

The Role of Fluorinated IL as an Interfacial Agent in P(VDF-CTFE)/Graphene Composite Films

Jing Yang, Sébastien Pruvost *, Sébastien Livi and Jannick Duchet-Rumeau *

Laboratory of polymer materials Engineering, UMR CNRS 5223, INSA Lyon, University of Lyon, 69621 Villeurbanne, France

* Correspondence: sebastien.pruvost@insa-lyon.fr (S.P.); jannick.duchet@insa-lyon.fr (J.D.-R.);

Tel.:+33-04-72438291 (S.P.); +33-04-72438548 (J.D.-R.)

1. No residual DMF in the resulting composite films

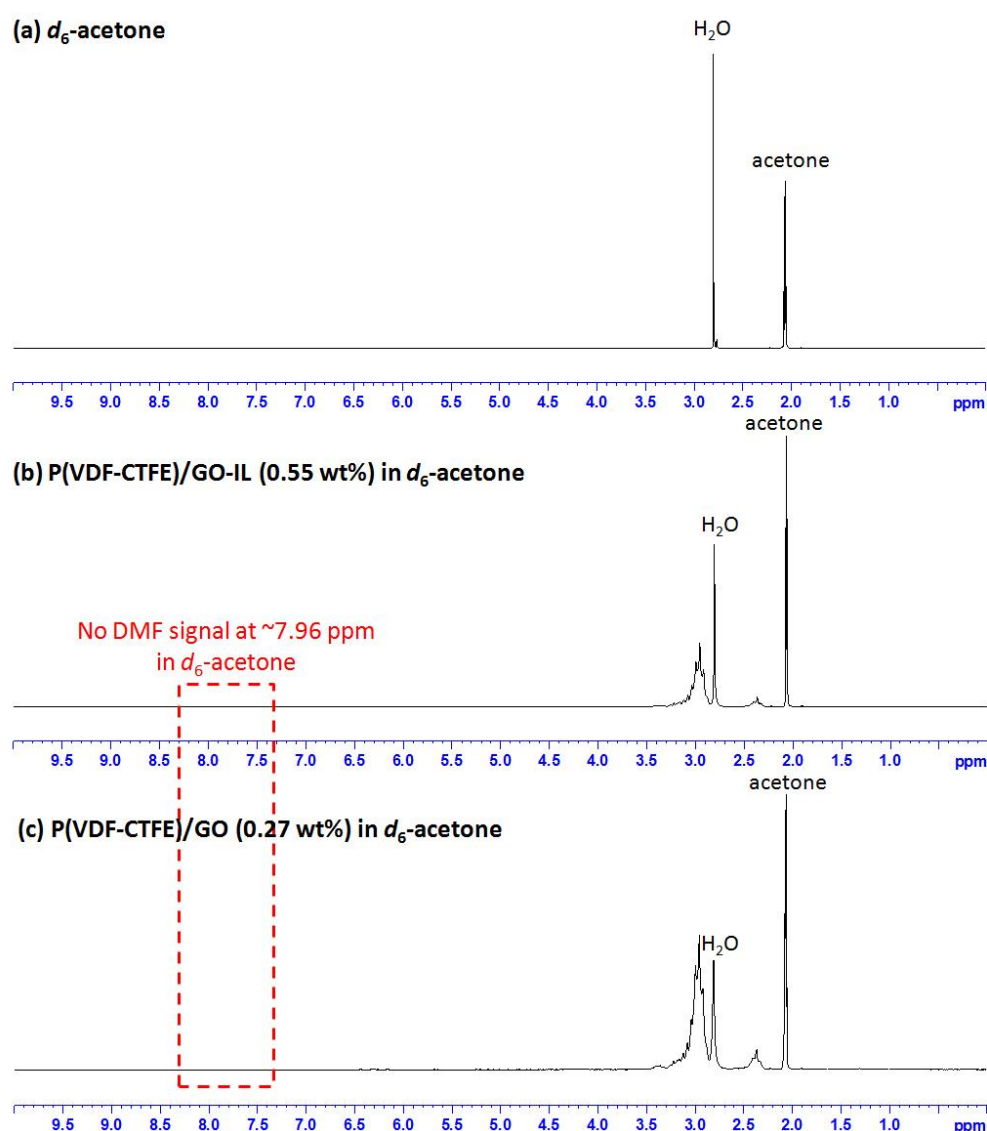
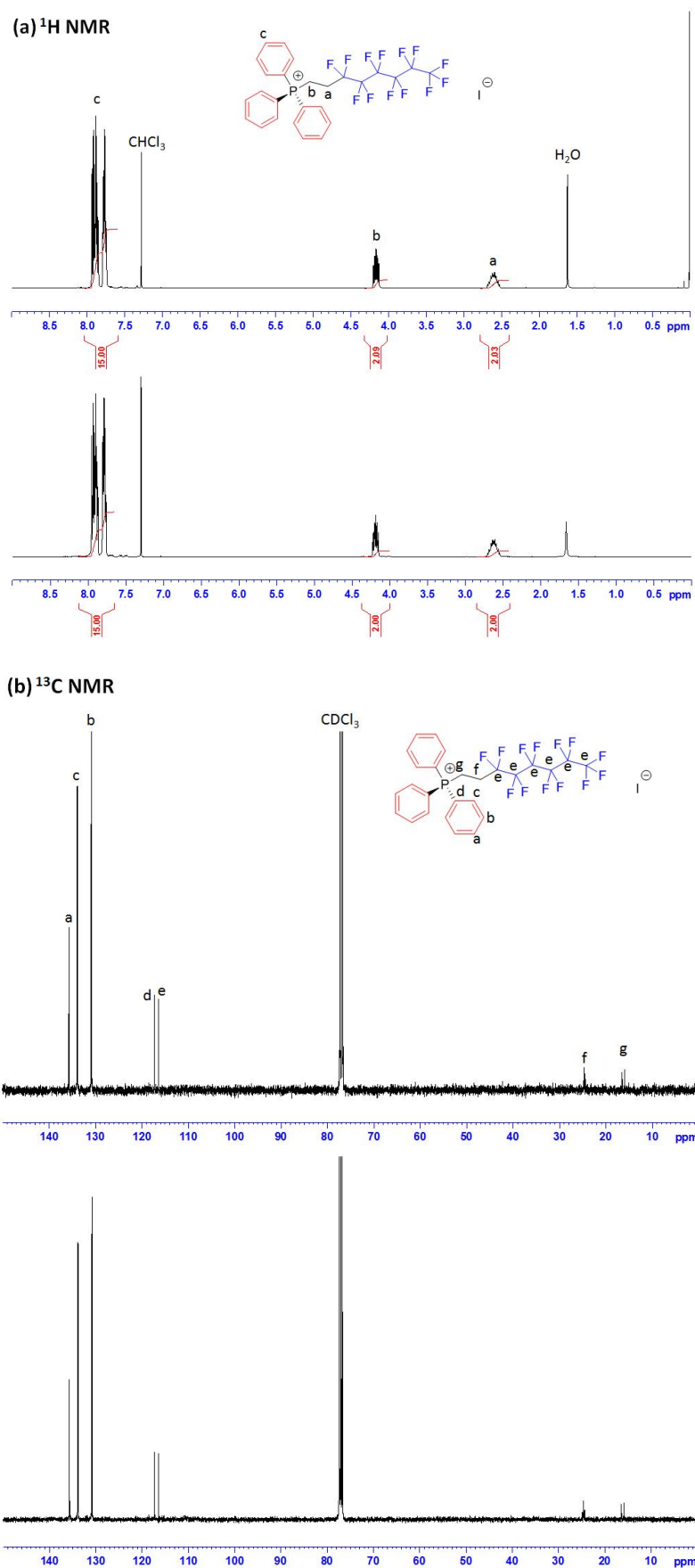


