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Organic optoelectronics: materials, devices and applications

LIU Yi, CUI Tian-hong

(*Department of Mechanical Engineering*, *University of*
Minnesota 111 *Church Street SE*, *Minneapolis*, *MN* 55455, *USA*)

Abstract: The interest in organic materials for optoelectronic devices has been growing rapidly in the last two decades. This growth has been propelled by the exciting advances in organic thin films for displays, low-cost electronic circuits, etc. An increasing number of products employing organic electronic devices have become commercialized, which has stimulated the age of organic optoelectronics.

This paper reviews the recent progress in organic optoelectronic technology. First, organic light emitting electroluminescent materials are introduced. Next, the three kinds of most important organic optoelectronic devices are summarized, including light emitting diode, organic photovoltaic cell, and photodetectors. The various applications of these devices are also reviewed and discussed in detail. Finally, the market and future development of optoelectronic devices are also demonstrated.

Key words: organic optoelectronics; material; device; application; overview

1　Introduction

Before the discovery of electrically conductive properties through doping, polymers were widely used as electrical insulators due to the superior insulating properties they exhibited. In 1977, the discovery of the first highly conductive polymer, chemically and electrochemically doped polyacetylene, was reported[1-2]. The discovery of doped polyacetylene has opened an entire new field for polymers and organic materials in their applications to both conductors and semiconductors. Although the initial emphasis was on the conductive properties obtained by the doping of conjugated polymers, over a decade the research has mainly focused on soluble and intrinsically conducting and semiconductive polymers. Many novel materials have been developed for their specific physical or chemical properties, and implemented in a variety of applications.

Electroluminescence (EL), the emission of light upon excitation by the flow of electric current, is a phenomenon that has been observed in a wide range of organic and inorganic semiconductors. In early 1960s, the first commercial display device employing inorganic semiconductor materials GaAsP was fabricated[3]. Electroluminescence from organic materials was first discovered in 1963[4]. Due to the very low electroluminescent efficiency of such single layered structures and very high driven voltage needed, very little activity was reported for a long period of time. The next report since the discovery of organic EL devices was from Tang and VanSlyke in 1987, and PhD described the first organic light emitting diodes (OLED). The OLED described in the 1987's paper produced light with a-

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bout 1 percent efficiency[5]. In 1990 the Cambridge group reported the emission of light from a plastic sandwich that operated at a low voltage[6]. The discovery of electroluminescence in conjugated polymers brought new interests to the development of polymer light-emitting diodes (PLEDs) for display and other applications. The history of EL devices can be seen from Fig. 1[7].

