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Supplementary Information

High-*k* polymer materials containing cyclic carbonate as gate dielectrics for application in low-voltage operating organic thin-film transistors

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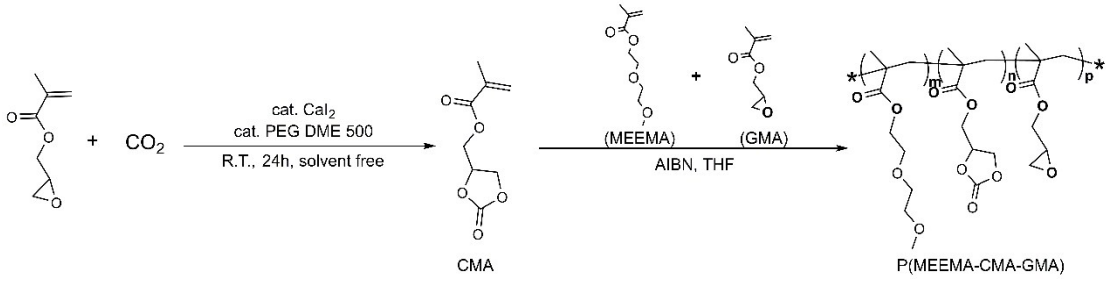
Materials

All materials such as glycidyl methacrylate (GMA), 2-(2-methoxyethoxy) ethyl methacrylate (MEEMA) and poly(ethylene glycol) dimethyl ether (PEG DME 500) were used as received. The CO_2 was used in a purity of 99.9%. The CaI_2 was used anhydrous with a purity of ≥ 99% and stored under N_2 conditions. 2,2-Azobisisobutyronitrile (AIBN) was recrystallized in ethanol before use. The tetrahydrofuran (THF), ethyl acetate (EA) and hexane were purified by distillation.



Scheme S1. The synthesis routes of P(MEEMA-CMA-GMA).

Synthesis of (2-oxo-1,3-dioxolan-4-yl)methyl methacrylate (CMA).

Compound CMA[1]: Glycidyl methacrylate (GMA) (28.43g, 200mmol) was added to a mixture of CaI_2 (2.94g, 10mmol) and poly(ethylene glycol) dimethyl ether (PEG DME 500) (5g, 10mmol) at

room temperature by bubbling CO_2 for 24 hours. The reaction mixture was filtered and column chromatography eluting with EA : hexane (1 : 5 v/v) was performed to purify the product obtained by removing the solvent under reduced pressure. The target compound was liquid of 27.94 g (75%). ^{1}H NMR (500 MHz, $CDCl_3$, δ, ppm): 6.15-6.17 (s, 1H), 5.66-5.68 (m, 1H), 4.95-5.0 (m, 1H), 4.56-4.60 (t, 1H), 4.41-4.46 (dd, 1H), 4.32-4.37 (m, 2H), 1.95-1.97 (s, 3H).

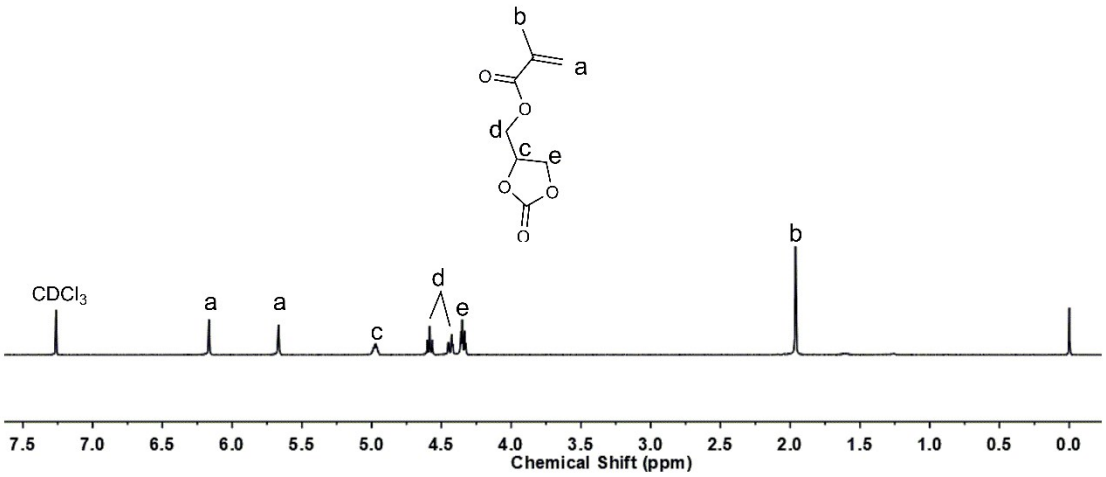


Fig. S1. The ^{1}H NMR spectrum of compound CMA ($CDCl_3$, 500 MHz).

