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Thick Film Deposition of Carbon Nanotubes by Alternating Electrophoresis

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ABSTRACT

lectrophoretic deposition of carbon nanotubes (CNTs) using alternating current electric fields (0.01-1000 Hz) is reported. Pure acetone was used as suspending medium. Two parallel gold electrodes were used as depositing substrate. The effect of depositing parameters such as frequency and three waveforms (sinusoidal, rectangular and triangular) on deposit yield was investigated. According to our experiments, thick films of CNTs are obtainable at frequencies as low as 0.01 to 10 Hz. Results showed that the deposit yield decreases as the frequency increases from 0.01 Hz to 1000 Hz. The rectangular waveform yields more deposit mass compared with sinusoidal and triangular waveforms. Comparison between AC and DC electric fields revealed that the deposition pattern is also different in AC and DC electric fields. Prog. Color Colorants Coat. 3(2010), 27-31. © Institute for Color Science and Technology.

1. Introduction

Because of their novel structure and remarkable properties, carbon nanotubes (CNTs) have attracted tremendous attention since the discovery by Ijima in 1991 [1]. CNTs are good candidates for fabrication of micro- and nano-electronic devices such as chemical and physical sensors, field emitters and so on. In order to deposit CNTs on proper substrates and fabricating such high-tech devices, various methods have been used. For example, chemical vapor deposition (CVD) is the main procedure for synthesis and preparation of CNT thick films. But it suffers from the presence of carbonaceous impurities and metallic catalysts which remain in the synthesized film after the CVD process.

Deposition of CNTs between two conductive electrodes has been done via dielectrophoretic deposition (DEP) technique in which a non-uniform high frequency alternating electric field (in the order of 1-100 MHz) is applied between two conductive electrodes [2-7]. In addition to its high frequency, the electrodes used in this method such as castellated or hyperbolic microelectrodes, have complex shapes [8]. Fabrication of such microelectrodes needs expensive and complex techniques such as photolithography or plasma enhanced chemical vapor deposition (PECVD) [9,10]. It should be mentioned that this technique is sometimes used somewhat ambiguously to refer to AC electrophoresis.

On the other hand, in the conventional electrophoretic deposition (EPD), direct (DC) electric fields are applied

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to force the particles to deposit on either of the electrodes depending on the surface charge of the particles [11-13]. Electrophoretic deposition (EPD) is an easy and cheap method for deposition of CNTs for development of electromechanical actuators, supercapacitors, batteries, and electrochemical sensors [14,15]. EPD has the advantages of short formation time, simple deposition apparatus and suitability for mass production. In contrast to conventional EPD in which DC electric field is applied to move particles towards one of the electrodes, we used alternating (AC) electric field for deposition of CNTs thick films on the surface of parallel gold electrodes. The effect of electric field parameters such as frequency and waveform on the deposit yield is also investigated. Comparison between deposition pattern in AC and DC electric fields is also presented in this paper.

2. Experimental

2.1. Materials

Multiwall carbon nanotubes (MCNTs) was synthesized by chemical vapor deposition (CVD) technique (70% purity) with a diameter range of 70-140 nm and average length of 1-2 micron in Electrochemistry Lab., Materials and Energy Research Center (MERC). The SEM micrograph of the CNTs is shown in Figure 1. 0.01 %wt of purified CNTs were dispersed in pure acetone after 60 min ultrasonic agitation. The top fraction of the suspension was used for deposition process.

2.2. Deposition

The electrode fabrication procedure was as follow: a thin layer of gold paste was applied on the surface of a glass plate (2 cm×2 cm×8 mm). The gold layer was heated at 700 °C for 30 minutes. A 100 micron gap was created between the two electrodes by a sharp alumina tip. optical microscopy image of the fabricated electrode is shown in Figure 2. After preparation of both the CNT suspension and the electrodes, the deposition process was accomplished at different frequencies and the samples were dried at ambient temperature. The set up used for deposition process is illustrated in [16] and was consisted of a function generator (hp 3312A), a voltage amplifier (hp 6826A) to amplify the output voltage of the function generator up to 60 V, and a storage oscilloscope (hp 1201A). A DC power supply (model hp 6282A) was used for DC electrophoretic deposition. The deposited mass was measured after the deposition process by a 10⁻⁵ balance.

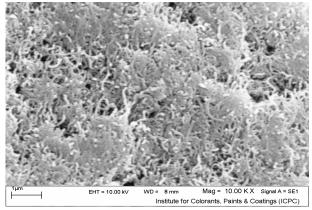


Figure 1: SEM image of the purified CNTs.

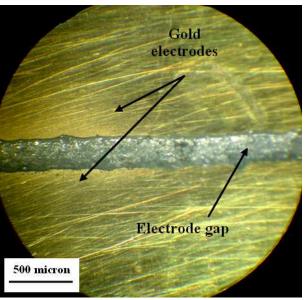


Figure 2: Optical microscopy image of the parallel gold electrodes before deposition. The white area between the gold electrodes is the glass substrate.