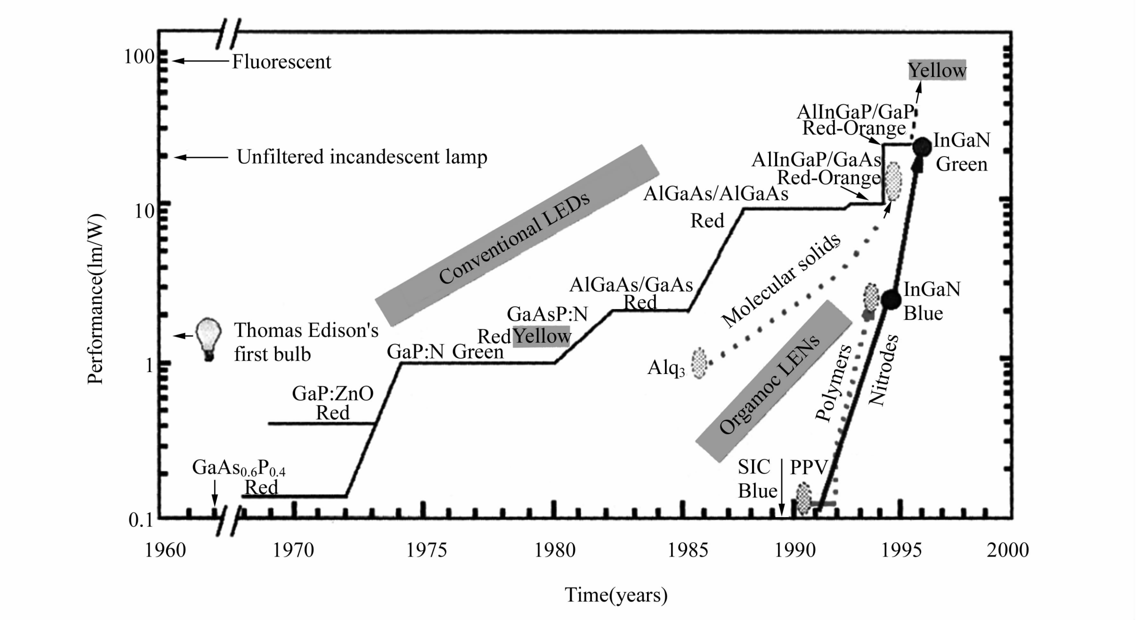


Fig. 1 Development of LED devices[7]

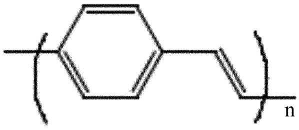
Electroluminescent organic and polymeric materials have many physical advantages over inorganic electroluminescent materials. Electroluminescent inorganic semiconducting materials have to be deposited by sublimation or vapor deposition under high vacuum, which is expensive and not suitable for the mass production of large-area displays. In contrast, uniform thin films of conjugated polymers can be deposited from diluted solutions in proper solvents by the process of spin coating or inkjet printing. Furthermore, the production of copolymers from a mixture of different monomers allows the fine-tuning of the physical properties for special interests.

In 1986, Wang reported the first results on organic-based photovoltaic cells, the principles of which can be regarded as the inverse of the electroluminescence process[8]. The novel work of Wang's created substantial interests in polymer and organic photovoltaic devices (PVDs). A photovoltaic cell is a specialized semiconductor diode that converts visible light into direct current (DC). Some photovoltaic cells can also convert infrared (IR) or ultraviolet (UV) radiation into DC electricity. Polymer photovoltaic offers a great technological potential as a renewable, alternative source of electrical energy. The potential advantages of polymer solar cell include: low cost, large area, flexibility, etc. The materials used for the production of polymer photovoltaic cells are hydrocarbon-based non-toxic molecules. These materials can easily be manufactured into thin film solar cells at room temperature without the production of dangerous exhaust gases. Since the band gap of polymeric semiconductors can be controlled and manipulated, it is possible to change the color of solar cells, which in return is uniquely interesting for architecture[9]. This can also allow unused large areas in addition to standard rooftops of buildings to be accessed for photovoltaic energy conversion by fabricating semitransparent photoactive thin films. These potential advantages bring a legitimate relevance for industrial interests to polymer photovoltaic devices.

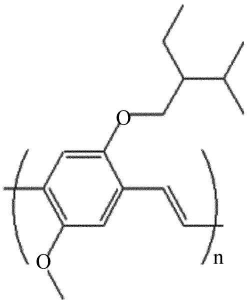
This paper reviews the recent progress of organic optoelectronics in the broad area of electroluminescence to photovoltaic effect. Materials, devices and various applications are to be discussed. Some recent reviews are also referred to the readers[7,10-13].

2 Organic optoelectronic materials

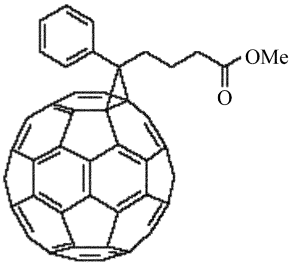
Conjugated organics and polymers have alternating single and double carbon-carbon (sometimes carbon-nitrogen) bonds. Single bonds are referred to as σ-bonds, and double bonds contain an σ-bond and a π-bond. The characteristics of the -bonds are the source of the semiconducting properties of these polymers. The -bonds are delocalized over the entire molecule and produces two orbits, a bonding (π) orbit and an anti-bonding (π^*) orbit. The lower energy -orbital produces the highest occupied molecular orbit (HOMO), and the higher energy π^*-orbital forms the lowest unoccupied molecular orbit (LUMO). The difference in energy between the two levels produces the band gap that determines the optical properties of the material. The structures of some popular optoelectronic polymers are shown in Fig. 2.



