Optimization of Surfactant Concentration in Carbon Nanotube Solutions for Dielectrophoretic Ceiling Assembly and Alignment: Implications for Transparent Electronics

Abdullah Abdulhameed,* Izhal Abdul Halin, Mohd Nazim Mohtar, and Mohd Nizar Hamidon

ABSTRACT: Surfactants such as sodium dodecyl sulfate (SDS) are used to improve the dispersity of carbon nanotubes (CNTs) in aqueous solutions. The surfactant concentration in CNT solutions is a critical factor in the dielectrophoretic (DEP) manipulation of CNTs. A high surfactant concentration causes a rapid increase in the solution conductivity, while a low concentration results in undesirably large CNT bundles within the solution. The increase in the solution conductivity causes drag velocity that obstructs the CNT manipulation process due to the electrothermal forces induced by the electric field. The presence of large CNT bundles is undesirable since they degrade the device performance. In this work, mathematical modeling and experimental work were used to optimize the concentration of the SDS surfactant in multiwalled carbon nanotube (MWCNT) solutions. The solutions were characterized using dynamic light scattering (DLS) and ultraviolet−visible spectroscopy (UV−Vis) analysis. We found that the optimum SDS concentration in MWCNT solutions for the successful DEP manipulation of MWCNTs was between 0.1 and 0.01 wt %. A novel DEP configuration was then used to assemble MWCNTs across transparent electrodes. The configuration was based on ceiling deposition, where the electrodes were on top of a droplet. The newly proposed configuration reduced the drag velocity and prevented the assembly of large MWCNT bundles. MWCNTs were successfully assembled and aligned across interdigitated electrodes (IDEs). The assembly of MWCNTs from aqueous solutions across transparent electrodes has potential use in future transparent electronics and sensor devices.

1. INTRODUCTION

Carbon nanotubes (CNTs) have attracted much attention in recent years due to their unique properties and usability in many scientific and research applications.¹ CNTs are produced in the form of agglomerates or bundles that are tightly attached to each other, making their dispersity and solubility in common solvents challenging.²,³ Solution-based processing is the main route available in manufacturing and engineering CNT-based devices, making the dispersion of CNTs in solutions an important research area.⁴

Two main approaches are used to improve the CNT dispersity in solvents, which are chemical and physical approaches. In the chemical approaches, CNTs are treated with acids to covalently attach functional groups onto their surface.⁵,⁶ The physical approaches (noncovalent treatment) use strong mechanical shear forces such as sonication to weaken the binding force between the CNTs with the help of agent materials (dispersants). Examples of dispersants are polymers,⁷,⁸−¹⁰ proteins,⁹,¹¹ and surfactants.¹²,¹³

Surfactants are preferred in dispersing CNTs because they do not cause any structural damage to the CNTs compared with the chemical approaches. Furthermore, the removal of the surfactant is much easier than that of polymers and proteins. Surfactants attached to the CNTs can be easily washed away after use using deionized water (DIW). There are three types of surfactants, which are anionic surfactants such as sodium dodecylbenzene sulfonate (SDBS),¹⁴ and sodium dodecyl sulfate (SDS),¹⁵ cationic surfactants such as dodecyl tri-methyl ammonium bromide (DTAB),¹⁶ and nonionic surfactants such as polyoxyethylene octyl phenyl ether (Triton X-100).¹⁷ Among the mentioned surfactants, SDS is widely used due to its low cost and easy processing procedures. SDS was used to prepare CNT solutions for several applications, such as nanocomposites,¹⁸ cement pastes,¹⁹ nanofluids,²⁰ antibacterial agents,²¹ and coating materials.²²

Many studies address the role of the SDS surfactant in CNT solutions in terms of the temperature effect.²³ SDS

Received: November 9, 2021
Accepted: December 30, 2021
Published: January 18, 2022
concentration,\textsuperscript{15} sonication power,\textsuperscript{24} and binding energy perspective.\textsuperscript{25} Yu et al. concluded that there are rules to disperse MWCNTs with the help of SDS and proper sonication.\textsuperscript{26} The first rule concerns the SDS/MWCNT ratio range, where the minimum weight ratio of SDS to MWCNTs to homogeneously dispersed MWCNTs in aqueous solutions was 1.5–1, while the maximum concentration was about 1.4 wt %. The second rule concerns the minimum sonication energy required, where a sonication time of 90 min (100 000 J) was enough to disperse MWCNTs at concentrations lower than 1.4 wt %. However, limited studies addressed the suitable SDS concentration in CNT solutions that are used in electrokinetic manipulation systems, such as the dielectrophoretic (DEP) deposition of MWCNTs.

