

# Conducting Polymers: Electrical Transport and Current Applications

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This review summarizes the experimental data on conducting polymers, and especially on polyacetylene, and will present several models currently used to explain the mechanisms of electrical conductivity in these materials. Much recent research has focused on electroluminescence in polymers, and a summary of advances in the fabrication of LEDs made with an electroluminescent polymer as the light-emitting layer will also be presented.

## I. Introduction

Significant electrical conductivity in polymers was first reported by Naarmann and Beck in 1961<sup>[1]</sup>, in polythiophene and polypyrrole (see Fig. 1). The conductivities reported were around 0.1 S/cm, which is comparable to the conductivity of pure germanium. In 1977, Chiang et al.<sup>[2]</sup> discovered that doping polyacetylene with iodine increased its conductivity to greater than 100 S/cm, heralding an upsurge in investigations of polyacetylene and other conducting polymers. Interest in conducting polymers has remained high ever since that discovery: polymers have chemical, mechanical and processing properties very different from those of metals and inorganic semiconductors, and the possibility to combine such properties with the electrical properties of metals has opened up many avenues of exploration, some of which have already resulted in viable industrial applications. Batteries utilizing polyaniline have been on the market since 1988<sup>[3]</sup>, for example.

Fig. 1 shows the chemical structure of several of the most important polymers which become electrically conductive when doped with oxidizing or reducing agents. All have conjugated double bonds along the main chain, and it is this chemical feature that makes conductivity possible. In the pristine state, conjugated polymers are semiconductors with band gaps between 1.5 and 3 eV. Oxidation or reduction either removes or adds electrons to the conjugated  $\pi$  system of electrons, leading to partially filled bands and therefore to metal-

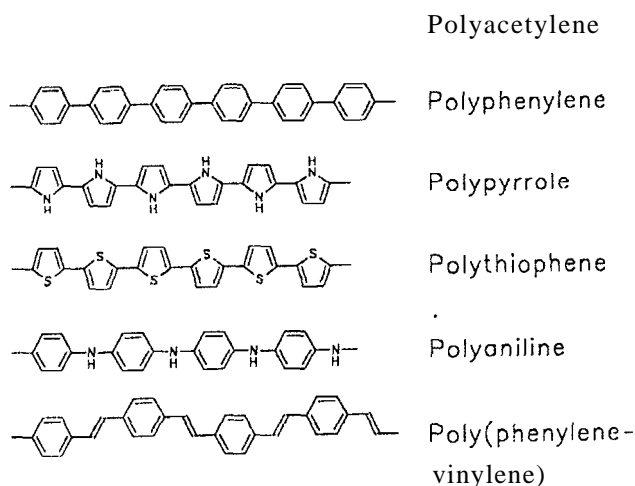


Figure 1: Chemical structure of some of the most important conducting polymers.

like conductivity. The simplest structure shown in Fig. 1, polyacetylene, is the polymer upon which the most intense research efforts have been focused, in part because of its attractively simple structure. The highest conductivity measured so far in doped polyacetylene is between  $10^5$  and  $10^6$  S/cm<sup>[4,5]</sup>, which is comparable to the room temperature conductivity of copper and considerably higher than the conductivity measured in any other conjugated polymer. A room temperature value of about  $10^7$  S/cm has been predicted for the ideal material with perfectly aligned strands and no structural defects, if it is considered a quasi-one-dimensional metal<sup>[6]</sup>.

## II. Conductivity in polyacetylene

The room temperature electrical conductivity of polyacetylene increases rapidly, by more than 6 orders of magnitude with increasing concentration of dopant<sup>[7]</sup>, up to about 1 mol%; thereafter it increases more slowly as the dopant concentration is increased, as shown in Fig. 2. The most common dopants are iodine, bromine, ferric chloride and arsenic pentafluoride, which are all p-type dopants. Alkali metals are used as n-type dopants. The total range of conductivity can span 15 orders of magnitude, from highly doped samples down to pristine, insulating samples in which unintentional dopants from the ambient, such as oxygen, or the polymerization catalyst, have been removed<sup>[7]</sup>.

