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Cite as: AIP Conference Proceedings **2170**, 020010 (2019); <https://doi.org/10.1063/1.5132729>
Published Online: 05 November 2019

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Temperature Modulated Field Effect in Organic Electrochemical Transistor with Ionic liquids

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Abstract. The paper deals with the description of the measuring method of temperature modulated electric current and influence of electric field on current in organic electrochemical transistors (OECT). The results of temperature dependences of current-voltage characteristics and their interpretation are presented. The measurements were performed on the organic electrochemical transistors with 3 ionic liquids: a) 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imid (No1), b) 1-ethyl-3-methylimidazolium ethyl sulfate (No4) and c) 1-ethyl-3-methylimidazolium tetrafluoroborate (No9). The method enables to measure the voltage and temperature dependence of the current in one step. Thus, the measured current response is influenced not only by temperature change but also by relaxation processes at the same time. The result is the dependence of activation energy on the source-drain voltage.

INTRODUCTION

The organic electrochemical transistor (OECT) is a special device where both electronic source-drain current and ionic gate-channel current influence the semiconductor behavior. Compared with usual FET transistor, there is no insulating layer between the electrolyte (gate) and channel. There are several possible applications of the OECT, e.g. driver in electrochromic display [1], switch [2], biological sensor [3]–[5] and gas sensor [6], [7]. For the preparation of drain-source channel organic semiconductive polymers are often used. It is possible to use soluble materials for complete transistor preparation (including channel, electrodes, gel electrolyte) – then transistor can be simply fabricated by cheap printing technique [8]. The working characteristics of OECTs were studied by many research groups [1], [2], [9]–[12] and their behavior is generally understood. However, there are some open questions concerning e.g. influence of temperature on physical characteristics, mechanism of ion injection from electrolyte to channel polymer, characterization of the junction between conducting polymer and electrolyte (ionic liquid), influence of dipolar field in ionic liquid on charge transport in conducting polymer, etc. In this paper we present electrical characteristics and their temperature dependences of OECTs based on poly(3,4-ethylenedioxy thiophene):poly(styrene sulfonate) (PEDOT:PSS) channel and 3 ionic liquids as electrolytes (gate): a) 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imid (No1), b) 1-ethyl-3-methylimidazolium ethyl sulfate (No4) and c) 1-ethyl-3-methylimidazolium tetrafluoroborate (No9). Emphasis is on the influence of temperature and applied voltage on source-drain current and its activation energy.

MATERIALS AND METHODS

Samples of OEETs, see Fig. 2a, were prepared by screen printing technique on a flexible substrate 175 μm Melinex® ST504 which was used as received. The printing process was divided into several steps: the carbon paste Gwent D1, modified by 10 wt. % of *N*-methyl-2-pyrrolidone (NMP), was used in the first step for the preparation of drain, source and gate electrodes. The sheet resistance of the resulting layers was about 60 Ω/sq . The active layer channel was printed using PEDOT:PSS polymer (Clevios™ SV3, purchased from Heraeus in the form of screen printing formulation); the sheet resistance of the resulting layer was $\sim 700 \Omega/\text{sq}$. Dielectric CSP-5210 purchased from Chang Sung Corporation CSC was used for the preparation of the mask for ionic liquid electrolyte. Liquid electrolyte was precisely dropped (0.5 μl) by automatic micropipette. The chemical structure of the ionic liquid gate is given in Fig. 1. OEETs were stored in environment with controlled humidity at room temperature. Transistors were contacted by spring probes.

