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United States Patent 12385129
Kind Code B2
Date of Patent August 12, 2025
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Graphene vapor deposition system and process

Abstract

A graphene vapor deposition system and process are disclosed. The system includes a support for a copper-plated sheet, a housing defining an interior region, a hydraulic cylinder to move the housing between a first and a second position, a pump to evacuate the interior region, carbon powder within the interior region, and a heat source to vaporize the carbon powder, for causing graphene vapor deposition on the copper. The process includes dissolving the copper and recovering the graphene.

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Family ID: **1000008751548**

Appl. No.: **18/187908**

Filed: **March 22, 2023**

Prior Publication Data

Document Identifier	**Publication Date**
US 20230416908 A1	Dec. 28, 2023

Related U.S. Application Data

us-provisional-application US 63366776 20220622

Publication Classification

Int. Cl.: **C23C16/26** (20060101); **C23C16/01** (20060101); **C23C16/02** (20060101)

U.S. Cl.:

CPC **C23C16/26** (20130101); **C23C16/01** (20130101); **C23C16/0209** (20130101);

Field of Classification Search

CPC: C23C (16/26); C23C (16/01); C23C (16/0209); C23C (14/0605); C23C (14/025); C23C (14/243); C23C (14/5873)

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Background/Summary

REFERENCE TO RELATED APPLICATION

(1) This nonprovisional application is based upon U.S. provisional application Ser. No. 63/366,776 filed Jun. 22, 2022, incorporated by reference in its entirety.

FIELD

(2) The present subject matter, in general, relates to graphene production and, more particularly, relates to a graphene vapor deposition system and process.

BACKGROUND

(3) Graphene refers to a two-dimensional material having a beehive-like lattice arrangement formed by carbon hybrid orbitals. Graphene is optically transparent and has extraordinarily high thermal conductivity and electron mobility properties at room temperature (i.e., from about 68 to 72 degrees Fahrenheit) as well as atomic levels of low thickness and high mechanical strength. These exceptional properties have provided graphene with unique, extensive industrial applicability in the functioning of products including photonic and electronic parts, fuel cells, electrochemical products, sensory devices, field emission, hydrogen storage, and power-supplying materials.

(4) Currently known methods of producing graphene include, for example, the following: (1) in a separation method, individual graphene plates are separated from a graphite crystal by mechanical or chemical means and their combination; and the size of assorted graphene plates synthesized from this method generally tend to be smaller than a graphite crystal; therefore, they are not suitable for use in large-area applications; (2) in another method, silicon carbide is heated to a high temperature to remove silicon, which results in single-layer or multilayer graphene; however, graphene made by this method cannot be adapted for uses on non-silicon carbide substances; moreover, it can cause problems when required to produce large area graphene sheets of uniform thickness; (3) chemical vapor deposition is currently the most popular, known preparation method for making graphene; yet, an inclination to produce monolayer and multilayer polycrystalline graphene having island-like, small crystalline domains and grain boundaries, make it difficult to achieve flat mono layer graphene having large crystalline domains, or large area monolayer graphene sheet.

(5) As there currently is no known way to produce flat, mono layer graphene having large crystalline domains, or large area monolayer graphene sheets, it can be appreciated that a process for producing flat, mono layer graphene having large crystalline domains or large area monolayer graphene sheets efficiently, is desirable.

SUMMARY

(6) The present subject matter is directed to a graphene vapor deposition system and process. A process for producing graphene according to an embodiment of the present subject matter may include the following steps. A sheet of metal could be electroplated with copper. Such a sheet of metal would be selected to not dissolve in an acidic solution (e.g., nitric acid or sulfuric acid in which copper dissolves), for producing sheets of graphene. The copper-plated sheet when transferred to a table, conveyor belt, or transfer medium, can be aligned with hydraulic cylinders supported above, which can be activated for moving a vacuum assembly down onto a copper-

plated sheet on the transfer belt, table, or conveyor. At least one tray may contain carbon powder within the vacuum assembly. Vacuum may be initiated, and a primary heat source activated to heat the interior of a vacuum assembly. Once vacuum is achieved, a secondary heat source may be activated to vaporize carbon by raising the temperature to at least the carbon vaporization temperature. Vaporized carbon next attaches to the copper plate via vapor deposition, thereby producing graphene. After a desired amount of graphene forms on a treated region of plate, a vacuum release valve releases vacuum, and the vacuum assembly is raised. The conveyor belt or table can move a plate to different positions, for desired graphene build-up. The plate can be moved to overlapping positions to fill gaps left by a first graphene application. Vacuum can be released again, the vacuum assembly raised, and the copper plated sheet transferred to a copper dissolving solution tank. Graphene can be separated from plates and stored. Process steps could be repeated, as needed.

