

## Effect of Plasma–Nitric Acid Treatment on the Electrical Conductivity of Flexible Transparent Conductive Films

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A flexible transparent electrically conductive film (FTCF) was formed on a poly(ethylene terephthalate) film by spraying single-walled carbon nanotubes dispersed with sodium dodecyl benzene sulfonate in water and, to improve the electrical conductivity of FTCF, the effect on plasma treatment followed by nitric acid treatment was investigated. The Ar plasma treatment was effective in dissociating the surfactant and removing impurities attached to the surface of the carbon nanotubes (CNTs). Therefore, through a cyclic treatment composed of an Ar plasma treatment and nitric acid treatment, more effective removal of surfactant and impurities attached to the spray-coated CNTs could be obtained than a cyclic treatment without the plasma treatment. With the optimized cyclic treatment, the sheet resistance ( $\Omega_s$ ) of the spray-coated CNTs could be decreased up to 45% by removing most of the surfactant and impurities. Using the repeated cyclic treatment, the FTCF having the  $\Omega_s$  of 160  $\Omega/\square$  at 81.5% optical transmittance at the wavelength of 550 nm could be obtained. © 2013 The Japan Society of Applied Physics

### 1. Introduction

Since the landmark paper on carbon nanotubes (CNTs) by Iijima in 1991, much research has been conducted to apply them for various applications due to their superior properties such as excellent electrical conductivity, high mechanical strength, high chemical stability, excellent thermal conductivity, large surface area, etc.<sup>1–5</sup> To use them as flexible transparent conductive materials, they must be dispersed in a solvent with a surfactant (or dispersant) and the dispersion needs to be coated on a flexible substrate. Various surfactants have been investigated to achieve highly nano-dispersed CNTs via dispersant. The challenge in the CNTs solubility is how to design the chemical structure of a surfactant to achieve improved nanodispersion of CNTs in an organic solvent. In general, as a surfactant, following conditions must be satisfied: 1) the head group of the surfactant should be strongly anchored to hydrophobic CNTs surface and 2) the number density of the stabilizing species (tails group) should be high enough to maintain a sufficient spatial volume to impart strongly repulsive forces to CNTs via steric hindrance.<sup>6</sup> Studies have showed that, among the various surfactants, sodium dodecyl sulfonate (SDS) and sodium dodecyl benzene sulfonate (SDBS) exhibited excellent characteristics as surfactant.<sup>7,8</sup> In the case of the solvent for CNTs dispersion, deionized (DI) water, ethanol, *N*-methyl-2-pyrrolidone (NMP), *N,N*-dimethylformamide (DMF), tetrahydrofuran (THF), and 1,2-dichloroethane (DCE) have been used to disperse CNTs.<sup>8–10</sup> For the coating methods of a dispersed CNTs solution, various methods such as filtration,<sup>11,12</sup> dip coating,<sup>13</sup> inkjet printing method,<sup>14</sup> spray coating,<sup>15,16</sup> etc. have been investigated. Among these methods, the filtration method has the advantage in removing surfactants and easy scale-up, the dip coating method has the advantage in the easy fabrication and the low cost of ownership, and the inkjet printing has the advantage in the micro-pattern formation. Especially, the spray coating method not only has the advantages of the dip coating method

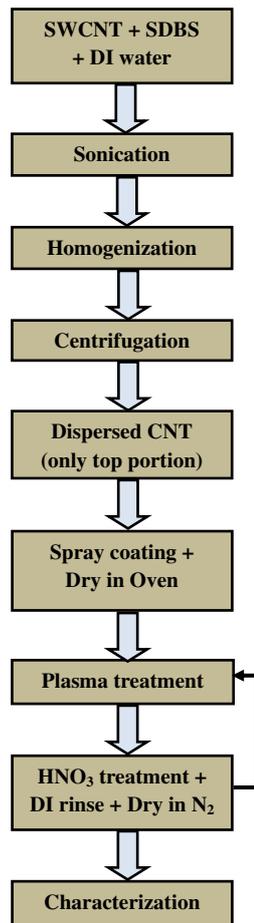
but also has excellent uniformity over a large substrate. Therefore, in this study, flexible transparent electrically conductive film (FTCF) was fabricated by using the spray coating method with the CNTs dispersed by SDBS in DI water.

