

SCIENCE
**THE INDIAN
JOURNAL
OF RESEARCH
ANVIKSHIKI**

Bi-Monthly International Journal of all Research

Volume-10

Number-2

March-April 2016



ISSN 0973-9777
GIS Impact Factor 2.4620
Volume-10, Number-2
March-April 2016



MPASVO

Published on behalf of the MPASVO in
association with the Member's of Anvikshiki

Anvikshiki

The Indian Journal of Research

Bi-Monthly International Journal of All Research

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Subscriptions

Anvikshiki, The Indian Journal of Research is Published every two months (January, March, May, July, September and November) by
mpasvo Press, Varanasi, U.P. India. A Subscription to The Indian Journal of Research : Anvikshiki Comprises 6 Issues in Hindi and
6 in English and 3 Extra Issues. Prices include Postage by Surface mail, or For Subscription in the India by Speed Post.
Airmail rates are also available on request. Annual Subscriptions Rates (Volume 10, 6 Issues in Hindi, 6 Issues in English and
Few Special Issues of Science 2016):

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Shukla and should be sent at the address given below. Sbi core banking cheques will also be accepted.

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Office Time : 3-5 P.M. (Sunday off)

Journal set by : Maheshwar Shukla, maheshwar.shukla@rediffmail.com

Printed by : Mpasvo Press

Date of Publication : 1 March 2016



Maneesha Publication
(Letter No.V-34564, Reg.533/2007-2008)
B-32/16-A-2/1, Gopalkunj, Nariya, Lanka
Varanasi, U.P., India

Anvikshiki

The Indian Journal of Research

Volume 10 Number 2 March 2016

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PRINT ISSN 0973-9777, WEBSITE ISSN 0973-9777

POLYFLUORENE-ACCEPTOR CONJUGATED POLYMER FORLED APPLICATIONS

AKANKSHA PATHAK*

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Abstract

Now a days, polymers are used in many electronic devices, with the challenge of obtaining highest efficiency. Many Organic polymers are used as an organic layer in organic light emitting diode (OLEDs) and organic photovoltaic cells (OPCs), which is due to the presence of the π -conjugated oligomers.

For our report we have reviewed many conjugated polymers including Polyfluorene. It has high efficiency in comparison to other conjugated polymer but since it is a hole transport dominated material, mobility of electrons is limited and hence Polyfluorene can be incorporated with different acceptors segments, like quinoxaline (Q), 2,1,3-benzothiadiazole (BT) and thieno[3,4-b]-pyrazine (TP). These three fluorene- acceptor (PFQ, PFBT, PFTP) were incorporated and investigated in the literature. Their properties were then compared with the parent Polyfluorene. It was found that tuning of various electronic and optoelectronic properties could be achieved by the incorporation of various acceptor segments and their concentration. Moreover, acceptor strength significantly affects the properties due to intramolecular charge transfer. However, high rate of charge transfer results in fluorescence quenching and thus reduces the photoluminescence efficiency. For this purpose binary blend structure (such as PF with PFQ, PF with PFBT) proved more significant as they show high rate of charge transfer along with the high photoluminescence efficiency.

1. Introduction

Light emitting polymers belongs to the unique class of aromatic molecules exhibiting semiconducting behaviour and give off light when electrically stimulated. They derive their semiconducting properties from the extensive delocalization of π -electron bonding along the polymer chain which makes them

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capable of absorbing sunlight, creating photo generated charge carriers and transporting these charge carriers ^[8]. Moreover, these significant properties can be altered by the inclusion of functional side groups as well as substitution of the intractable conducting polymers backbone with alkyl and alkoxy substituent. These materials exhibit mechanical, electrical, and luminescent properties that hold great potential for display application. Light is produced in the polymer by the fast decay of excited molecular states, the colour of which depends on the energy difference between those excited states and the molecular ground level.

The polymer LED is a dual carrier injection device. The electrons are injected from cathode to the Lowest unoccupied molecular orbital (LUMO) of the polymer and holes are injected from anode to the Highest occupied molecular orbital (HOMO) of conducting polymer and they recombine radioactively within the polymer to give off light ^[12]. The injection occurs due to tunnelling and thermionic emission. Basically it comprises of a thin- film structure of one or two layers typically no more than 0.1 micron thick, sandwiched between two electrodes. Optimal device efficiency is achieved if the two electrodes are dissimilar, specifically where the respective electrode materials possess Fermi levels or electronic work functions that closely match the valence (ground) and conduction (or excited state) energy levels in the polymer. It is also necessary that one of the electrodes is transparent to the wavelength of light generated. This can be achieved with very thin metal films or more preferably the oxide of indium and tin, which retains good electrical conductivity but which is transparent in layers below 0.2microm thick.

