

[0059] While the grit-shearing stage 10 has been described above in connection with a rotational tumbler for agitating the grit particles to cause shearing action, it should be understood that alternative or additional agitation mechanisms and devices may be used, such as stirrers, shakers, etc., to generate grit shearing action.

[0060] In an optional embodiment reflected in FIG. 1C, prior to the magnetic phase separation stage 12, fractions withdrawn from compartment(s) 42b and/or 44b are separately or together subjected to a filtration stage 11. The filtration stage 11 of FIG. 1C may be performed in the tumbler 32 after it has been emptied and cleaned. It should be understood that a separately provided/additional tumbler or other filtration equipment may be used for optional filtration stage 11. According to one implementation of the filtration stage 11, the materials withdrawn from compartment(s) 42b and/or 44b are reintroduced into the compartment 40b of the cleaned tumbler 32, and the rotating mechanism 34 is reactivated. The grit particles may be included or excluded from the compartment 40b during the filtration stage 11. Residual metallic catalyst particles will be attracted to the magnet 59 during filtration 11, while the desired CNTs will flow through the meshes 56.

[0061] According to the embodiment illustrated in the flowchart of FIG. 1, the magnetic separation stage 12 follows the grit-shearing stage 10. The materials in the compartment(s) 42b and/or 44b of the downstream cylindrical segment(s) 42 and/or 44 are withdrawn from the tumbler 32 and subjected to the magnetic phase separation stage 12 separately.

[0062] Generally, the magnetic phase separation stage 12 operates to substantially separate the debundled, sheared CNT materials from the amorphous carbon and crystalline graphene, tube bundles, catalyst, and grit taken from the tumbler 32 based on magnetic attractive forces and buoyancy. Separation in stage 12 usually is not 100 percent efficient, although enrichment of debundled, sheared CNTs as high as, for example, 100-fold has been estimated for processed, low grade CNT materials.

[0063] In certain implementations, magnetic phase separation stage 12 is comparable to a centrifugation operation because of its capability of separating the shortened CNT materials into fractions based on their hydrodynamic size.

[0064] An exemplary embodiment of an apparatus adapted for the magnetic phase separation stage 12 is illustrated in FIG. 5. It should be understood that other apparatus may be used to carrying out this stage 12. The apparatus is generally designated by reference numeral 70 in FIG. 5 and has the general appearance of a column. The apparatus 70 has a cylindrical outer body 72 standing on its lower end. At least one magnet 74 is disposed below the lower end of the outer body 72. An exemplary magnet 74 is a static neodymium magnet. Other magnetic materials may be used instead or in addition to magnet 74. For example, a non-static magnet, such an electro-magnet may be used for the magnetic phase separation stage 12.

[0065] The materials withdrawn from the compartment(s) 42b and/or 44b of the cylindrical segment(s) 42 and/or 44 of the tumbler 32 are introduced into the chamber of the apparatus 70 through a port in the upper end closure 78. Glycerol is preferably either pre-loaded into the chamber of the apparatus 70 or added with the extracted fluid of stage 10 through the end closure 78 port. Equal volumetric amounts of 100% glycerol and 1% SDS have been found adequate for conducted tests. Preferably, the glycerol, which has a higher

density than the aqueous SDS-solution and therefore sits below the aqueous solution in the apparatus 10, is fed in a relatively small amount to provide a relatively small column height, e.g., about 0.5 to 1 cm. By employing a small glycerol column height in the apparatus 70, the magnetic forces of the magnet 74 are better able to attract solid materials on the opposite side of the glycerol layer.

[0066] The chamber contents of the apparatus 70 are allowed to settle without agitation, preferably for 24 to 48 hours, although the time may vary depending upon various factors, including the strength of magnet 74, the amount of glycerol, the temperature of the liquid, and the effectiveness with which CNT byproducts such as catalysts and tube bundles were removed in grit-shearing stage 10. An exemplary neo-magnet 72 which has been employed by the inventors generates a 1000-1500 gauss field. The magnetic phase separation can be carried out, for example, at temperatures ranging from about 4° C. to about 37° C. Temperature variation can be used to alter the density of glycerol, which could be used to improve separation properties of different CNT materials, e.g., the density of glycerol could be increased at lower temperature to match the density of MWCNTs, which are denser than SWCNTs, to maintain and/or improve the resolution of this CNT band generated with these CNTs during magnetic phase separation. The magnetic separator may include a temperature controlling device, such as a water jacket or small enclosing temperature regulating box, for controlling temperature and temperature variation. The temperature range for this separation is restricted by the primary solvent (water) and the dispersant properties. While water freezes at 0° C., SDS precipitates at 10-12° C. Substitution of SDS with Lithium Dodecyl Sulfate (LDS), which has a lower precipitation temperature (4° C.), lowers the range of allowable temperature for carrying out operating this phase separation.

[0067] The magnetic phase separation stage 12 causes the chamber contents of apparatus 70 to segregate (or more precisely stratify) into a plurality of phases based on magnetic attractive forces and buoyancy. Greater magnetic attractive properties and higher mass of a material (e.g., catalyst particles and grit, respectively) cause it to sink within the chamber, whereas weaker magnetic attractive properties and lower masses cause a material (e.g., amorphous carbon) to rise towards the top of the chamber. The magnetic attractive properties and mass or buoyancy of each material essentially reach equilibrium at a level along the height of the chamber.

[0068] FIG. 5 shows an embodiment in which the materials settle and segregate into five (5) major discernible phases. The bottom phase contains metallic catalysts and CNTs. The high buoyancy of the metallic catalysts is offset by the attraction of the magnet 74, which pulls the magnetic catalysts to the bottom of the column. The bottom phase may also contain CNTs in which catalysts are entrapped or remain enjoined, and bundles of carbon nanotubes entangled with each other. The second of the five phases is a glycerol phase. Above the glycerol phase are debundled CNTs in a third phase. It should be understood that the debundled carbon nanotubes are not completely isolated from contaminants in this third phase. Additional steps discussed below may still be carried out for separating amorphous carbon, SDS and glycerol from the CNTs. Above the CNTs is a fourth phase of SDS and water. The top or fifth phase is known as the frass, and contains most