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Field-effect transistors based on short organic molecules

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The performance of organic based field-effect transistors (FETs) has recently known significant improvements. The mobility of organic FETs now approaches 1 cm^2 V^{-1} s^{-1} with short molecules, and 0.1 cm^2 V^{-1} s^{-1} with polymers. Here, we review recent results on these devices. Attention is paid to the models developed to account for charge transport in organic semiconductors, which present significant differences from their inorganic counterparts. In particular, the mobility is gate bias dependent, which actually mirrors a dependence of the mobility on charge concentration. This has been explained in terms of trap limited transport. The temperature dependence of the mobility is usually thermally activated, which is also consistent with trap limited transport, but could also be explained by a mechanism of small polaron hopping. Furthermore, recent measurements show that the mobility can become temperature independent, which could open the way to further improvements of the performance of organic transistors.

Introduction

In microelectronic devices, polymers have generally been exploited for their insulating properties, for instance as microlithographic photo-resists or in packaging. However, more recently, organic conjugated polymers such as polythiophenes (PTs) or poly-*para*-phenylenevinylenes (PTVs) are being used as active semiconductor materials. The worldwide research in this area has blossomed during the past decade in both academic and industrial institutions. A large majority of this work has been dedicated to organic light-emitting diodes (OLEDs),[1,2] which are expected to be launched into the market very soon. A bibliography on organic LEDs can be found elsewhere in this special issue. The activity has been less intense in the area of organic field-effect transistors (OFETs).[3–11] However, several reviews on OFETs have been published in recent years,[12–15] and significant breakthroughs have been reported recently.

The vast majority of microelectronic devices are currently based on crystalline silicon, and it is generally recognised that this will continue to be the case for at least the next ten years. Theoretical limitations on the performance of organic semiconductors prevent these materials from being suitable in applications where crystalline silicon excels. The advantages of organic semiconductors can be viewed from two standpoints. In the short term, organic materials could be used in large area devices, such as flat panel displays and smart cards, where the use of crystalline silicon is limited by the size of the single crystals. In this field, amorphous and polycrystalline silicon are currently used, but the relatively high temperatures needed in their fabrication process prevent their utilisation with polymeric substrates. In the longer term, one can think of single molecular devices, the so-called 'molecular electronics', which would result in a further dramatic reduction of the size of microelectronics (or rather 'nanoelectronics'). Despite the huge research interest in this area, it is outside the scope of the present review. The paper is organised as follows. In the first part, we describe the organic thin film transistor and outline its operating mode. Then, we review the various active materials used in OFETs and give their performance. The last part is devoted to the description of models that account for the charge transport mechanisms in organic semiconductors. Special attention will be paid to gate bias and temperature dependence of the mobility in these materials.

Thin film transistors

Description

Most of the OFETs are built according to the geometry of the thin film transistor (TFT),[16] an architecture mainly used in active matrix liquid crystal displays, where, because large areas are needed, crystalline silicon is replaced by amorphous silicon.[17] The basic scheme of a TFT is pictured in Fig. 1. As with a conventional MOSFET, the TFT is an insulated gate FET. It can be viewed as a capacitor where one plate consists of a metal electrode, the gate, and a semiconducting thin film constitutes the other one. The latter is equipped with two parallel electrodes, the source and the drain. Two independent voltages drive a field-effect transistor: one applied across the insulator serves to create charges at the insulator/semiconductor interface, and a second bias applied between source and drain that drives the charges induced by the first bias. In practice, the device behaves as a variable resistance that can be modulated by the voltage applied to the gate electrode.

