Supplementary Information (SI) for ChemComm.

Supporting Information

Exploration of metal-ionic liquids for enhancing electron conductivity and thermoelectric properties of single-walled carbon nanotube composites

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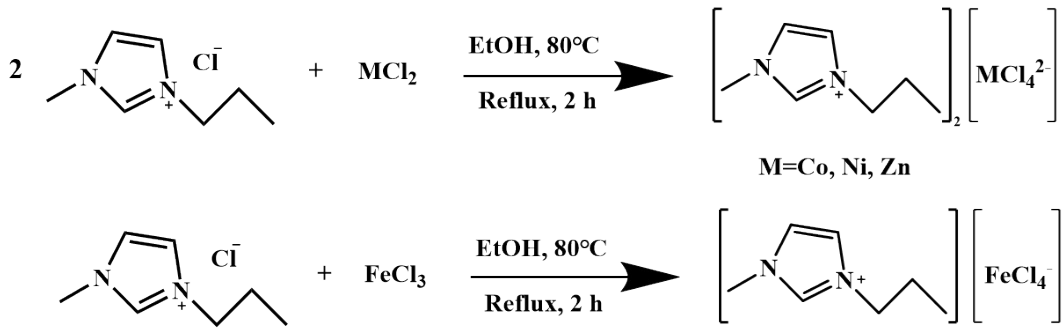

Fig. S1 Synthesis route of four metal-ionic liquids.

The synthesis route for metal ionic liquids (MILs) is illustrated using CoIL as an example: 1-methyl-3-propylimidazolium chloride ([C_3-mim]Cl, 1.6 g, 10 mmol) and $CoCl_2$ (0.65 g, 5 mmol) were added to a 100 mL three-neck flask, followed by the addition of 20 mL anhydrous ethanol. Argon gas was continuously purged through the mixture for 5 min to remove oxygen, after which the reaction was carried out under reflux at 80 ℃ for 2 h. Upon completion of the reaction, the mixture was allowed to stand until phase separation occurred. The supernatant was decanted, and the washing step with anhydrous ethanol was repeated three times to remove unreacted starting materials. The residual ethanol solvent was then removed by rotary evaporation, and the product was dried under vacuum at 80 ℃ for 6 h, yielding the blue viscous liquid [C_3-mim]$_2$[$CoCl_4$] (CoIL). Using the same procedure, [C_3-mim]$_2$[$NiCl_4$] (NiIL, blue), [C_3-mim]$_2$[$ZnCl_4$] (ZnIL, white), and [C_3-mim][$FeCl_4$] (FeIL, red) were synthesized. All resulting products were viscous liquids at room temperature.

[C_3-mim]Cl

^{1}H NMR (400 MHz, Deuterium Oxide) δ 8.66 (s, 1H), 7.40 (dd, J = 17.9, 1.9 Hz, 2H), 4.10 (t, J = 7.0 Hz, 2H), 3.84 (s, 3H), 1.82 (q, J = 7.3 Hz, 2H), 0.84 (t, J = 7.4 Hz, 3H).

[C_3-mim]$_2$[$ZnCl_4$]

^{1}H NMR (400 MHz, Deuterium Oxide) δ 8.63 (s, 1H), 7.37 (dt, J = 17.8, 2.1 Hz, 2H), 4.07 (t, J = 7.0 Hz, 2H), 3.81 (s, 3H), 1.79 (hd, J = 7.3, 1.0 Hz, 2H), 0.82 (td, J = 7.4, 0.9 Hz, 3H).

[C_3-mim]$_2$[$NiCl_4$]

^{1}H NMR (400 MHz, Deuterium Oxide) δ 8.57 (s, 1H), 7.32 (d, J = 17.4 Hz, 2H), 4.02 (s, 2H), 3.76 (s, 3H), 1.74 (s, 2H), 0.77 (s, 3H).

[C₃-mim]₂[CoCl₄]

^{1}H NMR (400 MHz, Deuterium Oxide) δ 7.07 (s, 1H), 5.82 (d, J = 22.3 Hz, 2H), 2.53 (s, 2H), 2.25 (s, 3H), 0.28 (d, J = 7.2 Hz, 2H), -0.67 (d, J = 7.4 Hz, 3H).

