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MOLECULAR PHOTODIODE CONSISTING OF FLAVIN-VIOLOGEN-TCNQ LB MULTILAYERS

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Abstract A moleculear photodiode was fabricated with the hetero-Langmuir-Blodgett(LB) film consisting of an electron acceptor(A), an electron relay(R) and a sensitizer(S). TCNQ, viologen and flavin derivatives were used as A, R and S units, respectively. By aligning hetero-LB film of A/R/S units on ITO glass with an aluminum thin film, a molecular photodiode with the structure of Metal/Insulator/Metal(MIM) was constructed. Due to excitation by irradiation with a 460 nm monochromatic light source, the photo-induced unidirectional flow of electrons in the MIM device could be achieved and was detected as photocurrents. The direction of energy flow was in accordance with the energy level profile across the LB films. The photoswitching function was achieved and the rectifying characteristic was observed in the molecular device.

INTRODUCTION

In the initial process of photosynthesis, a biological electron transfer system, photoelectronic conversion occurs and then long-range electron transfer takes place very efficiently in one direction through the biomolecules.[1] The specific energy and electron transfer takes place on a molecular scale due to the redox potential difference as well as the electron transfer property of functional molecules, especially the electron-acceptor and sensitizer.[2-3] Various artificial molecular devices have been fabricated by mimicking the electron transport function of biological photosynthesis.[2-5] The electrochemical photodiode consisting of Langmuir-Blodgett(LB) films of three functional molecules or an aligned triad on the electrode which worked in electrolyte solution have been made.[3] The MIM structured device consisting of hetero-LB film of a sensitizer and an electron acceptor was fabricated and photo-induced electron transfer was investigated.[2,5] In the present paper, the MIM structured device was fabricated with the hetero-LB film consisting of TCNQ, viologen and flavin derivatives, as A, R and S, respectively. Molecules of three functional materials were arranged on ITO glass regularly in space normal to the electrode surface, e.g. A/R/S, by the LB method. Finally by depositing aluminum on the hetero-LB film, a molecular device with MIM

structure was constructed. Photocurrent properties of the MIM structured device were investigated to evaluate the direction of electron transfer and photoswitching function.

EXPERIMENTAL DETAILS

Three kinds of functional materials were used. N-Dococylquinolium-TCNQ(TCNQ), N-Allyl-N'-[3-propylamido-N",N"-di(n-octadecyl)]-4,4'-bipyridium Dibromide(viologen), 7,8-dimethyl-10-dodecyl isoalloxazine(flavin) were used as A, R and S units, respectively. These three materials were synthesized. The measurement of surface pressure-area isotherms and the deposition of LB films were carried out with a circular Langmuir trough (Nima Tech., England).

Cyclic voltammetry was carried out at 25°C with a CV-75 potentiostat (BAS, Germany). The working (Pt plate), reference (Ag/AgCl) and counter (Pt plate) electrode were used. Acetonitrile with $NaClO_4$, KCl aqueous solution and acetonitrile with $TBABF_4$ were used as the electrolyte solution. The working electrode was deposited with LB film of functional materials for the measurement of redox potential.

The molecular photodiode was fabricated with the hetero-LB film consisting of A, R and S units, respectively. A schematic diagram of the apparatus for photocurrent measurement was shown in Fig. 1. An input exciting light of wavelength 460nm was generated with a xenon lamp system. The photocurrent was detected through a current-voltage amplifier, A/D converter and personal computer. I-V measurement was carried out using Hewlett Packard 4145B parameter analyzer.

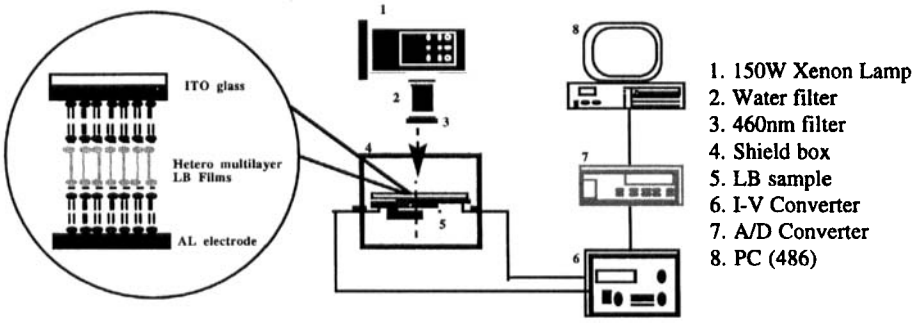

FIGURE. 1. Schematic illustration of a hetero-type cell and experimental configuration

RESULT AND DISCUSSION

The π-A isotherm of each material was shown in Fig. 2. The isotherm of the monolayer of S has two condensed regions at 20mN/m and 40mN/m. The monolayer was more stable at 40mN/m and the limiting area per molecule was 24Å^2. The

isotherms for the monolayer of S and A were similar pattern. The monolayer of A was more stable at 45mN/m and the limiting area per molecule is 35Å^2. In contrast, the expanded and condensed regions was clearly observed for the monolayer of R. At a surface area less than 60Å^2/molecule, there was an abrupt increase of slope due to a phase transition. The limiting area per molecule is 60Å^2.

