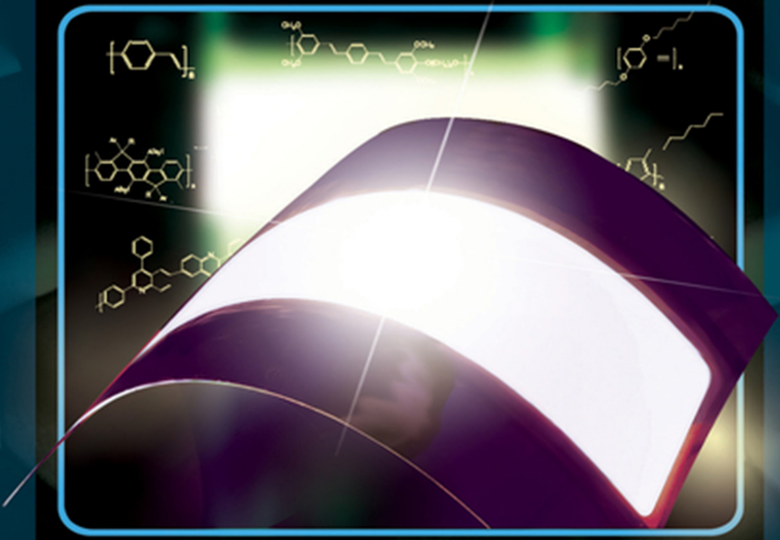
POLYMERS *for* LIGHT-EMITTING DEVICES *and* DISPLAYS



Edited by Inamuddin, Rajender Boddula,
Mohd Imran Ahamed, & Abdullah M. Asiri

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Contents

Preface

Polymer light-emitting diodes (PLEDs) or organic light-emitting diodes (OLEDs) are organic semiconductor light sources that emit light in response to an electric current. These PLEDs are promising devices with the aforementioned features to convert electrical energy to light energy, which is the necessary component of any display technology. OLEDs have received increasing attention since they were first developed in 1989. The development of OLEDs has attracted considerable interest in innovations for our daily life and future. They have promising applications in flat panel displays, electronic products, automotive, flexible displays, industrial products, and future wearables due to unique electrical and optical properties including low-cost, easy processing, low-operating voltage, energy-saving, eco-friendliness, thinner and smaller in size, lightweight, flexibility, and cost-effective fabrication process.

Polymers for Light-Emitting Devices and Displays provides an in-depth overview of fabrication methods and unique properties of polymeric semiconductors, and its potential applications for LEDs, organic electronics, displays, optoelectronics, and so on. Engineers, chemists, material science, and research scholars, students, and faculty members working in the area of organic electronics will benefit by understanding the materials used in optoelectronics. Based on thematic topics, the book edition contains the following eight chapters:

Chapter 1 is a detailed summary of the working principles of PLEDs. Different polymers used in PLEDs and limitations of PLEDs in the illumination system where the intensity of light is very high compared with displays like televisions and laptops are discussed.

Chapter 2 presents an overview of the newest polymeric materials and processes beyond the classical structure of PLED, leading to the low-cost and all-solution processed devices with enhanced parameters, which are closer to commercial production line requirements and custom needs.

Chapter 3 seeks to obtain a better understanding of the fluorescence quenching behavior and intramolecular charge transfer (ICT) character of two kinds of cyclopentadithiophene (CPDT) derivatives. A comparative study based on the optoelectronic properties of CPDT dimers for their highly efficient blue emitters in OLEDs is developed using the density functional theory (DFT) approach.

Chapter 4 deals with conjugated polymers and their application in the light-emitting diodes (OLEDs and PLEDs) as optoelectronic devices. It provides basic information on the classification of polymers and their modification via functionalization, copolymerization, doping, etc., for device fabrication, function, and use of conjugated polymers in blue, red, green, and multicolored light-emitting diodes along with challenges and their future perspectives. Additionally, the chapter focuses on the advantages/disadvantages and application of OLED technology in various fields.

Chapter 5 discusses the novel and noteworthy work carried out on electrospun nanofibers used for LEDs. It mainly focuses on the fabrication technology for producing electrospun nanofibers and how metal oxide semiconducting, perovskite, rare earth ion-doped, and coordination polymeric electrospun nanofibers are useful in designing smart clothes and LEDs.

