<table>
<thead>
<tr>
<th><strong>Title</strong></th>
<th>Organic field-effect transistors fabricated with N, N′-substituted dialkyl-1,3,8,10-tetramethylquinacridone compounds</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Author(s)</strong></td>
<td>Xu, ZX; Xiang, HF; Roy, VAL; Chui, SSY; Wang, Y; Lai, PT; Che, CM</td>
</tr>
<tr>
<td><strong>Citation</strong></td>
<td>Applied Physics Letters, 2009, v. 95 n. 12</td>
</tr>
<tr>
<td><strong>Issued Date</strong></td>
<td>2009</td>
</tr>
<tr>
<td><strong>URL</strong></td>
<td><a href="http://hdl.handle.net/10722/124070">http://hdl.handle.net/10722/124070</a></td>
</tr>
</tbody>
</table>

**Rights**

This work is licensed under a Creative Commons Attribution-NonCommercial-NoDerivatives 4.0 International License.; Applied Physics Letters. Copyright © American Institute of Physics.; After publication: Copyright (year) American Institute of Physics. This article may be downloaded for personal use only. Any other use requires prior permission of the author and the American Institute of Physics. The following article appeared in (citation of published article) and may be found at (URL/link for published article abstract).

Before publication:
The following article has been submitted to/accepted by [Name of Journal]. After it is published, it will be found at (URL/link to the entry page of the journal).
Organic field-effect transistors fabricated with $N, N'$-substituted dialkyl-1,3,8,10-tetramethylquinacridone compounds

Zong-Xiang Xu,1 Hai-Feng Xiang,1,2 V. A. L. Roy,1,3 Stephen Sin-Yin Chui,1 Yue Wang,4 P. T. Lai,2 and Chi-Ming Che1,a)
1Department of Chemistry and HKU-CAS Joint Laboratory on New Materials, The University of Hong Kong, Pokfulam Road, Hong Kong, Hong Kong
2Department of Electrical and Electronic Engineering, The University of Hong Kong, Pokfulam Road, Hong Kong, Hong Kong
3Department of Physics and Materials Science, City University of Hong Kong, Tat Chee Avenue, Kowloon, Hong Kong
4Key Laboratory of Supramolecular Structure and Materials, College of Chemistry, Jilin University, Changchun 130021, People's Republic of China

(Received 8 April 2009; accepted 26 August 2009; published online 22 September 2009)

A series of robust and inexpensive $p$-type organic semiconductors $1,3,8,10$-tetramethyl quinacridone compounds was prepared. These quinacridone compounds bearing $N, N'$-disubstituted long $N$-alkyl chains self-organize into highly oriented crystalline films, leading to high performance organic thin film transistors with the best field-effect mobility, on/off ratio, and threshold voltage being $1.6 \times 10^{-1} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, $1 \times 10^4$, and $-17 \text{ V}$, respectively. The effects of methyl substituent and $N$-alkyl chain length of the quinacridone compounds together with the molecular packing on the field-effect mobility are discussed. © 2009 American Institute of Physics. [doi:10.1063/1.3233961]

Organic thin film transistors (OTFTs) have important applications in electronics including sensors, light-emitting devices, and memory devices. Compared with silicon-based devices, the fabrication process of OTFT is simple and has a high compatibility with conventional soft lithography. To develop inexpensive and robust organic semiconductor materials that possess high field-effect mobility, much attention has been focused on $p$-type pentacene and derivatives. Nonetheless, multisteps are needed for the synthesis and modification of pentacene compounds. In the context of developing OTFT materials, $\pi$-conjugated organic compounds with a rigid and fused-ring structure are of particular interest.

Quinacridone compounds are chemically stable and widely utilized as light-emitting and photoconductive materials. These materials are relatively inexpensive, soluble in common organic solvents, and could be readily modified. In this work, a series of $N, N'$-dialkyl quinacridone (QA) compounds—QA-C$_2$, QA-C$_4$, QA-C$_6$, and QA-C$_8$—and $N, N'$-dialkyl-$1,3,8,10$-tetramethylquinacridone (TMQA) compounds—TMQA-C$_4$, TMQA-C$_6$, TMQA-C$_8$, and TMQA-C$_{16}$ (Fig. 1)—were prepared according to the literature methods. The charge transport properties of these compounds for OTFT applications have been investigated.

The QA and TMQA compounds were used to fabricate organic transistors using a bottom-contact substrate-gate structure by high-vacuum evaporation and measured using the same literature method. The surface morphology and crystallinity of the thin films in the OTFT devices fabricated with these compounds were studied by scanning electron microscopy (SEM) and grazing incidence X-ray diffraction (GIXRD).

