PRODUCTION OF GRAPHENE

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ABSTRACT

An apparatus for large-scale production of graphene and graphene oxide is provided. The apparatus includes a first electrode, a second electrode, an electrobath, a power supply, and a module for filtering and separating the graphene products. Large amounts of graphene and graphene oxide can be produced rapidly using electrochemical exfoliation. High-quality graphene and graphene oxide can be produced under the room temperature in a simple and cost-effective way.
Immerse a first electrode and a second electrode in an electrolyte, the first electrode including graphite

Apply a first voltage across the first and second electrodes to cause intercalation of the graphite to form a graphite intercalation compound

Apply a second voltage across the first and second electrodes to exfoliate the graphite intercalation compound to produce graphene sheets

Filter the electrolyte using a first filter that blocks un-exfoliated graphite particles and allows graphene sheets to pass through

Filter the electrolyte using a second filter to collect the graphene sheets

FIG. 10
PRODUCTION OF GRAPHENE

CROSS-REFERENCE TO RELATED APPLICATIONS

[0001] This application claims priority to Taiwan application 100148453, filed on Dec. 23, 2011, the content of which is incorporated by reference.

TECHNICAL FIELD

[0002] The disclosure relates to production of graphene.

BACKGROUND

[0003] A graphene sheet is composed of carbon atoms occupying a two-dimensional hexagonal lattice. Graphene can have a high carrier mobility and excellent thermal conductivity. Graphene can be used in, e.g., semiconductor devices, touch panels, and solar cells. Graphene can be fabricated by, e.g., mechanical exfoliation, epitaxial growth, chemical vapor deposition, and chemical exfoliation. Methods for producing graphene have been described in U.S. Pat. No. 7,790,285, U.S. Pat. No. 7,892,514, and U.S. patent application Ser. No. 13/170,624, filed on Jun. 28, 2011.

SUMMARY

[0004] In one aspect, an apparatus is provided for preparing high-quality graphene and graphene oxide in a simple and fast manner at a low cost. The graphene and graphene oxide can be produced by electrochemical exfoliation.

[0005] In one aspect, an apparatus for producing graphene and/or graphene oxide is provided. The apparatus includes a first electrode that includes graphite; a second electrode; a container that contains an electrolyte, in which the first and second electrodes are immersed in the electrolyte; a power supply to supply bias voltages across the first and second electrodes to cause intercalation of graphite and exfoliation of graphene; and a filtration module to separate the graphene from un-exfoliated graphite particles and collect the graphene.

[0006] In one aspect, a method for producing graphene and/or graphene oxide includes immersing a first electrode and a second electrode in an electrolyte, the first electrode including graphite; applying a first voltage across the first and second electrodes to cause intercalation of the graphite to form a graphite intercalation compound; applying a second voltage across the first and second electrodes to exfoliate the graphite intercalation compound to produce at least one of graphene or graphene oxide; filtering the electrolyte using a first filter that blocks un-exfoliated graphite particles and allows the at least one of graphene or graphene oxide to pass through; and filtering the electrolyte using a second filter to collect the at least one of graphene or graphene oxide.

[0007] In one aspect, an apparatus for producing graphene and graphene oxide includes a first electrode that has an electrode holder having a starting graphite material, a second electrode, an electro bath, a power supply, and a module for filtering and separating the graphene products.

[0008] Implementations of the apparatus can include one or more of the following features. The first electrode can be an electrode holder that includes the starting graphite material, and the second electrode can be either an electrode holder that includes the starting graphite material or a metal. The starting graphite material can include a mixture of graphite and metal. To electrochemically exfoliate graphene, the first electrode and the second electrode can be placed in an electrolyte. Intercalation of the graphite material is performed using a first bias voltage, and the exfoliation of the graphite material is performed using a second bias voltage. The solid graphene products are taken out of the electrolyte. The final graphene product is not necessarily limited to being dissolved in the electrolyte. In some examples, the final graphene product can also be collected in an electrode holder.

