Electric-double-layer transistors of organic Mott insulators

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1. Introduction
Carrier density control is one of the key issues in the development of solid state physics and novel functional devices. Beyond the simple enhancement of conductivity, high charge carrier accumulation can realize various phenomena, such as phase transition, magnetic ordering, and superconductivity. Particularity, Electric double layers, formed at solid/electrolyte interfaces, can induce extremely large electric fields, huge specific capacitance and high charge carrier accumulation \((n_{2D} \approx 10^{12}/\text{cm}^2)\). Therefore, electric-double-layer transistors (EDLTs), which use electric double layers as gate dielectric layers, are attracting much interest due to their wide range controllability of carrier density and resulting novel functionalities, opening new route for novel functional devices. However, owning to the heavy reactivity of electrolytes, the applicable materials were mainly limited to inorganic materials.

As the next step, because molecular materials have large variety of electronic and magnetic properties, we are trying the combination of molecular solid and EDLTs. Previously, we succeeded in the EDLT fabrication of van der Waals materials, such as Rubrene, Pentacene and BP3T \([1]\). Very recently, to explore material variation, we are challenging the EDLT fabrication of organic Mott insulators, resulting in the rapid dissolution of samples by electrolytes. To slow down the sample dissolution rate, we focus on an ionic gel, which is a mixture of ionic liquid and organic polymer, because the ion gel might prevent the sample from direct contact with electrolytes. Therefore, in this study, we examined a variety of ionic gel in order to resolve this problem.

2. Experiment
We examined more than 20 kinds of combinations among ionic liquids, organic polymers and organic solvent. We measured the ion conductivity from impedance measurements and checked dissolution of organic Mott insulators from optical microscopic observations. As an organic Mott insulator, in this study, we selected \(\kappa-(\text{BEDT-TTF})_2\text{Cu}[\text{N(CN)}_2]\text{Cl}\) and applied the EDLT fabrication technique into single crystals.

3. Result and Discussion
Finally, after testing 20 types of ion gels, we found suitable ion gel for \(\kappa-(\text{BEDT-TTF})_2\text{Cu}[\text{N(CN)}_2]\text{Cl}\) and succeeded in the fabrication of EDLTs with negligible dissolution. The finally obtained ion gel is the mixture of DEME-TFSI and P(VDF-HFP) (Fig1). Fig2 shows the optical image of single crystal after one week from ion-gel deposition. Fig3 shows gate voltage dependence of the sheet resistivity, suggesting the clear ambipolar carrier density modulation of \(\kappa-(\text{BEDT-TTF})_2\text{Cu}[\text{N(CN)}_2]\text{Cl}\) single crystal by the mixture of DEME-TFSI and P(VDF-HFP).

![Fig. 1. Ionic liquid (top) polymer (bottom)](image1.png)

![Fig. 2. Single crystal after one week from ion-gel deposition](image2.png)

![Fig. 3. Gate voltage dependence of the sheet resistivity](image3.png)