Supplementary Information

Enhancement of the Electrical Conductivity of Defective Carbon Nanotube Sheets for Organic Hybrid Thermoelectrics by Deposition of Pd Nanoparticles

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Fig. S9. SEM photographs (magnifications = 5,000, 50,000, and 500,000) of sheets of blank SGCNTs and chemically prepared Pd NP-decorated SGCNTs (Pd@SGCNT) at the charged ratio of Pd:SGCNT = 1:9, 2:8, 3:7, and 5:5.

Fig. S10. Size distribution histogram of Pd NPs in Pd@SGCNT (Pd content =9.47 wt.%) using "count" as the ordinate instead of "fraction" shown in Fig. 2b. The 200 Pd NPs were counted in this case.

Fig. S11. Temperature dependence of the thermoelectric figure-of-merit (*ZT*) of the sheets composed of SGCNTs only (blank) and Pd NP-covered SGCNTs (Pd@SGCNT) with various Pd contents. The error bars are clearly shown by minimizing the sizes of the average points in this Figure compared to those in Fig. 4.

Table S1. The relation of electrical conductivity and Pd content in the sheet of Pd@SGCNT, (Pd NP + SGCNT), and (Pd Black + SGCNT)

Table S2. The thermal conductivity (κ) through plane of the sheets fabricated from the blank SGCNTs only and the palladium NP-decorated SGCNTs (Pd@SGCNT) with various Pd contents (at rt)

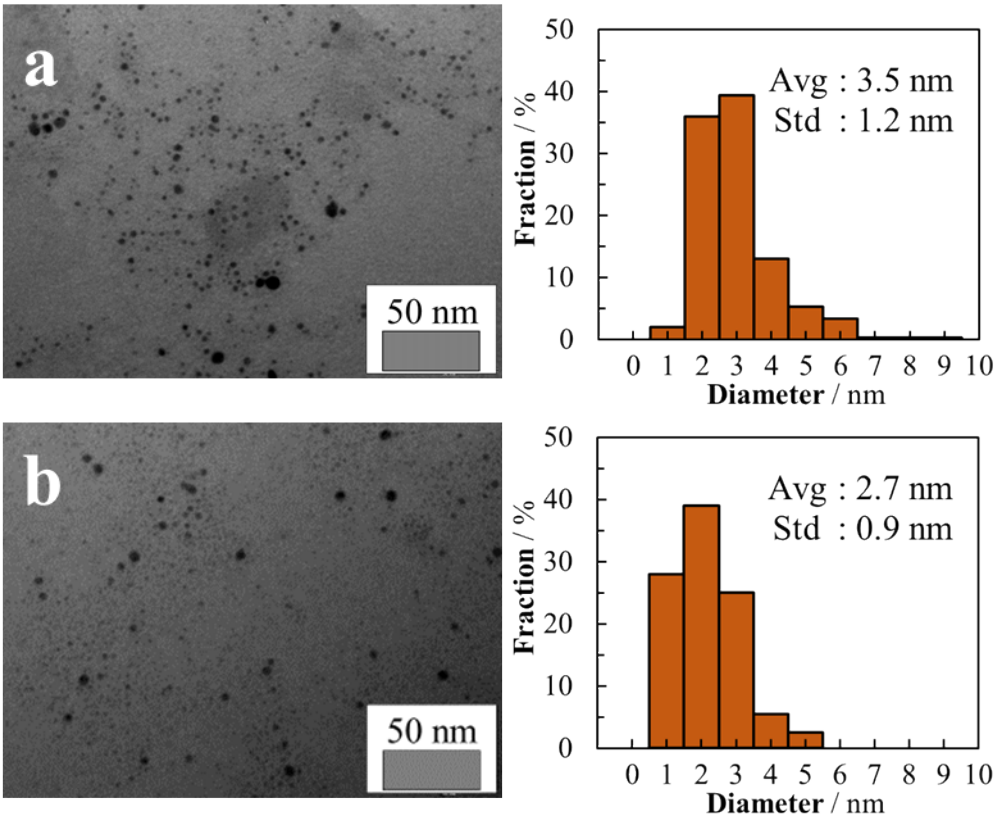


Fig. S1. TEM images and size distribution histograms of palladium black (a) and separately prepared Pd NPs (b).