DEP is an electrokinetic phenomenon that can be utilized to manipulate CNTs within the solution using nonuniform electric fields.\textsuperscript{27} An example of CNT manipulation is the deposition of CNTs across microelectrode structures in an aligned form.\textsuperscript{28,29} Despite the importance of the surfactants in dissolving CNTs, understanding their role in altering the aligned form\textsuperscript{28,29} is required to ensure successful deposition.\textsuperscript{30}

Controlling the electrical conductivity of CNT solutions by adjusting the surfactant concentration is one of the current challenges to avoid the occurrences of electrothermal phenomena in DEP systems, such as the joule heating effect and medium circulation due to the heat convection.\textsuperscript{31} Furthermore, solutions with high electrical conductivity allow more carriers to pass through the circuit, which might damage the microelectrodes at low frequencies or break the CNT connections across the electrode gaps.\textsuperscript{32} Optimizing the SDS concentration in DEP solution is required to ensure successful deposition while maintaining strong CNT dispersity. Additionally, introducing a new DEP setup compatible with CNT-SDS solutions is desirable in fabricating CNT-based devices such as transparent sensors.\textsuperscript{32}

In this article, the role of the SDS surfactant in dispersing MWCNTs in aqueous solutions and how it alters the electrical conductivity were studied. The study aims to optimize the concentrations of SDS in MWCNT solutions in order to use them in electrokinetic manipulation systems such as DEP systems. The article also presents a novel DEP configuration to assemble MWCNTs from the optimized solution and align them across microelectrodes.

2. THEORY AND MODELING

The electrical conductivity of CNT solutions plays a critical role in the manipulation of CNTs in DEP systems. Solutions with high electrical conductivity cause other forces to appear in the systems along with the DEP force. Examples of these forces are the electrothermal (ETH) and AC electroosmosis (ACEO) forces. In this section, the direct effect of the solution’s electrical conductivity on these forces is theoretically investigated. The DEP force is the motion of polarized particles in medium subjected to nonuniform electric fields. The magnitude of the DEP force depends on three main factors which are the particle geometry factor, \( v \), particle polarizability, \( \alpha \), and electric field, \( E \) as described by eq 1.\textsuperscript{34}

\[
F_{\text{DEP}} = \frac{1}{4} \alpha v E^2
\]  

(1)

The CNT geometrical factor depends on the CNT radius, \( r_{\text{cat}} \), and CNT length, \( l_{\text{cat}} \), and given by \( v = (2\pi r_{\text{cat}}^2 l_{\text{cat}})/3 \), while the effective polarizability factor depends on the complex permittivity of the CNT, \( \varepsilon_{\text{cat}} \), the complex permittivity of the medium, \( \varepsilon_{\text{m}} \), and the depolarization factor, \( L \), as expressed by eq 2.

\[
\alpha = \varepsilon_0 \varepsilon_{\text{m}} Re \left[ \frac{\varepsilon_{\text{cat}} - \varepsilon_{\text{m}}}{\varepsilon_{\text{m}} + (\varepsilon_{\text{cat}} - \varepsilon_{\text{m}}) L} \right]
\]  

(2)

The complex permittivity of the medium and CNT is described by eq 3.

\[
\varepsilon_{\text{m}} = \varepsilon_0 - \frac{\sigma_{\text{m}}}{\omega}, \quad \varepsilon_{\text{cat}} = \varepsilon_0 - \frac{\sigma_{\text{cat}}}{\omega}
\]  

(3)

At high frequencies (\( \omega \to \infty \)), the effective polarizability factor can be approximated by \( \alpha = \varepsilon_0 \varepsilon_{\text{m}} Re [(\varepsilon_{\text{cat}} - \varepsilon_{\text{m}})/(\varepsilon_{\text{cat}})] \), while at low frequencies (\( \omega \to 0 \)), the effective polarizability factor can be approximated by \( \alpha = \varepsilon_0 \varepsilon_{\text{m}} Re [(\sigma_{\text{cat}} - \sigma_{\text{m}})/(\sigma_{\text{cat}})] \). This means that medium electrical conductivity directly affects the DEP force at low frequencies.

The ACEO is the second force present in the electrokinetic system. ACEO occurs due to the existence of charges (negative or positive) at the solid–liquid interface. These charges form an electric double layer (EDL) due to the tangential component of the electric field. The EDL causes nonzero time-average Coulombic force on the ions at the electrode surface. This force causes medium drag velocity above the electrodes. The time-averaged velocity due to ACEO, \( U_{\text{aceo}} \), is expressed by the Smoluchowski formula (eq 4).\textsuperscript{34}

\[
U_{\text{aceo}} = \Lambda \varepsilon_0 \varepsilon_{\text{m}} \varepsilon_{\text{p}} \Omega^2
\]  

(4)

where \( \Lambda \), \( \varepsilon_0 \), \( \varepsilon_{\text{m}} \), \( V_{\text{p}} \), \( \eta_{\text{m}} \), and \( x \) are the EDL capacitance ratio, vacuum permittivity, medium relative permittivity, voltage potential, medium viscosity, and distance from the electrode gap center to the calculation point, respectively. \( \Omega \) is a dimensionless frequency expressed by eq 5.

\[
\Omega = \frac{\Lambda \varepsilon_0 \varepsilon_{\text{m}} \varepsilon_{\text{p}}}{x} \frac{x}{2\sigma_{\text{m}} \lambda_{\text{De}}}
\]  

(5)

The dimensionless frequency also depends on the signal angular frequency, \( \omega \), medium conductivity, \( \sigma_{\text{m}} \), and Debye length, \( \lambda_{\text{De}} \). The Debye length equals the square root of the product of diffusivity and medium permittivity conductivity ratio (\( \lambda_{\text{De}} = \sqrt{(\varepsilon_0 \sigma_{\text{m}} D)/\sigma_{\text{m}} \varepsilon_{\text{p}}} \)). From the equations given above, medium electrical conductivity directly affects the Debye length and the dimensionless frequency. Thus, the ACEO velocity is a function of medium electrical conductivity.

