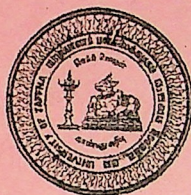# UNIVERSITY OF JAFFNA
SRI LANKA

Prof. Sivapathasuntharam

Mageswaran

Memorial Lecture
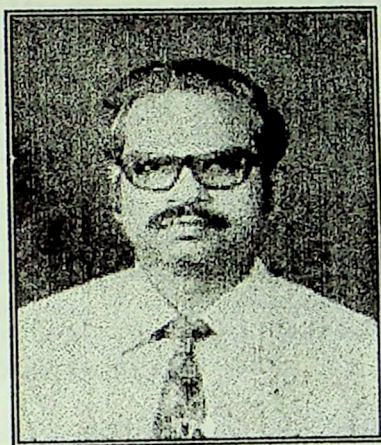# 2001

By

Dr. K. Kandasamy

Prof. Sivapathasuntharam

Mageswaran

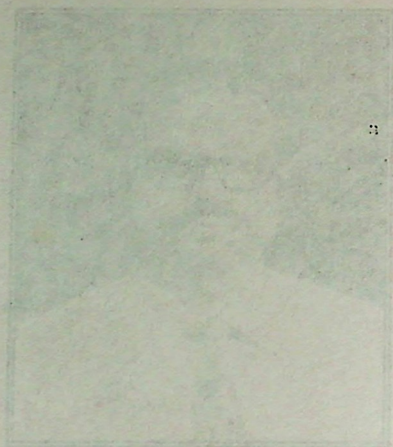# Memorial Lecture
2001

By

Dr. K. Kandasamy

☞ Third Memorial Lecture - 2001

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Prof.Sivapathasuntharam Mageswaran Memorial Lecture - 2001
Introduction

Late Professor Sivapathasuntharam Mageswaran is one of the pioneers of the University of Jaffna. He was one of the most dedicated and dynamic pioneers who played a major role in guiding the development of this University.

Prof.Mageswaran was one of the finest synthetic organic chemists Sri Lanka produced. His students and well-wishers have graciously established a fund for conducting a memorial lecture annually to honour Prof.Mageswaran and to remember his yeoman services.

I am happy to see Prof.K.Kandasamy , another pioneer of the University delivering this third memorial lecture. Prof.Kandasamy, a young and dynamic academic, is speaking on a topic relevant to technological development in the 21st century. Such activities should be appreciated and encouraged.

I wish Prof.Kandasamy every success and for the continuation of activities of this nature.

Prof.P.Balasundarampillai
Vice-Chancellor.
University of Jaffna.
21 March 2001.

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Organic Semiconductors for the 21ˢᵗ century

K.KANDASAMY.

Ladies and Gentlemen

I am fortunate to have got this rare privilege of presenting today's memorial lecture of Late Prof.Sivapathasuntharam Mageswaran, which is the third lecture of this series. Since the inception of Science discipline in the University of Jaffna from 1975, I have had the opportunity to work with him and observed him closely as an academic colleague. I firmly believe that Prof. Mageswaran was one of the finest academic administrators we had in the history of the University of Jaffna. While I was preparing for my final year examination at the University of Peradeniya in the latter part of 1973, Prof.Mageswaran returned to Sri Lanka after his postgraduate studies at the University of Sheffield. My roommate in the Arunachalam Hall, University of Peradeniya, who was a chemistry special student projected him as a talented academic and all rounder on many occasions. I have witnessed this rare quality in Prof. Mageswaran until I met him last at the Jaffna hospital before I left for Japan in 1998 to spend my sabbatical leave. Even during his last lapse of life his only worry was about the university. He was particularly worried about the attitude of the probationary lecturers who were resigning their posts after obtaining postgraduate degrees abroad. I would like to use this opportunity to make an appeal to our probationary staff: If you really wish to

honour Prof.Mageswaran's dedicated services to the university kindly return to the university after your postgraduate studies abroad. With this request, I shall commence my lecture.

In this lecture with my experience as a material scientist, I wish to talk on organic semiconductors. Since the fabrication of the first transistor by Shockley , Brattain and Bardeen in 1948 inorganic semiconductors offered an interesting ground for investigation of condensed matter physics and also for the development of technology based electronic devices. As such, in the past material scientist concentrated mainly on metals, alloys , semiconductors and other inorganic compounds. Organic compounds were studied by chemists and biologists. Now the situation has changed . A conducting polymer called polyacetylene was first synthesized by accident in the early 1970s by Hideki Shirakawa of the Tokyo Institute of Technology. Its conducting nature was identified in 1977 by MacDiarmid and his colleagues at the University of Pennsylvania. The discovery that organic materials can conduct electricity and emit light not only broke the boundaries of various disciplines but also contributed to a rich interdisciplinary research in science that is capable of making immense impact on technology. This is because these organic materials offer many advantages over their inorganic counterparts. These organic materials are durable, flexible and cheap. Many electronic devices such as field effect transistors (FET), light emitting diodes (LED) , photovoltaic (PV) cells, Laser light of all wavelengths in the visible region etc using organic materials are now under development. Many investigators foresee broad marketing opportunities in the future for

flexible graphic displays, smart windows, rechargeable batteries, electrolytic capacitors, artificial muscles, electromagnetic screens, antistatic coatings, camouflage coatings etc.