Table I. Most of the compounds examined have a decomposition temperature ($T_d$) in the range of $373–416$ °C. Interestingly all TMQA compounds exhibited $p$-type field-effect behavior but all QA compounds exhibited no field-effect behavior. TMQA-C$_4$ showed a hole mobility ($\mu$) of $2.4 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for TMQA-C$_6$ and $3.0 \times 10^{-2} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for TMQA-C$_8$. However, further increase in the $N$-alkyl chain length led to a decrease in the mobility value, as revealed in the case of TMQA-C$_{16}$ ($\mu = 2.1 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$). As depicted in Table I, the thin films derived from QA-C$_2$, QA-C$_4$, QA-C$_6$, and QA-C$_8$ showed no field effect. Octadecyltrichlorosilane (OTS) was deposited onto the substrate prior to the OTFT device fabrication to reduce the hydrophilicity of the SiO$_2$ surface. Figure 2 shows the output and transfer characteristics of TMQA-C$_8$-based device with OTS-treated SiO$_2$/Si substrate before and after 100 cycles measurement. Upon increasing the gate voltage $V_g$, the drain current $I_d$ increases, revealing a $p$-type field-effect transistor (FET) behavior. The fresh sample showed a mobility of $0.16 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and current on/off ratio of $10^4$. After 100 cycles measurement, the shape of the successive transfer curve remained unchanged, revealing an unchanged threshold voltage $V_t$, and the output curve...
showed a decrease in the saturated current by less than 10% and a mobility of 0.15 cm² V⁻¹ s⁻¹ corresponding to less than 10% decrease when compared with the initial mobility value. In addition, the devices could be operated under open atmosphere at room temperature and there was no change in the device performance. We did transient measurements on a TMQA-C₁₈-based FET device where a constant voltage was applied to the gate and drain (−40 V), while the current was measured as a function of time on the TMQA-C₁₈-based FET for more than 100 cycles. No decrease in the channel current was observed. This device was kept in a glovebox. After a period of 2 years, this device showed a mobility of 0.15 cm² V⁻¹ s⁻¹ corresponding to less than 10% decrease in the initial mobility value, revealing the device stability.

Both crystallinity and morphology play important roles in the FET performance. FET behavior is usually observed in the FET performance. We did transient measurements on a TMQA-C₁₈-based FET device where a constant voltage was applied to the gate and drain (−40 V), while the current was measured as a function of time on the TMQA-C₁₈-based FET for more than 100 cycles. No decrease in the channel current was observed. This device was kept in a glovebox. After a period of 2 years, this device showed a mobility of 0.14 cm² V⁻¹ s⁻¹ corresponding to less than 10% decrease in the initial mobility value, revealing the device stability.

Both crystallinity and morphology play important roles in the FET performance. FET behavior is usually observed in the FET performance. We did transient measurements on a TMQA-C₁₈-based FET device where a constant voltage was applied to the gate and drain (−40 V), while the current was measured as a function of time on the TMQA-C₁₈-based FET for more than 100 cycles. No decrease in the channel current was observed. This device was kept in a glovebox. After a period of 2 years, this device showed a mobility of 0.14 cm² V⁻¹ s⁻¹ corresponding to less than 10% decrease in the initial mobility value, revealing the device stability.

Both crystallinity and morphology play important roles in the FET performance. FET behavior is usually observed in the FET performance. We did transient measurements on a TMQA-C₁₈-based FET device where a constant voltage was applied to the gate and drain (−40 V), while the current was measured as a function of time on the TMQA-C₁₈-based FET for more than 100 cycles. No decrease in the channel current was observed. This device was kept in a glovebox. After a period of 2 years, this device showed a mobility of 0.14 cm² V⁻¹ s⁻¹ corresponding to less than 10% decrease in the initial mobility value, revealing the device stability.

Both crystallinity and morphology play important roles in the FET performance. FET behavior is usually observed in the FET performance. We did transient measurements on a TMQA-C₁₈-based FET device where a constant voltage was applied to the gate and drain (−40 V), while the current was measured as a function of time on the TMQA-C₁₈-based FET for more than 100 cycles. No decrease in the channel current was observed. This device was kept in a glovebox. After a period of 2 years, this device showed a mobility of 0.14 cm² V⁻¹ s⁻¹ corresponding to less than 10% decrease in the initial mobility value, revealing the device stability.

Both crystallinity and morphology play important roles in the FET performance. FET behavior is usually observed in the FET performance. We did transient measurements on a TMQA-C₁₈-based FET device where a constant voltage was applied to the gate and drain (−40 V), while the current was measured as a function of time on the TMQA-C₁₈-based FET for more than 100 cycles. No decrease in the channel current was observed. This device was kept in a glovebox. After a period of 2 years, this device showed a mobility of 0.14 cm² V⁻¹ s⁻¹ corresponding to less than 10% decrease in the initial mobility value, revealing the device stability.

Both crystallinity and morphology play important roles in the FET performance. FET behavior is usually observed in the FET performance. We did transient measurements on a TMQA-C₁₈-based FET device where a constant voltage was applied to the gate and drain (−40 V), while the current was measured as a function of time on the TMQA-C₁₈-based FET for more than 100 cycles. No decrease in the channel current was observed. This device was kept in a glovebox. After a period of 2 years, this device showed a mobility of 0.14 cm² V⁻¹ s⁻¹ corresponding to less than 10% decrease in the initial mobility value, revealing the device stability.