Operation

The performance of an FET is generally visualised by drawing its drain current I_d *vs.* drain voltage V_d curves for various gate voltages. This is called the output characteristic of the device. A set of such curves is displayed in Fig. 2. Note that the sign of the drain current is governed by that of the majority carriers of the semiconductor. For an n-type semiconductor, I_d and V_d are both positive, while they are negative in a p-type material. The curves can be divided into two regimes. At low drain voltage, the current follows Ohm's law and is proportional to V_d. This is the linear regime, where eqn. (1) is valid.

$$I_d = \frac{Z}{L} C_i \mu (V_g - V_0) V_d \qquad (1)$$

Here, Z and L are the channel width and length, C_i the capacitance of the insulator, μ the mobility of the majority carriers, V_g the gate bias and V_0 the threshold voltage. As the

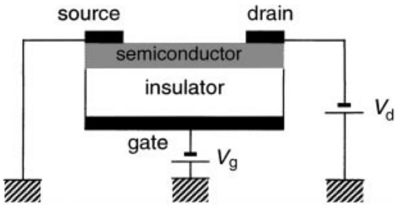

Fig. 1 Schematic view of the thin film transistor (TFT) structure used in organic field-effect transistors (OFETs).

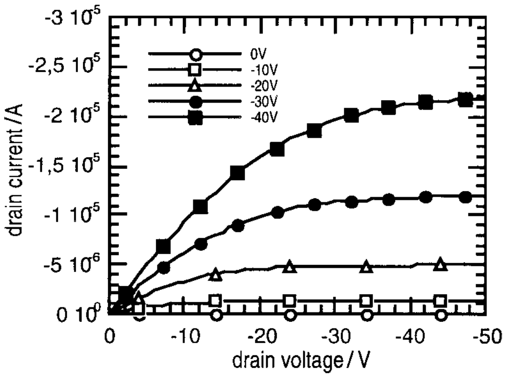

Fig. 2 Typical output characteristic of an OFET. Drain current is measured as a function of drain voltage for various gate biases. One can clearly observe the linear regime (at low drain voltage) and the saturation that occurs when the drain voltage overpasses the gate bias. Data were measured on a sexithiophene (6T) based device.

drain voltage increases, the device gradually enters the saturation regime where the drain current becomes independent of the drain bias. Eqn. (2) is often used to estimate the saturation current.

$$I_{\text{dsat}}=\frac{Z}{2L}C_i\mu(V_g-V_0)^2 \qquad (2)$$

The mobility of the majority carriers is one of the most important parameters of a FET. We see that it can be estimated from both the linear and the saturation current. The two estimations often disagree because eqn. (2) relies on several assumptions that are not actually fulfilled in organic semiconductors. One of these is that the mobility is constant; that is, it does not depend on control parameters such as the gate voltage.

Another way of characterising a FET is to draw the drain current as a function of the gate voltage at a constant drain bias, as shown in Fig. 3. This representation, also called transfer characteristic, is in fact more useful for practical applications, in which the device is used as a controlled switch. It gives access to another important parameter, namely the on–off ratio, that is, the ratio between the drain current at a given gate voltage to that at zero gate voltage. The on–off ratio actually measures the efficacy of the switch.

Materials

The dramatic improvement of the performance of OFETs during the past decade is illustrated in Fig. 4, where the mobility of various organic materials is drawn as a function of the publication date. Data are given for four materials, namely one polymer, polythiophene and its derivatives, and three small molecules, sexithiophene (6T), dihexylsexithiophene (DH6T) and pentacene. The best devices at present are made with pentacene and show a mobility of around 1 cm^2 V^{-1} s^{-1}, which compares well with that of amorphous silicon, used in active matrix liquid crystal displays. We also see that the performance of polythiophene remains roughly one order of magnitude below that of the small molecules, yet increasing at the same rate.

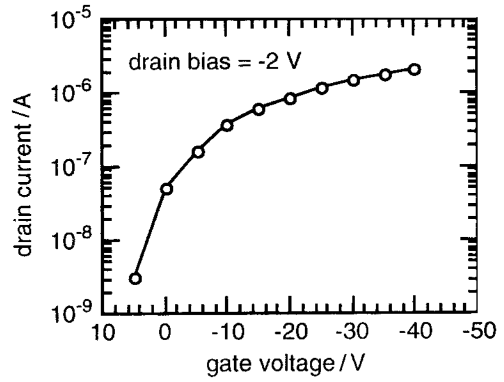

Fig. 3 Transfer characteristic of the same device as in Fig. 2. The drain current is measured as a function of the gate voltage for a fixed drain bias of -2 V.