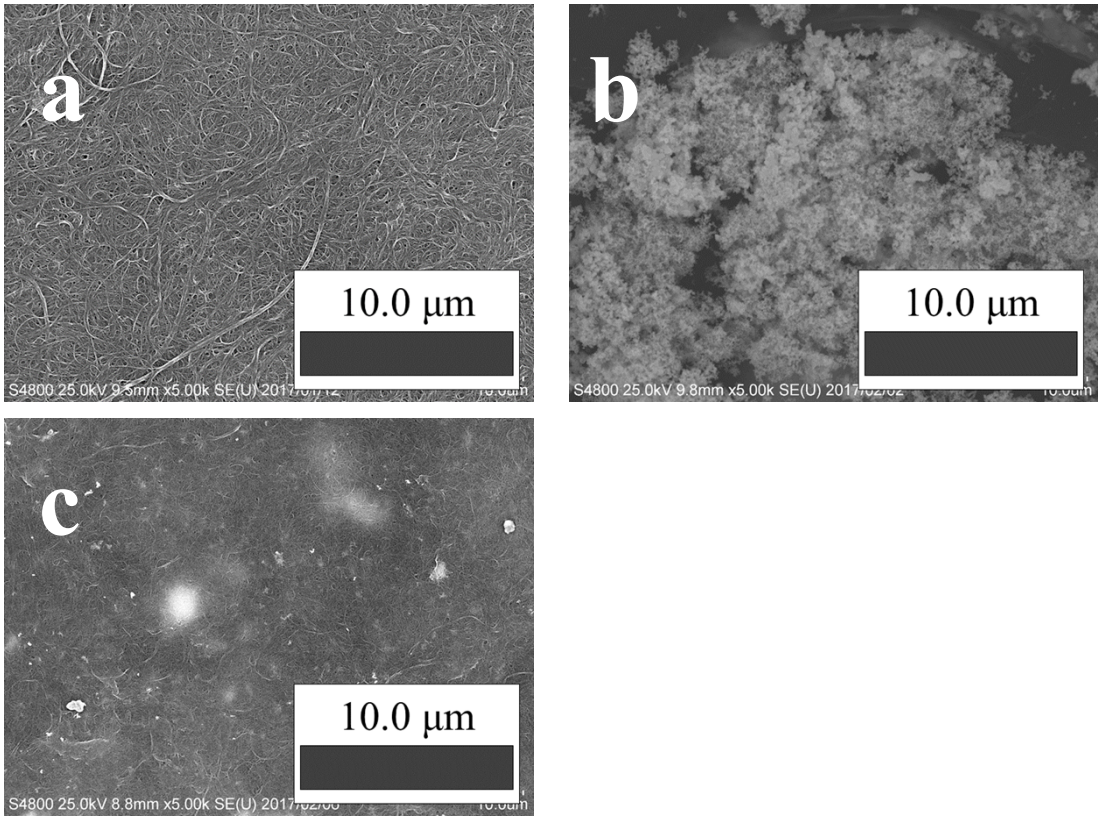


Fig. S2. SEM images of the sheets composed of SGCNTs without NPs (a), with Pd black (Pd content = 8.16 wt.%) (b) and separately prepared Pd NPs (Pd content = 6.44 wt.%) (c).

