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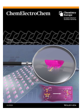
Supporting Information



Adhesive Ion-Gel as Gate Insulator of Electrolyte-Gated Transistors

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Contents

Experimental Section

Synthesis of adhesive ion gel: Poly(vinyl alcohol) (PVA, Mw: 100,000, Sigma Aldrich) and poly(methyl vinyl ether-alt-maleic anhydride) (PMVE-MA, Mw: 1,080,000, Sigma Aldrich) were separately dissolved in DMSO, and stirred at 60 ºC for 2-3 hours until those were perfectly dissolved. After each solution was cooled down to room temperature, 1-ethyl-3-methylimidazolium triflate ([EMIM][OTf], Sigma Aldrich) was added to PMVE-MA solution and stirred for 10 min at room temperature. Both solutions were mixed and stirred for a few minutes at room temperature. Before the gelation, the ink was poured on glass substrate and aged overnight. The weight ratio of ion gel ink; PVA : PMVE-MA : [EMIM][OTf] : DMSO = 5.6 : 1.4 : 16 : 77 wt%.

Characterization of adhesive ion gel: For the measurement of impedance spectroscopy, ion gel film (diameter: 12 mm, thickness: 0.76 mm) is sandwiched with stainless steel electrodes and assembled with Swagelok cell. Bio-logic SP 150 device is utilized to measure the impedance spectroscopy ranging from 300 kHz to 1Hz. Voltage amplitude is 10 mV. Fourier-transform infrared (FTIR) spectroscopy (Perkin Elmer) was utilized. PVA, PVA/PVME-MA film and PVA/PVME-MA ion gel film were prepared by drying at 50 °C for 5 hrs, and PMVE-MA pellet was prepared by blending with KBr (Potassium Bromide for FT-IR) and pressing using hydraulic pressure. The samples were scanned 12 times from 2000 cm^{-1} to 500 cm^{-1}.

Fabrication and characterization of in-plane, film-attached EGTs: Gate, source, and drain electrodes were prepared by patterning indium tin oxide (ITO)-coated glass substrate (ITO thickness: 150 nm) using a pulsed IR-laser (Trumpf, TruMicro 5000). Indium oxide precursor ink which 0.05 M of $In(NO_3)_3$ is dissolved into deionized water (D.I. water) with glycerol was printed by Sonoplot Microplotter on the area between the source and drain electrodes and annealed at 400 °C for 2 hr. Channel width and length are 2 mm and 0.5 μm. After completing to prepare the electrodes and semiconductor on the substrate, adhesive ion gel film was simply attached to the substrate by tweezer. The fabricated EGTs was characterized by Agilent 4156 C analyzer.

Supplementary Figures

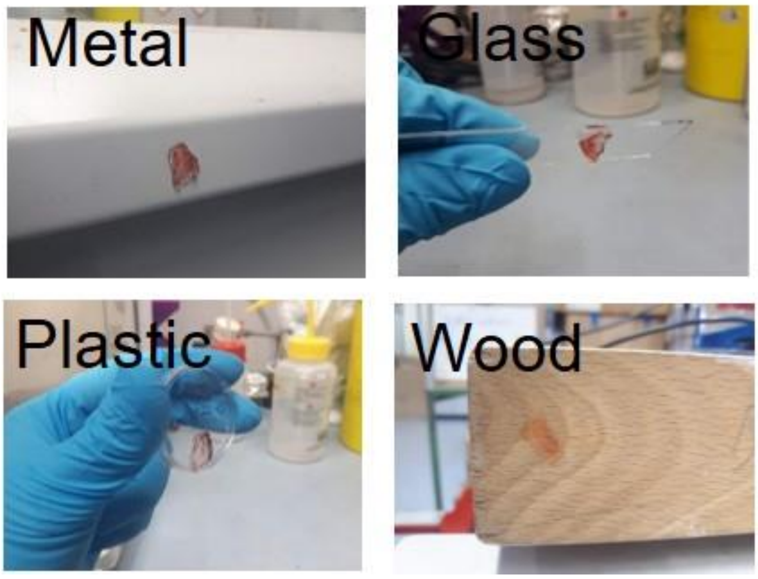

Figure S1. Pictures of adhesion test of ion gel on (a) metal, (b) glass, (c) plastic, and (d) wood.