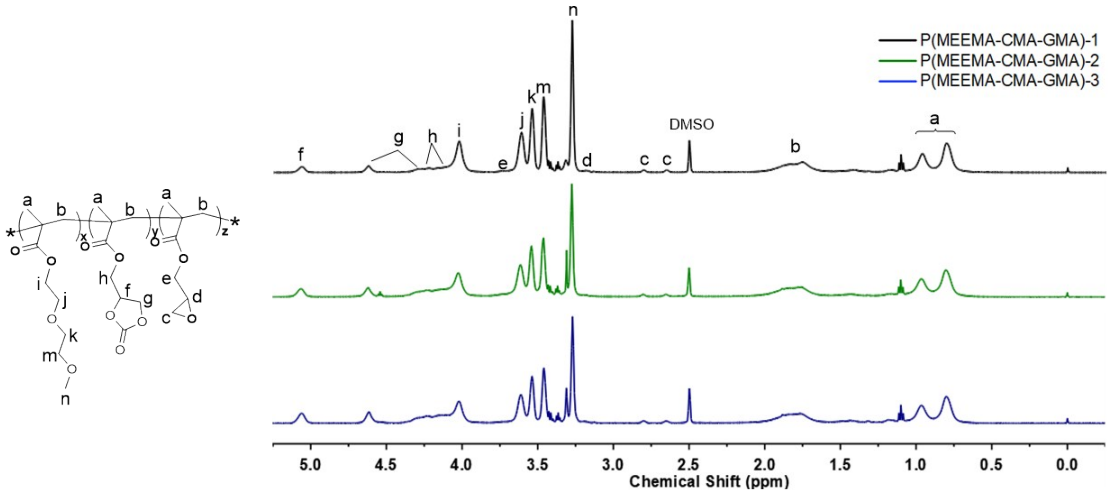


Fig. S2. The ^{1}H NMR spectra and structure of three copolymers ($CDCl_3$, 500 MHz).

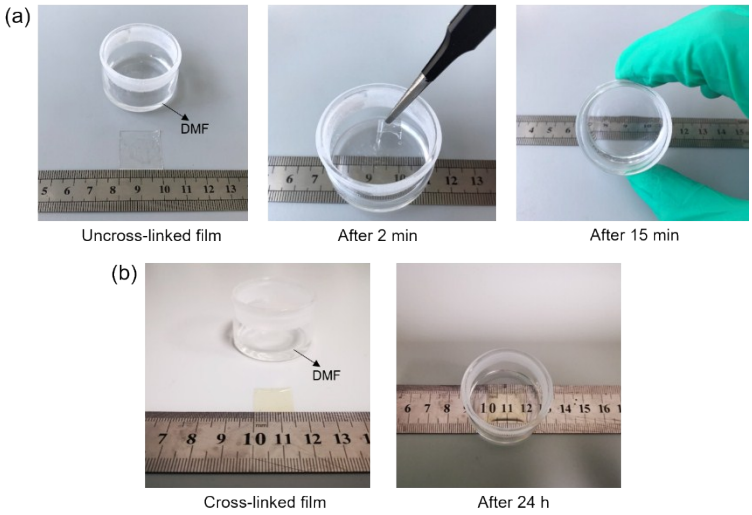


Fig. S3. Optical images of uncross-linked film (a) and cross-linked film (b) after soaking in DMF for different time period.