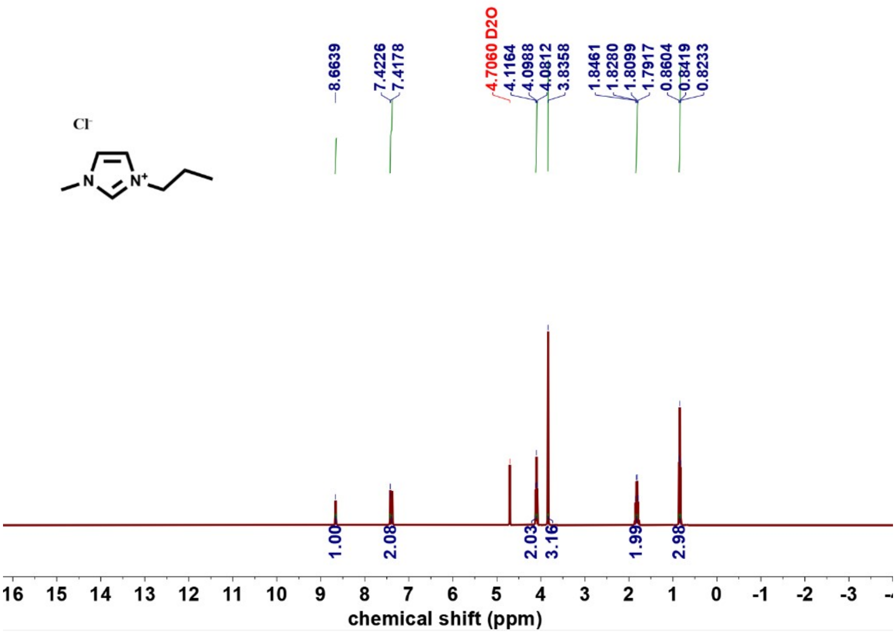

Figure S2. ^{1}H NMR spectrum of [C_3-mim]Cl (Deuterium Oxide)

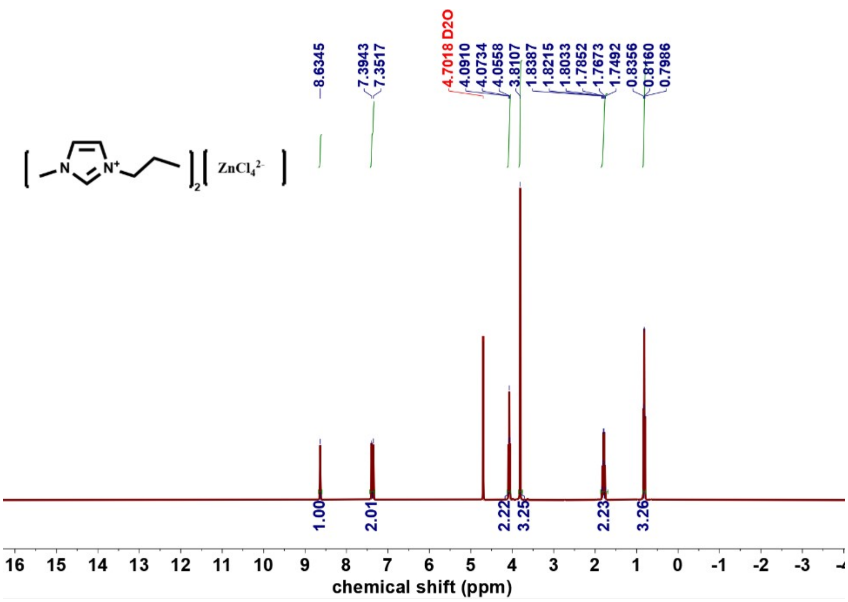

Figure S3. ^{1}H NMR spectrum of [C_3-mim]$_2$[$ZnCl_4$] (Deuterium Oxide)

