Title	Fabrication and Characteristics of C_<84> Fullerene Field-Effect Transistor
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Citation	Applied Physics Letters, 84(14): 2572-2574
Issue Date	2004-04
Type	Journal Article
Text version	publisher
URL	http://hdl.handle.net/10119/3367
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APPLIED PHYSICS LETTERS VOLUME 84, NUMBER 14 5 APRIL 2004

Fabrication and characteristics of C_{84} fullerene field-effect transistors

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(Received 25 November 2003; accepted 9 February 2004)

Fullerene field-effect transistors (FETs) were fabricated with thin films of C_{84}, which showed n-channel normally-on depletion-type FET characteristics. The C_{84} FET device exhibited the highest mobility, μ, of $2.1\times10^{-3}\ \mathrm{cm^2\,V^{-1}\,s^{-1}}$ among normally-on fullerene FETs. The carrier transport of this FET device can be interpreted as thermally activated hopping transport. Carrier type (n-channel) and transport mechanism (hopping) reflect the electronic properties of the C_{84} molecule. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1695193]

A large number of field-effect transistors (FETs) with thin films of organic molecules have been fabricated and their characteristics have been studied for next-generation electronics during the past decade.[1,2] The field-effect mobilities μ of organic FETs are lower by four orders of magnitude than those of conventional FETs with inorganic materials. Nevertheless, organic FETs are known to have many advantages such as large-area coverage, structural flexibility, and low-temperature and low-cost processing in comparison with inorganic FETs. In 1997 the mobility of a pentacene p-channel FET reached $1.5\ \mathrm{cm^2\,V^{-1}\,s^{-1}}$.[3] Subsequently, a p-channel FET on single crystals of rubrene showed a mobility of $1.0\ \mathrm{cm^2\,V^{-1}\,s^{-1}}$.[4]

The first fullerene FET was fabricated with thin films of C_{60} by Haddon *et al.*[5] This FET device showed a high mobility of $0.08–0.3\ \mathrm{cm^2\,V^{-1}\,s^{-1}}$. The high μ value of this device implies that C_{60} FETs can play an important role in future applications such as identification tags, smart cards, and drivers for active-matrix displays based on the integration with organic light-emitting diodes. The mobility values of n-channel organic FETs except for the C_{60} FET are much lower than those of p-channel organic FETs.[1,2] Furthermore, such high μ value attracts interest in the physics and chemistry of C_{60} FETs. The improvement of properties of the C_{60} FET device has long been examined, and very recently the mobility reached $0.56\ \mathrm{cm^2\,V^{-1}\,s^{-1}}$, which is one of the highest mobilities among n-channel organic FETs.[6] The highest μ of $0.6\ \mathrm{cm^2\,V^{-1}\,s^{-1}}$ was reported for N,N'-dialkyl-3,4,9,10-perylene tetracalboxylic diimide derivative (PTCDI-C8H) FET device.[7]

Recently we reported the transport properties of C_{60} FET.[8] Temperature dependence of the mobility suggested a hopping transport as a conduction mechanism for these FETs. Furthermore, a complementary metal–oxide–semiconductor logic gate circuit was fabricated with C_{60} and pentacene FETs,[8] which leads us to realize various types of logic circuits such as NOR and NAND for computing and memory. The first FETs with metallofullerenes have been realized with Dy@C_{82} and La_2@C_{80}.[8,9] These FETs operated as n-channel normally-on device, which is substantially different from enhancement-type FETs with C_{60} and C_{70}.[5,6,8,10] The output characteristics of fullerene FETs, therefore, reflect intrinsic electronic structures of individual fullerene molecules. This letter reports on the fabrication of higher-fullerene FET devices with thin films of C_{84}, and the FET characteristics and their temperature dependence.

Schematic representation of C_{84} (D_{2d} isomer) and a cross-sectional view of the C_{84} FET device are shown in Fig. 1(a). Commercially available C_{84} (99%) was used for the fabrication of the thin film. Commercially available $SiO_2/Si(100)$ wafers were used as substrates after cleaning with acetone, methanol, and H_2SO_4/H_2O_2. The thin film of C_{84} was formed by a thermal deposition under a vacuum of 10^{-8} Torr. The channel length L and the channel width W of this device were 75 and 4000 μm, respectively. The characteristics of the C_{84} FET device were measured under 10^{-6} Torr after annealing for 24 h at 120 °C under 10^{-6} Torr.

