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 CO₂ was used in a purity of 99.9%. The CaI₂ was used anhydrous with a purity of ≥ 99% and stored under N₂ 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 CMA1: Glycidyl methacrylate (GMA) (28.43g, 200mmol) was added to a mixture of CaI₂ (2.94g, 10mmol) and poly(ethylene glycol) dimethyl ether (PEG DME 500) (5g, 10mmol) at
room temperature by bubbling CO₂ 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, CDCl₃, 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 (CDCl₃, 500 MHz).

Fig. S2. The $^1$H NMR spectra and structure of three copolymers (CDCl₃, 500 MHz).
Fig. S3. Optical images of uncross-linked film (a) and cross-linked film (b) after soaking in DMF for different time period.

Fig. S4. (a), (b) and (c) Output characteristic curves and (d), (e) and (f) transfer characteristic curves of n-type F16CuPc TFTs with these P(MEEA-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

<table>
<thead>
<tr>
<th>Comp. mol %</th>
<th>MEEMA</th>
<th>CMA</th>
<th>GMA</th>
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<tbody>
<tr>
<td>P(MEEMA-CMA-GMA)-1</td>
<td>13</td>
<td>4</td>
<td>1</td>
</tr>
<tr>
<td>P(MEEMA-CMA-GMA)-2</td>
<td>11</td>
<td>6</td>
<td>1</td>
</tr>
<tr>
<td>P(MEEMA-CMA-GMA)-3</td>
<td>10</td>
<td>7</td>
<td>1</td>
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Table S2. Transistor parameters for F_{16}CuPc based on P(MEEMA-CMA-GMA) copolymer dielectric layers

<table>
<thead>
<tr>
<th>OS</th>
<th>Dielectric layer</th>
<th>Mobility ( \mu ) ( (\text{cm}^2 \text{V}^{-1} \text{s}^{-1}) )</th>
<th>On/off ratio ( I_{on}/I_{off} )</th>
<th>Threshold voltage ( V_{th} ) (V)</th>
</tr>
</thead>
<tbody>
<tr>
<td>F_{16}CuPc</td>
<td>P(MEEMA-CMA-GMA)-1</td>
<td>0.0051</td>
<td>( 2.22 \times 10^2 )</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td>P(MEEMA-CMA-GMA)-2</td>
<td>0.0024</td>
<td>( 3.15 \times 10^2 )</td>
<td>-1</td>
</tr>
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</table>
P(MEEMA-CMA-GMA)-3

0.0028          1.93 × 10⁻¹          -0.2

*Average field-effect mobility, *average on/off ratio and *average threshold voltage of 10 OTFTs.

REFERENCES