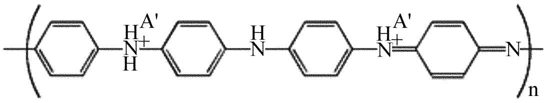
2a.PPV



2b.MEH-PPV



2c.PCBM



2d.PANI

Fig. 2 Structures of some polymer optoelectronic materials

The first discovered light electroluminescent polymer is poly(p-phenylenevinylenes) (PPV, shown in Fig. 2(a)), which was reported by J. H. Burroughes, et al. in 1990[6]. PPV can produce yellow-green luminescence during photoexcitation, and can also be used for photovoltaic devices. Marks and his co-workers described the fabrication of PPV-containing photodiodes in which PPV layer was obtained by spin coating on the sulfonium salt precursor, followed by heating the polymer to 250 ℃ in vacuum[14]. One of the popular light emitting polymers that can produce orange-red light is poly[2-methoxy-5-(2 prime-ethylhexyloxy)-1, 4- phenylene vinylene] (MEH-PPV, Fig. 2(b)). Davids, et al. reported the PLED based on MEH-PPV, and also demonstrated device model calculations for the current-voltage characteristics of the polymer diodes[15]. They found that the low mobility of the organic materials leads to a large backflow of injected carriers into the injecting contact. One popular conjugated polymer served as the electron acceptor for photovoltanic cell is [6,6]-phenyl C_{60} butyric acid methyl ester (PCBM, Fig. 2(c)). Zhang, et al. has investigated the influence of buffer layers on the performance of PCBM solar cells[16]. Polyaniline (PANI, Fig. 2(d)) was used as the hole transport material to fabricate solid-state dye-sensitized TiO_2 solar cells, which had an open-circuit voltage up to 310 mV and a short-circuit current up to 21 μA/cm^2 under solar radiation[17].

3 Organic light emitting diode

3.1 Operation physics

The basic structure of an organic LED includes one or more organic films between two electrodes, one of which is transparent. Fig. 3 shows the basic physics of a polymer light emitting diode by means of a scheme of energy levels[18]. Under a forward bias, electrons are injected into the polymer from cathode, and holes are injected from anode. The charge carriers driven by an applied electric field move along the polymer chain, then recombination occurs and light is emitted from the transparent electrode. Therefore, three processes are involved within the PLED: charge injection, charge transport, and recombination.

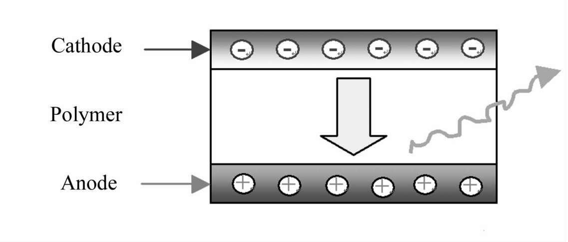


Fig. 3 Schematic of energy diagram for a single layer PLED

Electrons are injected into the HOMO and holes are injected into the LUMO of the polymeric semiconductor using metallic electrodes. The injected electrons and holes can then diffuse towards each other and finally recombine, and create neutral excitations at the same time. These neutral excitations are bond states of electron-hole pairs, and can easily decay into their ground state, generating a characteristic fluorescence simultaneously. The excitons may be either in a singlet-state that spontaneously generates fluorescence or in a triplet-state that are created due to spin elections. When electrons and holes that flow in opposite directions between two electrodes meet, the electrons will recombine with the holes and release their extra energy as light. At greater difference in energy between the holes and the electrons, the light will give off further from red to blue[18].

Fig. 4 shows the structure of the first PLED. It consisted of a thin layer of PPV between two electrodes, deposited on a glass substrate. The cathode was chosen to be aluminum or calcium, which has a low work function. It can easily withdraw an electron from a metal surface, and is very efficient at injecting electrons from the cathode into the PPV layer. The anode is a layer of indium tin oxide (ITO), which has a relatively high work function and is a good material for injecting holes into the PPV layer. Recombination occurs at the layer interfaces, and then the generated light will exit the device through transparent electrode.