3. Results and discussion

The stereo optical microscopy image of the deposited sample at 40 V, 10 min and 0.1 Hz is shown in Figure 3(a). It is obvious that the CNTs have deposited on the surface of the parallel gold electrodes, because in such a low frequency alternating current (AC) mode, the electric field lines are distributed from the surface of one electrode to the surface of the other. So, the CNT particles deposit on the surface of both electrodes.

For comparison, deposition under the same conditions is repeated except that this time a direct current (DC) electric field is applied (Figure (3b)). In this case, CNT particles are deposited only on one electrode. This is due to the fact that in DC mode, electric field lines are only distributed from one electrode edge to another. Hence, the margins of one electrode which has

opposite sign with respect to particles surface charge is coated with CNTs. In general, by applying AC electric field, in the first semicycle, particles move towards one of the electrodes with the opposite charge. In the next semicycle in which the direction of the electric field inverses, the particles move towards the other electrode. After some minutes (which depends on the applied voltage, the frequency and the suspension concentration, etc.) a thick coating of CNTs deposits on the surface of both electrodes.

But this behavior depends on the electric field frequency. For example, Figure 4 shows the deposited CNTs after 10 min at the frequency of 1000 Hz and the voltage of 40 V. It is obvious that carbon nanotubes are aligned between the electrodes. It can be seen that the CNTs are deposited along the electric field lines from one electrode to another (aligned deposition). This aligned deposition does not occur at lower frequencies (in comparison with Figure 3(a)). We suggest that at high frequencies as 1000 Hz, the field lines are bounded within the gap, hence the CNT particles only can move between the two electrodes in the gap. Because of the tubular shape of the CNTs, they align parallel to electric field lines so that the electrodes connected to each other.

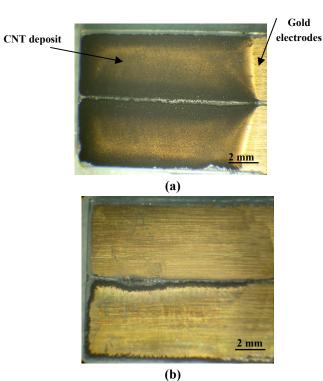


Figure 3: Stereo optical microscopy images of the samples obtained after 10 min deposition at (a) 0.1 Hz (rectangular wave) and (b) 40 V_{DC} .

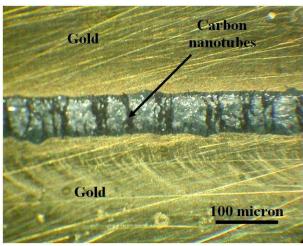


Figure 4: Stereo optical micrograph showing CNTs deposited at 1000 Hz (rectangular waveform), 10 min and 40 V. CNT bundles which are deposited parallel to each other are shown by black arrow.

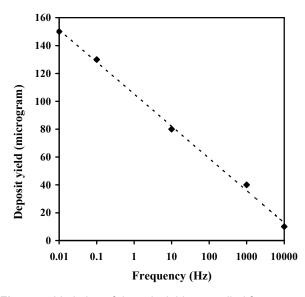


Figure 5: Variation of deposit yield vs. applied frequency at 40 V and 10 min (rectangular waveform).

For quantitative investigation, the variation of the deposit yield was measured versus electric field frequency. The deposit yield is calculated as deposited mass divided by the deposited area (in g/cm²). The deposited area was kept constant in all experiments, so the deposit yield is reported in microgram. According to the curve which is illustrated in Figure 5, the deposit yield decreases as the frequency of the electric field increases from 0.01 Hz to 10 kHz. This behavior is due to the fact that at higher frequencies, the particles have less time to response to the electric field so that at 10 kHz, the deposited yield is near to zero.

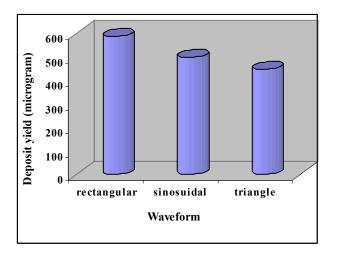


Figure 6: Variation of deposit yield vs. waveform at 40 V, 10 min and 0.1 Hz.

The effect of different waveforms on deposit yield is also investigated in this work. Figure 6 shows that the deposit yield increases as the waveform changes from triangular to sinusoidal and rectangular. This is because of the fact that in rectangular waveform, the effective time and voltage is more than that of the two other waveforms; consequently, the deposit yield obtained from rectangular wave form is slightly higher in comparison with triangular and sinusoidal wave forms This results is in good agreement with our previous works on SnO₂ and Si powders [17,18].

4. Conclusions

Alternating electric field was used for deposition of CNTs by electrophoretic technique successfully. The deposition pattern may varies depending on the applied frequency i.e. deposition may occur either on the surface or in the gap between the electrodes. This is due to the distribution of the electric field lines between the electrodes. The deposited yield decreased as frequency increased from 0.01 Hz to 10 kHz. The rectangular waveform had the most deposit yield because of its higher effective time and voltage compared with sinusoidal and triangular waveforms. This method may be used for mass production of CNTs-based devices such as gas sensors, field emission electrodes, etc.

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