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(54) **METHOD AND DEVICE FOR PRODUCING CARBON NANOTUBES**

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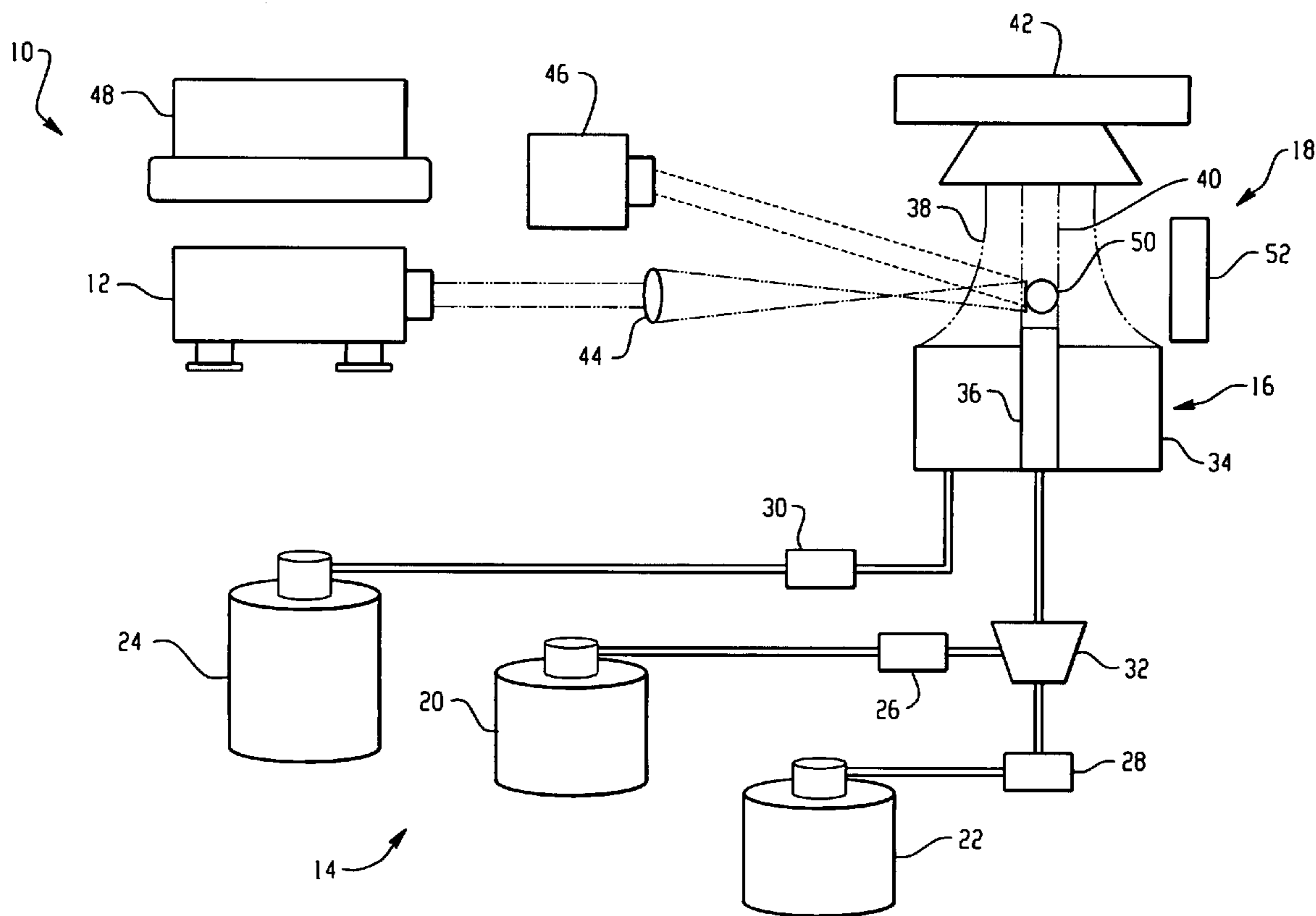
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(57) **ABSTRACT**

Methods and devices for producing carbon nanotubes are disclosed herein. These methods and devices are based on chemical vapor deposition (CVD) in an open environment under atmospheric pressure, which eliminates the need for a vacuum chamber or evacuation process to remove oxygen and/or impurities prior to carbon nanotube growth.



