SYNTHESIS OF COILED CARBON NANOTUBES BY MICROWAVE CHEMICAL VAPOR DEPOSITION

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ABSTRACT
The present invention provides a coiled carbon nanotube and a method for its manufacture. The coiled carbon nanotube comprises a specific non-hexagonal/hexagonal carbon ring ratio, a specific pitch, and a specific diameter. The invention employs a microwave chemical vapor deposition system with novel processing conditions and specialized catalysts to synthesize the coiled carbon nanotubes.
Fig. 4
SYNTHESIS OF COILED CARBON NANOTUBES
BY MICROWAVE CHEMICAL VAPOR DEPOSITION

REFERENCE TO RELATED APPLICATIONS

This application claims the benefit of U.S. Provisional Patent Application No. 60/431,888 filed Dec. 6, 2002.

FIELD OF THE INVENTION

The present invention relates to the field of carbon nanotubes. More particularly, the invention relates to mass-produced coiled carbon nanotubes and to a method for their synthesis using microwave chemical vapor deposition (CCVD).

BACKGROUND OF THE INVENTION

Prior art methods of catalytic chemical vapor deposition (CCVD) have been used to prepare carbon fibers/tubes with different morphologies in different sizes. Among them, fibrous carbon materials with coil morphology have especially attracted a wide interest. This interest relates not only to academic research interests but also to potential versatile commercial applications. Motojima et al. first reported regular coiled carbon fibers in micron size by CCVD with thiophene vapor as an impurity gas, named carbon microcoils [Motojima S. et al., “Preparation of coiled carbon fibers by pyrolysis of acetylene using a Ni catalyst and sulfur or phosphorus compound impurity” Appl. Phys. Lett. 62 2322-3, 1993]. In this paper, the growth mechanism of carbon microcoils was postulated as resulting from the anisotropic properties of the catalyst for carbon deposition. Such carbon microcoils found applications in EM absorbers, micro springs, etc. Subsequently, Pan et al. reported the synthesis of carbon tubule nanocoils using iron-coated indium tin oxide as a catalyst [Pan L. et al., “Growth and density control of carbon tubule nanocoils using catalyst of iron compounds” J. Mater. Res. 17 145-8, 2001]. Recently, Varadan et al. synthesized carbon nanocoil fibers by using Ni particles [V. K. Varadan, J. Xie, “Synthesis of carbon nanocoils by microwave CVD” Smart Materials and Structures, 11 728-34, 2002]. Such nanocoiled carbon fibers/tubes have been shown to be good candidates for commercial applications, especially field emission display technology [Pan L. et al., “Field emission properties of carbon tubule nanocoils” Jpn. J. Appl. Phys. 40 1 235-7, 2001]. Note that all of the coiled carbon fibers/tubes mentioned above are prepared using amorphous carbon materials.

In contrast to the conventional method of producing carbon nanocoils, the present invention relates to coiled carbon nanotubes. Since the discovery of carbon nanotubes by Iijima, coiled carbon nanotubes have become objects of widespread interest. The primary difference between coiled carbon nanotubes and carbon nanocoils lies in the crystalline graphic structures of the nanotubes. Also, the diameter of coiled carbon nanotubes (<100 nm) is much smaller than the diameter of carbon nanocoils. The coil morphology, together with as well as the extraordinary properties of nanotubes, make coiled carbon nanotubes a promising material for hydrogen storage, field emission, EM absorber and nanotechnology applications in general.

For example, accidentally coiled carbon nanotubes were reported in extremely low yield by Hernadi et al. [“Fe-catalyzed carbon nanotube formation” Carbon 34 1249-57, 1996]. Also, Amelinckx et al. reported a formation mechanism for catalytically grown helix-shaped graphite nanotubes [“A formation mechanism for catalytically grown helix-shaped graphite nanotubes” Science 265 635-9, 1994]. According to their results, the coil morphology of carbon nanotubes is due to the mismatch between the extrusion velocity and the rate of carbon deposition.

Thus far the synthesis of coiled CNTs has been reported as a byproduct of regular CNT synthesis. It would be more accurate to say that coiled CNTs have been found under microscope by accident because there was no control for the synthesis of coiled CNT. To date, neither specific process conditions nor special catalyst compositions have been identified for the effective and consistent synthesis of coiled CNTs. Despite the early indications of the usefulness of coiled CNTs, there continues to be a strong need for the effective synthesis of regular carbon nanotubes.