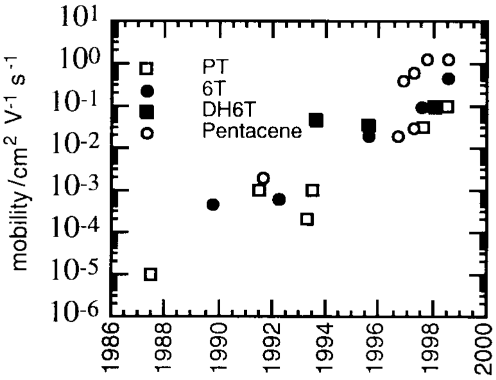

Fig. 4 Evolution of the field-effect mobility of OFETs during the past decade. Data are given for one polymer (polythiophene) and three short molecules: sexithiophene (6T), dihexylsexithiophene (DH6T) and pentacene.

Polymers

Polymers are attractive for thin film transistors because thin films of these materials can be obtained through simple and well-mastered techniques such as casting and spin coating. Conducting polymers are generally insoluble in most organic solvents, but this can be overcome by using soluble derivatives (*e.g.* alkyl chains) or soluble precursors. However, these techniques result in disordered materials, at the expense of charge transport. Accordingly, the field-effect mobility of polymer based OFETs is generally poor. Evidence for disorder limited charge transport is brought by the study of the temperature dependence of the mobility, as will be shown in the following.

One way to increase the performance of polymer TFTs consists of doping the polymer.[18] It has been shown that the mobility follows a power law with the doping level. However, doping also induces a substantial increase of the conductivity, thus resulting in a high current at zero gate voltage, at the expense of the on–off ratio.

More recently, a mobility up to 0.1 cm^2 V^{-1} s^{-1} has been reported with regioregular poly(alkylthiophene).[19,20] Undoubtedly, the use of a regioregular polymer leads to an enhancement of the ordering of the thin film, though the exact source of the improvement is not yet fully understood.

Small molecules

The use of small molecules in OFETs presents several advantages. First, their performance is better than with polymers. More importantly, the influence of various molecular parameters, such as length and crystal structure, on the charge transport can be easily controlled. An example is given by the oligothiophene series, where several reports show that the mobility tends to increase as the chain length increases. This is illustrated in Table 1.

The higher mobility in short molecules has been attributed to their ability to pack into well-organised polycrystalline films. Both pentacene and oligothiophenes, probably the most popular molecules used in OFETs, can be viewed as rigid rods. In the solid state, the molecules are fully planar and pack along parallel layers, as illustrated in Fig. 5, which shows

Table 1 Field-effect mobility/cm^2 V^{-1} s^{-1} of unsubstituted oligothiophenes of various lengths, and of their dihexyl derivatives

	Unsubstituted			
Compound	Evaporated film	Single crystal	Dihexyl substituted	References
4T	10^{-4}–6×10^{-3}	0.05	0.01–0.03	21,22,23,24
5T	1.5×10^{-3}			22
6T	0.01–0.03	0.1–0.5	0.04–0.13	11,24,25,26,27,28
8T	0.01–0.05		0.01	21,29

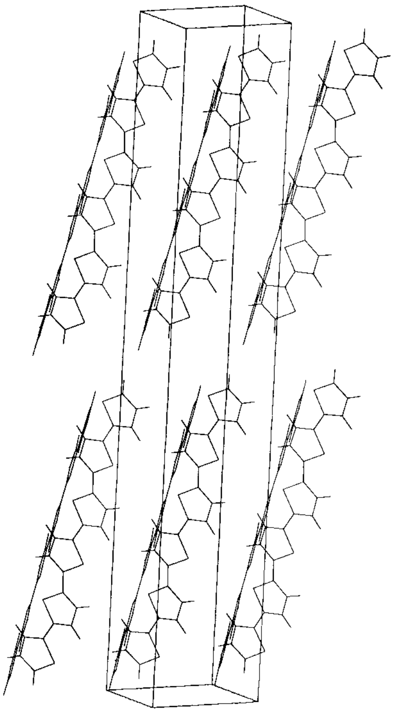

Fig. 5 Crystal structure of sexithiophene, with its quasi two-dimensional layered architecture. Note that because of the herringbone structure, one molecule out of two is viewed edge on.