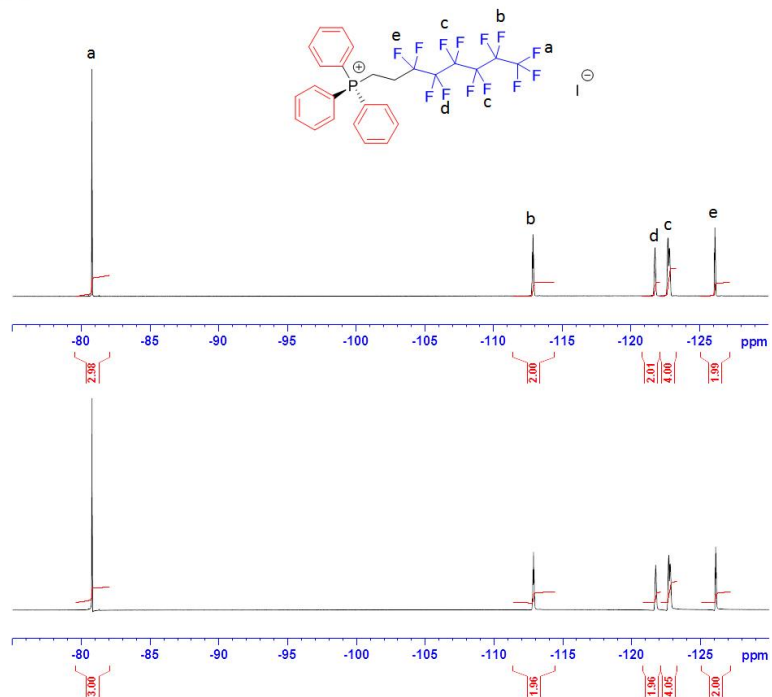
Figure S1. ^1H NMR spectra of deuterated acetone (d_6 -acetone) solvent (a), P(VDF-CTFE)/GO-IL (0.55 wt%) (b) and P(VDF-CTFE)/GO (0.27 wt%) composites in d_6 -acetone. Note that the signal of DMF in d_6 -acetone at around 7.96 ppm corresponding to $(\text{CH}_3)_2\text{N}-\text{C}(\text{O})\text{H}$ (*J. Org. Chem.* **1997**, 62, 7512-7515) is not visible in the spectra of P(VDF-CTFE)/GO-IL and P(VDF-CTFE)/GO composites, suggesting total removing of DMF by using the drying conditions: first dried at 60 °C in air oven overnight, and then under vacuum at 80 °C for 48 h.

2. Stability of IL-C8F13 during the chemical reduction by hydrazine

The IL-C8F13 is chemically stable under the reduction conditions by hydrazine (95 °C for 3 h in DMF), which can be confirmed by NMR spectra of IL before and after hydrazine treatment.