CNTs, along with coiled carbon nanotubes, are the most promising materials anticipated to impact future nano-science and nanotechnology. Their unique structural and electronic properties have generated great interest for use in a broad range of nanodevices. A significant amount of work has been done in the past decade to reveal the unique structural, electrical, mechanical, electromechanical and chemical properties of carbon nanotubes and to explore the key applications of this novel material. Most of these applications will require efficient fabrication methods capable of producing pure CNTs, including coiled carbon nanotubes, to meet device requirements. Another advantage of coiled carbon nanotubes is that they are capable of forming in situ semiconductor-metallic or semiconductor-insulator junctions which one can utilize for the fabrication of nanodevices. Coiled carbon nanotubes also have a greater surface area than CNTs which increases their functionalization. Accordingly, it is an object of the present invention to provide effective and efficient methods of fabricating CNTs, especially coiled carbon nanotubes, to enable the commercialization of applications using such.

SUMMARY OF THE INVENTION

The invention is a method for synthesizing coiled carbon nanotubes using a microwave CVD system with inventive processing conditions and specialized catalyst(s). Preferred conditions include the use of acetylene as a hydrocarbon source gas in a microwave CVD system without using an impurity gas and using an iron supported on magnesium carbonate as a catalyst. The invention also includes the resulting coiled carbon nanotubes.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 shows a schematic diagram of a conventional thermal filament CVD system.

FIG. 2 shows a schematic diagram of a microwave CVD system used in the present invention.

FIG. 3 shows an illustration of a coiled carbon nanotube.

FIG. 4 shows a flow diagram for the flow control systems.
FIG. 5 shows a SEM micrograph of coiled nanotube synthesized using a microwave CVD system in accordance with the present invention.

FIG. 6 shows a TEM image of coiled nanotubes obtained from a microwave CVD system in accordance with the present invention.

DETAILED DESCRIPTION OF THE INVENTION

Referring now to FIG. 1, an apparatus designed to perform this method is designated in its entirety by the reference numeral 8. The method generally involves placing a catalyst 3 inside of a reaction chamber 1 and then heating up the reaction chamber 1. In one embodiment of the method, a quartz reaction tube 2 is used to transport the catalyst in and out of the reaction chamber 1. The temperature is monitored by a thermocouple 7. When the temperature inside has reached a certain level (usually 700°C) a hydrocarbon source gas 4 (such as acetylene) is pumped into the reaction chamber 1 through an intake valve. The hydrocarbon source gas 4 is then broken down into its elements which interact with the catalyst 3 resulting in the growth of carbon nanotubes. The exhaust gas 6 is removed from the reaction chamber 1. In one embodiment of this method, argon 5 is pumped into the reaction chamber 1 for purging.

FIG. 2 shows a microwave CVD system 9 according to a preferred embodiment of the present invention. Until now, this method has involved the use of a furnace to heat the reaction chamber 33. The present invention uses a magnetron 10 in place of a furnace. Although a magnetron 10 capable of producing 750 W is preferred, any commercially-available magnetron may be used. The magnetron 10 creates a microwave field inside the reaction chamber 33.

In the preferred method of the invention, a known amount of the catalyst and catalyst support 15 are dispersed onto the substrate 34. The substrate 34 is then loaded into the reaction chamber 33. In this embodiment, the substrate 34 is loaded in and out of the reaction chamber 33 in a quartz container 19. The magnetron 10 is then switched on to heat the substrate 34 to the reaction temperature. In this embodiment, the reaction temperature is set to 700°C. During heating, an inert gas 22, at an optimized flow rate, can be used for purging, although the use of an inert gas 22 is not required for the present invention. When the reaction temperature is reached, a hydrocarbon source gas 21 is introduced into the reaction chamber at an optimal flow rate. The gases are pumped into the reaction chamber 33 through the gas inlet 17 and blown onto the substrate 34 using a quartz gas distributor 16. Exhaust gas leaves the chamber through the gas outlet 18. In this embodiment the reaction is set to 30 minutes. After the reaction, the resulting product is scratched from the substrate. Previous experiments by Varadan, V. et al. [Synthesis of carbon nanocylinders by microwave CVD] Smart Mater. Struct. 11 (2002) 728-734 did not involve the production of carbon nanocylinders, and utilized different processing materials and conditions than described in the present invention.