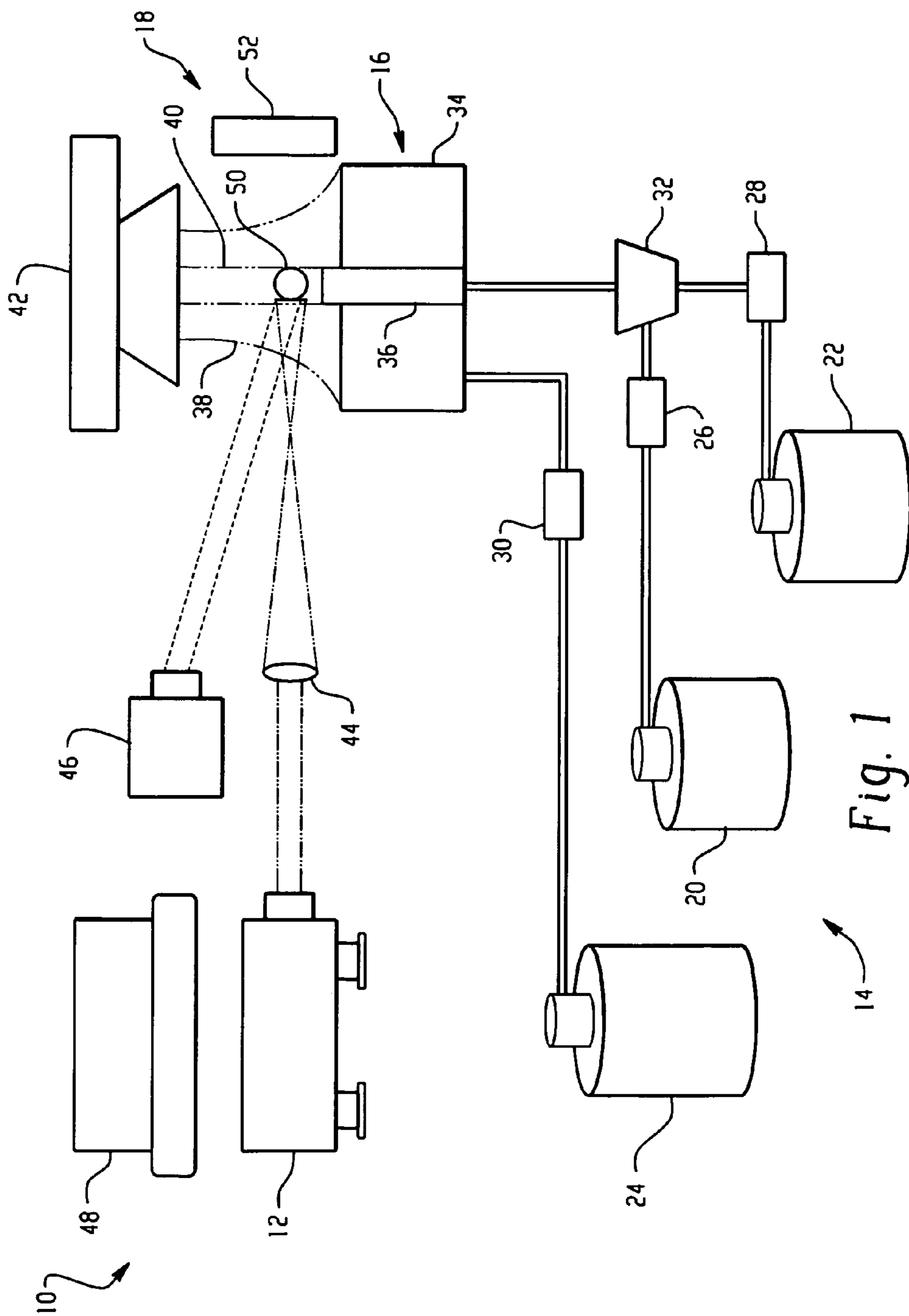


Fig. 1

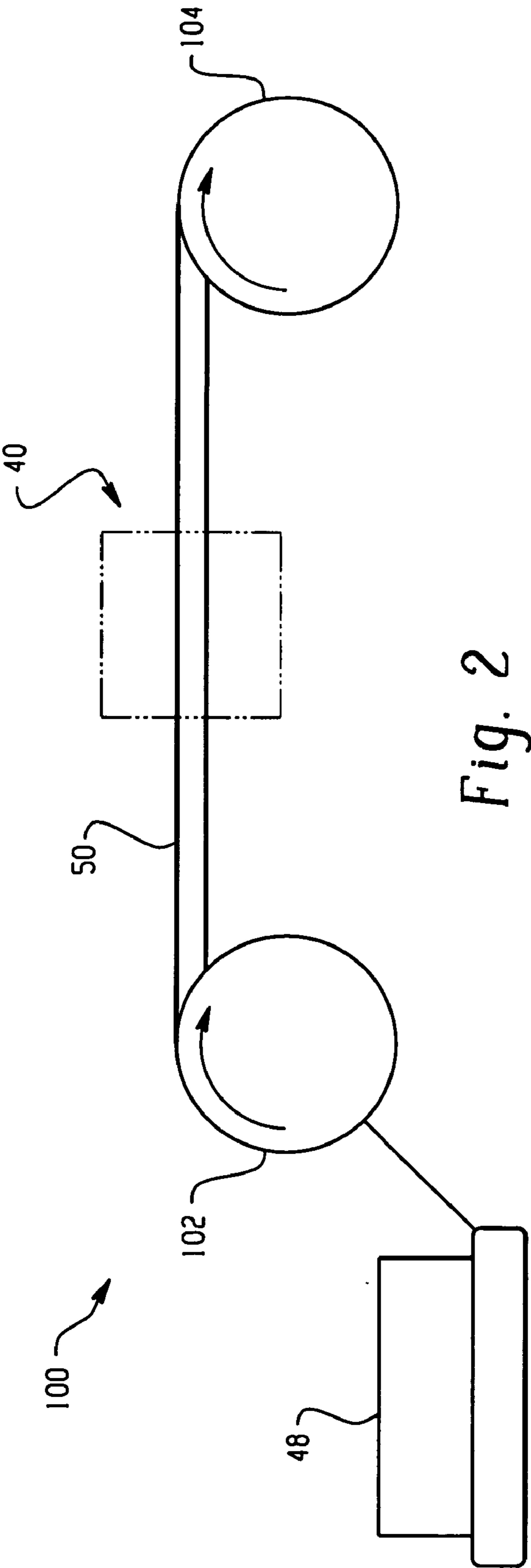


Fig. 2

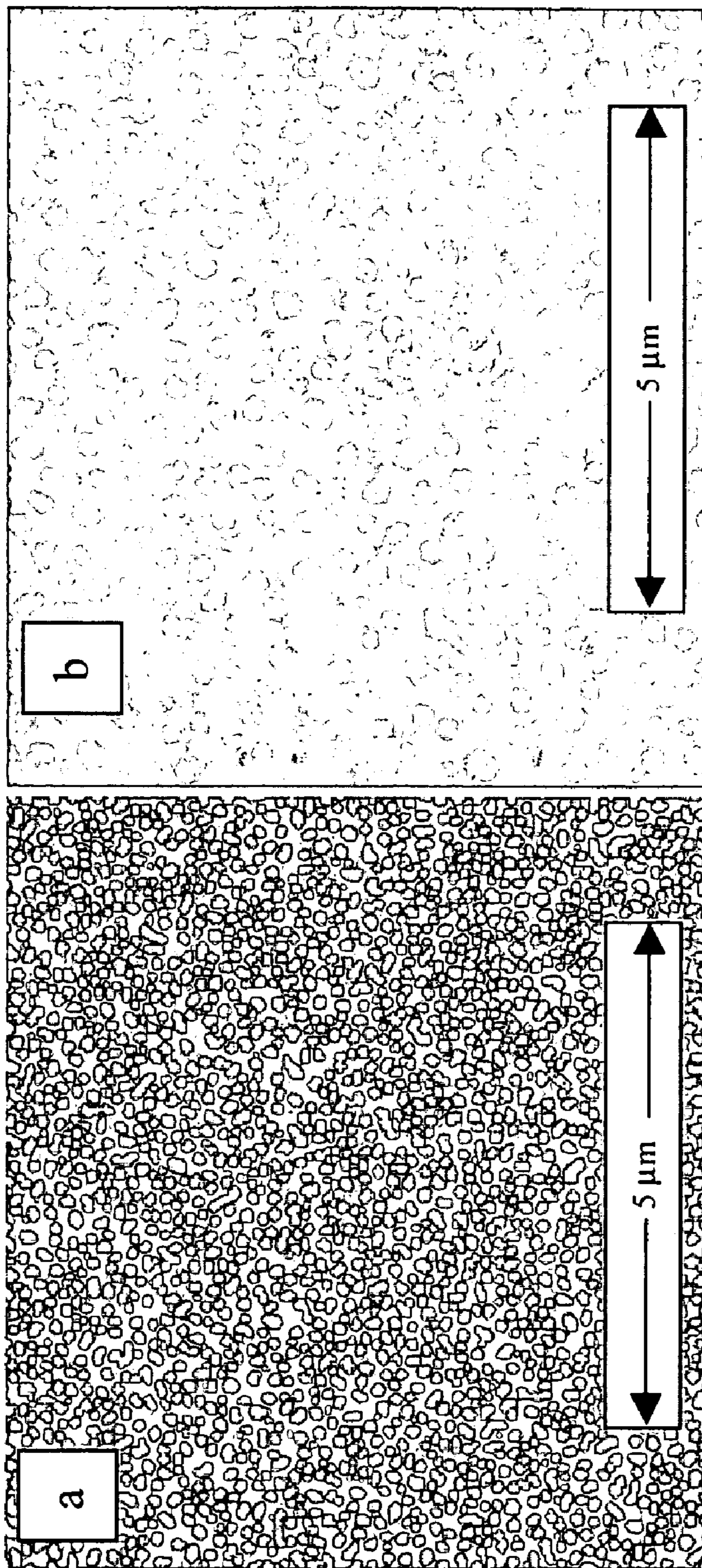


Fig. 3



Fig. 4

## METHOD AND DEVICE FOR PRODUCING CARBON NANOTUBES

### STATEMENT REGARDING FEDERALLY SPONSORED RESEARCH

[0001] The United States Government has certain rights in this invention pursuant to National Science Foundation Grant No. CTS 0093544.

### CROSS REFERENCE TO RELATED APPLICATIONS

[0002] The present application relates to, and claims the benefit of, U.S. Provisional Patent Application No. 60/683,627, which was filed on May 23, 2005, and is incorporated herein in its entirety.

### BACKGROUND

[0003] This disclosure relates generally to carbon nanotubes and, more specifically, to a method and device for producing carbon nanotubes in open environments.

[0004] Carbon nanotubes (also referred to as carbon fibrils) have received much attention owing to their promising electronic and mechanical properties. The nanotubes can be thought of as rolled up sheets of graphite, which may be open or capped at each end. Measurements have shown that their diminutive size belies a host of practical applications, such as molecular wires, transistors, nanoswitches, diodes, chemical sensors, high strength materials, electron field emitters, and tips for atomic force microscopes, among others. Carbon nanotubes are metallic, semimetallic, or semiconducting depending on their diameter and the hexagonal pattern along the tube axis (i.e., their chirality). They are also extremely strong, with a Young's Modulus similar to diamond and significantly greater than most known metals and alloys.