A special class of organic materials known as conjugated polymers have the right physical and chemical properties to conduct electricity. Some heavily doped conjugated polymers can have high conductivities even exceeding the values of well known inorganic semiconductors. In fact some conjugated polymers can function both as a semiconductor and a conductor depending on the level of doping. Some examples for conducting polymers are given in figure 1.
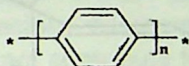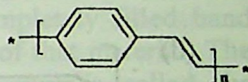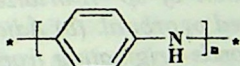
1. Polyacetylene
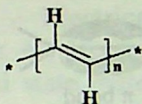
2. Polypyrrole
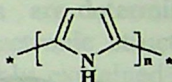
3. polyphenylene

4. polyphenylene vinylene

5. Polyaniline
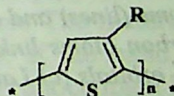
6. Poly(3-alkylthiophene)

Figure 1:*Chemical structures of some conjugated polymers.*

These carbon based materials offer a unique opportunity for looking at different kind of physics, largely determined by the formation of a partly localized π (pi) - orbital . This π- orbital originates from the lateral overlap of the p_z orbitals of adjacent sp^2 hybridized carbon atoms as illustrated in figure 2.
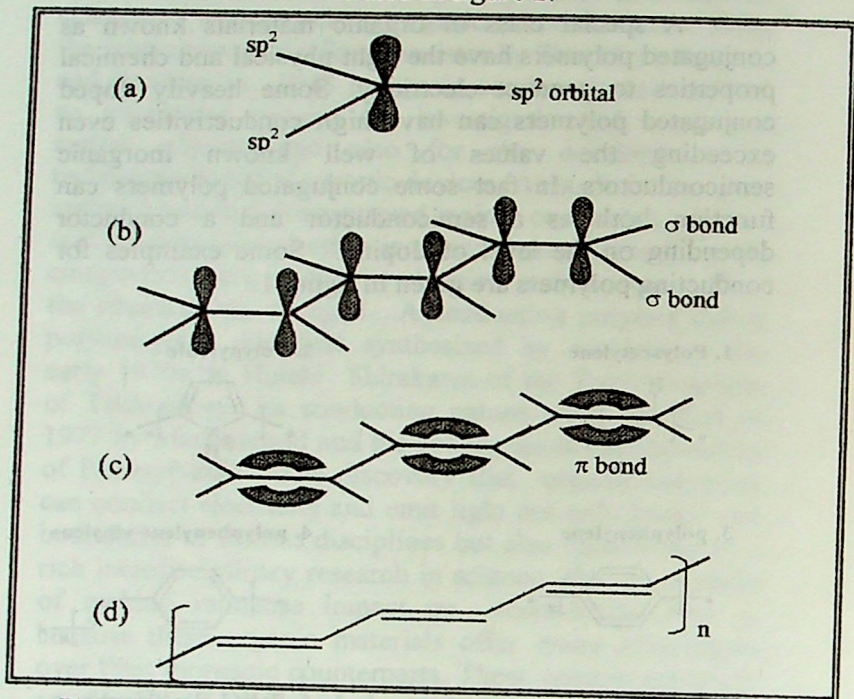

Figure 2: *(a) Schematic representation of sp^2 hybridized C atoms (lines) and of the non-hybridized p_z orbital. (b) Adjacent carbon atoms linked by the sigma bonds originating from the sp^2 orbitals (c) Lateral overlap of the p_z orbitals from adjacent carbons forming one π bond between every two carbons. (d) The conjugated structure of polyacetylene.*

The material formed in this way are called conjugated (figure 2.c) and the whole structure is rationalized in terms of alternating single and double bonds as in polyacetylene (figure 2d). But it would be more appropriate to look at the π-electron wave function extended over single and double bonds. The presence of π- electrons in these materials introduce remarkable differences with respect to σ bonded organic molecules. They are responsible for the low value of the redox potentials and reasonably large intrachain mobility (Hoofman *et al.* 1998). Precise physical mechanisms that enable conjugated polymers to conduct electricity is not yet well understood but purity and the arrangement of polymer chains in particular seems to be crucial. By stretching polyacetylene, for instance, material scientists make polyaceteylene to conduct electricity to a level comparable to that of copper.