Figure S1: The adhesive properties of the AIG can be approximately guessed. Pressure-sensitive adhesive MVE alone shows an adhesive strength of < 3×10^6 dynes cm^{-2}.[1] In general, polymer-based pressure-sensitive adhesives have probe tack values of 1 to 8×10^5 dynes cm^{-2} and peel force values of 5×10^3 to 3×10^5 dynes cm^{-2}.[2] Therefore, we roughly guess that the AIG shows approximately similar adhesive forces.

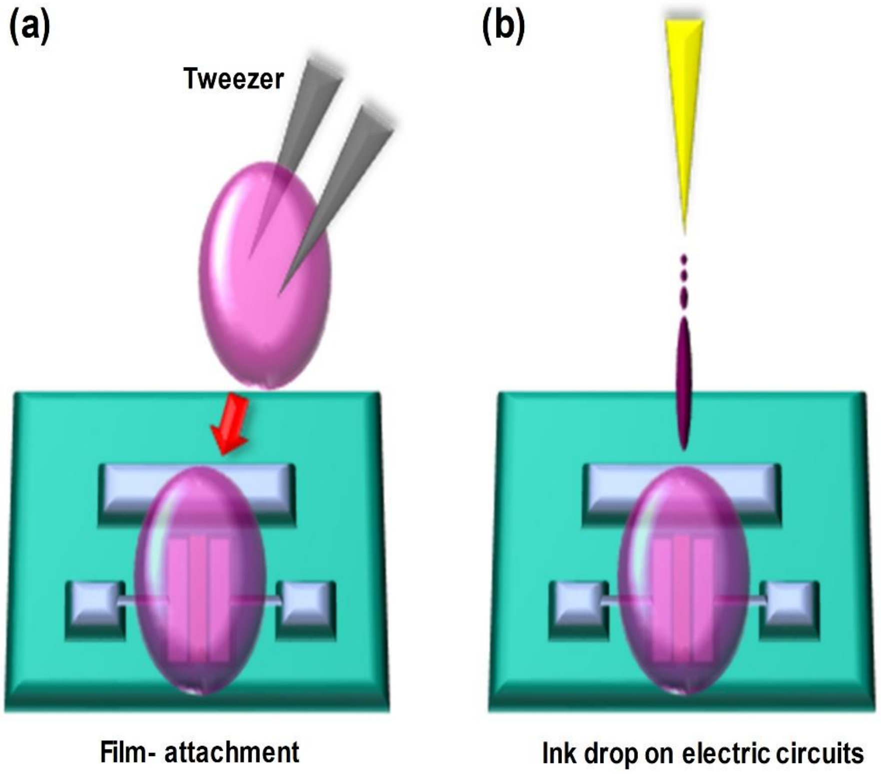

Figure S2. Illustration of film-attachment and ink drop-casting procedure for EGT fabrication