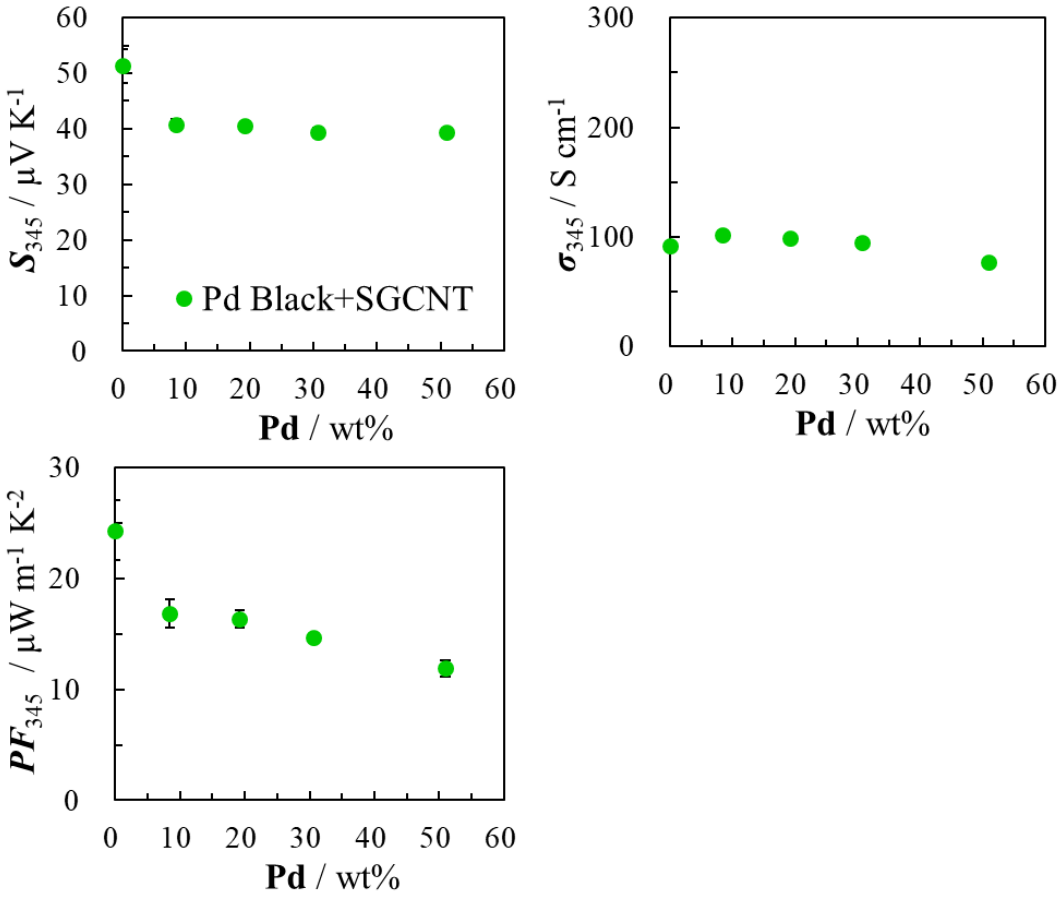
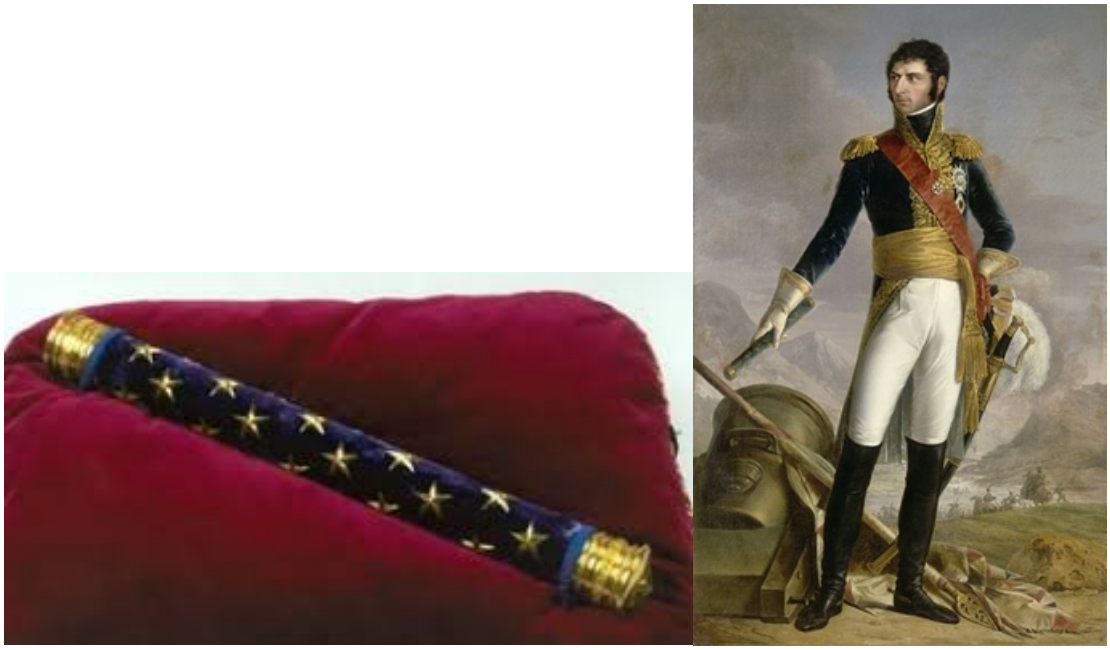


Fig. S3. Palladium content dependence of the Seebeck coefficient (S), electrical conductivity (σ) and power factor ($PF=S^2\sigma$) of the sheet of SGCNTs decorated with commercially available palladium black NPs at 345 K.