Electrical properties of solids are determined by their electronic structure . The electronic structure of solid state materials can be reasonably explained by the simple band theory. According to the band theory of solids, electrons in solid state materials occupy different energy bands in accordance with Pauli exclusion principle. Electrons occupying a completely filled band do not contribute to the conductivity of that material. The highest occupied band at absolute zero is called the valence band. If this band is partially filled the material is a conductor. If the valence band is completely filled at absolute zero then there is no conduction. The material is either an insulator or a semiconductor. If the band gap between the valance band and the lowest unoccupied band at absolute zero is narrow then thermal excitation of

electrons from the valence band to this unoccupied band at normal temperatures is possible. Therefore at normal temperatures these kinds of materials start to conduct electricity and are called semiconductors. The band that accepts electrons from the valence band due to thermal agitation is called the conduction band in the study of semiconductors. Both the valance band and the conduction band contribute to conductivity in semiconductors.

The electronic structure of organic semiconductors can also be described using the band theory. Of the two types of bonds between carbon atoms along the chain of a conjugated polymer, the σ -bonds are strong. This is because these bonds originate from the overlap of sp^2 hybridized orbitals of two adjacent carbon atoms along the inter nuclear axes. Since these bonds are strong and electrons in them are strongly localized, they do not contribute to the conductivity of the materials. The π-bonds are weak because they originate from the lateral overlap of p_z orbitals of the adjacent carbon atoms in the chain. These bonds are delocalized and the electrons are mobile. These mobile electrons are responsible for the conductivity in conjugated polymers. A slight distortion in the bond length between the adjacent carbon atoms along the conjugated chain contributes a potential which opens up a gap between the bonding band and the anti-bonding band of π-electrons. The energy gap between these two bands is comparable to the band gap in group IV and group III/V semiconductors. There are two valence electrons in a π-bond per repeat unit of the chain, which leads to filled bonding band and empty antibonding band and give semiconducting properties to

the material. In analogy to the inorganic semiconductors, the bonding band is called valence band and the antibonding band is called conduction band. But not all electrical and optical properties of these organic semiconductors could be explained well with this simple band picture for their electronic structure. Polymers, by nature, are disordered materials with many types of morphological and chemical defects. These defects very often can result in structural deformation and hence modification in their electronic structure. For example when electrons are removed from the top of the valence band of a conjugated polymer by oxidative doping/p-doping using iodine or oxygen or introduced to the conduction band by reductive doping/n-doping using alkali metals, the charge carriers created in the polymer do not delocalize completely as we would have expected in band theory. Only partial delocalization occurs, extending over a few repeat units of the conjugated polymer and causing them to deform structurally. This deformation results in the formation of energy levels within the band gap (in analogy with the rise in energy when an electron is removed from a filled bonding molecular orbital). These levels are called polaron levels if doping (number of holes created) is low and bipolaron levels if doping is high. In the polaron state only one hole is spread over a few repeat units of a polymer. But in bipolaron state a pair of holes is spread over a few repeat units of a polymer. A polaron and bipolaron in a polypyrrole are shown in figure 3.

Polaron spread over four units
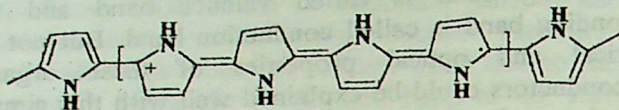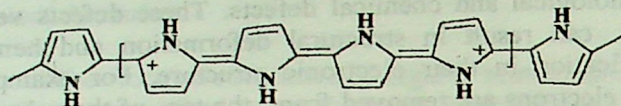

Bipolaron spread over four units

Figure 3: *A polaron and bipolaron in polypyrrole*

Both polarons and bipolarons are mobile and can move along the polymer chain by the rearrangement of double and single bonds in the conjugated systems. Conduction by polarons and bipolarons is now thought to be the dominant mechanism of charge transport in semiconducting polymers. This concept also explain very well the changes in optical absorption observed in these polymers with doping. Therefore a knowledge about the structural deformation induced modifications in the electronic structure of organic semiconductors is necessary to explain their electrical and optical properties. The semiconducting property of organic conjugated polymers makes them usable in the design of many electronic devices. Here we shall look in detail the use of organic conjugated polymers in a few important devices such as the Light Emitting Diode (LED), Photovoltatic cell (PVC) and Field Effect Transistor(FET).