Description

BRIEF DESCRIPTION OF THE DRAWING FIGURES

(1) FIG. **1** presents an exploded view of the components of a system and process for producing graphene according to principles of the present subject matter.

(2) FIG. **2** is a cross-sectional, side elevational view of a graphene-producing plated feature of the present subject matter, on an enlarged scale relative to FIG. **3**.

(3) FIG. **3** is a side elevational view, partially in section, of the system and process for producing graphene according to principles of the present subject matter.

(4) FIG. **4** is a cross-sectional, side elevational view of a feature of the present subject matter shown in FIG. **2**, with graphene particles now formed thereon.

(5) FIG. **5** is another cross-sectional, side elevational view of the subject matter shown in FIG. **4**, after its transfer into a metal (e.g., copper) dissolving tank.

(6) FIG. **6** is a diagram of an illustrative process presenting exemplary steps for producing graphene in accordance with principles of the present subject matter.

(7) FIG. **7** is an exploded view of the components of a graphene production system according to another embodiment of the present invention.

DETAILED DESCRIPTION

(8) Graphene produced by a system or apparatus according to embodiments of the present subject matter could be produced using subtractive manufacturing techniques. For instance, graphene produced by a system or apparatus according to embodiments of the present subject matter could be formed to a predetermined shape or size to optimize efficiency. The graphene could next be laser cut, to size. Hydraulics, lasers, heat sources, carbon, vacuum pumps, and conveyors may be purchased from commercial sources. To create a vacuum seal, please refer to “How to make a vacuum seal” on YouTube. Suitable vacuum seals must be heat resistant.

(9) Graphene vapor deposited onto copper-plated sheet is separated from the copper-plated sheet by dissolving copper on which the graphene was deposited.

(10) In some embodiments, mobile robotic arms on guide tracks may be used to transfer plates through the production process. A mobile plating tank and a mobile plate stacking table, a mobile dissolving fluid tank and a mobile vacuum plate holder table may be automated, computer controlled, and chain driven. For example, a large robotic arm may be used to transfer a plate into an automated electro-plating tank under a vacuum housing and to transfer it to a vacuum press table (or vice versa). Carbon may form on the plate on the vacuum table. The large robotic arm may transfer the carbonized plate onto a dissolving tank to remove carbon. the dissolving tank may be fitted with anti-sloshing guards. The large robotic arm may stack clean vacuum plates onto a plate holding table and may place the clean vacuum plates onto the electro-plating tank. Once the plate

holding table is full, it may move autonomously or manually to a position in which the robotic arm can transfer plates from the stacking table to the electroplating tank. The electro-plating tank may be mounting on a mobile table that may move into line for plating and may move out of the way for a subsequent plating tank to move into position. In some cases, dozens of automated plating tanks may take turns moving into position. Small robotic arms may be used to place carbon and/or radioactive blocks (e.g., nuclear batteries) onto a carbon holder.

(11) Referring initially to FIG. **1** a graphene production system **10** according to an embodiment of the present subject matter includes a press frame (not shown) and an elongated table **14**. The system **10** also includes an elongated copper-plated sheet of metal **12** disposed longitudinally atop the table **14**. The system **10** further includes a plurality of vacuum housings **20** disposed above the copper-plated sheet of metal **12**. The system **10** includes an associated plurality of hydraulic cylinders **28**. Two of the plural hydraulic cylinders **28** are associated with each one of the plural vacuum housings **20**. Each of the plural hydraulic cylinders **28** has a main body portion **101**, a first end portion **102** secured to the press frame, and a second end portion **104** opposite the first end portion **102**. The second end portion **104** of each of the plural hydraulic cylinders **28** is extendable from and retractable into the main body portion **101**. A distal end of the second end portion **104** of each of the plural hydraulic cylinders **28** defines a through bore **108** disposed transverse to the direction of extension-and-retraction of the second end portion **104**. An upper surface **110** of each of the plural vacuum housings **20** includes a spaced-apart pair of mounts **112** unitary with the upper surface **110**. Each of the two mounts **112** of each of the plural vacuum housings **20** defines a through bore **114** oriented to align with the through bore **108** of the second end portion **104** of an associated hydraulic cylinder **28**, so that a pin (not shown) is passed though the bores **108** and **114** for securing a spaced-apart pair of hydraulic cylinders **28** to each vacuum housing **20**.