The third electrokinetic force in the electrokinetic system is the ETH force. The ETH force occurs in the medium due to the nonuniform heating caused by the flow of the electric current in the medium. The ETH is expressed by eq 6.\textsuperscript{34}

\[
F_{\text{ETH}} = \rho_{\text{A}} E - \frac{1}{2} \varepsilon_0 E^2 \nabla \varepsilon + \frac{1}{2} \sqrt{\frac{\rho_{\text{A}}}{\rho_{\text{v}}}} \left| \frac{\partial \varepsilon}{\partial \rho_{\text{v}}} \right| \left| \varepsilon E^2 \right|
\]  

(6)

where \( \rho_{\text{A}} \) and \( \rho_{\text{v}} \) are the charge and mass densities, respectively. The three terms at the right-hand side of the equation are the Coulomb force, the dielectric force, and the electrostriction pressure. The last term can be ignored since its gradient of a scalar quantity does not affect the incompressible fluid dynamics. Generally, the expression defines the electrical body force and fluid motion in terms of local variations in
permittivity and conductivity. The time-averaged body force can be written in terms of temperature gradient as described by eq 7.35,36

\[
\sigma \varepsilon \alpha \beta = - \nabla \cdot \left( - \frac{1}{\varepsilon} \nabla T \right) + \nabla \cdot \left( - \frac{1}{\sigma} \nabla \sigma \right)
\]

where \( E^* \) indicates the complex conjugation of the electric field. The approximation values of \( \alpha = (1/\varepsilon)(\partial \varepsilon / \partial T) \) and \( \beta = (1/\sigma)(\partial \sigma / \partial T) \) for aqueous solution are \(-0.4\%\) and \(+2\% K^{-1}\), respectively.37,38 The body force equation has two terms; the first term represents the Coulomb force, which is dominant at low frequency and the second term represents the dielectric force and dominates at high frequencies. Thus, the electrical conductivity of the medium directly affects the ETH force, especially at low frequencies.

Unlike the first three forces, the gravitational force, \( F_{grav} \), does not depend on the electric field. It depends on the CNT volume, \( v \), and the density difference between the medium and the CNT (\( \rho_{cnt} - \rho_m \)). The gravitational force acting on a CNT is described by eq 8.35

\[
F_{grav} = \gamma v (\rho_{cnt} - \rho_m) g
\]

where \( g \) is the gravitational acceleration. The magnitude of the CNT velocity induced by gravity is calculated by dividing the gravitational force described in eq 8 by a friction factor, \( \gamma \), represents the CNT mass flow rate \( (\rho_{cnt} l_{cat}/(ln(l_{cat}/r_{cat})) \).35 The CNT total velocity, \( U_{CNT} \), is the sum of the velocity induced by the DEP force, \( U_{DEP} \), velocity induced by the gravitational force, \( U_{grav} \), and medium drag velocity due to the ETH and ACEO, \( U_{drag} \) (eq 9).

\[
U_{CNT} = U_{DEP} + U_{grav} + U_{drag}
\]

CNTs are required to be deposited (assembled and aligned) between ITO electrodes. Figure 1 illustrates the conditions of the velocities to ensure successful deposition. The velocity induced by the DEP force is the only velocity in the direction toward the deposition area. Thus, the DEP velocity must be greater than the sum of the other velocities present in the DEP system. If the DEP velocity is less than the sum of the other velocities, the concentration of the SDS surfactant must be optimized again to reduce the medium conductivity. Details

Figure 1. Flow chart illustrates the conditions under which the CNTs can be successfully deposited across ITO electrodes. The colors of the arrows in the medium correspond to the velocity boxes in the flow chart, which show the velocity direction.

Figure 2. Electrode fabrication protocol and the fabricated electrode geometry. (1) ITO-coated substrate was cleaned using acetone, IPA, and DIW. (2) Positive photoresist (AZ 5214E) spin-coated the ITO and then baked for 2 min at a temperature of 90 °C. (3) Photoresist was exposed to UV light through a polyester photomask. (4) Developing process followed by hard baking for 2 h at a temperature of 120 °C. (5) ITO was etched using a mixture of HCL and HNO3 (4:1). (6) Final product was cleaned using acetone and IPA.
The dynamic light scattering (DLS) technique (Malvern Instruments Nano S) was used to measure the size distribution of MWCNTs in solutions. The solubility of MWCNTs and the quality of the solutions were determined by their absorbance to a specific wavelength using ultraviolet–visible (UV–Vis) spectroscopy (Perkin Elmer Lambda 35).