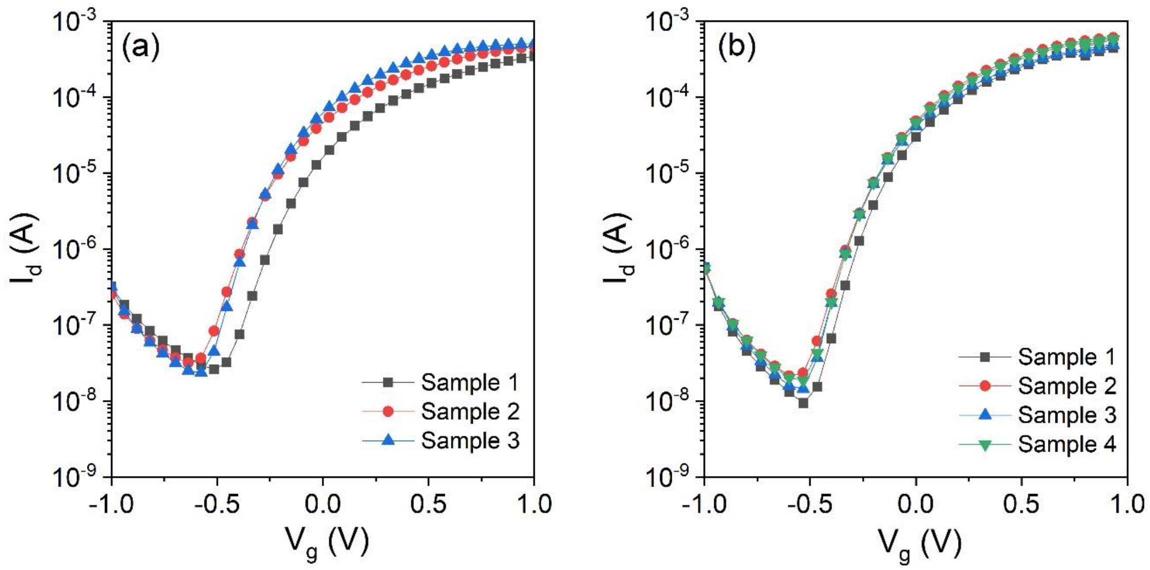

Figure S3. Gate voltage (V_g) versus drain current (I_d) plots of (a) film-attached EGTs and (b) ink drop-cased EGTs for reproducibility test.

Table S1. Electrical parameters of EGTs in Figure 5.

Type of EGTs	On-current [A]	Off-current [A]	On/off current ratio [10^4]	Subthreshold Swing [mV dec^{-1}]	Threshold voltage [V_{th}]
Film-attached EGTs	5.02×10^{-4}	2.36×10^{-8}	2.24	117.24	−0.36
Ink drop-casted EGTs	5.65×10^{-4}	1.86×10^{-8}	3.03	119.06	−0.32