(12) The system **10** includes a plurality of primary heating sources **16** and a plurality of carbon trays **18** disposed between the elongated copper-plated sheet of metal **12** and the plurality of vacuum housings **20**. Three of the plural primary heating sources **16** and two of the plural carbon trays **18** are disposed beneath each one of the plural vacuum housings **20**. The system **10** also includes a plurality of vacuum pumps **24** and a plurality of secondary heating sources **26** disposed between the plurality of vacuum housings **20** and the plurality of hydraulic cylinders **28**. The upper surface **110** of each vacuum housing **20** defines a spaced apart pair of through bores **116** sized and configured to receive an end portion of each one of the spaced-apart pair of secondary heating sources **26** through the upper surface **110** (FIG. **3**) of each associated one of the plural vacuum housings **20**. The upper surface **110** of each one of the plural vacuum housings **20** defines a recess **118** which is dimensioned and configured to receive a lower end portion of one of the plural vacuum pumps **24**.

(13) Referring next to FIGS. **1-4**, additional features and advantages of the present subject matter shall now be described in detail. The elongated copper-plated sheet of metal **12** (FIG. **2**) includes an upper layer **12**B of a metal such as copper that will dissolve in an acid such as nitric acid or sulfuric acid and a lower layer **12**A of a metal that will not dissolve in an acid such as nitric acid or sulfuric acid. For instance, while most metals dissolve in nitric acid (HNO3), the following precious metals: gold (Au), platinum (Pt), and palladium (Pd) do not dissolve even in concentrated HNO3. Moreover, certain other non-precious metals, namely, iron (Fe), nickel (Ni), chromium (Cr), and aluminum (Al) do not dissolve in HNO3 because of the formation of an oxide layer on the surface of these metals, which prevents further reaction with HNO3. To manufacture an elongated copper-plated sheet of metal **12** (FIG. **2**), the upper may be **12**B could be electroplated onto the lower layer **12**A. Each vacuum housing **20** defines an interior region **120** (FIG. **3**) and includes a vacuum seal **22** (FIGS. **1**, **3**) along an underside surface. When operative, the second end portion **104** of each of the plural hydraulic cylinders **28** (FIG. **1**) is connected to an associated mount **112** (unitary with the upper surface **110**) of a vacuum housing **20**; and each pair of hydraulic cylinders **28** is actuated, to extend the spaced-apart pair of extendable second end portions **104**, to

move the associated vacuum housing **20** downward to provide a vacuum seal at an upper surface of upper layer **12**B (FIG. **2**).

(14) Before initiating graphene production, the plural carbon trays **18** (FIG. **1**) are arranged above the copper-plated sheet of metal **12** a predetermined distance by means (not shown); and each tray **18** is provided a block **18**A of carbon powder, as shown in FIG. **3**. (Only one tray **18** is shown.) For operational purposes, each of the vacuum pumps **24** includes an evacuation line **24**B (FIG. **3**) used to evacuate the interior region **120** of an associated vacuum housing **20**; and each evacuation line **24**B includes a common “open/close” valve **24**A to be used as needed. When the interior region **120** of an associated vacuum housing **20** is sufficiently evacuated, the primary heaters **16** (only two are shown in FIG. **3**) and the secondary heaters **26** (only one is shown) are used to heat the block **18**A of carbon powder, converting carbon powder to vaporized carbon **18**B (FIG. **3**), which initially forms graphene deposits **36**A (FIG. **4**) on the upper layer **12**B of the copper-plated sheet of metal **12**. The primary heaters **16** and the secondary heaters **26** are each powered by a source (not shown). During operation of the system **10** (FIG. **1**), the graphene deposits **36**A ultimately form a graphene layer **36** on the upper layer **12**B of the sheet of metal **12**.