(c) ^{19}F NMR



(d) ^{31}P NMR

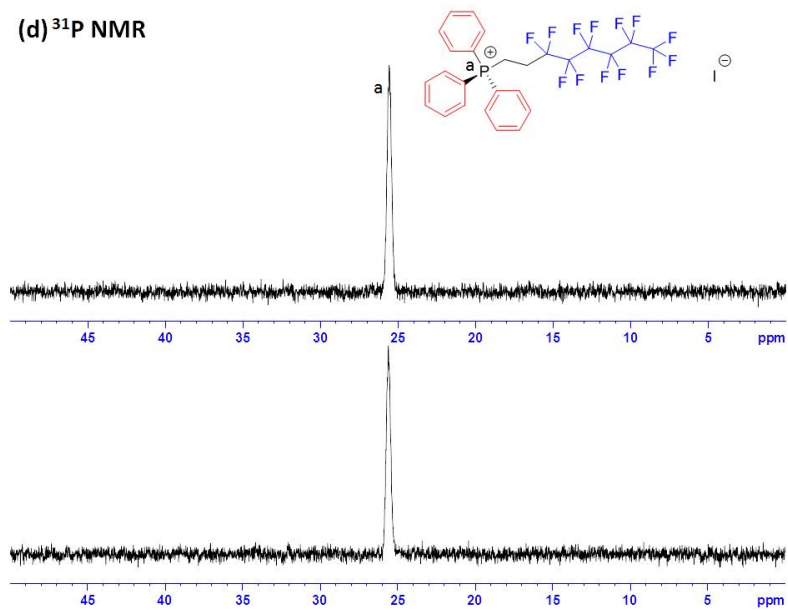
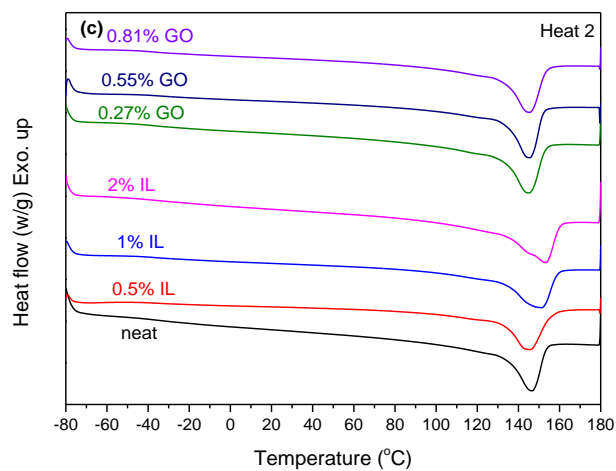
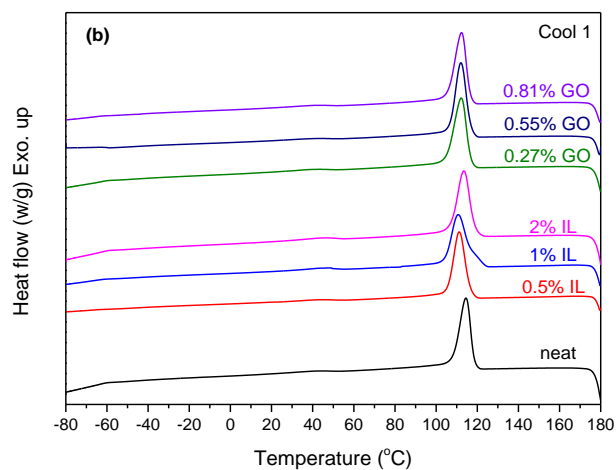
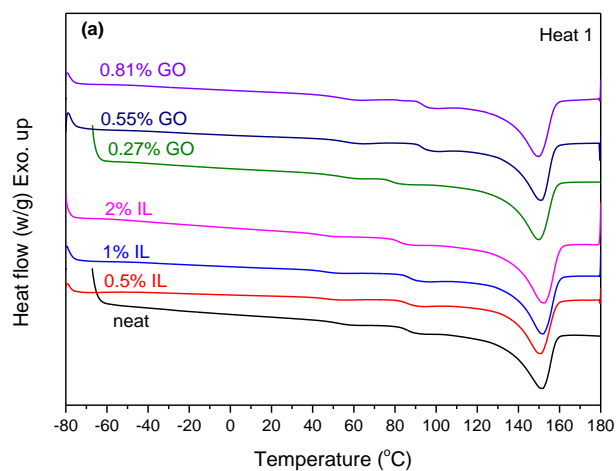
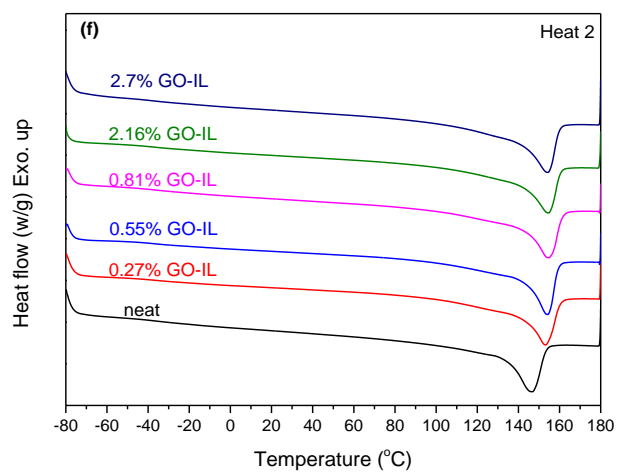
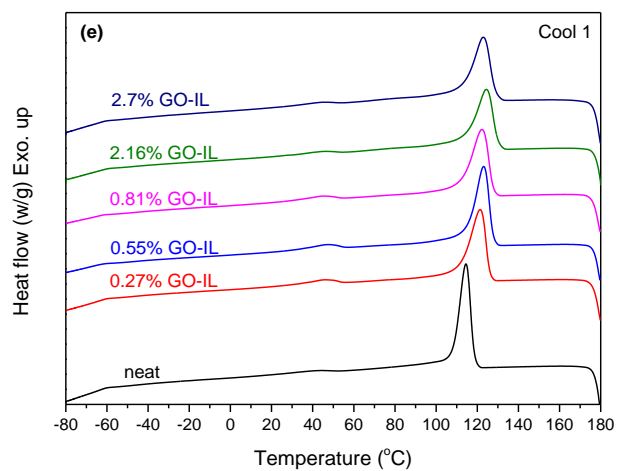
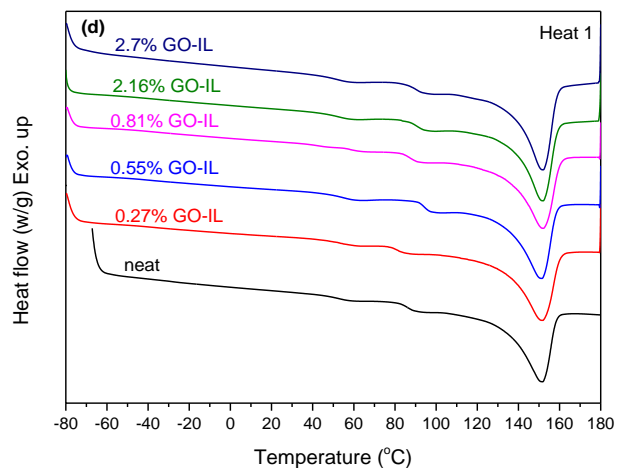