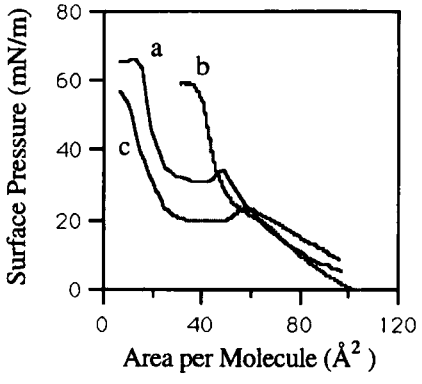

FIGURE. 2. Surface pressure-area isotherms of functional materials: a, A; b, R; c, S.

The cyclic voltammogram of A, R and S were shown in Fig. 3. During positive potential sweep, the anode currents showed a maximum value at 0.33V, -0.15V and -0.16V. When the cyclic direction was reversed, the oxidized forms of A, R and S were reduced back to the original starting material at 0.28V, -0.44V and -0.5V. Thus the redox potential of A, R and S were 0.3V, -0.3V and -0.36V, respectively.

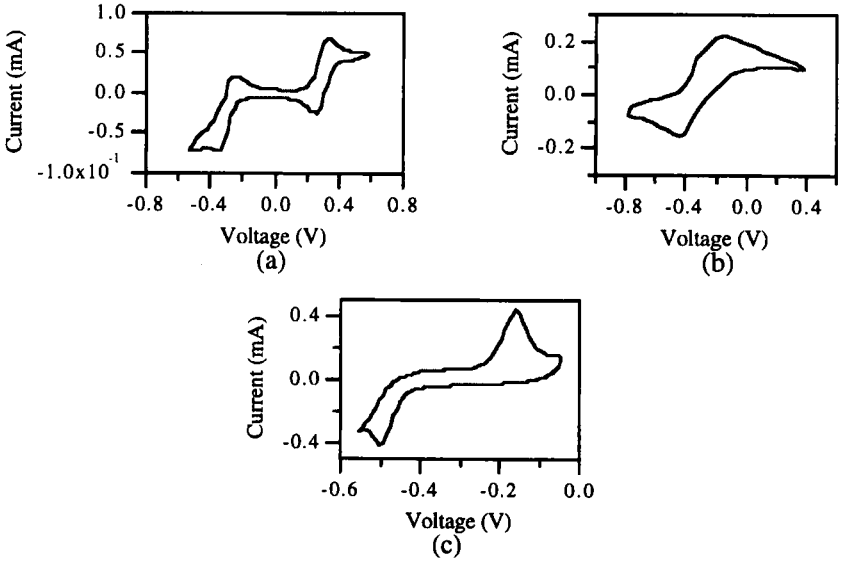

FIGURE. 3. Cyclic voltammograms of functional materials : a, A; b, R; c, S

The photocurrent-time response with the irradiation of a 460nm monochromatic light by a xenon lamp was shown in Fig. 4. When a forward bias was applied in accordance with the energy level profile in the MIM device, a stable photocurrent was generated. With repeated step illumination, the reproducible photocurrent was generated accordingly. The photocurrents were very stable and level of responses was consistent during the repeated cycle over 30 minutes. The results indicated that the photoswitching function of the MIM device was achieved. When reverse bias was applied, photocurrent was much smaller than that of forward case. In the proposed molecular device, the photo-induced unidirectional flow of electrons could be achieved due to the redox potential difference as well as electronic coupling between the functional molecules. As shown in Fig. 5. the rectifying characteristic was observed from the measurement of photocurrent(I)-voltage(V).

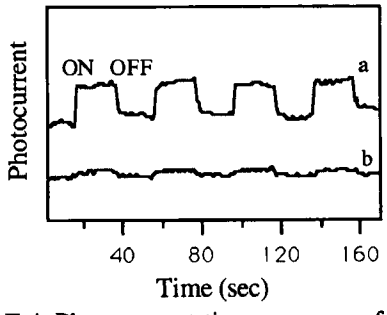

FIGURE.4. Photocurrent-time response of MIM device.(a : forward bias, b : reverse bias)

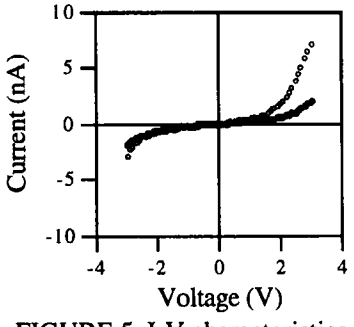

FIGURE.5. I-V characteristics for the MIM device.
(O : photocurrent , ● : dark current)

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