

Figure S.1.

X-ray diffraction profiles of high-purity metallic SWCNTs (Metal, red line) and SWCNTs before metal-semiconductor (MS-) separation (Bf, blue line). Metal sample was prepared after MS-separation/purification/annealing processes described in the text. Dotted lines are simulation results using a model described in a previous paper (M. Abe et al., *Phys. Rev. B* **68**, 041405-041408, 2003). As the results of analyses, average diameters of nanotubes in Metal and Bf were estimated to be 1.46 ± 0.05 nm, and 1.46 ± 0.08 nm, respectively. Average diameter of nanotube-bundles in Metal and Bf were estimated to be 11.4 nm and 12.5 nm, respectively.

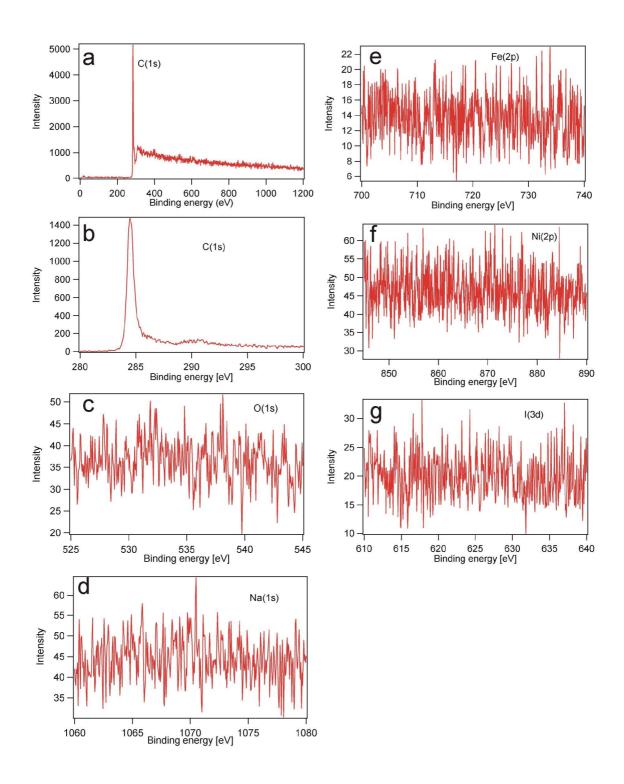


Figure S.2.

Typical X-ray photoelectron spectroscopy (XPS) spectra of sheets of SWCNTs used in this study. The sheet was prepared after metal-semiconductor-separation/purification/annealing processes described in the text. XPS spectra were recorded using a Thermo Fisher Scientific K.K., Theta Probe XPS system with a monochromatic Al-K α X-ray source (1486.6 eV). The base pressure of the XPS system was less than 1.3 x10⁻⁷ Pa. Panel (a) shows the whole spectra of the sample, and (b)-(g) panels focus on the regions in which signals from residual metals would be observed. We could not detect any significant signals from residual metals. The amount of residual metals was too small to be correctly evaluated and estimated to be less than 1 %.

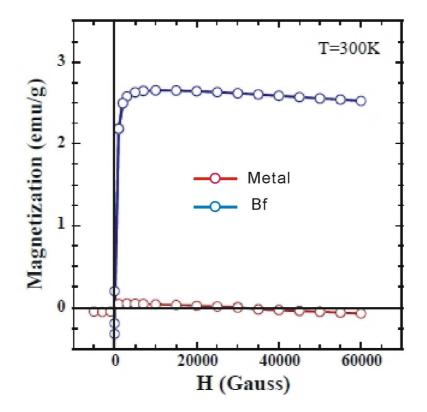


Figure S.3.

Magnetic characteristics of high-purity metallic SWCNTs (Metal, red) and SWCNTs before metal-semiconductor (MS-) separation (Bf, blue). Metal sample was prepared after MS-separation/purification/annealing processes described in the text. In Bf samples (no purification treatments), presence of ferromagnetic components, which would be residual Ni catalyst, was clearly detected. However, in Metal sample, there was no significant contribution of such components. When we assume that the ferromagnetic components were caused by Ni catalyst, the amount of the ferromagnetic components was estimated to be 2000 ppma in Bf, but 30 ppma in Metal.

