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# The electroluminescence and photoluminescence characteristics of films of poly(phenylene vinylene)/gel-glass interpenetrating networks

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A series of poly(phenylene vinylene) (PPV)/gel-glass composites were prepared from mixtures of PPV precursor and silicon alkoxide methanol solution by employing a sol–gel process. The PPV/gel-glass composite, which is an interpenetrating network (IPN) structure, is used as the emissive layer in a polymeric light-emitting diode (LED). The addition of a small quantity of titanium ethoxide catalysed the reaction of gel-glass formation, making the PPV/gel-glass formation successful without phase separation and the luminescence of the composite film tunable. The luminescence characteristics of these PPV/gel-glass composites are dependent on the raw-material composition of silicon alkoxide and the molar ratio of PPV to gel-glass. From the ultra-violet/visible absorption spectrum, the conjugated length of these PPV/gel-glass composites can be seen to be shorter than that of pure PPV. The resultant PPV/gel-glasses emit light over a wide range from yellow-green (550 nm) to blue (484 nm) in the photoluminescence spectrum. Electroluminescent devices made from PPV/gel-glass 4 composites in the molar ratios of 1/1.17 and 1/2.33 shows a maximum emission peak at 515 and 510 nm, respectively. These PPV/gel-glass 4 LEDs showed a higher relative electroluminescence efficiency and a longer lifetime than pure PPV. The improvement in the device stability is related to the reduction of the thermal vibration of PPV chains during operation and the hindrance of carrier mobility in the IPN structure. Copyright © 1996 Elsevier Science Ltd.

(Keywords: poly(phenylene vinylene); gel-glass IPNs; luminescence)

## INTRODUCTION

Since the first poly(phenylene vinylene) (PPV) light-emitting diode (LED) reported by Burroughes[1], conjugated polymers have attracted a lot of attention, especially for electroluminescence applications. A variety of conjugated polymers with different $\pi$–$\pi^*$ band-gap energies have now been developed to emit various wavelengths in the whole visible spectrum[2–14]. Polymeric LEDs show many advantages over inorganic LEDs, such as the easy production of large-area flat displays, low cost and easy design of polymer structure for emitting a specific wavelength of light via molecular-level engineering. The blue-light LED is the most attractive subject among LEDs. However, there are many problems encountered in the commercialization of polymeric LEDs. The main challenges include long-term stability, luminescence intensity and efficiency.

Among many conjugated polymers, PPV and its derivatives are the most intensively studied as very promising in LED application. Chemists have designed and synthesized various PPV-related polymers for LEP purposes from red to blue light emission[1,2,4,6,13,14], but the long-term stability problem is still in need of solution.

The problem may be solved from both physics and chemistry aspects.

The problem of long-term stability may include oxidation, ionic doping, spike effect, recrystallization, etc. The improvement of long-term stability of polymeric LEDs from physics aspects has seen some remarkable achievements. These methods to improve the lifetime of polymeric LEDs include operating or sealing the system in high vacuum or in an inert atmosphere[2,9,15–17], or using an a.c. source to drive the polymeric LED[17]. The approach to improving the long-term stability from chemistry aspects is still not clear.

In this study, we aimed at developing a new class of interpenetrating network (IPN) structures of PPV/silica gel-glass for LED applications to lock the PPV chain in the gel-glass in order to improve the lifetime and efficiency and to tune the emitting light frequency. The silica gel-glass used here has good transparency and thermal stability, high mechanical properties and good mixing with PPB precursor without phase separation after final heat treatment. In this sol–gel process, the PPV precursor is homogeneously mixed with the various composites of silicon alkoxide in the common solvent methanol and then converted to the final state by heat treatment under high vacuum. In this study, the PPV chains were designed to be locked in the network of silica

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**Table 1** Abbreviation of alkoxides used in this experiment

| No. | Alkoxide | Chemical formula | Abbreviation |
|---|---|---|---|
| 1 | Tetraethyl orthosilicate | $Si(OC_2H_5)_4$ | TEOS |
| 2 | Methyltriethoxysilane | $CH_3Si(OC_2H_5)_3$ | MTEOS |
| 3 | Diethoxydimethylsilane | $(CH_3)_2Si(OC_2H_5)_2$ | DEODMS |
| 4 | Titanium ethoxide | $Ti(OC_2H_5)_4$ | TIE |