OLEDs offer many advantages over both LCDs and LEDs^[9]. They are:

- OLEDs have low cost than LCDs or plasma displays and they can be printed onto any suitable substrate using an inkjet printer or even screen printing technologies.
- The ability of OLEDs to be printed onto displays embedded in fabrics.
- OLED pixels directly emit light, thus provides a greater range of colours, brightness, and viewing angle than LCDs.
- It has the color tuning property, that is its major advantage.
- It has energy saving potential and high luminescence efficiency.
- OLED substrates can be plastic rather than the glass used for LEDs and LCDs.

2. Literature review of the developments in Light Emitting Polymers

2.1 Polyacetylene; In 1977, Alan J. Heeger, Alan MacDiarmid and Hideki Shirakawa found out that a thin film of Polyacetylene could be oxidized with iodine vapours, turning the material into a conductor. This led to win the 2000 Nobel Prize in Chemistry ^[12]. However the photo luminescence efficiency was just 0.1% with wavelength corresponding to 350-370nm (violet).

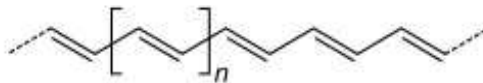


Figure 1 Polyacetylene

2.2 Poly phenylene vinylene; In 1990s the Cambridge group of Friend announced that they had achieved green + yellow using the conjugated polymer poly phenylene vinylene (PPV) in a single-layer device structure. It was a highly stable conjugated polymer. Its yellow color was due to an absorption band centred at ~400 – 420 nm (depending on the method of synthesis) with an on-set corresponding to a band gap of ~2.5eV. The HOMO and LUMO levels in PPV can be accessed in cyclic voltammetry experiments which under proper conditions, reveal chemically reversible oxidation and reductions waves. The deduced electrochemical gap corresponds reasonably well to the optical band gap ^[12].

Being a relatively good electron donor, PPV and its derivatives can be chemically doped by strong oxidising agents and strong acids affording highly conductive p doped materials .PPV is featured by a vibronic ally structured emission band with peaks maxima at 520 and 551 nm.

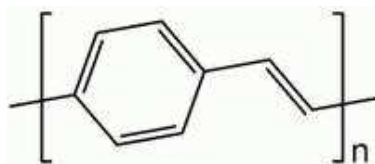


Figure 2 Poly Phenylene Vinylene

2.3 *Polythiophenes*; Schopf and Koßmehl published a comprehensive review of the literature published between 1990 and 1994. Polythiophenes (PTs) (including oligothiophenes) are one of the most studied and important classes of linear conjugated polymers. Versatile synthetic approaches to PTs, both chemical and electrochemical, easy functionalization and unique electronic properties, which can be widely tuned, focus a tremendous interest to this class of polymers ^[6].

Due to electron-rich character of the thiophene ring, PTs can be easily and reversibly oxidized by chemical or electrochemical mean to form p-doped, usually highly conducting materials. The first electronic transition of undoped PT (which strongly depends on the structure) lies between 300 and 500 nm. PTs have not been studied as widely as PPVs or polyfluorenes, yet they present an important class of LEP. PTs can show good red emission, consistent with their band gap of 2 eV, a colour, which is difficult to produce with other LEP. Often the luminescence efficiency of PTs in the solid state is relatively low, much lower than that of PPV and polyfluorenes, which originates from their solid state structure – a tendency of strong interchain interactions (especially for low molecular weight oligomers). This feature is an advantage of PTs in some electronic applications as, e.g., field-effect transistors. However, it becomes one of the most critical drawbacks for application as emissive materials in LED. Whereas in solution the photoluminescence efficiency of poly (3-alkylthiophenes) is ~30–40 %, it drastically drops down to 1–4 % and lower in the solid state, due to increased contribution of non-radiative decay via interchange interactions and intersystem crossing caused by the heavy-atom effect of sulphur. Thiophene-based polymers have stronger spin-orbital coupling than phenylene-based polymers due to the internal heavy atom effect of the sulphur heteroatom and hence triplet state processes play a greater role in their photo physics. Another feature of PTs is the phenomenon of thermochromism, which has been shown for poly (3-alkyl) thiophene in many publications. It is believed that the thermochromism observed in poly (3-alkyl) thiophene films originates from the thermal movement of the side chains, shifting predominantly planar structure of chains at low temperatures to a random coil conformation when the temperature is increased, thus forcing the polymer backbone out of planarity. This leads to decrease of orbital overlap and effective conjugation length, resulting in increase of the band gap and blue shift in the polymer absorbance (from red to purple or purple-blue). The process is completely reversible and on cooling the initial colour of the film is restored. However thermochromism ^[11,7] is undesirable for LED application, as it could lead to changes of the emission wavelength and the quantum efficiency of the device during the operation.