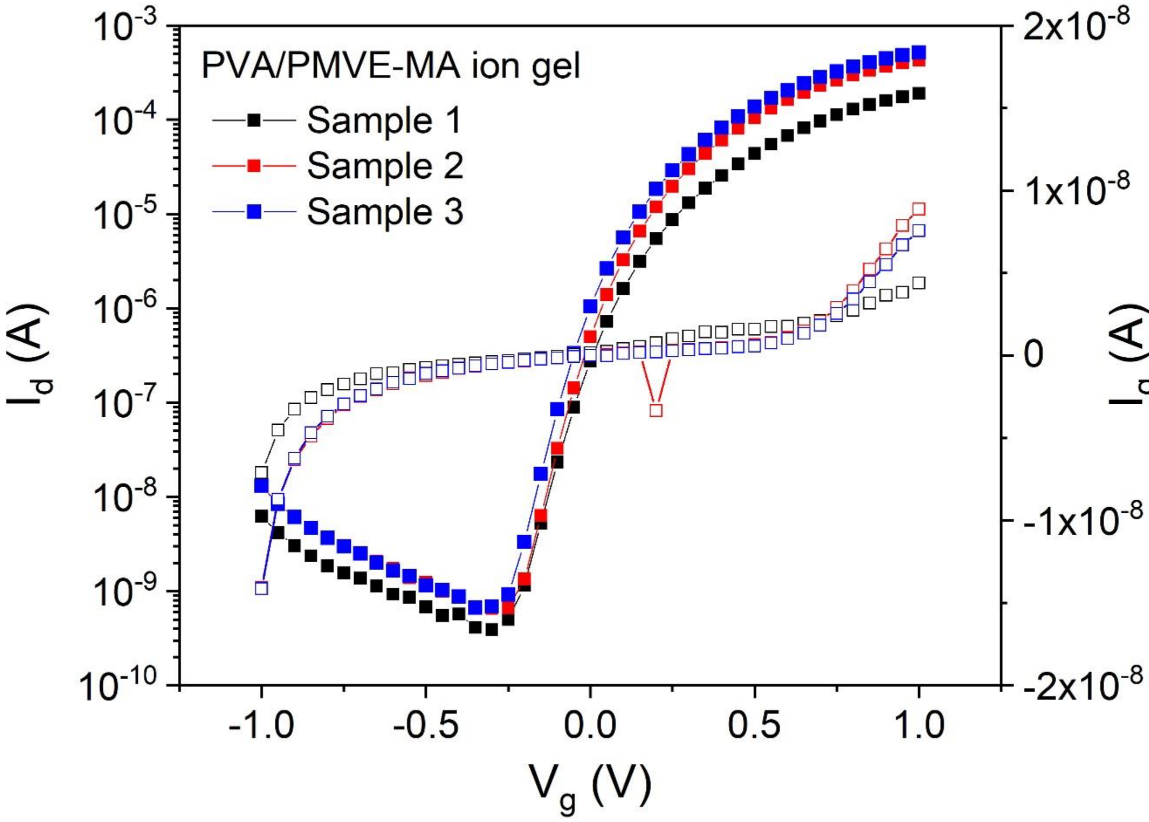

Figure S4. V_g versus I_d and I_g plot of top-gated, ion gel-gated transistors fabricated by ink-jet printing. The dilute ink of adhesive ion-gel (AIG) was printed by ink-jet printer. V_d is 1 V and RH is 20 %. The channel width and length are 600 µm and 20 µm.

Figure S4: The transfer curves result from top-gated, ion-gel-gated transistors fabricated by ink-jet printing with the dilute inks of adhesive ion gel (AIG). In previous research, we developed the dilute, ink-jet printable ion gel inks to control the self-assembled gelation.[3] By the same approach, we made the dilute inks of AIG and fabricated the micro-scale EGTs (channel width and length are 600 µm and 20 µm). Drain, source, and gate electrodes were prepared by e-beam lithography (EBL) on a glass substrate, and PEDOT:PSS was printed on ion gel by ink-jet printer as a top-gate electrode.

Ink-Jet printing and film-attaching are different manufacturing methods, each with its particular advantages and disadvantages. Film-attaching is a solvent-free method, which could circumvent solvent-based issues during the manufacturing process like swelling, dissolution of polymers or organic components, drying effects (e.g., coffee-ring effect) or molecular penetration. These issues arise especially in multilayer systems (e.g., multi-layered thin-film transistors).[4,5]

As an example, during the fabrication of OFETs, orthogonal solvents are utilized for polymer dielectrics to prevent dissolution and swelling of the organic semiconductors.[4,5] Film-attaching does not need to consider these effects of solvents, and it can shorten manufacturing time by removing the drying process to remove the solvents. Nevertheless, if these advantages can be really applied later in roll-to-roll processes, it has to be investigated.