3.2. Electrode Fabrication. Figure 2 illustrates the standard lithography method that was used to fabricate ITO electrodes on glass substrates. First, the ITO layer was covered and spin-coated with a positive photoresist. The substrate was then heated on a hotplate to harden the photoresist layer. A standard lithography method that was used to fabricate ITO electrodes on glass substrates. First, the ITO layer was covered and spin-coated with a positive photoresist. The substrate was then heated on a hotplate to harden the photoresist layer.

4. RESULTS AND DISCUSSION

4.1. Solution Characterization. Figure 3a presents the variation in the electrical conductivity of the solutions as a function of SDS concentration. The solution’s electrical conductivity increased linearly when the concentration of the surfactant was increased. The fitting formula of the conductivity curve is expressed by eq 10

\[ \log_{10}(\text{conductivity (S/m)}) = -0.76 + (0.91 \times \log_{10}(\text{concentration (wt %)}) \]

Although SDS is a well-studied surfactant in terms of how it alters the conductivity of solutions, it is essential to experimentally measure the conductivity in the presence of MWCNTs. Figure 3b presents the solution conductivity at a constant SDS concentration (0.05 wt %) and varied MWCNT concentration (0.01–0.00125 wt %). The conductivity was in the same order (10−3 S/m) with standard deviations in the

Figure 3. Conductivity measurements. (a) Measured electrical conductivities of DIW at different SDS concentrations. (b) Measured electrical conductivities of MWCNT solutions at different MWCNT concentrations and a fixed SDS concentration (0.05 wt %). Tables S3 and S4 in the Supporting Information show the raw data, mean value, and standard deviation of the measured conductivities.
order of $10^{-4}$. This indicates that MWCNTs did not alter the medium electrical conductivity in the same way as the SDS did at the mentioned concentrations. The solutions with varied MWCNT concentrations are shown in Figure S2, along with field emission scanning electron microscopy (FESEM) and high-resolution transmission electron microscopy (HRTEM) images.

Figure 4a presents the intensity of the scattered light from suspended MWCNTs. The intensity peaked in a larger size range in the solution that does not contain SDS. However, when the SDS surfactant was added to the MWCNT solution, the intensity curve was shifted to the left, indicating strong solubility of large MWCNT bundles to individual tubes.

Figure 4b shows the size distribution of the dispersed MWCNTs. Before adding the MWCNTs, the particle size distribution of pure SDS solution was around 2–3 nm, representing the SDS micelle diameter. In the solution containing SDS and MWCNTs, the total volume percentage in small size ranges was more than the volume percentage in the solution with only MWCNTs. For example, the total volume percentage of tubes with sizes less than 350 nm in solution with SDS solution was 38%, which was 4% more than the total volume percentage of tubes in solution without SDS in the same size range. In conclusion, the difference in the particle size distribution when SDS was used with MWCNTs proves the success of the surfactant in dissolving large bundles into individual tubes.

Figure 5a shows the UV–Vis absorbance results of the solution at different SDS concentrations (the DIW curve was used as a baseline to compare different SDS concentration curves). The absorbance intensity was higher at higher SDS concentrations, which was expected for quantitative analysis. However, there was a fixed peak at 240–242 nm, and a concentration-dependent peak ranged from 208 nm at a concentration of 1.25 wt % to below 190 nm at concentrations less than 0.005 wt %.

Figure 5b shows the absorbance due to the presence of MWCNTs at a fixed MWCNT concentration of 0.001 wt % and different SDS concentrations (curves in Figure 5a were used as baselines to subtract the absorbance due to the SDS). High absorbance peaks were observed at 260–264 nm, which entirely agrees with other studies showing that the absorbance peak of an individual MWCNT was around 260 nm. The intensity peaks were convergent regardless of the concentration of the surfactant. The absorbance curve drops at concentrations of 0.5 and 1 wt %, which indicates an adverse effect of the surfactant at high concentrations in addition to its pre-effect in increasing the medium electrical connectivity. The absorbance also decreased at a concentration of 0.01 wt %, indicating low solubility of MWCNTs at an SDS concentration below 0.01 wt %. In Figure 5c, the SDS concentration was maintained at 0.05%, while the MWCNT concentration was varied from 0.00125 to 0.01 wt %. The higher concentration of MWCNTs resulted in more single tubes and thus a higher absorbance peak. The selection of the MWCNT concentration

Figure 4. DLS analysis and results of MWCNT solutions with and without the SDS surfactant. (a) Intensity of the scattered light from suspended MWCNTs. (b) Size distribution of MWCNTs with and without SDS. The concentrations of SDS and MWCNTs were 0.01 and 0.001 wt %, respectively.

Figure 5. UV–Vis analysis and results. (a) UV–Vis absorbance of the surfactant solution at different SDS concentrations. (b) Different SDS concentrations at an MWCNT concentration of 0.001 wt %. The inset figure shows the absorbance at wavelengths between 250 and 300 nm. (c) Different MWCNT concentrations at an SDS concentration of 0.05 wt %.