The advantage of using a microwave field is that it is uniform throughout the reaction chamber 33. This uniform field allows for a higher quantity of coiled carbon nanotubes with consistent properties to be produced during each reaction. In addition, a microwave field can be instantly turned off whereas a reaction chamber heated by a furnace must be allowed to cool to room temperature before any nanotubes can be extracted. This dramatically reduces the lag time between production rounds. In the preferred embodiment of the invention, a commercially available stub tuner 12 is used prevent any reflected power from flowing into the magnetron 10. In one example, the inventors manually adjusted the stub tuner 12. In another example, a commercially available three-port circulator 13 was used to automatically adjust the stub tuner 12. In addition, the invention may comprise a circulating chiller 14 which cools the magnetron 10 and therefore extends its life. The invention may further comprise a stirrer 27, which assists in making the microwave field uniform. The stirrer 27 is driven by a motor 28.

The reaction chamber 33 can be made from any number of materials without departing from the scope of the present invention. For instance, in one embodiment the reaction chamber is constructed out of aluminum. In a further embodiment, the reaction chamber is made of steel. In the preferred embodiment, the reaction chamber is a cylinder. The inventors used two reaction chambers manufactured by HVS Technologies. The smaller reaction chamber had dimensions of 14" in length and 5.75" in diameter. The larger reaction chamber had dimensions of 70" in length and 35" in diameter.

Although the catalyst can be made from various materials without departing from the scope of the present invention, a preferred catalyst is iron. Iron is preferred because it produces the highest yield of coiled carbon nanotubes. Alternatively, other transition metal catalysts can be used; including combinations of transition metals (e.g., bimetallic catalysts). It is important to note here that the indium-tin-iron catalyst disclosed in U.S. Pat. No. 6,583,085 to Nakayama et al. is not preferred for this invention. The presence of tin in the catalyst would cause the catalyst to spark when placed in the microwave field. In addition, the tin-iron catalyst would not be preferred does not easily absorb the microwaves.

The specific support used in the method of the present invention is critical. The support must contain pores giving rise to the growth of coiled carbon nanotubes according to the invention as opposed to other formations, such as fibers. The supports must also be able to easily absorb microwaves. Some non-limiting examples include silica, zeolite, and magnesium carbonate (preferred). Preferred pore sizes lie in the range of 0.1 to 10 nm with a surface area of 250-300 m²/g. The following are three examples of catalyst supports and catalysts with which they were combined (being just three examples of “supported metal catalyst”).

EXAMPLES

Catalyst and Catalyst Support #1

Iron nitrate and magnesium carbonate were weighed 1:1 weight ratio. Iron nitrate was dissolved in water and the resulting solution was added to magnesium carbonate, followed by continuous stirring to obtain a semi-solid mixture. The semi-solid mixture was kept inside an overnight at 500°C. After allowing the mixture to cool to room temperature, the resulting brown color solid was powdered. While the pore size of the magnesium carbonated varied somewhat throughout its surface, a majority were 10 nm in diameter.
Iron nitrate and silica were weighed 1:1 weight ratio. Iron nitrate was dissolved in water and the resulting solution was added to silica, followed by continuous stirring to obtain a semi-solid mixture. The semi-solid mixture was kept inside an oven at 120ºC overnight. After allowing the mixture to cool to room temperature, the resulting brown color solid was powdered. As a porous substance, the pore sizes for silica varied throughout its length.

The inventors used “hydrothermal processing” to manufacture zeolite (although commercial grade zeolite may be used). The hydrothermal processing method is described in Cundy, C. et al. [“The Hydrothermal Synthesis of Zeolites: History and Development from the Earliest Days to the Present Time” Chem. Rev. 2003, 103, 663-701]. Nickel acetate was dissolved in water and a proper amount of zeolite was added into the solution with a Ni percentage in zeolite of 14.5 wt%. The gel solution was stirred and kept in an oven at 120ºC overnight. After drying, the solid was crushed into a fine powder. While the pore size for zeolite varied throughout its surface, the majority were 1 nm in diameter.

