% nitrogen, 0 atomic % sulfur, and 13-22 atomic % oxygen. It is believed that the reduced graphene oxide in this sample was produced by a modified Hummer's and subsequent chemical reduction. For the sake of convenience, this material is referred to as "GRAPHENEA RGO" herein.

A second such reference material was obtained from Thomas Swan & Co. Ltd. (Rotary Way, Consett, County Durham, DH8 7ND, United Kingdom) under the trademark "ELICARB GRAPHENE." The datasheet for this material is available at http://www.thomas-swan.co.uk/advanced-materials/elicarb%C2%AE-graphene-products/elicarb%C2%AE-graphene. According to this datasheet, the graphene in this sample was produced by solvent exfoliation and particle size is in the 0.5 to 2.0 micrometer range. For the sake of convenience, this material is referred to as "ELICARB GRAPHENE" herein.

A third such reference material is expanded graphite (EG), produced by thermal expansion of conventional graphite intercalation compounds that are typically produced by chemical oxidation. One example expanded graphite is "L2136," a non-commercial material made available by Schunk Hoffmann Carbon Technologies AG (Au 62, 4823 Bad Goisern am Hallstättersee, Austria). The company does not disclose details about the manufacturing at the present time. For the sake of convenience, this material is referred to as "L2136" herein.

SUMMARY

Graphene and the production of graphene, including an apparatus and a method for expansion of graphite to graphene, are described herein.

In a first example, a composition includes dehydrogenated graphite comprising a plurality of flakes. The flakes have at least one flake in 10 having a size in excess of 10 square micrometers, an average thickness of 10 atomic layers or less, and a defect density

characteristic of at least 50% of μ-Raman spectra of the de-hydrogenated graphite collected at 532 nm excitation with a resolution better than 1.8 reciprocal centimeters having a D/G area ratio below 0.5.

In a second example, a composition includes dehydrogenated graphite comprising a plurality of flakes having at least one flake in 10 having a size in excess of 10 square micrometers, a coefficient of determination value of 2D single peak fitting of μ-Raman spectra of the de-hydrogenated graphite collected at 532 nm excitation with a resolution better than 1.8 reciprocal centimeters larger than 0.99 for more than 50 % of the spectra, and a defect density characteristic of at least 50% of μ-Raman spectra of the de-hydrogenated graphite collected at 532 nm excitation with a resolution better than 1.8 reciprocal centimeters having a D/G area ratio below 0.5.

The first or second example can include one or more of the following features. More than 60%, for example, more than 80%, or more than 85 % of μ-Raman spectra of the de-hydrogenated graphite can have the coefficient of determination value larger than 0.99. More than 40%, for example, more than 50% or more than 65 % of the μ-Raman spectra of the de-hydrogenated graphite can have the coefficient of determination value larger than 0.995. At least one flake in six can have a size in excess of 10 square micrometers, for example, at least one flake in four. At least one flake in ten can have a size in excess of 25 square micrometers, for example, at least two flakes in ten. The average thickness can be seven atomic layers or less, for example, five atomic layers or less. The defect density can be characteristic of at least 80% of the collected spectra having a D/G area ratio below 0.5, for example, at least 95% of the collected spectra having a D/G area ratio below 0.5. The defect density can be characteristic of at least 80% of the collected spectra having a D/G area ratio below 0.5, for example, at least 95% of the collected spectra having a D/G area ratio below 0.5. The defect density can be characteristic of at least 50% of the collected spectra having a D/G area ratio below 0.2,