[0009] The starting graphite material can include natural graphite in a layered structure, artificial graphite, composite material prepared from graphite powder, or a combination of the above. The starting graphite material can include natural graphite, highly-oriented pyrolytic graphite (HOPG), pitch-based graphite, resin-based graphite, PAN-based carbon fibers, pitch-based carbon fibers, coal, a carbon material containing graphite layers, and/or a carbon material containing graphite flakes. The starting graphite material can be a crystalline graphite layer material in the form of large particles, fragments, or powder, or having an irregular shape. The starting graphite material can also be a block of graphite material made of pieces having the forms described above and held together by an electrically conductive adhesive.

[0010] To increase the efficiency of mass production of graphene, each of the two electrodes can include two or more sub-electrodes connected in parallel or in an array. Each sub-electrode can include a starting graphite material or an electrode holder that includes the starting graphite material.

[0011] The metal in the second electrode can be a precious metal that is resistant to acid and alkaline, such as platinum (Pt), silver (Ag), gold (Au), iridium (Ir), osmium (Os), palladium (Pd), rhodium (Rh) or ruthenium (Ru). Other electrically conducting material that is resistant to chemical etching, such as copper (Cu), stainless steel, graphite, glassy carbon, or conducting polymer, can also be used in the second electrode.

[0012] The electrolyte can be placed in a container made from glass, polymer material (e.g., acrylic, polypropylene, polystyrene, or polyvinyl chloride), stainless steel, or another metal material. The electrolyte can include hydrogen bromide, hydrochloric acid, or sulfuric acid. Alkaline such as potassium hydroxide or sodium hydroxide can be added to the electrolyte. The electrolyte can include an oxidant, which can include potassium bichromate, permanganic acid or potassium permanganate.

[0013] To enhance the graphene or graphene oxide production efficiency and to enhance the graphene product quality, the electrolysis process may be supplemented with heating, application of ultrasound or microwave, application of high-energy light radiation, and/or stirring the electrolyte by a rotor as needed to facilitate exfoliating of graphene or graphene oxide.

[0014] To implement a continuous production process, the exfoliated products can be fed into a module for filtering and separating the graphene products. The first component of the module is a microporous sieve having a size of about 18 mesh to 1250 mesh, preferably from 35 mesh to 500 mesh, for filtering large graphite particles that have not been exfoliated. The microporous sieve allows products having the desired sizes to pass through. The second component of the module is a filtration membrane that collects the graphene sheet product that passed through the sieve. The membrane can have a pore diameter of about 200 nm to 10 μm, preferably from about 500 nm to 5 μm. The graphene product collected from the filtration membrane can be treated with deionized water or
other ionic solutions (such as hydrochloric acid) capable of dissolving or replacing the residual ions (such as potassium ion or sulfate ion) to remove the residual electrolyte.

The bias voltage can be provided by a direct current (DC) or alternating current (AC) power supply. The power supply can be controlled in a constant voltage mode or a constant current mode. In the examples below, the power supply operates in a constant voltage mode to provide controlled bias voltages. The first bias voltage can range from 0.5 V to 10 V, preferably from 2.5 V to 5 V. The second bias voltage can range from 5 V to 220 V, preferably from 10 V to 100 V.

In one aspect, a method is provided for mass production of graphene and graphene oxide. The method includes placing the first electrode and the second electrode in an electrolyte, in which the first electrode is an electrode holder that includes a starting graphite material, and the second electrode is an electrode holder that includes a starting graphite material or a metal. The starting graphite material can include a mixture of graphite and metal. The intercalation of the starting graphite material is performed under a first bias voltage, and the exfoliation of the starting graphite material is performed under a second bias voltage. The exfoliated products are fed into a module for filtering and separating the products. The module includes a first component, which can be a microporous sieve, and a second component, which can be a filtration membrane. The graphene product and/or graphene oxide are collected from the filtration membrane.