Figure S2. ^1H , ^{13}C , ^{19}F and ^{31}P NMR spectra of IL-C8F13 before (upper) and after (lower) hydrazine treatment.

3. DSC traces of neat P(VDF-CTFE) and its composites containing IL, GO, rGO, GO-IL, rGO-IL with different loading contents during heat/cool/heat runs





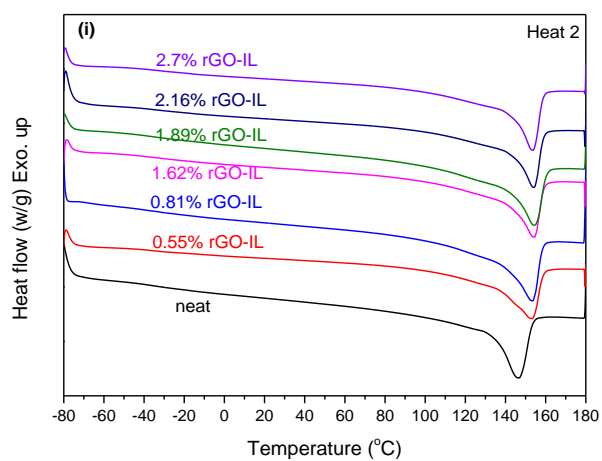
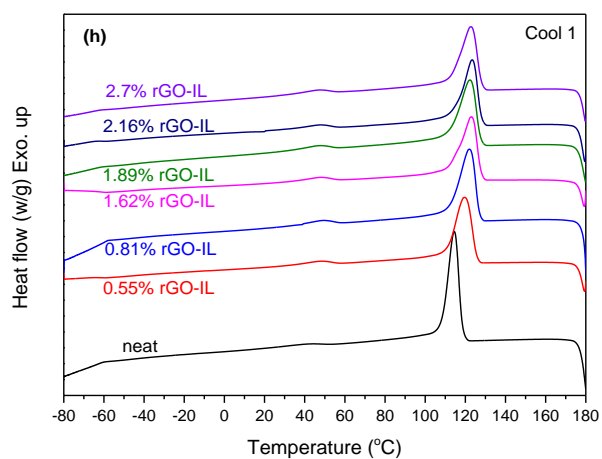
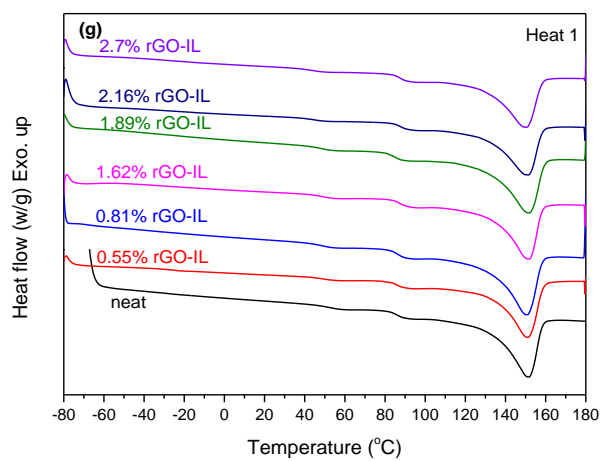