ACS Omega Article
http://pubs.acs.org/journal/acsodf
https://doi.org/10.1021/acsomega.1c06323
ACS Omega 2022, 7, 3680–3688
3684
usually depends on the required density of the deposited MWCNT layer.

In conclusion, the conductivity of MWCNT solutions exponentially increased with the increase in the SDS concentration. On the other hand, DLS and UV–Vis analysis showed that the addition of the SDS surfactant improves the MWCNT dispersity and solubility in aqueous solutions. SDS concentrations of ≥0.5 wt % are not desirable due to their

Figure 6. Simulation results of the DEP, ETH, and ACEO at different SDS concentrations. (a) DEP and ETH forces versus SDS concentration at a point located 10 μm below the electrode edge. The inset figure is the DEP force versus SDS concentration. (b) ACEO velocity vs SDS concentration at different frequencies at a point located 10 μm below the electrode edge.

Figure 7. Simulation results of the DEP and drag velocity at different SDS concentrations. (a) At location below the electrode edge. The inset figure illustrates the drag velocity at depths between −50 and −100 μm. (b) At location below the electrode center. The inset figure illustrates the drag velocity at depths between −50 and −100 μm.

Figure 8. Velocities induced on MWCNTs in the DEP system. (a) Drag velocity at an SDS concentration of 0.01 wt %. (b) Drag velocity at an SDS concentration of 0.1 wt %. (c) Drag velocity at an SDS concentration of 1 wt %. (d) DEP velocity at an SDS concentration of 0.01 wt %. (e) DEP velocity at an SDS concentration of 1 wt %. (f) Gravitational velocity at an SDS concentration of 1 wt %. The black arrows in the figures represent the direction of the velocity. Note that the simulation results in figure are at a location near the electrode surface. Further simulation results across the entire geometry can be found in Figure S3 (Supporting Information).
adverse effect on MWCNT solubility in addition to the massive increase in the medium electrical connectivity. SDS concentrations lower than 0.01 wt % were not inefficient in dispersing MWCNTs in DIW.

4.2. Simulation Results. The role of the SDS concentration in altering the electrokinetic forces can be realized by solving the equations discussed in the theory section. The resulting DEP force, ETH force, and ACEO velocity as a function of SDS concentration are discussed in the following paragraphs.

Figure 6a shows the magnitude of the ETH and DEP forces as a function of SDS concentration. Assuming that the permittivity of the medium is merely affected by SDS concentrations, the DEP force was almost constant as the SDS concentration increased from 0.001 to 1 wt %. On the other hand, there was a significant increase in the ETH force from $10^2$ to $10^6$ N/m$^3$ as the SDS concentration increased from 0.001 to 1 wt %. Figure 6b shows that the ACEO velocity increases linearly with the increase in the SDS concentration. For example, at a frequency of $10^5$ Hz, the ACEO velocity increased from $4.67 \times 10^{-7}$ m/s at a concentration of 0.01 wt % to $6.33 \times 10^{-6}$ m/s at a concentration of 0.1 wt %. The ACEO velocity is also a function of signal frequency where the velocity can be decreased by 2 orders of magnitude by increasing the signal frequency by 1 order of magnitude at the same SDS concentration.

Figure 7a shows the drag velocity and the DEP velocity at different depths using different SDS concentrations. The DEP was the dominant velocity near the electrode surface up to $-60 \mu m$ depth. At depths beyond $-60 \mu m$, the drag velocity becomes significant at an SDS concentration above 1 wt %. This means that MWCNTs located below 60 $\mu m$ are dragged away by the medium motion and cannot reach the deposition area. When the SDS concentration was reduced to 0.1 wt %, the drag velocity became lower than the DEP velocity at depths between $-60$ and $-80 \mu m$. Thus, the DEP velocity can attract MWCNTs from deeper locations. Further reduction in the SDS concentration has no effect as the DEP velocity attenuation is very strong. Figure 7b shows the velocities at depths below the electrode center. The velocity due to the DEP force was much weaker than the velocity at the electrode edge. However, the drag velocity dominated the DEP velocity at depths beyond $-40 \mu m$.

The results discussed in Figure 7 were taken at a specific location of the system geometry (electrode edge and electrode center). Figure 8 presents the velocity vectors at three different SDS concentrations below selected electrodes. Figure 8a–c shows that the intensity of the drag velocity increased with the increase in the SDS concentration. The increase in the velocity was significant at SDS concentrations of >1 wt %. Figure 8d,e shows that the SDS concentration does not affect the velocity due to the DEP force because no matter how concentrated the solution is, the electrical conductivity will not exceed that of the MWCNTs. The SDS concentration becomes critical in determining the DEP velocity direction (+DEP or −DEP) only when the manipulated particles have electrical conductivity in the same order as the solution.