Chapter 6 summarizes the roles of diversified architectures, layers, components, and their structural modifications in determining efficiencies and parameters of PLEDs as high-performance devices. Additionally, some recently developed materials and concepts, including white PLEDs, quantum dots, thermally activated delayed fluorescence, and transparent PLED, are discussed in detail.

Chapter 7 gives a general idea of polymer liquid crystal devices (PLCs), their synthesis, and applications in various liquid crystal devices (LCs) and displays.

Chapter 8 reviews the state-of-art of materials and technologies to manufacture hybrid white light-emitting diodes based on inorganic light sources and organic wavelength converters. It takes stock of the benefits—but also the weak spots—of the hybrid technology to envisage its future impact among the well-established inorganic lighting technologies.

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Applications of Polymer Light-Emitting Devices and Displays

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Abstract

This chapter gives information of polymer light-emitting diodes (PLEDs) and their applications. Besides, background, types, and the development of PLEDs also discussed. Further, the behavior of different PLEDs has been discussed with respect to various parameters, brightness, color purity, light conversion efficiency, and color stability are discussed.

Keywords: Polymer, light-emitting diodes, efficiency, color purity

1.1 Introduction

In the past one decade, the display technology has undergone several technological advancements and industries and household are looking for low cost, flexible, power efficient, and durable displays. Polymer light-emitting diodes (PLEDs), which convert electric energy into light, are promising devices with aforementioned features to convert electrical energy to light energy, which is the necessary component of any display technology. High temperature resistance, short response time, smooth brightness, and a large viewing angle are the additional advantages with PLEDs [1].

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These special characteristics of PLEDs give the scope to use them in the applications where a large array of displays is required [2]. At present, inorganic light-emitting diodes are widely used. The advancement of technology demands advancement of display also, some times, the display device needs to be flexible, this flexibility can be easily provided by PLEDs. In this chapter, the basic structure of PLED, the mechanism of light emission, and different applications are discussed.

1.2 Background

In 1990, an article was first published in Nature on “Light-emitting polymers” by J. H. Burroughes, Richard Friend, and others [3].

Basic structure of PLED

The basic structure of a PLED is illustrated in Figure 1.1. It consists of thin layers of light-emitting polymers film sandwiched between a transparent electrode which is anode and a non-transparent electrode which is cathode. Indium tin oxide (ITO) layer coated on glass substrate is most commonly used as the transparent anode. The glass provides the mechanical support for the PLED. ITO being transparent to light allows the light photon created inside the diode to escape from the device. There are two polymer layers in a typical PLED structure; among them is the hole transporting layer and the other is the light-emitting layer. Generally, the metal cathode is deposited over of the polymers by means of thermal evaporation.

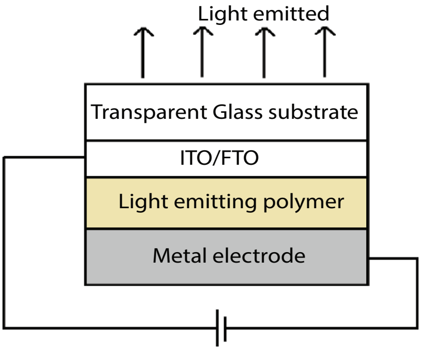

Figure 1.1 The basic structure of a PLED.

1.3 The Mechanism of Light Emission

Electron-hole recombination causes emission of a photon in visible region. Electrons are injected from the cathode to the LUMO (lowest unoccupied molecular orbit) and the holes are injected from the anode to the HOMO (highest occupied molecular orbit) of a conducting polymer. The reliability and the efficiency of the diode are strongly influenced by the materials which form the cathode, anode, and the emissive layers. A typical PLED may either be a single- layer device or a multilayer device. One example of PLED is the one fabricated from conjugated polymers including polyacetylene, polythiophene polypyrrole (PPy), poly (para-phenylene vinylene), and polyaniline (PANI) [4]. Another example of the active element used in PLED is the poly (p-phenylene vinylene) (PPV).