The surfactants used to disperse CNTs and to uniformly coat on the substrate need to be removed because they are attached all around the CNTs and block the direct contact among CNTs, therefore, they tend to increase the sheet resistance ( $\Omega_s$ ) of the spray-coated CNTs. Much kind of acids are chosen for removing the surfactant on the spray-coated CNTs such as peroxytrifluoroacetic acid (PTFAA), hydrochloric acid (HCl), nitric acid (HNO<sub>3</sub>), sulfuric acid (H<sub>2</sub>SO<sub>4</sub>), and thionyl chloride (SOCl<sub>2</sub>).<sup>9,17,18</sup> Research showed that HNO<sub>3</sub> demonstrated the best effect in reducing the  $\Omega_s$  of FTCF by effectively removing the surfactant on CNTs.<sup>17</sup> However, the resulting  $\Omega_s$  of spray-coated CNTs changes with the treatment time of HNO<sub>3</sub> and, in general, the treatment time takes a long time in removing the surfactant effectively.

Plasma treatment has been used to remove the impurities on the surface of the various materials,<sup>19,20</sup> in addition to remove the impurities on the CNTs surface and to change the CNTs surface characteristics by modifying the surface chemistry of CNTs,<sup>21</sup> in addition to use for the growth of CNTs at a low temperature due to its gas dissociation characteristics by the plasma. In this study, as a new strategy for removing the surfactant and impurities on the spray-coated CNTs, a technique which includes both the plasma and acid treatment is investigated to decrease the  $\Omega_s$  by removing the surfactant and impurities. Compared to the  $\Omega_s$  of the FTCF without any treatment or that with HNO<sub>3</sub> treatment only, a cyclic treatment composed of argon (Ar) plasma treatment and HNO<sub>3</sub> treatment was very effective and showed the lower  $\Omega_s$ .

### 2. Experimental Methods

2.1 Preparation of flexible transparent conducting film Single-walled carbon nanotubes (SWCNTs); ASP-100F,



**Fig. 1.** (Color online) Fabrication processes of CNTs spray-coated PET for FTFC.

purity 60–70 wt %, (Ijin Nanotech.) synthesized using an arc discharge method was used as the CNTs material for FTFC. The SWCNTs contain approximately 10 wt % of catalyst metal and 20 wt % of graphite impurity. Also, it has the diameter of 1.3–1.5 nm and exists in bundles measuring almost 10 nm.

The fabrication of FTFC in Fig. 1 presents a schematic of CNTs dispersion strategy used in this study. It includes three steps: (i) the CNTs solution preparation, (ii) the CNTs treatment, and (iii) the film preparation. 10 mg SWCNTs was dispersed in 50 ml DI water added with 100 mg of SDBS (Sigma-Aldrich) as a surfactant and followed by the sonication in a bath-type sonicator (Bandelin Electronic) for 10 min to release the random CNTs entangled bundles. Because some unreleased CNTs bundles and impurities are still remaining after the sonication, a homogenizer is used for the next step in order to separate to individual metallic SWCNTs completely and followed by the centrifugation as a last step to remove the large size bundles and metal catalysts.<sup>10</sup> Therefore, the CNTs solution was homogenized (Ulss-Hi-Tech) for 12 min, then continuously centrifuged (Hanil Science Industrial) at 15000 rpm for 10 min. Finally, the purified CNTs solution was sprayed onto a poly(ethylene terephthalate) (PET) substrate ( $3 \times 3 \text{ cm}^2$ ) with an Ar gas brush pistol (Fuso Seiki) with the CNTs solution amount of 0.25–1.5 ml/sample in a spray coater. The PET substrate was kept on the substrate holder at the temperature of 110 °C.

