Electronic Supplementary Information

Enhancing Thermoelectric Performance of Single-Walled Carbon Nanotube/Reduced Graphene Oxides Composites with Small Organic Molecules as a Novel Additive

Jae Gyu Jang[a b], Tae-hoon Kim[a], Sung Hyun Kim[b], and Jong-In Hong[a]**

[a]Department of Chemistry, Seoul National University, Seoul 08826, Republic of Korea
E-mail: jihong@snu.ac.kr

[b]Department of Carbon Convergence Engineering, Wonkwang University, Iksan, Jeonbuk 54538, Republic of Korea
E-mail: shkim75@wku.ac.kr

1. Experimental Section

Single crystal X-ray structure analysis: Single crystals (**1**, C70H78N4S3 (2,2'-(3,3"-dihexyl-[2,2':5',2"-terthiophene]-5,5"-diyl)bis(1-octyl-1H-phenanthro[9,10-d]imidazole)) were obtained by vapor diffusion of dimethylforamide into a concentrated solution of compound **1** in a mixture of dichloromethane and dichlorobenzene (4:1, v/v). A suitable crystal was taken by using a nylon loop coated with Paratone® N oil and mounted on an Agilent SuperNova, Dual, Cu at home/near, AtlasS2 diffractometer. The crystal was kept at 292.9(8) K. The X-ray diffraction analysis was conducted using Cu Kα radiation (λ= 1.542 Å). A total number of 15369 reflections were obtained ($5.83° \leq 2\Theta \leq 134.154°$) with 1° steps (ω scan). From Olex2, the structure was solved with the ShelXT structure solution program using Intrinsic Phasing and refined with the ShelXL refinement package using Least Squares minimization. CCDC(1854222) contains the supplementary crystallographic data of this paper. These data can be obtained free of charge from the Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.

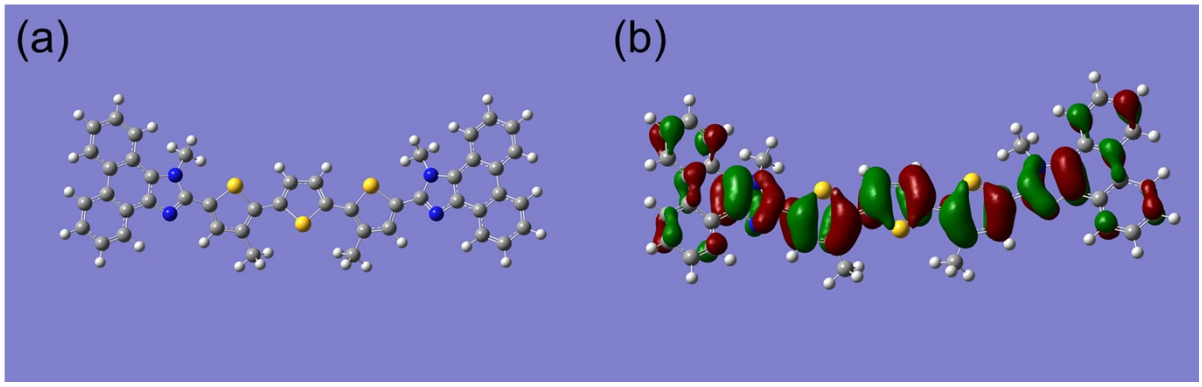

Fig. S1. (a) Ground state geometry and (b) HOMO distribution of **1** in vacuum from density theory calculations at the B3LYP/6-31G* level using the Gaussian03 package.