[0005] Presently, there are three main approaches towards the synthesis of carbon nanotubes. These include electric arc discharge of graphite, laser ablation of graphite, and chemical vapor deposition (CVD) of hydrocarbons. The arc discharge and laser ablation techniques involve rather extreme conditions (e.g., temperatures greater than about 3000° C.), with low nanotube yield. CVD methods have emerged as viable alternatives because, in part, they have shown that milder conditions are feasible and that bulk (e.g., kilogram or even ton) quantities of carbon nanotubes can be produced. Other techniques, which are in their early stages of development and will require much effort in order to overcome the technical challenges for mass production of carbon nanotubes, include open-air flame synthesis of carbon nanotubes and a template synthesis technique. The template synthesis technique has successfully been used to grow vertically-aligned carbon nanotube arrays; however, this technique leads to the formation of structural irregularities and defects.

[0006] There are several drawbacks associated with existing CVD processes. For example, the relatively low deposition temperature (e.g., less than about 1200° C.) results in carbon nanotubes with high defect concentrations and lower degrees of graphitization compared to those grown by arc discharge and laser ablation techniques. Additionally, deposition rates for CVD techniques are generally limited to about 10 micrometers per minute, and the cost of nanotubes can range from 60-750 dollars per gram, which is more expensive than gold. Finally, high vacuum growth chambers are needed

in order to eliminate oxidation of as-grown carbon nanotubes, which complicates the synthetic procedure and increases the overall cost of the equipment.

