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Multi-Walled Carbon Nanotube Ultrathin Film Using a Liquid-Liquid Interface: Effect of Alcohol Type to the Film Property

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Ultrathin film of multi-walled carbon nanotubes (MWCNTs) was fabricated using a liquid–liquid interface. Water dispersible MWCNTs, which were synthesized using an anodic aluminum oxide film as a template were used as a water phase. Hexane solution was added to the water dispersion as an oil phase. The MWCNTs were assembled at the liquid–liquid interface by adding 10 vol% of alcohol (methanol, ethanol, and 2-propanol) to the MWCNTs water dispersion/hexane solution. The assembled film was transferred to a solid substrate and the effect of alcohol type to the film properties was discussed.

Keywords: carbon nanotubes; liquid–liquid interface; ultrathin film

INTRODUCTION

Carbon nanotubes (CNTs) have strongly attracted attention to electrical and optical device applications because of their unique electrical, optical, and mechanical properties [1]. Because of these unique

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properties, CNTs have been applied to fabricate flexible electric devices, such as field effect transistors [2], transparent electrodes [3], and so on. To apply CNTs to flexible electric devices, it is important to fabricate uniform CNTs film using a solution based technique, such as spin coating [4], layer-by-layer assembly [5], and Langmuir-Blodgett technique [6,7]. Recently we have applied a liquid–liquid interface to assemble water dispersible multi-walled carbon nanotubes [8,9]. The surface wettability of the MWCNTs is one of the important physical values to induce the MWCNTs to assemble at the interface. In the previous paper, ethanol was used as a solvent to control the surface potential of the MWCNTs [9]. In this paper, we used other alcohols to assemble MWCNTs at the liquid–liquid interface and effect of surface potential controller solvent to the film properties was discussed.

EXPERIMENTAL SECTION

MWCNTs were synthesized using a template method reported elsewhere [10]. The size of the MWCNTs was 5 μm in length and 10–15 nm in diameter. After the synthesis, the MWCNTs were dispersed in ethanol (0.17 mg/mL). A silicon substrate was cleaned using O_3 treatment and immersed into 1 mM of 3-aminopropyltriethoxysilane in toluene for 40 min at 80°C to form an amino-terminated surface. The transferred films were observed by atomic force microscope (AFM, SPI400; Seiko Instruments Inc.). The ζ-potential analysis of MWCNTs were performed at 20°C with an electrophoretic light scattering spectrometer (ELS-8000; Otsuka Electronics).

RESULTS AND DISCUSSION

Decrease of MWCNT Surface Potential by Addition of Alcohol

We have reported that the surface potential of the MWCNTs decrease with increasing the amount of ethanol added [9]. The decrease is attributed to the adsorption of ethanol to the MWCNTs through hydrogen bonding. In this paper, methanol was added to the dispersion to study the effect of alcohol type to the reduction of ζ-potential. Figure 1 shows the ζ-potential of the MWCNTs measured by adding different amount of methanol. As similar to ethanol, the ζ-potential of MWCNTs was decreased with increasing the amount of methanol added. This result supports the idea that the adsorption of alcohol through hydrogen bonding compensates the negative charge at the MWCNTs surface. Therefore the interfacial energy of the MWCNTs is controllable by adding alcohol to the dispersion.

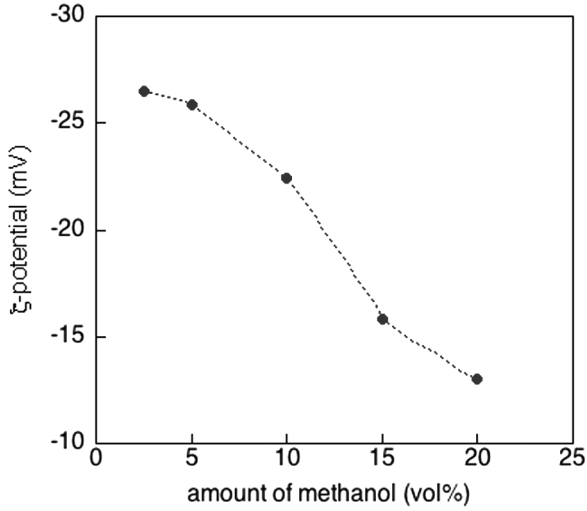

FIGURE 1 Values of ζ-potential for the water dispersion of the MWCNTs at different ethanol concentrations.

Fabrication of MWCNTs Film at the Liquid-Liquid Interface by Adding Alcohols

The assembling process of MWCNTs by addition of alcohols is schematically shown in Figure 2. First 147 μL of MWCNTs-ethanol dispersion (0.17 mg/mL) was added to the vessel with a diameter of 20 mm and a height of 50 mm (Fig. 2(a)). The ethanol dispersion was diluted with adding 8 mL of water (Fig. 2(b)). After addition of water, the vessel was sonicated for a few seconds to completely mix the solvent. Then 2 mL of hexane was added to the vessel to create the liquid–liquid interface (Fig. 2(c)). Ten vol% of alcohols were dropped from the top of the vessel using a syringe pump with a rate of 0.1 mL/min (Fig. 2(d)). After the addition of alcohols, we observed that MWCNTs were assembled at the liquid–liquid interface. The film prepared at the liquid–liquid interface was transferred to a silicon substrate by vertically immersing the substrate at a rate of 10 mm/min (Fig. 2(e)). Figure 3 shows AFM images of the film prepared by adding methanol, ethanol, and 2-propanol to the vessel. As reported previously, a densely packed ultrathin film was formed by an addition of 10 vol% of ethanol (Fig. 3(b)). On the other hand fabrication with adding the same volume amount of methanol results in a sparse film (Fig. 3(a)) and of 2-propanol results in an aggregated film (Fig. 3(c)). These phenomena indicate that the solvent hydrophobicity also plays an important role in film properties.