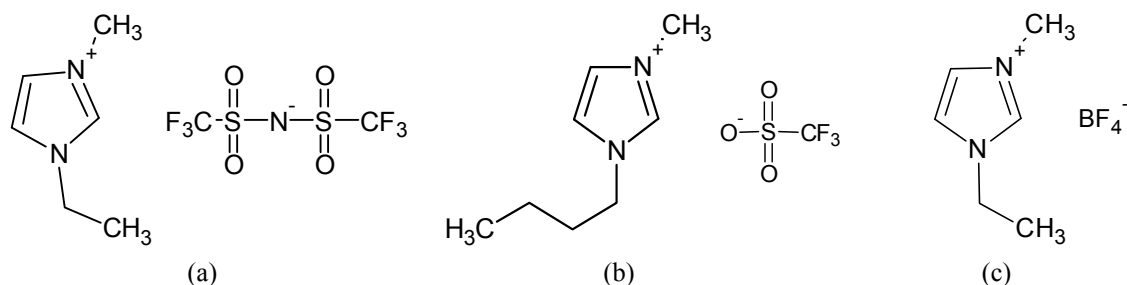


FIGURE 1. Chemical structure of ionic liquid (a) 1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide (No1), (b) 1-ethyl-3-methylimidazolium ethyl sulfate (No4) and (c) 1-ethyl-3-methylimidazolium tetrafluoroborate (No9)

EXPERIMENT

The measurements were performed on automatic apparatus. Temperature was swept using thermostat Lauda ECO RE415 in temperature range (20–40) $^{\circ}\text{C}$ (30 min linear heating, 30 min linear cooling, 450 measured values of the current). During the temperature sweep, drain-source current was measured in voltage range from 1 to -2 V (voltage step 5 mV, 75 measured values of the current at constant voltage) using electrometer Keithley 6517b. Several gate-drain voltages (U_{GD}) were used, however, they were constant during the measurements.

RESULTS AND DISCUSSION

In Fig. 2b is shown the dependence of step change of U_{SD} voltage, temperature and resulting current vs. time for OEET with ionic liquid No1. It is evident from this Fig. 2 that at constant voltage the current increases during both increase and decrease of temperature. Thus, the change of the current is caused not only by temperature influence but also by relaxation processes ongoing during the voltage change. Further it is clear that during the temperature decrease both effects compose each other and the current is growing slower. At temperature increase both effects are compensated.

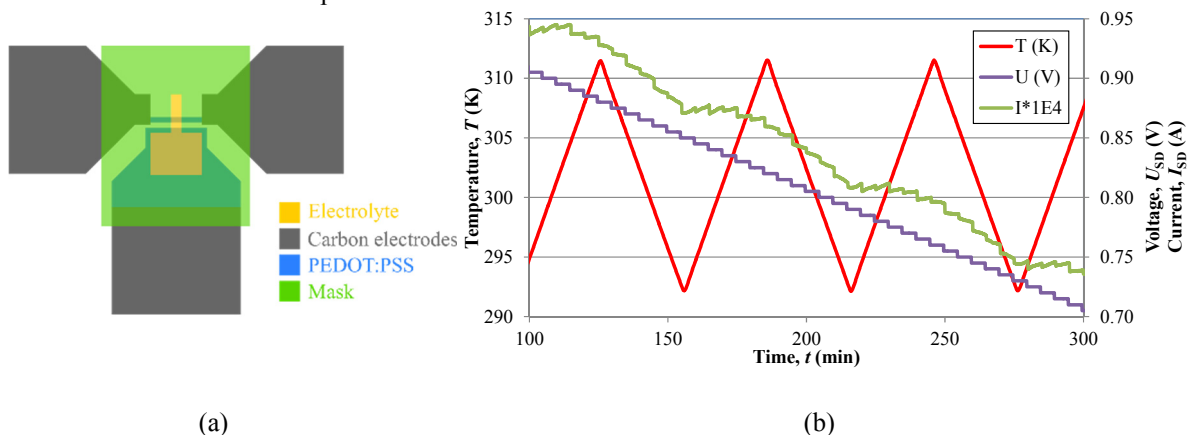


FIGURE 2. (a) Configuration of the OEET transistor, (b) dependence of temperature T , step change of U_{SD} voltage and resulting current I_{SD} vs. time (OEET with ionic liquid No1)

As the temperature rises, both temperature and voltage influence the process. In this case, when temperature increases, the apparent “activation energy” obtained from Arrhenius plot is higher than the real activation energy (thermal). When the temperature falls, the influence of temperature and voltage is subtracted, the apparent “activation energy” obtained from Arrhenius plot has a lower (sometimes negative) value than the real activation energy. Subsequently, a linear regression of the Arrhenius current-temperature dependence was performed for the individual jumps of voltage. For drain-source current the Eq. (1) can be written in the form

$$I_{SD} = I_0 \exp\left(-\frac{E_a \pm eU_{SD}(t)}{k_B T}\right), \quad (1)$$

where I_0 is the pre-exponential factor involving the relaxation of the current after applying the voltage, E_a is the activation energy, $U_{SD}(t)$ is the change of the voltage, k_B is the Boltzmann constant, and T is the temperature. From these values the activation energy for one temperature cycle was determined (450 points).

