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## **Supporting Information**

A Flexible Memory with Low-Voltage and High-Operation Speed Using an Al<sub>2</sub>O<sub>3</sub>/Poly(α-methylstyrene) Gate Stack on Muscovite Substrate

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**Fig. S1.** Typical I<sub>DS</sub>-V<sub>GS</sub> characteristics for various V<sub>GS</sub> sweeping ranges for OFET memory devices with various P $\alpha$ MS thicknesses: (a) 8 nm; (b) 10 nm; (c) 12 nm; (d) 14 nm; (e) 18 nm; (f) 20 nm. The Al<sub>2</sub>O<sub>3</sub> is 20 nm thick and V<sub>DS</sub> = -3 V.



Fig. S2. Typical  $I_{DS}$ - $V_{GS}$  characteristics at different  $V_{GS}$  sweeping ranges for OFET memory devices with various  $Al_2O_3$  thicknesses: (a) 10 nm; (b) 20 nm; (c) 30 nm; (d) 40 nm. PaMS is 10 nm thick and  $V_{DS} = -3$  V.



Fig. S3. Gate leakage current Characteristics of OFET memory devices with different thickness P $\alpha$ MS films: (a) 8 nm, (b) 10 nm. AFM surface topographies of pentacene film on different thickness P $\alpha$ MS films: (c) 8 nm, (d) 10 nm.



**Fig. S4.** Typical I<sub>DS</sub>-V<sub>GS</sub> curves for reading out V<sub>th</sub> values for OFET memory with Al<sub>2</sub>O<sub>3</sub>(20 nm)/ P $\alpha$ MS (10 nm) gate stack: (a) pulse height is 6 V; (b) pulse height is 7 V; (c) pulse height is 8 V; (d) pulse width is 100  $\mu$ s. V<sub>DS</sub> = -3 V for all measurements.



**Fig. S5.**  $V_{th}$  for various samples with different channel lengths after the +6 V, 100 µs program, -4 V, 100 µs erase. Each device has an Al<sub>2</sub>O<sub>3</sub> (20 nm)/ PaMS (10 nm) gate stack.



**Fig. S6.** Comprehensive comparison of operating voltage, operating speed, and endurance of proposed device with that of the representative flexible flash-type OTFT memory devices reported in recent years. The references here are from the main text.



Fig. S7. Electrical properties of  $Al_2O_3$  films fabricated at various temperatures. (a) Dielectric constant as a function of frequency. (b) Leakage current as a function of electric field.



Fig. S8. (a) Band alignments of the  $Al_2O_3$  dielectric with different deposition temperatures from the XPS;(b) XPS valence band spectra of  $Al_2O_3/Si$  with different deposition temperatures; (c) XPS O 1s electron energy loss spectra of  $Al_2O_3/Si$  with different deposition temperatures



**Fig. S9.**  $I_{DS}$ - $V_{GS}$  hysteresis characteristics of OFET memory devices with  $Al_2O_3$  films fabricated at various temperatures: (a) 80 °C; (b) 200 °C; (c) 300 °C. The memory performance is lower for  $Al_2O_3$  devices deposited at lower temperatures.



**Fig. S10.** Characteristics of gate leakage current of OFET memory devices with  $Al_2O_3$  films fabricated at various temperatures: (a) 80 °C; (b) 200 °C; (c) 300 °C. (d) Comparison of gate leakage current during voltage scans between ±15 V. A higher gate leakage current occurs for  $Al_2O_3$  devices deposited at lower temperatures.



Fig. S11. Mechanical-bending properties of ALD-grown 20 nm  $Al_2O_3$  film. (a) Leakage current as a function of electric field for  $Au/Al_2O_3/Au$  after various numbers of bending cycles at a fixed bending radius of 10 mm. (b) Leakage current density as a function of number of bending cycles with a bending radius of 10 mm and for various electric fields.



**Fig. S12.** Mechanical-bending properties of OFET memory device with PαMS (10 nm)/Al<sub>2</sub>O<sub>3</sub> (20 nm) gate stack fabricated on a muscovite substrate. (a) Typical I<sub>DS</sub>-V<sub>GS</sub> curves for reading out V<sub>th</sub> at various bending radii. (b) Schematic diagram of study of bending endurance with various bending radii. The threshold voltage was periodically recorded after a certain number of bending-unbending cycles. (c) Typical I<sub>DS</sub>-V<sub>GS</sub> curves for reading out V<sub>th</sub> after various bending cycles with different bending radii. (d) Typical I<sub>DS</sub>-V<sub>GS</sub> curves for reading out V<sub>th</sub> after various bending cycles with different bending radii. (d) Typical I<sub>DS</sub>-V<sub>GS</sub> curves for reading out V<sub>th</sub> after various bending cycles at a fixed bending radius of 10 mm. All I<sub>DS</sub>-V<sub>GS</sub> curves were measured after the memory device was subjected to a +6 V, 100 μs program or a -4 V, 100 μs erase pulse at a V<sub>DS</sub> = -3 V.