PLEDs like polymer light-emitting diodes and polymer light-emitting electrochemical cells gain huge interests owing to their high capabilities to serve as next generation illuminants and displays. Contrasted to inorganic light-emitting materials, conducting polymers possess very good film-forming behavior enabling the deposition uniformly by solution-based techniques, for example, screen printing and spin-coating that are competent of upscaling to industrial scale manufacturing. Polyfluorene, poly(p-phenylene vinylene), polycarbazole, and poly(p-phenylene) are widely researched; their solubility, morphology, stability, doping, etc., are proved to increase the device performance.

For example, poly(p-phenylene) films are widely synthesized by precursor methods because of its insolubility in commonly used organic solvents. Its solubility can be increased by the preparation of conducting polymers (ladder type) which leads to improved co-planarity [5]. Furthermore, a complete color display may be achieved through adjusting the structure of the molecule to regulate the energy gap of the HOMO-LUMO. Also, small amount of molecule doping proved to give desired luminous properties. Other than light emitting, color changes (electrochromic devices) and strain (electromechanical actuator) can also be stimulated by applying electric energy. Further, the electromechanical actuators directly convert the electrical energy to mechanical energy. These materials find applications in fabricating robotics, artificial muscles, etc. Electrochromic devices produce revocable color variation in reaction to the applied electric field, which makes it suitable for electronic skins and smart windows.

Owing to the tunable redox states under electricity, the conducting polymers are the fascinating materials for high performance electrochromic devices and electromechanical actuators. Further enhancement of the electrochromic or actuating ability of the polymer and the response speeds is improved by incorporating the graphene and other nanocarbon materials. For example, the multiwalled carbon nanotubes incorporated with polyaniline through an electrochemical deposition technique to aid as composite electrodes which can exhibit large conductivity ranges from 100 to 1,000 S/cm^{-1} that enables reversible and rapid electrochromic developments within short time [6]. PLEDs are not loaded with only merits. They do have a main disadvantage of weathering of the polymers with time. The disadvantage of the PLED technology is the sensitivity of the organic light-emitting materials to the atmospheric oxygen and water vapor. Hence, to protect PLEDs, a weather proof transparent polymer which is chemically and physically stable must be used for encapsulation.

1.4 Widely Used Polymers in PLED Applications

Among wide choices of polymers, some particular polymers gained special attention owing to their processability and functionality advantages that are discussed below. The advancement of important polymer material groups has been discussed here. This report gives the group of materials which can exhibit utmost potential till date to be espoused as the emissive materials in PLED applications, for example, the poly(fluorene)s, the poly(phenylenevinylene)s, etc. Polyfluorene homo- and co-polymers are purposefully emphasized because they are not well re-viewed, much progress has been made only recently, and this group of polymers are rapidly developed as a most promising viable LED polymeric material of widespread commercial interest.

1.4.1 Polyfluorene-Based Luminescent Polymers

Fukuda *et al.* reported the first fluorene-based polymers, by ferric chloride oxidative polymerization of 9-alkyl-fluorene and 9,9-dialkylfluorene [7, 8]. Their molecular weight was relatively low, with some-extent of separating and non-conjugated connections across locations except 2 and 7 [7, 8]. By using the transition-metal-catalyzed reactions of monomeric 2,7-dihalogenatedfluorenes, researchers introduced the homo-polymers for minimization of branching, improving regiospecificity. Further, Suzuki and co-workers discovered the palladium-catalyzed synthesis of mixed biphenyls from aryl bromide and phenylboronic acid [9, 10].

1.4.2 Polyfluorene Homo-Polymers

In general, polyfluorenes with substituents C6 or C9 are solvable in traditional organic diluters like aromatic hydro-carbons, chlorinated-hydrocarbons, etc. [11]. The polymers with large molecular weight does not consist separate glass transition. The polymers with straight alkyl substituents exhibit liquid crystallinity and tend to be semicrystalline. For example, F8, exhibits constant liquid crystallinity up to the temperature 270°C [12]; however, the polymer-materials having diverged alkyl-substituents exhibit non-crystallinity. Further, entire polymers while excited with UV emits a strong blue light, either in solution or in their solid state. They have a wide and drab absorption spectrum, whereas the photoluminescence spectrum shows distinct vibrionic-structures [13]. In general, the Stoke's shift lower than 50 MeV indicates a prolonged conformation.