Implementations of the method can include one or more of the following features. The first electrode and the second electrode can be used as the anode and the cathode, respectively, and can be wrapped with an electrode wire and immersed in an electrolyte. The graphite material can be subject to the intercalation step under the first bias voltage, after which the anions in the electrolyte, such as sulfate ions and nitrate ions, are intercalated into the interlayer space between two adjacent graphite layers or their grain boundary due to the electric field produced by the bias voltage. The first bias voltage can range from 0.5 V to 10 V, preferably from 2.5 V to 5 V, and the reaction time can range from about 1 minute to 30 minutes, preferably from 1 minute to 5 minutes.

Subsequently, the graphite material is subject to the exfoliation step using the second bias voltage, in which the second bias voltage is greater than the first bias voltage and can range from 5 V to 220 V, preferably from 10 V to 100 V, and there is no restriction on the reaction time.

The method can further include performing the exfoliation step for the graphite material using a third bias voltage that is different from the second bias voltage. For example, the third bias voltage and the second bias voltage may have opposite polarities or have the same polarity but different values, in which the second bias voltage and the third bias voltage may be direct current and alternating current, respectively. If the third bias voltage and the second bias voltage have opposite polarities, then the positive voltage can facilitate exfoliating and oxidizing the graphene and subsequently the negative voltage can reduce the oxidized graphene. For example, the second bias voltage can be 10 V and the third bias voltage can be −10 V, in which the reaction time is 2 seconds and 5 seconds, respectively, and the two voltages are alternatively applied for a certain period of time.

The intercalation step and the exfoliation step can be performed by switching between different first and second bias voltages, depending on the composition and acidity of the electrolyte to achieve the optimal quality and yield.

In some implementations, a first bias voltage (e.g., 0.5 V) can be applied to the electrodes for a period of time to cause intercalation, and a second bias voltage (e.g., 5 V) can be applied to the electrodes to cause exfoliation. In some implementations, during the exfoliation step, a second bias voltage (e.g., 5 V) and a third bias voltage (e.g., −5 V) are alternately applied to the electrodes, each of the second and third bias voltages being applied for a certain period of time (e.g., 2 seconds). The graphene obtained using the second method of alternating between the second and third bias voltages may have a higher quality, compared to using only the second bias voltage.

The bias voltage can be selected based on a desired production rate and quality of products. For example, when the absolute value of the bias voltage is higher, the exfoliation rate may be faster but the quality of the produced graphene may be lower. When the absolute value of the bias voltage is lower, the exfoliation rate is slower but the graphene quality may be better.

After the exfoliation step, the exfoliated products may be provided to a module for filtering and separating the products. The first component of the module is a microporous sieve (e.g., 35 mesh) for filtering un-exfoliated large graphite particles and obtaining the products in suitable sizes that pass through the sieve. The second component of the module is a filtration membrane having a pore diameter of, e.g., 400 nm for collecting the graphene sheet product passing through the sieve. The graphene product and graphene oxide collected from the second component can be treated with a large amount of deionized water to remove the residual electrolyte, or be treated with other ionic solutions (such as hydrochloric acid) capable of dissolving or replacing the residual ion (such as potassium ion or sulfate ion) to remove the residual electrolyte. The module mainly relies on an air pump to accelerate the filtration process. In some examples, filtration and separation can be achieved by vacuum filtration.

Advantages of the aspects, systems, and methods may include one or more of the following. High quality graphene and graphene oxide can be produced in mass quantities. The cost for producing the graphene and graphene oxide is low. During the production process, it is not necessary to subject the graphene or graphene oxide to a high temperature.

BRIEF DESCRIPTION OF DRAWINGS

FIG. 1 is a diagram of an apparatus for producing graphene and graphene oxide.

FIG. 2 is a diagram of an exemplary electrode holder that includes starting graphite material.

FIG. 3 is a photo showing graphene dispersed in 250 mL of DMF.

FIG. 4 is a photo showing solid graphene after separation and filtration.
FIG. 5 is a scanning electron microscope image of exemplary solid graphene powder.

FIG. 6A is an image of an exemplary graphene sheet.

FIG. 6B is a graph showing exemplary measurements by an atomic force microscope of a portion of the graphene sheet shown in FIG. 6A.

FIG. 7 is a graph showing exemplary measurements by a confocal Raman microscopic system for analyzing the bonding of graphene sheets.