Both crystallinity and morphology play important roles in the FET performance. FET behavior is usually observed in the FET performance. We did transient measurements on a TMQA-C₁₈-based FET device where a constant voltage was applied to the gate and drain (−40 V), while the current was measured as a function of time on the TMQA-C₁₈-based FET for more than 100 cycles. No decrease in the channel current was observed. This device was kept in a glovebox. After a period of 2 years, this device showed a mobility of 0.14 cm² V⁻¹ s⁻¹ corresponding to less than 10% decrease in the initial mobility value, revealing the device stability.

Both crystallinity and morphology play important roles in the FET performance. FET behavior is usually observed in the FET performance. We did transient measurements on a TMQA-C₁₈-based FET device where a constant voltage was applied to the gate and drain (−40 V), while the current was measured as a function of time on the TMQA-C₁₈-based FET for more than 100 cycles. No decrease in the channel current was observed. This device was kept in a glovebox. After a period of 2 years, this device showed a mobility of 0.14 cm² V⁻¹ s⁻¹ corresponding to less than 10% decrease in the initial mobility value, revealing the device stability.

Both crystallinity and morphology play important roles in the FET performance. FET behavior is usually observed in the FET performance. We did transient measurements on a TMQA-C₁₈-based FET device where a constant voltage was applied to the gate and drain (−40 V), while the current was measured as a function of time on the TMQA-C₁₈-based FET for more than 100 cycles. No decrease in the channel current was observed. This device was kept in a glovebox. After a period of 2 years, this device showed a mobility of 0.14 cm² V⁻¹ s⁻¹ corresponding to less than 10% decrease in the initial mobility value, revealing the device stability.

Both crystallinity and morphology play important roles in the FET performance. FET behavior is usually observed in the FET performance. We did transient measurements on a TMQA-C₁₈-based FET device where a constant voltage was applied to the gate and drain (−40 V), while the current was measured as a function of time on the TMQA-C₁₈-based FET for more than 100 cycles. No decrease in the channel current was observed. This device was kept in a glovebox. After a period of 2 years, this device showed a mobility of 0.14 cm² V⁻¹ s⁻¹ corresponding to less than 10% decrease in the initial mobility value, revealing the device stability.
ents of platinum

N

platelike grains [Fig. 4(a)] and the QA-C₈ film was covered by continuous crystalline grains [Fig. 4(d)], whereas the thin films of TMQA compounds revealed continuous tightly packed crystals (Fig. 5) and this is favorable for charge transport. Upon increasing the N,N'-diaryl chain length from N-butyl in TMQA-C₄ to N-octyl in TMQA-C₈, TMQA-C₈ thin film was found to have a densely packed crystal grain texture with little defect content.

The thick films of all QA compounds were poorly crystalline [Fig. 3(a)], consistent with the lack of field-effect mobility, whereas thin films of all TMQA compounds have a good field-effect mobility. This should be attributed to the effect of the peripheral 1,3,8,10-tetramethyl groups at the quinacridone core. The four C–H···π interactions (d = 2.36 Å) in the case of TMQA-C₄ substantially stabilize arrangement of molecules in the solid phase whereas the aryl hydrogen atoms of QA-C₈ merely lead to weak intermolecular interactions (2.54 Å). Our previous study also showed that replace an ethyl group by a methyl group can significantly affect the field-effect mobility. Compared with platinum(II) 2,3,7,8,12,13,17,18-octaethyl-21H,23H-porphyrin, the unsymmetrical peripheral substituents of platinum(II) etioporphyrin-I enables the molecules to efficiently pack with extensive lateral aggregations and π···π stacking interactions consequently leading to a high mobility of 0.32 cm² V⁻¹ s⁻¹. Among all QA and TMQA compounds, the TMQA-C₈ thin film showed the strongest diffraction peak at 5.17° (2θ) (Fig. 3), revealing that this film had the highest degree of crystallinity. The unique spatial orientation of TMQA-C₈ molecules might reinforce the lateral molecular stacking parallel to the surface of the substrate, facilitating the in-plane charge transport of the film Therefore, the TMQA-C₈ thin film with a good morphology (Fig. 5) has the best field-effect mobility. But further increase in the N-alkyl chain as in the case of TMQA-C₁₆ leads to a decrease in the field-effect mobility. The long N-alkyl chain of TMQA-C₁₆ decreases the molecular packing, leading to a low crystallinity [Fig. 3(b)] and a low-field-effect mobility.

In summary, a series of QA and TMQA was synthesized to fabricate OTFTs by vacuum deposition method. The charge transport properties are strongly affected by the surface morphology, crystallinity, and molecular packing of the thin film. The crystal packing and field-effect mobility of thin films of TMQA compounds are affected by the length of N-alkyl chains. TMQA-C₈ was found to be a simple and low-cost organic semiconductor for the fabrication of OTFT device with potential practical application.

This work was supported by The Chinese Academy of Sciences–Croucher Foundation Funding Scheme for Joint Laboratories and HKU Seed Funding for Strategic Research Theme-Molecular Materials. We acknowledge Clover & Sunny Systems Ltd. for their support with the fabrication system housed at The University of Hong Kong (Grant No. HKU 200807176003).

See EPAPS supplementary material at http://dx.doi.org/10.1063/1.3233961 for the details of the synthesis, and TGA and transient curves.