Fig. 3 shows output current-voltage characteristics in the course of individual temperature cycles for gate voltage $U_{GD} = -0.5$ V. A linear dependence for positive U_{SD} voltages is evident from the current-voltage characteristic, which corresponds to the PEDOT:PSS properties (ohmic current, hole conductivity). For negative U_{SD} voltage the PEDOT:PSS hole conductivity again plays a role, for greater negative voltages the current saturation is observed.

From the slope of the relation $\ln(I_{SD}) = f(1/k_B T)$ the voltage dependence of the “activation energy” of the current was determined (see Fig. 4). The differences between the values determined at the temperature increase and decrease are due to a step-wise increase in voltage during the experiment. In some cases (for $U_{SD} > -0.5$ V), the values of the slope were negative. This means that the influence of the change in voltage prevails over the influence of temperature. For the U_{SD} voltage ~ -1 V, the voltage change does not affect the slope and the value is directly the real activation energy. For the voltage $U_{SD} < -1.2$ V the influence of the voltage change is obvious again, but the differences are not so pronounced.

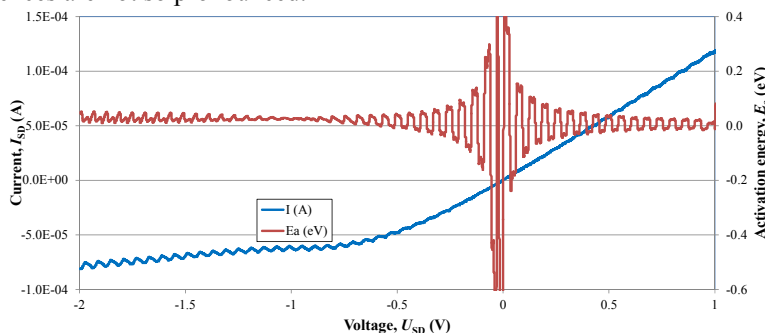


FIGURE 3. Current-voltage characteristic and the dependence of the apparent “activation energy” on voltage for OECT with ionic liquid No1 – the voltage of gate electrode was $U_{GD} = -0.5$ V.

The value of the activation energy (undistorted by the change of voltage during the measurement) can be determined as the arithmetic mean of the slope of the Arrhenius dependence determined either when the temperature rises ($E_a + eU_{SD}$) or when the temperature drops ($E_a - eU_{SD}$). This mathematical operation compensates the voltage effect, which has the same trend (it gradually decreases, see Fig. 2).

