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**Improved Thermoelectric Properties of Dispersant-Free  
Semiconducting Single-Walled Carbon Nanotubes Sorted by  
a Flavin Extraction Method**

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## Thermoelectric Properties of Dispersant-Free Semiconducting Single-Walled Carbon Nanotubes Sorted by a Flavin Extraction Method

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**Semiconducting single-walled carbon nanotubes (s-SWNTs) were extracted from SWNT mixtures using a flavin derivative (FC12). We evaluated the thermoelectric properties of the s-SWNT sheets. Electrical conductivity, power factor and figure of merit values of the sheets were increased by two orders of magnitude after removing FC12 simply by dipping in dichloromethane.**

Single-walled carbon nanotubes (SWNTs) are promising thermoelectric (TE) materials because of their excellent conductivity, light weight, flexibility, low toxicity, abundant component element, simple film fabrication, and production scalability.<sup>1</sup> In particular, semiconducting single-walled carbon nanotubes (s-SWNTs) are attracting considerable attention because of their unique electronic properties such as high mobility and sharp van Hove singularity band structures,<sup>2, 3</sup> which are favorable for high TE conversion efficiency.<sup>4-6</sup> TE conversion efficiency is often estimated by power factor ( $PF$ ) =  $\sigma S^2$  and figure of merit ( $zT$ ) =  $\sigma S^2 T / \kappa$ , where  $S$ ,  $\sigma$ ,  $\kappa$ , and  $T$  are the Seebeck coefficient ( $V K^{-1}$ ), electrical conductivity ( $S m^{-1}$ ), thermal conductivity ( $W m^{-1} K^{-1}$ ), and absolute temperature (K), respectively.<sup>7</sup> Both theoretical and experimental reports have recently demonstrated that s-SWNTs possess an extremely large  $S$  of over  $1000 \mu V K^{-1}$  at room temperature,<sup>2, 3, 8</sup> which is much higher than that of inorganic semiconducting materials.<sup>9</sup> Another attractive feature of s-SWNTs is that unlike inorganic or metallic TE candidates, whose  $\sigma$  and  $\kappa$  are interrelated through the Wiedemann–Franz law,<sup>1, 10</sup>  $\sigma$  and  $\kappa$  of s-SWNTs can be separately controlled because phonon and carrier

conductions are mediated by different mechanisms;<sup>11, 12</sup> thus, higher  $zT$  ( $\propto \sigma / \kappa$ ) of s-SWNTs is achievable. In addition,  $\sigma$  of s-SWNTs can be increased substantially by carrier doping with chemical dopants; therefore, optimization of  $PF$  and  $zT$  is possible.<sup>2, 5, 6</sup>

In reality, SWNTs are produced as a 1:2 mixture of metallic (m-) and s-SWNTs<sup>13</sup> and high purity s-SWNT can be obtained by many extraction methods.<sup>14-19</sup> In particular, the polyfluorene (PFO)-based extraction method has often been used for TE research<sup>2</sup> because of its simple process and high sorting purity of s-SWNTs.<sup>18</sup> In 2016, Ferguson et al. produced an s-SWNT network with extremely high  $PF$  ( $340 \mu W m^{-1} K^{-2}$ ) and  $zT$  ( $2.1 \times 10^{-2}$ ) obtained by PFO extraction.<sup>2</sup> Recently, the same group pointed out that PFO-based polymer residue left on s-SWNTs hampered the charge carrier transport inside the s-SWNT network and demonstrated that removal of the polymer using “cleavable” H-bonded supramolecular PFO<sup>20</sup> led to a further increase of  $PF$  (by  $\sim 2.5$  times), which was mainly derived from the large increase in  $\sigma$  ( $\sim 2$  times).<sup>5</sup> However, this removal process required trifluoroacetic acid to break down the H bonding of the “cleavable” PFO,<sup>5, 6, 20</sup> which is not favorable for the device process. In addition, the depolymerized monomer needed to be polymerized to allow recycling.<sup>6</sup> Therefore, more mild method to obtain dispersant-free s-SWNTs is required.