**Table 2** The compositions of the gel-glasses and their hardness

| Sample | Composition (wt%) | | | | Hardness |
|---|---|---|---|---|---|
| | TEOS | MTEOS | DEODMS | TIE | |
| Gel-glass 1 | 0 | 49 | 49 | 2 | 31 |
| Gel-glass 2 | 0 | 98 | 0 | 2 | 43 |
| Gel-glass 3 | 49 | 0 | 49 | 2 | 54 |
| Gel-glass 4 | 49 | 49 | 0 | 2 | 69 |
| Gel-glass 5 | 98 | 0 | 0 | 2 | fragile |

matrices, restraining the movement of the PPV chains. This may reduce the thermal vibration of the polymer chains and the mobility of ions due to Joule heating during operation. Therefore, it may promote the lifetime of the devices. The effect of gel-glass on the behaviours of polymeric LEDs was also under investigation.

## EXPERIMENTAL

### The sulfonium precursor route for preparing PPV

The PPV precursor was prepared by the addition of 20 ml of 0.22 M NaOH aqueous solution into 20 ml of 0.2 M *p*-xylylenebis(tetrahydrothiophenium chloride) aqueous solution with 80 ml of pentane. Both solutions were first cooled to 0–5°C by an ice bath. The reaction was allowed to proceed for an hour and then terminated by the addition of 0.1 M HCl aqueous solution to neutralize the reaction solution. After the pentane was decanted off, the PPV precursor aqueous solution was dialysed (molecular weight cut-off, *MWCO* of 6000) against deionized water for several days. The PPV film was obtained by spin-coating the PPV precursor solution on an indium tin oxide (ITO) glass and then heating in a high-vacuum oven at 220°C for 2 h.

### The preparation of gel-glasses

All alkoxides used in this experiment and their abbreviations are listed in *Table 1*.

Based on the total weight of alkoxides, 2% titanium ethoxide was added either to one-component silicon alkoxide (98%) or to an equal weight of two components of silicon alkoxide mixture (49%, respectively), as listed in *Table 2*. These alkoxide mixtures were homogeneously stirred for three days under ambient conditions. These solutions were cast on Teflon discs or containers and then placed in temperature- and humidity-controlled chamber for three days (typically at 30°C and 50% r.h.).

### The preparation of PPV/gel-glass composites

The PPV precursor aqueous solution was freeze-dried to remove the solvent water. The PPV precursor was then stored in a refrigerator. The PPV precursor solid was redissolved in methanol solution to form 1% (by weight) PPV precursor methanol solution. These alkoxide solutions mentioned above were prestirred for three

days under ambient conditions. Adequate molar fractions of PPV precursor methanol solutions were added to these alkoxide solutions. After vigorous stirring, these solutions were spin-coated onto the precleaned ITO glasses and then heat treated in a high-vacuum oven at 200°C for 2 h.

### The preparation of the LED device

PPV and PPV/gel-glass composite films (80–100 nm) on ITO glass were coated with Al metal (5000 Å, area 7 mm²) by thermal evaporation in vacuum ($4 \times 10^{-6}$ Torr) to give Al/polymer/ITO sandwich devices. These sandwich devices were then annealed in a high-vacuum oven at 160°C for 3 h to improve the performance[18].

### Characterization

The thickness of PPV and PPV/gel-glass films was measured with a Dektak 3030 surface profilometer. Infra-red absorption spectra of PPV and PPV/gel-glass films were taken using a Bui-Rad FTS-165 *FT*i.r. spectrometer. U.v.-vis. absorption spectra of PPV and PPV/gel-glass films were measured using a Beckman 7400 spectrometer. A Shimadzu RG-5000 spectrofluorophotometer was used to get photoluminescence spectra of PPV and PPV/gel-glass films with an excitation light source at 365 nm. A Jasco FR-770 spectrometer was employed to measure the electroluminescence spectra of Al/polymer/ITO sandwich devices. The hardness of gel-glass was measured according to the method of ASTM-D220 Shore D. The current–voltage curves of these sandwich devices were measured with a programmable Keithley 237 electrometer. The electroluminescence intensities of these sandwich devices were recorded using a photodiode detector connected with a Newport (model 1815-C) Power Meter. The electroluminescence characterizations were done in a vacuum ($\sim 10^{-2}$ Torr) chamber.