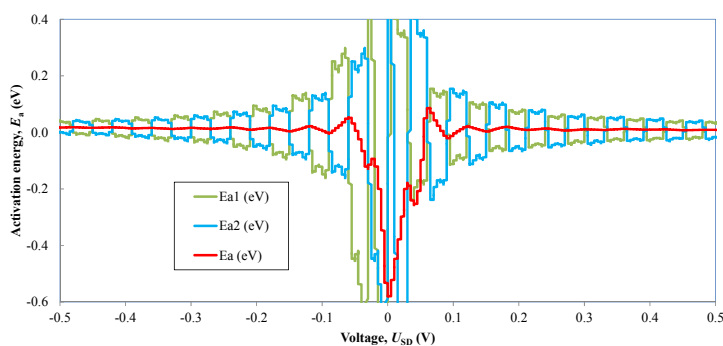
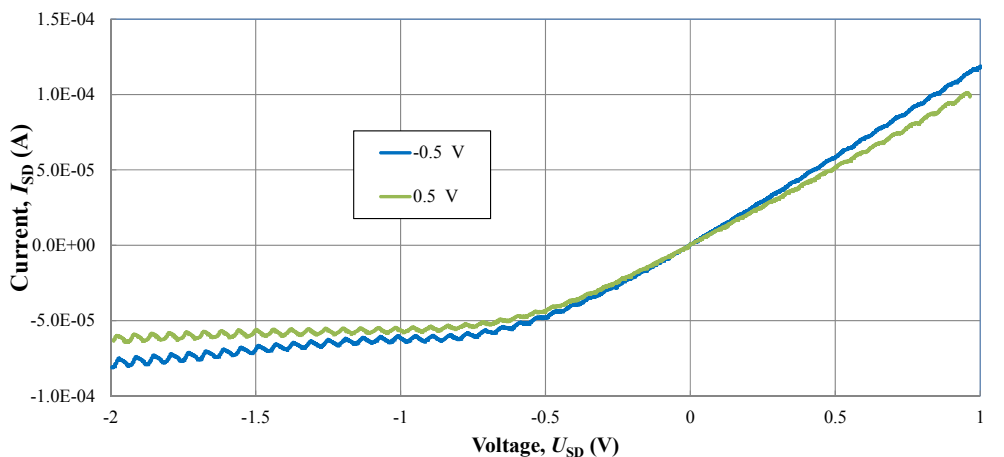


FIGURE 4. Determination of the dependence of activation energy on voltage for OECT with ionic liquid No1 for the voltage of gate electrode $U_{GD} = -0.5$ V.

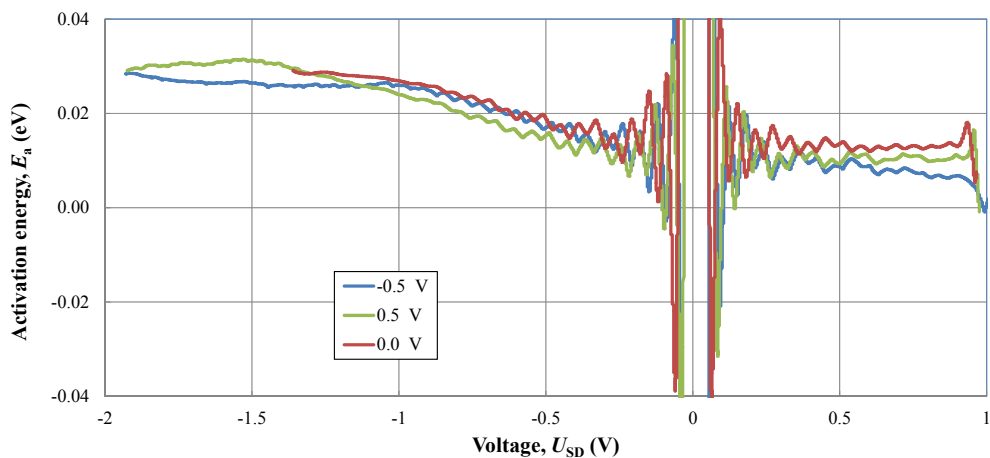
In Fig. 5 are given the overall results of output characteristics of OECT for two gate electrode voltages . From these plots it is evident a resistive (active) behavior of the transistor for voltage $U_{SD} > -0.5$ V. For the saturation region can be written

$$I_{SD} = I_0 \left(\frac{U_{GD}}{U_P} - 1 \right)^2, \quad (2)$$

where U_P is the threshold value of voltage between active and saturation regions. For high negative U_{SD} voltages the transistor avalanche breakdown occurs. In our case, this effect begins at the voltage $U_{GD} = -0.5$ V.



(a)



(b)

FIGURE 5. (a) Current-voltage characteristics and (b) the dependences of activation energies on voltages for OECT with ionic liquid No1 for the gate electrode voltage $U_{GD} = 0.5$ V (reverse direction) and -0.5 V (forward direction).