In the undoped material, and at low doping levels, the structure of polyacetylene appears as drawn in Fig. 1: alternating single and double carbon-carbon bonds, with the double bonds shorter than the single bonds. If the bond lengths and the bond orders were equal, solid state physics predicts that pure polyacetylene would be a one dimensional metal with one non-bonding electron per carbon atom and a half filled highest-energy band. That the bond lengths are not equal is due to a Peierls distortion: polyacetylene is more stable if a lattice deformation occurs which creates an energy gap at the Fermi level of about 1.4 eV<sup>[8]</sup>. The energy of the electrons is thereby lowered<sup>[9]</sup>, and the polymer is transformed into a semiconductor. At high doping levels, above about 1% iodine for example, a semiconductor-to-metal transition is induced, thought to be due to a suppression of the Peierls transition.

## III. Temperature dependence of the conductivity and charge transport mechanisms

The temperature dependence of the conductivity of iodine doped polyacetylene is shown in Fig. 3. Even for the highest doping levels, in the data shown, the conductivity increases with temperature, which is typical for semiconductors but not for metals. (Some highly doped samples do show a decrease of conductivity with increasing temperature, but only near room temperature<sup>[10]</sup>, see Fig. 4). However, the sign of the temperature coefficient of the conductivity is not always

used to classify a material as metallic or not. Rather we use the zero temperature limit: if the conductivity goes to zero as  $T \rightarrow 0$  then the material is a semiconductor; if the conductivity remains finite it is a metal. The specific shapes of the temperature dependence curves do provide plentiful amounts of information about possible charge transport mechanisms, although no one mechanism seems to apply to polyacetylene over the whole range of doping. Even for a particular level of doping there may be several models that can explain the shape of the temperature dependence of the conductivity<sup>[11]</sup>.

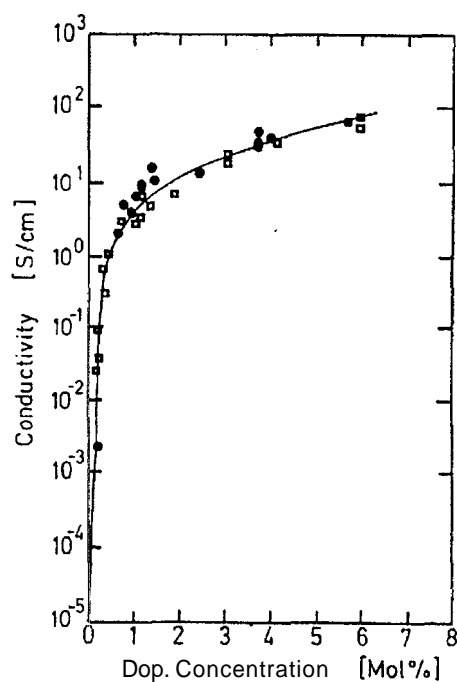


Figure 2: Change of electrical conductivity of polyacetylene upon doping with iodine.

Empirically, the electrical conductivity  $\sigma$  of very lightly doped samples of polyacetylene (dopant concentrations of less than 0.1%) shows a strong temperature dependence that is similar to that of crystalline semiconductors<sup>[12]</sup>, namely

$$\sigma \propto \exp[-\Delta E/kT], \quad (1)$$

where  $\Delta E$  is the semiconductor energy gap,  $k$  is Boltzmann's constant and  $T$  is the absolute temperature. In a crystalline semiconductor, such as silicon, electrons from the valence band are thermally excited across the energy gap into the conduction band and are able to contribute to current flow. As the temperature is increased, more electrons have enough energy to jump

to the conduction band; the number of mobile charges increases and hence the conductivity increases. These electrons are delocalized and free to move about the crystal.