[0007] There accordingly remains a need in the art for new and improved methods and devices for producing carbon nanotubes. It would be particularly desirable if these methods and devices provided the advantages of CVD over arc discharge and laser ablation while simultaneously offering performance advantages (e.g., lower defect concentrations, higher degrees of graphitization, higher deposition rates, less complicated procedures, and/or lower equipment costs) over existing CVD methods.

### BRIEF SUMMARY

[0008] A device for producing carbon nanotubes includes a fluid delivery portion comprising a carbon-containing precursor reservoir, a reducing agent reservoir, and an inert gas reservoir; a coaxial jet reactor in fluid communication with the fluid delivery portion, wherein the coaxial jet reactor comprises an outer nozzle configured to receive an inert gas and an inner nozzle configured to receive a carbon-containing precursor and a reducing agent; a deposition portion in fluid communication with the coaxial jet reactor, wherein the deposition portion comprises an outer zone configured to receive the inert gas from the outer nozzle, an inner zone configured to receive the carbon-containing precursor and reducing agent from the inner nozzle, and a substrate having at least a portion within the inner zone, wherein the outer zone, inner zone and the substrate are in an open environment; and a heating source configured to heat at least the portion of the substrate within the inner zone effective to thermally decompose at least a portion of the carbon-containing precursor in the presence of the reducing agent and produce the carbon nanotubes on the substrate.

[0009] In another embodiment, the device for producing carbon nanotubes includes a fluid delivery portion comprising a carbon-containing precursor reservoir, a reducing agent reservoir, an inert gas reservoir, a carbon-containing precursor mass flow controller, a reducing agent mass flow controller, an inert gas mass flow controller, and a mixer at a junction downstream of the carbon-containing precursor reservoir and the reducing agent reservoir, and a coaxial jet reactor in fluid communication with the fluid delivery portion, wherein the coaxial jet reactor comprises an outer nozzle configured to receive an inert gas and an inner nozzle configured to receive a carbon-containing precursor and a reducing agent; a deposition portion in fluid communication with the coaxial jet reactor, wherein the deposition portion comprises an outer zone configured to receive the inert gas from the outer nozzle, an inner zone configured to receive the carbon-containing precursor and reducing agent from the inner nozzle, a substrate having at least a portion within the inner zone, a nanotube growing catalyst disposed on a surface of the substrate, an exhaust, and a scattered light absorber, wherein the outer zone, inner zone, substrate, nanotube growing catalyst exhaust, and scattered light absorber are in an open environment; a laser, wherein a beam produced by the laser is configured to heat the at least the portion of the substrate within the inner zone effective to thermally decompose at least a portion of the carbon-containing precursor in the presence of the reducing agent and produce the carbon nanotubes on the substrate; and a controller in operative communication with the laser, carbon-containing precursor mass flow controller,

reducing agent mass flow controller, inert gas mass flow controller, mixer, substrate or a combination comprising at least one of the foregoing.

**[0010]** A method for growing carbon nanotubes includes heating at least a portion of a substrate in an open environment; forming a first jet of reactants comprising a carbon-containing precursor and a reducing agent, wherein the first jet is surrounded by a second jet of inert gas that is effective to protect the reactants from the open environment; thermally decomposing at least a portion of the carbon-containing precursor in the presence of the reducing agent in proximity to the heated substrate; and growing the carbon nanotubes on the at least the portion of the substrate.

**[0011]** The above described and other features are exemplified by the following figures and detailed description.

#### BRIEF DESCRIPTION OF THE DRAWINGS

**[0012]** Referring now to the Figures, which are exemplary embodiments, and wherein the like elements are numbered alike:

**[0013]** FIG. 1 is a schematic representation of an open environment chemical vapor deposition device;

**[0014]** FIG. 2 is a schematic representation of a moving substrate device;

**[0015]** FIG. 3 illustrates environmental scanning electron microscope (ESEM) images of (a) 80 nanometer and (b) 300 nanometer gold-palladium catalyst particles disposed onto a 3 millimeter fused quartz rod substrate; and

**[0016]** FIG. 4 illustrates ESEM images of carbon nanotubes of (a) 50 nanometer and (b) 220 nanometer diameters deposited onto a 3 millimeter fused quartz rod substrate.

#### DETAILED DESCRIPTION

**[0017]** Chemical vapor deposition (CVD) methods and devices for making carbon nanotubes are described herein. In contrast to the prior art, the methods and devices are based on chemical vapor deposition under open environment conditions, which provide several advantages over existing carbon nanotube production methods and devices. For example, deposition of the carbon nanotubes occurs under atmospheric pressure, which eliminates the need for a costly vacuum chamber and therefore a time-consuming evacuation process to remove oxygen and/or impurities prior to deposition. In addition to the elimination of this process step, open environment CVD allows for deposition rates that may be up to three orders of magnitude greater than other CVD techniques. Other advantages of open environment CVD based methods and devices will be apparent to those skilled in the art in view of this disclosure. It is to be understood that “open environment”, as used herein, may include the use of an exhaust, vent, or the like.

**[0018]** As used herein, the term “carbon nanotube” is inclusive of a variety of carbon fibers having average diameters of less than or equal to about 2000 nanometers (nm) and having graphitic or partially graphitic structures. These include semi-conducting, semimetallic, and metallic nanotubes as well as single-walled nanotubes (SWNT) and multi-walled nanotubes (MWNT). They may have lengths of a few nanometers to many centimeters. Furthermore, each carbon nanotube may be derivatized or functionalized, for example including an oxygen-containing group such as a carbonyl, carboxylic acid, carboxylic acid ester, epoxy, vinyl ester, hydroxy,

alkoxy, isocyanate; amide group; or derivatives thereof, for example, sulfhydryl, amino, or imino groups; or the like.