From the dependence of activation energy on voltage (Fig. 5 bottom) follows that for positive voltage U_{SD} the activation energy is 0.01 eV (activation energy of PEDOT:PSS). For negative voltage U_{SD} the activation energy increases up to 0.03 eV. It is caused by the inhomogeneity of electric field on the interface ionic liquid – PEDOT:PSS. It results in the formation of an energy barrier of approximately 0.02 eV. For voltages lower than -1.5 V in the forward direction (blue curve) the activation energy increases slightly, which corresponds to the increase in current. On the contrary, for reverse direction, the activation energy is slightly reduced.

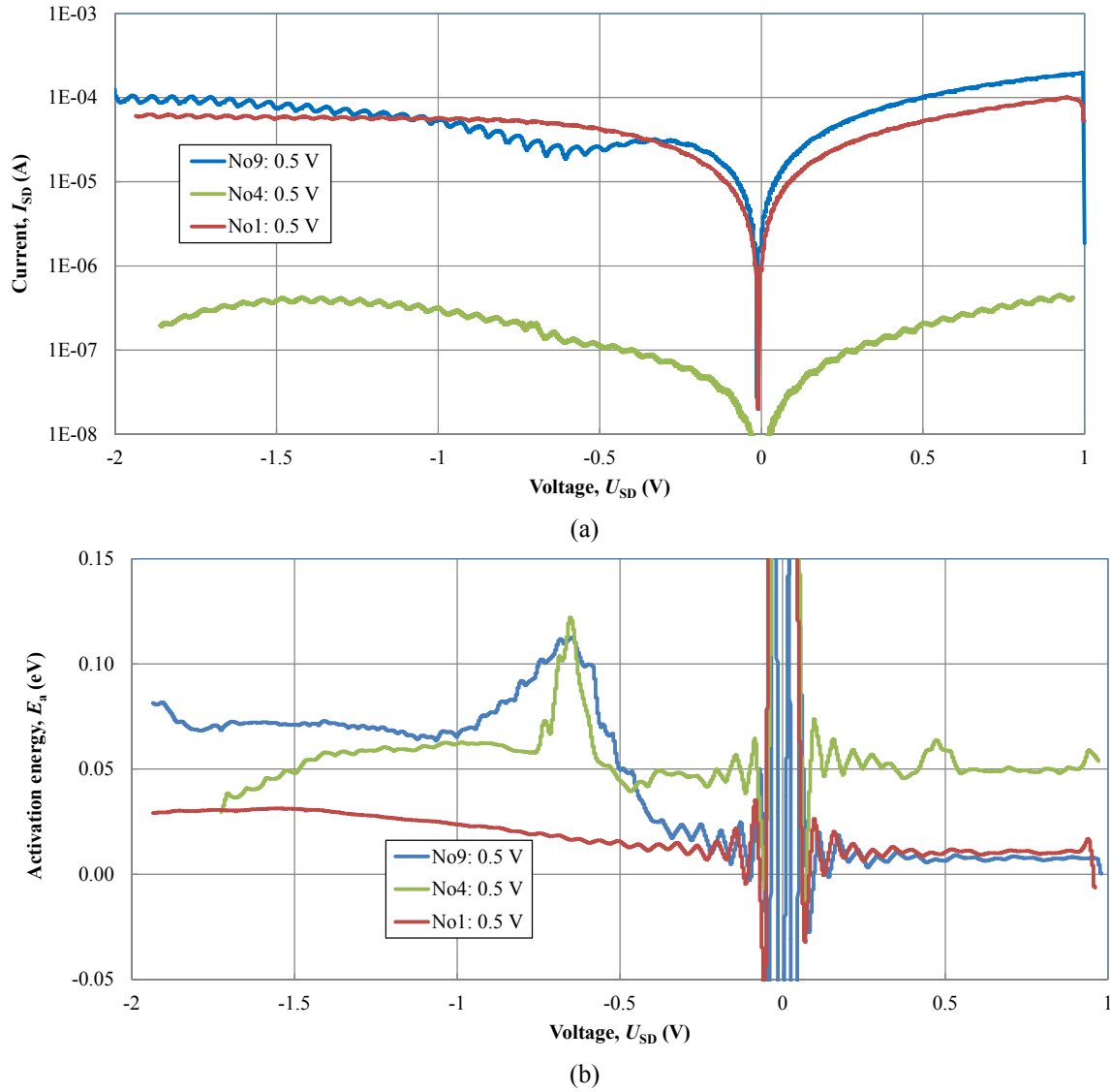


FIGURE 6. Comparison of (a) current-voltage characteristics and (b) activation energies E_a on U_{SD} voltage for OECTs with ionic liquids No1, No4 a No9 for $U_{GD} = 0.5$ V (reverse direction).