The substrate 34 is made of silicon carbide. Although the hydrocarbon source gas 21 can be any gas containing carbon, in the preferred embodiment the hydrocarbon source gas is acetylene. The inventors found that the optimal flow rate for acetylene is 30 sccm for the smaller reaction chamber (14.5±5.75") and 600 sccm for the larger one (70±35"). Other non-limiting examples include methane, ethane and propane.

When an inert gas 22 is also used as described herein, helium is preferred, although any inert gas can be used (such as argon). The inventors found the optimal flow rate for helium is 190 sccm for the smaller reaction chamber and 3500 sccm for the larger one. It is important to mention here that for the synthesis of carbon nanotubes by a conventional CVD method, the presence of an impurity gas (e.g. thiophene) is necessary, while in the microwave CVD method, no impurity gas is required.

To optimize the processing conditions, the temperature of the reaction chamber 33 and the gas flow rates can be monitored by a computer 26. FIG. 4 is a software flow chart for the flow control systems. The temperature of the reaction chamber 33 is monitored by a pyrometer 25, which in the preferred embodiment is an optical pyrometer 25. The temperature readings taken by the pyrometer 23 are transmitted to a computer 26. The computer 26 then compares the temperature of the reaction chamber 33 with the set temperature for processing (preferably 700ºC). The computer 26 then controls the switching power supply 11 which in turn controls the magneto 10. If the reaction chamber temperature is too low, the computer 26 will tell the switching power supply 11 to turn on the magneto 10 and increase the temperature. If the reaction chamber temperature is too low, the computer 26 will tell the switching power supply 11 to turn off the magneto 10. The computer 26 also communicates with the master flow controller 20 which controls the mass flow controllers 24. The mass flow controllers 24 control the flow rates of the inert 22 and hydrocarbon source gas 21.

An illustration of a coiled carbon nanotube is shown in FIG. 3. The distance between the coils is substantially uniform throughout the length of each coiled carbon nanotube. In addition, the diameter of each coiled carbon nanotube will also be substantially uniform. The coiled carbon nanotube 29 is composed of carbon rings in the shapes of pentagons 32, hexagons 30, and heptagons 31. Depositing the various shapes in specific locations along the surface of the carbon nanotube causes the carbon nanotube to assume a coiled shape. Depending upon the distance between the coils, the non-hexagonal/hexagonal ratio of the carbon rings ranges from 0.1:1 to 1:1. A non-hexagonal/hexagonal ratio of 0.1 produces a “loose” coil with a large pitch. A non-hexagonal/hexagonal ratio of 1:1 produces a “tight” coil with a small pitch. The exact morphology of coiled carbon nanotubes will depend on the catalyst/catalyst support that is used and the conditions of the microwave CVD.

A scanning electron microscope (SEM 3000N manufactured by Hitachi) was used to investigate the morphology of the coiled carbon nanotubes. Due to the conducting property of carbon nanotubes, no gold coating is necessary for SEM operation. A transmission electron microscope (TEM 420T manufactured by Philips) was used to study the nanostructure of the coiled carbon nanotubes. TEM samples were prepared by ultrasonic vibration of a small amount of material in acetone followed by dropping on a TEM grid (Lacey carbon film on 300 mesh copper grid, Electron Microscopy Science).

SEM micrograph shown in FIG. 5, the coiled morphology for the microwave CVD samples is clearly revealed. From the TEM micrograph shown in FIG. 6, the hollow structure as well as coiled morphology for the microwave CVD samples was confirmed.

Under optimized conditions, the inventors have achieved ~90% yield of coiled carbon nanotubes. While the invention has been particularly shown and described with reference to preferred embodiments thereof, it will be understood by those skilled in the art that various alterations in form and detail may be made therein without departing from the spirit and scope of the invention. In particular, the particular metal catalysts, supports, source gas, and flow rates used can vary significantly and still be within the optimization scope of the present invention.

