Temperature Dependence of Current in Plasma Polymerized 1-Benzyl-2-Methylimidazole Thin Film

Rama Bijoy Sarker[1*], A. H. Bhuiyan[2]

Abstract: Plasma polymerized thin films in Al/polymer/Al sandwich configuration was prepared using 1-benzyl-2-methylimidazole as organic monomer by a parallel plate capacitively coupled glow discharge reactor. It has been found that the current-voltage characteristics of the PPBMI thin films obey space charge limited current model. The dependence of current density on inverse absolute temperature was investigated at constant voltages. The electrical conductivity of the deposited films increases with increasing temperature. For 5 V, the activation energies were estimated from the measured data and were found to be 0.16 eV and 0.78 eV in the low and high temperature regions, whereas for 15V, the low and high temperature activation energies have been found to be 0.17 and 0.81 eV respectively.

Keywords: Plasma polymerization, capacitively coupled, glow discharge, space charge limited conduction, activation energy.

Introduction

A plasma is a partially ionized gas consisting of ions, electrons and neutral atoms. It is a state of matter that can be formed by different techniques. The technique of most interest to plasma polymerization is the glow discharge, in which free electrons acquire energy from the imposed electric field and loses it through collisions with the neutral molecules present in the gas phase. This transfer of energy to gas molecules leads to the formation of a host of chemically reactive species, some of which becomes precursors for the plasma polymerization process. Polymerized thin films can be formed by several methods: such as glow discharge [1], electron bombardment [2] and vacuum evaporation [3].

Thin films prepared by plasma polymerization have been found versatile applications in a wide variety of fields such as solar mirror protective coatings [4], microelectronics [5], optoelectronics, biomedical [6], humidity sensors [7], chemical sensors [8] etc. The films formed by glow discharge technique are pinhole free, highly cross-linked and exhibits a number of unique and desirable properties. These films are insoluble in organic solvents and resistant to high temperatures. The glow discharge method is simpler than the other

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conventional methods from a processing point of view. Plasma polymer films are highly coherent and adherent to a variety of substrates including conventional polymer, glass and metal surfaces.

The plasma polymerization process differs from the conventional molecular polymerization by that it is an atomic polymerization, in which the molecular structure of the starting monomer is not retained. Jalal et al. [9] **s**tudied the conduction mechanism in plasma polymerized m-xylene (PPm-X) thin films and inferred that the Poole-Frenkel conduction mechanism was operative in PPm-X thin films.

The activation energies in plasma polymerized N,N,3,5-tetramethylaniline thin films were estimated to be 0.19 ± 0.02 and 0.85± 0.05 eV by Akther et al. [10]. Guermat et al. [7] studied plasma-polymerized films of hexamethyldisiloxane as sensing layer for humidity sensor development by using a capacitively coupled parallel plate reactor. They reported that thickness and discharge power are two main parameters that govern sensor characteristics for moisture detection. Films with lower thickness and low discharge power provide the optimum deposition condition for fabricating high performance humidity sensors. In this context, ac plasma polymerization technique has been used for deposition of PPBMI thin films. In the present study samples in the Al/PPBMI/Al sandwich form were prepared by conventional metal coating unit for the dc measurement. Our attention is focused to investigate the temperature dependence of the current density of the sample. We also report the dc conductivity of the PPBMI thin films as a function of voltage and temperature.

Experimental Procedure

Film preparation

1-Benzyl-2-methylimidazole ($C_{11}H_{12}N_2$) (Tech. grade 90%, Aldrich chemical company, USA) in liquid form was used as monomer in this work (Fig. 1). Glass slides (18 mm × 18 mm × 1mm) (Sail brand, China) were used as substrates. In order to produce high quality PPBMI thin films the glass substrates were carefully cleaned using acetone and distilled water in an ultrasonic bath sequentially. The cleaned glass slides were dried and preserved in well-cleaned desiccators. A schematic diagram of the apparatus used for plasma polymerization is given in figure 2. The polymerization process took place in a home-made capacitively coupled parallel plate reactor. The system consist of a cylindrical chamber (0.16 m diameter, 0.20 m high), a pair of symmetrical electrodes (0.09 m diameter) separated by a distance of 0.04 m, vacuum system and a monomer inlet system.