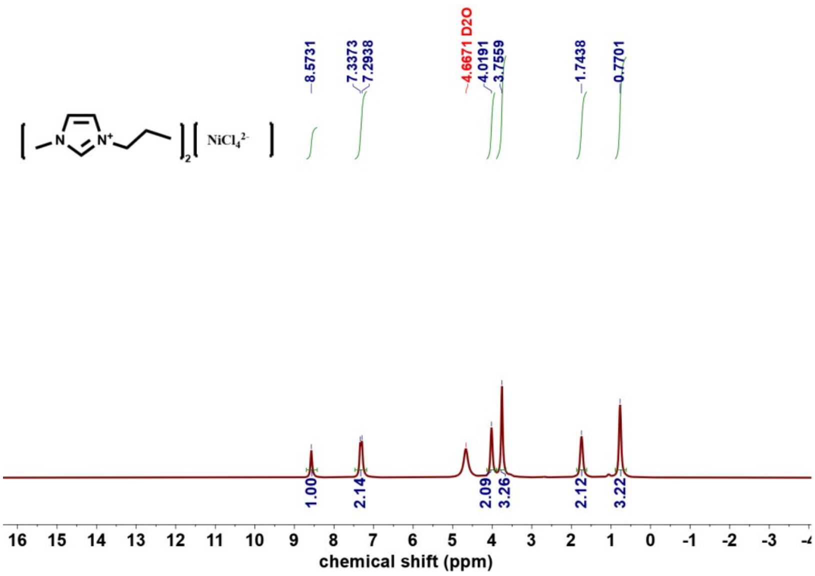

Figure S4. ^{1}H NMR spectrum of $[C_3\text{-mim}]_2[NiCl_4]$ (Deuterium Oxide)

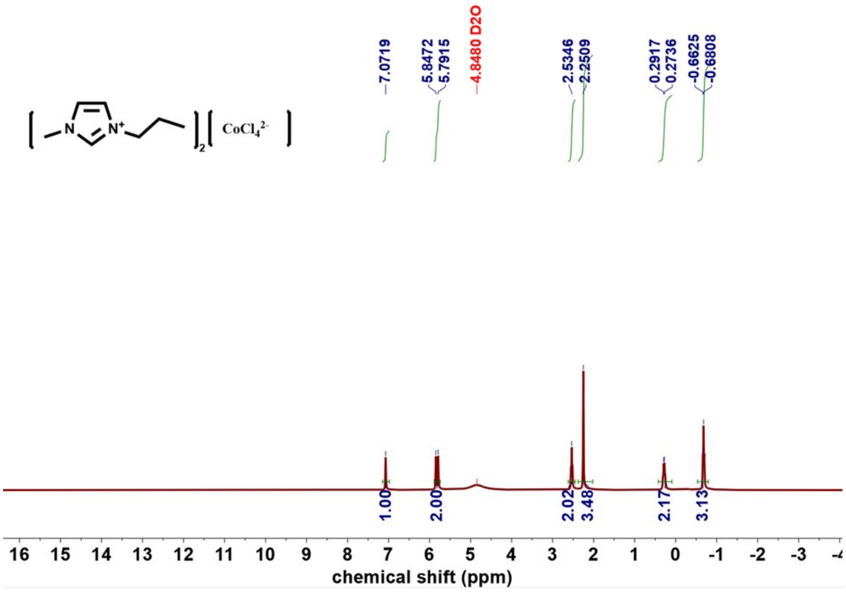

Figure S5. ^{1}H NMR spectrum of $[C_3\text{-mim}]_2[CoCl_4]$ (Deuterium Oxide)