1.4.3 Polyfluorene Alternating Copolymers

Tertiary aromatic amines are very good hole-transport materials, viable for photoconductors and LEDs. Preparation of large molecular weight, varying co-polymers containing different aromatic amines and 9,9-dialkylfluorene is possible through the Pd-catalyzed polymerization process. These alternating polymers are all soluble in conventional organic solvents, excellent film formers, and are good blue emitters. These polymer films exhibit discrete and adjustable oxidation capacities through cyclic-voltammetry that could be cycled exclusive of any significant alteration. The mobilities of positive charge carriers of the above said polymers are relatively large (3×10^{-4} to 1×10^{-3} cm^2/Vs) [14–16]. Due to these large mobilities of holes, these polymeric materials recommended for the applications in photoconductors also in LEDs for transportation of holes. Attempting to create polymers with distinctive properties, the alternating copolymer approach has been extended to other conjugated monomers, such as triarylamine, thiophene, etc. [11]. The co-polymers consisting large molecular weight exhibit high photoluminescence and emission spectra of the co-polymers may be associated with degree of co-monomers delocalization, e.g., the copolymers of bithiophene produce the spectra in yellow region, cyano-stilbene produces the spectra in green region, and thiophene in bluish green region [11].

1.4.4 Derivatives of PPV

The emission of yellow-green light by PPV under electrical stimulation was discovered in past decade, since then, several researchers focused on optimization of PPV and to make this as a potential material [17, 18, 19]. Most importantly, some of the advancements have taken place in preparation, regulating the balance in charge carriers, improving the efficiency in power, and enhancing the life-time, also in adjusting the emission of wavelength. Owing to the vinylene linkages, photo-oxidatively, the PPV chemical structure is unstable, also there is some restriction in the improving the saturated blue rich and red rich emitters. Even though these problems continuing to encounter the initiation of PPV into the display devices commercially, noteworthy development has been made towards the controlling and optimization of the PPV materials to make these are potential aspirants for the applications in PLED devices [11].

1.4.5 Soluble Precursors of PPV

For poly (arylene vinylene) series, the parent structure is PPV owing to absence of functional groups to improve solubility, rigid structure, and propensity to develop crystalline morphology, these materials are stubborn and directly not processable form the solution. Meanwhile for polymeric emission systems solvent process ability is a necessary characteristic; further, the soluble precursors to the PPV which can be molded as films then transformed to PPV through heating have been established.

1.4.6 Derivatives of PPV for Solution-Processing

PPVs are generally difficult to process but possess properties that are capable of good PLED candidates. To overcome the processing difficulty, plenty of research is devoted towards the advancement of soluble PPVs. Making of thin films is easy with soluble PPVs, exclusive of successive thermal-conversion. In 1991, Heeger *et al.* reported the applications of PPVs particularly 2,5-dialkoxy functional PPVs; according to them, the alkoxy groups having at least one bulky or long polymer groups are soluble in diverse organic solvents which include xylene, chloroform, etc. The functionality of the bulkier materials has been described to interrupt the propensity of PPV in order to increase the efficiency of EL.

1.4.7 Polyphenylenes

In the area of polymer light-emitting devices, there exist another class of conjugated polymer group called PPP (poly(1,4-phenylene)); these PPP materials consist large bandgap and permit blue light emission. Subsequently, the design of blue emitters which are having high efficiency and long life time endures a major task in advancement of the polymers. Therefore, activities of research in focused on PPP to emphasize the methods towards PPP thin-films through solvable precursor polymer materials which are thermally converted, in addition to the improvement of soluble PPPs. These class polymers have high molecular weights that are sufficient to mold films including excellent integrity that have been accomplished besides diodes with blue-emission that have been built and reported with considerable efficiencies [20].

1.5 Parameters to be Considered for Display Applications

The following parameters are considered for making different display technologies:

i. Color purity and brightness,
ii. Light conversion efficiency, and
iii. Durability.