FIG. 8 is a graph showing the characteristic Raman peaks of graphene oxide.

FIG. 9 is a diagram of an apparatus for producing graphene and graphene oxide.

FIG. 10 is a flowchart of an exemplary procedure for producing graphene.

**DETAILED DESCRIPTION**

Graphene sheets can be mass produced by a process that includes intercalation, exfoliation, and filtration. The intercalation and exfoliation are performed in an electrolyte. Graphene and graphene oxide are separated from un-exfoliated graphite particles by a filter having a mesh size selected to block the un-exfoliated graphite particles and allow the graphene and graphene oxide to pass. The graphene and graphene oxide are collected by a filtration membrane having a pore size smaller than the sizes of the graphene and graphene oxide to be collected.

Referring to FIG. 1, an exemplary graphene production system 100 includes a first electrode 102 and a second electrode 104. In this example, the first electrode 102 is an anode and the second electrode 104 is a cathode. Each of the electrodes 102 and 104 is wrapped with an electrode wire (not shown) and immersed in an electrolyte 106. The first electrode 102 can be made of a starting graphite material or include a holder that contains starting graphite material. The second electrode 104 can be made of a starting graphite material or metal, or include a holder that contains a starting graphite material. For example, the starting graphite material can include highly-oriented pyrolytic graphite (HOPG), pitch-based graphite, resin-based graphite, polycrylonitrile-based carbon fibers, pitch-based carbon fibers, or coal. The metal used for the second electrode 104 can be a precious metal that is resistant to chemical etching, such as platinum, silver, gold, iridium, osmium, palladium, rhodium, or ruthenium. The second electrode 104 can also be made of other conducting materials, e.g., copper, stainless steel, glassy carbon, or conducting polymer.

The graphene production system 100 can also be used to produce graphene oxides.

Each of the first electrode 102 and the second electrode 104 can be made of crystalline graphite layer material in the form of large particles, flakes, or powder, or having an irregular shape. The electrodes can also be a bulk material composed of graphite particles, flakes, or powder bonded together by an electrically conductive adhesive. In some implementations, to increase throughput in mass production, an electrode can include two or more sub-electrodes connected in parallel or in an array.

In some implementations, the electrolyte 106 can be placed in a container 108 made from glass, polymer material (e.g., acrylic, polypropylene, polystyrene, or polyvinyl chloride), stainless steel, or other metals. The electrolyte can include hydrogen bromide, hydrochloric acid, or sulfuric acid. Potassium hydroxide can be added to the electrolyte. The electrolyte can include an oxidant, such as potassium bichromate, permanganic acid or potassium permanganate.

A voltage supply 114 provides a bias voltage to the first and second electrodes 102 and 104. The voltage supply 114 can supply a direct-current (DC) or alternating-current (AC) bias voltage. When a first bias voltage (e.g., in a range from 0.5 V to 10 V) is applied to the first and second electrodes 102 and 104, ions in the electrolyte 106 can penetrate spaces between layers in the starting graphite material to form a graphite intercalation compound. This is referred to as the intercalation step. When a second bias voltage (e.g., in a range from 5 V to 220 V) is applied to the first and second electrodes 102 and 104, the graphite intercalation compound is exfoliated to form graphene sheets that are dispersed in the electrolyte 106.

The graphene sheets are then removed from the electrolyte using a filtration process. A pump 118 pumps the electrolyte 106, which contains the graphene sheets, to the filtration module 116. The filtration module 116 includes a microporous sieve 120 and a filtration membrane 122. The microporous sieve 120 filters out un-exfoliated large graphite particles and allows graphene sheets of suitable sizes to pass. For example, the microporous sieve 120 can have a size in a range from 18 mesh to 1250 mesh, preferably 35 mesh. The size of the sieve can be selected depending on the size of the exfoliated products. The filtration membrane 122 collects the graphene sheets that passed through the sieve 118. The filtration membrane 122 can have a pore diameter in a range from 200 nm to 1,200 nm, preferably 400 nm. The size of the pore diameter can be selected depending on the size of the exfoliated products. The graphene sheets collected at the filtration membrane 122 can be washed with a large amount of deionized water to remove the residual electrolyte. The graphene sheets can also be washed by using other ionic solutions (such as hydrochloric acid) capable of dissolving or replacing the residual ion (such as potassium ion or sulfite ion) to remove the residual electrolyte. In some examples, the pump 118 is stopped periodically, the filtration membrane 122 is removed from the filtration module 116 and rinsed. The graphene is removed from the filtration membrane 122. An air pump 124 provides an additional suction force to accelerate the filtration process. The electrolyte can be recycled after being filtered by the filtration module 116.

