

## Supporting Information

### Quantitative Method for Analyzing Dendritic Carbon Nanotube Agglomerates in Dispersions Using Differential Centrifugal Sedimentation

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#### SYMBOLS AND PARAMETERS

Symbols and parameters used for calculations are given below.

$d$ : average diameter of Multi-walled carbon nanotubes (MWCNTs).

$l$ : average length of MWCNTs.

$\omega$ : rotational velocity (309–2308 rad/s).

$R$ : disk radius, which is the distance of the particles from the center of the disc cavity. We used a representative disk radius (39.83 mm)  $\frac{R_1 - R_0}{\ln(R_1/R_0)}$  for approximate calculations.

$R_0$ : diameter of liquid surface, which is the starting distance of the particles from the center of the disc cavity, determined by the volume of spin fluid used in the rotor (36.82 mm).

$R_1$ : photodetector location, which is a fixed distance from the center of the disc cavity (43.00 mm).

$R_2$ : inner radius of disc (46.25 mm).

$R_1 - R_0$ : settling distance (6.18 mm).

$c$ : sucrose mass concentration.

$c_o$ : sucrose mass concentration of outer fluid.

$c_i$ : sucrose average mass concentration of inner fluid.

$\Delta c = c_o - c_i$ : sucrose mass concentration difference between outer fluid and inner fluid.

$\rho_{f,o}$ : outer fluid density. We used outer fluid densities at a representative disk radius for approximate calculation (Table S1).

$\rho_{f,i}$ : inner fluid density.

$\rho_a$ : apparent particle density (1780 and 2100 kg/m<sup>3</sup> for Single-walled carbon nanotube (SWCNT), and MWCNT, respectively, measured by the He gas method).

$\Delta\rho = (\rho_a - \rho_{f,o})$ : density difference between apparent particle density and outer fluid density.

$\Delta\rho_{\text{fluid}} = \rho_{f,o} - \rho_{f,i}$ : density difference between outer fluid and inner fluid.

$\frac{d\rho_{f,o}}{dR}$ : density gradient (Table S1) .

$\eta$ : fluid viscosity. We used fluid viscosities at a representative disk radius (Table S1).

$\eta_{\text{dilute}}$ : fluid viscosity of water at 25°C (0.890×10<sup>-3</sup> kg/m s).

$D_d$ : diffusion constant of sucrose. We used the diffusion constant in dilute solution at 25 °C,  $D_{d,\text{dilute}}$  (5.22×10<sup>-10</sup> m<sup>2</sup>/s).<sup>1</sup>

$D$ : particle diameter used in simulating ideal spherical particle sedimentation.

$V$ : particle volume used in simulating ideal spherical particle sedimentation.

$D_{\text{stokes}}$ : stokes diameter calculated from the Stokes equation (eqS2) or from measured values.

$D_{\text{corrected}}$ : corrected hydrodynamic diameter calculated from eqS22 and measured values.

$\phi$ : porosity, which is the volume occupancy of the open pore volume with respect to the envelope volume in hydrodynamics.

$t$ : sedimentation time.

$t_r$ : representative sedimentation time. We typically used the sedimentation time representing the maximum value of extinction.

$u$ : settling velocity. We used the average settling velocity of  $\frac{R_1 - R_0}{t_r}$  for approximate calculations and plots.

**Table S1.** Density, viscosity, and density gradient of fluids inside the disk centrifuge apparatus.

Sucrose mass concentration	$\rho_{f,o}$ (kg/m <sup>3</sup> )	$\eta$ (kg/m s)	$\frac{d\rho_{f,o}}{dR}$ (kg/m <sup>4</sup> )
8-24%	1047.9	$1.351 \times 10^{-3}$	7026.9
2-40%	1049.8	$1.375 \times 10^{-3}$	17338.4
2-18%	1023.2	$1.089 \times 10^{-3}$	6732.0
2-8%	1011.4	$0.992 \times 10^{-3}$	2430.4
2-4%	1006.8	$0.957 \times 10^{-3}$	803.4