## 2.2 Plasma and acid treatment

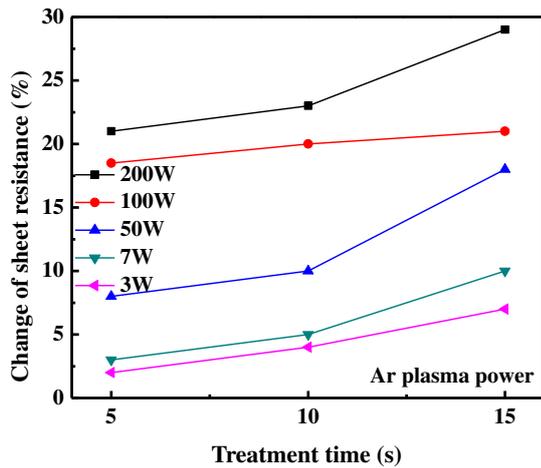
After the spray coating was terminated, the PET coated with CNTs was treated with an Ar plasma and  $\text{HNO}_3$ , repeatedly. The plasma treatment system was inductively coupled plasma (ICP)-type plasma system where 3–200 W of 13.56 MHz radio frequency (RF) power was applied to the top ICP source while the substrate was located on the bottom electrode. The distance between the dielectric window of the ICP source and the substrate was kept at about 5 cm. For all of the experiments, 170 mTorr Ar was used as the process gas condition and the substrate was kept at room temperature. For the acid treatment, the PET spray-coated with CNTs were immersed in  $\text{HNO}_3$  solution (OCI purity 70%) for 30 min to remove the SDBS surfactant. After the  $\text{HNO}_3$  treatment, the samples were rinsed in DI water and dried in oven during 10 min at 70 °C to neutralize until a hydrogen ion concentration (pH) is reached to 7 by removing the remaining nitric acid in FTFC. Such a cycle of both an Ar plasma and  $\text{HNO}_3$  treatment was carried out for a few times. A previous study showed no significant chemical reaction or oxidation of CNT was observed during the  $\text{HNO}_3$  immersion treatment.<sup>17</sup> Also, during the plasma treatment, the PET coated with CNT was physically bombarded by the  $\text{Ar}^+$  ions to dissociate the surfactant molecules without damaging the CNT. Therefore, through the cyclic  $\text{HNO}_3$  and Ar plasma treatment, we tried to increase the chemical reaction of surfactants only without increasing the chemical reaction of CNT.

## 2.3 Characterization

The optical properties of the FTFC after the treatment process were characterized by an ultraviolet–visible (UV–vis) absorption spectroscopy (Varian) in the visible range (400–800 nm). The  $\Omega_s$  of the samples were obtained using a four-point-probe method (Keithely). The surface morphology of the film was observed by field emission-scanning electron microscopy (FE-SEM; JEOL). Raman spectroscopy (Renishaw) with excitation energy of 514 nm ( $\text{Ar}^+$  ion laser) was used for characterizing the optical properties of the CNT films on PET substrates.

## 3. Results and Discussion

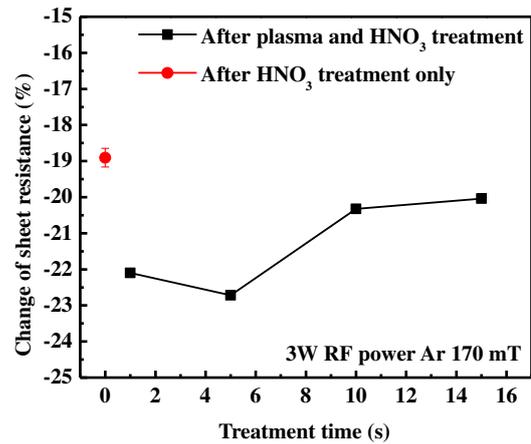
Plasma has been used to modify and clean the material surface including CNTs through the chemical reaction and physical ion bombardment. In this study, Ar plasma was applied to the spray-coated CNTs, and the effect of an Ar plasma treatment followed by  $\text{HNO}_3$  treatment on the change of  $\Omega_s$  has been investigated. Figure 2 shows the effect of plasma treatment on the change of  $\Omega_s$  of spray-coated CNTs for various plasma treatment time and RF power. The spray-coated CNTs were plasma-treated without rinsing after the spray coating. The RF power and plasma treatment time were varied from 3 to 200 W and from 5 to 15 s using Ar gas at the room temperature. The PET film was coated with 0.5 ml CNTs solution/sample. As shown in Fig. 2, the increase of treatment time from 5 to 15 s at a fixed RF power increased the  $\Omega_s$  in the range of 3–10% and the increase of RF power from 3 to 200 W at a fixed treatment time increased the  $\Omega_s$  in the range of 18–19%. Therefore, the increase of RF power from 3 to 200 W at a fixed treatment time increased the  $\Omega_s$  of the CNTs more significantly than the increase of treatment time from 5 to 15 s at a fixed RF



