 mol% oxygen.

30. The method of claim 17, further comprising:
at least partially carbonizing the composite powder at a temperature ranging from about 700°C to about 1800°C in an environment comprising about 0.1 mol% oxygen or below to at least partially convert the petroleum pitch to carbon with a degree of graphitization up to 60%.

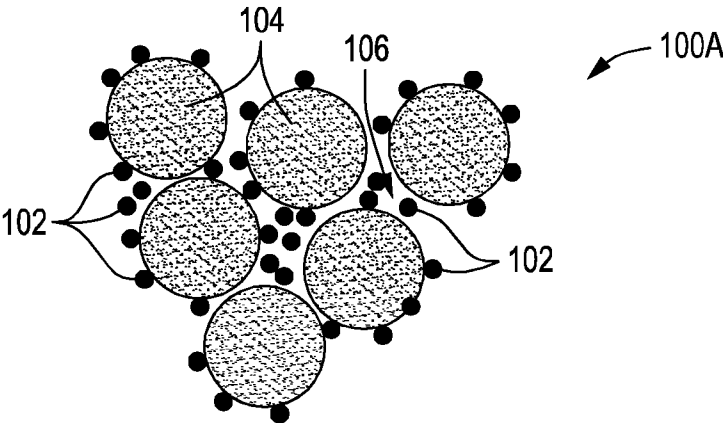


FIG. 1A

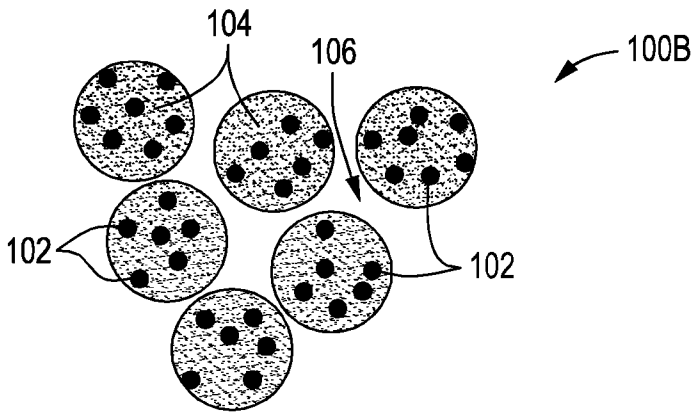


FIG. 1B

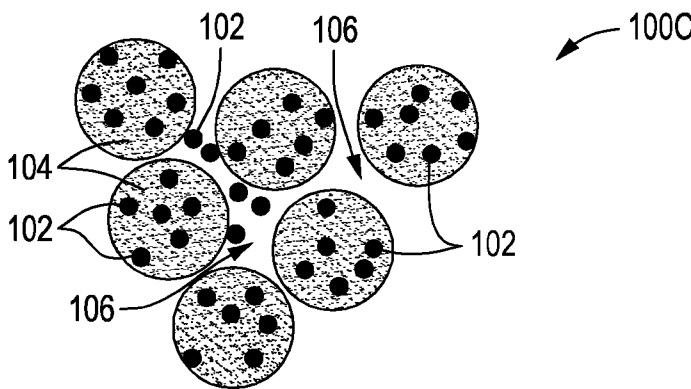


FIG. 1C

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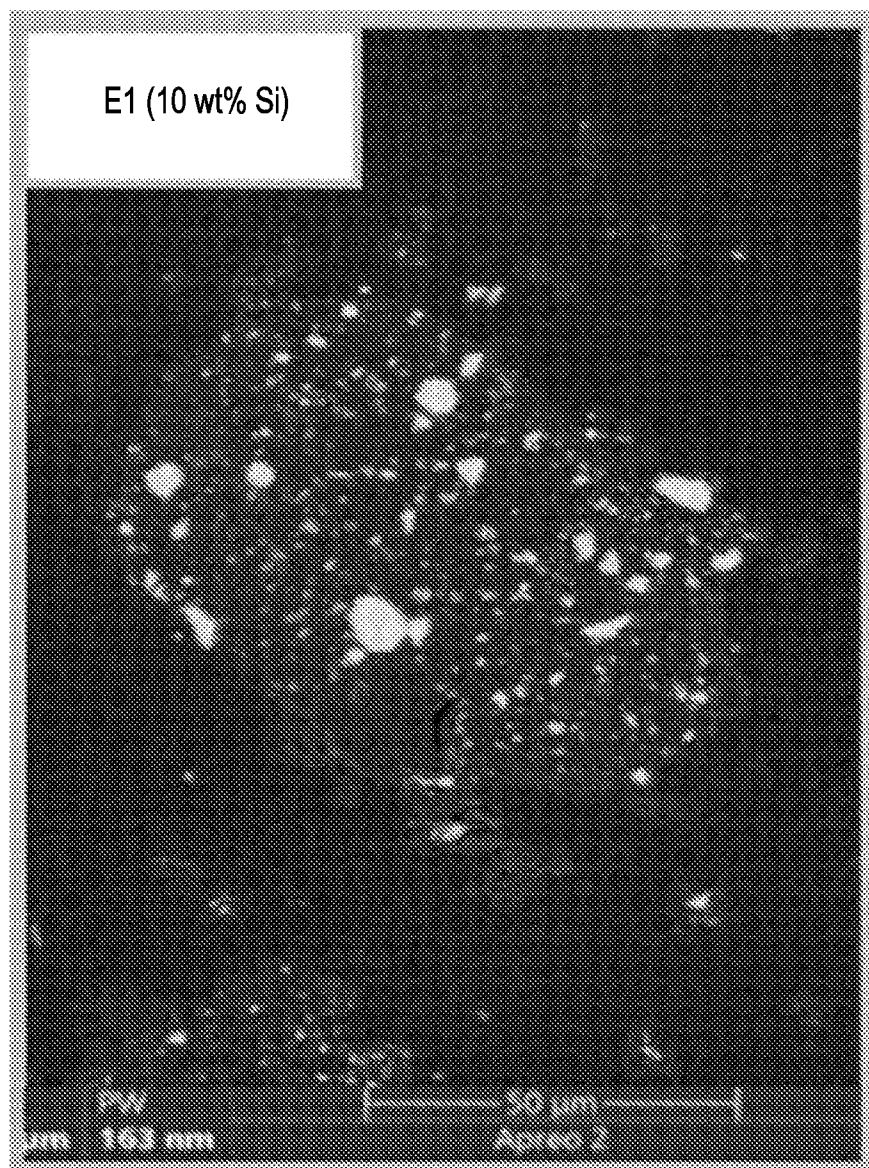


FIG. 2A

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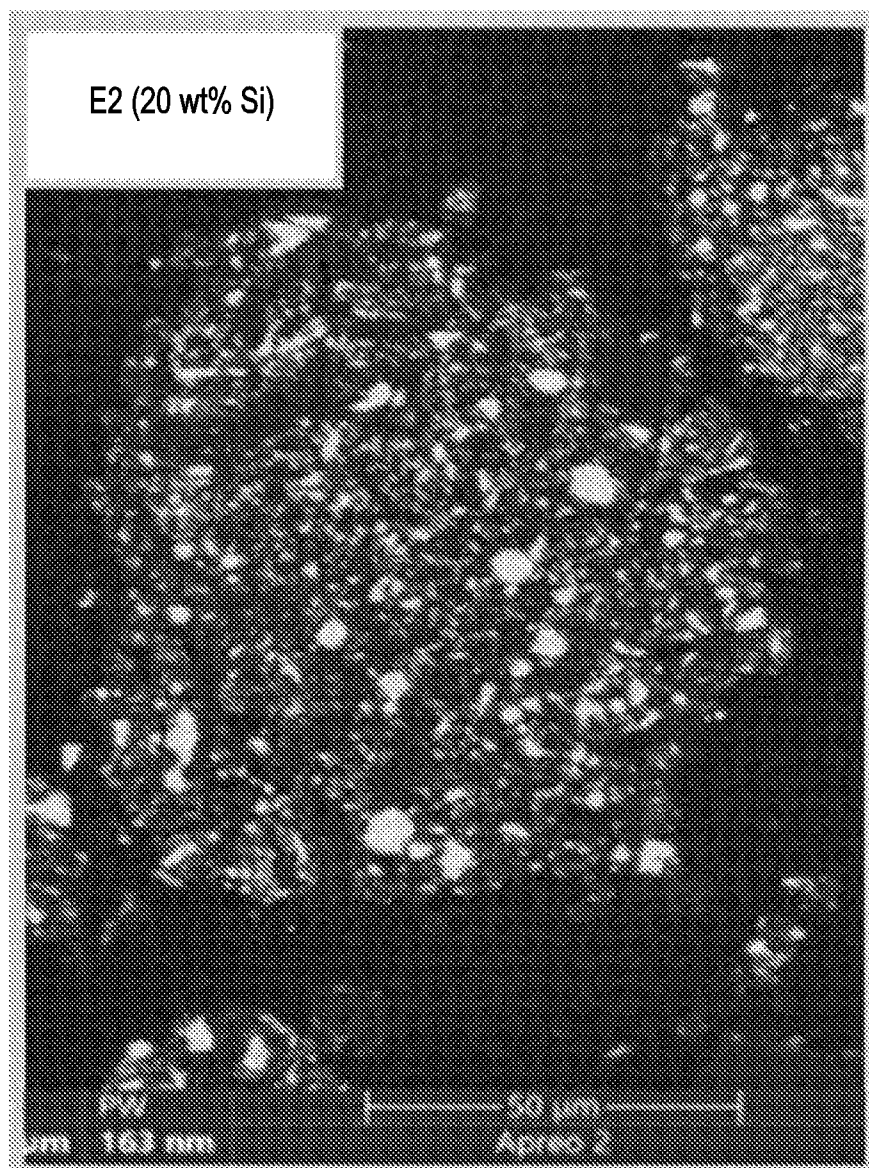