In this study, we used Flavin derivative 10-dodecyl-7,8-dimethyl-10H-benzo[g]pteridine-2,4-dione (FC12, Fig. 1) to extract s-SWNTs from unsorted SWNTs since the Flavin extraction provides high-purity of s-SWNT (>99%) and FC12 was easily removed using organic solvents such as methanol and dichloromethane ( $CH_2Cl_2$ ).<sup>21-23</sup> In addition, FC12 is expected to be easy to recycle, which makes it suitable for large-scale synthesis of dispersant free s-SWNTs. By using FC12-free

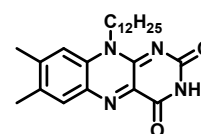


Fig. 1 Chemical structure of FC12.

Electronic Supplementary Information (ESI) available: [details of any supplementary information available should be included here]. See DOI: 10.1039/x0xx00000x

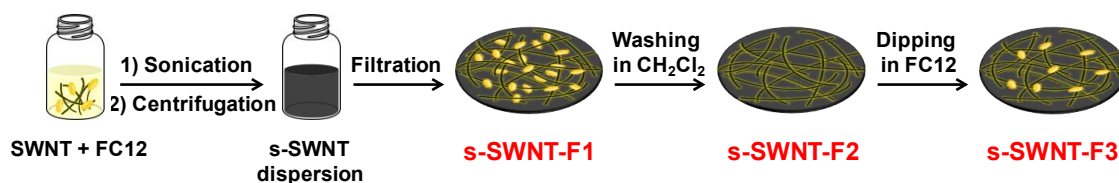


Fig. 2 Schematic illustration of the fabrication process of s-SWNT-F1, s-SWNT-F2, and s-SWNT-F3.

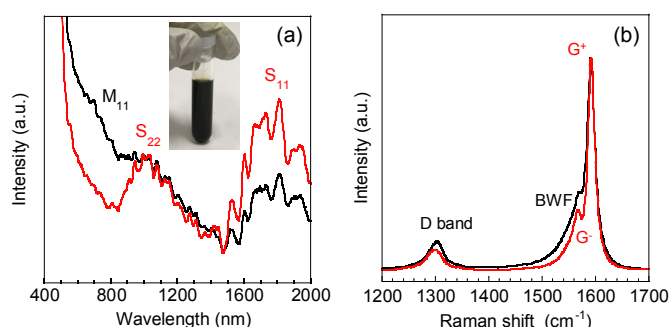


Fig. 3 (a) UV-vis-NIR absorption spectra of s-SWNT dispersion (red) and unsorted SWNT dispersion (black) in toluene. (b) Raman spectra of s-SWNT-F2 (red), and unsorted SWNT (black) obtained at an excitation wavelength of 785 nm. Inset; Photo of s-SWNT dispersion.

s-SWNT free-standing sheets fabricated by FC12 extraction method, TE properties of the sheets were evaluated. As the result, a large increase in  $PF$  and  $zT$  was achieved when FC12 was removed by  $\text{CH}_2\text{Cl}_2$  washing.

s-SWNT extraction and sheet fabrication process were shown in Fig. 2. For s-SWNT extraction, SWNTs were added to FC12 solution in toluene and dispersed by a bath sonicator.<sup>21, 23</sup> The dispersion was centrifuged and then the SWNT supernatant was collected (For the details of experiments, see ESI†). Fig. 3a shows the UV-vis-NIR absorption spectra of the unsorted SWNT dispersion and SWNT supernatant in toluene after centrifugation. The broad m-SWNT peak centered at around 800 nm ( $M_{11}$ ) observed for the unsorted SWNT dispersion was largely decreased and clear  $S_{11}$  and  $S_{22}$  peaks centered at 1800 and 1000 nm, respectively,<sup>24</sup> were observed in the SWNT supernatant (red line in Fig. 3a), which strongly indicated that the supernatant was s-SWNT dispersion (inset in Fig. 3a).<sup>21, 23</sup> This result is also supported by the Raman spectra presented in Fig. 3b, in which a clear  $G^+$  peak was observed at  $1566.6\text{ cm}^{-1}$  for the s-SWNTs, which was related to the decrease in the intensity of the Breit–Wigner–Fano (BWF) peak from the m-SWNTs.<sup>25, 26</sup> The purity of the s-SWNTs estimated from the conductivity measurements based on dimensionality of the samples obtained from variable range hopping (VRH) model fitting was  $\sim 98\%$  (Fig. S1 and Table S1, ESI†),<sup>27, 28</sup> which was in accordance with the previous reports.<sup>21, 22</sup>

