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Contact resistance of flexible, transparent carbon nanotube films with metals

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We studied the contact properties of different metals to optically-transparent single-walled carbon nanotube (SWCNT) films using fabrication processes compatible with flexible electronic applications. The SWCNT films are deposited on flexible polyethylene terephthalate substrate and patterned in test structures optimized for contact resistance measurements for a particular metal contact. Specific contact resistance and current transfer length is determined for Pt, Cr, Cu, and Au contacts. We also evaluate effects of chemical doping and thickness of SWCNT films on the contact resistance. We find that the current transfer length, defining the minimal dimensions of efficient contacts to transparent SWCNT films, ranges from 8×10^{-6} to 10^{-4} m for different metals. © *2010 American Institute of Physics*. [doi:10.1063/1.3496465]

Many applications of flexible electronics devices such as displays, solar cells, and touch screens require transparent and flexible highly-conducting electrodes. Optically transparent and electrical conductive thin films of randomly distributed carbon nanotubes have been broadly investigated as an alternative to commonly used conductive oxides.[1–7] Single-wall carbon nanotube (SWCNT) networks demonstrate sheet resistances and optical transparencies comparable to indium tin oxide films on plastic substrates and, in addition, display better mechanical stabilities.[1–7] Characterization of contact interfaces between common metals and SWCNT films is essential for many applications. For example, to improve efficiency of solar cells, SWCNT films are often combined with metallic grids.[8] The contact resistance at the interface of the SWCNT films and metals contributes an additional resistive loss and directly affects the efficiency of the device. The trade-off between the contact area to improve the conducting properties and the resulting loss of the optical transparency has to be carefully considered. While there have been multiple investigations of transport properties of SWCNT films, sparse data are available addressing the contact resistance between metals and SWCNT films. Most of the previous research has focused on contacts to individual nanotubes.[9–13]

Quantitative characterization of the contact interfaces is an interesting and challenging problem[14–17] that can be affected by multiple parameters. Recent reports list widely different specific contact resistances between metal interfaces with nominally comparable SWCNT films, ranging from 2×10^{-6} Ω m^2 for Ag (Ref. 8) to 3×10^{-9} Ω m^2 for Au (Ref. 16) and down to 1×10^{-10} Ω m^2 for Pt.[17] Besides the chemical composition at the contact interface, the contact resistance can be affected by metal-specific three-dimensional nanoscale morphology, particular processing conditions, or substrate material.

The goal of this paper is to compare the properties of contact interfaces between SWCNT films and typical metals fabricated under similar processing conditions relevant to flexible electronic applications. We developed a multilayer fabrication process for patterning flexible and transparent SWCNT films on polyethylene terephthalate (PET) substrates, and measured the contact resistances in well-defined geometries. A common and reliable technique, the transfer length method,[18,19] was used to determine the specific contact resistance ρ_c. We also examined the variability of the contact resistances resulting from the changes in SWCNT sheet resistance and doping level. The specific contact resistances and the current transfer lengths in this paper provide essential guidance for designing, fabricating, and optimizing flexible electronic devices based on SWCNT films.

The SWCNT films on flexible PET substrate were fabricated by the spraying method using ink made from 0.8 mg/ml laser ablation SWCNTs suspended in water with 1% sodium dodecylbenzene sulfonate surfactant.[20] Then the films were carefully rinsed in water and dried. The SWCNT film used for the measurement has a sheet resistance of 650 Ω with high uniformity (measured resistance in 1×1 in.^{2} area with standard deviation less than 3%) and 92% transmittance at 550 nm, which is typical for transparent electrodes. To prepare the test structure, we developed a multilayer optical lithography process to pattern the SWCNT films. First, S1813 on LOR3A (Ref. 21) dual layer resists were spin coated on the SWCNT-PET film. Then the resist was exposed by UV light through a photomask in a mask aligner. MF-319 was used as the developer to wash away the resists at the exposed area. An argon plasma was used to remove the exposed SWCNT film, creating the patterned SWCNT strips on the substrate. Any residual resist was cleaned by REMOVER PG. A similar photolithography procedure was performed to define the metal contacts over the patterned strips. In Figs. 1(b) and 1(c), we show the interface between nanotube films and metal contact layers, which illustrates good penetration of metal into SWCNT film. The distance between the contact leads S range from 10 μm to 2 mm, the strip width L and contact lead width w range from 10 to 200 μm. For a particular metal contact-film combination, a set of contacts was chosen to minimize the measurement error.