The drain current I_D versus drain–source voltage V_{DS} plots for the C_{84} FET at 290 K are shown in Fig. 1(b). The

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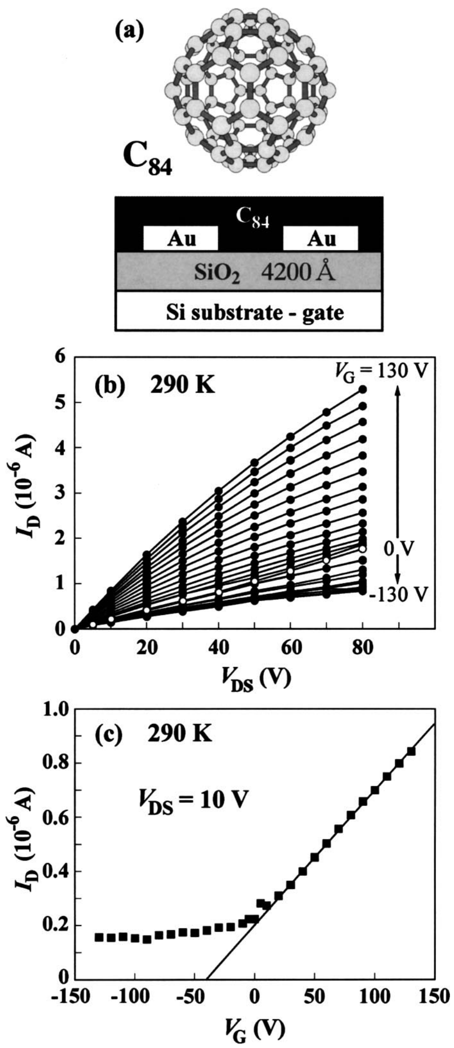

FIG. 1. (a) Schematic picture of $C_{84}(D_{2d})$ and cross-sectional view of C_{84} FET. (b) $I_D - V_{DS}$ plots of C_{84} FET at 290 K. Closed and open circles refer to the points measured at $V_G \neq 0$ V and $V_G = 0$ V, respectively. (c) $I_D - V_G$ plot at $V_{DS} = 10$ V. Solid line is the line fitted with equation $I_D = (\mu W C_0 / L)(V_G - V_T) V_{DS}$, where C_0 refers to capacitance of SiO_2; $C_0 = 8.22 \times 10^{-9}$ F cm^{-2}.

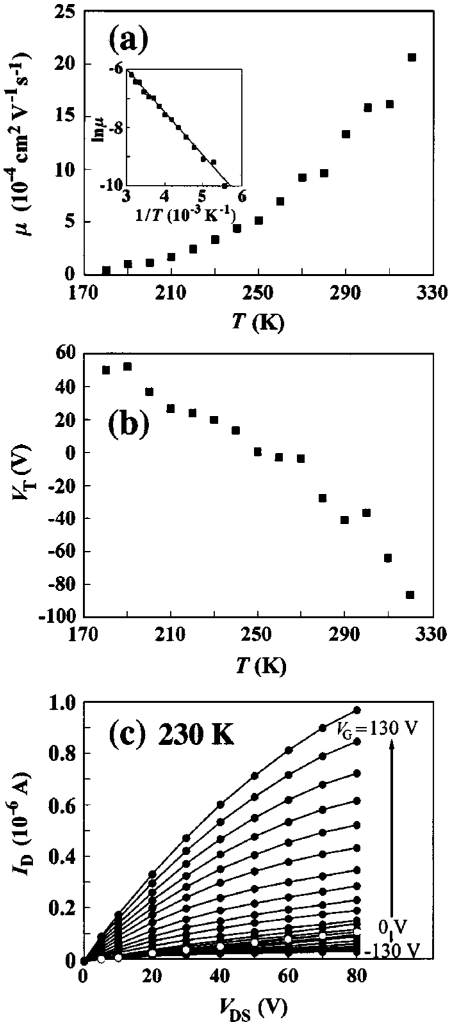

FIG. 2. (a) $\mu - T$ plot of C_{84} FET and ln μ-(1/T) plot in the inset. Solid line is the line fitted with the equation $\ln \mu \sim E_a / k_B T$, where k_B is Boltzmann constant. (b) $V_T - T$ plot of C_{84} FET. (c) $I_D - V_{DS}$ plots of C_{84} FET at 230 K. Closed and open circles refer to the points measured at $V_G \neq 0$ V and $V_G = 0$ V, respectively.