(a) **(b)**

Fig. S4. Images of Marshal's Baton (a), and Charles XIV and III John holding his Marshal's Baton in his right hand (b).

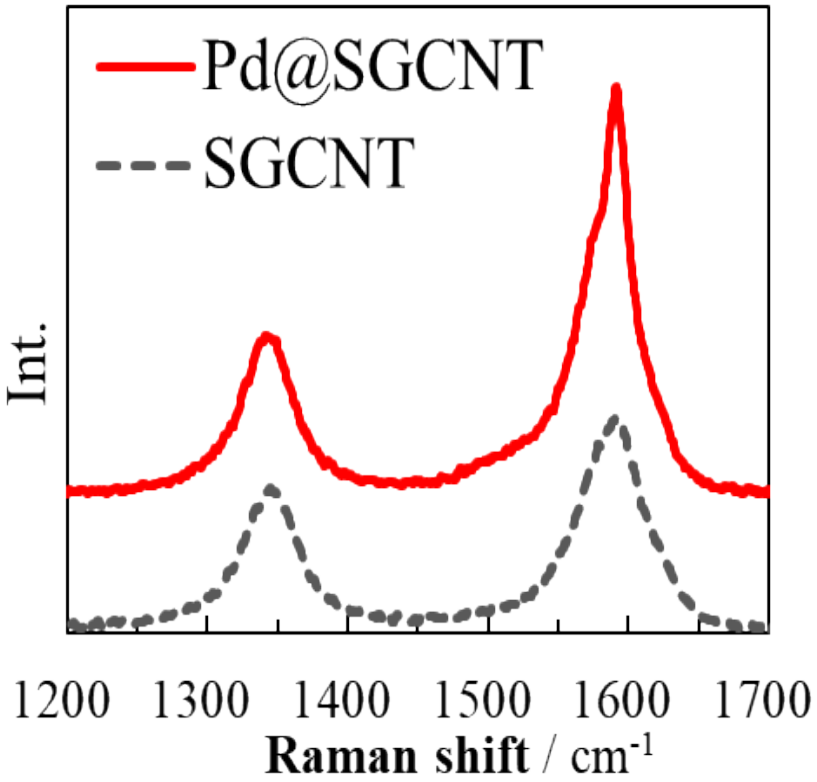


Fig. S5. Raman spectra of the SGCNT and Pd NP-covered SGCNT with an accumulated chemical reduction method (Pd@SGCNT, Pd content = 11.0 wt.%).

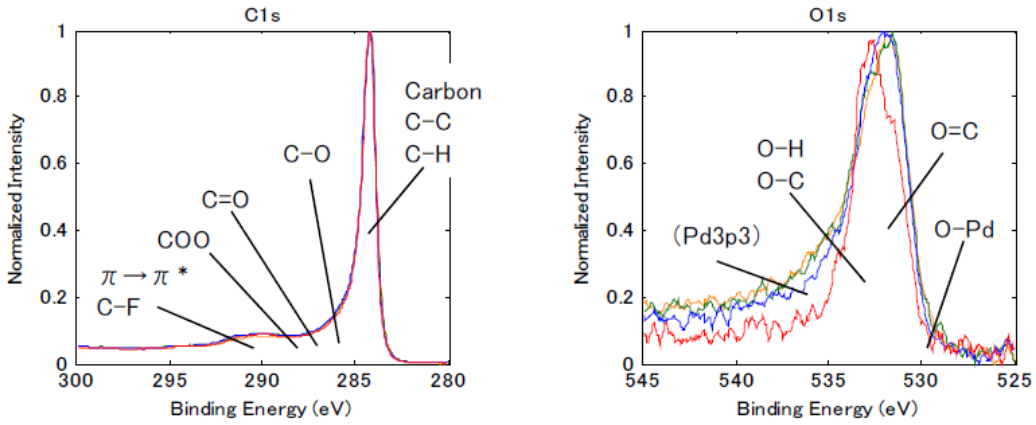


Fig. S6. C1s and O1s parts of normalized narrow XPS spectra of the sheets of blank SGCNTs (red), Pd@SGCNTs with the Pd content of 4.62 wt.% (blue), 9.47 wt.% (green), and 17.43 wt.% (orange).