**Fig. 2.** (Color online) Effect of Ar plasma treatment on the change of  $\Omega_s$  of the spray-coated CNTs for various Ar plasma treatment time and RF power. The CNTs were plasma-treated without rinsing after the spray coating.

power. The Ar plasma generated by the RF power contains  $\text{Ar}^+$  ion formed by the ionization reaction ( $e^- + \text{Ar} \rightarrow \text{Ar}^+ + 2e^-$ ) in the plasma. These  $\text{Ar}^+$  ions in the plasma will bombard the spray-coated CNTs surface during the plasma treatment with the energy defined by the voltage difference between the Ar plasma and the PET substrate which is the sheath potential developed on the substrate. Generally, the plasma potentials increase with the increase of RF power to the plasma system in addition to the increase of  $\text{Ar}^+$  ion density, therefore, it is believed that the increased  $\Omega_s$  with the increase of RF power at a fixed plasma treatment time is related to the increased damage of the CNTs surface on the FTCF by the physical bombardment by the increased  $\text{Ar}^+$  bombardment energy and flux. Also, the increase of  $\Omega_s$  with the increase of plasma treatment time at the fixed RF power in the range from 3–200 W is also related to the increased surface damage of the CNTs during the plasma treatment. In fact, the plasma treatment was investigated to dissociate the surfactant molecules enclosing the CNTs-coated FTCF and to remove the broken surfactant molecules and impurities more easily during the following  $\text{HNO}_3$  treatment, however, it is believed that, by the plasma treatment, CNTs surface was physically damaged in addition to the dissociation of surfactant molecules on the CNTs surface.

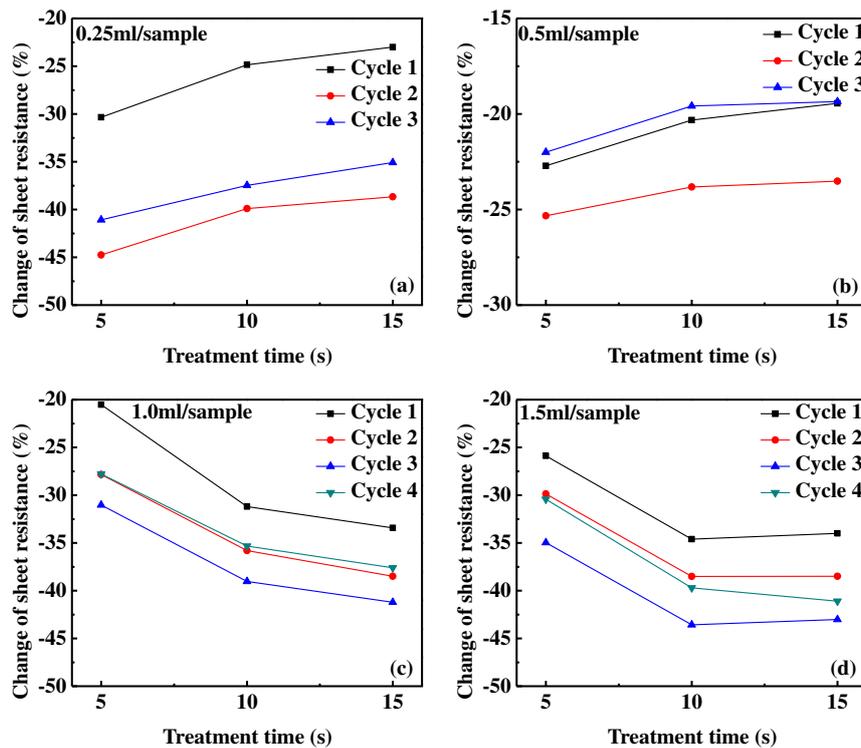
To dissociate the surfactant molecules on the CNTs surface without significantly damaging the CNTs surface, the RF power for Ar plasma treatment was reduced to 3 W and the spray-coated CNTs were treated with the Ar plasma and the plasma treatment time was varied from 1 to 15 s. After the plasma treatment, the CNTs were dipped in  $\text{HNO}_3$  for 30 min, rinsed in DI water and dried in  $\text{N}_2$ . Figure 3 shows the change of  $\Omega_s$  of the pristine spray-coated CNTs after both the plasma treatment and  $\text{HNO}_3$  treatment. For the spray-coated CNTs, 0.5 ml CNTs solution/sample was sprayed on the PET film. As a reference, the  $\Omega_s$  change of the spray-coated CNTs after the  $\text{HNO}_3$  treatment only without the plasma treatment, which is one of the widely used methods to remove the surfactant after the spray-coating of CNTs, was included. As shown in Fig. 3, the  $\text{HNO}_3$  treatment decreased the  $\Omega_s$  by showing the  $\Omega_s$  change