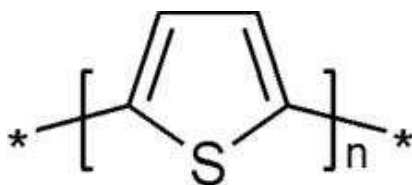


Figure 3 Polythiophene

2.4 *Polyfluorene*; Polyfluorene belong to the class of rigid-rod polymers. The monomer unit consists of rigid planar biphenyl units, bridged by a carbon atom in position 9, ensuring a high degree of conjugation. This carbon-9 atom may also carry additional substituents to modify the polymer process ability and the inter chain interactions in films, without significantly altering the electronic structure of the individual chains ^[5]. Suitable substituents enable solubility in common organic solvents as well as the processing of high-quality thin films by spin casting. The thermal stabilities of the homo and copolymers are excellent with decomposition temperatures exceeding 400C. The absorption of poly (9, 9-dialkylfluorene) consists of a strong featureless p-p* transition (Figure 1) that peaks at about 3.2 eV (380 nm) and has a relatively narrow line width PF layers appear brightly sky blue under excitation with UV light. The photoluminescence quantum efficiency is large and values of more than 50% have been reported both in solution and in the solid state.

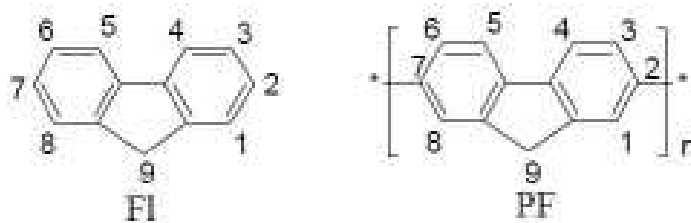


Figure 4 Polyfluorene ^[12]

The 2, 7-Positions in Fl are the most reactive sites towards electrophilic attack, which allows Construction of a fully conjugated rigid-rod polymer chain by substitution reactions, whereas the methylene bridge provides an opportunity to modify the process ability of the polymer by substituents, without perturbing the electronic structure of the backbone. The varieties, excellent optical and electronic properties, and high thermal and chemical stability of polyfluorenes (PFs) make them an attractive class of materials for polymer light-emitting diodes (PLEDs). Different aspects of syntheses, properties and LED applications of fluorene-based conjugated polymers and co-polymers have been highlighted in several recent reviews. In fact, polyfluorenes are the only class of conjugated polymers that can emit a whole range of visible colours with relatively high quantum efficiency.

The electronic absorption spectra of dilute (typically 5–10 mg/L) solutions of poly (9,9dialkylfluorenes) show a sharp peak with Lambda max ~ 385–390 nm (3.2 eV) of electronic transition. The thin solid films (spin-coated from 15–20 mg/mL solutions) reveal similar absorption with slightly red-shifted (~10 nm) and a relatively broader peak (due to. intramolecular interaction). The photo luminescent spectra of the PFs show well-resolved structural features with maxima at 420 nm, 445 nm, and 475 nm respectively .

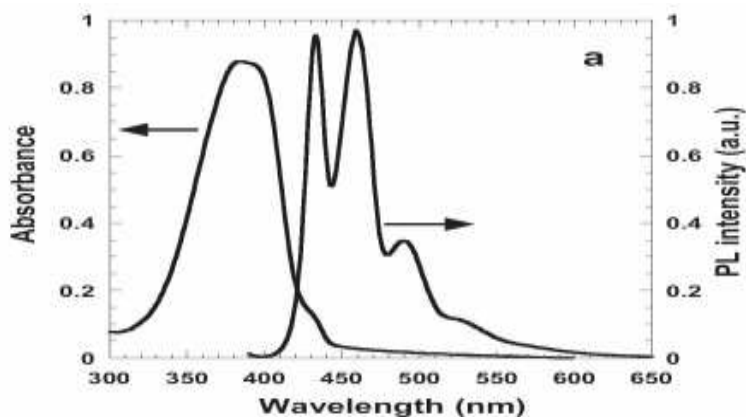


Figure 7 Absorption curve for PF ^[8]