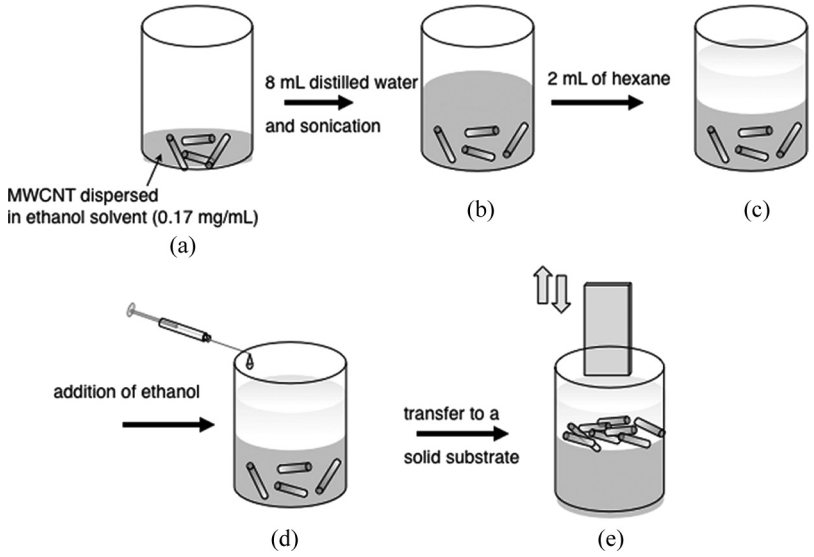

FIGURE 2 Procedure to fabricate MWCNTs monolayer at the liquid–liquid interface. (a) MWCNT-ethanol dispersion was added to the vessel with a diameter of 20 mm and height of 50 mm; (b) addition of 8 mL of distilled water to the dispersion; (c) addition of 2 mL of hexane to the vessel; (d) addition of 10 vol% of alcohol with a speed of 0.1 mL/min; (e) transferred onto a substrate by dipping the substrate with a speed of 10 mm/min.

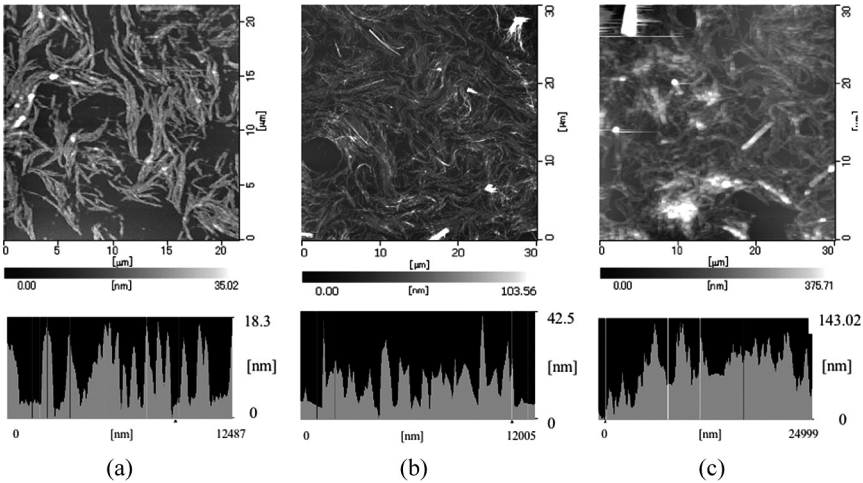

FIGURE 3 AFM image and cross section of the MWCNTs monolayer fabricated with addition of different alcohol. (a) with addition of 10 vol% of methanol; (b) with addition of 10 vol% of ethanol; (c) with addition of 10 vol% of 2-propanol.

Considering the carbon number, the hydrophobicity may follow the order of 2-propanol > ethanol > methanol. Therefore if we assume that the amount of alcohol adsorbed to the MWCNTs are irrespective to the alcohol type, the MWCNTs with methanol shows the highest hydrophilicity and the MWCNTs with 2-propanol shows the highest hydrophobicity in the three conditions. Due to the hydrophilic nature of the MWCNTs with methanol, most MWCNTs are dispersed in water, which results in a sparse film formation. On the other hand, the MWCNT with 2-propanol were aggregated together due to their hydrophobic nature. In a similar reason, the MWCNTs form an aggregated structure due to the hydrophobic nature of MWCNTs with 2-propanol.

CONCLUSION

Effect of the type of alcohol to assemble the MWCNTs to the liquid–liquid interface was studied. It was concluded that the alcohol hydrophobicity is effective to the film properties.

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