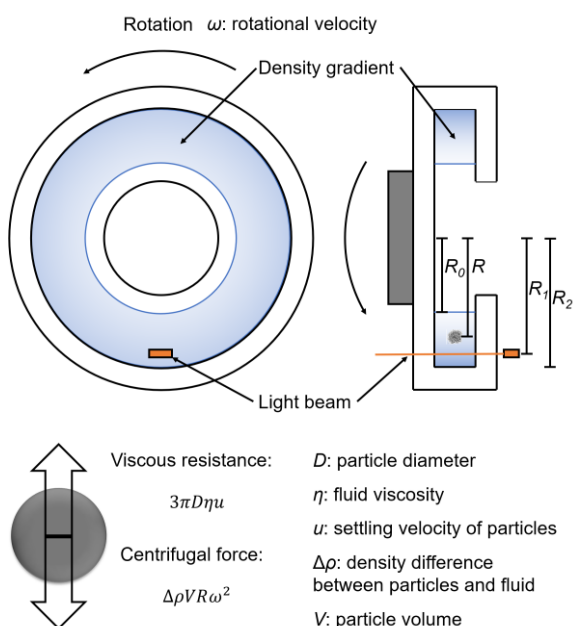
## EXPERIMENTAL

### *Carbon nanotube dispersions*

The SWCNTs we used in this study were synthesized by water-assisted chemical vapor deposition. The SWCNTs were grown as a vertical array on a Si substrate using iron nanoparticles as the catalyst and C<sub>2</sub>H<sub>4</sub> as the carbon source, with water-vapor as the growth enhancer.<sup>2</sup> The as-grown SWCNTs were removed from the substrate to obtain powders, which were used as starting materials

for suspension preparation. We obtained sodium deoxycholate-aided SWCNT suspensions by sonication according to the literature<sup>3</sup> (probe-type, Vibracell, power: 500 W, 40% power output). We mixed SWCNT powder with 10 wt/v% sodium deoxycholate aqueous solution. Then we dispersed SWCNT by ultrasonication for 2 h, resulting in a viscous 2.5 mg/mL SWCNT suspension. We concentrated the suspension by heating it to obtain a 4 mg/mL SWCNT paste. We diluted the paste to 0.0025 mg/mL, typically. We obtained 0.2 mg/mL multi-walled carbon nanotubes (MWCNTs) (vapor grown carbon fiber (VGCF), Showa Denko) with 1 wt/v% sodium deoxycholate aqueous solution by ultrasonication for 30 min.

#### ***Sedimentation time measurement with a disc centrifuge***



**Figure S1.** Schematic illustration of a disc centrifuge particle size analyzer (Front view, upper left, and side view, upper right) and schematic of viscous resistance and centrifugal force acting on a spherical settling particle.

We measured the sedimentation time of the suspensions with a disc centrifuge particle size analyzer (CPS Instruments, Disc Centrifuge CPS 24000 UHR (CR-39)) (Figure S1). The measurement conditions were as follows. Wavelength of optical beam: 405 nm, optical path length: 0.65 cm, temperature: 22–30°C. The density and viscosity values at 25°C were used for the calculations.

Calibration was performed using poly(vinyl chloride) particles (0.237 or 0.476  $\mu\text{m}$ , CPS Instruments). In detail, in order to compensate for the variation of the fluid volume by evaporation and density changes by temperature, the settling time was calibrated by the sedimentation of the spherical standard particles using eqS1.

$$t = t_{\text{measured}} \frac{18\eta \ln(R_1/R_0)}{(\rho_{\text{STD}} - \rho_{f,0})\omega^2 t_{r,\text{STD}}} \frac{1}{D_{\text{STD}}^2} \quad (\text{S1})$$

Here,  $t_{\text{measured}}$  is the recorded settling time,  $\rho_{\text{STD}}$  is the density of the standard particle (1385  $\text{kg/m}^3$ ),  $t_{r,\text{STD}}$  is the settling time of the standard, and  $D_{\text{STD}}$  is the diameter of the standard (0.237 or 0.476  $\mu\text{m}$ ). The density of the standard particle is discussed precisely by Vikram Kestens,<sup>4</sup> but in this paper, the above values were used as-is.

### ***Dynamic flow image analysis***

The sizes of the SWCNT particles in the suspensions were estimated by a wet-suspension type particle image analyzer (measurement range: 1  $\mu\text{m}$ –400  $\mu\text{m}$ , IF-400nano, Jasco International Co., Ltd.) fitted with a circulation pump. The thickness of the flow-cell was adjusted in accordance with the sizes of SWCNT particles using a paper spacer of 400  $\mu\text{m}$  thickness. After 1 min circulation of SWCNT suspensions by the pump and 5 s of sedimentation, the suspensions (10 mL) were circulated to capture 3000 to 4000 particles in a sampling volume of 1.2%. For particle imaging, we used a high pixel density (6.6 Megapixel) camera with telecentric lens, enabling a large field of focal depth.

The SWCNT particles in the suspension were illuminated from the back by a collimated light source (monochromatic blue light, 440 nm) to capture the projected images of the particles. To quantify the particle sizes, PIA-Pro particle image analysis software was employed. The size of each SWCNT particle in the suspensions was analyzed using the Feret diameter, which is the distance of the longest straight line connecting any two points on the perimeter, as a size parameter.