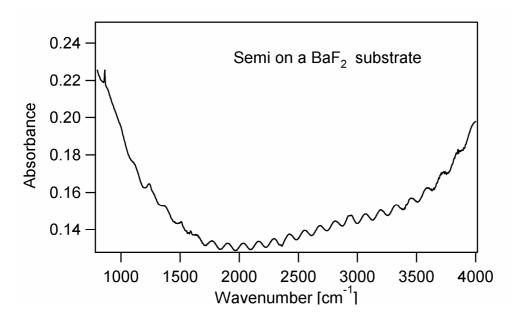


Figure S.4.

FT-IR spectrum of a thin film of a high-purity semiconducting SWCNT sample (Semi) on a BaF_2 substrate. The spectrum was obtained using FT-IR-6100 (JASCO). Modulated signals were caused by interference of incident laser light on the film.

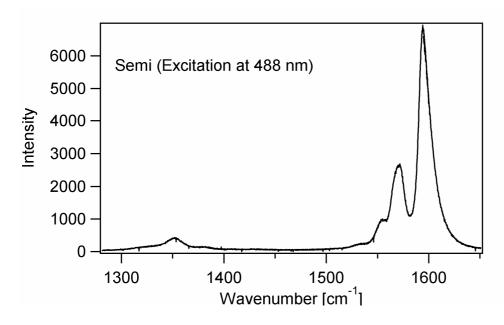


Figure S.5.

Raman spectrum of a high-purity semiconducting SWCNT sample (Semi) at excitation of 488 nm, which was obtained using an Ar^+ ion laser (Stabilite 2017, Spectra Physics Co) and a double monochromator system (Jobin Yvon U1000). The G/D ratio of the sample was estimated to be 20.

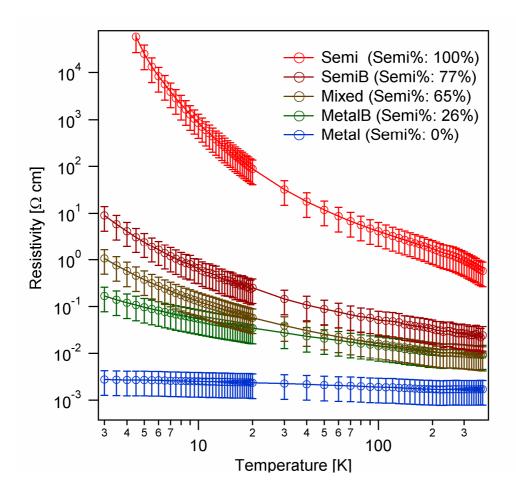


Figure S.6.

Resistivity of the sheets of five SWCNT samples with different MS ratio, Semi, SemiB, Mixed, MetalB, and Metal.

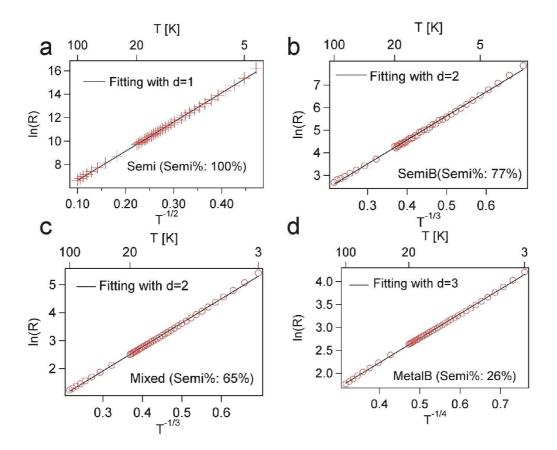


Figure S.7.

The results of analyses of temperature dependence of resistance of four SWCNT samples with different MS content ratio; Semi, SemiB, Mixed, and MetalB, using VRH model,

$$R(T) = R_0 \exp\left[-\left(\frac{T_0}{T}\right)^{\frac{1}{d+1}}\right].$$
 Here *d* is 1, 2, or 3. $\ln(R)$ data are plotted as a function of

 $T^{-1/(d+1)}$. Linear fitting results are shown in solid lines for guide. (a) Semi was well reproduced by VRH with d = 1, (b) SemiB was by VRH with d = 2, (c) Mixed was by VRH with d = 2, (d) MetalB was by VRH with d = 3.