Table 2 N-Channel organic field-effect transistors

Compound	Mobility/cm^2 V^{-1} s^{-1}	References
TCNQ[a]	1.9×10^{-5}	36
C_{60}	0.002–0.008	37,38
C_{60}+TDAE[b]	0.3	37
C_{70}	0.002	39
Perylenediimide	1.5×10^{-5}	40
NDTCA[c]	0.002	41
PDTCA[d]	10^{-4}	41
F_{16}–CuPc[e]	0.03	42

[a]TCNQ: tetracyanoquinodimethane. [b]TDAE: tetrakis(dimethylamino)ethylene. [c]NDTCA: naphthalenetetracarboxylic dianhydride. [d]PDTCA: perylenetetracarboxylic dianhydride. [e]F_{16}–CuPc: copper hexadecafluorophthalocyanine.

the crystal structure of sexithiophene (6T).[30] In each layer, molecules are arranged according to a so-called herringbone structure, where the planes of the molecules form a zigzag pattern. Also, the molecular close packing induces a tilt angle of the rods with respect to the normal of the layers. When deposited onto an insulator such as silica, the layers align parallel to the surface of the substrate, so that charge transport occurs in the direction perpendicular to the rods. A thin film can therefore be viewed as a two-dimensional medium where charges travel in parallel layers, where hopping of charges from one layer to the next is unlikely. Such a description is strengthened by reports on oligothiophenes substituted by alkyl chains at both ends. In these cases, the separation between layers is enhanced by the presence of alkyl layers between them, so the two dimensional character is even more apparent.[23,25,27] Actually, it has been reported that alkyl substitution leads to an improvement of charge transport, as shown by the increase of the field-effect mobility (see Table 1).

More recently, OFETs made with oligothiophene and pentacene single crystals have been reported.[24,28,31] Their mobility ranges between 0.1 and 1.5 cm^2 V^{-1} s^{-1}, which can be considered as the ultimate magnitude achievable with these materials. Other prominent features such as the variation of the mobility as a function of chain length have also been confirmed.[24]

Several other small molecules have been successfully used in OFETs, among which we can quote phthalocyanines[4,32,33] and various fused ring compounds.[34,35] All of these are p-type semiconductors. There have also been reports on n-type organic semiconductors. Some of them are listed in Table 2.

A general problem with n-type compounds is their chemical instability, and in particular their extreme sensitivity to oxygen. They are therefore less easy to handle than the p-type materials. Nevertheless, perfluorinated copper phthalocyanine has been claimed to be air-stable, with an attractive mobility of 0.03 cm^2 V^{-1} s^{-1}. A very interesting mobility has also been measured on C_{60}, which makes this compound a potential candidate to fabricate complementary organic devices. This was achieved by inserting TDAE between the source and drain electrodes and the semiconductor film in order to improve charge injection.

Models

In this section, we shall study in more detail the charge transport mechanisms in organic semiconductors.

Extended *vs.* localised states

The high mobility found in conventional inorganic semiconductors rests on the fact that charges in crystalline materials move freely in delocalised bands. These bands result from the coalescing of discrete levels; when a large number of individual atoms are gathered together in a three dimensional lattice, the discrete atomic levels widen into bands. By contrast, the lower mobility found in organic semiconductors is because the molecular levels, which become the building blocks for the solid, do not interact with each other so easily. Localisation of the states in organic semiconductors may have various origins.

