# Limiting Factor Analysis of Device Operation of Organic Thin Film Transistors by Field-Induced Electron Spin Resonance

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### 1. Introduction

Organic thin-film transistors (OTFTs) have recently attracted considerable worldwide attention because they permit production of large-area and low-cost electronic devices on flexible plastic substrates [1]. Owing to the development of new semiconductor materials and thin-film processing of the materials, the reported mobility now exceeds 10  $\text{cm}^2/\text{Vs}$ , which is one order higher than that of amorphous silicon TFTs [2]. However, the origin of the effective mobility calculated from transfer characteristics has not been fully understood yet. Generally speaking, the charge transport in polycrystalline OTFTs can be divided into three parts: transport inside crystalline domains, across domain boundaries, and at electrode contacts. The effective mobility should be more or less determined by these contributions, while it is not clear which one is the most important factor in respective OTFTs.

Here we show that electron spin resonance (ESR) analysis allows separate evaluation of the intra- and interdomain charge transport in polycrystalline OTFTs. We utilized the field-induced ESR technique to probe the charge carriers accumulated in OTFTs [3-6]. By comparing the results with the effective mobility calculated from the transfer characteristics, we identified the most limiting factors in device operations of respective OTFTs.

## 2. Experimental

Four kinds of semiconductor small molecules and polymers were used as channel materials (Fig. 1): pentacene, dinaphtho(2,3-*b*:2,3-*f*)thieno(3,2-*b*)thiophene (DNTT) [7], poly(2,5-bis[3-hexadecylthiophene-2-yl]thieno[3,2-*b*]thiophene) (PBTTT) [8] and poly(2,7-bis-(3-icosylthiophene-2-yl)naphtho[1,2-*b*:5,6-*b*']dithiophene) (PNDTBT) [9]. For small molecules, we used polyethylene naphthalate films as substrates and parylene C as gate insulators. The channel layers were deposited in vacuum onto the parylene C layers. For polymers, we used n-doped silicon wafers with thermal oxide layers as substrates, and formed the PBTTT and PNDTBT layers by spin coating and push coating [10], respectively.

FI-ESR measurements were carried out by using a cw X-band microwave (JES-FA200, JEOL). Gate voltages of -200 V or -100 V were applied during the ESR measurements, while the source and drain electrodes were shorted to one another. We measured the temperature dependence of the ESR spectra under the two different conditions: static magnetic field perpendicular and parallel to the substrate. The spectral analysis was carried out according to literature [6]: The intra- and interdomain diffusion rates of charge carriers were evaluated by fitting analysis of the ESR spectra measured under the perpendicular and parallel conditions, respectively.



Fig. 1 Molecular structures of channel materials.

## 3. Results & Discussions

Figure 2(a) shows the temperature dependence of the ESR spectra for PNDTBT TFTs at  $B \parallel$  substrate. Although the lineshape at 285 K agrees well with a single Lorentz curve, it clearly begins to deviate from the Lorentz curve as the temperature decreases. Such a non-Lorentz feature is due to the variation in resonance magnetic fields for respective polycrystalline domains of OTFTs. They are averaged out at high temperature because of the thermally-activated motional narrowing effect [5,6]. By

analyzing the spectra, we evaluated the interdomain carrier diffusion rate and its activation energy; the former describes how often the carriers get across domain boundaries and the latter describes how high the energetic barriers are. On the other hand, we evaluated the intradomain carrier diffusion rate and its activation energy by analyzing ESR linewidth at  $B \perp$  substrate (Fig. 2(b)) [4].

Figure 3 shows the activation energies of intra- and interdomain carrier diffusion rates for four kinds of OTFTs. The intradomain activation energies were smaller than  $k_{\rm B}T = 25$  meV at room temperature for all the OTFTs. It indicates that the intradomain transport is dominated by shallow traps, from which carriers can be easily released at room temperature. By contrast, the interdomain activation energies were typically 2–4 times larger than  $k_{\rm B}T$ . Since the activation energies can be considered as the heights of potential barriers at domain boundaries, the result indicates that the carrier conduction across the domain boundaries is limited even at room temperature.

The effective mobility of the same devices was calculated from the transfer characteristics. For DNTT and



Fig. 2 Temperature dependence of the ESR spectra for the PNDTBT TFT measured at (a)  $B \parallel$  substrate and (b)  $B \perp$  substrate.



Fig. 3 Activation energies of intra- and interdomain carrier diffusion rates evaluated by ESR in addition to those of effective mobility evaluated by transfer characteristics. The intradomain activation energy of the pentacene TFT can not be evaluated due to the small *g* anisotropy.

PBTTT TFTs, the activation energies of the effective mobility provide good agreement with the interdomain diffusion rates. The result indicates that the most limiting factor of the two devices is the domain boundaries. On the other hand, the effective mobility of the PNDTBT TFT had the activation energy larger than both the intra- and interdomain diffusion rates. It implies that the Shottky barrier at electrode contacts should be the most important factor in PNDTBT TFTs. We believe that these analyses should provide a useful perspective on further improvements of the device fabrication processes.

#### 4. Conclusions

To conclude, we evaluated separately the intra- and interdomain carrier diffusion rates in addition to the effective mobility in polycrystalline OTFTs. Based on the analysis, we attributed the most important factors to domain boundaries in DNTT and PBTTT TFTs, and to the electrode contacts in PNDTBT TFT.

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