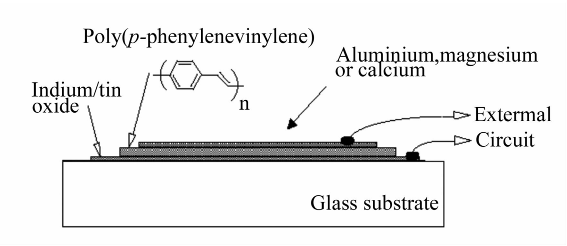


Fig. 4 Structures of the first reported light emitting polymer and PLED[6].

3.2 Fabrication and characterization

Organic thin films within light emitting diode can be fabricated by evaporation, spin-coating, inkjet-printing, screen-printing, self-assembly, etc. Ink-jet printing technology has gained a lot of interests, and is widely investigated for PLEDs fabrication. Hebner, et al. reported the patterned luminescent doped polymers of PVK films, with dyes of coumarin 6, coumarin 27 and nile red, which were directly deposited by ink-jet printing[19]. Hybrid ink-jet printing (HIJP) technology was developed for electrolumnescent devices by Yang's group at University of California, Los Angeles[20]. This technology consists of an ink-jet printed layer in conjunction with another uniform spin-coated polymer layer. This uniform layer called the buffer layer serves to seal the pin holes while the ink-jet printed layer consists of the desired patterns. Using this method, polymer light-emitting logo, dual-color polymer light-emitting diodes and multicolor organic light-emitting diodes were fabricated. An example of polymer light-emitting logo via ink-jet printing is shown in Fig. 5.

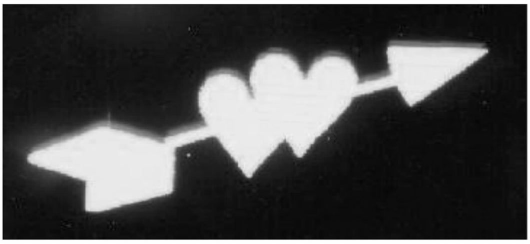


Fig. 5 Polymer light-emitting device (logo) by ink-jet printing[20].

3.3 Applications

3.3.1 Full color display

Forrest, et al. reported a stacked OLED (SOLED) in which color, grey scale, and intensity can be independently achieved[21]. The device consists of three separately contacted, red-, green- and blue-emitting OLEDs placed in a vertical stack and separated by transparent, conducting electrodes. The SOLED could generate a high-intensity optical output whose color was a linear superposition of spectra of the individual emitting elements in the device. Makoto Shibusawa, et al. developed a 17-inch WXGA full-color polymer OLED display using ink-jet printing method[22]. Both droplet volume and landing position were precisely controlled pixel by pixel. A uniform picture with 260,000 colors and wide viewing angle was achieved. Nijs C. Van Der Vaart, et al. developed a multi-head polymer OLED ink-jet-printing technology to make large-screen OLED television displays[23]. The printer was used to make a 13-in diagonal 16:9-format P/OLED prototype driven by an active matrix with a pixel circuit. A novel scrolling-bar addressing scheme was used to reduce motion artifacts and to make sparkling images with a high local peak brightness.

3.3.2 Flexible display

Chwang, et al. reported the encapsulated passive matrix OLED display on 178-μm-thick barrier coated polyethylene terephtalate (PET)[24]. A multilayer of Al_2O_3 and polyacrylate can protect the OLEDs from moisture and oxygen. The flexible display has a resolution of 80 dpi, and lifetime of 2 500 h under DC test. Sugimoto, et al. investigated the flexible OLED using plastic substrate[25]. Silicon nitride was used as the passivation film to protect the OLED from the moisture and oxygen. Silicon oxynitride (SiON) is served as the moisture barrier film on the plastic substrate to meet the requirements of moisture impermeability and optical transparency. The lifetime of the device on a plastic substrate could be more than 6 000 h. A 3-in full-color OLED display on a plastic substrate driven by a passive matrix method was also demonstrated. Kwon, et al. demonstrated the MEH-PPV LED on a flexible substrate, ITO-coated flexible PET[26]. The structure of the flexible PLED is shown in Fig. 6. Triple-layer passivation composed of high-density polyethylene (HDPE) and metal layers were used to make the diode more stable in ambient conditions. The luminance of 2 000 cd/m^2 was obtained at a current density of 165 mA/cm^2 (6.5 V).