**Fig. 3.** (Color online) Change of  $\Omega_s$  of the pristine spray-coated CNTs after both the plasma treatment and  $\text{HNO}_3$  treatment. As a reference, the  $\Omega_s$  change of the pristine spray-coated CNTs after the  $\text{HNO}_3$  treatment only without the plasma treatment was included. The CNTs solution was 0.5 ml/sample and the Ar plasma operated at 3 W with 170 mTorr was used.

of the spray-coated CNTs about  $-19\%$  ( $-$  means the decrease of  $\Omega_s$ ) by removing the surfactant on the spray-coated CNTs surface. However, as shown in the Fig. 3, when the spray-coated CNTs was treated both by the Ar plasma and  $\text{HNO}_3$ , the further decrease of  $\Omega_s$  down to about  $-23\%$  could be obtained for 5 s plasma treatment time and about  $-20\%$  for 15 s plasma treatment time. Therefore, the combined plasma and  $\text{HNO}_3$  treatment process was more effective than the  $\text{HNO}_3$  treatment only. The improved property after the combined plasma and  $\text{HNO}_3$  treatment is believed to be related to the dissociation of the surfactant molecules during the plasma treatment followed by more effective removal of the broken surfactant and impurities on the CNTs surface. However, when the plasma treatment time was increased more than 5 s, the amount of the  $\Omega_s$  change was decreased with increasing the plasma treatment time possibly due to the increased damage of CNTs surface.

The spray-coated CNTs were treated by a number of cyclic treatments composed of the Ar plasma treatment and  $\text{HNO}_3$  treatment and the change of  $\Omega_s$  after the cyclic treatments was measured. For the Ar plasma treatment, the plasma generated at 3 W RF power with the pressure of 170 mTorr was used and the plasma treatment time was varied from 5 to 15 s. To investigate the effect of CNTs thickness coated on the substrate, the PET films spray-coated with 0.25–1.5 ml CNTs solution/sample were used as the samples. The change of  $\Omega_s$  after the cyclic treatment is shown in Fig. 4 as a function of the plasma treatment time and the number of cyclic treatments for the PET coated with (a) 0.25, (b) 0.5, (c) 1.0, and (d) 1.5 ml/sample of CNTs solution. As shown in Fig. 4(b), for 0.5 ml/sample, after the first cyclic treatment of first plasma treatment and first  $\text{HNO}_3$  treatment, compared to the  $\Omega_s$  of pristine spray-coated CNTs, the  $\Omega_s$  were decreased about 22.7% for 5 s plasma treatment to 19.4% for 15 s plasma treatment similar to those in Fig. 3. For the PETs coated with 0.25 or 0.5 ml/sample, the  $\Omega_s$  increased with the increase of Ar plasma treatment time because the bindings of the surfactants were sufficiently broken for 5 s of Ar plasma treatment time due to the thin thickness of CNT on the PET substrate and



**Fig. 4.** (Color online) Change of  $\Omega_s$  after the cyclic treatment composed of the Ar plasma treatment and  $\text{HNO}_3$  treatment is as a function of the plasma treatment time and the number of cyclic treatments for the PET coated with (a) 0.25, (b) 0.5, (c) 1.0, and (d) 1.5 ml/sample of CNTs solution. The Ar plasma operated at 3 W with 170 mTorr was used.

