

Research paper

Highly bendable luminescent semiconducting organic single crystal



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ABSTRACT

Mechanical flexibility is one of the key benefits of organic electronics and optoelectronics. Although organic single crystals show the best performance in field-effect and light-emitting transistors, they are commonly considered to be brittle. In this work, we show that organic single crystals can be flexible, luminescent and semiconducting. We grew ~1 cm-size single crystals of a thiophene/phenylene co-oligomer, 1,4-bis(5'-hexyl-2,2'-bithiophene-5-yl)benzene, from solution and resolved their structure by x-ray analysis. The crystals were bendable with a radius down to 0.2 mm corresponding to a mechanical strain of 5%, had molecularly flat surface, showed the photoluminescence quantum yield up to 17% and charge mobility up to 0.07 cm²/Vs. Multiple bending cycles of the crystals did not affect their photoluminescence but resulted in minor degradation of the charge mobility. These findings indicate that organic single crystals can be a platform for flexible optoelectronic devices.

1. Introduction

Organic semiconducting single crystals provide the highest performance in various electronic and optoelectronic devices [1–4] because of their highly ordered structure. However, single crystals are usually considered to be highly brittle and only very thin crystals were implemented in flexible devices. For example, Briseno and co-workers demonstrated 150nm-thick rubrene single crystal organic field-effect transistors (OFETs) on flexible kapton substrates [5]; Raghuvanshi et al. reported flexible TIPS-pentacene single crystals which could be bent to a radius of 5 mm, and the OFETs performance was slightly affected by the mechanical strain [6]. Fang et al. demonstrated a flexible optically-pumped laser based on 200-nm-thick single crystal of 1,4-bis(4-methylstyryl)benzene with bending radius c.a. 1 cm [7]. However, organic single crystals combining high flexibility, semiconducting and luminescent properties have not yet been reported.

Single crystals of thiophene/phenylene co-oligomers (TPCOs) are among the most promising materials for various optoelectronic devices such as organic light emitting transistors and electrically-driven organic

lasers [2,3,8] as they combine efficient charge transport [3] and high luminescence efficiency [9,10]. By synthetic variation of the number/position [8] of thiophene and phenylene units as well as the terminal substituents [11–13] TPCO materials with various charge-transport and emission properties have been demonstrated [3].

In this work, we report on solution-processed single crystals combining high mechanical flexibility with semiconducting and light-emitting properties. As a material, we used a TPCO 1,4-bis(5'-hexyl-2,2'-bithiophene-5-yl)benzene with five conjugated rings TTPTT (T – thiophene, P – phenylene) terminated by hexyl groups (Fig. 1e) referred below as DH-TTPTT. DH-TTPTT single crystals were grown, and their structure was solved by x-ray analysis. The ~20-μm-thick DH-TTPTT crystals had the molecularly flat surface, showed yellow photoluminescence (PL) with quantum yield (QY) up to 17%, charge-carrier mobility up to 0.07 cm²/Vs, and were mechanically bendable to a radius as low as 0.2 mm.