Figure 8f shows that the velocity resulting from the gravitational force was a constant velocity directed to the ground (opposite of the deposition direction). The gravitational velocity of a suspended MWCNT depends on the MWCNT structure and dimensions. The variation in the gravitational velocity was not significant at different MWCNT lengths and densities. However, the increase in the MWCNT diameter significantly increased the gravitational velocity (see Figure S4 in the Supporting Information). Individual tubes have a diameter in the range of a few nanometers up to a few hundred nanometers, while the diameter of MWCNT bundles is equal to the average diameter of a single tube multiplied by the number of the tubes that form the bundle. Large MWCNT bundles that SDS fails to dissolve experience a stronger gravitational force. Thus, ceiling deposition is expected to eliminate the deposition of large MWCNT bundles and result in clean and homogeneous MWCNT networks.

In conclusion, low-conductivity media are required to avoid undesirable electrothermal and electroosmotic flows. Surfactant concentrations higher than 0.1 wt % caused a massive increase in the drag velocity at depths near the electrodes, which obstructs the suspended MWCNTs from reaching the deposition area. Minimum SDS concentration must be used with the help of ceiling deposition to avoid the assembly of undissolved and large MWCNT bundles.

4.3. Deposition Results. Deposition of MWCNTs across ITO was successfully conducted using ceiling deposition, as shown in Figure 9. The solution used in the deposition process has the SDS surfactant at a concentration of 0.05 wt % and MWCNTs at a concentration of 0.001 wt %. MWCNTs were accumulated instantaneously at the electrode edges because of the high-intensity DEP force at the electrode edges (Figure 9a). The MWCNTs continued to chain and attach to each other until complete connections were formed across the electrode gap (Figure 9b,c).

Figure 9d shows that the MWCNT connections were broken during the removal process. SDS molecules penetrate...
the gap between the tubes in the solution. However, these molecules break down while drying the medium, reducing the MWCNT–MWCNT contact force. This problem can be solved by diluting the droplet with DIW after forming the MWCNT connections, which could help in maintaining the quality of the aligned MWCNTs.

5. CONCLUSIONS

Surfactants such as SDS are used to improve the dispersity and solubility of MWCNTs in aqueous solutions to form MWCNT suspensions. MWCNT suspensions are used in many applications, including the manipulation of MWCNTs in a microfluidic channel using an electric field. Furthermore, the deposition of MWCNTs from a solution to an electrode structure is widely used in the fabrication of CNT-based devices such as transistors and sensors. In this work, we focused on optimizing the SDS concentration in MWCNT solutions used in DEP systems. The simulation results showed that SDS concentrations of more than 0.1 wt % were not desirable because they caused a massive increase in the medium drag velocity. SDS concentrations lower than 0.01 wt % were inefficient in dispersing MWCNTs in DIW. Thus, the optimum SDS concentration in MWCNT solutions for DEP deposition was between 0.1 and 0.01 wt %. The proposed DEP setup successfully assembled MWCNTs from the optimized deposition method and optimized bundles to move toward the drop surface away from the concentration because long-duration deposition allows large MWCNT assembly from solution with low SDS signal of 20 Vpp and 1 MHz. Ceiling deposition was preferable setup, simulation results, and large-scale pictures of the aligned MWCNTs.

5. CONCLUSIONS

Surfactants such as SDS are used to improve the dispersity and solubility of MWCNTs in aqueous solutions to form MWCNT suspensions. MWCNT suspensions are used in many applications, including the manipulation of MWCNTs in a microfluidic channel using an electric field. Furthermore, the deposition of MWCNTs from a solution to an electrode structure is widely used in the fabrication of CNT-based devices such as transistors and sensors. In this work, we focused on optimizing the SDS concentration in MWCNT solutions used in DEP systems. The simulation results showed that SDS concentrations of more than 0.1 wt % were not desirable because they caused a massive increase in the medium drag velocity. SDS concentrations lower than 0.01 wt % were inefficient in dispersing MWCNTs in DIW. Thus, the optimum SDS concentration in MWCNT solutions for DEP deposition was between 0.1 and 0.01 wt %. The proposed DEP setup successfully assembled MWCNTs from the optimized solution and aligned them across ITO electrodes using an AC signal of 20 Vpp and 1 MHz. Ceiling deposition was preferable in MWCNT assembly from solution with low SDS concentrations because long-duration deposition allows large bundles to move toward the drop surface away from the deposition area. The proposed method and optimized materials have potential use in the fabrication of future transparent wearable electronics such as sensors and detection devices.