1.5.1 Color Purity and Brightness

Entire spectrum of colors (and infrared) is possible with different PLEDs. For orange and green emitting PLEDs, life cycles of over 10,000 hours have been reported. However, till now the data is not available, blue devices with high cyclic life. This causes the loss of blue component of light emitted with time and the PLED will eventually lose the entire blue light, and hence, the visible light emitted cannot maintain color purity with time. So, a blue emitting polymer with lifetime equal to orange and green ones is barely needed in realizing a display device to maintain the emission color through the lifetime of the device.

High luminance values may be achieved at small voltages. For orange PLEDs, the observed starting value of emission is of around 1.79 V which is above the bandgap. A brightness of 100 cd/m^2 is reported around 2.5 V for the same PLED. It might be associated with a distinct brightness of 60 cd/m^2 for computer and laptop displays. Even a 50-nm thin film polymers are reported to emit light. If the layer thickness increases, for instance, 100 nm, the voltage rises approximately by 1V. The low-voltage process enables device operation possible in ordinary less-expensive integrated chips. Up to 10,000 cdm^{-2} brightness can be achieved at as low as 6 V. In pulsed open, even 100,000 cdm^{-2} brightness is possible to achieve. Even some groups proved laser action is possible in polymer devices with intensity more than 1,000,000 cdm^{-2}.

1.5.2 Light Conversion Efficiency

The efficiency of a PLED depends on the external efficiency which is calculated in forward direction. However, the value of external efficiency observed in forward direction is large compared to the values observed using integrated spheres. In addition, sometimes, the samples behave like an optical fiber, in which total internal reflection takes place when the angle of incidence is greater than the critical angle, due to this in PLEDs considerable quantity of light which is produced in the emissive-layer escapes through the sideways of the sample. Ching Tang *et al.* [21] proposed a simple solution to determine the whole value in terms of candela per ampere. Green PLEDs exhibit highest efficiency of 75 cd/A. Further enhancements can be achieved through rising in the efficiency of photoluminescence and also by enhanced electron-injection. In general, PLEDs' efficiencies are far superior than normal bulbs and LEDs. In current display technologies, PLED is far superior and simple to fabricate.

1.5.3 Color Stability

Stability of polymers and their properties are main concerns regarding this polymer technology. In the present scenario, PLEDs' displays and backlights for LCDs can meet customer specification only in orange light-emitting materials. Quick evolution is happening to grow blue and green emitting polymers to similar extent of constancy. Some researchers proved that it's possible to fabricate device on elastic substrates as an alternative of rigid glass-substrates. But these devices are reported to have short lifetime around 1 day. These devices are ruined by diffusion of water through the plastic film because of the sensitivity of the device for O_2 and H_2O, it may lead to rapid corrosion of the positive electrode of the device. Recently, advancement in the production of elastic films consisting good water barrier properties, flexible light-emitting films, and displays was made possible.

1.6 Applications in Large and Small Area Devices

1.6.1 Displays

PLED technology can be employed to fabricate small and simple unicolor segmented displays to complicated and large full-colored displays. The application ranges are typically classified based on the size of the display.

1.6.1.1 Matrix and Small Segmented Displays, ≤25 cm^2

The appliances are where the information displayed is limited, for example, in car dash boards, professional-equipment, etc. Utmost of the display area is typically monochrome. Comparing PLEDs with other technologies, reflective LCD is advantageous as a function of power consumption, but it has a poor contrast, it has a dull visual aspect, it has low viewing angle, and it is not readable in the dark. PLED exhibits a discrete advantages in slimness and an improved power consumption factor in the range of 10 to 100, combining with a backlight and LCD. Some of the advantages of PLED devices are thin, response time is fast, graphics resolution is very high, display brightness is high, and contrast is also high.

1.6.2 Thin and Flat Light Sources

In present scenario, application of PLEDs as sources of light, apart from some special cases, is questionable. When compared with power efficiency of 19.99 Lm/W for florescent sources and 60 Lm/W for incandescent tubes and the PLED power efficiency is as low as of 4 to 10 Lm/W. Also, the lifetime of polymer-based LED devices is limited by the high intensity of light needed for illumination. Nevertheless, PLED devices might be utilized in all types of applications in signaling such as brake lights for cars, decorative light sources, potentially rear lights, etc. LCD backlighting is one explicit application where the source of the light is really lightweight, thin, and flat. The usage PLEDs in comparatively less in large-area applications, less than 100 cm^2, can be distinguished based on the purpose for which they are used. Also, the backlight not only used to increase the contrast of the display in daylight but also used to illuminate the display in dark for example car stereo, radio sets, etc.