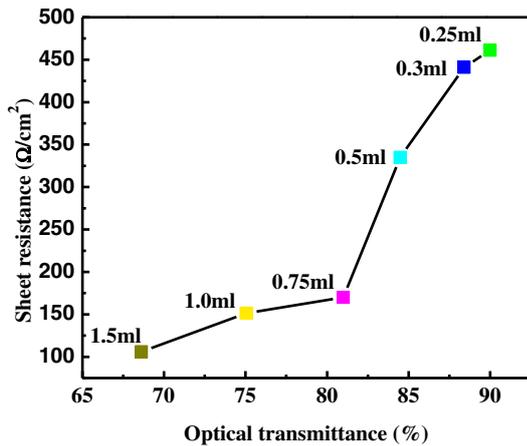
the further increase of Ar plasma treatment time increased the bombardment of CNT surfaces, and which appears to damage the CNT surface. After the second cyclic treatment, compared to pristine spray-coated CNTs, the  $\Omega_s$  of the CNTs was decreased to 25.3% (for 5 s plasma treatment)  $\sim$ 23.5% (for 15 s plasma treatment). Therefore, after the second cyclic treatment, compared to the first cyclic treated CNTs, the  $\Omega_s$  was further decreased up to 2.6% (for 5 s plasma treatment)  $\sim$ 4.1% (for 15 s plasma treatment). Therefore, the cyclic treatment composed of an Ar plasma treatment and  $\text{HNO}_3$  treatment is believed to be effective in removing the surfactant and impurities on the CNTs surface, therefore, by decreasing the  $\Omega_s$  of spray-coated CNTs. Figure 4(b) also shows the  $\Omega_s$  change after the third cyclic treatment and, as shown in Fig. 4(b), the  $\Omega_s$  was increased after the third cyclic treatment by showing the change of  $-22.0\%$  (for 5 s plasma treatment) and  $-19.3\%$  (for 15 s plasma treatment) compared to that of pristine CNTs. Therefore, by the third cyclic treatment, the CNTs surface appeared to be damaged more rather than removing the surfactant and impurities on the CNTs surface. Therefore, the optimum number of cyclic treatment and Ar plasma treatment time for the cyclic treatment for the PET coated with 0.5 ml/sample of CNTs solution were two cycles and 5 s of Ar plasma treatment.

The optimum number of cyclic treatment and the Ar plasma treatment time for the spray-coated CNTs can be different depending on the thickness of CNTs coated on the substrate that is, depending on the ml/sample of CNTs solution coated on PET. When the amount of CNTs solution per sample was 0.25 ml/sample, as shown in Fig. 4(a), the  $\Omega_s$  was decreased until the CNTs sample is treated by two cycles and the change of  $\Omega_s$  was decreased with the increase of Ar

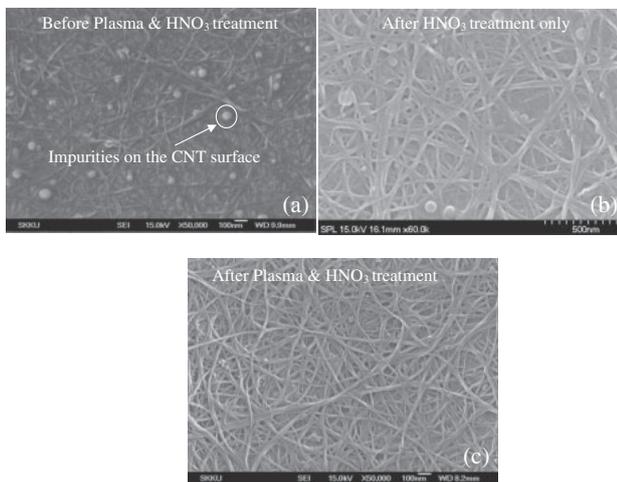
plasma treatment time, therefore, 5 s of Ar plasma treatment and second cyclic treatment showed the optimum result similar to the case of 0.5 ml/sample in Fig. 4(b). However, compared to the  $\Omega_s$  change for 0.5 ml/sample in Fig. 4(b), the  $\Omega_s$  change for 0.25 ml/sample in Fig. 4(a) was more significant by showing  $-44.8\%$  for the optimum condition of 5 s of Ar plasma treatment and second cyclic treatment. More significant change in the  $\Omega_s$  for 0.25 ml/sample appears to indicate more effective removal of surfactant on the CNTs surface due to thinner thickness of CNTs on the substrate.