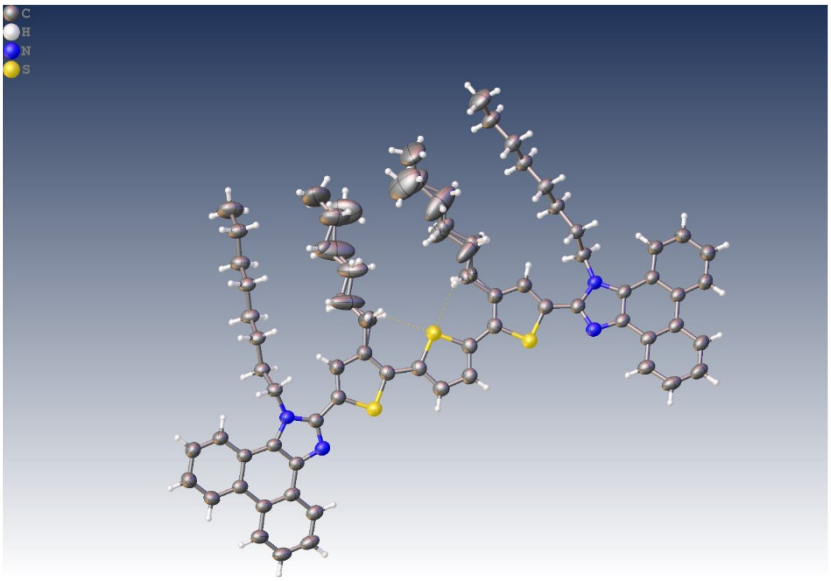

Fig. S2. Single-crystal X-ray structure of **1**.

Table S1. Crystallographic data and structure refinement information for 2,2'-(3,3"-dihexyl-[2,2':5',2"-terthiophene]-5,5"-diyl)bis(1-octyl-1H-phenanthro[9,10-d]imidazole)

Identification code	dihexylterthiophenediyl bisoctylphenanthroimidazole
Empirical formula	$C_{70}H_{78}N_4S_3$
Formula weight	1071.54
Temperature/K	292.9(8)
Crystal system	orthorhombic
Space group	Pbcn
a/Å	25.5231(13)
b/Å	18.8440(5)
c/Å	12.4455(3)
α/°	90
β/°	90
γ/°	90
Volume/Å^3	5985.8(4)
Z	4
ρ_{calc}g/cm^3	1.189
μ/mm^{-1}	1.467
F(000)	2296.0
Crystal size/mm^3	0.366 × 0.101 × 0.061
Radiation	CuKα (λ = 1.54184)
2Θ range for data collection/°	5.83 to 134.154
Index ranges	$-20 \le h \le 30$, $-14 \le k \le 22$, $-10 \le l \le 14$
Reflections collected	15369
Independent reflections	5342 [R_{int} = 0.0207, R_{sigma} = 0.0194]
Data/restraints/parameters	5342/0/404
Goodness-of-fit on F^2	1.036
Final R indexes [I>=2σ (I)]	R_1 = 0.0430, wR_2 = 0.1091
Final R indexes [all data]	R_1 = 0.0543, wR_2 = 0.1203
Largest diff. peak/hole / e Å^{-3}	0.16/-0.26

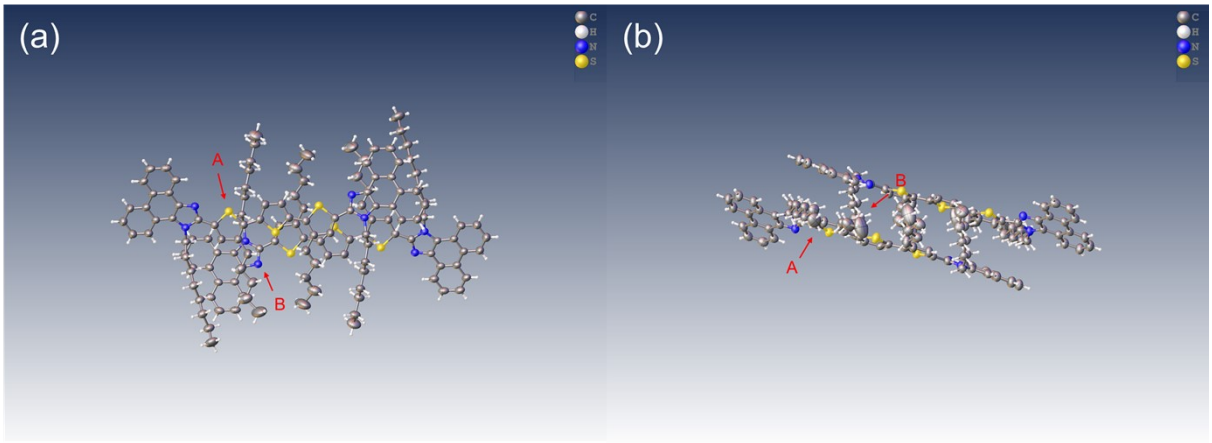

Fig. S3. Packing patterns of **1**. The arrows are eye guides.