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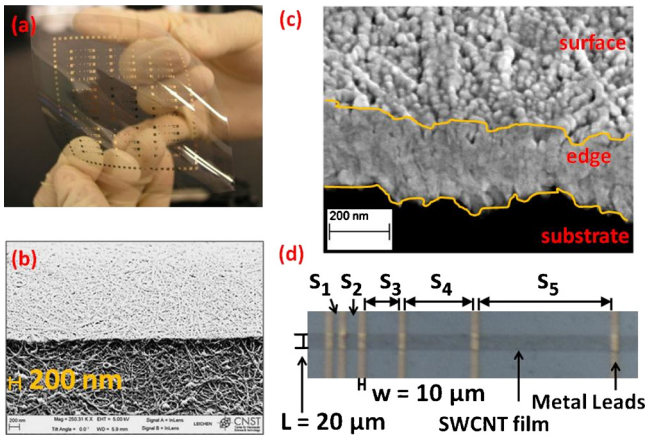

FIG. 1. (Color online) (a) Picture of the patterned sample on PET substrate. (flexible and transparent). (b) SEM image of the interface between nanotube film and metal contact layer. (c) Cross-section SEM images of interface of metal and SWNCT film. Cross-sectioning is performed using a microtome. Metal morphology is the same across the section indicating good metal penetration and the contact formation throughout the SWCNT film. (d) Schematic and optical image of the patterned strip and metal leads.

Figure 2(a) shows a plot of measured 2-point resistance between metal leads at different spacing. The 2-point resistance R_T can be approximately written as follows:

$$R_T = 2R_c + R_{sh}S_i/L + R_0, \tag{1}$$

where R_c is the contact resistance between the metal leads and SWCNT film, $R_{sh}S_i/L$ is the resistance of the SWCNT film strip, and R_0 is the serial resistance of the measurement cables, the metal leads, and the contacts between cables and the metal leads (10 Ω).

The straight lines in Fig. 2(a) are the least square fits to R_T. The intersections with the vertical axis give $2R_c$ and the standard deviations, $(1.39 \pm 0.10) \times 10^4$ Ω and $(6.60 \pm 1.0) \times 10^3$ Ω for 10 μm wide leads with $L = 100$ μm and 200 μm, respectively, as illustrated in the figure. We also measured the current-voltage characteristics of the devices verifying that the contact between the metal contacts and SWCNT films is Ohmic up to 1 V.

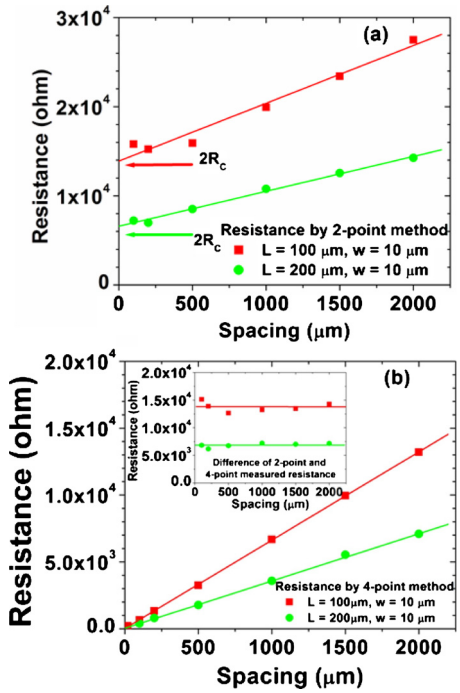

FIG. 2. (Color online) Measured resistance at different spacings for copper leads. (a) 2-point resistance as a function of lead spacing. Contact resistance R_c is determined from intersect with y-axis. (b) Resistance measured by 4-point method vs lead spacing. The inset shows the difference, $2R_c$, of 2-point and 4-point resistances.

TABLE I. Specific contact resistance and transfer lengths of SWCNT films with various metal contacts.

	Pt	Cr	Cu	Au
ρ_c (Ω m^2)	$5.2 \pm 1.0 \times 10^{-8}$	$2.0 \pm 0.5 \times 10^{-6}$	$6.7 \pm 2.0 \times 10^{-6}$	$5.0 \pm 2.3 \times 10^{-6}$
L_T (m)	9.1×10^{-6}	5.5×10^{-5}	1.0×10^{-4}	8.8×10^{-5}

The difference between the 2-point resistance and 4-point resistance provides another estimate of the contact resistance, shown in the inset to Fig. 2(b). The averages of the differences between 2-point and 4-point measurements at different spacings, giving $2R_c$, $(1.38 \pm 0.07) \times 10^4$ Ω and $(6.60 \pm 0.30) \times 10^3$ Ω, respectively, are consistent with the values determined from the 2-point measurements.

The specific contact resistance, ρ_c, independent of contact area is an important parameter usually used to evaluate the quality of electrical contacts. For a transmission line model structure, the contact resistance R_c and the specific contact resistance ρ_c has the following relations:[18,19]

$$R_c L/(R_{sh} \times \rho_c)^{1/2} = \coth[w \times (R_{sh}/\rho_c)^{1/2}],$$
$$\text{or} \quad R_c L/(R_{sh} \times L_T) = \coth(w/L_T), \tag{2}$$

where the characteristic transfer length $L_T = (\rho_c/R_{sh})^{1/2}$ defines the length over which 1/e of the current is transferred from metal contact to the film. Equation (2) can be simplified as $R_c = (\rho_c \times R_{sh})^{1/2}/L = R_{sh} \times L_T/L$, for $w \gg L_T$ (long contact limit) or $R_c = \rho_c/(Lw)$, for $w \ll L_T$ (short contact limit). From Eq. (2), the specific contact resistance $\rho_c = 6.7 \times 10^{-6}$ Ω m^2 is calculated.