### ***“Common” centrifuge experiment***

To characterize the particles being detected by differential centrifugal sedimentation (DCS), we centrifuged 12 mL of the SWCNT suspension in a 15 mL centrifugal tube using a standard centrifuge (Kokusan H-36, RF-110 rotor, MC-110 bucket) and collected supernatant and precipitate. We obtained  $\omega$  as 367 rad/s and  $t$  as  $5.0 \times 10^2$  s from the Stokes equation (eqS2) with  $D_{\text{stokes}}$ : 0.43  $\mu\text{m}$  (see Figure 1c at 2222 rad/s),  $\Delta\rho$ : 782 kg/m<sup>3</sup> ( $\rho_a=1780$  kg/m<sup>3</sup>,  $\rho_{f,o} = 998$  kg/m<sup>3</sup>),  $\eta$ :  $0.890 \times 10^{-3}$  kg/m s,  $R_0$ : 0.088 m, and  $R_1$ : 0.162 m.

$$D_{\text{stokes}} = \sqrt{\frac{18\eta \ln(R_1/R_0)}{\Delta\rho\omega^2 t}} \quad (\text{S2})$$

Therefore we centrifuged at 367 rad/s for 10 min.

After centrifugation, we collected both 6 mL of the supernatant and 1 mL of the precipitate. We performed optical microscopy (Keyence, VHX-1000), dynamic flow image analysis to obtain the Feret diameter (JASCO, IF-400 nano), and DCS (2-18% sucrose density gradient, 1020 rad/s).

## **RESULTS AND DISCUSSION**

### ***Standard Stokes diameter description***

Here,  $D_{\text{stokes}}$  was calculated using the density and viscosity values at the position of the

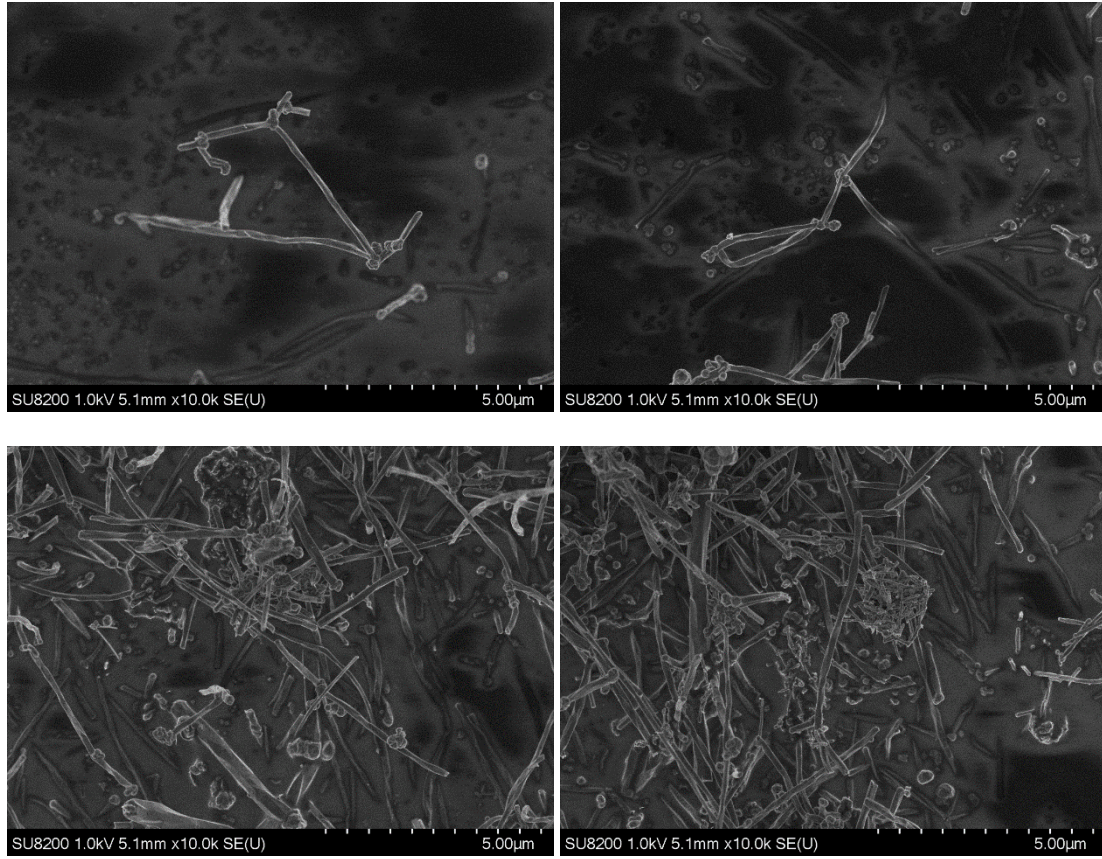
representative disk radius from eqS2.  $\omega$  is a parameter that has not been considered in DCS under the assumption that the  $D_{\text{stokes}}$  obtained for a particle does not depend on  $\omega$ . This premise enables a wide dynamic range of DCS.<sup>5-8</sup> The centrifugal force applied to a particle is expressed by  $\Delta\rho VR\omega^2$ . On the other hand, the viscous resistance acting on a settling spherical particle is expressed by  $3\pi D\eta u$ . Here,  $D$  and  $V$  are respectively the particle diameter and particle volume used in simulating an ideal spherical particle sedimentation. On the other hand, the value obtained by eqS2 from the measured value is called  $D_{\text{stokes}}$ , and is distinguished in this paper. Because the centrifugal force and the viscous resistance are balanced at the terminal settling velocity  $u$ , the Stokes equation (Eq2 below) is obtained from the centrifugal force  $\Delta\rho VR\omega^2$  and the viscous resistance  $3\pi D\eta u$ , and the relationship between the diameter and the volume of the sphere.