## RESULTS AND DISCUSSION

A series of silicon alkoxides were employed including TEOS, MTEOS and DEODMS, which have four, three and two functional reactive sites, respectively, as a host matrix in this study. Titanium ethoxide was used to promote the rate of hydrolysis and condensation of these silicon alkoxides in the sol–gel process. Titanium ethoxide itself showed a very fast hydrolysis, even under normal moisture conditions, as observed in our publication elsewhere[19]. The alkoxide mixture containing titanium ethoxide became a gel-glass in several minutes under ambient conditions after casting on glass plates. Without titanium ethoxide, these silicon alkoxides exhibited a very low reactivity under ambient conditions. In the sol–gel process, the reactions were preceded by diffusion of moisture from the atmosphere into the alkoxide mixtures. The PPV/gel-glass composites were prepared by spin-coating the methanol solution of PPV precursor and alkoxides onto ITO glasses, and then heating the thin films in a high-vacuum oven. The PPV and gel-glass could be mixed in a wide range without phase separation and the resultant PPV/gel-glass could be mixed in a wide range without phase separation and the resultant PPV/gel-glass composites were of transparency suitable for optoelectronic and electroluminescence applications.

The preparation of PPV/gel-glass was first reported by Wung and coworkers[20] for a non-linear optical material. Wung's method cannot be used to tune the emitted light frequency of the PPV-based LED. In Wung's method, PPV precursor was mixed with silicon ethoxide in methanol at 60°C for 30 min to wait for the formation of sol before casting. At 60°C a lot of sulfonium groups were eliminated from the PPV precursor before casting. Therefore, the final PPV/gel-glass-based polymeric LED emitted very weak yellow-green light. This method may be good for non-linear optical material, but is not suitable for LED purposes. In this study, we developed a novel way to freeze the movement of PPV precursor in the network of gel-glass formed at room temperature in order to tune the emitted light frequency as well as to improve the lifetime of the polymeric LED.

As seen in *Table 2*, the hardness of these gel-glasses was dependent on the composition of the raw material. The gel-glass 1 made from silicon alkoxides with three and two functional reactive sites showed the lowest hardness in this series of gel-glasses. The hardness of these gel-glasses increased with the number of functional reactive sites on the silicon alkoxides. It was reasonable that the gel-glass prepared from more functional reactive sites could form a relatively compact three-dimensional network structure and became harder.

The chemical structures of fully converted PPV film and PPV/gel-glass composite film were characterized with an *FT*i.r. spectrometer as shown in *Figures 1a* and *1b*. The absorption band near $963 cm^{-1}$ was due to C–H out-of-plane bending of the *trans* configuration of the vinylene group. The band near $3024 cm^{-1}$ was assigned to the *trans*-vinylene C–H stretching node. The absorption bands near 831 and $1515 cm^{-1}$ were assigned to *p*-phenylene C–H out-of-plane bending and C–C ring stretching, respectively. The absorption bands are characteristic absorptions of PPV shown in both *Figures 1a* and *1b*. In *Figure 1b*, additional bands were observed near 1053 and $450 cm^{-1}$ corresponding to the Si–O stretching of silica glass network and the deformation vibration of Si–O–Si–O respectively. The bands near 1273 and $778 cm^{-1}$ were assigned to the systematic $CH_3$ deformation of $SiCH_3$ and $Si-CH_3$ rocking vibration, respectively. The i.r. absorptions for the non-conjugated methylene units of PPV in the composite film were covered by the strong absorptions of the methyl group of the gel-glass at 2977 and $2863 cm^{-1}$. The absorption band at $3450 cm^{-1}$ corresponded to the O–H stretching mode of Si–OH. The i.r. spectra could not tell if there is any chemical bonding between PPV and gel-glass because the potential C–O–Si stretching peak located in the range of 1000 and $1200 cm^{-1}$ overlapped the Si–O stretching characteristic peak of the gel-glass. Wung in his paper reported that no chemical bonding between PPV and silicate glass in their composite film was seen from Raman spectra[21].