Light emitting diodes

Research on polymer electroluminescence was first reported in 1983 by Roger Partridge of the National Physical Laboratory, U.K. But the research activities gathered momentum in 1990 when Richard Friend and his colleagues at Cambridge University reported emission of Green-yellow light from conjugated polymer poly phenylene vinylene or PPV. A typical design of a LED is shown in figure 4.
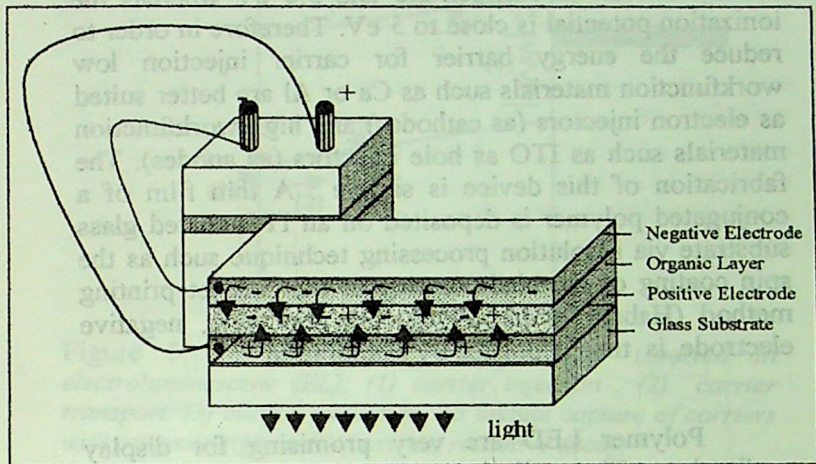

Figure 4: *Schematic structure of an organic polymer LED. A voltage applied to the LED sends negative and positive charges from the electrodes to the organic polymer where they combine and give off light.*

This LED consists of a thin film of conjugated polymer such as PPV sandwiched between two electrodes. When a d.c. voltage is applied to the electrodes, negative charges are injected from one contact and positive charges (holes) from the other. Some of the opposite charges injected into the polymer combine and emit light. At least one of the electrodes has to be transparent in order to ensure efficient light out put. The indium tin oxide (ITO) is a transparent and conducting material and therefore a suitable choice as one of the electrodes. The electron affinity of most organic semiconductors is between 2.5 and 3.0 eV whereas the ionization potential is close to 5 eV. Therefore in order to reduce the energy barrier for carrier injection low workfunction materials such as Ca or Al are better suited as electron injectors (as cathodes) and high workfunction materials such as ITO as hole injectors (as anodes). The fabrication of this device is simple . A thin film of a conjugated polymer is deposited on an ITO coated glass substrate via a solution processing technique such as the spin coating or the blade casting or the ink-jet printing method (Habner *et al.* 1998). The top layer, negative electrode, is then deposited by evaporation.

Polymer LED are very promising for display applications. They offer a fast response combined with high brightness, visibility ,contrast, and viewing angle. They can be flat, flexible, could be operated at low voltage and are relatively simple to manufacture. Major drawbacks of polymer LEDs are their emission inefficiency and short lifetime. The emission efficiency of LED can be expressed as the ratio of the number of photons emitted to the number of charge carriers passing

through the device. This is sometimes known as quantum efficiency. The emission efficiency is determined at least by four subprocesses namely (see figure 5)

     (1) charge injection,
     (2) charge transport,
     (3) electron-hole pair (exciton) formation,
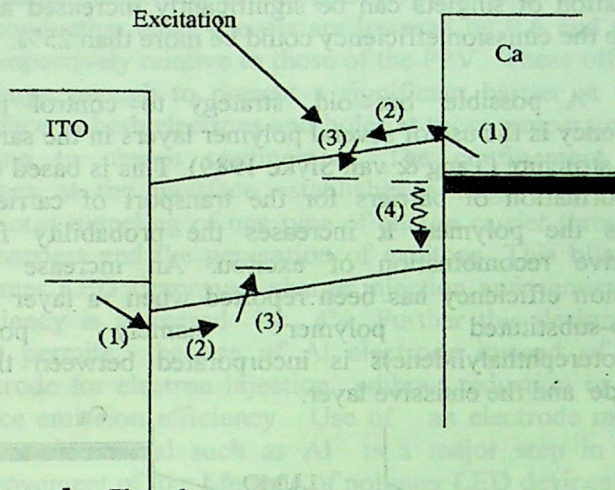   (4) radiative recombination of excitons.

Figure 5: *The four main processes involved in electroluminescene (EL); (1) carrier injection , (2) carrier transport, (3) exciton formation via mutual capture of carriers with opposite sign and (4) exciton radiative decay.*

Upon the application of an electric field charge carriers of opposite sign are injected from the electrodes and transported across the polymer. Once injected, each carrier can interact with an oppositely charged carrier and form coulombically bound exciton or can swept across the whole polymer film and finally be ejected into the other electrode. Of the excitons formed it is believed that 75%

will be in the triplet state (with spin 1) and the rest 25% in the singlet state (with spin 0). Only the singlet state excitons can undergo radiative recombination. A part of the light generated in the polymer layers can also escape from the device. As such maximum emission efficiency expected is 25%. Recent theoretical studies (Shuai *et al.* 2000), however, have indicated that the probability of formation of singlets can be significantly increased and hence the emission efficiency could be more than 25%.