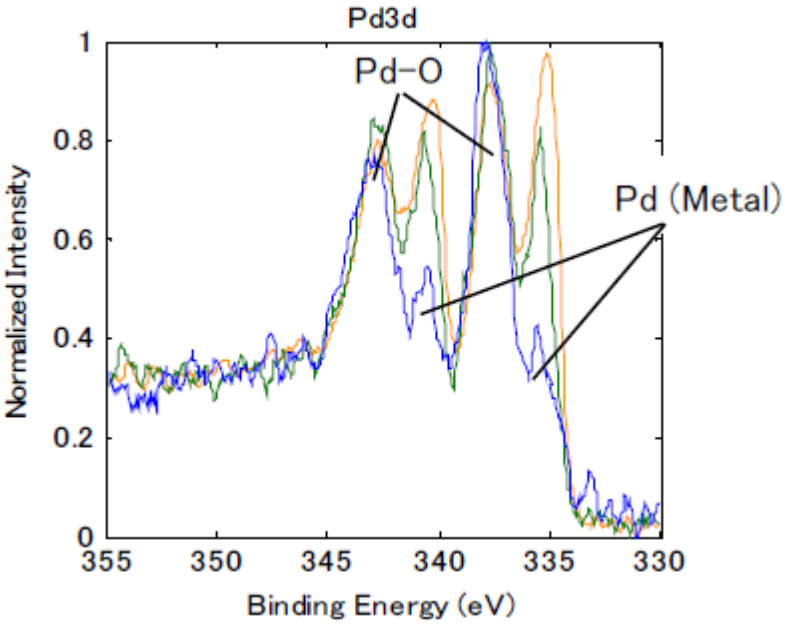
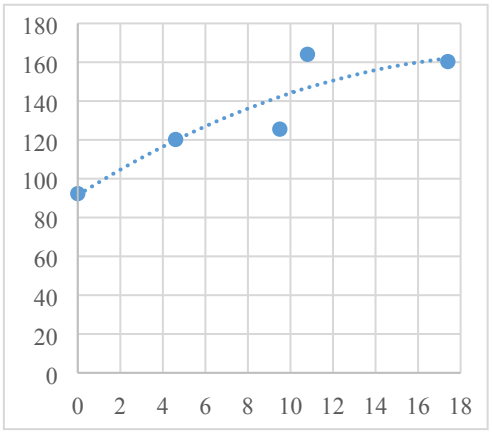
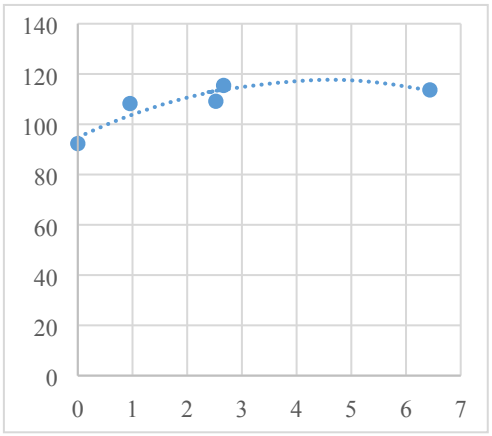


Fig. S7. Pd3d part of normalized narrow XPS spectra of the sheets of Pd@SGCNTs with the Pd content of 4.62 wt.% (blue), 9.47 wt.% (green), and 17.43 wt.% (orange).