Having thus described the invention, what is claimed is:

1. A coiled carbon nanotube having a non-hexagonal/hexagonal carbon ring ratio in the range of 0.1:1 to 1:1.
2. The coiled carbon nanotube of claim 1 wherein the non-hexagonal/hexagonal carbon ring ratio is 0.1:1.
3. The coiled carbon nanotube of claim 1 wherein the non-hexagonal/hexagonal carbon ring ratio is 1:1.
4. The coiled carbon nanotube of claim 1 wherein the nanotube comprises a substantially uniform distance between coils throughout its length.
5. The coiled carbon nanotube of claim 1 wherein the nanotube comprises a substantially uniform diameter throughout its length.
6. The coiled carbon nanotube of claim 1 wherein the nanotube comprises a substantially uniform distance between coils and diameter throughout its length.
7. The coiled carbon nanotube of claim 1 wherein the nanotube comprises a diameter of less than 1000 nm.
8. The coiled carbon nanotube of claim 1 wherein the nanotube comprises a diameter of less than 100 nm.
9. The coiled carbon nanotube of claim 1 wherein the nanotube comprises a distance between coils of less than 1000 nm.
10. The coiled carbon nanotube of claim 1 wherein the nanotube comprises a distance between coils of less than 200 nm.
11. The coiled carbon nanotube of claim 1 wherein the nanotube comprises a diameter of less than 1000 nm and a distance between coils of less than 1000 nm.
12. The coiled carbon nanotube of claim 1 wherein the nanotube comprises a diameter of less than 100 nm and a distance between coils of less than 200 nm.
13. A coiled carbon nanotube having a substantially uniform diameter throughout its length.
14. The coiled carbon nanotube of claim 13 wherein the nanotube comprises a diameter of less than 1000 nm.
15. The coiled carbon nanotube of claim 13 wherein the nanotube comprises a diameter of less than 100 nm.
16. A coiled carbon nanotube wherein the nanotube comprises a substantially uniform distance between coils throughout its length.
17. The coiled carbon nanotube of claim 16 wherein the nanotube comprises a distance between coils of less than 200 nm.
18. The coiled carbon nanotube of claim 16 wherein the nanotube comprises a distance between coils of less than 1000 nm.
19. A coiled carbon nanotube having a substantially uniform diameter and a substantially uniform distance between coils throughout its length.
20. The coiled carbon nanotube of claim 19 wherein the nanotube comprises a diameter of less than 1000 nm and a distance between coils of less than 1000 nm.
21. A method of manufacturing coiled carbon nanotubes, comprising:
   placing a supported metal catalyst inside of a reaction chamber;
   creating a microwave field inside said reaction chamber;
   introducing a hydrocarbon source gas into said reaction chamber; and
   reacting for a time and at a temperature sufficient to form said coiled carbon nanotubes.
22. The method of claim 21, wherein an inert gas is introduced into said reaction chamber.
23. The method of claim 21, wherein said source gas is acetylene.
24. The method of claim 21, wherein said metal catalyst comprises a metal selected from the group consisting of iron, nickel, cobalt, and vanadium.
25. The method of claim 21, wherein said catalyst support is selected from the group consisting of silica, zeolite, and magnesium carbonate.
26. The method of claim 21, wherein said metal catalyst is iron and said catalyst support is magnesium carbonate.
27. The method of claim 21, wherein said metal catalyst is iron and said catalyst support is silica.
28. The method of claim 21, wherein said metal catalyst is nickel and said catalyst support is zeolite.
29. The method of claim 21, further comprising the use of a stirrer to make said microwave field uniform.
30. The method of claim 21, further comprising a stub tuner.
31. The method of claim 30, further comprising a port circulator for controlling said stub tuner.
32. The method of claim 21, further comprising a circulating chiller.
33. A method for manufacturing coiled carbon nanotubes, comprising:
   placing a supported metal catalyst inside of a reaction chamber;
   creating a microwave field inside said reaction chamber;
   introducing a hydrocarbon source gas into said reaction chamber;
   using a feedback system to control the temperature inside said reaction chamber and the flow rate of said hydrocarbon source gas; and
   reacting for a time and at a temperature sufficient to form said coiled carbon nanotubes.
34. The method of claim 33, wherein an inert gas is introduced into said reaction chamber.
35. The method of claim 33, wherein said source gas is acetylene.
36. The method of claim 33, wherein said metal catalyst comprises a metal selected from the group consisting of iron, nickel, cobalt, and vanadium.
37. The method of claim 33, wherein said catalyst support is selected from the group consisting of silica, zeolite, and magnesium carbonate.
38. The method of claim 33, wherein said metal catalyst is iron and said catalyst support is magnesium carbonate.
39. The method of claim 33, wherein said metal catalyst is iron and said catalyst support is silica.
40. The method of claim 33, wherein said metal catalyst is nickel and said catalyst support is zeolite.
41. The method of claim 33, further comprising the use of a stirrer to make said microwave field uniform.
42. The method of claim 33, further comprising a stub tuner.
43. The method of claim 42, further comprising a port circulator for controlling said stub tuner.
44. The method of claim 33, further comprising a circulating chiller.
45. The method of claim 33, wherein said feedback system comprises:
   a pyrometer;
   a switching power supply;
   a computer;
   a master flow controller; and
   a mass flow controller.
46. A coiled carbon nanotube produced by the process of:
   placing a supported metal catalyst inside of a reaction chamber;
   creating a microwave field inside said reaction chamber;
   introducing a hydrocarbon source gas into said reaction chamber; and
   reacting for a time and at a temperature sufficient to form said coiled carbon nanotubes.
47. The coiled carbon nanotube of claim 46, wherein argon is introduced into said reaction chamber.