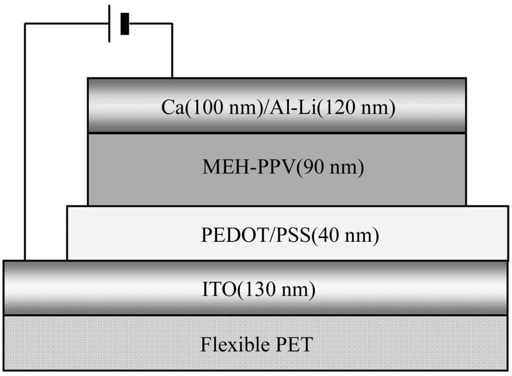


Fig. 6 Structure of the flexible MEH-PPV PLED[26].

4 Organic photovoltaic cells

4.1 Operation principle

Organic solar cells normally have a planar-layer structure, in which the organic light-absorbing layer is sandwiched between two electrodes. Its operational principle is the reverse of the principle in light emitting diode. When the active organic layer is illuminated, an electron is promoted from the HOMO to the LUMO, and forms an exciton, or an electron-hole pair. The resulted electron-hole pair needs to be dissociated into separate charges, which is called exciton dissociation. This can be driven by the built-in potential field between two electrodes with asymmetrical work functions. To dissociate the exciton, materials to function as an electron donor and an electron acceptor need to be used, in which the electron affinity of the electron acceptor material should be larger than the ionization potential of the donor material. The light harvesting process in the organic photovoltaic cell is illustrated in Fig. 7, where Φ is the work function.

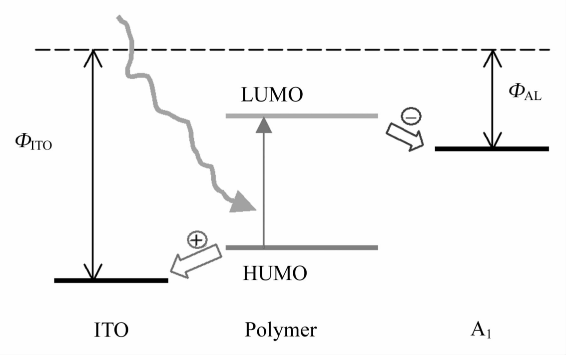


Fig. 7 Light harvesting process of organic PV cell

Efficiency is the essential parameter for solar cells, and is calculated by the following formula[28]:

$$\eta_{eff} = \frac{V_{OC} \cdot I_{SC} \cdot FF}{I_{LIGHT}}$$

where V_{OC} is the open circuit voltage, I_{SC} is the short circuit current density in A/m^2, FF is the fill factor, and I_{LIGHT} is the incident solar radiation in W/m^2. The fill factor of solar cells is determined by the following formula[28]:

$$FF = \frac{I(\times 10^{-6}) \cdot V(\times 10^{-6})}{I_{SC} \cdot V_{OC}}$$

where $I(\times 10^{-6})$ and $(V \times 10^{-6})$ are the current and voltage at the maximum power, respectively.