The efficiency of exfoliation can be enhanced by heating the electrolyte 106 using a temperature controller 112 and/or stirring the electrolyte 106 using a rotor 110. The efficiency of exfoliation can also be enhanced by applying ultrasound microwave to the electrolyte 106 and/or irradiating the electrolyte 106 with high-energy light.

If the starting graphite material is in a block form having an edge length of, e.g., 1 centimeter or more, the starting graphite material may be directly connected to the voltage supply 114 and be used as an electrode. Referring to FIG. 2, if the starting graphite material is fragmented or in powder form, an electrode holder 140 can be used as part of the first electrode 102 and/or the second electrode 104. The electrode holder 140 includes a separation sieve 142 that holds a starting graphite material 144, which may be crystalline graphite layer material that is fragmented or in powder form, or other formats that prevent direct connection to the voltage supply 114. The separation sieve 142 can be made of,
e.g., a filter sieve (e.g., having a pore size in a range from 0.1 mm to 5 mm), a porous membrane, or a component with pores.

The separation sieve 142 can have one or more of the following functions. The separation sieve 142 can allow dispersed starting graphite material to maintain a compact and conducting state and be electrically connected to the voltage supply 114. The separation sieve 142 can allow the starting graphite material to access the electrolyte 106 to effect electrochemical exfoliation. The separation sieve 142 can allow exfoliated graphene sheets to diffuse into the electrolyte 106 so that remaining unreacted starting graphite material can continue to react with the electrolyte. The separation sieve 142 can have a pore size that is related to the dimensions of the starting graphite material and should be configured to prevent the unreacted starting graphite material from passing through.

In some examples, the separation sieve 142 can have a pore size selected to be sufficiently small to trap the produced graphene sheets inside the separation sieve 142. This way, the graphene sheets can be collected in the electrode holder 140.

The separation sieve 142 can be made of an electrically insulating material that is resistant to acid and alkaline. For example, the separation sieve 142 can be made of glass, acrylic, polypropylene, polystyrene, polyvinyl chloride, other polymer materials, or metal materials that have been processed by insulation and anti-corrosion treatments. A metal electrode 146 electrically connects the starting graphite material to an external circuit that is electrically connected to the voltage supply 114. The metal electrode 146 applies a fixed pressure to the starting graphite material 144 so that the graphite fragments or powder are coupled more closely together to improve better conductance.

FIG. 3 is a photo 150 of a solution obtained using the graphene production system 100 (FIG. 1). The solution includes graphene sheets that are dispersed in 250 mL of dimethylformamide (DMF). The solution is sometimes referred to as a “graphene ink” and can be used to produce graphene thin films.

FIG. 4 is a photo 160 showing solid graphene powder obtained using the graphene production system 100.

The graphene sheets were observed by using a scanning electron microscope (SEM), model JEOL-6330F, and an atomic force microscope (AFM), Veeco Dimension-Ion system. FIG. 5 is a scanning electron microscope image 170 of the solid graphene sheet powder obtained using the graphene production system 100. The graphene powder includes graphene sheets stacked in layers and has a high purity.

FIG. 6A is an image 180 of a graphene sheet 182 that was produced by droplet plating graphene ink (made using the graphene production system 100) onto a silica (SiO₂) substrate. The graphene sheet 182 was observed using an atomic force microscope.