48. The coiled carbon nanotube of claim 46, wherein said source gas is acetylene.

49. The coiled carbon nanotube of claim 46, wherein said metal catalyst comprises a metal selected from the group consisting of iron, nickel, cobalt, and vanadium.

50. The coiled carbon nanotube of claim 46, wherein said catalyst support is selected from the group consisting of silica, zeolite, and magnesium carbonate.

51. The coiled carbon nanotube of claim 46, wherein said metal catalyst is iron and said catalyst support magnesium carbonate.

52. The coiled carbon nanotube of claim 46, wherein said metal catalyst is iron and said catalyst support is silica.

53. The coiled carbon nanotube of claim 46, wherein said metal catalyst is nickel and said catalyst support is zeolite.

54. The coiled carbon nanotube of claim 46, further comprising the use of a stirrer to make said microwave field uniform.

55. The coiled carbon nanotube of claim 46, further comprising a stub tuner.

56. The coiled carbon nanotube of claim 55, further comprising a port circulator for controlling said stub tuner.

57. The coiled carbon nanotube of claim 46, further comprising a circulating chiller.

58. The coiled carbon nanotube of claim 46, further comprising the use of a feedback system to control the temperature inside said reaction chamber and the flow rate of said hydrocarbon source gas.

59. A coiled carbon nanotube produced by the process of claim 58, wherein said feedback system comprises:

- a pyrometer;
- a switching power supply;
- a computer;
- a master flow controller; and
- a mass flow controller.

60. An article of manufacture produced by the process of:

- placing a supported metal catalyst inside of a reaction chamber;
- creating a microwave field inside said reaction chamber;
- introducing a hydrocarbon source gas into said reaction chamber; and
- reacting for a time and at a temperature sufficient to form said coiled carbon nanotubes.

61. The article of manufacture of claim 60, wherein argon is introduced into said reaction chamber.

62. The article of manufacture of claim 60, wherein said source gas is acetylene.

63. The article of manufacture of claim 60, wherein said metal catalyst comprises a metal selected from the group consisting of iron, nickel, cobalt, and vanadium.

64. The article of manufacture of claim 60, wherein said catalyst support is selected from the group consisting of silica, zeolite, and magnesium carbonate.

65. The article of manufacture of claim 60, wherein said metal catalyst is iron and said catalyst support is magnesium carbonate.

66. The article of manufacture of claim 60, wherein said metal catalyst is iron and said catalyst support is silica.

67. The article of manufacture of claim 60, wherein said metal catalyst is nickel and said catalyst support is zeolite.

68. The article of manufacture of claim 60, further comprising the use of a stirrer to make said microwave field uniform.

69. The article of manufacture of claim 60, further comprising a stub tuner.

70. The article of manufacture of claim 69, further comprising a port circulator for controlling said stub tuner.

71. The article of manufacture of claim 60, further comprising a circulating chiller.

72. The article of manufacture of claim 60, further comprising the use of a feedback system for controlling the temperature inside said reaction chamber and the flow rate of said hydrocarbon source gas.

73. The article of manufacture of claim 72, wherein said feedback system comprises:

- a pyrometer;
- a switching power supply;
- a computer;
- a master flow controller; and
- a mass flow controller.