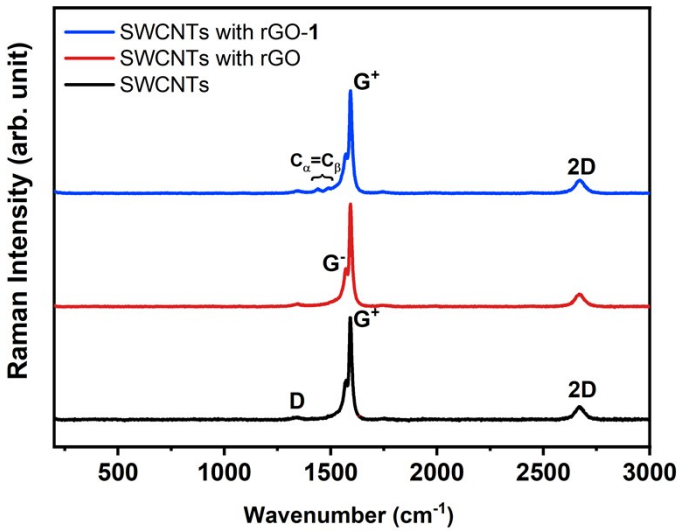

Fig. S4. Raman spectra of SWCNTs, SWCNTs with pristine rGOs, and SWCNTs with rGOs-**1**.

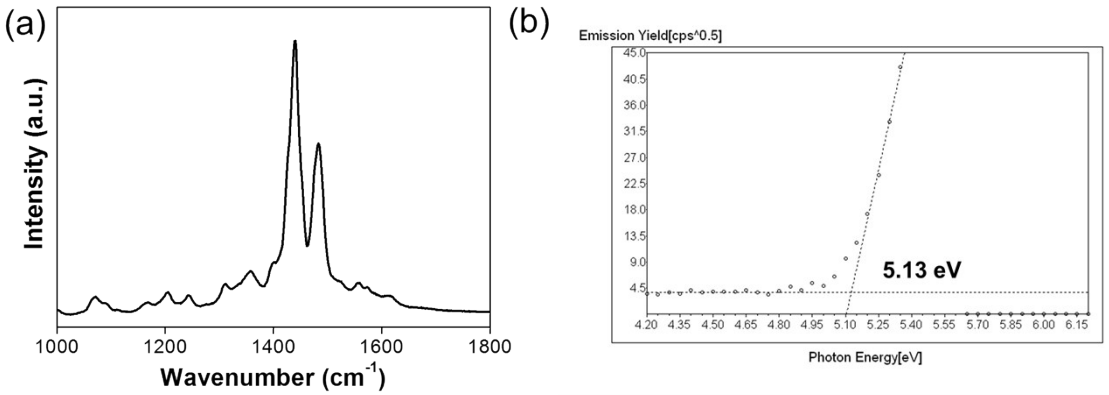

Fig. S5. (a) Raman spectrum of **1** using an excitation wavelength of 532 nm. (b)Work function of **1**.

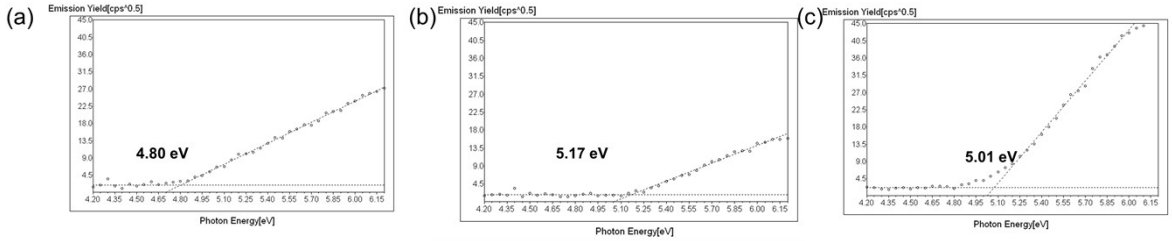

Fig. S6. Work functions of (a) SWCNTs, (b) pristine rGOs, and (c) rGOs-**1**

Table S2. Thermoelectric parameters of SWCNTs with pristine rGO and rGO-**1**.