A possible, but old, strategy to control the efficiency is the use of several polymer layers in the same LED structure (Tang & van Slyke 1989). This is based on the formation of barriers for the transport of carriers across the polymer. It increases the probability for radiative recombination of exciton. An increase of emission efficiency has been reported when a layer of cyano-substituted polymer namely poly (cyanoterephthalylidene)s is incorporated between the cathode and the emissive layer.
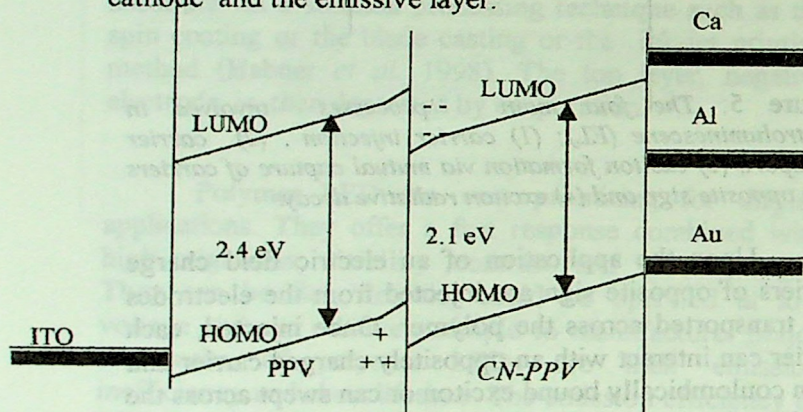

Figure 6: *Energy band diagram of a PPV/CN-PPV junction.*

Figure 6 shows the band modification introduced by the inclusion of a cyano derivative of PPV in the LED between the light emitting layer PPV and the cathode of LED. Experimental studies reveal that in CN-PPV the highest occupied molecular orbital (HOMO)- the equivalent of the valence band- and the lowest unoccupied molecular orbital (LUMO)- the equivalent of the conduction band - levels are lowered by 0.6 and 0.9 eV respectively relative to those of the PPV. These offset are large enough to present a significant barrier at the interface to both electrons and holes at room temperature, leading to charge confinement. The confinement of charges at the interface establishes a large field and promotes tunneling of one type of charge carrier through the barriers and the generation of excitons. This bilayer polymer LED improved charge injection and emission efficiency is increased by 4%. Further this design of LED permits the use of Al electrode instead of Ca electrode for electron injection without reduction in the device emission efficiency. Use of an electrode made from stable metal such as Al is a major step in the improvement of the life time of polymer LED devices.

The injection process at the anode also has some effect on the emission efficiency of LED. Substantial improvement in emission efficiency has been observed by inserting a polymeric hole transport layer (HTL) between the emissive layer and ITO. These HTLs can be poly (styrene Sulphonate), (PSS), (Cacialli 2000) , poly 3,4 ethylene dioxythiophene, (PEDOT) (Carter *et al*. 1997), polyaniline doped with camphor-sulphonic acid (Karg *et al*. 1996) or other materials with high ionization

potential. It was also reported that inserting a hole transport layer such as PEDOT between the ITO and the emissive layer enhanced the lifetime of the LED (Kim *et al.* 1999). The efficiency luminance and lifetime of this device can also be improved by giving oxygen-plasma treatment to the ITO (Kim *et al.* 1998). A major drawback of this multilayer technique is the increase in the electric field required for a given luminosity. This causes a higher power dissipation and hence accelerate device degradation. In this respect another method of increasing the efficiency using blends of different organic semiconductors is more promising, since it allows a large interfacial area between different polymers without introducing energy barriers but increasing the probability for formation of excitons (Cacialli et al 1998). Yet another interesting way of increasing the luminescence is via incorporation of the LED into an optical resonator (Dodabalapur et al.1994). This gives substantial narrowing down of the emission spectrum and enhancement of the emission in the forward direction.

Research activities during the last ten years have helped the technologists to increase the quantum efficiency of LED from 0.001% to more than 10% (Samuel *et al.* 2000). This is a marvelous progress. At present the efficiency of the devices made with organic semiconductors exceed those of many inorganic LEDs. Improvements in the materials used and the fabrication techniques have helped the designers to make LEDs with life time greater than 10,000 hours. This means that now organic LEDs are ready to replace the inorganic LEDs in lighting and display applications. However organic LED technology is not yet matured. In the future we hope to

see many useful advancement and applications of this technology. This is evident from the competition between many firms to share global market worth over $ 50 bn per year for electronic display. In fact some of these firms are ready for large scale production (Physics world March 1999).