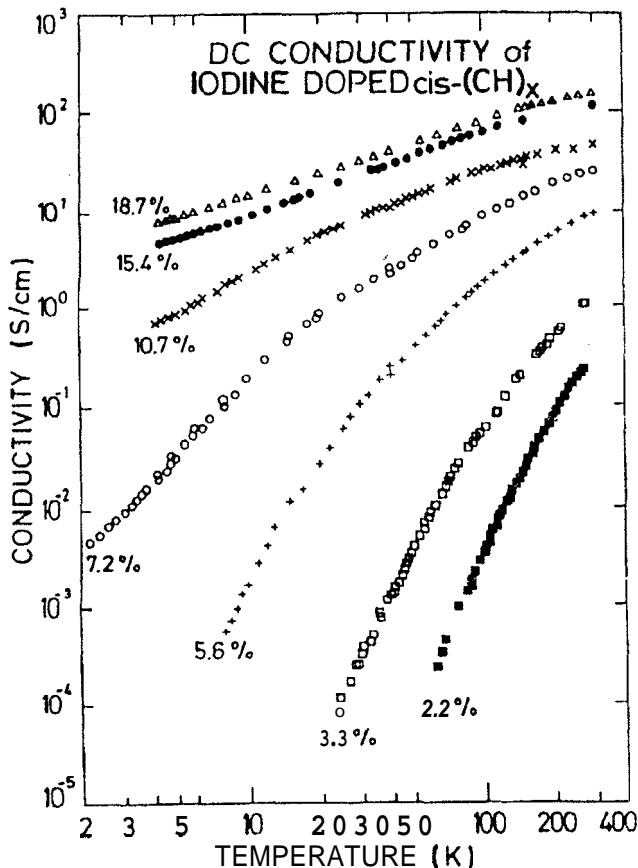


Figure 3: Temperature dependence of the conductivity of polyacetylene for various doping levels.

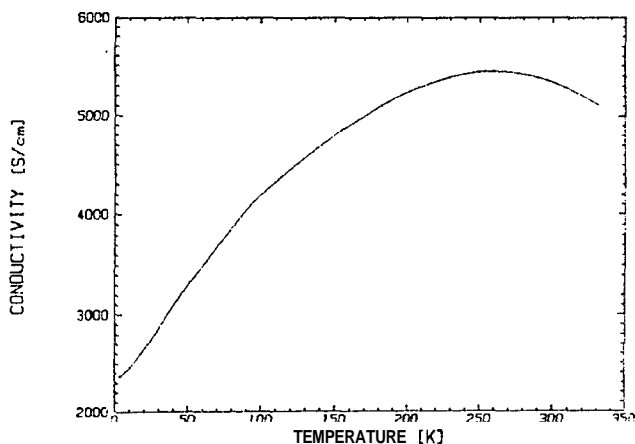


Figure 4: Some samples of highly conducting polyacetylene show a decrease in conductivity with increasing temperature, near room temperature.

The picture in very lightly doped polyacetylene is somewhat different. Undoped polyacetylene contains

conjugational defects in the form of domain walls separating regions in which the double and single bonds are interchanged, as shown in Fig. 5. Such a defect is called a soliton, a term that more generally refers to the exact solutions of certain nonlinear differential equations. In physical systems, a soliton is a nonlinear pulse-like excitation, a quasi-particle that travels freely and without changing shape. Many physicists suppose that a conjugational defect in polyacetylene can likewise move easily along the polymer chain, interchanging the double and single bonds; but the domain wall remains intact and no dispersion occurs. For review articles about solitons in polyacetylene, see references [7,13,14].

The soliton as shown in Fig. 5 contains one electron, but is electrically neutral since the electron is simply a neutral free radical. The electron occupies a localized state at the center of the energy gap<sup>[8]</sup>. Oxidation by doping removes the electron, but the conjugational defect remains, now positively charged with respect to the polymer chain. Reduction adds a second electron to the defect, giving it a negative charge. Doping can itself create charged solitons by breaking double bonds. Charged solitons are electrostatically trapped by the dopant counter-ions and so only neutral solitons are mobile. Kivelson has proposed that the electrical conductivity in lightly doped polyacetylene is due to intersoliton hopping, a process in which the mobile neutral solitons move along the chains until they approach charged solitons, and then the solitons exchange charge by hopping<sup>[15]</sup>.



Figure 5: Conjugational defect, or soliton, on a trans-polyacetylene chain.

Light to moderately doped samples have a somewhat lesser dependence on the temperature - the lower curves in Fig. 3 rather follow the law

$$\sigma \propto \exp[-\Delta E/kT]^{1/4} \tag{2}$$

which is associated with a transport mechanism called variable-range hopping, described by Mott in 1969 for amorphous semiconductors<sup>[16]</sup>. In this model, the electrons tunnel between localized states randomly located

within the energy gap. The states are close enough together in energy so that by absorbing or emitting a phonon the electron can "hop" or tunnel between sites. Since increasing the temperature increases the number and the mean energy of the lattice vibrations (phonons), the conductivity via hopping is expected to increase with temperature. The term *variable-range* refers to the variation in hopping distance of the electron: there is a physical separation as well as an energy difference between the localized states, and therefore a compromise arises between the distance the electron hops and its energy change. At a particular temperature, it may be more probable for an electron to hop over a large spatial distance to a site only slightly higher in energy, rather than to hop a short distance but to a level further away in terms of energy. As the temperature changes, the probability of the electron hopping a particular distance changes<sup>[17]</sup>. The model can be slightly modified to account for the structural anisotropy of polymers by assuming that hops within a chain are more likely than interchain hops, in which case it is called anisotropic variable-range hopping<sup>[18]</sup>.