ASSOCIATED CONTENT

Supporting Information
The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.1c06323.
Prepared solutions, FESEM, HRTEM, ceiling deposition setup, simulation results, and large-scale pictures of the aligned MWCNTs (PDF)

AUTHOR INFORMATION

Corresponding Author
Abdullah Abdulhameed — Department of Electronic Engineering, Faculty of Engineering, Hadhramout University, Mukalla 50511, Yemen; orcid.org/0000-0002-6122-8995; Email: gs50667@student.upm.edu.my

Authors
Izhal Abdul Halin — Department of Electrical and Electronic Engineering, Faculty of Engineering, Universiti Putra Malaysia, Serdang 43400, Malaysia
Mohd Nazim Mohtar — Department of Electrical and Electronic Engineering, Faculty of Engineering, Universiti Putra Malaysia, Serdang 43400, Malaysia
Mohd Nizar Hamidon — Institute of Advanced Technology (ITMA), Universiti Putra Malaysia, Serdang 43400, Malaysia; orcid.org/0000-0001-9189-4465

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.1c06323

Notes
The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was supported by Universiti Putra Malaysia under Grant GPB/2017/9563800. Also, the authors would like to express their heartfelt gratitude to the Hadhramout Foundation for supporting this research. This work is also dedicated to the fond memories of Dr. Nurul Amziah Mohd Yunus (1975-2019), who has spearheaded applications of DEP research in Universiti Putra Malaysia.

ABBREVIATIONS

ACEO, AC electroosmosis; CNTs, carbon nanotubes; DEP, dielectrophoresis; DLS, dynamic light scattering; DTAB, dodecyl tri-methyl ammonium bromide; EDL, electric double layer; ETF, electrophoretic; IDE, interdigitated electrodes; MWCNT, multiwalled carbon nanotube; SDBS, sodium dodecylbenzene sulfonate; SDS, sodium dodecyl sulfate; UV−Vis, ultraviolet–visible spectroscopy

REFERENCES

(1) Abdulhameed, A.; Wahab, N. Z. A.; Mohtar, M. N.; Hamidon, M. N.; Shafie, S.; Halin, I. A. Methods and Applications of Electrical Conductivity Enhancement of Materials Using Carbon Nanotubes. J. Electron. Mater. 2021, 50, 3207–3221.
(2) Ma, P.-C.; Siddiqui, N. A.; Marom, G.; Kim, J.-K. Dispersion and Functionalization of Carbon Nanotubes for Polymer-Based Nano-composites: A Review. Composites, Part A 2010, 41, 1345–1367.
(3) Feng, T.; Liu, N.; Wang, S.; Qin, C.; Shi, S.; Zeng, X.; Liu, G. Research on the Dispersion of Carbon Nanotubes and Their Application in Solution-Processed Polymeric Matrix Composites: A Review. Adv. Nano Res. 2021, 10, 559–576.
(4) Maillaud, L.; Poulin, P.; Pasquali, M.; Zakri, C. Effect of the Rheological Properties of Carbon Nanotube Dispersions on the Processing and Properties of Transparent Conductive Electrodes. Langmuir 2015, 31, 5928–5934.
(5) Karousis, N.; Tagmatachis, N.; Tasis, D. Current Progress on the Chemical Modification of Carbon Nanotubes. Chem. Rev. 2010, 110, 5366–5397.
(6) Abdulhameed, A.; Mohtar, M. N.; Hamidon, M. N.; Halin, I. A. Mild Nitric Acid Treatments to Improve Multi-Walled Carbon Nanotubes Dispersion and Solubility in Dielectrophoresis Mediums. Fullerences, Nanotub. Carbon Nanostruct. 2021, 29, 832–839.
(7) Ntim, S. A.; Sae-Khow, O.; Wittmann, F. A.; Mitra, S. Effects of Polymer Wrapping and Covalent Functionalization on the Stability of MWCNT in Aqueous Dispersions. J. Colloid Interface Sci. 2011, 355, 383–388.
(8) Voge, C. M.; Johns, J.; Raghavan, M.; Morris, M. D.; Stegemann, J. P. Wrapping and Dispersion of Multiwalled Carbon Nanotubes Improves Electrical Conductivity of Protein-Nanotube Composite Biomaterials. J. Biomed. Mater. Res., Part A 2013, 101A, 231–238.
(9) Salavagione, H. J.; Diez-Pascual, A. M.; Lázaro, E.; Vera, S.; Gómez-Fatou, M. A. Chemical Sensors Based on Polymer Composites with Carbon Nanotubes and Graphene: The Role of the Polymer. J. Mater. Chem. A 2014, 2, 14328–14338.
(10) Antonucci, A.; Kupis-Rozmysłowicz, J.; Boghossian, A. A. Noncovalent Protein and Peptide Functionalization of Single-Walled Carbon Nanotubes for Biodelivery and Optical Sensing Applications. ACS Appl. Mater. Interfaces 2017, 9, 11321–11331.
(11) Raffaini, G.; Ganazzoli, F. Surface Topography Effects in Protein Adsorption on Nanostructured Carbon Allotropes. Langmuir 2013, 29, 4883–4893.
(12) Cui, H.; Yan, X.; Monasterio, M.; Xing, F. Effects of Various Surfactants on the Dispersion of MWCNTs-OH in Aqueous Solution. Nanomaterials 2017, 7, 262.
Fernandes, R. M. F.; Abreu, B.; Claro, B.; Buzaglo, M.; Regev, O.; Furó, I.; Marques, E. F. Dispersing Carbon Nanotubes with Ionic Surfactants under Controlled Conditions: Comparisons and Insight. Langmuir 2015, 31, 10955–10965.