FIG. 6B is a graph 190 showing measurements made by an atomic force microscope along a line 184 on the graphene sheet 182. A curve 192 indicates that the thickness of the graphene sheet 182 is not greater than 3 nm. Additional measurements indicate that about 65% of the graphene sheet has a thickness of less than 2 nm.

FIG. 7 is a graph 200 shows data obtained from a NT-MDT confocal Raman microscopic system for analyzing the bonding of graphene sheets. In this example, a 1.6-μm thick graphene sheet (based on measurements from an atomic force microscope) was excited at a wavelength of 473 nm by the NT-MDT confocal Raman microscopic system, and the molecular bonding structure of the graphene sheet was analyzed. A curve 202 indicates that a G peak 204 at around 1580 cm⁻¹ is narrow and shows a high intensity, indicating that the graphene obtained according to the production process described above has excellent graphitization. In general, the 2D/G intensity ratio of a single-layer graphene is greater than the 2D/G intensity ratio of a double-layer graphene, and the intensity ratio of a 2D peak 206 at around 2720 cm⁻¹ to the G peak 204 is greater than that of the single-layer reduced graphene oxide produced using conventional methods. This shows the graphene produced by the graphene production system 100 has excellent graphitization.

The graphene production system 100 can also be used to produce graphene oxide by using a process similar to that for producing graphene described above, but with increased DC bias voltage for electrolysis or increased acidity for the electrolyte. In some examples, applying a bias voltage having a higher absolute value and using an electrolyte having a higher acidity level tend to produce more graphene oxide (compared to applying a bias voltage having a lower absolute value and using an electrolyte having a lower acidity level). The graphene oxide can be purified by filtration and rinsing by water. FIG. 8 is a graph showing the characteristic Raman peaks of graphene oxide prepared by using this method.

Referring to FIG. 9, an exemplary graphene production system 220 includes a controller 222 that automatically controls the bias voltages applied to the first and second electrodes 102, 104. The graphene production system 220 includes other components similar to those of the graphene production system 100 (FIG. 1).

For example, the controller 222 can have a user interface (not shown) that allows a user to select pre-stored modes of operation. The controller 222 can have a first mode of operation in which a first bias voltage is applied to the electrodes 102, 104 for a period of time to cause intercalation and a second bias voltage is applied to the electrodes 102, 104 to cause exfoliation. The controller 222 can have a second mode of operation in which a first bias voltage is applied to the electrodes 102, 104 for a period of time to cause intercalation, and a second bias voltage is alternately applied to the electrodes 102, 104 to cause exfoliation. The controller 222 may allow the user to choose between a first mode for producing higher throughput but lower quality graphene, or a second mode for producing lower throughput but higher quality graphene. The controller 222 may be programmable such that the user can set a sequence of voltages levels to be applied to the electrodes 102, 104 over time.

The graphene production system 220 may include sensors 224 that detect the amount of graphene being collected on the filtration membrane 122. The sensor signals are sent to the controller 222. The controller 222 may stop the pump 118 and initiate a process for collecting the graphene. For example, the controller 222 may control a robotic arm (not shown) to retrieve the filtration membrane 122, wash the filtration membrane 122 with deionized water, scrape the graphene off the filtration membrane 122, collect the graphene in a container, wash the filtration membrane 122 again, and place the filtration membrane 122 back in the filtration module 116. The controller 222 may start the pump 118 so that the filtration membrane 122 can continue to collect graphene sheets.
The graphene production system 220 may determine the amount of graphene produced per unit of time, and adjust the voltage applied to the electrodes 102, 104 to adjust the production rate. For example, the amount of graphene recovered from the filtration membrane 122 can be divided by the amount of time used for collection of the graphene to generate an estimate of the production rate. The graphene may be automatically determined to determine its quality, and the quality information is provided to the controller 222, which in turn adjusts the bias voltage level to adjust the quality of the graphene. By using various sensors to detect the production throughput and the graphene quality and sending the sensor information to the controller 222, the graphene production system 220 can control the production throughput and the quality of the graphene to meet preset requirements.