3. Properties of early Polyfluorene devices

The first paper on electroluminescence from PF by Ohmori et al. reported a single-layer LED consisting of an indium tin oxide (ITO) anode, a 100 ± 200 nm emission layer of poly(9,9-dihexylfluorene-2,7-diyl) (DHF) and a Mg in cathode. The device turned on at approximately 10 V. In contrast to the solid-state photoluminescence, which exhibited a well-resolved vibronic structure, the EL emission spectrum was very broad with the maximum at 470 nm. The emission colour was blue, thus this was the first report on blue electroluminescence from PF. No information on efficiency and brightness was provided. Injection due to the large band gap of PF. Based on cyclic voltammetry and the optical gap, a valence band energy of 5.5 eV and a conduction band energy of 2.9 eV below the vacuum level was reported.

Later, Pei and Yang reported photoluminescence and electroluminescence from poly [9, 9-bis (3, 6-dioxaheptyl) - fluorene-2, 7-diyl] (BDOH-PF), a PF homo polymer substituted with oligo (ethylene oxide) side chains. This special side chain was chosen to promote ion transport as required for light-emitting electrochemical cells (LECs). The polymer was found to exhibit efficient photoluminescence in solution and in the solid state, with PL quantum efficiencies (PL) of 70% and 73%, respectively. Compared to the PL in solution, the thin film PL spectrum showed an additional broad green contribution, assigned to interchain excimers. Single-layer LEDs with an ITO anode, a ca. 120 nm thick BDOH-PF emission layer and a Ca cathode turned on at 9 V and reached 100 cd/m² (the brightness required for good visibility of a display) at 30 V. The external quantum efficiency for electroluminescence (EL) was 0.3%.

Two years later, Grice et al. reported a double-layer device based on poly (9, 9-dioctylfluorene-2, 7-diyl) (PFO), as supplied by Dow Chemical Corporation. The device comprised a 60 nm thick hole-transporting layer of a polymeric triphenyldiamine derivative (polyTPD). The device turned on at 5 V and the brightness was 600 cd/m² at 20 V. At that voltage, luminance and luminous efficiency were 0.25 cd/A and 0.04 lm/W, respectively. The maximum external quantum efficiency was found to be just 0.2%, however the solid-state PL quantum efficiency was 55%.

S. Janietz et al. reported CV measurements for PFO in solution and in the solid state. In the solid state, the onset potentials for oxidation and reduction were $E_{ox} = 1.4$ V and $E_{red} = \pm 2.28$ V relative to an Ag/AgCl reference electrode. With an absolute energy of Ag/AgCl of 4.4 eV, this corresponds to HOMO and LUMO energies of 2.12 eV and 5.8 eV, respectively, and an HOMOLUMO gap of 3.7 eV. This is significantly larger than the optical gap $E_g = 2.95$ eV as defined by the absorption onset. According to these numbers one expects a large injection barrier for holes from ITO (ionization energy ca. 4.7 ± 5 eV) and for electrons from Ca (work function ca. 2.9 eV). Indeed, charge transport-limited currents (which require ohmic contacts) have by now neither been reported for electrons nor for holes in PF homo polymers.

4. Limitations of PF Based Polymers and Incorporation of Acceptor Units

PF-based polymers are hole-transport-dominated materials that exhibit low electron mobility in EL devices. Their unbalanced charge injection and transport properties limit the efficient recombination of holes and electrons, resulting in a decrease in the EL efficiency. The incorporation of an alternating electron donor-acceptor system into a PF based polymer can lower its LUMO energy and reduce its band-gap, and therefore can be used to balance device hole/electron mobility and realize long-wavelength light emission. Full colour emission can be obtained by incorporating different acceptor segments

(quinoxaline (Q), 2, 1, 3-benzothiadiazole (BT) and thieno [3, 4-b]-pyrazine (TP)). Three fluorine acceptor conjugated polymers were investigated and was compared with parent fluorine and there properties are as follow:

4.1 Thermal properties; The TGA and DSC curves of the PF, PFQ, PFBT, and PFTP is studied. The thermal decomposition temperatures (Td, 95 wt% residue) and glass transition temperatures (Tg) estimated from their TGA and DSC curves are summarized in the table below. The Td of the PFQ, PFBT, and PFTP are 446, 434, and 447 °C respectively, which are higher than that of the parent PF with 409 °C. As shown in the Figure 7, Tg of the PF, PFQ, PFBT, and PFTP are 67, 132, 115, and 160 °C respectively.