Using the above s-SWNT dispersion, we prepared free-standing s-SWNT sheet by filtration (denoted as s-SWNT-F1). Then, s-SWNT-F1 was dipped in  $\text{CH}_2\text{Cl}_2$  several times to remove FC12 until the yellowish color of FC12 disappeared to afford s-SWNT-F2 (inset in Fig. 4b). The yield of s-SWNTs is 20% out of

the starting material. To confirm the effect of residual FC12, control sheet containing FC12 was prepared by dipping s-SWNT-F2 in FC12 solution in toluene for 2 h (denoted as s-SWNT-F3). In addition, to confirm the effect of s-SWNT extraction, an unsorted SWNT sheet (denoted as unsorted SWNT) was also fabricated in the same manner as s-SWNT-F2 using unsorted SWNT dispersion.

Fig. 4 showed scanning electron microscopy (SEM) images of s-SWNT-F1 (Fig. 4a) and s-SWNT-F2 (Fig. 4b). A clear network structure of bundled SWNTs was observed for both s-SWNT-F1 and s-SWNT-F2. Bundled SWNTs was also observed for s-SWNT-F3 and unsorted SWNT (Fig. S2, ESI†)

To estimate the amount of the residual FC12, X-ray photoelectron spectroscopy (XPS) spectra of N1s peaks at around 400 eV were measured (Fig. 5a). The mass percentages of carbon to nitrogen in the sheets estimated from XPS were summarized in Fig. 5b. We found that s-SWNT-F2 and unsorted SWNT possessed quite low N1s contents similar to that of the SWNTs (0.5%), clearly indicating that FC12 was successfully removed by dipping in  $\text{CH}_2\text{Cl}_2$ .

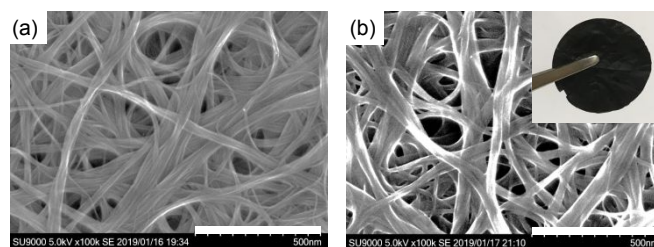


Fig. 4 SEM images of (a) s-SWNT-F1 and (b) s-SWNT-F2. Scale bars: 500 nm. Inset in (b) is a photograph of the free-standing s-SWNT-F2.

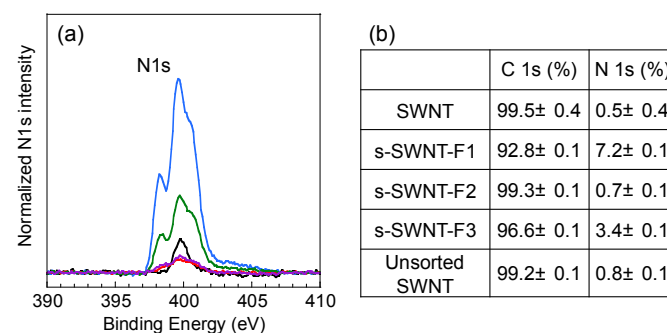
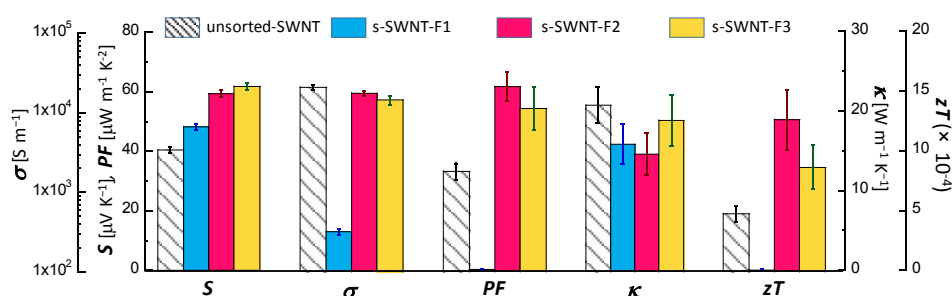