U.v.-vis. absorption spectra of PPV and PPV/gel-glass composites are illustrated in *Figure 2*. The onset absorption of PPV was about 530 nm and that of pure gel-glass 4 showed a good transparency in the visible region (400–800 nm). However, when the PPV precursor was mixed with metal alkoxide solutions and heat converted into PPV/gel-glass composites, the onset absorption of these PPV/gel-glass composites shifted to a higher energy in comparison with that of pure PPV.
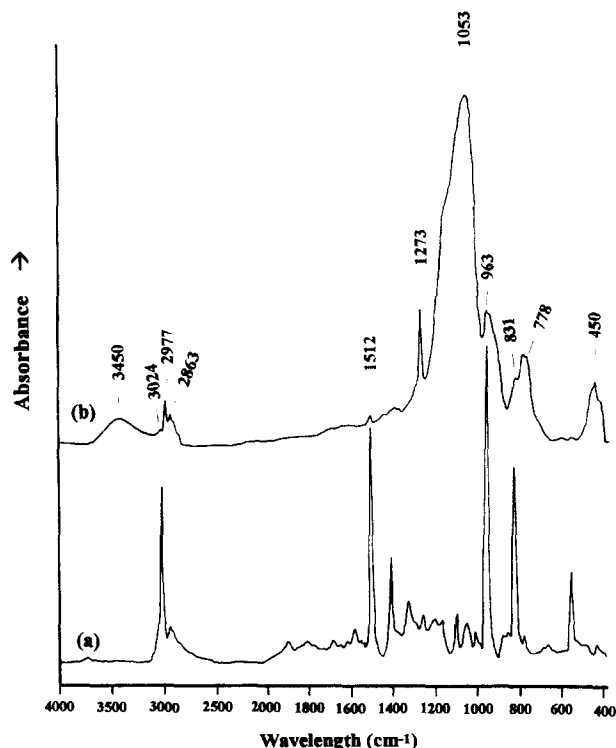


**Figure 1** I.r. spectra of (a) PPV and (b) PPV/gel-glass 4 (1/1.17 in molar ratio)
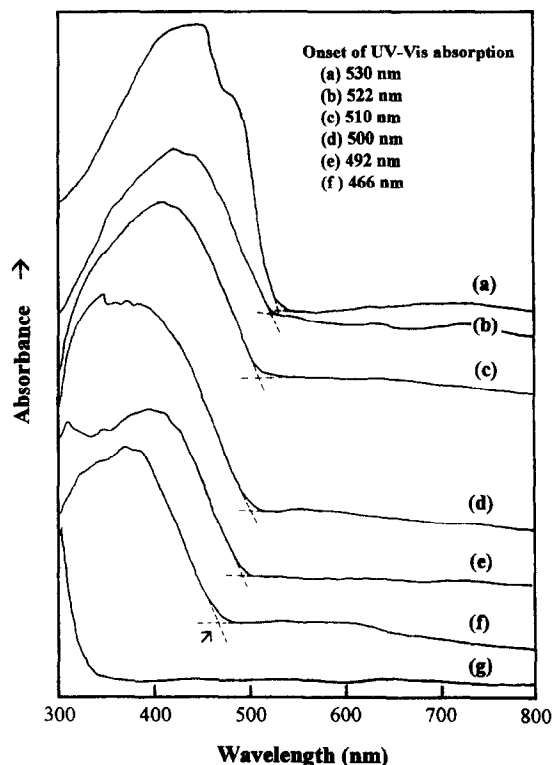