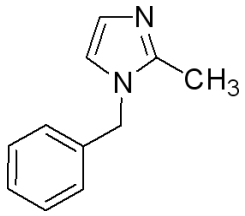

Fig. 1: *1-Benzyl-2-methylimidazole structure*

Substrates were placed in the lower electrode and the deposition chamber was evacuated to 1.33 Pa with a rotary pump (Vacuubrand, Germany). In order to evaporate the liquid monomer and to avoid condensation, the monomer container and the supply line was heated to about 150 °C by a heating element of nichrome wire. For electrical measurements samples of Al/PPBMI/Al sandwiched structures were prepared. Aluminum (Al) of high purity (purity 4N, British chemical standard) was used as the metal electrode. The Al electrode was evaporated from a tungsten filament onto substrates through suitable mask to form the base electrode in a conventional metal coating unit (Model: Edwards 306, England, UK]. To perform the electrical measurements under dynamic vacuum, the vacuum chamber was pumped down to a pressure of about 1.33×10^{-3} Pa. The aluminum deposited glass substrates were placed at the center of the lower electrode for polymer thin film deposition. Then PPBMI thin films were deposited through a square shaped mask to form the film over the electrode.

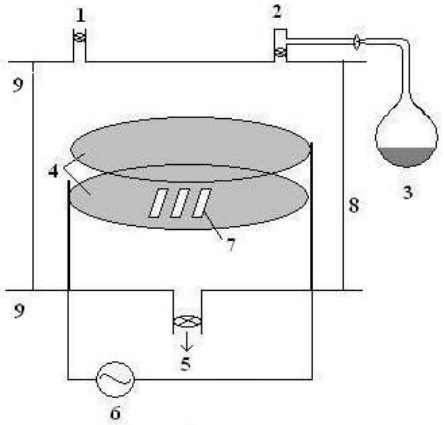

Fig. 2: *Plasma polymerization setup: 1. Pressure gauge, 2. Monomer inlet through a flowmeter, 3. Monomer container, 4. Steel plate electrode, 5. Vacuum pump, 6. AC power supply, 7. Substrates, 8. Glass cylinder 9. Brass flanges.*

After the deposition of PPBMI film onto first Al electrode the substrates were taken out of the polymerization chamber to deposit the second Al electrode. The lower and upper Al electrodes are rectangular in shape with an effective cross-sectional area 10^{-4} m^2. The deposition was carried out for a period of 45 minutes to 1 hour. PPBMI thin films of different thicknesses were deposited under optimum conditions. Good electrical contacts between the Al electrodes and the connecting Cu wire have been achieved by means of silver paste.

Measurements

For electrical measurements, dc bias voltage was provided by a power supply (Model: Agilent 6545A, Agilent Technologies, Malaysia) and the current was recorded by an electrometer 614 (Keithley Instruments Inc., USA). The temperature of the sample was

monitored using Cr-Al thermocouple attached to the film substrate and connected to a digital microvoltmeter (Model: 197A, Keithley Instruments, USA). The thickness of the PPBMI films was measured by Fizeau interferrometric method.

Results and Discussion

Voltage dependence of current density

Figure 3 shows the voltage dependence of current density, *J* of PPBMI thin film of thickness 150 nm at various temperatures. According to the data presented in Figure 3 it is observed that each curve is characterized by two different slopes on this double logarithmic plot. In the lower voltages a slope of approximately 1 indicates ohmic conduction, whereas in the higher voltage regions, the slopes of approximately 2.5 indicates space charge limited conduction (SCLC) in PPBMI thin films. In absence of the traps, for SCLC the charge transport through a thin polymer film can be written as [11]

$$J = \frac{9}{8}\mu\varepsilon\varepsilon_0\frac{V^2}{d^3} \qquad (1)$$

Where ε is the dielectric constant of the material, ε_o is the permittivity of free space. Equation (1) shows that the current density depends quadratically on the applied voltage *V*. The current density at higher temperatures is increased significantly revealing a temperature dependence of the current density. It is seen that as the temperature increases the knee voltage corresponding to the sharp rise in current density in PPBMI thin film shifts toward higher voltages.