(a)



(b)



(c)

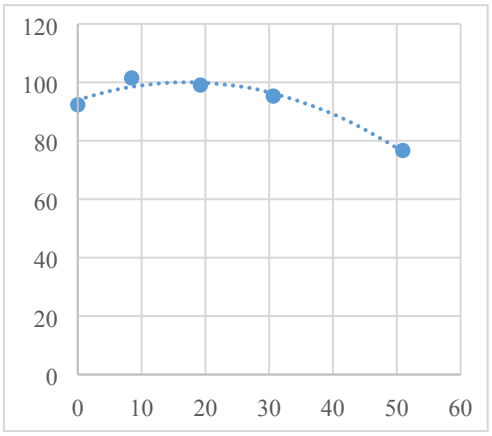
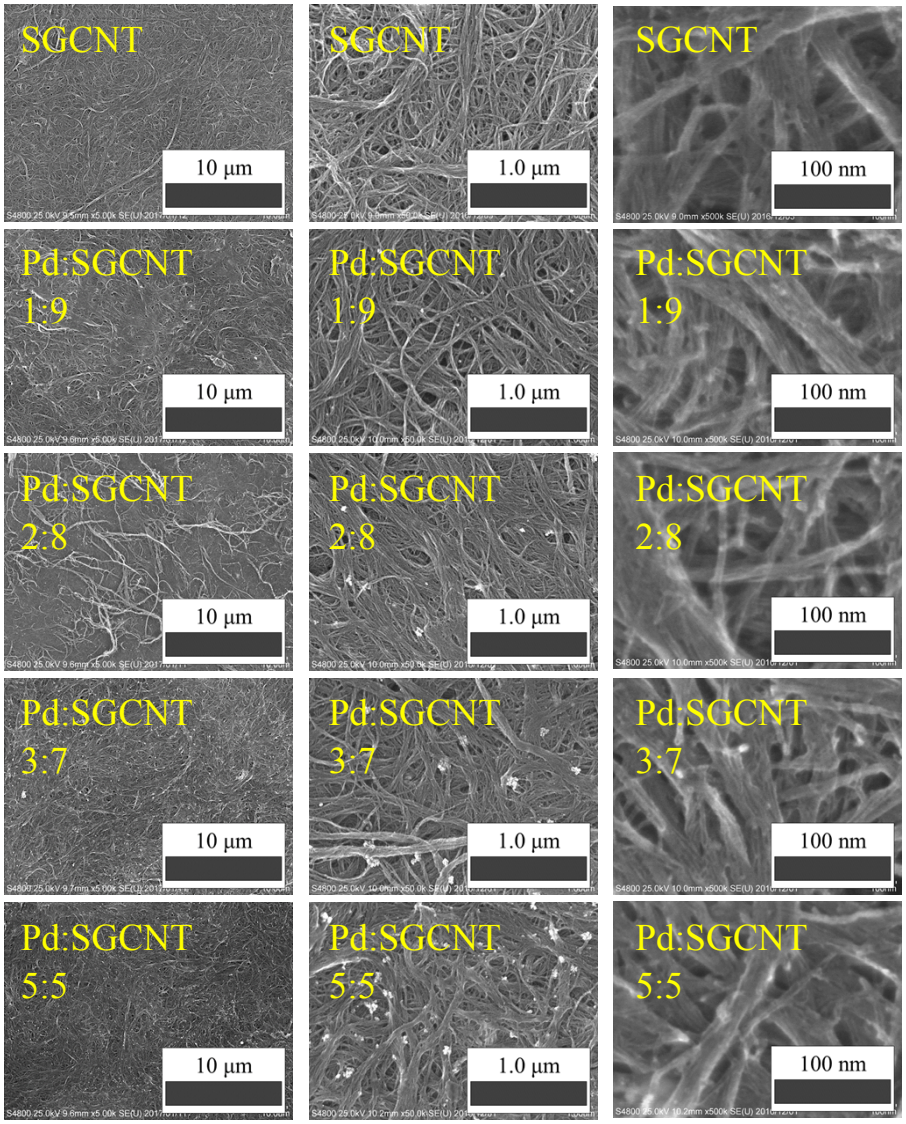
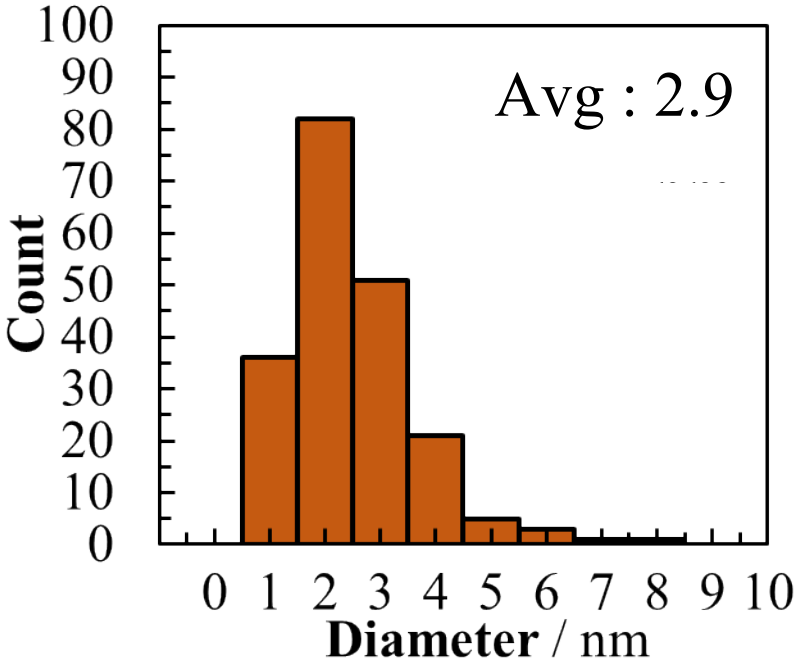