**[0019]** Also as used herein, the terms “first,” “second,” “bottom,” “top,” and the like do not denote any order or importance, but rather are used to distinguish one element from another; and the terms “the”, “a” and “an” do not denote a limitation of quantity, but rather denote the presence of at least one of the referenced item. Furthermore, all ranges reciting the same quantity or physical property are inclusive of the recited endpoints and independently combinable. The modifier “about” used in connection with a quantity is inclusive of the stated value and has the meaning dictated by the context or includes at least the degree of error associated with measurement of the particular quantity.

**[0020]** For illustrative purposes, an exemplary open environment CVD device **10** is shown in FIG. 1. The CVD device **10** generally comprises a heating source **12**, a fluid delivery portion **14**, a coaxial jet reactor **16**, and a deposition portion **18**. The fluid delivery portion **14** is in fluid communication with the coaxial jet reactor **16**, which itself is in fluid communication with the deposition portion **18**.

**[0021]** The fluid delivery portion **14** includes a carbon-containing precursor reservoir **20**, a reducing agent-containing reservoir **22**, and an inert gas reservoir **24**, all of which are in fluid communication with the coaxial jet reactor **16**. The fluid delivery portion **14** may optionally include flow meters or mass flow controllers **26**, **28**, **30** to regulate the amount of fluid dispensed by each of the various reservoirs **20**, **22**, **24**, respectively, to the coaxial jet reactor **16**. The fluid delivery portion **14** may further optionally include a mixer **32** disposed on one side at a downstream junction of the carbon-containing precursor reservoir **20** (or optional flow meter **26**) and the reducing agent-containing reservoir **22** (or optional flow meter **28**) and the coaxial jet reactor **16** on the other side. The optional mixer **32** may be used to mix the reducing agent and the carbon-containing precursor before flowing them downstream towards the coaxial jet reactor **16**.

**[0022]** The coaxial jet reactor **16** comprises an inner nozzle **36** and an outer nozzle **34**. In embodiments wherein more than two nozzles are used (not shown), each nozzle is concentric about inner nozzle **36**. Regardless of the number of nozzles, the outermost nozzle is configured to receive the inert gas and the inner nozzle(s) is configured to receive the reactants (e.g., reducing agent and the carbon-containing precursor) from the fluid delivery portion **14**.

**[0023]** The deposition portion **18** comprises an innermost reaction zone **40** and an outermost zone **38**. The number of zones in the deposition portion **18** corresponds to the number of nozzles used in the coaxial jet reactor **16**. Accordingly, each zone is therefore concentric about the circular innermost zone **40**. Regardless of the number of nozzles/zones, the outermost zone **38** comprises an inert gas jet from the outermost nozzle of the coaxial jet reactor **16**. The thickness of outermost zone **38** (i.e., the outer diameter minus the inner diameter) must be of sufficient size to prevent the introduction of oxygen and/or impurities into any inner reaction zone(s). This outermost zone **38** thus functions as a shield or “curtain”, which peripherally encloses the innermost zone to enable the reactions to occur in an open environment and also eliminates the need for a vacuum chamber. The ratio of the thickness of the outermost zone **38** to all inner zones is greater than or equal to about 0.1:1.

**[0024]** The deposition portion **18** further comprises a substrate **50**, a portion onto which the carbon nanotubes are

grown. The nanotube growth portion of the substrate **50** is located inside the inner reaction zone(s) **40**. The substrate **50** material must be chosen such that it is stable (e.g., does not melt, decompose, volatilize, react, or the like) with respect to the thermal energy absorbed from the heating source **12**, the reactants, and the optional catalyst. Suitable substrate **50** materials include quartz, fused quartz, silica, fused silica, sapphire, diamond, or the like. The substrate **50** may have any shape that will allow growth of nanotubes including irregular shapes such as flakes as well as regular shapes such as spheres, rods, sheets, and films.

[0025] The deposition portion **18** may optionally include an exhaust **42**, downstream of the substrate, to collect of the inert gas, any of the various fluids not consumed by the carbon nanotube production, as well as any material created during the process that does not remain on the substrate **50**.

[0026] The heating source **12** can be any device capable of heating the substrate **50** such that the desired deposition temperature is attained once the substrate **50** is in the deposition portion **18**. In one embodiment, the substrate **50** is heated directly inside of the deposition portion **18**. In another embodiment, the substrate **50** is heated outside of the deposition portion **18** to a temperature slightly above the desired temperature, such that any cooling that occurs during its placement into the deposition portion **18** does not reduce the temperature below the desired temperature. Alternatively, the substrate **50** can be "pre-heated" outside of the deposition portion **18** and subsequently heated to the desired temperature once inside the deposition portion **18**. The heating source **12**, regardless of where the substrate **50** is heated, can be a resistive heater, a microwave generator, a flash lamp, an infrared light, a laser, a thermoelectric device, a plasma source, and the like.

[0027] In an exemplary embodiment, as depicted in FIG. 1, the heating source is a laser. An advantageous feature of laser induced chemical vapor deposition (LCVD) is that the laser permits greater control and spatial resolution of the deposition temperature than other CVD processes. In addition, the deposition temperatures involved in LCVD may be higher than other CVD processes, which allows for lower defect concentrations and increased graphitization of the carbon nanotubes.

[0028] A laser beam generated by the laser **12** is used to locally heat at least a portion of the substrate **50** within the deposition portion **18**. The laser **12** may be any type of laser such as a solid state laser, a gas laser, an excimer laser, a dye laser, a semiconductor laser, or the like. The laser beam may be pulsed or may be continuous wave. The choice of laser **12**, and the specific lasing material employed in the laser **12**, is based on the absorption characteristics of the substrate **50** and the thermal stability of the carbon-containing precursor. Specifically, the laser **12** desirably has an emission wavelength at which the substrate **50** absorbs and a power level sufficient to provide enough thermal energy to heat at least the portion of the substrate **50** to a temperature greater than or equal to the reaction temperature of the carbon-containing precursor. The appropriate laser may be readily selected by those skilled in the art in view of this disclosure without undue experimentation.