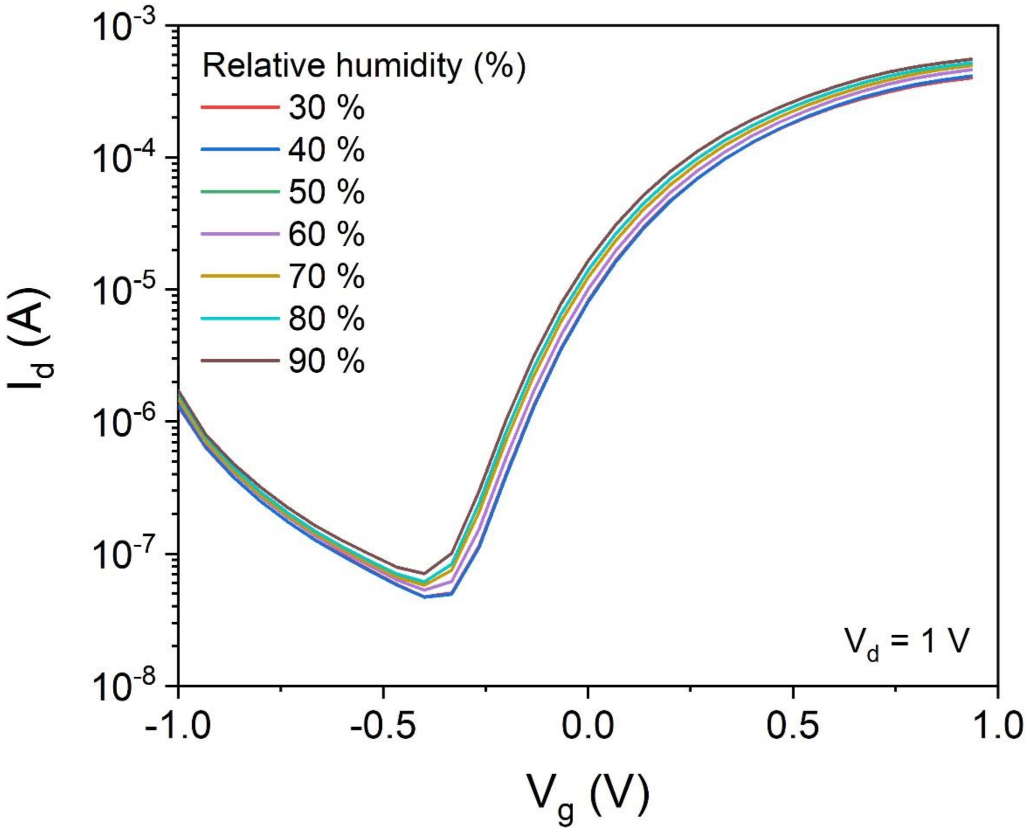

Figure S5. Transfer curves of AIG-gated EGTs measured at different relative humidities (R.H.) for humidity stability test. The R.H. conditions were controlled from 30 % to 90 %, and source-drain voltage (V_d) was set to 1 V. Gate voltages (V_g) were swept from -1 V to 1 V during measurement. The width and length of In_2O_3 semiconducting channels are 2 mm and 50 μm, respectively.

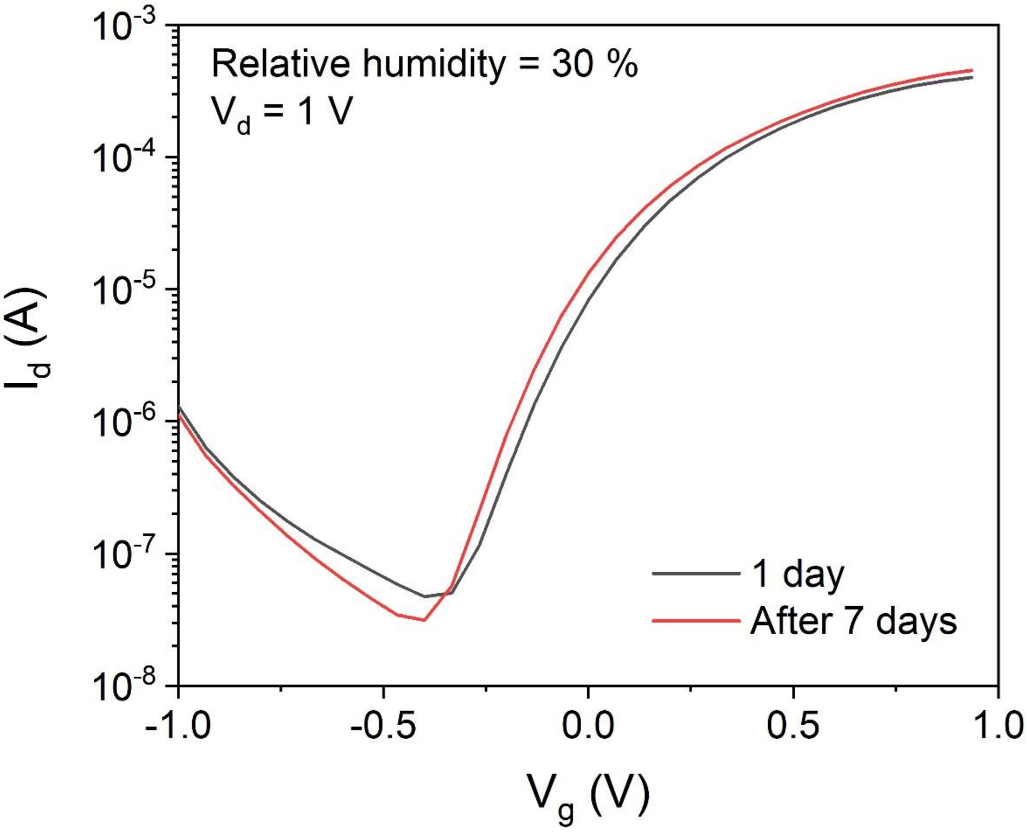

Figure S6. Durability test of AIG-gated EGTs. Drain-source voltage(V_d) was set to 1 V, and relative humidity was 30 %. The width and length of In_2O_3 semiconducting channels were 2 mm and 50 μm, respectively. The sample was stored at room condition for 7 days after first measurement.

Reference

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