(15) Referring next to FIGS. **1** and **5**, additional features and advantages of the present subject matter shall now be described in detail. After a graphene layer **36** has formed to predetermined thickness on the upper layer **12**B of a sheet of metal **12**, the graphene layered sheet of metal **12** is transferred into a copper dissolving tank **32**, noted by step **1** (“S**1**”) in FIG. **1**; another elongated copper-plated sheet of metal **12** is moved from a plate loading station **30** placed atop vacuum table **14**, noted by step **2** (“S**2**”); and the process described, for forming a layer of graphene on the fresh sheet of metal **12**, is repeated.

(16) After the graphene layered sheet of metal **12** has been transferred into the copper dissolving tank **32** and spaced above the (inner) bottom of the tank **32** by means (not shown), the copper dissolving tank **32** is filled initially with an acidic liquid substance, e.g., nitric acid or sulfuric acid to an upper level **35**. After the layer of copper **12**B has been dissolved by the acidic liquid, an upper level **34** of the liquid in the tank **32** may have a turquoise color typical for a copper-ion containing solution.

(17) In FIG. **5**, differences between levels **34**, **35** are illustrative (not to scale). After the copper layer **12**B has been totally dissolved by the acid liquid, a sheet of graphene **36**B (FIG. **5**) is available. Commercial-quality, industrial-grade sheets of graphene **36**B produced by the system and process of the present subject matter are generally able to be rolled up and stored until needed or shipped to customers.

(18) The system **10** includes an operational control system **111** operatively connected to the various components of system **10** described, for enabling efficient and effective control of the system **10**, for producing graphene as described above.

(19) FIG. **6** presents a flowchart **40** (now described in detail) that is illustrative of a graphene vapor deposition process of the present subject matter. At step **402**, a metal sheet, not dissolvable in a solution that dissolves copper, is electroplated with elemental copper. At step **404**, a copper-plated sheet of metal is positioned, e.g., by conveyor or other means, onto a table or other surface upon which at least one vacuum housing is placed. At step **406**, hydraulic cylinders are actuated to cause a vacuum housing to be lowered onto the table or other surface. At step **408**, vacuum within the housing is initiated and a primary heat source is powered on. At step **410**, once predetermined vacuum conditions and a desired temperature are achieved, a secondary heat source activates to vaporize the carbon. At step **412**, the vaporized carbon attaches and condenses on the surface of the copper to produce graphene.

(20) At step **414**, once graphene has been created, a vacuum release valve is opened and the associated vacuum housing moved a predetermined distance from a first position to a second or so-called “overlapping” position, for achieving more efficient graphene coverage on the copper portion of a copper-plated sheet of metal. At step **416**, after the copper-plated sheet of metal has

been moved, predetermined vacuum conditions and the desired temperature are re-established, for producing graphene coverage at overlapping positions. At step **418**, after the copper-plated sheet of metal has efficiently been coated with a layer or sheet of graphene, the vacuum and temperature conditions end; and a copper-plated sheet of metal with a layer or sheet of graphene thereon, is transferred to a copper-dissolving tank. At step **420**, as copper dissolves in the tank, graphene is separated from the sheet of metal. At step **422**, the now free graphene sheet or layer is thereafter recovered and stored.

(21) FIG. **7** illustrates an alternate embodiment of the apparatus. A large robotic arm **152** on guide tracks **150** transfers a plate **12** into an automated electro-plating tank **156** and transfer it to a vacuum press table **214** under a vacuum housing **220**. The large robotic arm **152** transfers the carbonized plate **12** onto a dissolving tank **132**. The plate holding table **130** moves to a position in which the robotic arm **152** can transfer plates **12** to the electroplating tank **156**. The electroplating tank **156** is also mobile to move into line for plating and to move out of the way for a subsequent plating tank to move into position. Small robotic arms place carbon **18**A and/or radioactive blocks **16** (e.g., nuclear batteries) onto a carbon holder **18**; see FIG. **1**.

(22) What has been illustrated and described in this patent application is a graphene vapor deposition system and process. While the present subject matter has been described with reference to exemplary embodiments, the present subject matter is not limited to these examples. On the contrary, many alternatives, changes, and/or modifications will become apparent to a person of ordinary skill in the art ("POSITA") after this application and its associated figures have been reviewed. Therefore, alternatives, changes, or modifications are to be treated as part of the present subject matter insofar as they fall within the spirit and scope of the claims.