[0029] In embodiments where a laser is used as the heating source **12**, several tools may be used to manipulate the laser beam. For example, in order to focus or tune the laser beam to a specific diameter, the CVD device **10** may further include an optical lens **44**. The composition of the material used to make

the lens, the shape of the lens, and the distance of the lens from the laser **12** can independently be adjusted to provide a desired laser beam diameter by those skilled in the art in view of this disclosure without undue experimentation. For example, the composition of the material used to make the lens would be selected such that the lens is transparent to the laser beam. The lens shape and distance from the laser can be determined using optics equations. Furthermore, a second, aligning laser (not shown) and/or a laser diode (not shown) may be used to align the laser beam with the substrate **50**. Other components that may be used in conjunction with the laser include a beam splitter (not shown) and/or mirror (not shown) to divide and/or re-focus, respectively, the beam.

[0030] Additionally, an optional scattered light absorber **52** may be disposed in the deposition portion **18** to absorb any portion of the laser beam (or any light beam used to heat the substrate **50**) that is not absorbed by the substrate **50**.

[0031] The deposition temperature (i.e., temperature of the substrate) may be determined using an optional temperature-sensing device **46** that is in thermal communication with the substrate **50**. Suitable temperature sensing devices include thermocouples, pyrometers, and the like.

[0032] The open environment CVD device **10** may further comprise a controller **48**, such as a computer, in operative communication with the heating source **12**. In addition, the controller may be used to monitor and/or control the optional flow meters **26**, **28**, **30**, the optional mixer **32**, the size and flow rate of the jets from the coaxial jet reactor **16**, or a combination comprising at least one of the foregoing. In embodiments where a laser is used as the heating source **12**, the controller may be used to monitor and/or control the optional optical lens **44**, the optional second laser, the optional laser diode, the optional beam splitter, the optional mirror, the optional temperature sensing device **46**, or a combination comprising at least one of the foregoing.

[0033] Producing the carbon nanotubes using the open environment CVD device **10** generally comprises heating the substrate or target **50** and at least partially thermally decomposing the reactants (e.g., the reducing agent and carbon-containing precursor) in an open environment to deposit carbon nanotubes at an interface between the reactants and a surface of the substrate **50**. Without being bound by theory, it is believed that once the temperature of the substrate **50** exceeds the reaction temperature of the carbon-containing precursor, the precursor decomposes, in the presence of the reducing agent and in proximity to the substrate **50**, into carbon radicals, which then precipitate to form the carbon nanotubes.

[0034] Carbon nanotube production may be facilitated by having the surface of the substrate **50** coated with a nanotube growing catalyst (not shown) prior to the heating of the substrate **50** using the heating source **12**. Alternatively, the nanotube growing catalyst can be generated during deposition from a catalyst precursor, such as ferrocene. The presence of the nanotube growing catalyst increases the rate of nanotube production. The nanotube growing catalyst may be coated onto the substrate **50** using any known deposition method and may comprise iron, nickel, aluminum, yttrium, cobalt, platinum, gold, silver, palladium, or the like; combinations comprising at least one of the foregoing; and material combinations or alloys comprising at least one of the foregoing.

[0035] The carbon-containing precursor may be any composition that can at least partially thermally decompose into carbon. Furthermore, the carbon-containing precursor may



be a subliming solid, a liquid, or a gas. Generally, hydrocarbons of less than about 16 carbon atoms will be suitable, although hydrocarbons with higher numbers of carbon atoms may also be suitable, for example, depending on their chemical structure, temperature, and pressure. The hydrocarbons may be of any type, including, for example, alkanes, alkenes, alkynes, alcohols, ethers, esters, carboxylic acids, aldehydes, ketones, carbonates, thiols, amines, and the like. The hydrocarbons may be straight chained, branched, or cyclic.

**[0036]** In one embodiment, the carbon-containing precursor is a mixture of hydrocarbons. The carbon-containing precursor may be a mixture of hydrocarbons that are all of the same type or it may contain a mixture of different hydrocarbons. Further, the mixture may be a mixture of hydrocarbons all having the same number of carbon atoms such as octane, octene and 1,3-dimethyl-cyclohexane or a mixture of hydrocarbons having different numbers of carbon atoms such as methanol and butane.

**[0037]** The reducing agent may be hydrogen, ammonia, or a combination comprising at least one of the foregoing, such as forming gas (non explosive hydrogen and nitrogen mixture).

**[0038]** The inert gas may be nitrogen, helium, neon, argon, krypton, xenon, radon, or a combination comprising at least one of the foregoing.

**[0039]** The volumetric ratio of the carbon-containing precursor to reducing agent may be about 0.01:1 to about 15:1. Specifically, the volumetric ratio of the carbon-containing precursor to reducing agent may be about 01:1 to about 1:1. Furthermore, the volumetric ratio of the reactants to the inert gas may be about 0.001:1 to about 10:1. Specifically, the volumetric ratio of the reactants to the inert gas may be about 0.01:1 to about 0.75:1.

**[0040]** In one embodiment, the substrate 50 may be a moving substrate. Owing to the lack of an enclosed chamber, the substrate may be a continuously moving substrate, such as on a conveyor. An exemplary moving substrate device 100 is shown in FIG. 2. The substrate 50 begins as a rolled sheet on a single feed roll 102 and is fed through the inner reaction zone 40 of the deposition portion 18. After the carbon nanotubes are deposited on the portion of the substrate 50 in the inner reaction zone 40, the substrate 50 exits the inner reaction zone 40 and is ultimately rolled onto a take-up roll 104. After exiting the reaction zone 40, the substrate 50 enters the outer zone 38, where the inert gas jet further serves to cool down the heated portion of the substrate 50. In this manner, ultra-long carbon nanotubes may be grown on the substrate 50 if they are grown parallel to the direction of motion of the substrate 50. Alternatively, a larger amount of carbon nanotubes may be produced if they are grown perpendicular to the direction of motion of the substrate 50.