- In molecular crystals, the cohesion between individual units is ensured by weak van der Waals forces rather than strong covalent bonding. This leads to narrow bands, which in turn reduces the delocalisation of the energy levels.
- Conjugated molecules tend to change their geometry upon charging. Solid state physicists would say that they show a strong electron–phonon coupling. The association of the charge with the geometrical deformation is termed *polaron.* When the charge moves in the solid, the associated defor-

mation follows it like its shadow. In other words, a polaron is a *self-localised* charge.

- A last source of charge localisation, which mainly occurs in polymer, is *disorder*.

The main consequence of localisation is the reduction of charge mobility. It also has a decisive effect on the temperature dependence of the mobility. When charges move in delocalised levels, their mean free path is much larger than the de Broglie's wavelength, so transport is only limited by scattering by phonons. In this case, the mobility increases when temperature decreases, and varies with temperature according to a power law. However, in materials with localised levels, charge transport is thermally activated, so the mobility *decreases* as the temperature is lowered.

Small polarons

As stated above, a polaron results from the coupling of a charge with the deformation of the lattice associated with this charge. When the deformation compares with the distance between lattice elements (which is the case in molecular solids, where the deformation is localised on the individual molecules), the polaron is termed *small.* (Chemists would call small polarons *radical cations* (or *radical anions*), a terminology that is also interesting because it shows that polarons have spin.) The theory of small polaron transport was developed in the late fifties by Yamashita and Kurosawa,[43] and Holstein.[44] In his theory, Holstein uses a Hamiltonian composed of three terms, one that accounts for the lattice, the second for the electron and the last one for the electron–phonon coupling. The lattice component consists of a sum of harmonic oscillators that vibrate at a unique frequency ω_0. The dominant parameter of the electron component is J, the so-called electron transfer, or overlap integral. Finally, the electron–phonon coupling magnitude can be quantified through the so-called polaron binding energy E_b, which corresponds to the energy gain due to the polarisation and deformation of the lattice. The small polaron limit corresponds to strong coupling, where $E_b \gg J$, in which case the electronic term can be treated as a perturbation. Polaron motion takes place *via* a succession of random jumps, in each of which the electron hops to a neighbour site. More recently, the concept of polaron hopping has been revisited by Emin.[45] He introduced the concept of *coincidence*, where site jumps occur when the energy state of the second site coincides with that of the first one. Such a coincidence is achieved by thermal deformation of the lattice. Emin also makes a distinction between adiabatic and non-adiabatic processes.[46] In the former, the lifetime of the coincidence is much larger than the electron transit time, in which case the electron has time to follow the lattice deformations. In the high temperature limit, the mobility of the adiabatic small polaron is given by eqn. (3).

$$\mu = \frac{\omega_0 q \delta^2}{2\pi\, kT} \exp\left(-\frac{E_b}{kT}\right) \qquad (3)$$

Here q is the absolute electron charge, δ the mean intermolecular distance, k Boltzmann's constant and T the absolute temperature. At low temperatures, the variation of the mobility is found to deviate from the thermally activated law.

Traps

Amorphous and polycrystalline silicon present significantly lower carrier mobility than the single crystalline material. This comes from localised levels in the energy gap, due to defects (dangling bounds in the amorphous material, grain boundaries in polycrystalline devices) acting as traps for the charge carriers. The multiple trapping and release model is widely used to account for charge transport in amorphous silicon.[47] The model is based on a mechanism in which carriers moving in the extended levels are trapped by the localised defects. The charges can eventually be thermally released and again contribute to charge transport. As the release mechanism is thermally activated, this results in a thermally activated mobility. In the simplest case where there is a single trap level, the activation energy corresponds to the distance between this level and the edge of the extended transport band. As shown in the following, we have successfully used this model to explain the characteristics of 6T and DH6T based devices.