$$u = \frac{D^2 \Delta\rho R \omega^2}{18\eta} \quad (2)$$

When  $\omega$  is doubled, for example, the particles exhibit a 4-fold  $u$ . Therefore, the time for settling to the fixed distance  $t$  is reduced to one-fourth. According to eqS2,  $D_{\text{stokes}}$  produces the same value. In fact,  $\omega^2$  is proportional to  $u$  for most micro particles, including the MWCNTs in this experiment.

### ***MWCNT SEM observation***

The average diameter and the length of the MWCNTs were estimated by a field-emission type SEM (SU8200, Hitachi, acceleration voltage: 1.0 kV, magnification:  $\times 10\text{k}$  or  $\times 5\text{k}$ ) instrument. Line-sections across SEM images were utilized to randomly sample MWCNTs to obtain the average diameter ( $d$ ) and length ( $l$ ). A total of 100 MWCNTs from 29 images (4 images are shown in Figure S2) were used for this purpose.



**Figure S2.** SEM images of the MWCNTs.

***Inner fluid buoyancy (force balance equation)***

We propose the hypothesis that  $D_{\text{stokes}}$  of a porous particle changes with  $\omega$  in a density gradient because of inner fluid buoyancy  $\Delta\rho_{\text{fluid}}\phi VR\omega^2$ . Here,  $\Delta\rho_{\text{fluid}}$  is the density difference between the inner and outer fluids. When a particle sediments, it is conceivable that fluid flows inside the particle or flows around the particle (Figure 3). In the latter case, the porous particle holds the fluid internally. Owing to settling in the density gradient, the inner fluid density  $\rho_{f,i}$  is less than that of the surrounding fluid  $\rho_{f,o}$ . Here we consider the single sedimentation of a uniform porous spherical particle. The equation of motion of the particle is expressed by the centrifugal force, the viscous resistance, and the inner fluid buoyancy.



$$\{\rho_a(1 - \phi) + \rho_{f,i}\phi\}Va = \Delta\rho(1 - \phi)VR\omega^2 - 3\pi Du\eta - \Delta\rho_{\text{fluid}}\phi VR\omega^2 \quad (\text{S3})$$

Here,  $a$  is the acceleration in the sedimentation direction of the particles. The force balance equation is obtained if the settling velocity of the particles has reached the terminal velocity.

$$\Delta\rho(1 - \phi)VR\omega^2 = 3\pi Du\eta + \Delta\rho_{\text{fluid}}\phi VR\omega^2 \quad (3)$$

### ***Fluid density and solute mass concentration***

Here we consider  $\Delta\rho_{\text{fluid}}$ , which is the outer-inner density difference. The external fluid density  $\rho_{f,o}$  and internal fluid density  $\rho_{f,i}$  are expressed by (S4) and (S5), respectively.