FIG. 2B

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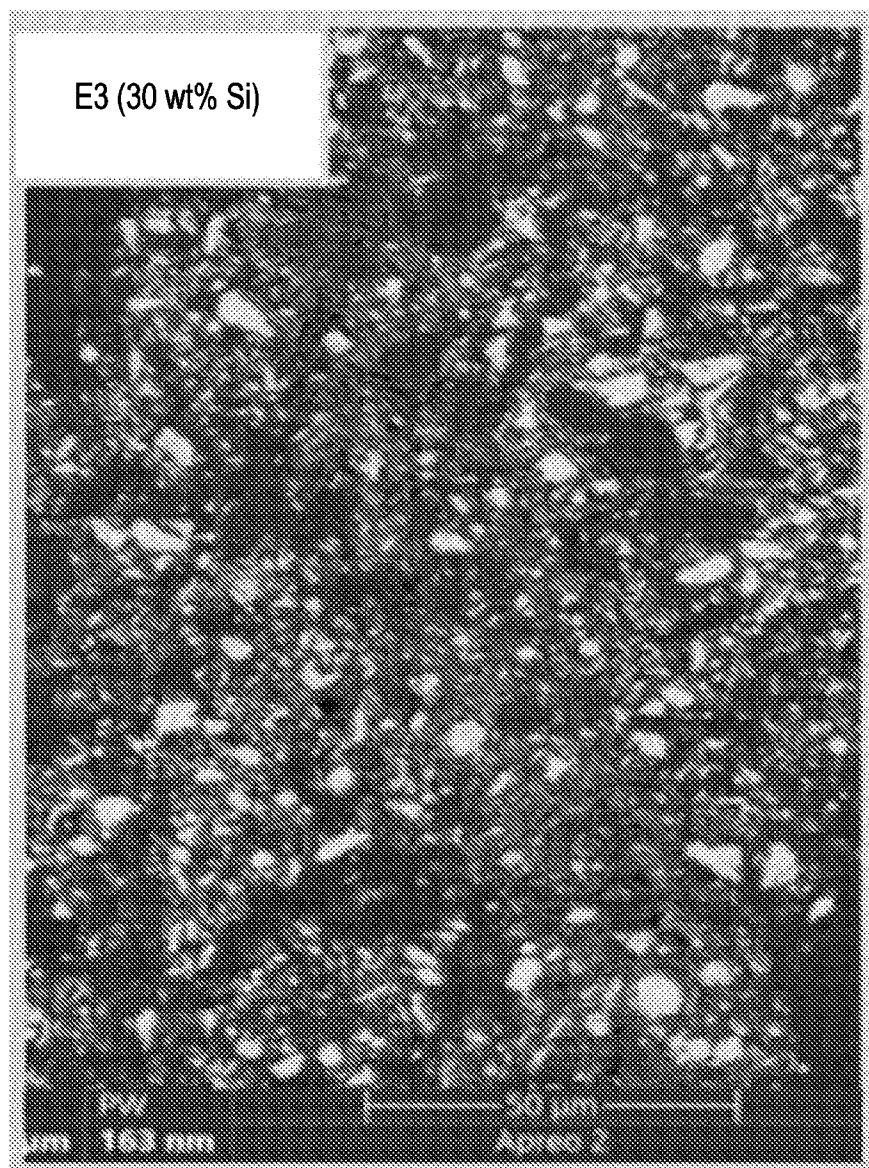