The conduction mechanism in lightly to moderately doped polyacetylene has been attributed by some authors<sup>[13-19]</sup> to intersoliton hopping. The variable-range hopping model does not rule out the intersoliton model, since some of the localized states could be due to soliton defects<sup>[18]</sup>. A percolation model has also been suggested<sup>[20]</sup>

Highly doped polyacetylene behaves in a rather puzzling manner. The conductivity is in the metallic regime; it extrapolates to finite values at zero temperature, as expected for a metal; and the thermopower increases linearly with temperature<sup>[21]</sup>, which suggests that the number of charge carriers does not increase with temperature, also as expected for a metal. So why is the temperature dependence of the conductivity similar to that of inorganic semiconductors and opposite to that expected for metals? What is the nature of the charge transport in this doping regime?

Highly doped, metallic polyacetylene is described as an anisotropic metal by Kivelson and Heeger<sup>[22]</sup>. In their model, the intrinsic conductivity along a defect

free chain is given by

$$\sigma = \sigma_a \exp[\hbar\omega_0/k_bT], \quad (3)$$

where  $\sigma_a$  is a constant and  $\hbar\omega_0$  is the energy of the phonons that backscatter electrons. This expression shows conductivity decreasing with increasing temperature, as expected for a metal, due to phonon scattering. The intrinsic room temperature conductivity of polyacetylene is predicted by this model to be very high, higher than that of copper, precisely because of the structural differences between isotropic metals and polymers: the quasi-one-dimensional conduction path of polyacetylene limits the possible electron-phonon scattering, and only phonons with large momentum and energy will limit conduction<sup>[16]</sup>. Kivelson and Heeger suggest that material imperfections limit the measured conductivities and are also responsible for the absence of a metallic temperature dependence.

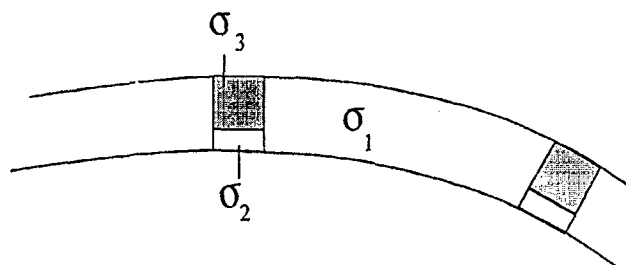


Figure 6: In a simplified model, highly doped polyacetylene is made up of highly conducting regions with conductivity  $\sigma_1$ , separated by thin barrier regions. The barriers consist of disordered metallic regions with conductivity  $\sigma_2$ , in parallel with regions of conductivity  $\sigma_3$  in which hopping or tunneling occurs. After [23].

To account for the observed temperature dependence of the conductivity in laboratory measurements, Kaiser and Graham<sup>[23]</sup> suggest a heterogeneous model. They attribute the high conductivity to quasi-1-D metallic conduction as suggested by Kivelson and Heeger, but reason that the observed temperature dependence is due to some other process or processes for which the conductivity is assisted by temperature. In the model proposed, highly conducting regions are separated by thin barrier regions, with each barrier region comprised of two different barrier regions in parallel as shown in Fig. 6, so that the total conductivity can

be written as

$$\sigma^1 = (g_1 \sigma_1)^{-1} + (g_2 \sigma_2 + g_3 \sigma_3)^{-1}, \quad (4)$$

where  $g_i = x_i/l_i$  are geometric coefficients that depend on the fraction  $l_i$  of the sample length and the fraction  $x_i$  of the sample cross-sectional area associated with conductivity  $\sigma_i$ . Using this model, the temperature dependence of the conductivity follows that of the barrier regions, and the magnitude of the total conductivity, while less than  $\sigma_1$ , is greater than either  $\sigma_2$  or  $\sigma_3$  due to multiplication by the geometric coefficient and can therefore be quite high. This model can be applied to a material with fibrillar structure such as polyacetylene<sup>[7]</sup>, each fibril made up of many strands and having a certain cross-sectional area.