for example, at least 70% of the collected spectra having a D/G area ratio below 0.2. The defect density can be characteristic of at the average D/G area ratio being below 0.8, for example, below 0.5 or below 0.2. The composition can be a particulate powder of dehydrogenated graphite flakes, for example, a black particulate powder of dehydrogenated graphite flakes. The plurality of the flakes of the dehydrogenated graphite can be wrinkled, crumpled, or folded, for example, wherein the plurality of the flakes are assembled into a 3-dimensional structure. The full width half maximum of the G peak in μ-Raman spectra of the de-hydrogenated graphite collected at 532 nm excitation with a resolution better than 1.8 reciprocal centimeters can be larger than 20 reciprocal centimeters, for example, larger than 25 reciprocal centimeters or larger than 30 reciprocal centimeters. The μ-Raman spectra of the de-hydrogenated graphite collected at 532 nm excitation with a resolution better than 1.8 reciprocal centimeters can show a broad peak in the range between 1000 and 1800 reciprocal centimeters with a full width half maximum of more than 200 reciprocal centimeters, for example more than 400 reciprocal centimeters. More than 1%, for example, more than 5% or more than 10% of the flakes can be of a thickness of more than 10 atomic layers. The composition can be a composite, for example, wherein the composite further includes activated carbon or wherein the composite further includes a polymer. The composition can be a composite and at least 30%, for example, at least 50% or at least 70% % of sp3 hybridized carbon sites of the composition are one or more of functionalized with a non-hydrogen chemical group, cross-linked with sp3 hybridized carbon sites of another flakes, or otherwise chemically modified.

An electrode can include the composite of the first or the second example. The electrode can be part of a battery or an electrochemical capacitor, for example, a lithium battery, a lithium-ion battery, a silicon anode battery, or a lithium sulfur battery.

In a third example, a composition can include hydrogenated graphite comprising a plurality of flakes. The flakes can have at least one flake in 10 having a size in excess of 10 square micrometers, an average thickness of 10 atomic layers or less, and a defect density characteristic of μ-Raman spectra of the hydrogenated graphite collected at 532 nm excitation with a resolution better than 1.8 reciprocal centimeters and an excitation power below 2 mW at the focus of an 100X objective having an average D/G area ratio being between 0.2 and 4, wherein the majority of the defects are reversible hydrogenation of sp3-hybridized carbon sites away from the edges of the flakes.

In a third example, a composition can include a reversibly hydrogenated graphite comprising a plurality of flakes having at least one flake in 10 having a size in excess of 10 square micrometers, a coefficient of determination value of 2D single peak fitting of μ-Raman spectra of the graphite after thermal treatment in inert atmosphere at 2 mbar and 800 °C, collected at 532 nm excitation with a resolution better than 1.8 reciprocal centimeters, of larger than 0.99 for more than 50 % of the spectra, and a defect density characteristic of μ-Raman spectra of the hydrogenated graphite collected at 532 nm excitation with a resolution better than 1.8 reciprocal centimeters and an excitation power below 2 mW at the focus of an 100X objective having an average D/G area ratio being between 0.2 and 4. The majority of the defects are reversible hydrogenation of sp3-hybridized carbon sites away from the edges of the flakes.

The third example and the fourth example can include one or more of the following features. More than 60%, for example, more than 80%, or more than 85 % of μ-Raman spectra of the graphite can have the coefficient of determination value larger than 0.99. More than 40%, for example, more than 50% or more than 65 % of the μ-Raman spectra of the graphite can have the coefficient of determination value larger than 0.995. At least one flake in six can have a size in excess of 10 square micrometers, for example, at least one flake in four. At least one flake in ten can have a size in excess of 25 square

micrometers, for example, at least two flakes in ten. The average thickness can be seven atomic layers or less, for example, five atomic layers or less. The defect density can be characteristic of at least 50% of the μ-Raman spectra collected at 532 nm excitation with a resolution better than 1.8 reciprocal centimeters and an excitation power below 2 mW at the focus of an 100X objective having a D/G area ratio above 0.5, for example, at least 80% or at least 95% of the collected spectra having a D/G area ratio above 0.5. The defect density can be characteristic of at least 50% of the collected spectra having a D/G area ratio above 0.8, for example, at least 60% or at least 90% of the collected spectra having a D/G area ratio above 0.8. The defect density can be characteristic of the average D/G area ratio being between 0.4 and 2, for example, between being 0.8 and 1.5. At least 60%, for example, at least 75% of the defects can be reversible hydrogenation of sp3-hybridized carbon sites away from the edges of the flakes. The composition can be a composite and at least 5%, for example, at least 10%, of sp3 hybridized carbon sites of the composition can be one or more of a) functionalized with a chemical group, b) cross-linked with sp3 hybridized carbon sites of another flakes, or c) otherwise chemically modified.