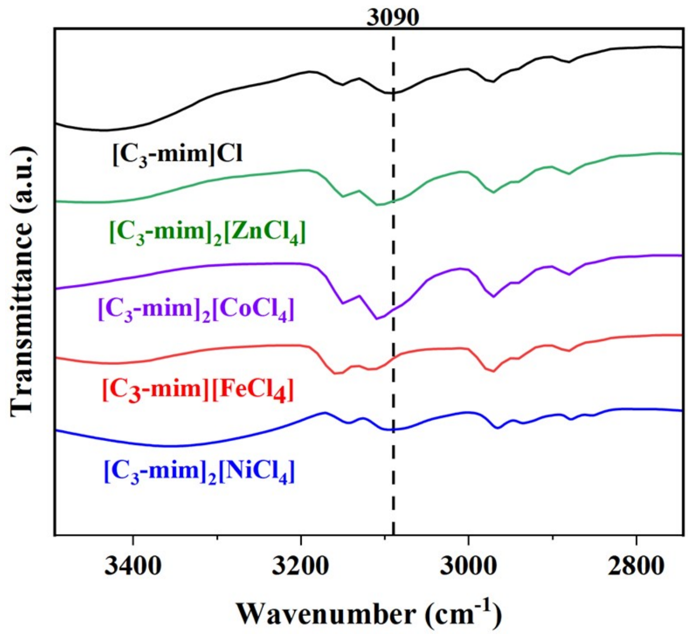

Figure S6. FTIR spectra of [MPIm]Cl and MILs magnified C–H stretching region (2700–3500 cm^{-1})

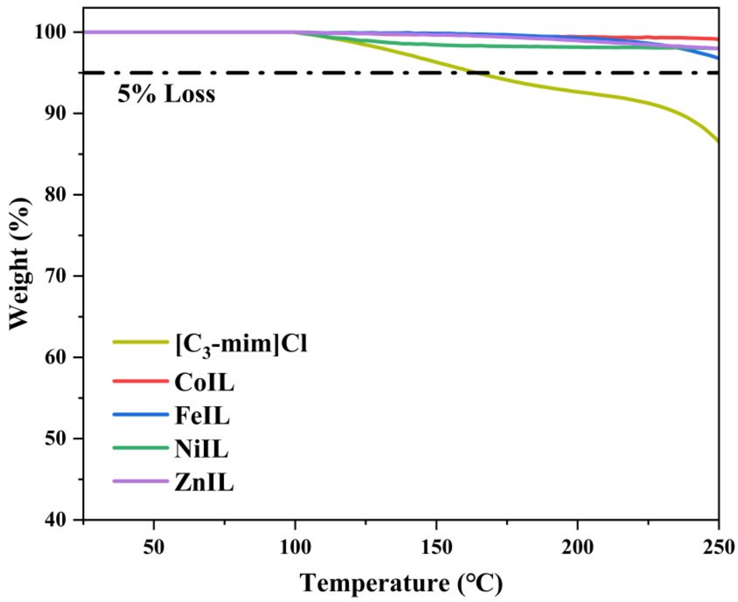

Fig. S7 TGA curves of [C3-mim]Cl、CoIL、FeIL、NiIL and ZnIL.

Table S1 The elemental analysis of metal ionic liquids.

Molecular Formula	C%（Calculated Value）	H%（Calculated Value）	N%（Calculated Value）
$[C_3\text{-mim}]_2CoCl_4$	37.36(37.27)	5.90(5.82)	12.44(12.42)
$[C_3\text{-mim}]_2NiCl_4$	37.31(37.29)	5.76(5.82)	12.40(12.43)
$[C_3\text{-mim}]\ FeCl_4$	26.01(26.04)	4.02(4.07)	8.77(8.68)
$[C_3\text{-mim}]_2ZnCl_4$	36.81(36.75)	5.68(5.74)	12.32(12.25)

Table S2 Hall test of pristine SWCNT, SWCNT/CoIL, SWCNT/FeIL, SWCNT/NiIL and SWCNT/ZnIL composite films .

Films	Carrier type	Carrier concentration(cm^{-3})	Carrier mobility ($cm^2\ V^{-1}\ s^{-1}$)
SWCNT	p	4.45×10^{21}	2.45
SWCNT/CoIL	p	6.97×10^{21}	4.18
SWCNT/FeIL	p	7.25×10^{21}	4.33
SWCNT/NiIL	p	6.89×10^{21}	4.02
SWCNT/ZnIL	p	4.75×10^{21}	3.65