Fig. S8. Electrical conductivity (S/cm) vs. Pd content (wt.%) of (a) Pd@SGCNT sheet, (b) Pd NP + SGCNT sheet, and (c) Pd Black + SGCNT sheet. Dotted lines show approximate expression by the secondary curves.



×5k ×50k ×500k

Fig. S9. SEM photographs (magnifications = 5,000, 50,000 and 500,000) of the sheets of the blank SGCNTs and chemically prepared Pd NP-decorated SGCNTs (Pd@SGCNT) at the charged ratios of Pd:SGCNT = 1:9, 2:8, 3:7 and 5:5.



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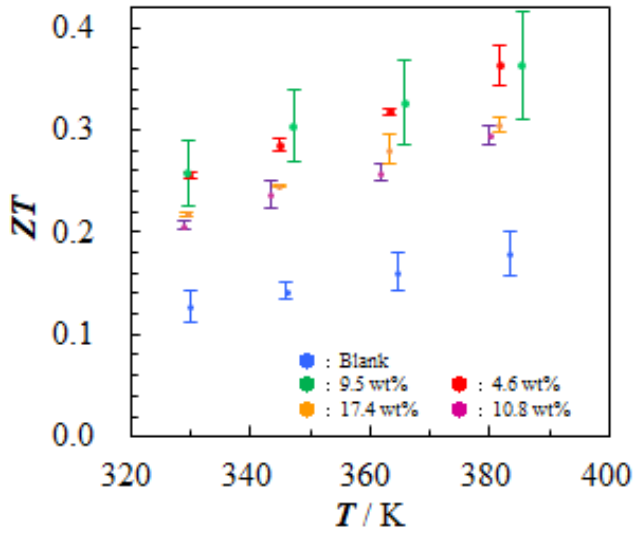

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Table S1. The relation of the Pd content and the electrical conductivity estimated from Fig. S8 in the sheets of Pd@SGCNT, (Pd NP + SGCNT) and (Pd Black + SGCNT)

Pd content	Pd@SGCNT		Pd NP + SGCNT		Pd Black + SGCNT	
wt.%	S/cm	%	S/cm	%	S/cm	%
0	92.3	100.0	92.3	100.0	92.3	100.0
6	127.0	137.6	115.1	124.7	97.5	105.6
8	136.2	147.6	103.8	112.5	98.3	106.5
10	144.1	156.1	83.3	96.2	99.0	107.3
12	150.7	163.3	53.9	58.4	99.5	107.8
14	156.0	169.0	15.3	16.6	99.8	108.1

Table S2. Calculation of the thermal conductivity (κ) through plane of the sheets fabricated from the blank SGCNTs and the Pd NP-decorated SGCNTs (Pd@SGCNT) with various Pd contents based on the data of the density (ρ), specific heat (C_p) and thermal diffusivity (α) at rt

Pd Conc.	ρ	C_p	α	κ
/ wt.%	/ g (cm)$^{-3}$	/ J g^{-1} K^{-1}	/ mm^2 s^{-1}	/ W m^{-1} K^{-1}
0 (Blank)	0.83	0.96	0.06±0.01	0.05
4.6	0.89	0.98	0.06±0.02	0.05
9.5	0.88	0.79	0.08±0.01	0.05
10.8	0.92	0.95	0.09±0.01	0.08