T A B L E 1 *Thermal properties*^[1,3,14,15]

	Mn	Tg (C)	Td (C)	Acceptor ratio
PF	57000	67	409	0
PFQ	19000	132	446	50
PFBT	19000	115	434	50
PFTP	9000	160	447	50

4.2 Absorption Spectra & Optical Band Gaps of Alternating Copolymer; The UV–visible absorption spectra of the PF, PFQ, PFBT, and PFTP films are shown in the figure 8 and the corresponding absorption maximum (λ_{max}) are summarized in the Table 2. The λ_{max} of the PF, PFQ, PFBT, and PFTP films are 368, 416, 470, and 578 nm, respectively. It suggests that the optical properties of these polymers could be tuned over a wide range through different acceptors. The optical band gaps (eV) estimated from the absorption edges are in the order of PF (2.95) PFQ (2.64) PFBT (2.34) PFTP (1.82). The much lower band gaps of the PFQ, PFBT, and PFTP than that of the parent PF are probably due to the intramolecular charge transfer between the fluorene and acceptor or the backbone planarity.

However, the decrement of band gaps of these alternating copolymers is not in consistence with the increasing order of the acceptor strength. The electron-accepting strength is in the order of BT>TP>Q. The smaller band gap of the PFTP than that of the PFBT might be because of the backbone planarity. The TP with the five member thiophene ring generally have smaller torsional angle with fluorene than that of the BT with the six-membered phenylene ring, which assists the efficient intramolecular charge transfer and results in a smaller band gap. The extended tails and/or shoulders near the absorption edges of PFQ, PFBT, and PFTP suggest stronger inter-chain interaction as compared to the parent PF. This could also result in a smaller band gap. Finally we can conclude that there is enhancement of π –electronic delocalization through the incorporation of acceptor moiety and the significance of intramolecular charge transfer.

T A B L E 2 *Optical Properties*^[1,3,14,15]

	λ_{max} (nm)	Eg (Ev)
PF	368	2.95
PFQ	416	2.64
PFBT	470	2.34
PFTP	578	1.82

4.3 PL Spectra & Emissive Colours of Alternating Copolymers; The variation of emission peaks shows the same trend as that of optical absorption spectra. The emissive colours of the PF, PFQ, PFBT, and PFTP are blue, green, yellow, and red, respectively covering the entire visible region. The results suggested that the colour tuning of the fluorene-based alternating copolymers by incorporating different acceptor segments in the backbone is feasible. Two additional emission peaks of the PFTP at 439 and 508 nm are observed in the solid state but not shown in dilute solution. They are probably attributed to the intermolecular interaction of the fluorine segments and the formation of excimers in the solid state, respectively, as compared to the PL spectrum of the PF.

T A B L E 3 *Optical Properties of alternating polymers*^[1,3,14,15]

	λ (nm)	η (%)	
PF	445	56.6	Blue
PFQ	493	22.4	Green
PFBT	540	18.5	Yellow
PFTP	674	2.1	red

4.4 EL Spectra & Emissive Colors of Alternating Copolymers; The emission maxima of EL devices based on the PF, PFQ, and PFBT were found to be 425,480 and 540 nm, respectively, which are similar to the corresponding photoluminescence maxima. This result suggested that the colour tuning on the EL devices of polyfluorenes through incorporation various

acceptors on the backbone is feasible. However, the EL device with the PFTP as the emissive layer was too weak to be detected. It was probably due to the intramolecular charge transfer or heavy-atom effect resulting in fluorescence quenching.

T A B L E 4 *Luminiscence characteristics*^[1,3,14,15]

	λ (nm)	V	η (%)	
PF	425	10.5	0.18	Sky Blue
PFQ	480	8.5	0.20	Blue Green
PFBT	540	7	0.13	Yellow
PFTP	-	-	-	-

4.5 PL and EL spectra of binary blends; Until now we have seen that as acceptor is incorporated into the polymer backbone, band gap decreases but simultaneously efficiency also get reduced. Here in order to overcome this reduce in efficiency, we can go for Binary spectra. That is we can use the combination of PF with PFQ or PF with PFBT etc. The Results for the Binary blend is shown below.

