Supporting Information

High Mobility Organic Field-Effect Transistor Based on Hexamethylenetetrathiafulvalene with Organic Metal Electrodes

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1. Crystal growth

HMTTF was synthesized according to the literature[9]. The material was purified by recrystallization and temperature-gradient sublimation in vacuum. In the vapour-transport crystal growth, we used a glass tube with size of about 20cm in length and 1.6 cm in diameter, sealed with 3 mbar nitrogen gas. Single crystals with size of about $0.8 \times 0.2 \times 0.2$ mm^3 were grown around the location at about 140 °C in the temperature-gradient furnace with the source temperature at about 180 °C. In the crystal growth from solution, 3-5 mg of HMTTF powder was dissolved in 10 ml of well-purified chlorobenzene by stirring at 80 °C. After it was cooled down and filtrated, the solution was placed in desiccator for two weeks with flowing argon gas. As the solvent gradually evaporates, the single crystals were obtained.

2. Device fabrication

Complex powders of tetrathiafulvalene-tetracyanoquinodimethane (TTF-TCNQ) were

obtained as precipitation by the reaction of TTF and TCNQ in acetonitrile solution, and were fully dried in vacuum. Vacuum-deposited TTF–TCNQ films with thickness of about 300 nm were obtained by direct deposition of complex powders at 5×10^{-4} Pa from alumina crucible which is set at 185 °C at the deposition rate of 0.3 nm/s. The sheet resistance of the obtained film is 1 ~ 1.5 kΩ. The thickness of parylene C used as the gate dielectric layer is around 1 μm (C = 1.90 ~ 2.12 nF/cm^2). Both of the channel length and width is 100 μm. The gate electrodes were fabricated by painting of a gold paste (Tokuriki).

3. Device characteristics with Ag or Au source/drain electrodes

Transfer characteristics of HMTTF single crystal FETs with (a) silver and (b) gold source/drain electrodes are shown in Fig 1. The mobility of the devices is estimated as 0.12 cm^2/Vs for the former and 0.02 cm^2/Vs for the latter.

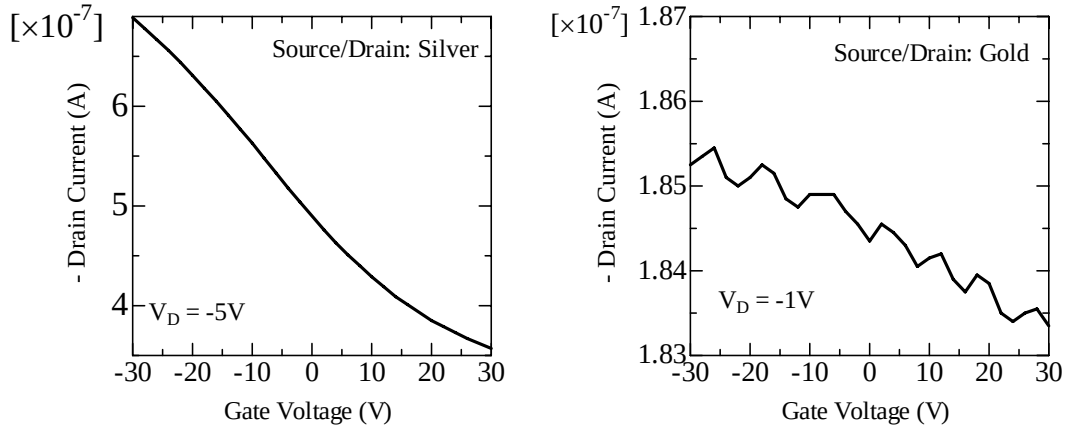

Fig. 1. Transfer characteristics of HMTTF single crystal FETs with (a) silver and (b) gold source/drain electrodes.

4. X-ray analysis

The x-ray measurements were carried out at the beam line BL-1A at the KEK (High Energy Accelerator Research Organization) Photon Factory in Japan. The incident beam was

monochromated (λ = 0.688 Å) by a pair of Si(111) single crystals. Cylindrical imaging plate diffractometer (Rigaku) is used for the detection of the Bragg reflections. The Rapid AUTO program (Rigaku) was used for the two-dimensional image processing. The crystallographic software package of CrystalStructure^{TM} (Rigaku/MSC) was used for refinements of the structure. The initial structure is modeled by the direct method (SIR2002) and is expanded using Fourier techniques. Non-hydrogen atoms were refined anisotropically. Hydrogen atoms were included but not refined. The final cycle of full-matrix least-squares refinement is based on 1,517 observed reflections with 157 variable parameters (reflection/parameter ratio ~ 9.66) and the final residual value is 5.7 %. The final atomic parameters, bond lengths, and angles are summarized as the crystallographic files in .cif format.

5. Intermolecular overlap

The intermolecular overlap integrals were calculated on the basis of extended Hückel molecular orbital method. Calculations of the overlap between highest-occupied molecular orbitals (HOMO) were done for various combinations of neighboring HMTTF molecules based on the atomic coordinates as obtained by the structure analyses. The intermolecular transfer integral t_i were estimated by using the relation; $t_i = -E\,S_i$ where E is assumed to be -10 eV.