The controller 222 may include a programmable system having at least one programmable processor coupled to receive data and instructions from, and to transmit data and instructions to, a data storage system for storing data and instructions. The at least one programmable processor can include, e.g., general purpose microprocessors, special purpose microprocessors, or digital signal processors.

FIG. 10 is a flowchart of an exemplary procedure 230 for producing graphene. The procedure 230 may be performed by, e.g., the graphene production system 100 (FIG. 1) or the graphene production system 220 (FIG. 9).

The procedure 230 includes immersing (232) a first electrode and a second electrode in an electrolyte, the first electrode including graphite. For example, the first electrode can include natural graphite, highly-oriented pyrolytic graphite, pitch-based graphite, carbon fibers, coal, a material including graphite layers, or a material comprising graphite flakes. The first electrode can include two sub-electrodes connected in parallel. The second electrode can include a holder that holds graphite or a mixture of graphite and metal. The metal can include a precious metal that is resistant to acid. The second electrode can include two sub-electrodes connected in parallel. The electrolyte can include hydrobromide, hydrochloric acid, or sulfuric acid.

A first bias voltage is applied (234) across the first and second electrodes to cause intercalation of the graphite to form a graphite intercalation compound. For example, the first bias voltage can be in a range from 0.5 V to 10 V. The first bias voltage can be a DC voltage or an AC voltage.

A second voltage is applied (236) across the first and second electrodes to exfoliate the graphite intercalation compound to produce graphene sheets. For example, the second bias voltage can be in a range from 5 V to 220 V. The second bias voltage can be a DC voltage or an AC voltage.

The electrolyte is filtered (238) using a first filter that blocks un-exfoliated graphite particles and allows graphene sheets to pass through. For example, the first filter can include a microporous sieve. The microporous sieve can have a size in a range from 18 mesh to 1,250 mesh.

The electrolyte is filtered (240) using a second filter to collect the graphene sheets. The second filter can include a filtration membrane. The filtration membrane can have a pore diameter in a range from 200 nm to 1,200 nm.

While this specification contains many specific implementation details, these should not be construed as limitations on the scope of any inventions or of what may be claimed, but rather as descriptions of features specific to particular embodiments of particular inventions. Certain features that are described in this specification in the context of separate embodiments can also be implemented in combination in a single embodiment. Conversely, various features that are described in the context of a single embodiment can also be implemented in multiple embodiments separately or in any suitable subcombination.

Similarly, while operations are depicted in the drawings in a particular order, this should not be understood as requiring that such operations be performed in the particular order shown or in sequential order, or that all illustrated operations be performed, to achieve desirable results. In certain circumstances, multitasking and parallel processing may be advantageous. Moreover, the separation of various system components in the embodiments described above should not be understood as requiring such separation in all embodiments.

A number of implementations of the invention have been described. Nevertheless, it will be understood that various modifications can be made without departing from the spirit and scope of the invention. In some cases, the actions recited in the claims can be performed in a different order and still achieve desirable results. In addition, the processes depicted in the accompanying figures do not necessarily require the particular order shown, or sequential order, to achieve desirable results. Other implementations and applications are also within the scope of the following claims.

What is claimed is:

1. An apparatus for producing at least one of graphene or graphene oxide, the apparatus comprising:
   a first electrode that includes graphite;
   a second electrode;
   a container that contains an electrolyte, in which the first and second electrodes are immersed in the electrolyte;
   a power supply to supply bias voltages across the first and second electrodes to cause intercalation of graphite and exfoliation of graphene; and
   a filtration module to separate the graphene from un-exfoliated graphite particles and collect the graphene.

2. The apparatus of claim 1 in which the first electrode comprises an electrode holder that holds graphite.

3. The apparatus of claim 2 in which the second electrode comprises an electrode holder that holds graphite.

4. The apparatus of claim 2 in which the electrode holder comprises a separation sieve having a pore size selected to allow the electrolyte to pass but prevent un-exfoliated graphite from passing.

5. The apparatus of claim 4 in which the separation sieve has a pore size selected to prevent a portion of the graphene from passing.