**Figure 2** U.v.-vis. absorption spectra of (a) PPV, (b) PPV/gel-glass 1, (c) PPV/gel-glass 2, (d) PPV/gel-glass 3, (e) PPV/gel-glass 4, (f) PPV/gel-glass 5, and (g) gel-glass 4. (All PPV/gel-glass = 1/118 in molar ratio)

This phenomenon indicated that the effective conjugated length of PPV in PPV/gel-glass composite IPN structure was shorter than that of pure PPV. In addition, the onset of u.v.-vis. absorption of these PPV/gel-glass composites showed a blue shift as the number of reactive functional
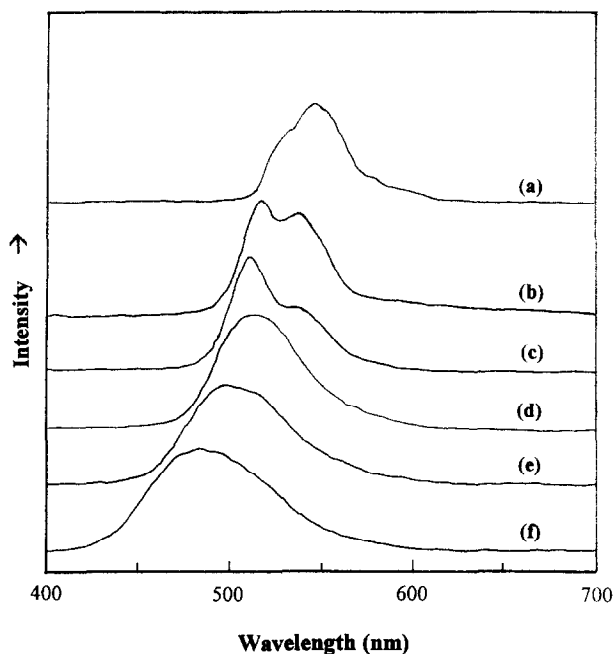
**Figure 3** PL spectra of (a) PPV, (b) PPV/gel-glass 1, (c) PPV/gel-glass 2, (d) PPV/gel-glass 3, (e) PPV/gel-glass 4 and (f) PPV/gel-glass 5. (All PPV/gel-glass = 1/118 in molar ratio)
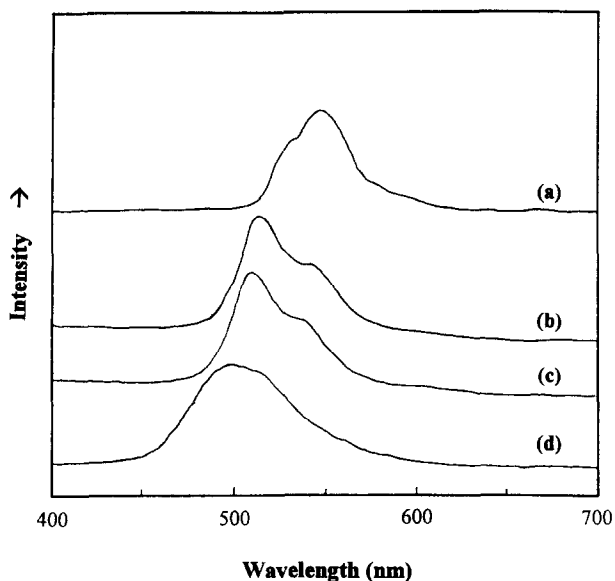


**Figure 5** *I–V* curves of (a) PPV, (b) PPV/gel-glass 4 (1/1.17) and (c) PPV/gel-glass 4 (1/2.33) LEDs



**Figure 4** PL spectra of (a) PPV, (b) PPV/gel-glass 4 (1/1.17), (c) PPV/gel-glass 4 (1/2.33) and (d) PPV/gel-glass 4 (1/118)

sites of silicon alkoxides increased. The greater the number of reactive functional sites of silane, the higher the network density in the gel-glass matrix. In turn, the movement of PPV precursor chains was restricted more severely by the network and it became harder to convert the PPV precursor to conjugated structures. Therefore, the conjugated length of the PPV in the composite became shorter as the hardness of the gel-glass (in *Table 2*) increased.

