% 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 one aspect of the present disclosure, there is provided a composition comprising:

dehydrogenated graphite comprising a plurality of flakes having

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 another aspect of the present disclosure, there is provided a composition comprising:

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.

In a further aspect of the present disclosure, there is provided an electrode comprising the composition disclosed herein.

In another aspect of the present disclosure, there is provided a battery or an electrochemical capacitor comprising the electrode disclosed herein, for example, wherein the battery is a lithium battery, a lithium-ion battery, a silicon anode battery, or a lithium sulfur battery.

In a further aspect of the present disclosure, there is provided a composition comprising:

hydrogenated graphite comprising a plurality of flakes having

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 another aspect of the present disclosure, there is provided a composition comprising:

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, wherein the majority of the defects are reversible hydrogenation of sp3-hybridized carbon sites away from the edges of the flakes.

In a further aspect of the present disclosure, there is provided an apparatus for the expansion of the graphite to graphene with at least one container provided for receiving an electrolyte, at least one anode and at least one cathode, characterized in that the cathode contains diamond or consists thereof.

In another aspect of the present disclosure, there is provided a method for the expansion of the graphite to graphene, wherein graphite particles and at least one electrolyte are introduced into at least one container and the graphite, through the application of an electrical voltage to at least one anode and at least one cathode is expanded,

characterized in that the cathode contains or consists of diamond and hydrogen is produced at the cathode.

The present disclosure provides a composition including 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.

The present disclosure also provides a composition including 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 present disclosure 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 disclosed herein. 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.

The present disclosure further provides a composition including 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 the present disclosure, 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 present disclosure 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 the present disclosure, 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.

The present disclosure 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 the present disclosure, 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.

The present disclosure 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

31. The composition of any of claims 21 to 30, wherein at least 60%, for example, at least 75% of the defects are reversible hydrogenation of sp3-hybridized carbon sites away from the edges of the flakes.

32. The composition of any of claims 21 to 31, wherein the composition is a composite and at least 5%, for example, at least 10%, of sp3 hybridized carbon sites of the composition are one or more of:

a) functionalized with a non-hydrogen chemical group,

b) cross-linked with sp3 hybridized carbon sites of another flakes, or

c) otherwise chemically modified.

33. An apparatus for the expansion of the graphite to graphene with at least one container provided for receiving an electrolyte, at least one anode and at least one cathode, characterized in that the cathode contains diamond or consists thereof.

34. The apparatus of claim 33, further comprising a separator which separates the anode from the cathode.

35. The apparatus according to claim 33 or 34, characterized in that the separator is in contact with the surface of the anode or that the separator is diamond and/or polytetrafluoroethylene and/or Al2O3 and/or ceramic and/or quartz and/or glass contains or consists thereof.

36. The apparatus according to one of claims 33 to 35, further comprising a drive means with which the separator, and optionally the anode, are rotatable.

37. The apparatus according to one of claims 33 to 36, characterized in that the separator, and optionally the anode are displaceably mounted, so that the distance between the cathode and the separator is changeable in operation of the apparatus.

38. The apparatus according to one of claims 33 to 37, further comprising 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.

39. The apparatus according to one of claims 33 to 38, further comprising a feed apparatus by which electrolyte and graphite particles can be fed as a dispersion into the at least one container and/or further comprising a discharge apparatus by which electrolyte and graphene flakes are dischargable from the at least one container as a dispersion.

40. A method for the expansion of the graphite to graphene, wherein graphite particles and at least one electrolyte are introduced into at least one container and the graphite, through the application of an electrical voltage to at least one anode and at least one cathode is expanded,

characterized in that the cathode contains or consists of diamond and hydrogen is produced at the cathode.

41. Method according to claim 40, characterized in that hydrogen is intercalated in the graphite particles and/or chemisorbed on the graphite particles, so that graphene flakes are exfoliated from the graphite particles.

42. Method according to any one of claims 40 or 41, characterized in that the anode is separated from the cathode by a separator.

43. Method according to claim 42, characterized in that the separator contains or consists of diamond and/or polytetrafluoroethylene and/or Al2O3 and/or ceramic and/or quartz and/or glass.

44. Method according to one of claims 40 to 43, characterized in that the separator, and optionally the anode are 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.

45. Method according to any one of claims 40 to 44, characterized in that 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 is applied between the anode and cathode.

46. Method according to any one of claims 40 to 45, characterized in that graphite particles are supplied continuously to the container and/or that graphene flakes are removed continuously from the container.

47. Method according to any one of claims 27 to 46, further comprising the step of photo-treating the graphene flakes for dehydrogenation, for example, wherein the photo-treating including illuminating the graphene flakes with visible light, UV, or microwaves, wherein more that 50% of hydrogenated sp3 hybridized carbon sites are de-hydrogenated.

48. Method according to any one of claims 40 to 47, further comprising the step of: subsequent thermal treatment of the graphene flakes at a temperature from about 100°C to about 800°C, or from about 300°C to about 650°C, and for a period of from about 1 min to about 60 min, or from about 15 min to about 40 min.

49. Method according to one of claims 40 to 48, characterized in that the graphene flakes have an average surface area of more than 10 um2 or more than 50 um2 or of more than 100 um2.