Fig. 5 (a) XPS N1s narrow scans of SWNT (black), s-SWNT-F1 (blue), s-SWNT-F2 (red), s-SWNT-F3 (green) and unsorted SWNT (purple). (b) Mass percentages of C1s and N1s calculated from XPS measurements.



**Fig. 6** In-plane Seebeck coefficient ( $S$ ), electrical conductivity ( $\sigma$ ), power factor ( $PF$ ), thermal conductivity ( $\kappa$ ), and  $zT$  of unsorted SWNT, s-SWNT-F1, s-SWNT-F2, and s-SWNT-F3 in a helium atmosphere at 303 K and 0.01 MPa.

In contrast, s-SWNT-F1 and s-SWNT-F3 contained larger contents of N1s (7.2% and 3.4%, respectively), clearly indicating that FC12 remained in the sheets. Suppose that all the N atoms are originated from the FC12. 7.2% mass ratio of N corresponded to ca. 45 wt% of FC12 in the s-SWNT-F1, which was comparable to the PFO/s-SWNT composite used for TE applications.<sup>2</sup> On the other hand, s-SWNT-F2 contained only 4 wt% of FC12. For these sheets, FC12 was adsorbed on the SWNT surfaces mainly through  $\pi$ - $\pi$  interactions in a supramolecular fashion as described in the previous reports.<sup>29, 30</sup> Compared to the PFO-based polymers that attached to the SWNT surface through multipoint interactions, FC12 molecules adsorbed dynamically on the SWNT surface in solution because FC12 is a small molecule.<sup>17, 31</sup> Therefore, FC12 was easily washed away by good solvent for FC12 such as  $\text{CH}_2\text{Cl}_2$ .<sup>21, 22</sup>

Fig. 6 summarizes the  $S$ ,  $\sigma$ ,  $\kappa$ ,  $PF$ , and  $zT$  values of the sheets at 303 K. The large increases of  $S$  for s-SWNT-F2 and s-SWNT-F3 (59.4 and 61.8  $\mu\text{V K}^{-1}$ , respectively) compared with that of unsorted SWNT (40.3  $\mu\text{V K}^{-1}$ ) clearly supported the successful extraction of s-SWNTs using FC12.<sup>32, 33</sup> The slightly smaller  $S$  observed for s-SWNT-F1 (48.2  $\mu\text{V K}^{-1}$ ) was explained probably by the n-type doping by large amount of FC12, similar to the doping behavior of other nitrogen-containing compounds.<sup>34</sup>

Regarding  $\sigma$ , we found that s-SWNT-F2 showed a much higher  $\sigma$  than s-SWNT-F1 ( $3.15 \times 10^2 \text{ S m}^{-1}$ ) and s-SWNT-F3 ( $1.43 \times 10^4 \text{ S m}^{-1}$ ). The presence of FC12 in s-SWNT-F1 and s-SWNT-F3 might hamper the charge carrier transport in these sheets, as previously reported,<sup>5, 6, 35</sup> which caused lower  $\sigma$  for s-SWNT-F1 and s-SWNT-F3. It is also noted that s-SWNT-F1 had much lower  $\sigma$  than s-SWNT-F3. Considering the sheet fabrication process, we assumed that for s-SWNT-F1, FC12 existed not only on the surface of the bundled SWNT network but also inside the network junctions, while for s-SWNT-F3, FC12 was attached mainly on the SWNT bundle surface. The lower  $\sigma$  for s-SWNT-F3 than s-SWNT-F2 again manifested the importance of the removal of the dispersant from the s-SWNT sheets. It is also worth mentioning that  $\sigma$  of s-SWNT-F2 was almost comparable to that of unsorted SWNT ( $2.06 \times 10^4 \text{ S m}^{-1}$ ). Other sorting processes typically lead lowering of the  $\sigma$  due to the lowering of m-SWNT content and shortening of the tubes.<sup>36, 37</sup> This result suggests that FC12-based sorting is able to conserve the structure of s-SWNTs and high  $\sigma$ , which is supported by the