However, when the CNTs solution coated on the substrate was higher as 1.0 and 1.5 ml/sample as shown in Figs. 4(c) and 4(d), respectively, the increase of Ar plasma treatment time during the cyclic treatment decreased the  $\Omega_s$  further and the  $\Omega_s$  was decreased until the CNTs were treated for three cycles possibly due to the insufficient CNTs surface treatment during the Ar plasma treatment in each cyclic treatment because of thick CNTs coating on the PET substrate. Therefore, the optimum condition for 1.0 and 1.5 ml/sample was third cyclic treatment with 15 s of Ar plasma treatment and the change of  $\Omega_s$  compared to the pristine spray-coated CNTs was about  $-41.2\%$  for 1.0 ml/sample and about  $-43.0\%$  for 1.5 ml/sample.

The increase of CNTs solution per sample increases the conductivity of the CNTs coated PET but it decreases the optical transmittance. The relationship between the optical transmittance of the CNTs coated PET film at the wavelength of 550 nm and the  $\Omega_s$  after the treatment with the optimized CNTs treatment conditions was investigated and the results are shown in Fig. 5. As shown in Fig. 5, the increase of CNTs solution decreased the optical transmittance while decreasing the  $\Omega_s$  due to the increased thickness of CNTs on the sub-



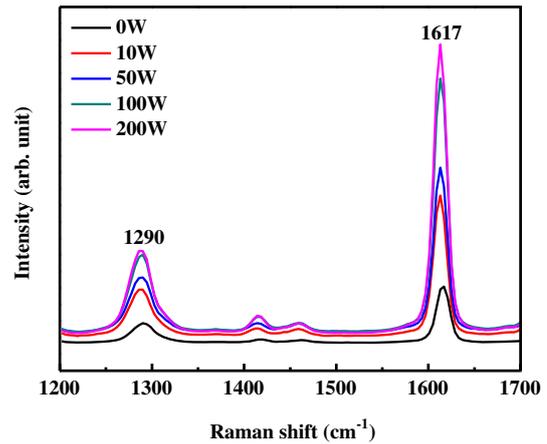
**Fig. 5.** (Color online) Relationship between the optical transmittance of the CNTs spray-coated PET film at the wavelength of 550 nm and the  $\Omega_s$  of the CNTs after the optimized cyclic treatments composed of an Ar plasma treatment and  $\text{HNO}_3$  treatment. The amount of CNTs solution per sample was varied from 0.25 to 1.5 ml/sample for the variation of optical transmittance.



**Fig. 6.** FE-SEM images of the spray-coated CNTs (a), the CNTs treated with only  $\text{HNO}_3$  treatment for two cycles (b), and the CNTs treated with the Ar plasma (5 s with 3 W at 170 mTorr) and  $\text{HNO}_3$  treatment for two cycles (c).

strate. In general, for the FTFCF, the optical transmittance needs to be higher than 85%. As shown in the Fig. 5, the optical transmittance close to 85% is obtained with the 0.5 ml/sample and, by the optimized cyclic treatment composed of two cycles with 5 s Ar plasma treatment (3 W and 170 mTorr), the  $\Omega_s$  of about 330  $\Omega/\square$  could be obtained.

The spray-coated CNT, the CNT treated with the  $\text{HNO}_3$  treatment only, and the CNT treated with the Ar plasma and  $\text{HNO}_3$  treatment were observed using FE-SEM, as shown in Figs. 6(a), 6(b), and 6(c), respectively. For Fig. 6(c), the CNT was treated for two cycles, and the Ar plasma treatment was done for 5 s with 3 W at 170 mTorr. It is known that the  $\Omega_s$  and optical transmittance of the CNT films strongly rely on the degree of bundles and entanglement. Large bundle size gives rise to higher adsorption of light. Consequently, the dispersion of the bundled CNT into individual CNTs and the increased entanglement of the CNT network are desired to increase the optical transmittance and



**Fig. 7.** (Color online) Raman spectra of CNT films before (black: 0 W) and after the plasma treatments with (red: 10 W), (blue: 50 W), (green: 100 W), and (pink: 200 W) for 30 s at 170 mTorr Ar.