the flexibility of crosslinked film.MP4

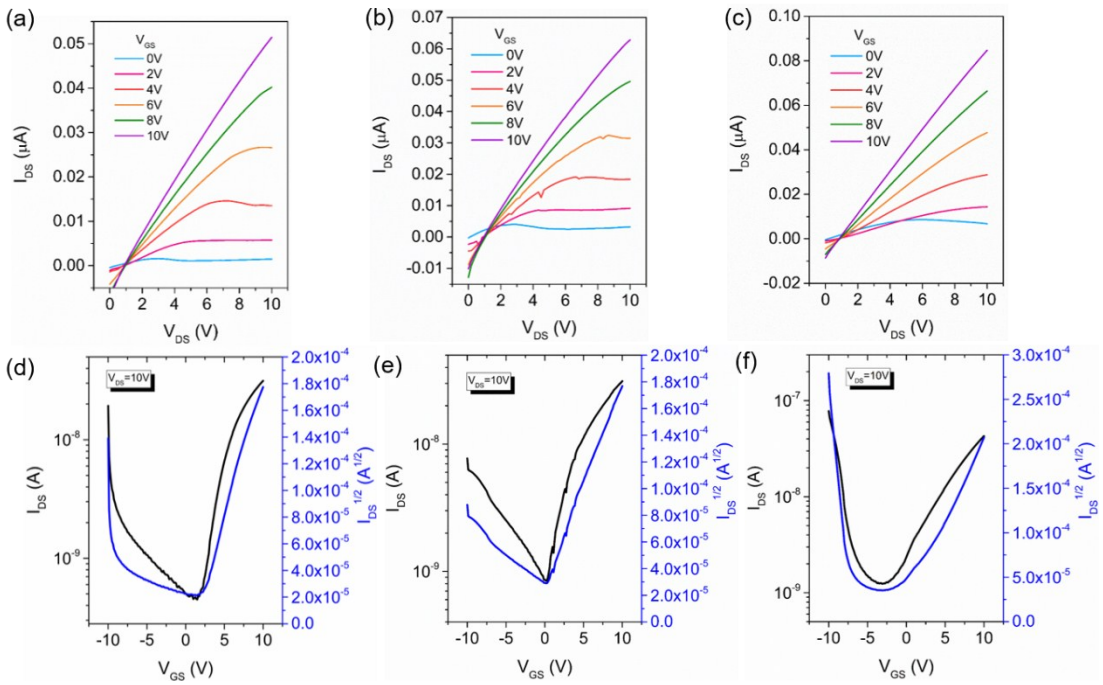


Fig. S4. (a), (b) and (c) Output characteristic curves and (d), (e) and (f) transfer characteristic curves of n-type F_{16}CuPc TFTs with these P(MEEMA-CMA-GMA) copolymers as the dielectric layers.

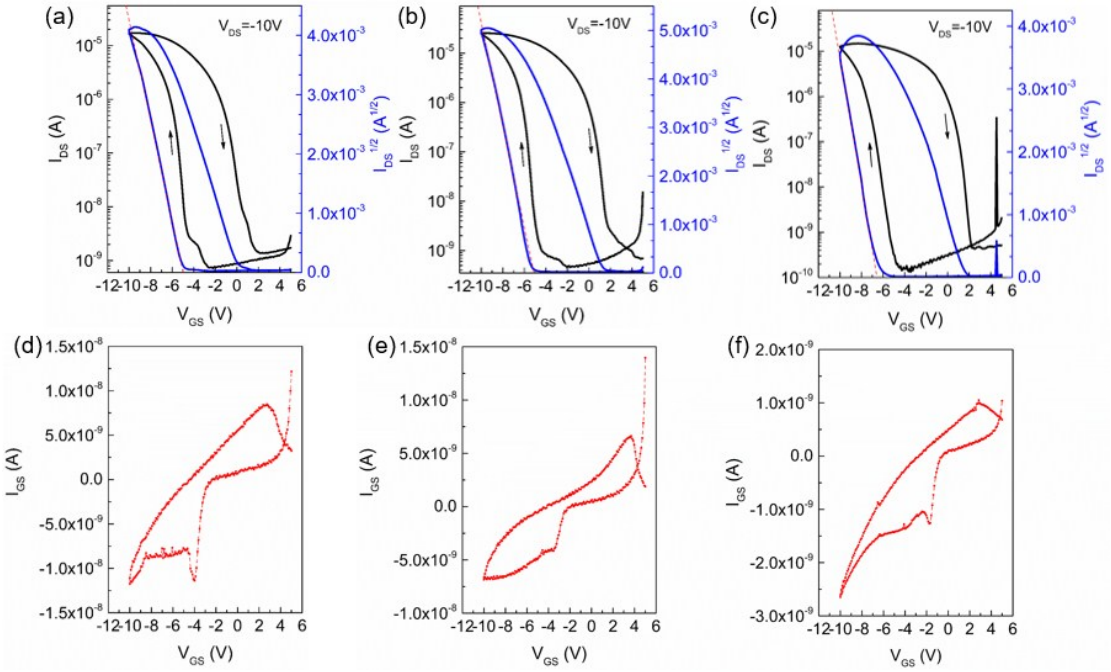


Fig. S5. (a), (b) and (c) transfer characteristic curves of p-type C_{10}-DNTT TFTs with these P(MEEMA-CMA-GMA) copolymers as the dielectric layers. (d), (e) and (f) gate leakage current curves of p-type C_{10}DNTT TFTs with these P(MEEMA-CMA-GMA) copolymers as the dielectric layers.

Table S1. The molar ratios of the monomers in the structures of P(MEEMA-CMA-GMA) copolymers

Name of copolymer	Comp. mol %		
	MEEMA	CMA	GMA
P(MEEMA-CMA-GMA)-1	13	4	1
P(MEEMA-CMA-GMA)-2	11	6	1
P(MEEMA-CMA-GMA)-3	10	7	1

Table S2. Transistor parameters for F_{16}CuPc based on P(MEEMA-CMA-GMA) copolymer dielectric layers

OS	Dielectric layer	Mobility[a] ($cm^2\ V^{-1}\ s^{-1}$)	On/off ratio[b]	Threshold voltage[c] (V)
F_{16}CuPc	P(MEEMA-CMA-GMA)-1	0.0051	2.22×10^2	2
	P(MEEMA-CMA-GMA)-2	0.0024	3.15×10^2	-1

P(MEEMA-CMA-GMA)-3	0.0028	1.93×10^1	-0.2

[a]Average field-effect mobility, [b]average on/off ratio and [c]average threshold voltage of 10 OTFTs.

REFERENCES

(1) Steinbauer, J.; Werner, T. Poly(ethylene glycol)s as Ligands in Calcium-Catalyzed Cyclic Carbonate Synthesis. *ChemSusChem* **2017,** *10,* 3025-3029.