T A B L E 5 *Optical Properties of Binary Blends*^[1,3,14,15]

	Composition	PL		Bias (V)	EL	
		λ (nm)	PL		λ (nm)	EQE (%)
PF	100:0	425	56.6	10.5	425, 445, 475	0.18
FQ1	95:5	483	53	9	424, 475	0.38
FQ2	90:10	484	53	9	424, 481	0.44
FQ3	75:25	487	51	9.5	481	0.13
PFQ	0:100	493	22.4	8.5	480	0.20
FBT1	95:5	532	56	8	423, 535	0.27
FBT2	90:10	534	48	7.5	423, 527	0.37
FBT3	75:25	538	33	7	530	0.27
PFBT	0:100	540	18.5	7	530	0.17

Two sets of emission peaks are shown by the PL spectrum of FQ1 at 424 and 478 nm attributed to PF and PFQ, respectively, with this we suggested that energy is transferred from PF to PFQ, And it is found that the energy transfer from PF to PFQ becomes more efficient with increasing PFQ content. Overlapping of the emission of PF and the absorption of PFQ explained the significant energy transfer. For FQ2–FQ3, the PL spectra display almost only the green emission around 485 nm from PFQ, indicating the almost complete energy transfer from PF to PFQ. The PL spectrum of FBT1 also shows two sets of emission peaks at 425– 445 and 525 nm attributed to PF and PFBT, respectively. The PL spectra of the FBT binary blends show a variation with the concentration of PFBT similar to that of FQ. Both binary blends show complete energy transfer from the donor PF to the acceptor (PFQ or PFBT) at a relatively low acceptor concentration of 5 wt %. These results suggest efficient energy transfer in the studied fluorene binary blend systems. The efficiency of Forster energy transfer from PF to PFQ or PFBT can be evaluated through the analysis of the Forster radius. Efficient Forster energy transfer can be expected from PF to PFQ (or to PFBT) in these blends, this can be indicated by large Forster radius. Forster radius can be found from the formula given below

$$R_{60} = \frac{9000(\ln_{10})k^2\Theta\phi_D}{128\pi^5 N_{AV}\eta^4} J$$

Table 5 summarized that the PL efficiencies of the binary blends, FQ and FBT, in solid states. The PL efficiencies of PF, PFQ, and PFBT are 56.6, 22.4, and 18.5%, respectively. For FQ and FBT, the binary blends, the PL efficiencies are in the ranges of 53–75 and 33– 56%, respectively. The much higher PL efficiencies of these binary blends, compared with those of PFQ and PFBT. This higher PL efficiencies suggest that adding a small amount of PFQ or PFBT to PF can significantly enhance the efficiency of the acceptor. The PL efficiencies of these binary blends decrease with an increasing concentration of PFQ or PFBT because of the possible fluorescence quenching attributed to the intramolecular charge transfer or heavy-atom effect.

After analysing the samples trends for EL and PL is obtained and it is found that the variation of the EL spectra is similar

to that of the PL spectra, that is, partial quenching of the PF emission with a concentration of 1 wt % (FQ1 and FBT1) and complete quenching of the PF emission with a concentration higher than 5 wt % (FQ2–FQ4 and FBT2–FBT4). However, the acceptor concentration for quenching the PF emission in the EL spectra of FQ1 and FBT1 is lower than that for their PL spectra, and this indicates more efficient energy transfer in EL than in PL. The more efficient energy transfer in EL can be attributed to the recombination zone for photo excitation and electric excitation. In addition to the Forster energy transfer in PL, the emission from PFQ (or PFBT) in EL might also arise from the charge-trapping mechanism. The corresponding electroluminescence data for the same is given in the table 5.

5. Conclusion

Tuning of various electronic and optoelectronic properties could be achieved by the incorporation of various acceptor segments and their concentration. Moreover, acceptor strength significantly affects the properties due to intermolecular charge transfer.

However, high rate of charge transfer results in fluorescence quenching and thus reduces the photoluminescence efficiency. The chemical structures and concentrations of the acceptor in the polymer blend significantly affect the energy transfer and the resultant luminescence characteristics. The miscible blend systems promote efficient energy transfer and result in emissions only from the acceptor. Thus, a significant enhancement of the luminescence quantum efficiency in comparison with the corresponding acceptor has been observed by the blending of a relatively small amount of the acceptor.

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