In addition to Cu, the specific contact resistance between SWCNT films and other metals commonly used in electrical devices was studied, including Pt, Cr, and Au. The results are shown in Table I. We carefully checked the interface of the SWCNT films with these metals and found all the deposited metals penetrate into the nanotube films well. Figure 1(c) is a scanning electron microscopy (SEM) cross-section images of nanotube films with gold deposited on top, which penetrated well to the nanotube films. Similar behavior has also been found for other metals. This attributed to highly porous structure of nanotube films with holes around tens of nanometer to hundreds of nanometer, which are larger than the typical grain size of the deposited metals. We also attempted to determine the contact resistance of SWCNT films with Al and Ti; however, the resistance was too high to measure. Pt has the lowest contact resistance to SWCNT films, followed by Cr. Au and Cu display relatively high contact resistance. While the details of contact interface formation can be different for single CNT devices and CNT films, the sequence of resistances is consistent with previous studies of contact resistances to individual nanotubes.[9–13] Calculated L_T are also listed in Table I. For good electric contact, the lead width should be larger than L_T. We note that the range of specific contact resistances determined for different metals is much narrower than that reported in the literature for these metals.[8,16,17]

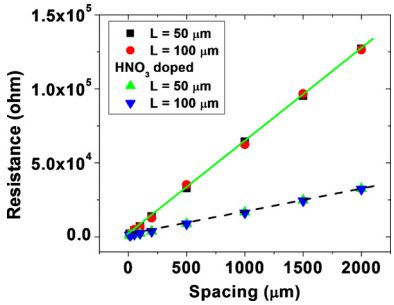

FIG. 3. (Color online) Influence of the doping process on the contact resistance. The resistance was measured by 2-point method for doped and undoped SWCNT films with Pt leads.

The contact resistance can be affected by various treatment process, CNT morphology, and compositions.[22] To evaluate this effect we used chemical doping and compared films of different thickness. Chemical doping is an effective method to improve the conductivity of SWCNT films without sacrificing optical transparency.[23,24] The SWCNT films were doped using concentrated nitric acid (4 mol/l HNO_3). Figure 3 shows the comparison of contact resistance measurements of SWCNT films with Pt leads with and without chemical doping. In both cases, the determined transfer length, L_T, is less than half of the contact pads width ($w > 2L_T$). In this situation, the contact resistance does not depend on the lead width [see Eq. (2)], which is clearly seen in Fig. 3. The results are summarized in Table II. We found that while the contact resistance decreases, the specific contact resistance and the transfer length actually increase for the doped film as a result of significant drop in the sheet resistance [see Eq. (2)]. This observation is consistent with the previous work on Ag contacts and the proposed microscopic explanation suggested in Ref. 8. The decrease in the sheet resistance is attributed to the doping and the improved conductivity of semiconductor CNTs within the film, while the conductance of metal-semiconductor CNTs contacts is less affected by the doping.

Although thicker SWCNT films can be also used to decrease the sheet resistance the optical transmittance becomes adversely affected for thick films. We selected a film with 250 Ω sheet resistance and 85% optical transmittance at 550 nm to evaluate the effect of the film thickness on the contact resistance. The results are listed in Table II. The thicker film has smaller contact resistance, and the specific contact resistance roughly scales with the sheet resistance of the sample. This scaling is likely caused by the highly porous structure of the carbon nanotube films and the penetration of the evaporated metal deep into the film. Note that such penetration can be seen in Figs. 1(b) and 1(c). In this case, the effective contact area is determined by a three-dimensional surface of the metal penetrating through the film that scales with the film thickness rather than the geometrical surface area of the contact.

TABLE II. Comparison of contact resistance of undoped and doped SWCNT films with Pt leads.

	R_{sh} (Ω)	Transmittance @550 nm (%)	R_c (Ω)	ρ_c (Ω m^2)	L_T (m)
Thin film (undoped)	630	92	575 ± 50	$(5.2 \pm 1.0) \times 10^{-8}$	9.1×10^{-6}
Thin film (doped)	160	92	385 ± 50	$(9.3 \pm 2.0) \times 10^{-8}$	2.4×10^{-5}
Thick film (undoped)	250	85	228 ± 25	$(1.5 \pm 0.2) \times 10^{-8}$	7.7×10^{-6}

In summary, the contact interfaces of different metals with conductive optically-transparent SWCNT films on flexible substrate were studied and the specific contact resistance and the current transfer lengths were determined. The variation in contact resistance caused by doping and thickness of the SWCNT films has also been evaluated. Pt is found to have the lowest contact resistance with SWCNT films while Au and Cu form relatively high resistance interfaces. The range of contact resistances is narrower than reported in the literature[8,16,17] indicating that specific contact morphology, processing condition and film substrate can be as significant as the chemical composition of the contacts. We have determined that, for the processing conditions relevant for transparent flexible electronic applications, the current transfer length ranges from 8×10^{-6} to 10^{-4} m, thus providing the guidance for the minimal dimensions of efficient contacts to transparent SWCNT films.

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