Supplementary Information

Large Work Function Difference Driven Electron Transfer from Electrides to Single-walled Carbon Nanotubes

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<table>
<thead>
<tr>
<th>Acronym</th>
<th>Description</th>
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<tbody>
<tr>
<td>HSWNT</td>
<td>HiPco SWNT (Diameter: 0.7-1.2 nm, Length: 400-700 nm)\textsuperscript{S1}</td>
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<tr>
<td>PSWNT</td>
<td>Purified plasma SWNT (Diameter: 1.2-1.7 nm, Length: 1-2 µm)\textsuperscript{S2}</td>
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<tr>
<td>C12A7:e\textsuperscript{-}</td>
<td>([\text{Ca}<em>{24}\text{Al}</em>{28}\text{O}_{64}]^{4+}\cdot4\text{e}^-)</td>
</tr>
<tr>
<td>Ca2N:e\textsuperscript{-}</td>
<td>([\text{Ca}_5\text{N}^+]\cdot\text{e}^-)</td>
</tr>
<tr>
<td>C12A7:e\textsuperscript{-}-HSWNT film</td>
<td>([\text{Ca}<em>{24}\text{Al}</em>{28}\text{O}_{64}]^{4+}\cdot4\text{e}^-)-HiPco SWNT-PVDF-HFP film</td>
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<td>C12A7:e\textsuperscript{-}-PSWNT-Ag paste</td>
<td>([\text{Ca}<em>{24}\text{Al}</em>{28}\text{O}_{64}]^{4+}\cdot4\text{e}^-)-Plasma purified SWNT-Ag-Epoxy paste</td>
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**Table S1.** Acronyms of base materials and synthesized specimens
Figure S1. Field emission characteristics of the PSWNT-Ag paste and HSWNT-Ag paste. The nanotubes were mixed with Ag pastes without electride particles. The nanotube, silver, and epoxy concentrations were 2.5, 80, and 17.5 wt.%. The field emission current density of PSWNT-Ag paste was 0.48 mA/cm$^2$ at 7.3 V/μm with a turn on voltage of 4.14 V/μm. The field emission current density of HSWNT-Ag paste was 0.19 mA/cm$^2$ at 7.34 V/μm with a turn on voltage of 4.2 V/μm.
**Figure S2.** Wide scan XPS data of pure HSWNTs, pure C12A7:e⁻ powder, and C12A7:e⁻-HSWNT powder mixture (50:50 wt.%).
Figure S3. The G-mode of Raman spectra measured at 6 different locations in pure HSWNTs, C12A7:e−-HSWNT, and Ca2N:e−-HSWNT powder mixtures.
Estimation of transferred electrons from electrides to nanotubes

The fitting to the calculation of density functional theory correlates the G-mode shift of nanotubes (\(\Delta \omega \text{ (cm}^{-1}\)) with the number of transferred electrons per carbon atom (\(f_c\)).\(^{S3}\)

\[ \Delta \omega = \Delta \omega_s + \Delta \omega_d \]  \hspace{1cm} (S1)

\[ \Delta \omega = 350 f_c + 101 \sqrt{f_c} \]  \hspace{1cm} (S2)

\[ f_c^* = (f_c \times N_{C-atoms})/L_{Tube} \]  \hspace{1cm} (S3)

where \(\Delta \omega_s\) is the Raman shift due to the lattice contraction (strain), \(\Delta \omega_d\) is the nonadiabatic effects (dynamical) due to electron-phonon coupling, \(f_c^*\) is the total number of electrons transferred per unit length of a nanotube (#/cm\(^{-1}\)), \(N_{C-atoms}\) is the total number of carbon atoms in a nanotube, and \(L_{Tube}\) is the length of a nanotube.

The following equations were used to calculate \(N_{C-atoms}\)\(^{S4}\)

\[ N_{C-atoms} = (M_{Tube} \times N_A)/M_{carbon} \]  \hspace{1cm} (S4)

\[ M_{Tube} = (\pi L_{Tube} \times D_{Tube})/1315 \text{ m}^2\text{g}^{-1} \]  \hspace{1cm} (S5)

where \(N_A\) is Avogadro's number (6.02\(\times\)10\(^{23}\) mole\(^{-1}\)), \(M_{carbon}\) is the molar mass of carbon (12.011 g mole\(^{-1}\)), \(M_{Tube}\) is the mass of a single nanotube, and \(D_{Tube}\) is the diameter of a nanotube.

The average length and diameter of HSWNTs were used for the calculation in this study \((L_{Tube} = 600 \text{ nm, } D_{Tube} = 1.03 \text{ nm})^{S1,S5}\) The resulting \(M_{Tube}\) was 1.48\(\times\)10\(^{-18}\) g and \(N_{C-atoms}\) was 7.4\(\times\)10\(^4\). The experimentally observed G-mode shifts of nanotubes were 1.8 and 2.7 cm\(^{-1}\) for C12A7:e\(^-\) and Ca2N:e\(^-\), respectively. The calculated \(f_c\) values were 2.8\(\times\)10\(^{-4}\) and 6.1\(\times\)10\(^{-4}\) for...
C12A7:e− and Ca2N:e−, respectively. The corresponding $f_{c}^*$ were $3.5 \times 10^5$ cm$^{-1}$ and $7.48 \times 10^5$ cm$^{-1}$ for C12A7:e− and Ca2N:e−, respectively.
**Figure S4.** Raman spectra of pure HSWNTs and electride-HSWNT powder mixtures. The time after exposure of the powder mixtures to air is shown in parenthesis. The G-mode shift is magnified in the inset. a) C12A7:e\textsuperscript{−}-HSWNT powder mixture. b) Ca2N:e\textsuperscript{−}-HSWNT powder mixture.
**Figure S5.** Optical images of C12A7:e−-HSWNT films. The letters SKKU are placed behind the films. The transmittance decreased from 84.8 % to 30 % at 550 nm with the addition of 0 to 2.5 wt.% C12A7:e− particles.
Figure S6. XPS and SEM analysis. a,b) Wide scan and C1s XPS data of pure PSWNTs, pure C12A7:e\(^{-}\) powder, and C12A7:e\(^{-}\)-PSWNT powder mixture (50:50 wt.%). The inset compares the shift of binding energies with the addition of C12A7:e\(^{-}\) to PSWNTs. The peaks of graphitic structure of PSWNTs (C=C bonds @284.6 eV), defect sites (sp\(^3\) carbon atoms), and surface functional groups (C=O, COOH, and carbonates) were investigated. c) SEM image of C12A7:e\(^{-}\)-PSWNT powder mixture.
Figure S7. Cross-sectional SEM image of the tape-activated PSWNT-Ag paste. The surface was activated 10 times using Scotch tapes.\textsuperscript{56} The yellow arrows indicate nanotubes.
Figure S8. Comparison of the turn on voltage ($V_{to}$), maximum current density ($I_{max}$), and the electric field at maximum current density ($V_{max}$). a) C12A7:e$^-$-PSWNT-Ag pastes. b) Ca2N:e$^-$-PSWNT-Ag pastes.
REFERENCES