6. The apparatus of claim 1 in which the second electrode is made of metal.

7. The apparatus of claim 6 in which the metal comprises a precious metal that is resistant to acid.

8. The apparatus of claim 1 in which the second electrode is made of a mixture of graphite and a metal.

9. The apparatus of claim 1 in which the first electrode comprises at least one of natural graphite, highly-oriented pyrolytic graphite, pitch-based graphite, carbon fiber, coal, a material comprising graphite layers, or a material comprising graphite flakes.

10. The apparatus of claim 1 in which the first electrode comprises two sub-electrodes connected in parallel.

11. The apparatus of claim 10 in which the second electrode comprises two sub-electrodes connected in parallel.
12. The apparatus of claim 1 in which the electrolyte comprises at least one of hydrogen bromide, hydrochloric acid, or sulfuric acid.
13. The apparatus of claim 1, comprising an air pump to pump the electrolyte through the filtration module.
14. The apparatus of claim 1 in which the filtration module comprises a microporous sieve and a filtration membrane.
15. The apparatus of claim 14 in which the microporous sieve has a size in a range from 18 mesh to 1250 mesh.
16. The apparatus of claim 14 in which the filtration membrane has a pore diameter in a range from 200 nm to 1,200 nm.
17. The apparatus of claim 1, comprising a controller to control the power supply to provide a first bias voltage to cause intercalation and a second bias voltage to cause exfoliation.
18. The apparatus of claim 1, comprising a controller to control the power supply to provide a first bias voltage during an intercalation step, and alternately provide a second bias voltage and a third bias voltage during an exfoliation step.
19. The apparatus of claim 18 in which the second bias voltage and the third bias voltage have opposite polarities.
20. A method for producing graphene, comprising:
immersing a first electrode and a second electrode in an electrolyte, the first electrode comprising graphite;
applying a first voltage across the first and second electrodes to cause intercalation of the graphite to form a graphite intercalation compound;
applying a second voltage across the first and second electrodes to exfoliate the graphite intercalation compound to produce at least one of graphene or graphite oxide;
filtering the electrolyte using a first filter that blocks unexfoliated graphite particles and allows the at least one of graphene or graphite oxide to pass through; and
filtering the electrolyte using a second filter to collect the at least one of graphene or graphite oxide.
21. The method of claim 20 in which the first filter comprises a microporous sieve.
22. The method of claim 20 in which the second filter comprises a filtration membrane.
23. The method of claim 20 in which the second electrode comprises a holder that holds graphite or a mixture of graphite and metal.
24. The method of claim 20 in which the first electrode comprises at least one of natural graphite, highly-oriented pyrolytic graphite, pitch-based graphite, carbon fibers, coal, a material comprising graphite layers, or a material comprising graphite flakes.
25. The method of claim 20 in which the first electrode comprises two sub-electrodes connected in parallel.
26. The method of claim 25 in which the second electrode comprises two sub-electrodes connected in parallel.
27. The method of claim 20 in which the metal comprises a precious metal that is resistant to acid.
28. The method of claim 20 in which the electrolyte comprises at least one of hydrogen bromide, hydrochloric acid, or sulfuric acid.
29. The method of claim 20, comprising an air pump to pump the electrolyte and cause the electrolyte to pass the first and second filters.
30. The method of claim 20 in which the first voltage is in a range from 0.5 V to 10 V.
31. The method of claim 20 in which the second voltage is in a range from 5 V to 220 V.
32. The method of claim 20 in which the voltage comprises a DC voltage.
33. The method of claim 20 in which the voltage comprises an AC voltage.
34. The method of claim 20 in which the first filter comprises a microporous sieve having a size in a range from 18 mesh to 1,250 mesh.
35. The method of claim 20 in which the second filter comprises a filtration membrane having a pore diameter in a range from 200 nm to 1,200 nm.
36. The method of claim 20, comprising providing a third voltage during an exfoliation step, the third voltage having a polarity that is opposite to that of the second voltage.
37. The method of claim 36, comprising during the exfoliation step, alternately providing the second voltage and the third voltage across the first and second electrodes.