FIG. 2C



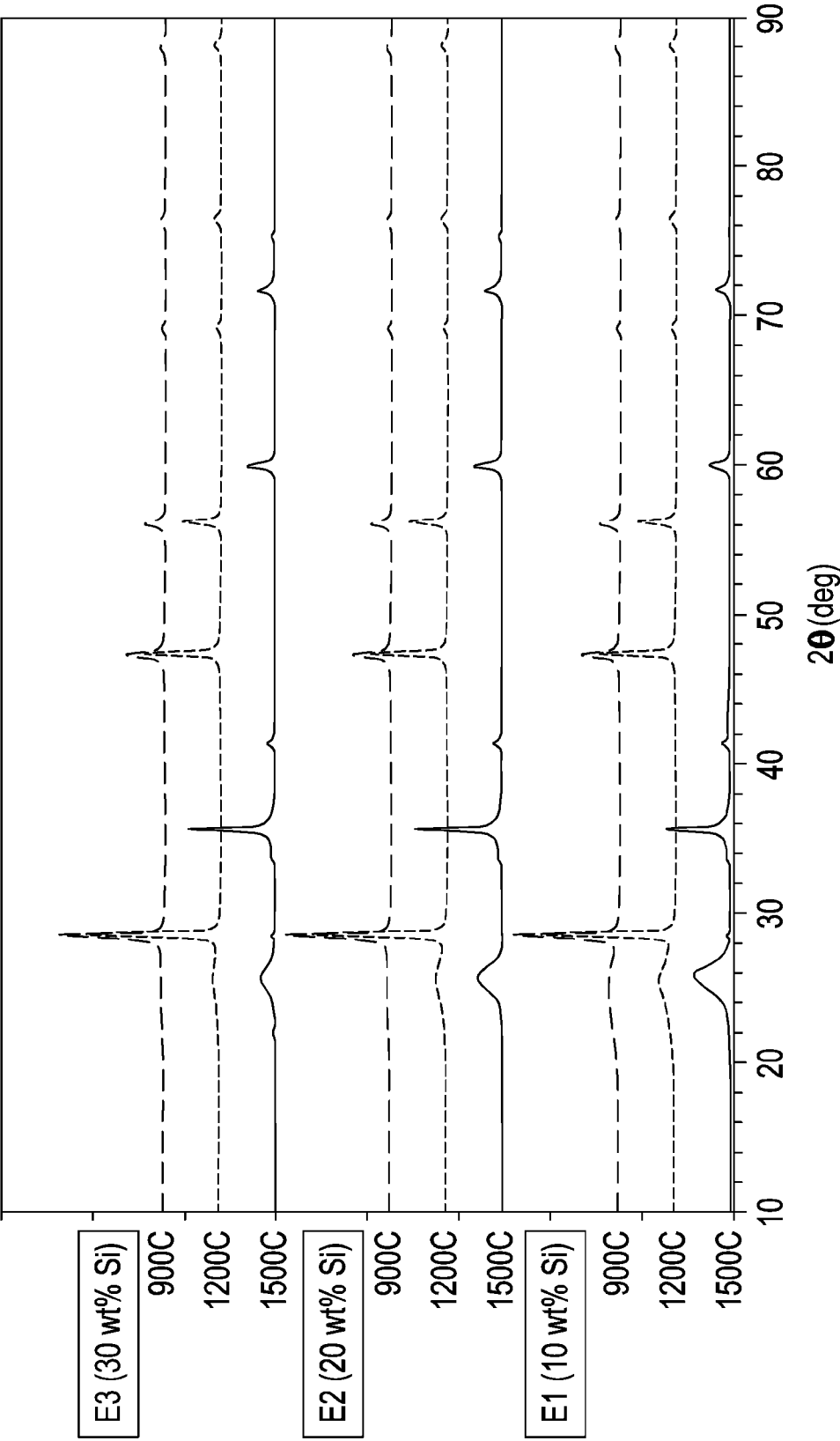


FIG. 3