$$\rho_{f,o} = f(c_o) \quad (\text{S4})$$

$$\rho_{f,i} = f(c_i) \quad (\text{S5})$$

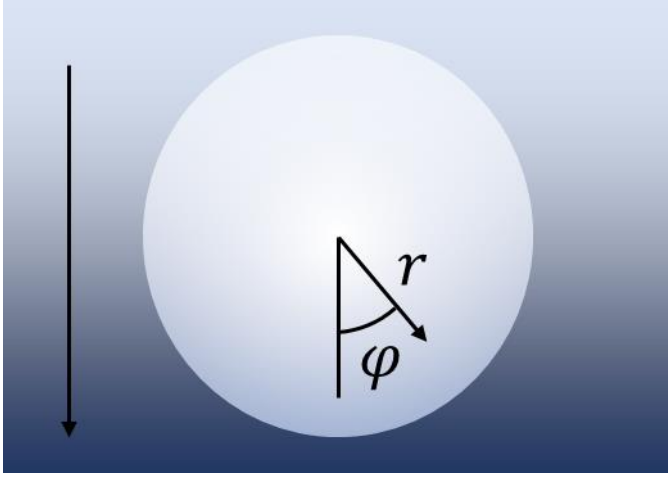
$f(c)$  is a function of fluid density to mass concentration of the solute, and  $c_o$  and  $c_i$  are the solute concentrations of the internal and external fluid, respectively. Here we consider  $\Delta\rho_{\text{fluid}}$ , which is the outer-inner density difference using  $\Delta c$ , which is the solute mass concentration difference.

$$\begin{aligned} \Delta\rho_{\text{fluid}} &= \rho_{f,o} - \rho_{f,i} \\ &= f(c_o) - f(c_i) \\ &= f(c_o) - f(c_o - \Delta c) \end{aligned} \quad (\text{S6})$$

$\Delta\rho_{\text{fluid}}$  can be expressed by eqS7 if  $\Delta c$  is small enough, so that the change in density is assumed to be proportional to the change in concentration.

$$\begin{aligned} \Delta\rho_{\text{fluid}} &= f(c_o) - f(c_o - \Delta c) \\ &= f(c_o) - \left\{ f(c_o) - \frac{\partial}{\partial c} f(c_o) \Delta c \right\} \\ &= \frac{\partial}{\partial c} f(c_o) \Delta c \end{aligned} \quad (\text{S7})$$

### ***Diffusion equation and the solute concentration distribution in non-equilibrium steady state of a spherical uniform porous particle settling at the terminal velocity in a linear density gradient***



**Figure S3.** Schematic illustration of porous sphere settling in a density gradient.

Next, we consider  $\Delta c$ . According to Fick's law, the diffusion equation in the spherical coordinate system  $(r, \theta, \varphi)$  of a uniform porous spherical particle is given by eqS8.

$$\frac{\partial c}{\partial t} = \left(\frac{1}{r^2}\right) \left\{ \frac{\partial}{\partial r} \left( D_d r^2 \frac{\partial c}{\partial r} \right) + \frac{1}{\sin^2 \varphi} \frac{\partial}{\partial \theta} \left( D_d \frac{\partial c}{\partial \theta} \right) + \frac{1}{\sin \varphi} \frac{\partial}{\partial \varphi} \left( D_d \sin \varphi \frac{\partial c}{\partial \varphi} \right) \right\} \quad (\text{S8})$$

The sedimentation direction origin is  $\theta = 0$ ,  $\varphi = 0$ , and  $\varphi$  is defined for the plane parallel to the particle sedimentation direction (Figure S3). We assume a non-equilibrium steady state in which the time increment of  $c_o$  as the particle settles at the terminal velocity in a gradient  $u \frac{\partial c_o}{\partial R}$  is equal to the time increment of  $c_i$  by diffusion  $\frac{\partial c_i}{\partial t}$ . We also assume that the diffusion constant  $D_d$  is constant within the particle. Under these assumptions, we solve the diffusion equation for the non-equilibrium steady state. We use a second-order partial differential equation of  $r$  to solve for  $c$  using constants A, B, C, and E.

$$c = Ar^2 + Br + C + Et, \quad \left( 0 \leq r < \frac{D}{2} \right) \quad (\text{S9})$$

On the other hand, the solute concentration of the external fluid  $c_o$  is expressed by the following equation.

$$c_o = c_{o,0} + u \frac{\partial c_o}{\partial R} t + \cos \varphi \frac{\partial c_o}{\partial R} r, \quad \left( \frac{D}{2} \leq r \right) \quad (\text{S10})$$

Here,  $c_{o,0}$  is the solute concentration of the external fluid at time  $t = 0$  and  $\varphi = \frac{\pi}{2}$ . We obtain  $B = \cos \varphi \frac{\partial c_o}{\partial R}$  from the boundary condition  $r = 0, \frac{\partial c}{\partial r} = \cos \varphi \frac{\partial c_o}{\partial R}$ . We define  $c_{c,0}$  as the solute concentration at the particle center at time  $t = 0$  ( $C = c_{c,0}$ ). We obtain  $A$  from the boundary condition  $r = \frac{D}{2}, \varphi = \frac{\pi}{2}, t = 0, c = c_{o,0}$ .