In a fifth example, an apparatus for the expansion of the graphite to graphene includes at least one container provided for receiving an electrolyte, at least one anode and at least one cathode, wherein the cathode contains diamond or consists thereof.

In one aspect, the present disclosure refers to an apparatus for expanding graphite into graphene, comprising: a container into which graphite particles and an electrolyte are introduced; a cathode disposed in the container; an anode disposed in the container; a separator separating the cathode and the anode in the container; and a voltage supply configured to apply a pulsed DC-voltage between 5V and 60V between the anode and the cathode, wherein the voltage supply is configured to apply the pulsed DC-voltage such that hydrogen is generated at the cathode, the generated hydrogen intercalated between lattice planes of the graphite particles and / or chemisorbed on the surface of the graphite particles, thereby exfoliating graphene flakes from the graphite particles.

The fifth example can include one or more of the following features. The apparatus can include a separator which separates the anode from the cathode. The separator can be in contact with the surface of the anode or the separator can be diamond and/or polytetrafluoroethylene and/or Al2O3 and/or ceramic and/or quartz and/or glass contains or consists thereof. The can include a drive means with which the separator, and optionally the anode, are rotatable. The apparatus can include a separator, and optionally an anode, that are displaceably mounted so that the distance between the cathode and the

separator is changeable in operation of the apparatus. The apparatus can include an electric voltage supply set up to apply a DC voltage of from about 5 V to about 60 V between the anode and cathode, or from about 15 V to about 30 V, wherein the voltage is optionally pulsed. The apparatus can include a feed apparatus by which electrolyte and graphite particles can be fed as a dispersion into the at least one container and/or a discharge apparatus by which electrolyte and graphene flakes are dischargable from the at least one container as a dispersion.

In a sixth example, a method for the expansion of the graphite to graphene includes introducing graphite particles and at least one electrolyte into at least one container, applying an electrical voltage to at least one anode and at least one cathode so that the graphite is expanded, wherein the cathode contains or consists of diamond and hydrogen is produced at the cathode.

In one aspect, the present disclosure refers to a method for expanding graphite into graphene, the method comprising: introducing graphite particles and an electrolyte into a container; expanding the graphite by applying a voltage to an anode and a cathode, including producing hydrogen at the cathode, wherein the hydrogen is intercalated into and / or chemisorbed onto the graphite particles, thereby exfoliating the graphene flakes from the graphite particles.

The sixth example can include one or more of the following features. Hydrogen can be intercalated in the graphite particles and/or chemisorbed on the graphite particles, so that graphene flakes are exfoliated from the graphite particles. The anode can be separated from the cathode by a separator. The separator can contain or consist of diamond and/or polytetrafluoroethylene and/or Al2O3 and/or ceramic and/or quartz and/or glass. The separator, and optionally the anode, can be set into rotation and/or in that the separator, and optionally the anode are shifted so that the distance between the cathode and the separator changes during operation of the apparatus. An electrical voltage from about 5 V to about 60 V, or an electrical voltage from about 10 V to about 50 V, or an electrical voltage from about 12 V to about 45 V, or an electric voltage from about 15 V to about 30 V can be applied between the anode and cathode. Graphite particles can be supplied continuously to the container and/or that graphene flakes are removed continuously from the container. The graphene flakes can be photo-treated for dehydrogenation, for example, wherein the photo-treating can include illuminating the graphene flakes with

WHAT IS CLAIMED IS:

1. An apparatus for expanding graphite into graphene, comprising:

a container into which graphite particles and an electrolyte are introduced;

a cathode disposed in the container;

an anode disposed in the container;

a separator separating the cathode and the anode in the container; and

a voltage supply configured to apply a pulsed DC-voltage between 5V and 60V between the anode and the cathode,

wherein the voltage supply is configured to apply the pulsed DC-voltage such that hydrogen is generated at the cathode, the generated hydrogen intercalated between lattice planes of the graphite particles and / or chemisorbed on the surface of the graphite particles, thereby exfoliating graphene flakes from the graphite particles.