Now consider the barrier regions: one conduction pathway is through a disordered metallic region, which ensures that there will be a finite conductivity in the zero temperature limit. The conductivity  $\sigma_2$  of this region is small, with only a small temperature dependence. Microscopically, it can be attributed to inter-chain transfer. In parallel with this region there is a region where transport can be described by fluctuation-induced tunneling<sup>[24]</sup>, a temperature assisted process which can account for the puzzling temperature dependence. Fluctuation-induced tunneling is a mechanism which describes transport in a disordered material with predominantly high metallic conduction but with conductivity limited by thin insulating barriers. The following expression is used to describe fluctuation-induced tunneling through a parabolic barrier:

$$\sigma_3 = \sigma_c \exp[-T_c/(T + T_s)], \quad (5)$$

where  $\sigma_c$ ,  $T_c$  and  $T_s$  are constants. Simple elastic tunneling is temperature independent, but the above mechanism assumes that the tunnel junctions, the barriers, are small and therefore subject to large fluctuations in voltage across the junction, caused by thermal fluctuations. The probability of tunneling through the barriers increases with temperature. Many research groups use the fluctuation-induced tunneling model to describe transport in highly doped polyacetylene<sup>[7,19,25-27]</sup> but some authors argue that the picture of a heterogeneous system such as the one described above is not applica-

ble to doped polyacetylene<sup>[11]</sup>. Paasch et al.<sup>[26]</sup> point out that whereas the fluctuation-induced model is often used assuming that the barriers are due to interfibrillar contacts, it is better to assume that the barriers arise from inhomogeneous distribution of dopants. In a simplified picture of Kaiser and Graham's model, Kaiser suggests that the fluctuation-induced tunneling occurs across chain breaks or defects<sup>[21]</sup>.

#### IV. Applications of conjugated polymers

In order to understand the mechanism of electrical conductivity in conjugated polymers, polyacetylene has been studied extensively because of the simplicity of its chemical structure. Unfortunately, polyacetylene is unstable in the presence of oxygen and water, and is unprocessable because it is insoluble and decomposes when heated instead of melting; it is not the material of choice for industrial applications despite its high conductivity. Many of the other conjugated polymers are stable, and even processable when their structures are modified with soluble sidechains<sup>[28,29]</sup>. There are currently several companies that market conducting polymers in various forms, especially polyaniline, polythiophene and polypyrrole<sup>[3]</sup>. None of these, or any other, conducting polymers have electrical conductivities nearly as high as that of polyacetylene, but for many applications high conductivity is not important. As an example, through-hole plating of printed circuit boards is a process that can be achieved by depositing doped polypyrrole on the walls of the holes in the circuit boards, and then electroplating copper on the polymer so that both sides of the board are electrically connected<sup>[3]</sup>. As a second example, high quality aluminum electrolytic capacitors have been made using doped polypyrrole as a solid electrolyte<sup>[30]</sup>. These capacitors are very stable in the presence of moisture and heat, and have excellent frequency characteristics. The polymer film can be made very thin, which makes high values of capacitance possible.

A current area of research is investigating processable, undoped conjugated polymers that exhibit electroluminescence (see Fig. 7), and using them as the emissive layer in light-emitting devices (LED's). A schematic representation of a polymer LED is shown

in Fig. 8. It is hoped that these polymers will make possible new types of display devices such as large area panels, which would be too expensive and difficult to make with inorganic semiconductor LED's or other conventional display technologies<sup>[31]</sup>. Polymers can be made quite easily into high quality, large area thin films and have the further advantage of requiring less stringent manufacturing conditions than inorganic devices<sup>[32]</sup>, which would further lower the cost. Imagine a television screen so thin and light that it could be mounted on the wall like a picture, or even a giant sized display screen that could be rolled up when not in use.