$$A = (c_{o,0} - c_{c,0}) \frac{4}{D^2} \quad (\text{S11})$$

Here, the solute average mass concentration of inner fluid  $c_i$  is expressed by:

$$\begin{aligned} c_i &= \frac{\int_0^{2\pi} \int_0^\pi \int_0^{\frac{D}{2}} c r^2 \sin \theta dr d\theta d\varphi}{V} \\ &= \frac{3}{5} c_{o,0} + \frac{2}{5} c_{c,0} + Et \end{aligned} \quad (\text{S12})$$

Therefore,  $\frac{\partial c_i}{\partial t} = E$ . We obtained the constant  $E$  from the assumption that  $\frac{\partial c_i}{\partial t}$  equals the time increment of  $c_o$  in the non-equilibrium steady state.

$$E = u \frac{\partial c_o}{\partial R} \quad (\text{S13})$$

Here we rewrite  $c_i$  below.

$$c_i = \frac{3}{5} c_{o,0} + \frac{2}{5} c_{c,0} + u \frac{\partial c_o}{\partial R} t \quad (\text{S14})$$

The difference between  $c_o$  at  $\varphi = \frac{\pi}{2}$  and  $c_i$  ( $\Delta c$ ) has a constant value, regardless of time, as follows.

$$\begin{aligned} \Delta c &= c_o - c_i \\ &= \frac{2}{5} (c_{o,0} - c_{c,0}) \end{aligned} \quad (\text{S15})$$

We rewrite  $c$  and  $c_i$ .

$$c = c_{o,0} - \frac{5}{2} \Delta c \left(1 - \frac{4}{D^2} r^2\right) + \cos \varphi \frac{\partial c_o}{\partial R} r + u \frac{\partial c_o}{\partial R} t, \quad \left(0 \leq r \leq \frac{D}{2}\right) \quad (\text{S16})$$

$$c_i = c_{o,0} - \Delta c + u \frac{\partial c_o}{\partial R} t \quad (\text{S17})$$

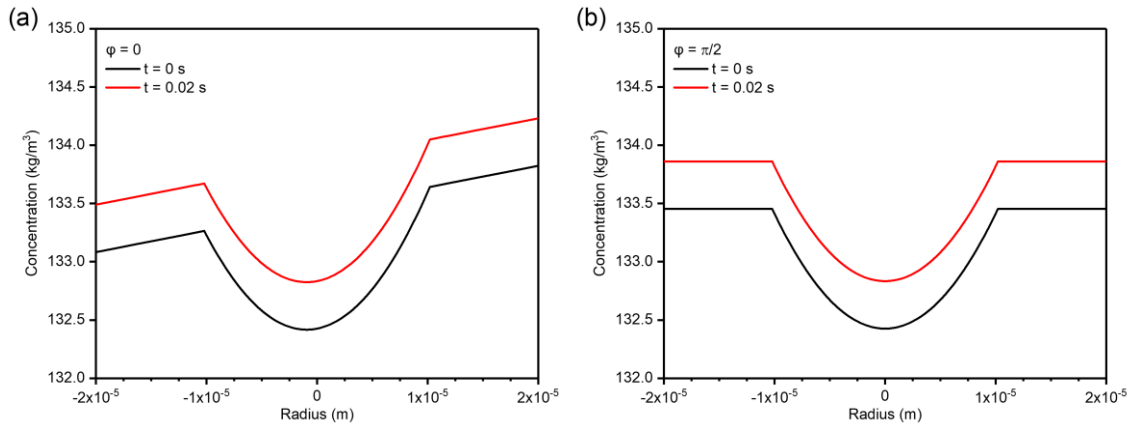
**Figure S4** shows the  $c$  distribution from eqS16 and the values of the SWCNT aggregate that settles in 8–24% sucrose aqueous solutions in the sedimentation direction ( $\varphi = 0$ ) and the vertical direction ( $\varphi = \frac{\pi}{2}$ ). The time  $t = 0$  s is the reference time after reaching the non-equilibrium steady state. It is

not the 0 s (the start) of the measurement time in the disc centrifuge. Substituting  $c$  into the diffusion equation (eqS8),  $\Delta c$  is expressed by the following equation using the diffusion constant of the solute  $D_d$ .