2. The apparatus of claim 1, wherein the anode contains diamond.

3. The apparatus according to claim 1 or 2, wherein the voltage supply is configured to supply a pulsed DC-voltage of between 15V to 30V between the anode and the cathode.

4. The apparatus according to any one of claims 1 to 3, wherein the separator is in contact with a surface of the anode and configured to apply pressure.

5. The apparatus according to any one of claims 1 to 4, wherein the separator contains diamond and / or polytetrafluoroethylene and / or Al2O3 and / or ceramic and / or quarts and / or glass.

6. The apparatus according to any one of claims 1 to 5, further comprising a driving unit, wherein the separator and the anode are coupled to be rotated by the driving unit.

7. The apparatus according to any one of claims 1 to 6, wherein the separator and the anode are displaceably mounted such that a distance between the cathode and the separator can be changed during operation of the device.

8. The apparatus according to any one of claims 1 to 7, further comprising a supply device capable of supplying an electrolyte and graphite particles as a dispersion to the container.

9. The apparatus according to any one of claims 1 to 8, further comprising a discharge device capable of discharging the electrolyte and the graphene flakes as a dispersion from the container.

10. A method for expanding graphite into graphene, the method comprising:

introducing graphite particles and an electrolyte into a container;

expanding the graphite by applying a voltage to an anode and a cathode, including producing hydrogen at the cathode, wherein the hydrogen is intercalated into and / or chemisorbed onto the graphite particles, thereby exfoliating the graphene flakes from the graphite particles.

11. The method of claim 10, wherein the anode contains diamond.

12. The method according to claim 10 or 11, wherein the anode is separated from the cathode by a separator.

13. The method of claim 12, wherein:

the separators contain diamond and / or polytetrafluoroethylene and / or Al2O3 and / or ceramic and / or quarts and / or glass.

14. The method according to claim 12 or 13, wherein the separator and the anode can be configured to rotate.

15. The method according to any one of claims 12 to 14, further comprising displacing the separator or the anode during operation, thereby changing a distance between the cathode and the separator during operation.

16. The method according to any one of claims 10 to 15, wherein the voltage between 5V and 60V is applied between the anode and the cathode.

17. The method according to any one of claims 10 to 15, wherein the voltage between 10V and 50V is applied between the anode and the cathode.

18. The method according to any one of claims 10 to 15, wherein the voltage between 12V and 45V is applied between the anode and the cathode.

19. The method according to any one of claims 10 to 15, wherein the voltage between 15V and 30V is applied between the anode and the cathode.

20. The method according to any one of claims 10 to 19, further comprising continuously feeding the graphite particles into the container.

21. The method according to any one of claims 10 to 20, further comprising continuously removing the graphene flakes from the container.

22. The method according to any one of claims 10 to 21, further comprising photo-treating the graphene flakes to dehydrogenate the graphene flakes, for example, the light treatment further comprises illuminating the graphene flakes with visible light, UV or microwave; and greater than 50% of the hydrogenated sp3 hybrid-carbon sites are dehydrogenated.

23. The method according to any one of claims 10 to 22, comprising a subsequent heat treatment of the graphene flakes at a temperature of from 100 °C to 800 °C for a period of from 1 minute to 60 minutes.

24. The method of claim 23, wherein in the subsequent heat treatment step, the treatment is performed at a temperature of 300 ° C to 650 ° C.

25. The method according to claim 23 or 24 wherein, in the subsequent heat treatment step, the treatment is performed for a time period of 15 minutes to 40 minutes.

26. The method according to any one of claims 10 to 25, wherein the graphene flakes have an average surface area of greater than 10 μ m2.

27. The method according to any one of claims 10 to 25, wherein the graphene flakes have an average surface area of greater than 50 μ m2.

28. The method according to any one of claims 10 to 25, wherein the graphene flakes have an average surface area of greater than 100 μ m2.

29. The method according to any one of claims 10 to 28,wherein the applied voltage is a pulsed DC voltage.