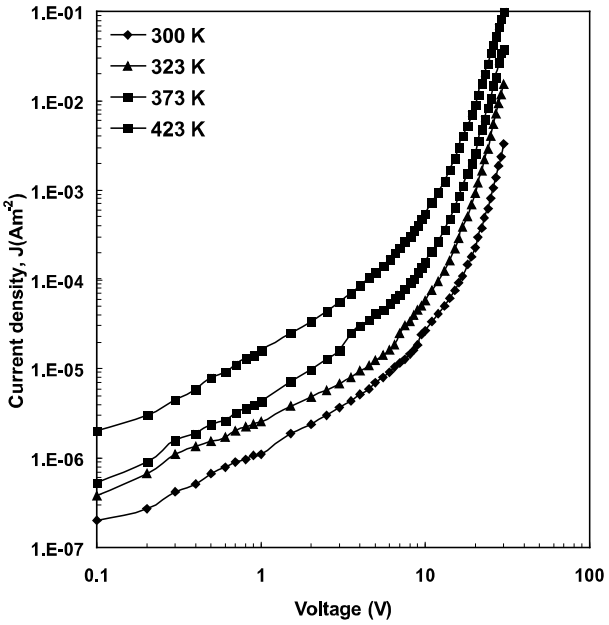

Fig. 3: *Voltage dependence of current density for150 nm thick PPBMI thin film.*

Temperature dependence of current density

The temperature dependence of *J* can be expressed by Arrhenius equation

$$J = J_0 \exp(-\tfrac{\Delta E}{kT})$$

Where J_0 is a constant, ΔE is the thermal activation energy of electrical conduction and *k* is the Boltzmann constant. Figure 4 shows the dependence of *J* on inverse absolute temperature, 1/T, for PPBMI thin film of thicknesses 150 nm, in the ohmic region for 5 volts and in the SCLC region for 15 volts. According to the data presented in the Figure 4 it is observed that both the curves have varying slope at low temperatures but becomes almost linear in the higher temperature region, corresponding to a well-defined activation energy. It is also observed that the current density increases slowly for temperatures <340 K and above this temperature *J* increases rapidly with temperature. This increase in *J* with temperature may be due to the increased movement of the adventitious ions and/or electrons. The activation energies associated with two temperature regions were calculated from the slopes of $\log_{J} - \frac{1}{T}$ plot for the sample of thicknesses 150 nm. For 5 V, at low temperature region the activation energies are found to be around 0.16 eV and at higher temperature region it is 0.78 eV, whereas, the low and high temperature region activation energies are found to be 0.17 eV and 0.81 eV for an applied voltage of 15 V. These small variations of the activation energies in the low and high temperature regions suggest the existence of the shallow traps levels in PPBMI thin films. The low activation energies in the low temperature region indicate that the thermally activated hopping conduction is operative in this material. This change in ΔE from lower to higher values may be attributed to a transition from a hopping regime to a regime dominated by distinct energy levels [12].

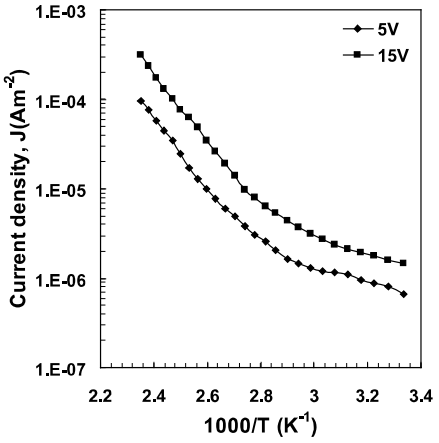

Fig. 4: *Variation of current density with inverse absolute temperature for PPBMI thin film in the ohmic and non-ohmic regions (Thickness, d=150 nm).*

Conclusions

The *J-V* characteristics of PPBMI thin film with Al electrodes show ohmic behaviour at the lower voltage region and SCLC in the higher voltage region. It is found that the conduction is a thermally activated process having average activation energies of 0.16 and 0.79 eV in the low and high temperature regions respectively. These values of ΔE in the lower and higher temperature regions indicate that at low temperatures the conduction is a hopping process and at higher temperatures it is dominated by distinct energy level transitions.

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References:

1. Cremona A., Laguardia L., Vassallo E., J., 2005, *Appl. Phys.* **97,** 023533(1-5).
2. Robert W., Christy, J., 1960, *Appl. Phys.* **31**(9), 1680 –1683.
3. Ibrahim A. A., 2006, *Vacuum*, **81,** 527–530.
4. Bieg R. W. and Wiscmann K. B., 1980, *Sol. Energy Matter.* **3**, 301.
5. Kubono A., Okui N., 1994, *Prog. Polym. Sci.* **19,** 389-438.
6. Muguruma H., 2007, *Anal. Tr. Chem.* **26** (5), 433-443.
7. Guermat N., Bellel A., Sahli S., Segui Y., Raynaud P., 2009, *Thin Solid Films* **517**(15)**,** 4455-4460.
8. Inagaki N. and J. Ohkubo, 1991, *J. Appl. Polym. Sci.* **43**, 793-800.
9. Shah Jalal A. B. M., Ahmed S., Bhuiyan A. H, and Ibrahim M., 2005, *Thin Solid Films*, **295,** 125-130.
10. Akther H. and Bhuiyan A.H., *Thin Solid Films* **488,** 93-97.
11. Mott N. F. and Gurney R. W., 1940, Electronic Processes in Ionic Crystals, Clarendon Press, Oxford, p168.
12. Gould R. D., 1982, *Appl. J. Phys.* **53**(4)**,** 3353-3355.