$$u \frac{\partial c_o}{\partial R} = \left( \frac{1}{r^2} \right) \left[ \frac{\partial}{\partial r} \left\{ D_d r^2 \frac{\partial}{\partial r} \left( \frac{5}{2} \Delta c \frac{4}{D^2} r^2 + \cos \varphi \frac{\partial c_o}{\partial R} r \right) \right\} + \frac{1}{\sin \varphi} \frac{\partial}{\partial \varphi} \left\{ D_d \sin \varphi \frac{\partial}{\partial \varphi} \left( \cos \varphi \frac{\partial c_o}{\partial R} r \right) \right\} \right]$$

$$u \frac{\partial c_o}{\partial R} = \frac{60 D_d}{D^2} \Delta c$$

$$\Delta c = u \frac{\partial c_o}{\partial R} \frac{D^2}{60 D_d} \quad (\text{S18})$$



**Figure S4.** Solute mass concentration distribution in the non-equilibrium steady state from eqS16.

We use the values of the SWCNT aggregate that settles in 8–24% sucrose aqueous solutions. (a) The sedimentation direction ( $\varphi = 0$ ). (b) The vertical direction ( $\varphi = \frac{\pi}{2}$ ).

### Inner fluid buoyancy

Substituting  $\Delta c$  of eqS18 into eqS7 yields the following equation.

$$\Delta \rho_{\text{fluid}} = u \frac{\partial \rho_{f,o}}{\partial R} \frac{D^2}{60 D_d} \quad (\text{S19})$$

Substituting  $\Delta \rho_{\text{fluid}}$  of eqS19 into inner fluid buoyancy  $\Delta \rho_{\text{fluid}} \phi V R \omega^2$ , inner fluid buoyancy is obtained  $u \frac{\partial \rho_{f,o}}{\partial R} \frac{D^2}{60 D_d} \phi V R \omega^2$ . Substituting  $\Delta \rho_{\text{fluid}}$  of eqS19 into eq3, the following equation is obtained.

$$\Delta\rho(1-\phi)VR\omega^2 = 3\pi D u \eta + u \frac{d\rho_{f,o}}{dR} \frac{D^2}{60D_d} \phi VR\omega^2 \quad (4)$$

From eq4 and the relationship between the diameter and the volume of the sphere, a simple equation for  $u$  is then obtained.

$$u = \frac{\alpha R \omega^2}{1 + \beta R \omega^2}$$

$$\alpha: \frac{\Delta\rho(1-\phi)D^2}{18\eta}$$

$$\beta: \frac{\frac{d\rho_{f,o}}{dR} \phi D^4}{1080\eta D_d}$$
(5)

Furthermore, in the solute concentration range in which the diffusion coefficient is inversely proportional to the solution viscosity,  $\beta$  can be written as follows using  $D_{d,dilute}$  and  $\eta_{dilute}$ .

$$\beta: \frac{\frac{d\rho_{f,o}}{dR} \phi D^4}{1080\eta_{dilute} D_{d,dilute}} \quad (S20)$$

If molecular diffusion can be assumed, it can also be written based on Boltzmann's constant  $k$ , temperature  $T$ , and solute molecular radius  $a'$  as follows.

$$\beta: \frac{\pi a' \frac{d\rho_{f,o}}{dR} \phi D^4}{180kT} \quad (S21)$$

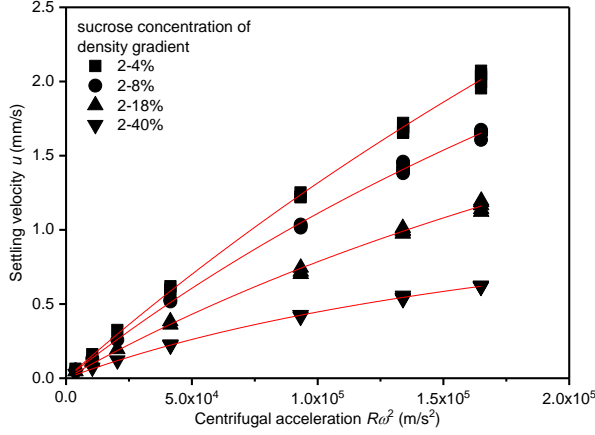
### ***Detailed fitting procedure***

Equation 5 explains the experimental results well. Here we define a corrected hydrodynamic diameter  $D_{corrected}$  as an approximate size calculated from eqS22 based on eq5 and measured values.

$$u \frac{18\eta \frac{\partial \rho_{f,o}}{\partial R}}{\Delta\rho} = \frac{(1-\phi) D_{corrected}^2 \frac{\partial \rho_{f,o}}{\partial R} R \omega^2}{1 + \frac{\phi D_{corrected}^4}{1080\eta_{dilute} D_{d,dilute}} \frac{\partial \rho_{f,o}}{\partial R} R \omega^2} \quad (S22)$$

Here,  $\eta$ ,  $\Delta\rho$ ,  $\frac{\partial \rho_{f,o}}{\partial R}$ , and  $R$  are the respective values for the representative disk radius, and  $u$  is the

average settling velocity. We first plotted  $u \frac{18\eta \frac{\partial \rho_{f,o}}{\partial R}}{\Delta \rho}$  versus  $\frac{\partial \rho_{f,o}}{\partial R} R\omega^2$  (Figure 4b) and fitted it with eqS22 to obtain the plot of  $u$  versus  $R\omega^2$  shown in Figure S5.



