# **Supplementary Information**

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

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Acronym	Description
HSWNT	HiPco SWNT (Diameter: 0.7-1.2 nm, Length: 400-700 nm) <sup>S1</sup>
PSWNT	Purified plasma SWNT (Diameter: 1.2-1.7 nm, Length: 1-2 $\mu m)^{S2}$
C12A7:e <sup>-</sup>	$[Ca_{24}Al_{28}O_{64}]^{4+}\cdot 4e^{-}$
Ca2N:e <sup>-</sup>	$[Ca_2N]^+ \cdot e^-$
C12A7:e⁻-HSWNT film	[Ca <sub>24</sub> Al <sub>28</sub> O <sub>64</sub> ] <sup>4+.</sup> 4e <sup>-</sup> -HiPco SWNT-PVDF-HFP film
Ca2N:e⁻-HSWNT film	[Ca <sub>2</sub> N] <sup>+</sup> ·e <sup>-</sup> -HiPco SWNT-PVDF-HFP film
C12A7:e <sup>-</sup> -PSWNT-Ag paste	[Ca <sub>24</sub> Al <sub>28</sub> O <sub>64</sub> ] <sup>4+.</sup> 4e <sup>-</sup> -Plasma purified SWNT-Ag-Epoxy paste
Ca2N:e <sup>-</sup> -PSWNT-Ag paste	[Ca <sub>2</sub> N] <sup>+</sup> ·e <sup>-</sup> -Plasma purified SWNT-Ag-Epoxy paste

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<sup>2</sup> at 7.3 V/ $\mu$ m with a turn on voltage of 4.14 V/ $\mu$ m. The field emission current density of HSWNT-Ag paste was 0.19 mA/cm<sup>2</sup> at 7.34 V/ $\mu$ m with a turn on voltage of 4.2 V/ $\mu$ m.



**Figure S2.** Wide scan XPS data of pure HSWNTs, pure C12A7:e<sup>-</sup> powder, and C12A7:e<sup>-</sup> HSWNT powder mixture (50:50 wt.%).



**Figure S3.** The G-mode of Raman spectra measured at 6 different locations in pure HSWNTs, C12A7:e<sup>-</sup>-HSWNT, and Ca2N:e<sup>-</sup>-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 ( $\Delta \omega$  (cm<sup>-1</sup>)) with the number of transferred electrons per carbon atom ( $f_c$ ).<sup>S3</sup>

$$\Delta \omega = \Delta \omega_s + \Delta \omega_d \tag{S1}$$

$$\Delta\omega = 350f_c + 101\sqrt{f_c} \tag{S2}$$

$$f_c^* = (f_c \times N_{C\text{-}atoms}) / L_{Tube}$$
(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<sup>-1</sup>),  $N_{C-atoms}$  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  $N_{C-atoms}$ <sup>S4</sup>

$$N_{C-atoms} = (M_{Tube} \times N_A) / M_{carbon}$$
(S4)

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

where  $N_A$  is Avogadro's number (6.02×10<sup>23</sup> mole<sup>-1</sup>),  $M_{carbon}$  is the molar mass of carbon (12.011 g mole<sup>-1</sup>),  $M_{Tube}$  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<sup>-1</sup> for C12A7:e<sup>-</sup> and Ca2N:e<sup>-</sup>, respectively. The calculated  $f_c$  values were  $2.8 \times 10^{-4}$  and  $6.1 \times 10^{-4}$  for

C12A7:e<sup>-</sup> and Ca2N:e<sup>-</sup>, respectively. The corresponding  $f_c^*$  were  $3.5 \times 10^5$  cm<sup>-1</sup> and  $7.48 \times 10^5$  cm<sup>-1</sup> for C12A7:e<sup>-</sup> and Ca2N:e<sup>-</sup>, 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<sup>-</sup>-HSWNT powder mixture. b) Ca2N:e<sup>-</sup>-HSWNT powder mixture.



**Figure S5.** Optical images of C12A7:e<sup>-</sup>-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<sup>-</sup> particles.



**Figure S6.** XPS and SEM analysis. a,b) Wide scan and C1s XPS data of pure PSWNTs, pure C12A7:e<sup>-</sup> powder, and C12A7:e<sup>-</sup>-PSWNT powder mixture (50:50 wt.%). The inset compares the shift of binding energies with the addition of C12A7:e<sup>-</sup> to PSWNTs. The peaks of graphitic structure of PSWNTs (C=C bonds @284.6 eV), defect sites (sp<sup>3</sup> carbon atoms), and surface functional groups (C=O, COOH, and carbonates) were investigated. c) SEM image of C12A7:e<sup>-</sup> 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.<sup>S6</sup> 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<sup>-</sup>-PSWNT-Ag pastes. b) Ca2N:e<sup>-</sup>-PSWNT-Ag pastes.

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