**[0041]** In another embodiment, the controller 48 controls the position of the applied heat and/or the position of the substrate 50 using the moving substrate device 100. In this manner, the carbon nanotubes may be selectively deposited in specific locations and/or configurations, such as in a patterned array.

**[0042]** The disclosure is further illustrated by the following non-limiting examples.

#### EXAMPLE 1

##### Carbon Nanotube Production via Laser-Induced Chemical Vapor Deposition (LCVD)

**[0043]** A 30 Watt (W) continuous wave carbon dioxide (CO<sub>2</sub>) laser (Synrad J48-2W) operating at a wavelength of

10.6 micrometers (μm) was used to heat the substrate. The laser beam profile was “TEM<sub>00</sub>”, which represents a Gaussian distribution. Plano-convex zinc selenide (ZnSe) lenses were used to focus the laser beam to the desired diameter on the substrate. Proper alignment of the laser beam and the substrate was also performed with the aid of a low power He—Ne laser beam and a laser diode. A verification and/or realignment of the laser optics was carried out for each deposition to maintain the consistency of the experimental conditions. The LCVD device was equipped with a two-color pyrometer for measuring the deposition temperature. LabVIEW (National Instruments Corporation) computer software was used for data acquisition and laser power control.

**[0044]** With regards to the coaxial jet reactor, the diameters of the inner and outer nozzles are 4.2 and 52.3 mm, respectively. The inert gas was nitrogen, the carbon-containing precursor was propane (C<sub>3</sub>H<sub>8</sub>) from Airgas with 99.95% purity, and the reducing agent was ultra-high purity grade hydrogen. Precision flow meters were used to regulate the flow rate of the various gases to the reaction region. The hydrogen and propane were mixed with a mixer. The volumetric flow rates of propane, hydrogen and nitrogen were 0.2, 1.1 and 10 standard liters per minute (SLPM), respectively.

**[0045]** 3 millimeter (mm) diameter rods of fused quartz (GE 214 from Quartz Plus, Inc.) were used as the substrates. The substrates were cleaned with methanol and distilled water and dried with dry air, then sputter-coated with a gold-palladium (Au—Pd) thin film (about 10 nm to 100 nm thick depending on sputtering time exposure). In order to convert the deposited Au—Pd thin film into nanoparticles, each coated substrate was annealed at 1000° C. for 15 minutes in a nitrogen environment at 15 Torr. FIG. 3 illustrates environmental scanning electron microscope (ESEM) (Philips 2020 ESEM) images of the Au—Pd nanoparticles for two different film thicknesses under identical annealing conditions. The average particle sizes obtained from films of about 20 nm and about 50 nm thickness were about 80 nm (FIG. 3a) and about 300 nm (FIG. 3b), respectively. The results indicated that thinner films would give rise to smaller particles sizes. The laser irradiation time for LCVD was generally about ten minutes and was followed by a five-minute cooling period. The laser beam diameter was set at 1.8 mm and the laser power ranged from 10-15 W. During each deposition, the temperature history of the deposition process was measured by the pyrometer.

**[0046]** The microstructure and chemical composition of the carbon nanotubes were analyzed using ESEM and Raman spectroscopy. These characterization techniques were used because they were non-destructive. A Renishaw Ramascope was used to acquire Raman spectra directly from the carbon nanotubes using a 514.5 nm argon ion laser (2.41 eV) at 25 mW as the excitation source. The argon ion laser beam was focused by a 50× objective lens to illuminate a 1 μm diameter spot on the substrate. The obtained spectrum contained information representing the average properties of the material inside the area of interest. Curve fitting was applied to the spectra in order to identify relevant Raman peaks. No smoothing was applied to the raw Raman data. In addition, transmission electron microscopy (JEOL 2010 FasTEM) was used to obtain high-resolution images of the carbon nanotubes.

**[0047]** FIG. 4 shows two ESEM images of the carbon nanotubes deposited on a fused quartz substrate at a laser power of 10 Watts for a period of 10 minutes. These images revealed that the deposited nanotubes were in tangled form with no

particular growth orientation, and catalyst particles were present along the length of the nanotubes in some regions of the sample. In addition, nanotubes with small diameter (10-40 nm) tended to grow from the metal catalyst with no metal particles attached at an end. These observations suggest that both tip-growth and base-growth modes were present. In tip-growth mode, the catalyst particle is lifted off from the substrate and carried along at the end of the nanotube as the nanotube grows in length. In base-growth mode, the catalyst particles remain adhered to the substrate surface as the nanotubes lengthen. The length of the carbon nanotubes obtained in this study ranged from a few microns to several hundred microns. Longer carbon nanotubes may have been deposited, but the tangling of nanotubes made it difficult to perform accurate length measurements.

**[0048]** It appeared that the size of the metal catalyst particles determined the diameter of the deposited carbon nanotubes. The average Au—Pd particle size used in the synthesis of nanotubes shown in FIGS. 3a and 3b are 80 and 300 nm, respectively. By comparing the diameter of the nanotubes, larger metal particles resulted in larger diameter carbon nanotubes. The average diameter of carbon nanotubes shown in FIGS. 4a and 4b are approximately 50 nm and 220 nm, respectively, which indicates that these nanotubes were MWNTs. The Raman spectrum of carbon nanotubes obtained in this study reveal several distinctive peaks located between 100 and 3500  $\text{cm}^{-1}$  representing the characteristic Raman peaks of multi-wall carbon nanotubes.

**[0049]** While the disclosure has been described with reference to exemplary embodiments, it will be understood by those skilled in the art that various changes may be made and equivalents may be substituted for elements thereof without departing from the scope of the invention. In addition, many modifications may be made to adapt a particular situation or material to the teachings of the invention without departing from the essential scope thereof. Therefore, it is intended that the invention not be limited to the particular embodiment disclosed as the best mode contemplated for carrying out this invention, but that the invention will include all embodiments falling within the scope of the appended claims.

