Supporting Information

Low-Voltage Organic Nonvolatile Memory Transistors with Water-Soluble Polymers Containing Thermally Induced Radical Dipoles

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Figure S1. Output (a) and transfer (b) characteristics (dark condition) for the OFETs with the PAMPSA layers according to the thermal treatment temperature (80-230 °C for 30 min). '*F*' and '*B*' denote forward and backward sweeps, respectively.



Figure S2. DSC thermogram for the PAMPSA sample. The scan rate was 5 °C/min. The measured glass transition temperature (Tg) (onset) was 116.8 °C.



Figure S3. Optical absorption spectra (normalized) for the PAMPSA films coated on quartz substrates according to the thermal treatment temperature (80–230 °C for 30 min).



Figure S4. GIWAXS results for the PAMPSA layers coated on the ITO-glass substrates: (a) 2D GIWAXS images, (b) 1D GIWAXS profiles (top: OOP direction; bottom: IP direction). The d-spacing (*d*) values for characteristic peaks are given on each graph in (b).



Figure S5. AFM images $(3 \ \mu m \times 3 \ \mu m)$ for the PAMPSA layers according to the thermal treatment temperature (80–230 °C for 30 min). Note that the root-mean-square roughness (Rg) was 0.165, 0.182, 0.195, 1.358, 0.356, and 0.240 nm at 80, 110, 140, 160, 170, and 230 °C, respectively. The thermal treatment time was 30 min for each case.



Figure S6. FT-IR spectra for the PAMPSA films according to the thermal treatment temperature (80–230 °C for 30 min). The major peaks are marked with the corresponding functional groups.



Figure S7. XPS spectra for the PAMPSA films according to the thermal treatment temperature: (a) Normalized spectra for the selected four temperatures (80, 160, 170, and 230 °C for 30 min), (b) normalized spectra for whole temperatures (80–230 °C for 30 min), and (c) raw spectra (before normalization) for the selected four temperatures (80, 160, 170, and 230 °C for 30 min).



Figure S8. ESR spectra for the PAMPSA films coated on polyimide film substrates according to the thermal treatment temperature. Note that the pronounced ESR signal was measured for the PAMPSA film treated at 170 °C even though no ESR signal was measured for the PAMPSA film treated at 150 °C.



Figure S9. Frequency (*f*)-dependent capacitance changes according to the thermal treatment temperature (applied voltage = 0 V): (a) MIM devices (glass/ITO/PAMPSA/Al), (b) MISM devices (glass/ITO/PAMPSA/P3HT/Al). Inset graphs show the variation of capacitance as a function of thermal treatment temperature at a selected frequency.



Figure S10. Frequency (*f*)-dependent capacitance changes according to the thermal treatment temperature (applied voltage = 3 V): (a) MIM devices (glass/ITO/PAMPSA/Al), (b) MISM devices (glass/ITO/PAMPSA/P3HT/Al). Inset graphs show the variation of capacitance as a function of thermal treatment temperature at a selected frequency.



Figure S11. STEM (atom mapping) images for the cross-section parts of the OFETs with the PAMPSA layers thermally treated at three different temperatures for 30 min: (a) 140 °C, (b) 170 °C, (c) 230 °C. No particular interactions could be observed from the interfaces between the P3HT and PAMPSA layers even though the interfacial region was investigated by atom mapping method focusing on three different atoms. It is worthy to note that the concentration of sulfur atoms was found to be noticeably reduced in the PAMPSA layer by thermal annealing at 230 °C, while the thickness of PAMPSA layers was decreased with the annealing temperature owing to the thermal degradation.



Figure S12. Capacitance as a function of applied voltage for the MISM devices (glass/ITO/PAMPSA/P3HT/Al) with the PAMPSA films treated at 170 °C (see arrows for scan direction). Note that the capacitance hysteresis became more pronounced at lower frequency than higher frequency.



Figure S13. Output and transfer characteristics (dark condition) for the OFETs with the PAMPSA layers treated at 170 °C for 30 min. The maximum sweep voltage (V_D and V_G): (a) –1 V; (b) –5 V. '*F*' and '*B*' denote forward and backward sweeps, respectively.



Figure S14. $I_D^{0.5} \sim V_G$ plot (from transfer curve in the dark) for the OFET with the PAMPSA layers treated at 170 °C for 30 min. '*F*', '*B*', 'V_{TH,F}', 'V_{TH,B}', and ' Δ V_{TH}' denote forward sweep direction, backward sweep direction, threshold voltage at forward sweep, threshold voltage at backward sweep, and threshold voltage shift between forward and backward sweeps, respectively.



Figure S15. (a) writing-once-reading-many (WORM) operation results (W: $V_G = -5$ V and $V_D = -5$ V; R: $V_G = -1$ V and $V_D = -5$ V), (b) writing-reading-reading (WRER) operation results (W: $V_G = -5$ V and $V_D = -5$ V; R1, R2: $V_G = -1$ V and $V_D = -5$ V; E: $V_G = +5$ V and $V_D = -1$ V), (c) retention characteristics during 11,630 WRER cycles with the same operation conditions as used for (b).



Figure S16. Transfer curves for the OFETs with the PAMPSA layers treated at 170 °C for 30 min: (a) Writing (W) condition, (b) reading-1 (R1) and reading-2 (R2) conditions, and (c) erasing (E) condition. 'F' and 'B' denote forward and backward sweep directions, respectively.