Photovoltaic Cell

Another area of application of organic semiconductors is in the fabrication of solar cells and photo detectors. The photovoltaic effect involves the production of electrons and holes in semiconductor devices under illumination and their subsequent collection at the electrode of opposite charges. In many inorganic semiconductors photon absorption produces free electrons and holes directly. But in polymer semiconductors light absorption creates excitons, which are bound at normal temperatures. Therefore charge collection requires the dissociation of the excitons and the quick transport of the products to the electrodes. To some extent the separation and collection of charges are problematic. But technologist show increased interest in developing organic photovoltaic cells for solar energy conversion because of the high installation cost of inorganic semiconductor photovoltaic cells. Exciton dissociation is efficient at interfaces between materials with different electron affinities and ionization potentials; the electrons are accepted by a material with higher electron affinity and the holes by a material with lower ionization potential (Tang 1989). This means that the materials and device requirements differ from those of LEDs. A mixture of acceptor and donor semiconductor polymers can provide phase-separated structures, which go some way in

meeting this requirement and providing high photoconductive efficiencies. A very successful way of achieving this is to make a blend of two such polymers. In the blend electrons will move into the polymer with high electron affinity and holes will move into the polymer with low ionization potential. The separated charges can then be transported through each polymer to the electrodes. A typical design of a photovoltaic cell is shown below in figure 7.
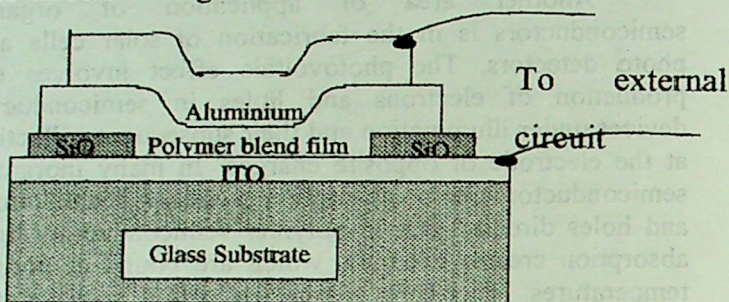

Figure 7: *Schematic diagram of a polymer photodiode*

Halls et al. (1995) in their design used CN-PPV and MEH-PPV to make a blend. They argued that in their device excitons are dissociated at the interface between CN-PPV and MEH-PPV polymers; the electrons are transferred to CN-PPV and holes to MEH-PPV. Graiström et al (1998) used MEH-CN-PPV as electron acceptor and PODT as hole acceptor in their design. The chemical structure of these polymers are shown below in figure 8.
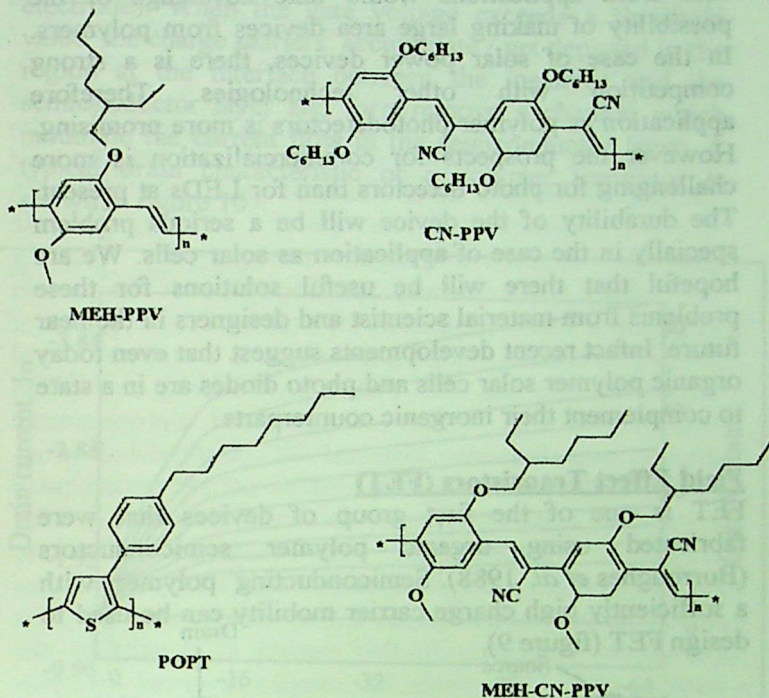
Figure 8: *Chemical structures of the polemers used in photovoltaic diodes; MEH-PPV-poly(2-methoxy-5-(2'-ethyl)-hexyloxy-p-phenylene vinylene, CN-PPV- cyano derivative of poly phenylene vinylene, POPT-phenyl-octyl substitued polythiophene and MEH-CN-PPV cyano derivative of poly(2-methoxy-5-(2'-ethyl)-hexyloxy-p-phenylene vinylene.*

The above design can be used for two purposes: When a bias is applied it will function as a photodiode; In the absence of an applied bias it can be used as a solar cell. Both applications would take advantage of the possibility of making large area devices from polymers. In the case of solar power devices, there is a strong competition with other technologies. Therefore application as polymer photodetectors is more promising. However the prospects for commercialization is more challenging for photo detectors than for LEDs at present. The durability of the device will be a serious problem specially in the case of application as solar cells. We are hopeful that there will be useful solutions for these problems from material scientist and designers in the near future. Infact recent developments suggest that even today organic polymer solar cells and photo diodes are in a state to complement their inorganic counterparts.