1.6.3 Cloth-Type PLEDs

The formation top emission OLEDs on a substrate of fabric is an easy way in advancement of OLEDs. The substrates of fabrics are categorized thru spatial voids and through these voids water can easily pass. Besides the assembly of constituent fibers forms the uneven surface; unfortunately, this is not compatible with the OLEDs. Hence, to prepare OLED-compatible fabric substrates, it is necessary to introduce the methods to eliminate the spatial voids then reformation of surface to be even [22–24]. There exist two stages to form a glass-like surface which can be used as fabric substrate to embed OLEDs [24]. Foremost, for partial planarization, i.e., fill the valleys, low viscous ductile polyurethane (PU) is spin coated on self-assembled fabric substrate. Next, to reduce the roughness of the surface, a high viscous polyurethane is deposited on the fabric before clean guide substrate is transported to the subsequent fabric substrate *via* lamination at room temperature. Subsequently, the distinguished methods of top-emission OLED production and multibarrier encapsulation, extremely robust wearable OLEDs were attained on the modified fabric substrate [24]. Even after this promising achievement, remaining practical problems should be overcome to use in realistic wearable displays. The predicted planarization procedure for fabric substrate well-suited with OLEDs weakens the nature of the fabric like softness, breathability, etc.

Furthermore, accomplishing consistent wash ability of the device might be another critical obstacle in the perspective of wearable electronic clothes. In view of the ability to concede actual light-emitting fabrics, the fiber-shaped OLEDs are much closer to cloth-type display concept, due to tiny and curved substrate, it is difficult to prepare OLEDs on the fiber substrate. B. O'Conner *et al.* reported a technique that the deposition of layers of OLEDs on the fiber is retain the rotation of the fiber in vacuum deposition [25]. Another reported technique is dip-coating method which is cost-effective and simple and can be employed for solution-based PLED-fibers [26]. Whereas both techniques surely proved the possibility of OLEDs on fibers, advanced studies on electrical addressing and reliable encapsulation schemes for the accumulated fibers are required for working as a display device [27].

1.6.4 PLEDs in Wearable Electronics

S. Choi *et al.* [28] have fabricated a light-emitting fabric, which is efficient and flexible, making it suitable for displays which can be worn like clothes. The PEN fibers are used for weaving the fabric, and the thermal lamination is used to form planarization layer onto this fabric. The surface roughness of $R_q = 2.073$ nm, these fabrics make them appear very smooth. Organic light-emitting diodes were placed thru thermal evaporation and the additive protective layers by transparent-flexible encapsulation effectively block the saturation of water and oxygen. Besides, the prepared device exhibits the luminance of around 35,844 Cd/m^2 and maximum current efficiency of about 70.43 Cd/A. In addition, the device on the material was found to operate stably after harsh bending, even at a bending radius of 2 mm for 3,000 cycles and a bending radius of 1 cm after 30,000 cycles. However, more bending of these fabrics results in leakage current within the device and cracks on the fabric. These fabrics can find various electronic textile industrial applications like curtain manufacturing, serves as functional and table clothes in and healthcare, fashion, as well as in the automobile industries.

1.7 Conclusion

The construction and working principles of PLEDs is discussed in detail. Different polymers used in PLEDs are discussed. The important parameters that are the key factors to be considered while adopting the polymer electroluminescent materials for PLEDs like brightness, color purity, light conversion efficiency, and color stability are discussed. Finally, the application of the PLEDs to small, midsize, and large size displays are discussed. Also, their application in flexible displays and cloth type wearable displays are discussed. These PLEDs are undergoing a rapid development and may soon be available in all forms of displays. The present limitations of PLEDs in illumination system where the intensity of light is very high compared with displays like television and laptops will soon be rectified with improved polymers.

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