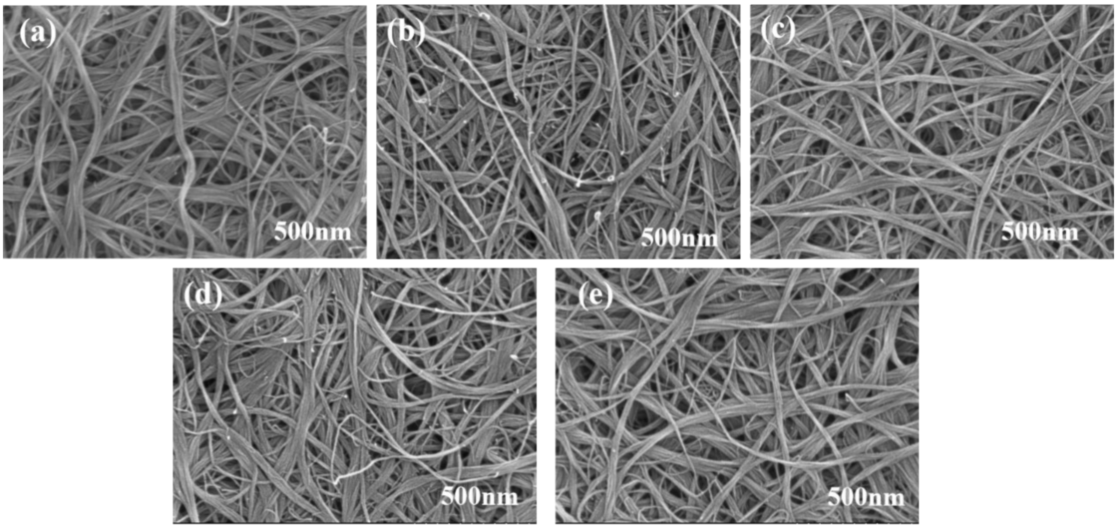

Fig. S8 Typical SEM images of (a) pristine SWCNT, (b) SWCNT/CoIL，(c) SWCNT/FeIL, (d) SWCNT/NiIL and (e) SWCNT/ZnIL.

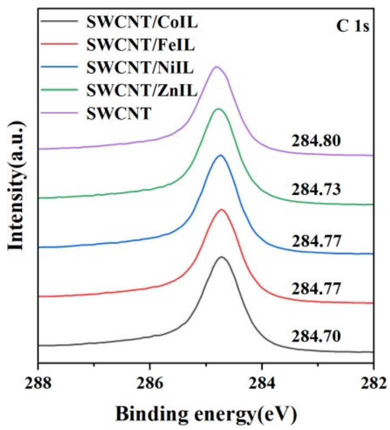

Fig. S9 C 1s XPS spectra of of pristine SWCNTand SWCNT/MIL composite films.

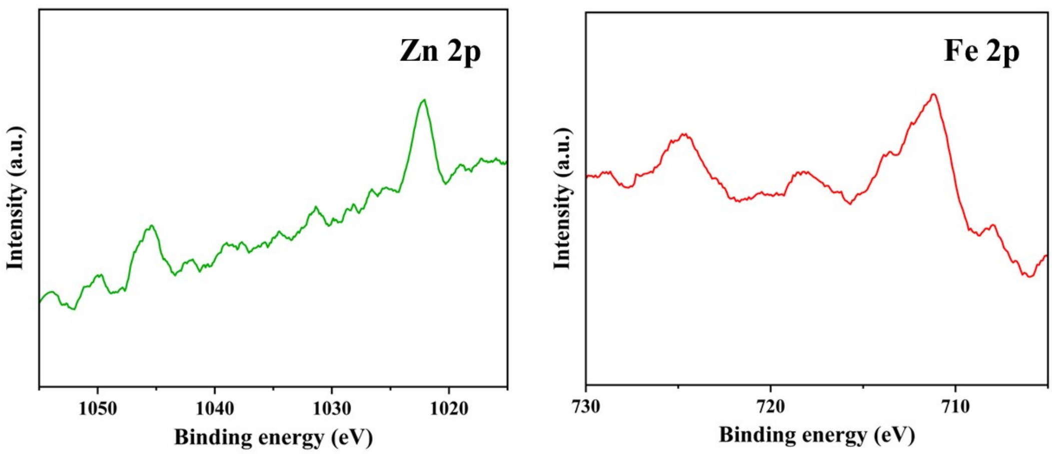

Fig. S10 The Zn 2p XPS spectra of the SWCNT/ZnIL composite film and the Fe 2p XPS spectra of the SWCNT/FeIL composite film.