Gate bias dependent mobility

The charge mobility in organic semiconductors is usually estimated from the gate voltage dependence of the saturation current. As shown by eqn. (2), a plot of the square root of I_{dsat} as a function of V_g should give a straight line, the slope of which would be proportional to the mobility. A typical plot is shown in Fig. 6. We note that the straight line is only followed at voltages higher than a few tens of volts. We have attributed the low voltage departure to a decrease of the mobility at low gate voltages.[48] Further evidence for this gate voltage dependence is brought by the plot of the drain current (the so-called transfer characteristic, displayed in Fig. 3) at low drain voltage. As shown by eqn. (1), the mobility is this time given by the slope of the line, which again departs from a straight line at low gate voltages.

The gate voltage variation of the mobility of 6T is shown in Fig. 7. We note that the mobility initially increases quasi-linearly with V_g, and seems to eventually saturate. However, we have shown more recently that this apparent saturation comes from the rather high contact resistance at source and drain.[49] The mobility corrected from the contact resistance is

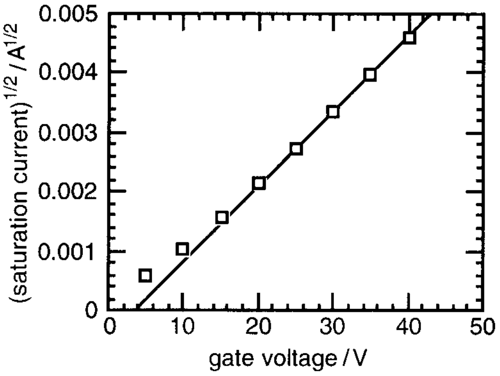

Fig. 6 Plot of the square root of the saturation current as a function of gate voltage for the same device as in Fig. 2 and 3. The deviation of the data from the straight line indicates that the mobility tends to increase with gate bias.

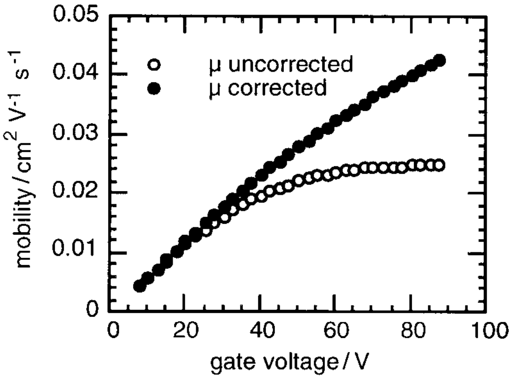

Fig. 7 Variation of the mobility as a function of gate bias for the same device as in Fig. 6. Data in full symbols are corrected for the contact series resistance.

shown by the full symbols in Fig. 7. A contact resistance of about $10^5\ \Omega$ was found.

The gate voltage produces two features in the device. First, it induces charges at the insulator/semiconductor interface, thus creating the conducting channel. Secondly, it generates an electric field directed perpendicular to the channel. As electric field dependent mobility is well documented in organic semiconductors,[50] one would think that the gate voltage dependence of the mobility in OFET could have a similar origin. However, this explanation can be disregarded since in the OFET, the electric field is mainly directed *perpendicular* to the charge transport. The field generated by the drain voltage (which is parallel to charge transport) is much weaker than the one generated by the gate voltage because of the much larger distance between source and drain than between source and gate. We can therefore safely assume that the gate voltage dependence can be related to the concentration of carriers in the conducting channel. This analysis has been confirmed recently by Dimitrakopoulos and co-workers from measurements on OFETs with insulating materials of various dielectric constants.[51]

We have used the above mentioned multiple trapping and release model to rationalise the gate voltage dependence of the mobility.[52] The model assumes a large concentration of localised states in the forbidden gap. At low gate voltage, most of the charges are trapped by these states, thus leading to a low mobility. As the gate voltage increases the Fermi level moves toward the extended band edge and more traps are filled. Eventually, all traps would be filled and any additional injected charge would be free to move in the extended states with a mobility independent of the gate bias. An analysis of both the gate bias and temperature dependence mobility allowed us to estimate the density of localised state in the forbidden gap. We found an exponential distribution of states (DOS) in both 6T and DH6T.