Field Effect Transistors (FET)

FET is one of the first group of devices that were fabricated using organic polymer semiconductors (Burroughes *et al.* 1988). Semiconducting polymers with a sufficiently high charge carrier mobility can be used to design FET (figure 9).
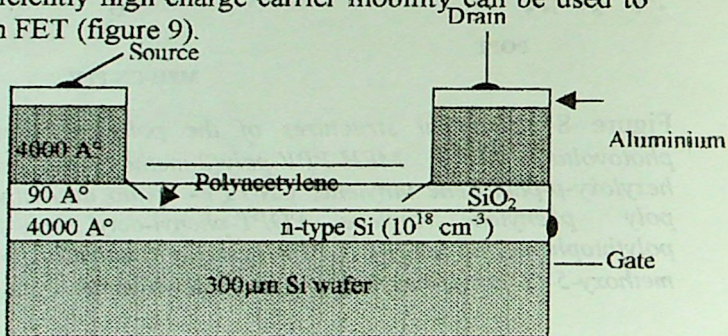

Figure 9: *Schematic diagram of a polymer Field Effect Transistor.*

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In this the gate is separated from the semiconductor polymer, polyacetylene, by an insulating layer. The current flow between the source and the drain is controlled by applying a voltage to the gate (the third electrode). When the gate voltage exceeds a threshold value the charge carriers accumulate / get depleted in the region at the interface between the insulator and the semiconductor layer and form/remove a channel which modulate the current flow in the semiconductor layer. A typical drain characteristic of a polymer transistor is shown in figure 10.
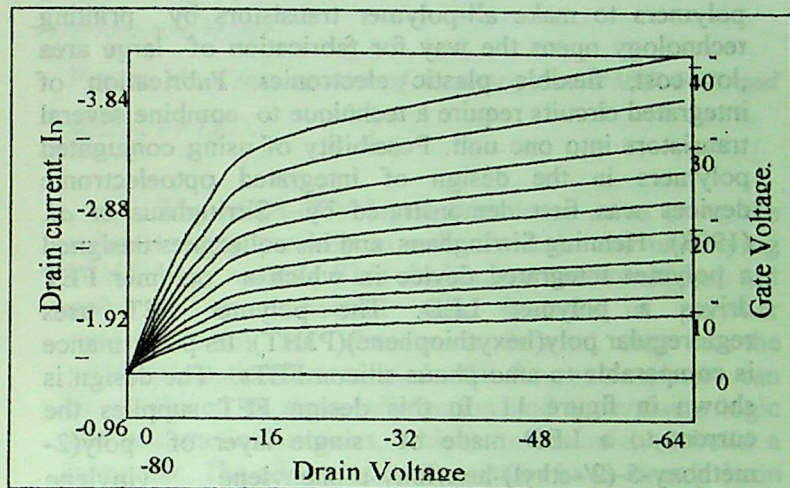

Figure 10: *Drain current (I_D) versus drain voltage (V_D) curves for various gate voltages.*

The performance of these transistors is very modest compared to the performance of inorganic devices made of silicon or GaAs. But it seems that they are healthy competitors to inorganic transistor devices.

The ability of polymer transistors to perform logic operations was demonstrated by Brown et al (1995). In the design of NOT and NOR gates, Brown and his colleagues used Metal-Insulator Semiconductor Field Effect Transistors (MISFET) with conducting channel made of poly thienylenevinylene (PTV). Further they demonstrated the suitability of these two gates for logic operations by designing a ring oscillator. In 1990 use of organic polymers was extended to the fabrication of the substrate of FET by Garnier. Later in 1994 he used printing techniques to fabricate all parts of FETs with organic polymers (Garnier 1994) . Use of organic polymers to make all-polymer transistors by printing technology opens the way for fabrication of large area ,low-cost, flexible plastic electronics. Fabrication of integrated circuits require a technique to combine several transistors into one unit. Possibility of using conjugated polymers in the design of integrated optoelectronic devices was first demonstrated by Sirringhaus *et al.* (1998). Henning Sirringhaus and his colleagues designed a polymer integrated device in which a polymer FET drives a polymer LED. The polymer FET uses regioregular poly(hexythiophene)(P3HT). Its performance is comparable to amorphous silicon FETs. The design is shown in figure 11. In this design FET supplies the current to a LED made of single layer of poly(2-methoxy-5-(2'-ethyl)-hexyloxy-p-phenylene vinylene (MEH-PPV).
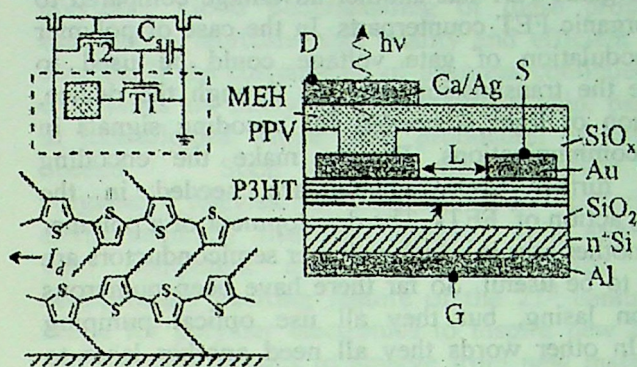
Figure 11: *Cross section of the integrated P3HT FET and MEH-PPV LED.*