In Fig. 6 are presented current-voltage characteristics of OECTs with different ionic liquids (see Fig. 1 for their detail description) in reverse direction ($U_{GD} = 0.5$ V). It is evident that the current of OECT with ionic liquid No.4 is two orders of magnitudes lower than the currents of OECTs with ionic liquids No1 and No9. Current-voltage characteristics for $U_{SD} > 0$ V show ohmic character with constant activation energies 0.01 eV and 0.05 eV, respectively. For negative U_{SD} voltage the current decreases resulting in an increase of activation energy. At higher negative voltage the saturation of the current occurs, which again leads to a constant but higher activation energy. The exception is the sample No4, where the current and activation energy are further reduced.

CONCLUSION

The paper describes a unique method of determining the activation energy from thermally modulated currents of the OECT transistor. The method is highly sensitive and allows very low activation energies to be determined, as it is described above. Three OECT transistors with different ionic liquids were used for demonstration. The results can be used to optimally select ionic liquids for OECTs especially in terms of response rate and transistor current.

ACKNOWLEDGMENTS

This work were supported by the Czech Science Foundation under Grant "New Nature Inspired Organic Semiconductors for Bioelectronics" No. 17-24707S, by the Ministry of Education, Youth and Sports of the Czech Republic under the RICE – New Technologies and Concepts for Smart Industrial Systems, project No. LO1607, an by the Technology Agency of the Czech Republic No. TE01020022)

REFERENCES

1. P. Andersson, D. Nilsson, P.O. Svensson, M. Chen, A. Malmström, T. Remonen, T. Kugler, M. Berggren, *Adv. Mater.* **14**(20), 1460–1464 (2002).
2. P. Andersson Ersman, D. Nilsson, J. Kawahara, G. Gustafsson, and M. Berggren, *Org. Electron. physics, Mater. Appl.*, **14**(5), 1276–1280 (2013).
3. P. Lin and F. Yan, *Adv. Mater.*, **24**(1), 34–51 (2012).
4. X. Strakosas and M. Bongo, *J. Appl. Polym. Sci.*, **132**(15), 41735 (1–14) (2015).
5. O. Yaghmazadeh, F. Cicoira, D. A. Bernards, S. Y. Yang, Y. Bonnassieux, and G. G. Malliaras, *J. Polym. Sci. Part B Polym. Phys.*, **49**(1), 34–39 (2011).
6. U. Lange and V. M. Mirsky, *Anal. Chim. Acta*, **687**(1), 7–11 (2011).
7. B. Crone et al., *Appl. Phys. Lett.*, **78**(15), 2229–2231 (2001).
8. P. A. Ersman et al., *Flex. Print. Electron.*, **2**, p. 045008 (2017).
9. G. C. Faria, D. T. Duong, and A. Salleo, *Org. Electron.*, **45**, 215–221 (2017).
10. N. D. Robinson, P. O. Svensson, D. Nilsson, and M. Berggren, *J. Electrochem. Soc.*, **153**(3), p. H39 (2006).
11. D. Khodagholy et al., *Nat. Commun.*, **4**, p. 2133, (2013).
12. J. T. Friedlein et al., *Appl. Phys. Lett.* **111** (2), p. 023301, (2017)
13. L. Mracek, S. Pretl, T. Syrový, and A. Hamacek, “Ionic liquid as an electrolyte for organic electrochemical transistor,” in 38th International Spring Seminar on Electronics Technology (ISSE)-2015, (Institute of Electrical and Electronics Engineers, Eger, Hungary, 2015), pp. 11–15.