Figure S3. DSC thermograms of neat P(VDF-CTFE) and its composites containing GO, GO-IL and rGO-IL during first and second heating run and the first cooling run.

Table S1. T_c , T_m and ΔH of neat P(VDF-CTFE) and composites from DSC thermograms.

Sample	Heating 1		Cooling 1	Heating 2	
	T_m (°C)	ΔH (J/g)	T_c (°C)	T_m (°C)	ΔH (J/g)
neat	152	32	114	147	30
0.5 wt% IL-C8F13	151	31	111	146	35
1 wt% IL-C8F13	152	34	111	151	35
2 wt% IL-C8F13	153	37	114	153	33
0.27 wt% GO	150	37	112	145	34
0.55 wt% GO	151	31	112	146	35
0.81 wt% GO	150	33	112	146	34
0.27 wt% GO-IL	152	37	121	153	35
0.55 wt% GO-IL	152	34	123	154	35
0.81 wt% GO-IL	152	37	122	155	34
2.16 wt% GO-IL	152	37	124	155	32
2.70 wt% GO-IL	152	40	123	154	34
0.55 wt% rGO-IL	151	33	120	153	32
0.81 wt% rGO-IL	151	35	122	153	32
1.62 wt% rGO-IL	152	32	123	155	30
1.89 wt% rGO-IL	152	35	122	155	30
2.16 wt% rGO-IL	152	30	123	154	30
2.70 wt% rGO-IL	151	31	123	154	30