Claims

1. A graphene vapor deposition system (**10**) comprising: an electroplating tank (**156**); a supporting surface (**14**) for supporting a copper-plated sheet of metal (**12**) adjacent to the electroplating tank (**156**); at least one housing (**20**) defining an interior region (**120**), wherein the at least one housing (**20**) includes a sealing surface (**22**) that is capable when urged against the supporting surface (**14**) of maintaining predetermined vacuum conditions within the interior region (**120**) of the at least one housing (**20**); a hydraulic cylinder means (**28**) operatively coupled to the at least one housing (**20**) for moving the at least one housing (**20**) between a first position above the supporting surface (**14**) and a second position to urge the sealing surface against the supporting surface and provide a vacuum-tight seal therebetween; a pump (**24**) to evacuate the interior region (**120**) to the predetermined vacuum conditions when the sealing surface is urged against the supporting surface; a predetermined amount of carbon disposed in the interior region; a primary heating means (**16**) powered by a source to heat the interior region (**120**); a secondary heating means (**26**) powered by the source to convert the carbon to vaporized carbon, for enabling graphene vapor deposition on the copper; and a dissolving tank (**132**) adjacent to the housing (**20**).
2. The graphene vapor deposition system (**10**) of claim 1, including a control system (**111**) operatively connected to the pump (**24**) and the primary heating means, for controllably enabling graphene vapor deposition on the copper.
3. The graphene vapor deposition system (**10**) of claim 1, wherein at least one of the dissolving tank (**132**) and the electroplating tank (**156**) are mobile, and further comprising a mobile plate holding table (**130**) operative to move the copper-plated sheet (**12**) from a first plate position to a second plate position.
4. The graphene vapor deposition system (**10**) of claim 3, further comprising a mobile robotic arm (**152**) operative to transfer a metal sheet (**12**) to and from the supporting surface (**214**).
5. The graphene vapor deposition system (**10**) of claim 4, wherein the mobile robotic arm (**152**) is operative to transfer a sheet of metal (**12**A) into the electroplating tank (**156**), to transfer the

copper-plated sheet (**12**) to the supporting surface (**14**), and to transfer the copper-plated sheet (**12**) into the dissolving tank (**132**).

6. The graphene vapor deposition system (**10**) of claim 3, wherein the dissolving tank (**132**), the electroplating tank (**156**), and the mobile plate holding table (**130**) are automated, computer-controlled, and chain driven.

7. The graphene vapor deposition system (**10**) of claim 1, wherein the at least one housing (**20**) comprises at least two housings (**20**).

8. A graphene vapor deposition process comprising: transferring a sheet of metal into an electro-plating tank; electro-plating the sheet to make a copper-plated sheet; positioning the copper-plated sheet on a surface; actuating hydraulic cylinder means to cause a vacuum housing defining an interior region to be lowered onto the surface, wherein the copper-plated sheet is disposed within the interior region; initiating vacuum conditions within the interior region of the vacuum housing; powering on a primary heating source within the interior region; once predetermined vacuum conditions and a predetermined temperature by the primary heating source are achieved, activating a secondary heating source to vaporize carbon; and once the vaporized carbon has coated the copper-plated sheet to form a graphene coated copper-plated sheet, transferring the graphene coated copper-plated sheet into a dissolving tank filled with a copper-dissolving liquid.

9. The graphene vapor deposition process of claim 8, further including: once graphene begins to form on the copper of the copper-plated sheet and before transferring the graphene coated copper-plated sheet into the dissolving tank, opening a vacuum release valve and moving the vacuum housing from a first position to a second position, and repeating the steps of actuating, initiating, powering on, and activating, to achieve more efficient graphene coverage on the copper.

10. The graphene vapor deposition process of claim 8, further including: once the copper has dissolved, removing the graphene from the dissolving tank.

11. The graphene vapor deposition process of claim 10, further including: after removing the graphene from the dissolving tank, storing the removed graphene.

12. The graphene vapor deposition process of claim 8, further comprising: once graphene begins to form on the copper of the copper-plated sheet and before transferring the graphene coated copper-plated sheet into the dissolving tank, opening a vacuum release valve, raising the vacuum housing, and moving the copper-plated sheet from a first plate position to a second plate position, and repeating the steps of actuating, initiating, powering on, and activating, to achieve more efficient graphene coverage on the copper.