*Figure 3* depicts the photoluminescence (PL) spectra of PPV and PPV/gel-glass composites. The PPV emitted yellow-green light with an emission maximum around 550 nm. The introduction of gel-glass into PPV lead to a significant blue shift in PL spectra. The PPV/gel-glasses 1
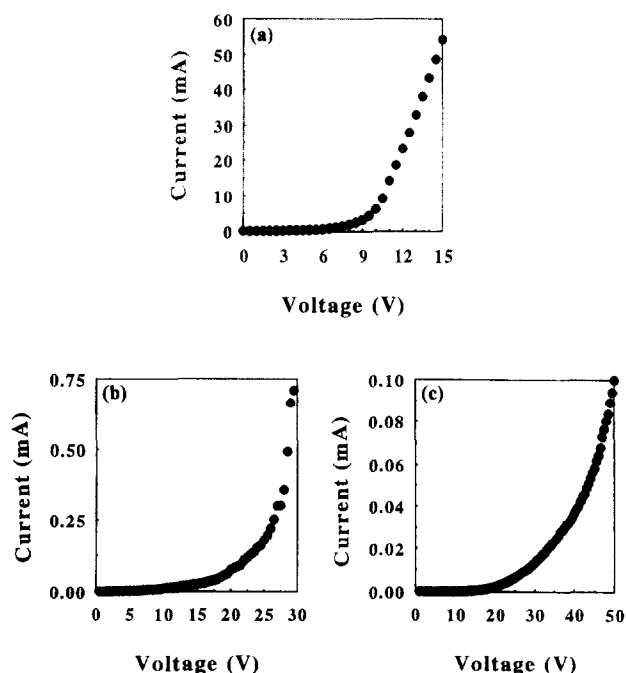
and 2 emitted a maximum peak at 515 nm with a shoulder at 540 nm and a maximum peak at 509 nm with a shoulder at 534 nm, respectively. Green light emission was observed from PPV/gel-glass 3 with an emission maximum at 510 nm. The PPV/gel-glass 4 emitted a green-blue light with an emission maximum at 496 nm. The PPV/gel-glass 5 emitted a blue light with a maximum peak at 484 nm. Besides, these PPV/gel-glass composites showed a better luminescence intensity than pure PPV by more than fourfold. PL intensity increasing with decreasing $\pi$-conjugated chain length has been demonstrated by us[22] and others[4,9]. *Figure 4* shows the PL spectra of gel-glass 4 with various molar ratios of PPV. The light emission shifted to longer wavelength as the content of gel-glass decreased. At lower gel-glass content, the ability of the network of the gel-glass to restrain the chain movement of PPV precursor converting to conjugated phenylene vinylene units in the gel-glass matrix became less significant. Therefore, the PPV conjugation became longer as more phenylene vinylene units were formed. It is clear now that, by adjusting the composition of raw materials or controlling the molar ratios of PPV to gel-glass, the light emission spectrum could be tuned in a wide range.