Details of procedures to prepare high-purity metallic and semiconducting SWCNTs.

We prepared the SWCNT samples through the following two schemes.

Scheme 1

1. 100 mg of SWCNTs (Arc-SO type, Meijo-Nanocarbon Co.) were dispersed in 100 ml of deoxycholate sodium salt (DOC, Tokyo Chemical Industry Co.) 1% solution by a bath-type ultrasonic cleaner (Sharp Co., UT-206H).

2. 30 ml of the solution was dispersed using a digital sonifier (Branson, 250DA) for 4 hours at 20 % output.

3. Dispersed solutions were centrifuged for 30 minutes at 40,000 rpm (Rotor P40ST, Hitachi Koki Co.). Supernatant was obtained.

4. Density-gradient was formed in a centrifuge tube (40 PA seal tube, 345321A, Hitachi Koki Co.) using the following 5 solutions; (1) iodixanol 25 %, sodium dodecyl sulfate (SDS, Aldrich) 1.5 %, and sodium cholate (SC, Aldrich) 1.5%, (2) iodixanol 30 %, SDS 1.5 %, and SC 1.5%, (3) iodixanol 32.5 %, SDS 1.5 % and SC 1.5%, (4) iodixanol 35 %, SDS 1.5 %, and SC 1.5%, and (5) iodixanol 40 %, SDS 2.4 %, SC 0.6 %, DOC 0.33 %, and SWCNTs (the solution was adjusted using the supernatant obtained at the previous step). Here the concentrations of iodixanol were adjusted using Optiprep (60 % iodixanol solution, COSMO BIO Co.).

5. The tube was centrifuged at 50,000 rpm for 9 hours, 22 °C (Rotor P50VT2, Hitachi Koki Co.). Then, high-purity metallic SWCNTs were obtained at the upper part of the centrifuge tube, and high-purity semiconducting SWCNTs were obtained at its lower part.

Scheme 2

1. 100 mg of SWCNTs were dispersed in 100 ml of DOC 2% solution by a bath-type ultrasonic cleaner (Sharp Co., UT-206H).

2. 30 ml of the solution was dispersed using a digital sonifier (Branson, 250DA) for 4 hours at 20 % output.

3. Dispersed solutions were centrifuged for 30 minutes at 40,000 rpm (Rotor P40ST, Hitachi Koki Co.). Supernatant was obtained.

4. Density-gradient was formed in a centrifuge tube (40 PA seal tube, 345321A, Hitachi Koki Co.) using the following 6 solutions; (1) SWCNT solution, which was obtained at the previous step, (2) iodixanol 25 %, SDS 2%, (3) iodixanol 30 %, SDS 2 %, (4) iodixanol 32.5 %, SDS 2 %, (5) iodixanol 35 %, SDS 2 %, (6) iodixanol 40 %, SDS 2 %.

5. The tube was centrifuged at 50,000 rpm for 9 hours, 22 °C (Rotor P50VT2, Hitachi Koki Co.). Then, high-purity metallic SWCNTs were obtained at the upper part of the centrifuge tube, and high-purity semiconducting SWCNTs were obtained at its lower part.

Details of washing procedures of SWCNTs after density-gradient separations

1. Methanol was added to SWCNT dispersed solutions, then the SWCNTs were re-bundled, and the solution was filtered (pore size 0.2 μ m, Millipore Co.), and then hot water (about 70 ° C ~ 100 ° C) was poured to the SWCNTs formed on the surface of the filter.

2. The SWCNTs were well dispersed into methanol solutions using a ultrasonic cleaner (Sharp Co., UT-206H), and the solutions were filtered, and hot water was poured. This step was repeated four times.

3. The SWCNTs were well dispersed into methanol solutions, and then filtered.

4. The SWCNTs were well dispersed into toluene solutions using a ultrasonic cleaner, and then filtered. This step was repeated two times.

5. The SWCNTs were well dispersed into methanol solutions, and the solutions were filtered, then finally a sheet of bucky-paper of SWCNTs was obtained.

6. The sheet was annealed at 500 ° C for 1 hour under vacuum, $\sim 10^{-6}$ torr.