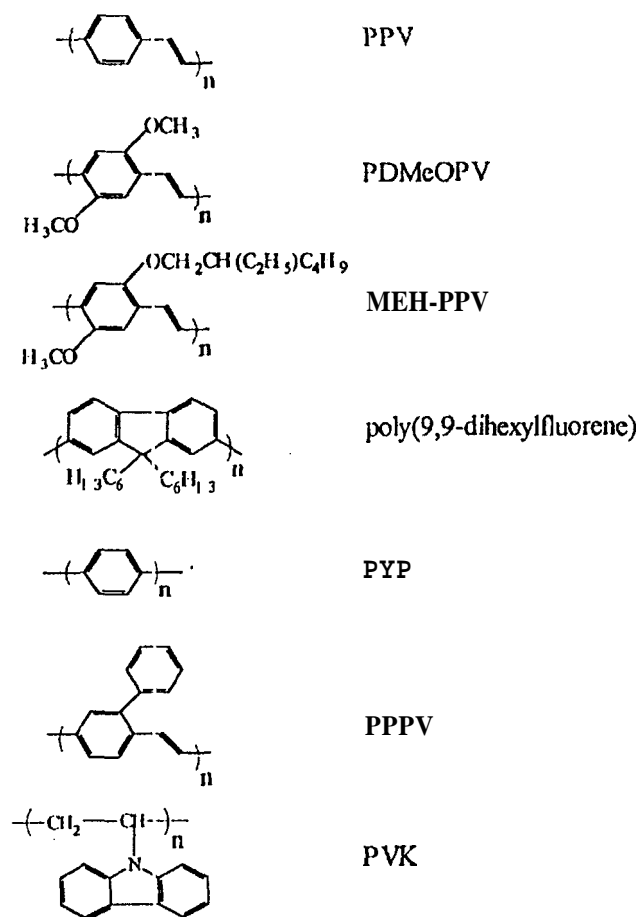


Figure 7: The chemical structures of some electroluminescent conjugated polymers: poly (p-phenylene vinylene) or PPV; poly (2,5-dimethoxy - p-phenylene vinylene) or PDMcOPV; poly (2-methoxy-5-(2-ethyl-hexoxy)- p-phenylene vinylene) or MEH-PPV; poly (9,9-dihexylfluorene); poly (p-phenylene) or PPP; poly (p-phenylphenylene vinylene) or PPPV; poly (9-vinylcarbazole) or PVK.

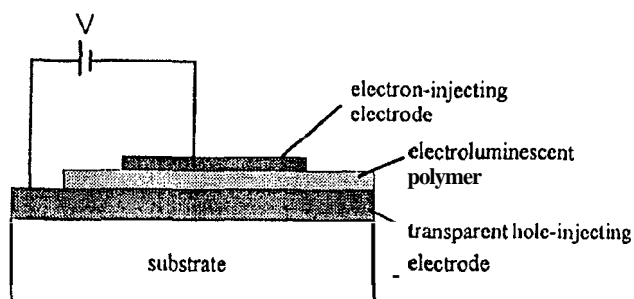


Figure 8: An LED fabricated with a conjugated polymer as the active, light-emitting layer.

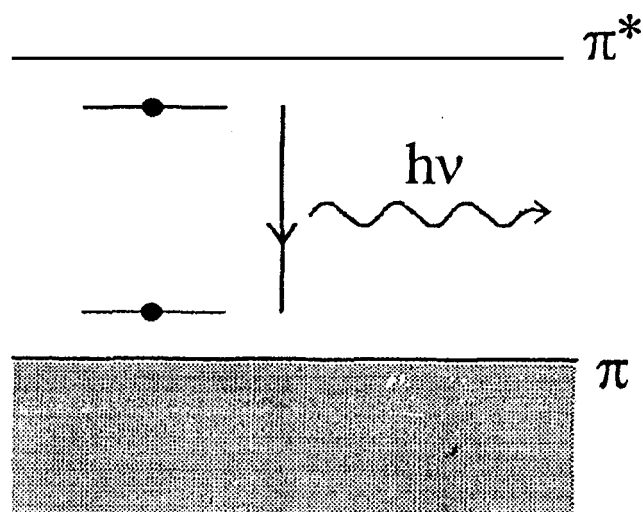


Figure 9: An exciton decaying radiatively.

Electroluminescence is the emission of light that results when electrons and holes injected into a luminescent material from separate electrodes travel through the material under the influence of an applied electric field, and combine to form an exciton, which subsequently decays radiatively<sup>[31]</sup>, as shown schematically in Fig. 9. The first report of an LED fabricated with a conjugated polymer as the active layer was made in 1990 by Burroughes et al.<sup>[33]</sup> in Cambridge. That group made a series of simple sandwich devices consisting of a thin layer of poly(*p*-phenylene vinylene) or PPV between a negative, electron-injecting electrode such as aluminum in some devices, and a positive, hole-injecting electrode such as gold or indium oxide. The PPV films were made by spin-coating a soluble precursor polymer on the bottom electrode followed by thermal conversion. The devices emitted yellow-green light visible under normal room lighting when just under 14 volts was applied, but the efficiencies were very low,

only 0.05%, or five photons emitted for every 10,000 charges injected.