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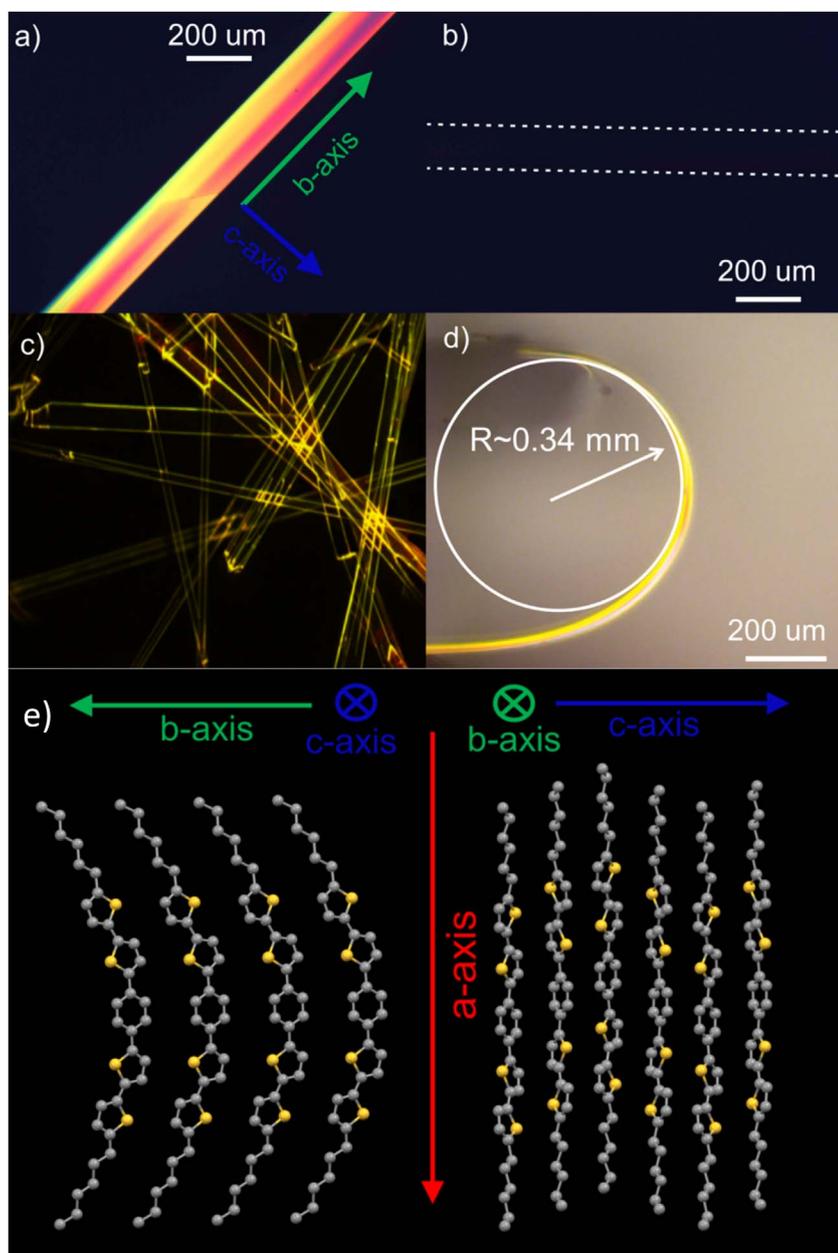


Fig. 1. Optical characterization of DH-TTPPT single crystals and their x-ray structure. Optical images of single crystals: in transmitted light through the crossed polarizers oriented at 45° (a) and 90° (b) with respect to the crystal plate; in UV (405 nm) irradiation (c); bent to a radius of ~0.34 mm (d). Crystal structure (e); the arrows indicate the orientation of crystallographic axes.

2. Experimental

DH-TTPPT (1,4-bis(5'-hexyl-2,2'-bithiene-5-yl)benzene) was prepared by Kumada reaction from 5'-hexyl-2,2'-bithienyl-5-yl magnesium bromide and 1,4-dibromobenzene in the presence of Pd(dppf)Cl₂ as a catalyst in dry tetrahydrofuran (THF) similar to the method described in ref [14]. The molecular structure and purity of the final product were proved by ¹H NMR spectroscopy and elemental analysis. ¹H NMR (250 MHz, CDCl₃, TMS/ppm): 0.89 (t, 6H, *J* = 6.7 Hz), 1.23–1.45 (overlapped peaks, 12H), 1.68 (m, 4H), 2.79 (t, 4H, *J* = 7.3 Hz), 6.68 (d, 2H, *J* = 4.3 Hz), 7.01 (d, 2H, *J* = 3.7 Hz), 7.05 (d, 2H, *J* = 3.7 Hz), 7.21 (d, 2H, *J* = 3.7 Hz), 7.57 (s, 4H). Calc. for C₃₄H₃₈S₄: C, 71.03; H, 6.66; S, 22.31. Found: C, 71.00; H, 6.68; S, 22.26%.

2.1. Crystal growth and analysis

DH-TTPPT was crystallized using solvent-antisolvent crystallization technique [13,15]. DH-TTPPT powder was dissolved in 1,2-dichlorobenzene (0.5 g/l) at room temperature using an ultrasonic bath. The

solution was filtered through a 0.2-μm PTFE filter and the vial with the solution was placed into a closed vessel containing isopropanol. The growth period was 2 days. The crystals were examined using a transmission optical microscope (Motic BA210) through crossed polarizers. For further studies the crystals with minimal number of visible defects (cracks, outgrowths, etc.) were selected. The flexibility of the crystals was studied in reflected light using an optical microscope (Axio Imager 2, Zeiss). A single crystal elongated plate was bent by pushing it at one end, whereas the other end was fixed. The smallest bending radius before crystal breaking was measured from the crystal image. To study the bending effect on luminescent and charge transport properties of DH-TTPPT crystals, they were exposed to multiple bending cycles with subsequent measurements of their PL intensity and OFET characteristics on the flat crystals.

The top surface of DH-TTPPT single crystals was investigated with an atomic force microscope (Integra Spectra, NT-MDT) in the tapping mode using HQ:XSC11/Hard/Al-C probes (MicroMash).

The x-ray diffraction data for DH-TTPPT single crystals were collected at 110 K by STOE diffractometer with Pilatus100 K detector

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