4.2 Fabrication and characterization

Krebs, et al. used industrial screen printing to produce large-area polymer solar cells[29]. Both the conducting silver epoxy for the electrical contacts and the active layer MEH-PPV were screen printed on the polyethyleneterephthalate substrate. The efficiency of the above solar cell was 0.0046% with an open-circuit voltage of 0.73 V and a short-circuit current density of 20 μAcm^{-2}. Alem, et al. fabricated the polymer PV cell based on polymer-interpenetrated network of MEH-PPV and [6,6]-phenyl C_{60} butyric acid methyl ester[30]. Spin-coating with surface treatment increased the performance of the devices, which reached the power consumption of 2.9%. Padinger, et al. fabricated the plastic PV cells by spin-casting the P3HT and PCBM as the active layers[31]. The postproduction treatment significantly increased the short-circuit current. A short-circuit current of 8.5 $mAcm^{-2}$ and an external quantum efficiency of 70% were demonstrated by the authors. Xue, et al. recently reported the organic PV cell with power energy efficiency as high as 4.2%[32]. The active organic layers of the device were made of copper phthalocyanine/C_{60} and were deposited by thermal evaporation. Xue, et al. predicted that the dominance of the bulk resistance of the ITO anode as a limiting factor in practical cell efficiencies. Later in this year, this group further improved the power efficiency of the organic PV cell up to 5.7% using hybrid planar-mixed molecular heterojunctions[33].

4.3 Applications - flexible organic solar cell

Kushto, et al. reported organic photovoltaic devices on fully flexible thermoplastic substrates using pre-patterned conducting polymer as electrodes[34]. N, N prime - (α-naphthyl) -N, N prime -diphenyl-1, 1 prime -biphenyl-4, 4 prime -diamine (α-NPD) worked as the electron donating /hole transporting material, and C_{60} served as the electron accepting /transporting material. The white-light power conversion efficiency could be up to 1%. Brabec, et al. presented solar cells on flexible ITO polyester substrates based on a soluble alkoxy poly(para phenylenevinylene) (MDMO-PPV) and a highly soluble fullerene derivative (PCBM)[35]. The energy conversion efficiency was larger than 1% and charge carrier collection efficiency was 20%. Maher Al-Ibrahim, et al. reported the cells using poly[2-methoxy-5-(3′,7′-dimethyloctyloxy)-1, 4-phenylenevinylene]: [6, 6]-phenyl-C_{61}-butyric acid methyl ester ($\omega = 1:3$) as a photoactive layer on flexible 5 cm×5 cm polyester foils[36]. Using these materials, a white-light power conversion efficiency of 3% was achieved.

5 Organic photodetector

In addition to the energy conversion, polymer diodes in a sandwich configuration can also be used for light detection. The principle of organic photodetector is similar to the photovoltaic cell, except for the parameters used to evaluate the device. The major parameters used to evaluate a photodetector include spectral response, photosensitivity (or quantum efficiency), dark current, dynamic range, noise equivalent power, response speed, etc. [37]. The spectral responses of polymer photodetector cover from visible to UV light.

Halls, et. al reported the interpenetrating network formed from a phase-segregated mixture of two semiconducting polymers[38]. This network provided both the spatially distributed interfaces necessary for efficient charge photogeneration, and the means for separately collecting electrons and holes. The detailed configuration is shown in Fig. 8. Devices using thin films of these polymer mixtures could be used for large-area photodetectors.

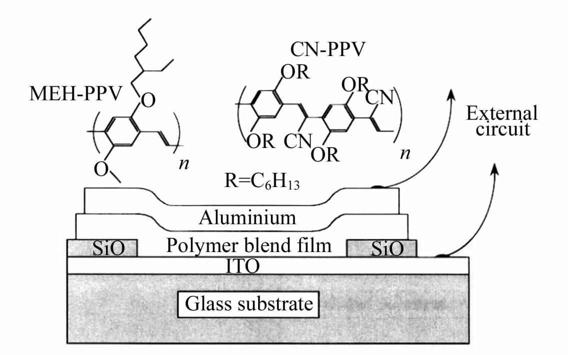


Fig. 8 Schematic diagram of the polymer photodiode[38]

Peumans, et al. reported the organic photodetectors incorporating an ultrathin donor-acceptor alternating multilayer stack as the optically active region[39]. The stacks exhibited an external quantum efficiency of 75% across the visible spectrum, and had response times below a nanosecond. The photogenerated carriers rapidly transported to their repective electrodes before charge recombination or deep trapping occurs, yielding high carrier collection efficiencies. Qi, et al. added the PbSe nanocrystals into MEH-PPV and observed a dramatic enhancement of the quantum efficiency of the polymer-based photodetectors[40]. The observed photocurrent gain could be attributed to the carrier multiplication in PbSe nanocrystals via multiple exciton generation, and the efficient charge conduction was through the host polymer material.