The MEH-PPV layer is made so as to cover the substrate only partially in order to make the underlying P3HT layer visible. This design can be considered not only as an important step forward in polymer optoelectronics but also as a forward step in the polymer/plastic electronics. In the future, transistors can be combined to store information and to evaluate logic and arithmetic functions, which are essential to build a computer. The all- polymer transistors have already been combined into several test devices and logic circuits such as inverters, two input NAND gates and flip-flops. Dago de Leeuw and his colleagues at the Phillips research laboratories had designed a programmable code generator that can be used to store a 15 bit digital number. This circuit combines 326 transistors, 200 interconnects, a 5-bit counter, a decoder logic and 15 programming pads. (Dago de Leeuw 1999).

Organic FET has another advantage compared to their inorganic FET counterparts. In the case of polymer FET, modulation of gate voltage could be used to modulate the transmission of light through the device. Modulation of light is needed for encoding signals in optical communications. But to make the encoding effective further improvements are needed in the switching action of FETs. The development of a polymer laser is another area in which polymer semiconductors are expected to be useful. So far there have been numerous reports on lasing, but they all use optical pumping method. In other words they all need another laser to excite the polymer medium to lase. However recently the design of a light emitting FET using crystalline organic semiconductor, α - sexithiophene has been reported. Design of this device is a promising start for the fabrication of low cost, large area organic semiconductor lasers in the future.

What is expected in the 21st century ?

21st century, no doubt, is going to witness a healthy competition between organic-polymer and inorganic electronic devices. In the field of technology, the organic electronic devices will be complementing their inorganic counterparts in many applications. The remarkable progress in polymer electronics has been due to excellent cooperation between physicists, chemists and material scientists. This will be expected to continue in the 21st century too. There are numerous questions that need to be answered. For example we have not clearly understood the factors controlling the charge transport in semiconducting polymers and about the excited states that

are responsible for emission of light from the conjugated polymers. Good understanding of these properties is essential to fabricate high quality and efficient polymer devices. If the current rate of progress continues, plastic electronics are likely to replace silicon chip, particularly in technologies where mass production of circuits are needed. Soon we might find ourselves watching "organic" televisions, reading news papers under environmentally friendly "organic" light and working with computers made of plastic chips. Another academically exciting feature of the 21st century is the scope for polymer electronics to inspire new chemistry and for new physics to emerge from new materials and devices.

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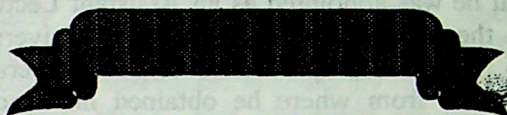
Professor Kathirgamanathan Kandasamy is the Head of the Department of Physics and an Associate Professor of Physics in the University of Jaffna, Sri Lanka. He entered the Faculty of Science of the then University of Ceylon, Peradeniya Campus in 1970 and graduated in Physics with Honours in 1974. After graduation, he served as a temporary Assistant Lecturer in Physics in the Peradeniya Campus of the University of Sri Lanka until he was appointed as an Assistant Lecturer in Physics in the Jaffna Campus of the same University in June 1975. In 1977, he proceeded to the University of Keele, England from where he obtained his Doctorate Degree in 1980. After obtaining the post graduate degree, he returned to the University of Jaffna and served as a Lecturer in Physics. He was promoted to the Senior Lecturership in 1986 and appointed as an Associate Professor of Physics in 1992. He served as the Head of the Department of Physics for several years since 1988.

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His research abilities have been recognised both nationally and internationally. The Abdus Salam International Centre for Theoretical Physics (ICTP), Italy, a prestigious institution founded by the Nobel Laureate Prof. Abdus Salam, and sponsored by UNESCO and IAEA appointed Prof. Kandasamy as an Associate of the Centre in 1997. He is now holding this position for a second term of three years duration. His research abilities have also been recognised nationally by his appointment as a member of the National Research Council of Sri Lanka by Her Excellency the President of SriLanka for a period of three years from August 1999.

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