small Raman D band for s-SWNT-F2 compared to that observed for unsorted SWNT (Fig. 3b). Importantly, s-SWNT-F2 achieved the highest  $PF$  among the four sheets because of the increases of  $S$  induced by s-SWNT sorting and the increases of  $\sigma$  caused by the removal of FC12.

$\kappa$  was also measured for these samples (Fig. 6) using a periodic heating method. The four samples possessed almost comparable  $\kappa$  values (14.8–21.0  $\text{W m}^{-1} \text{K}^{-1}$ ).<sup>38</sup> In our previous research, we experimentally studied the effect of s-SWNT purity on  $\kappa$  and found that  $\kappa$  was not sensitive to the s-SWNT purity,<sup>37</sup> which agrees well with the small difference of  $\kappa$  between unsorted SWNT and sorted s-SWNT sheets (Fig. 6). In s-SWNTs, heat is transported mainly by phonons, and therefore  $\sigma$  and  $\kappa$  are only weakly related,<sup>11, 36</sup> which is one of the attractive features of s-SWNTs for TE applications. In addition, the above results also indicated that, different from  $\sigma$ , the removal of dispersant did not strongly affect  $\kappa$ .<sup>5</sup>

Based on the above TE parameters,  $zT$  values were calculated for the sheets (Fig. 6). Most importantly, we found that s-SWNT-F2 showed the highest  $zT$  value of  $1.27 \times 10^{-3}$ , which was two orders of magnitude higher than that of s-SWNT-F1 ( $1.38 \times 10^{-5}$ ). The increase of  $zT$  was attributed to the removal of the dispersant, as reported previously. It is also important to note that  $zT$  of s-SWNT-F2 was 2.6 times higher than that of unsorted SWNT, clearly indicating the importance of s-SWNT sorting. As a non-doped s-SWNT sheet, the obtained  $zT$  value ( $1.27 \times 10^{-3}$ ) is comparable to the other non-doped s-SWNT sorted by different method ( $1.91 \times 10^{-3}$ , Table S2, ES†).<sup>37</sup> Further improvement of the  $zT$  value is possible by doping of s-SWNT sheet as listed in Table S2 (ES†).

This time, we measured the  $^1\text{H}$  NMR spectrum of the FC12 obtained after the s-SWNT extraction process, which was collected from the filtrate upon washing the s-SWNTs (Fig. S3, ES†). The spectrum was identical to that of as-synthesized FC12, strongly suggesting that FC12 structure was unchanged and can be recycled without further treatments. Overall, FC12-based method for s-SWNT extraction is advantageous not only because of its simplicity but also from economic and environmental perspectives.

In summary, we prepared s-SWNT sheets using s-SWNT dispersions extracted from SWNTs using FC12 as a sorting dispersant. FC12 was removed simply by dipping the obtained

s-SWNT sheet in  $\text{CH}_2\text{Cl}_2$  to provide an FC12-free s-SWNT sheet. The TE properties of the s-SWNT sheets with and without FC12 removal including  $S$ ,  $\sigma$ , and  $\kappa$  were evaluated and found that  $PF$  and  $zT$  dramatically increased when FC12 was fully removed, which mainly because the increase of  $\sigma$ . Our method to obtain s-SWNT sheets free from the extraction dispersant is much simpler than previous polymer-based extraction methods. Because of the possible recyclability of FC12, the present method is attractive for large-scale synthesis of dispersant-free s-SWNT TE materials.

### Conflicts of interest

There are no conflicts to declare.

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