*I–V* characteristics of Al/PPV/ITO and Al/(PPV/gel-glass)/ITO sandwich devices are shown in *Figure 5*. In these LED sandwich devices, Al was used as the negative electrode (electron-injecting electrode), and ITO as the positive electrode (hole-injecting electrode). When the applied bias on these devices exceeded their threshold voltages, the light emission started with their own spectra. As shown in *Figure 5*, the threshold voltages of the LEDs were raised as the component ratio of gel-glass 4 increased. The currents in *I–V* curves, as shown in *Figures 5a–c*, reduced as the gel-glass 4 content increased. This resulted from a combination of the shorter conjugation of PPV with wider band-gap and the higher non-conjugated gel-glass content impeding the
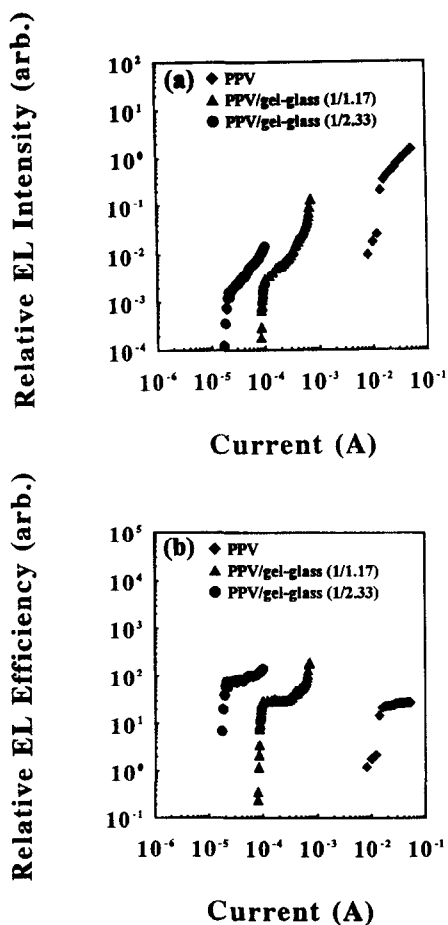
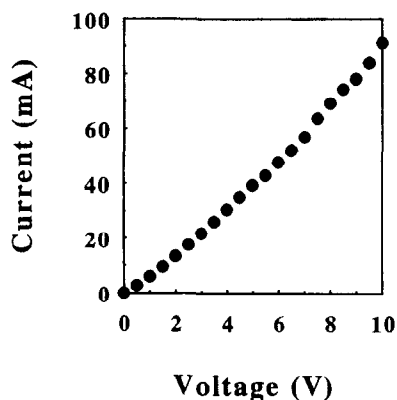**Figure 6** (a) Relative EL intensity vs. current and (b) relative EL efficiency vs. current for these LEDs



**Figure 7** *I–V* curve of PPV LED operating for 36 h

interchain transport of charge carriers. When the PPV/gel-glass 4 films of *Figure 4d* with the highest fraction of non-conjugated gel-glass were fabricated into LED sandwich devices, the current in the *I–V* curve was less than $10^{-6}$ A and no luminescence was observed under bias. The phenomenon was quite different from that in the PL process, because no interchain and/or intrachain motion of carriers are necessary in the PL process. In the EL process, however, the holes and electrons were injected from two counter-electrodes into the emissive layer to form singlet excitons within the emissive layer via interchain and/or intrachain motion. The singlet excitons could then decay radiatively. The formation
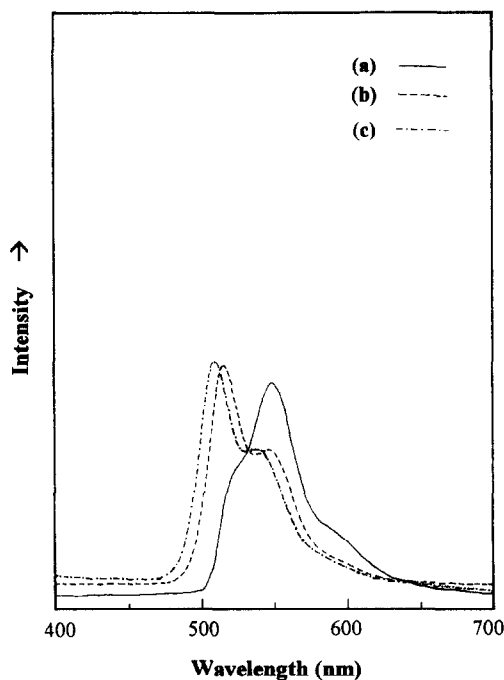


**Figure 8** EL spectra of (a) PPV, (b) PPV/gel-glass 4 (1/1.17) and (c) PPV/gel-glass 4 (1/2.33) LEDs

probability and decay mechanism of singlet excitons dominate the electroluminescence characteristics. The high fraction of the non-conjugated gel-glass may impede the motion of carriers and the formation of singlet excitons. Therefore, no electroluminescence was observed in the sandwich device made from PPV/gel-glass 4 of *Figure 4d*.