6 Market and outlook

Organic materials have transformed the world of circuit and display technology as never before. The display marketplace exhibits average annual growth rates between 12% and 13%, ensuring a vast market for OLEDs once certain challenges can be overcome[41]. Many corporations, such as Kodak, Sanyo, Philips and Pioneer Electronics, Cambridge Display Technology Corp., Universal Display Corp., Uniax, etc. are vigorously attempting to mine the tremendous opportunity for the low-cost organic electronic and optoelectronic devices. In 2003, Kodak announced the world′s first digital camera with OLED display.

OLED materials and displays have made significant improvements in lifetime and operational stability. Up to date, lifetimes are about 8 000 h, almost long enough to last for one year of continuous use. When OLED displays can reach the 10 000 h range, more applications and potential profits will be explored, including the use of OLEDs in main cell phone displays.

Worldwide OLED panel market revenue was $251 million in 2003, and then expanded to $408 million in 2004[42]. Thirty one million OLED panels were sold in 2004, which was nearly double of that in 2003. With the rising usage of OLEDs as an alternative to LCDs for mobile-phone displays, the market is expected to grow to $615 million in 2005. Looking into the long-term market, as shown in Fig. 9, the OLED market is forecasted to rise to $2.9 billion in 2011.

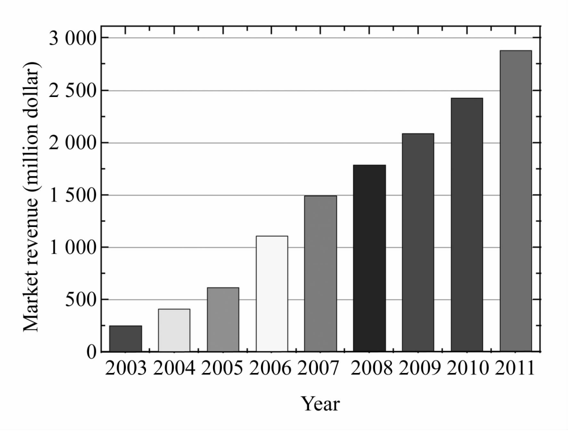


Fig. 9 OLED display forecast (Courtesy of iSuppli Corp.)

Using organic light-emitting devices, organic full-color displays may eventually replace liquid-crystal displays (LCDs) for the applications to cell phones, laptop and even desktop computers. Such displays can also be deposited on flexible plastic foils, eliminating the fragile and heavy glass substrates used in LCDs, and can emit bright light without the pronounced directionality inherent in LCD[43]. The future of organic optoelectronics shows tremendous potentials and a huge market.

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Brief professional biographies of the authors:

LIU Yi received the B.S. degree in telecommunication engineering from Beijing University of Posts and Telecommunications, Beijing, China, in 1998. He is currently a Ph.D. candidate at Department of Mechanical Engineering, University of Minnesota, Minneapolis, Minnesota. His main research interests include micro/nanofabrication, polymer/nanoparticle based microelectronic devices, and biosensors.

CUI Tian-hong received the B.S. degree from Nanjing University of Aeronautics and Astronautics in 1991, and the Ph.D. degree from Changchun Institute of Optics and Fine Mechanics, Chinese Academy of Sciences in 1995. He is currently a Nelson Associate Professor of Mechanical Engineering and Nanofabrication Center at the University of Minnesota. From 1999 to 2003, he was an Assistant professor of Electrical Engineering and Institute for Micromanufacturing at Louisiana Tech University. Prior to that, he was a STA fellow at National Laboratory of Metrology in Japan, and served as a Postdoctoral Research Associate at the University of Minnesota and Tsinghua served as a Postdoctoral Research Associate at the University of Minnesota and Tsinghua University, respectively. He has more than 100 publications and 2 US patents pending. He received many research awards including Richard and Barbara Nelson Professorship from the University of Minnesota, Research Foundation Award from Louisiana Tech University, and STA & NEDO fellowships in Japan. His current research interests include MEMS/NEMS and nanotechnology.