Thermoelectric Materials		Seebeck coefficient (μV K^{-1})	Electrical conductivity (S cm^{-1})	Power factor (μW m^{-1} K^{-1})
SWCNTs with pristine rGOs	0	51.9 ± 7.6	225.0 ± 22.6	62.9 ± 8.7
	10	51.4 ± 3.6	145.0 ± 16.8	38.6 ± 6.2
	20	63.5 ± 3.4	137.0 ± 8.7	55.4 ± 5.2
	30	68.5± 6.4	134.0 ± 13.6	55.1± 9.5
	40	74.1 ± 1.8	114.0 ± 9.6	62.8 ± 2.9
	50	71.5 ± 3.6	79.2 ± 8.6	40.5 ± 5.0
SWCNTs with rGOs-**1**	0	51.9 ± 7.6	225.0 ± 22.6	62.9 ± 8.7
	10	66.4 ± 7.6	187.0 ± 11.8	84.0 ± 10.3
	20	73.2 ± 3.6	179.0 ± 10.2	95.7 ± 8.1
	30	73.2 ± 3.1	107.0 ± 9.4	57.6 ± 7.9
	40	75.1 ± 6.9	74.5 ± 12.5	43.1 ± 11.5
	50	80.0 ± 5.4	62.0 ± 14.9	39.4 ± 4.3

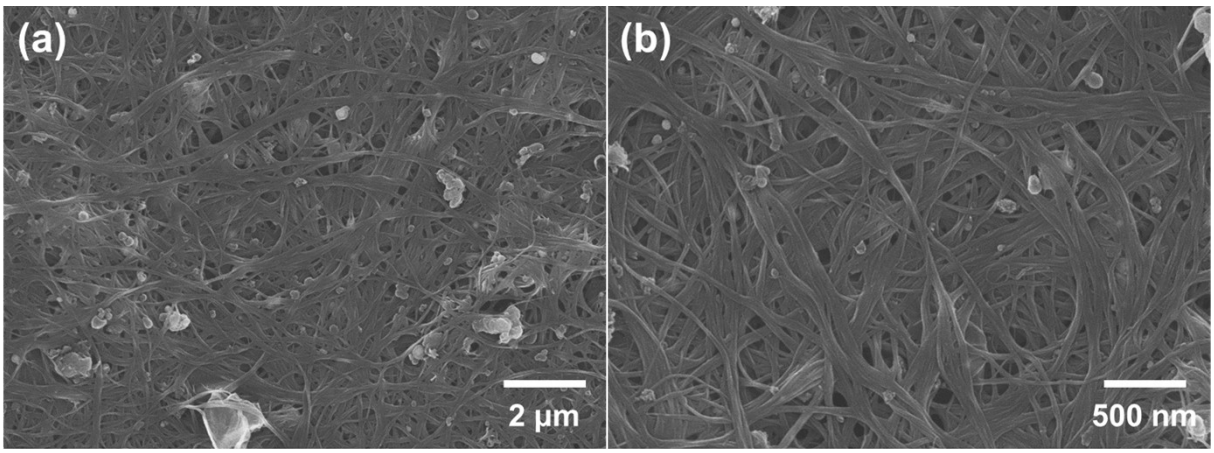

Fig. S7. (a) and (b) SEM images of SWCNTs. The white particles observed in the round shape are metal catalysts.

Table S3. Detailed fitting parameters, σ_0, n, and T_0 values, obtained by normalized temperature-dependent resistances of SWCNTs, SWCNTs with 20 wt% and 40 wt% pristine rGOs, and SWCNTs with 20 wt% and 40 wt% rGOs-**1**.

Fitting parameters	σ_0	T_0	n
SWCNTs	332.57	19.37	1
SWCNTs with 20 wt% pristine rGOs	323.83	162.09	3
SWCNTs with 20 wt% rGOs-**1**	268.23	47.86	1
SWCNTs with 40 wt% pristine rGOs	301.67	274.45	3
SWCNTs with 40 wt% rGOs-**1**	308.3	1964.05	3

Table S4. Estimated theoretical power outputs of thermoelectric modules having different thermoelectric elements.

Number of thermoelectric element	Temperature gradient (K)	Current (mA)	Voltage (mV)	Theoretical power output, P_{Th} (nW)
1	3.3	4.19	25.02	0.11
	5.2	56.01	406.5	22.8
	7	77.6	603.3	46.81
5	3.3	-		0.53
	5.2			113.85
	7			234.07
8	3.3	-		0.84
	5.2			182.16
	7			374.51