What is claimed is:

1. A device for producing carbon nanotubes, the device comprising:

- a fluid delivery portion comprising a carbon-containing precursor reservoir, a reducing agent reservoir, and an inert gas reservoir;
- a coaxial jet reactor in fluid communication with the fluid delivery portion, wherein the coaxial jet reactor comprises an outer nozzle configured to receive an inert gas and an inner nozzle configured to receive a carbon-containing precursor and a reducing agent;
- a deposition portion in fluid communication with the coaxial jet reactor, wherein the deposition portion comprises an outer zone configured to receive the inert gas from the outer nozzle, an inner zone configured to receive the carbon-containing precursor and reducing agent from the inner nozzle, and a substrate having at least a portion within the inner zone, wherein the outer zone, inner zone and the substrate are in an open environment; and
- a heating source configured to heat the at least the portion of the substrate within the inner zone effective to thermally decompose at least a portion of the carbon-con-

taining precursor in the presence of the reducing agent and produce the carbon nanotubes on the substrate.

2. The device of claim 1, wherein the fluid delivery portion further comprises a mixer, a mass flow controller, or both, wherein the mixer is at a junction upstream of the inner nozzle of the coaxial jet reactor and downstream of the carbon-containing precursor reservoir and the reducing agent reservoir, and wherein the mass flow controller configured to regulate an amount of fluid dispensed by the carbon-containing precursor reservoir, reducing agent reservoir, or inert gas reservoir.

3. The device of claim 1, wherein a ratio of a thickness of the outer zone of the coaxial jet reactor to the inner zone of the coaxial jet reactor is greater than or equal to about 0.1:1.

4. The device of claim 1, wherein the deposition portion further comprises an exhaust, a scattered light absorber, or both.

5. The device of claim 1, wherein the heating source is a laser.

6. The device of claim 1, further comprising an optical lens, an aligning laser, a laser diode, a beam splitter, a mirror, a temperature-sensing device, or a combination comprising at least one of the foregoing.

7. The device of claim 1, further comprising a controller in operative communication with the heating source, mass flow controller, mixer, optical lens, aligning laser, laser diode, beam splitter, mirror, temperature sensing device, substrate, or a combination comprising at least one of the foregoing.

8. The device of claim 1, wherein the substrate is a moving substrate.

9. The device of claim 1, wherein a surface of the substrate comprises a nanotube growing catalyst.

10. The device of claim 1, wherein a volumetric ratio of the carbon-containing precursor to the reducing agent is about 0.01:1 to about 15:1.

11. The device of claim 1, wherein a volumetric ratio of the carbon-containing precursor and the reducing agent to the inert gas is about 0.001:1 to about 10:1.

12. A device for producing carbon nanotubes, the device comprising:

- a fluid delivery portion comprising a carbon-containing precursor reservoir, a reducing agent reservoir, an inert gas reservoir, a carbon-containing precursor mass flow controller, a reducing agent mass flow controller, an inert gas mass flow controller, and a mixer at a junction downstream of the carbon-containing precursor reservoir and the reducing agent reservoir, and
- a coaxial jet reactor in fluid communication with the fluid delivery portion, wherein the coaxial jet reactor comprises an outer nozzle configured to receive an inert gas and an inner nozzle configured to receive a carbon-containing precursor and a reducing agent;
- a deposition portion in fluid communication with the coaxial jet reactor, wherein the deposition portion comprises an outer zone configured to receive the inert gas from the outer nozzle, an inner zone configured to receive the carbon-containing precursor and reducing agent from the inner nozzle, a substrate having at least a portion within the inner zone, a nanotube growing catalyst disposed on a surface of the substrate, an exhaust, and a scattered light absorber, wherein the outer zone, inner zone, substrate, nanotube growing catalyst exhaust, and scattered light absorber are in an open environment;

- a laser, wherein a beam produced by the laser is configured to heat the at least the portion of the substrate within the inner zone effective to thermally decompose at least a portion of the carbon-containing precursor in the presence of the reducing agent and produce the carbon nanotubes on the substrate; and
- a controller in operative communication with the laser, carbon-containing precursor mass flow controller, reducing agent mass flow controller, inert gas mass flow controller, mixer, substrate or a combination comprising at least one of the foregoing.
- 13.** The device of claim **12**, wherein a ratio of a thickness of the outer zone of the coaxial jet reactor to the inner zone of the coaxial jet reactor is greater than or equal to about 0.1:1.
- 14.** The device of claim **12**, further comprising an optical lens, an aligning laser, a laser diode, a beam splitter, a mirror, a temperature-sensing device, or a combination comprising at least one of the foregoing.
- 15.** The device of claim **14**, wherein the controller is further in operative communication with the optical lens, aligning laser, laser diode, beam splitter, mirror, temperature sensing device, or a combination comprising at least one of the foregoing.

- 16.** The device of claim **12**, wherein the substrate is a moving substrate.
- 17.** A method for growing carbon nanotubes, comprising: heating at least a portion of a substrate in an open environment; forming a first jet of reactants comprising a carbon-containing precursor and a reducing agent, wherein the first jet is surrounded by a second jet of inert gas that is effective to protect the reactants from the open environment; thermally decomposing at least a portion of the carbon-containing precursor in the presence of the reducing agent in proximity to the heated substrate; and growing the carbon nanotubes on the at least the portion of the substrate.
- 18.** The method of claim **17**, further comprising mixing the carbon-containing precursor with the reducing agent prior to the forming the jet of reactants in an open environment.
- 19.** The method of claim **17**, further comprising disposing a nanotube growing catalyst onto the at least a portion of a substrate.
- 20.** The method of claim **17**, further comprising moving the substrate contemporaneously with the growing.

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