**Figure S5.** Plot of average settling velocity versus the centrifugal acceleration of SWCNT aggregate measured for different density gradients and rotational velocities.

We fitted the plot with the conditions below using Origin 2017J;

Formula:

$$Y = \frac{(1 - \phi) D_{corrected}^2 X}{1 + \frac{\phi D_{corrected}^4}{1080 \eta_{dilute} D_{d,dilute}} X} \quad (S23)$$

Independent variable:  $X: \frac{\partial \rho_{f,o}}{\partial R} R\omega^2$

Dependent variable:  $Y: u \frac{18\eta \frac{\partial \rho_{f,o}}{\partial R}}{\Delta \rho},$

Parameters (initial values):  $\phi$  (0.99),  $D_{corrected}$  ( $2.0 \times 10^{-5}$ )

Constant:  $\eta_{dilute} D_{d,dilute}$  ( $4.648 \times 10^{-13}$ )

### ***Estimation of hydrodynamic size and porosity***

Here we show how to estimate the hydrodynamic size and  $\phi$  of a particle with  $\omega$ -dependent

$D_{\text{stokes}}$ . (1) For higher precision, we recommend that you perform numerical simulations for solving the equation of motion (eqS3) and the diffusion equation (eqS11) of the particle at one-minute time intervals for two reasons. First, there is an initial fluid density difference between the sample layer and the density gradient. Second, note that  $\eta$  and  $\rho_{f,o}$  are functions of  $R$ . (2) Approximately and practically, you can estimate both  $D_{\text{corrected}}$  and  $\phi$  simultaneously by numerical analysis with eqS23. You should measure  $u$  for different values of  $\omega$ . Representative values of  $\eta$ ,  $\Delta\rho$ ,  $\frac{\partial\rho_{f,o}}{\partial R}$ , and  $R$  are sufficient for the approximate estimations. (3) If  $\phi$  is known, then  $D_{\text{stokes}}$  should be measured at the lowest possible  $\omega$  using the minimum necessary  $\frac{d\rho_{f,o}}{dR}$  to stabilize the sedimentation of the particles. Equation 5 indicates that the inner fluid buoyancy decreases by decreasing  $\frac{d\rho_{f,o}}{dR}$  and  $\omega$ . It is not desirable to measure buoyancy when using a uniform solution in the disc to prevent streaming effects.<sup>9</sup>

### ***Bulk density of SWCNT suspension in a liquid***

We prepared 0.4 wt/v% SWCNT dispersed in propylene glycol by sonication (probe-type, 1 h, Vibracell, power: 500 W, 40% power output). We diluted it into 0.00032, 0.016, 0.04, and 0.06 wt/v% solutions. We stirred them for 5 min at 500 rpm and let them stand for 7 days. The SWCNT suspension of 0.06 wt/v% occupied the full volume of liquid (Figure S6, rightmost column, bottom right), while that of 0.04 wt/v% did not (Figure S6, rightmost column, second from bottom right). Thus, the bulk density of a SWCNT suspension in a liquid is approximately 0.06 wt/v%.

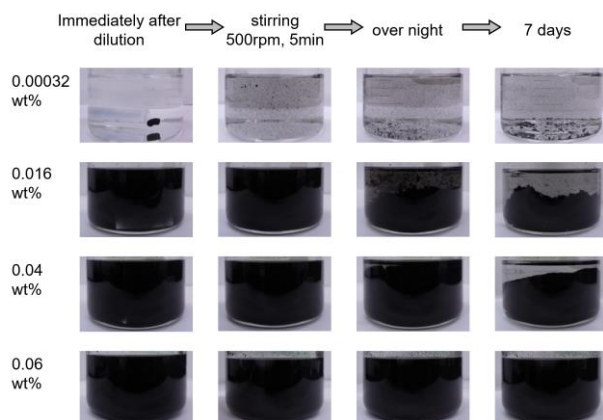


Figure S6 Photos of SWCNT suspensions.

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