A typical dependence of relative EL intensity on applied current is shown in *Figure 6a*. The relative EL intensity of PPV LED was higher than that of PPV/gel-glass 4 (in the molar ratios of 1/1.17 and 1/2.33) LEDs. However, when the relative EL intensity was normalized by the current, i.e. the emission intensity per unit current (relative EL efficiency), these PPV/gel-glass 4 LEDs showed a higher relative EL efficiency than pure PPV LED, as evidenced in *Figure 6b*.

When the PPV LED was continuously operated under a bias voltage of 10 V, the device failed in about 40 h and the emitted light intensity decayed continuously with time. However, a significant improvement in the lifetime of the polymeric LED as observed in PPV/gel-glass LED. These devices could operate continuously for more than 100 h, although the emitted light intensity of the PPV/gel-glass 4 LEDs also reduced with the testing time. The differences in the degradation mechanism of these devices between PPV LED and PPV/gel-glass 4 LEDs are not clear at present. When we checked the *I–V* curve of a PPV LED that operated for 36 h prior to failure, the device exhibited ohmic behaviour as shown in *Figure 7*. Also the current observed at a specific voltage was higher than that in *Figure 5a*. Device failure might result from the migration of metal atoms or ions into the emissive layer to dope this layer and/or to short the device as reported in the literature[17,23]. The introduction of gel-glass into PPV resulting in improvement in the performance might be due to the migration mobility of metal atoms or ions in PPV/gel-glass being more difficult than in PPV. The reduction of mobility of ions is also related

to the decrease of the thermal vibration of PPV chain in the IPN structure. There is another physical way to reduce the effect of metal atom or ion migration into the bulk polymer layer from either electrode. The research group in Cambridge[17] have applied an a.c. bias (50% duty cycle, 1070 Hz) to operate the PPV LED, and showed a good improvement in the lifetime of the device to above 1000 h. It may be expected that the combination of the chemical approach in this article and the physical method can further improve the performance of polymeric LEDs.

The EL spectra of these LED devices were similar to their PL spectra, as shown in *Figure 8*. The PPV emitted yellow-green light with a maximum peak at 550 nm. The PPV/gel-glass 4 in the molar ratio of 1/1.7 emitted a maximum peak at 515 nm and a shoulder at 547 nm. The PPV/gel-glass 4 in the molar ratio of 1/2.33 emitted a maximum peak at 510 nm and a shoulder at 538 nm. The result of PPV/gel-glass composite prepared successfully for application in LEDs was encouraging in this study. More research in this field is promising on the way towards low threshold voltage, efficient blue light emission and long lifetime.

## CONCLUSION

A number of PPV/gel-glass composites were prepared in a novel way from PPV precursor and silicon alkoxide solutions in the presence of methanol cosolvent via a sol–gel process under ambient conditions and followed by heat treatment. The addition of titanium ethoxide shortens the PPV/gel-glass film formation time and makes it practicable for LED preparation. The conjugated chain lengths of these PPV/gel-glass composites were strongly dependent on the raw-material compositions of silicon alkoxides and their molar ratios. The u.v.–vis. absorption data showed that the conjugated lengths of these PPV/gel-glass composites were shorter than that of pure PPV. The higher the network density of the gel-glass matrix, the shorter the conjugated length of PPV in PPV/gel-glass. It may be seen that the gel-glass network at room temperature impeded the chain movement of PPB precursor converting to conjugate phenylene vinylene units during heat treatment. The resultant PPV/gel-glass composites were of transparency applicable to the optoelectric field. The incorporation of gel-glass into PPV resulted in an improvement of the lifetime of the LEDs by reducing the thermal vibration of the PPV chain in the structure, which, in turn, reduced the ion mobility. And in a LED its light emission wavelength is tunable. The photoluminescence of these PPV/gel-glass composites was in a wide range from yellow-green (550 nm) to blue (484 nm), depending on components

and composites. The PPV/gel-glass 4 LEDs, in the molar ratios of 1/1.17 and 1/2.33, emitted light with a maximum peak at 515 and 510 nm, respectively. These PPV/gel-glass 4 LEDs also showed a higher EL efficiency and a longer lifetime than PPV LED.

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