plots show output characteristics of n-channel normally-on depletion-type FET. Relatively large I_D is observed even at zero gate voltage V_G. By varying V_G from -130 to 130 V, I_D increases as V_G increases as shown in Fig. 1(b). It can be seen that the channel conductance decreases by applying a negative V_G, presumably due to the depletion of carriers in the channel. I_D remained constant below $V_G = -70$ V. This can be explained by assuming the existence of bulk current which cannot be reduced by applying the negative V_G. The threshold voltage V_T was estimated to be -42 V from the $I_D - V_G$ plot at $V_{DS} = 10$ V [Fig. 1(c)]. The negative V_T supports that the C_{84} FET is normally-on type. Very recently such a normally-on depletion-type property was observed in $La_2@C_{80}$ FET.[9]

The field-effect mobility for the C_{84} FET was estimated to be 1.1×10^{-3} cm^2 V^{-1} s^{-1} from the $I_D - V_G$ plot [Fig. 1(c)]. This value is higher by one order of magnitude than those reported for $Dy@C_{82}$ and $La_2@C_{80}$ FETs.[8,9] This implies that C_{84} is superior to metallofullerenes such as $Dy@C_{82}$ and $La_2@C_{80}$ as materials for an active layer in normally-on depletion-type FET. The μ increases monotonically as temperature increases up to 320 K [Fig. 2(a)]; the mobility reached 2.1×10^{-3} cm^2 V^{-1} s^{-1} at 320 K. FET behavior was not clearly observed below 180 K because of very small I_D. The activation energy E_a was estimated to be 0.13 eV from the ln μ versus T^{-1} plot shown in the inset of Fig. 2(a). These results show that the channel conduction of the C_{84} FET device follows a thermally activated hopping-transport model ($\mu \sim \exp - E_a / k_B T$). One of the origins of high μ in the C_{84} FET may be attributed to the fact that the channel conduction occurs through a hopping between the delocalized lowest unoccupied molecular orbitals (LUMOs) in C_{84}, which is different from that between the localized LUMOs dominated by encapsulated metal ions in metallofullerenes.[9]

The threshold voltage V_T decreases monotonically with increasing temperature up to 320 K [Fig. 2(b)]. Such temperature dependence might be caused by an increase in bulk current. Actually the bulk current increased with increasing temperature,[11] which results in an apparent variation in V_T. The V_T values are positive at temperatures below 250 K, and the FET property apparently changes from the normally-on to a normally-off with decreasing temperature. As an example of the normally-off like FET, the I_D–V_{DS} plots at 230 K are shown in Fig. 2(c).

The high I_D at $V_G = 0$ V in the high-temperature region can be explained by a small gap energy, E_g, of ~0.55 eV which was estimated from the temperature dependence of resistivity ρ for the C_{84} thin-film ($V_G = 0$ V).[11] The E_g value of the C_{84} thin film is much smaller than those determined for C_{60} (1.8 or 2.1 eV) and C_{70} (2.2 eV).[12–14] The C_{60} and C_{70} FETs showed enhancement-type normally-off properties.[5,6,8,10] Consequently, the normally-on type FET property in the C_{84} FET device can be interpreted within the framework of high bulk current in the C_{84} thin film. However, the origin of carriers of the high bulk current in the C_{84} thin film remains to be clarified. The on–off ratio, I_D ($V_G = 130$ V)/I_D ($V_G = -130$ V), at $V_{DS} = 10$ V of the C_{84} FET was ~6 at 290 K, as shown in Fig. 1(b). This low on–off ratio is owing to high bulk current of the C_{84} thin film. On the other hand, the on–off ratio increases at low temperatures, and the value reached ~37 at 230 K [Fig. 2(c)]. This increment is caused by the reduction of bulk current at low temperature. This will open a way to fabricate practical devices with higher fullerene FETs for computing and memory.

Y.K. acknowledges Dr. Yoichi Nonogaki of the Institute for Molecular Science for his valuable discussion. The authors appreciate financial supports from CREST of Japan Science and Technology Corporation and from Mitsubishi foundation, and a Grant-in-Aid (15350089) of Ministry of Education, Culture, Sports, Science and Technology, Japan.

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