4. DMA curves of neat P(VDF-CTFE) and its composites containing IL, GO, GO-IL and rGO-IL

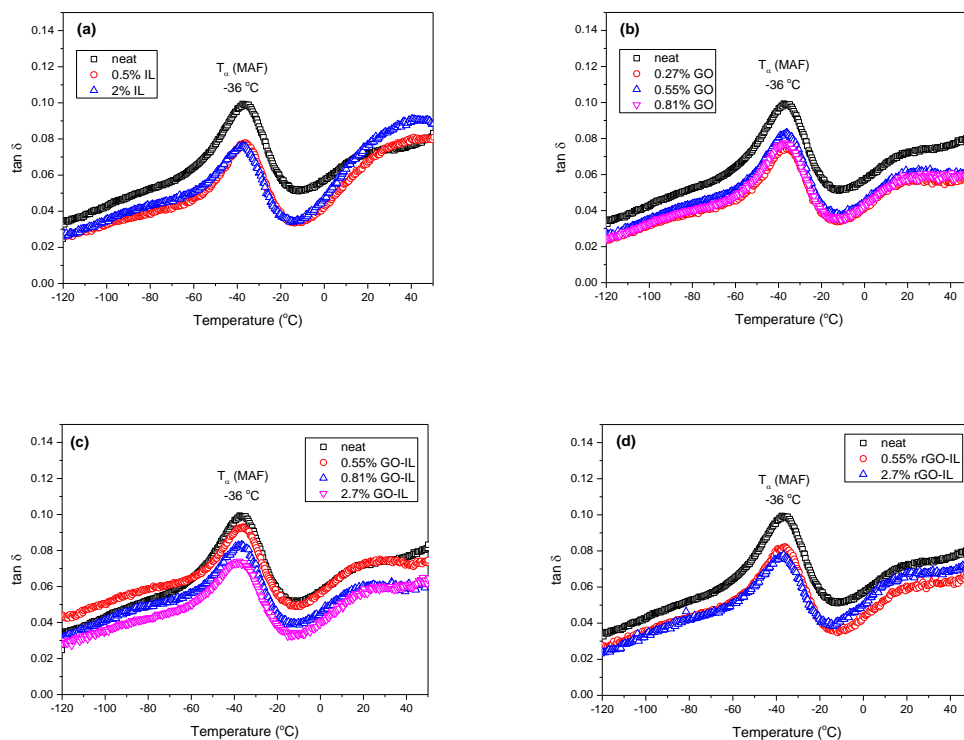
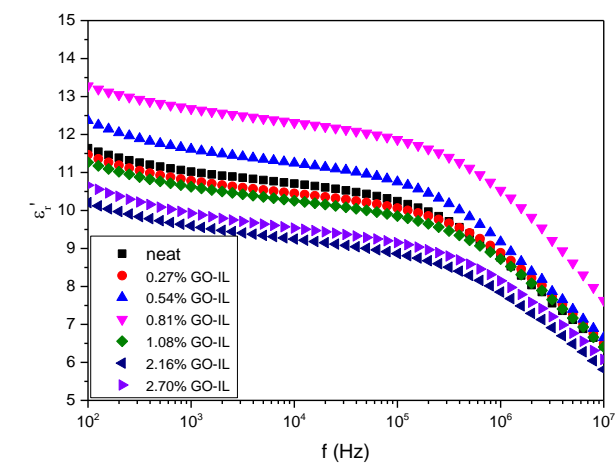
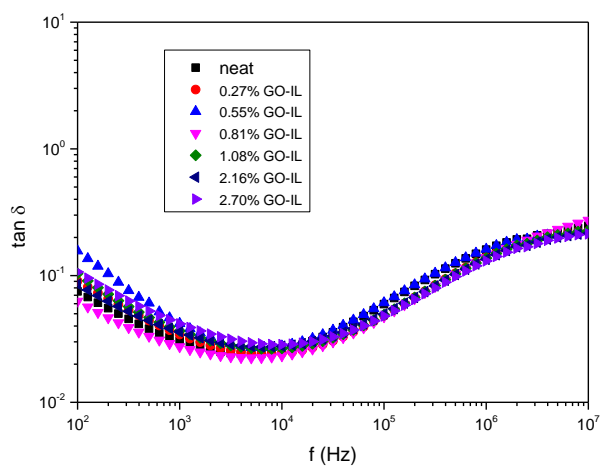


Figure S4. Dynamic mechanical loss tangent ($\tan \delta$) as a function of temperature for neat P(VDF-CTFE) and composites containing IL (a), GO (b), GO-IL (c) and rGO-IL (d).

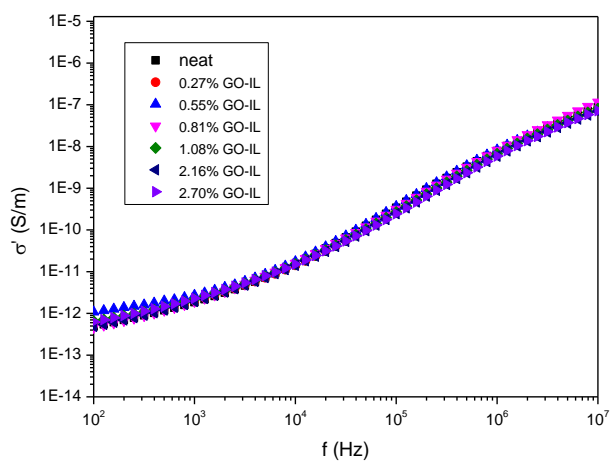
5. Dielectric properties of P(VDF-CTFE)/GO-IL composites



(a)



(b)



(c)

Figure S5. Relative dielectric permittivity (ϵ') (a), loss tangent ($\tan \delta$) (b) and AC conductivity (σ') (c) of P(VDF-CTFE)/GO-IL composites as a function of frequency at RT with different compositions.