It is perhaps surprising that electroluminescence in conjugated polymers was not investigated until 1990. One reason may be that polyacetylene, the most widely studied conjugated polymer, exhibits only very weak photoluminescence<sup>[33]</sup>, a process very similar to electroluminescence. Another reason is that most research with conjugated polymers in the past has been done with doped samples, and doping inhibits the emission of light by increasing the number of exciton quenching sites<sup>[34]</sup>. But since the discovery of electroluminescence in polymers, researchers seem to be making up for lost time.

LEDs made with other conjugated polymers, and copolymers with PPV, with increased efficiency, were reported<sup>[35]</sup> soon after the first LED was reported. The increased efficiency in LEDs made with the copolymers was attributed to trapping of created excitons in shallow potential wells, which in turn are due to variation in the  $\pi - a^*$  energy gap along the copolymer chain. The trapped excitons are more likely to decay radiatively, as mobile excitons have several available routes to efficient non-radiative decay. A maximum efficiency of 0.3% was seen for a copolymer of PPV, which has an energy gap of 2.4 eV, and 10% PDMeOPPv, or poly(2,5-dimethoxy-p-phenylene vinylene) with an energy gap of 2.1eV<sup>[35]</sup>.

The next advance was reported by Braun and Heeger<sup>[36]</sup>. They fabricated a device using a substituted PPV which has the advantage of being soluble, and so the film could be cast directly from solution without further processing. The device emitted yellow-orange light, but no improvement in efficiency was reported.

Besides advantages in processing, the wide range of conjugated polymers with different energy gaps and the opportunity to further tune the energy gaps means that a large range of emitted colors is possible<sup>[31,32,36]</sup>. One way of tuning the energy gap is by chemical substitution: for example, adding methoxyl sidechains to PPV changes the  $a - a^*$  energy gap from 2.5 eV (yellow emission) to 2.1 eV (red emission)<sup>[37]</sup>. Using copolymers can allow good control of the emission: Burn et al.<sup>[38]</sup> ob-

served a linear shift in the emission spectra with varying composition of PPV and methoxyl substituted PPV copolymers.

By now, all of the basic colors of the spectrum have been achieved in polymer LEDs: red, orange, yellow, green and blue<sup>[31,39]</sup>. It is difficult to make blue LEDs out of inorganic materials, but the first blue LED based on a conjugated polymer was reported by Ohmori et al.<sup>[40]</sup> in late 1991. Their device was made with poly(9,9-dihexylfluorene) as the active layer, which has an energy gap of 2.9 eV. The efficiency was not reported. Very shortly after, Grem et al.<sup>[41]</sup> reported a blue LED using poly(p-phenylene) or PPP, with an efficiency of 0.01-0.05%. Recently, Zhang et al.<sup>[39]</sup> reported blue light emission from diodes with efficiencies of up to 0.16%, by using polymer blends of poly(p-phenylphenylene vinylene) or PPPV and poly(9-vinylcarbazole) or PVK.

The above devices were fabricated with rigid substrates and electrodes. One of the electrodes must be semi-transparent and the material of choice is usually indium/tin oxide on glass<sup>[33,36,39-43]</sup>. The indium/oxide is brittle, as well as the glass. Gustafsson et al.<sup>[44]</sup> fabricated the first fully flexible polymer LED by using poly(ethylene terephthalate) as the substrate, doped polyaniline as the transparent hole-injecting electrode, a substituted PPV as the electroluminescent layer and calcium as the electron-injecting electrode. The devices could be bent sharply, and furthermore exhibited fairly high efficiencies, among the highest so far reported, of about 1%, and turn-on voltages of only 2-3 volts.

## V. Conclusions

In the 17 years since high electrical conductivity was first measured in doped polyacetylene, and in the 4 years since the first LED was made using PPV, much progress has been made in understanding transport in conjugated polymers, and in developing applications that take advantage of their versatile properties. It is possible that conjugated polymers will become indispensable for the manufacture of large area display devices, an up-and-coming field in which, thus far, no

other technologies have shown as much promise in terms of flexibility, competitive efficiency and ease of manufacturing, combined with reasonable cost.

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