Alafogianni, P.; Dassios, K.; Farmaki, S.; Antiohos, S. K.; Matikas, T. E.; Barkoula, N.-M. On the Efficiency of UV-Vis Spectroscopy in Assessing the Dispersion Quality in Sonicated Aqueous Suspensions of Carbon Nanotubes. Colloids Surf. A Physicochem. Eng. Asp. 2016, 495, 118–124.

Blanch, A. J.; Shapter, J. G. Surfactant Concentration Dependent Spectral Effects of Oxygen and Depletion Interactions in Sodium Dodecyl Sulfate Dispersions of Carbon Nanotubes. J. Phys. Chem. B 2014, 118, 6288–6296.

Govda, J. I.; Gunjigavvi, D. G.; Sunagar, N. B.; Bhat, M. N.; Nandibewoor, S. T. MWCNT-CTAB Modified Glassy Carbon Electrode as a Sensor for the Determination of Paracetamol. RSC Adv. 2015, 5, 49045–49053.

Van Nguyen, H.; Tun, N. M.; Rakov, E. G. Dispersion of Carbon Nanomaterials in an Aqueous Medium Using a Triton X-100 Surfactant. Russ. J. Inorg. Chem. 2015, 60, 536–540.

Li, Z.; Zhang, Y.; Liang, S.; Li, G. Effect of Surfactant on Microstructure, Surface Hydrophilicity, Mechanical and Thermal Properties of Different Multi-Walled Carbon Nanotube/Poly(styrene) Composites. Mater. Res. Express 2018, 5, 055035.

Mendoza Reales, O. A.; Ocampo, C.; Arias Jaramillo, Y. P.; Ochoa Botero, J. C.; Quintero, J. H.; Silva, E. C. C. M.; Toledo Filho, R. D. Reinforcing Effect of Carbon Nanotubes/Surfactant Dispersions in Portland Cement Pastes. Adv. Civ. Eng. 2018, 2018, 2057940.

Singh, K.; Sharma, S. K.; Gupta, S. M. Preparation of Long Duration Stable CNT Nanofluid Using SDS. Integ. Ferroelectr. 2020, 204, 11–22.

Bai, Y.; Park, I. S.; Lee, S. J.; Bae, T. S.; Watari, F.; Uo, M.; Lee, M. H. Aqueous Dispersion of Surfactant-Modified Multiwalled Carbon Nanotubes and Their Application as an Antibacterial Agent. Carbon 2011, 49, 3663–3671.

Ghasemi-Kahrizsangi, A.; Neshati, J.; Shariatpanahi, H.; Akbarinezhad, E. Effect of SDS Modification of Carbon Black Nanoparticles on Corrosion Protection Behavior of Epoxy Nanocomposite Coatings. Polym. Bull. 2015, 72, 2297–2310.

Vo, M. D.; Papavassiliou, D. V. Effects of Temperature and Shear on the Adsorption of Surfactants on Carbon Nanotubes. J. Phys. Chem. C 2017, 121, 14339–14348.

Huang, Y. Y.; Terentjev, E. M. Dispersion of Carbon Nanotubes: Mixing, Sonication, Stabilization, and Composite Properties. Polymers 2012, 4, 275–295.

Duan, W. H.; Wang, Q.; Collins, F. Dispersion of Carbon Nanotubes with SDS Surfactants: A Study from a Binding Energy Perspective. Chem. Sci. 2011, 2, 1407–1413.

Yu, J.; Grossiord, N.; Koning, C. E.; Loos, J. Controlling the Dispersion of Multi-Wall Carbon Nanotubes in Aqueous Surfactant Solution. Carbon 2007, 45, 618–623.

Kang, J.; Hong, S.; Kim, Y.; Baik, S. Controlling the Carbon Nanotube-to-Medium Conductivity Ratio for Dielectrophoretic Separation. Langmuir 2009, 25, 12471–12474.

Abdulhameed, A.; Mohtar, M. N.; Hamidon, M. N.; Mansor, I.; Halin, I. A. Characterization and Selective Deposition of Carbon Nanotubes from Carbon Nanoparticles Mixture Using Mild Acid Treatment and Electrokinetic Manipulation. Mater. Res. Express 2021, 8, 055603.

Kang, J.; Hong, S.; Jung, S.; Strano, M. S.; Choi, J.; Baik, S. Dielectrophoresis of Surface Conductance Modulated Single-Walled Carbon Nanotubes Using Cationic Surfactants. J. Phys. Chem. B 2006, 110, 1541–1545.

Naeni, A. K.; Nojeh, A. Effect of Solution Conductivity and Electrode Shape on the Deposition of Carbon Nanotubes from Solution Using Dielectrophoresis. Nanotechnology 2012, 23, 495606.

Song, L.; Yu, L.; Brunme, C.; Shaw, R.; Zhang, C.; Xuan, X. Joule Heating Effects on Electrokinetic Flows with Conductivity Gradients. Electrophoresis 2021, 42, 967–974.