Temperature dependent mobility

Although they are of primary importance to clarify the mechanisms of charge transport, temperature dependent measurements in organic FETs are still scarce. We note that both the small polaron theory and the multiple trapping and release model predict a thermally activated mobility. Such dependence was indeed observed in the early measurements reported by Hotta and co-workers,[53] who interpreted their results in terms of the small polaron model. They found a polaron binding energy of 0.24, 0.18 and 0.16 eV for 4T, 5T and 6T, respectively, in good agreement with the spectroscopic data for chemically generated polarons in oligothiophene. On the other hand, and as noted above, we used the multiple trapping and release model to interpret measurements on 6T and DH6T, and found an exponential distribution of traps.[52]

More recently, Brown and co-workers reported on temperature dependence of the mobility of pentacene and polythienylenevinylene (PTV) down to 100 K. Again, the mobility was found to be thermally activated.[18] A comprehensive model was developed by Vissenberg and Matters to account for these data.[54] The model is based on a variable range hopping system with an exponential distribution of localised levels. In a sense, this model combines that of Waragai and Hotta and that used by us.

In the foregoing reports the mobility was measured down to around 100 K. In more recent reports, the temperature range was extended down to around 10 K.[55,56] Measurement on oligothiophenes revealed that the mobility at very low temperature tends to become temperature independent. An example is shown in Fig. 8, in the case of octithiophene. The curve can be divided into three domains. In the high temperature range (down to 100 K), the mobility is thermally activated with an activation energy of 0.12 eV that compares well with that reported by Waragai and Hotta for shorter molecules. In this temperature range, charge transport is therefore in good agreement with the small polaron model. At temperatures ranging from 25 to 100 K, the mobility is again thermally activated, but the activation energy is substantially reduced. Finally, the mobility is practically temperature independent for temperatures lower than 25 K. We note that data reported by Nelson and co-workers[57] on pentacene showed devices with a temperature independent mobility from 10 K up to room temperature. Remarkably, the magnitude of the mobility ranged between 1 and 2 $\mathrm{cm^2\ V^{-1}\ s^{-1}}$, which is probably the highest value for an organic FET to date. It is worth pointing out that a temperature independent mobility cannot be explained by available theories on charge transport in solids. Transport in extended levels generally results in mobility decreasing with temperature according to a power law, while hopping in localised levels leads to a thermally activated mobility. Obviously, much theoretical work remains to be done to understand charge transport in organic semiconductors.

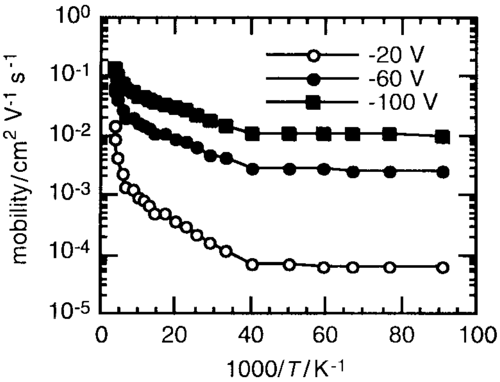

Fig. 8 Arrhenius plot of the temperature dependent mobility of octithiophene (8T). Data were recorded at various gate biases. Note that the mobility becomes practically temperature independent at low temperatures ($T < 25$ K).

Conclusions

The performance of organic FETs has now reached a level where practical applications can be envisioned. The main interest of these devices resides in the low temperature required in their processing, and their full compatibility with plastic materials. However, their operation mechanism substantially differs from that of their inorganic counterparts. The following features can be highlighted.

- The mobility depends on the concentration of carriers. This results in a gate voltage dependent mobility.
- The mobility is generally thermally activated. However, it has been reported in oligothiophene that the mobility becomes temperature independent at low temperatures. Moreover, pentacene may occasionally present a temperature independent mobility from room temperature down to 10 K.

The first feature is generally interpreted in terms of trap limited transport. Traps can also account for the thermally activated mobility. However, because of the relatively low magnitude of the mobility, hopping models are generally preferred to describe the temperature dependence of the mobility. Finally, none of these models is able to account for a temperature independent mobility.

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