network resistance.<sup>22)</sup> As shown in Fig. 6, most of the CNTs were remaining as bundles before and after the treatments, and no significant change in the dispersion and entanglement of CNT bundles could be observed after the treatments. However, the spray-coated CNT and the CNT treated with the  $\text{HNO}_3$  only contained impurity particles and were coated with some material which appeared to be related to the metal catalyst and SDBS surfactant. As shown in Fig. 6(c), after the second cyclic treatments of Ar plasma and  $\text{HNO}_3$ , the particles and the material enclosing the CNT bundles appeared to be removed almost completely. Therefore, the cyclic treatment is believed to be effective for removing surfactant and impurities on the spray-coated CNTs.

It is well known that Raman spectra are highly sensitive to the microstructure, especial for carbon materials.<sup>23,24)</sup> In the experiment, Raman spectra were carried out to further characterize the microstructure and defects of CNTs after the plasma treatment. Figure 7 exhibits the Raman spectra taken with 514 nm excitation for the samples before and after the plasma treatment of 10–200 W, at 170 mTorr for 30 s. The Raman spectra showed the G peaks at 1617  $\text{cm}^{-1}$  and D peaks at 1290  $\text{cm}^{-1}$ . Previous studies show that the D peak in the Raman spectra of CNTs originates from the defect which breaks the basic symmetry of the graphitic sheet thus leading to a lattice distortion, while the G peak corresponds to graphite or ordered carbon.<sup>25,26)</sup> The intensity ratio ( $I_D/I_G$ ) of the D peak ( $I_D$ ) to the G peak ( $I_G$ ) is an indication of the amount of disorder in the CNTs.<sup>17,27–30)</sup> As shown in the figure, the  $I_D/I_G$  ratios increased from 1.77 (no plasma treatment) to 1.89 (10 W), 1.97 (50 W), 2.32 (100 W), and 2.38 (200 W), indicating a continuous increase of the structural defect density with the increase of the plasma treatment power. Therefore, the higher treatment power induced the higher defect structural density.

#### 4. Conclusions

To improve the optical transmittance and conductivity of CNTs spray-coated on PET for FTFCF, the spray-coated CNTs were treated by a cyclic treatment composed of an Ar plasma treatment and  $\text{HNO}_3$  acid treatment. The  $\Omega_s$  of spray-coated CNTs was increased when the CNTs were treated with Ar plasma only with increasing RF power and

treatment time due to the damage of the CNTs surface. However, when the CNTs were treated with HNO<sub>3</sub> after the plasma treatment, the  $\Omega_s$  of the spray-coated CNTs was decreased further compared to the CNTs treated with HNO<sub>3</sub> only. The lower  $\Omega_s$  was related to the easier removal of the impurities and the surfactant enclosing the CNTs surface by dissociating surfactant with Ar<sup>+</sup> ion bombardment in the Ar plasma even though the Ar<sup>+</sup> ion bombardment can also damage the CNTs surface during the Ar plasma treatment. Repeated cyclic treatment of the spray-coated CNTs composed of an Ar plasma treatment and HNO<sub>3</sub> treatment decreased the  $\Omega_s$  further and the optimum number of cyclic treatment and its Ar plasma treatment time for the lowest  $\Omega_s$  of the CNTs was dependent on the thickness of CNTs coated on PET. The PET coated with thicker CNTs needed more cyclic treatment and longer Ar plasma treatment time while the PET coated with thinner CNTs needed less cyclic treatment and shorter Ar plasma treatment time. For the FTCF, the optical transmittance of CNTs coated PET needs to be higher than 85% while maintaining the  $\Omega_s$  as low as possible. In our experiment, by using 2 cyclic treatments with the plasma treatment condition of 5 s at 3 W and Ar 170 mTorr, the CNTs coated PET having of the  $\Omega_s$  of 330  $\Omega/\square$  at 85% optical transmittance (or 160  $\Omega/\square$  at 81.5%) at 550 nm could be obtained. It is believed that, if the SWCNTs with higher quality (purity, >90%) and the spray-coated CNTs are doped with dopants such as AuCl<sub>3</sub>, trifluoromethanesulfonimide, etc. after the optimized cyclic treatment, the further decrease in the  $\Omega_s$  can be expected while maintaining a high optical transmittance.

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