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Thesis outline

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This thesis is entitled "Study on Photoelectrocatalysis of Structured p/n Heterojunction Electrode" and it consists of 4 chapters.

Chapter 1 is "Introduction" and it describes the overview of the electronic properties of organic semiconductor materials (OSM), difference between organic heterojunction and inorganic heterojunction, p/n junction OSM photocatalyst and its mechanism, features and the challenge in p/n junction OSM photocatalyst, analysis technique for structured p/n junction and finally about the objectives of the study.

Chapter 2 is "Enhanced Oxidation Power as Photoelectrocatalysis Based on Micrometer-Localized Positive Potential in Terrace Hetero p/n Junction" and it describes about the enhancement of oxidation power in the photocatalyst based on the localized positive charge. In this chapter, terraced bilayer (TB) p/n heterojunction structure was fabricated and consisting of metal free phthalocyanine (H_2Pc, a p-type) and 3,4,9,10-perylenetetracarboxylic-bis-benzimidazole (PTCBI, an n-type) on the indium tin oxide (ITO) glass substrate. There are single layer region (SLR), bilayer region (BLR) and boundary region (BDR) in the TB structure. Scanning Kelvin probe microscopy (SKPM) was used to measure the contact potential difference (V_{CPD}) value of each region and their electronic properties was evaluated. By comparing the V_{CPD} data, the electron transfers would happen from PTCBI into ITO, from H_2Pc into ITO and from H_2Pc to PTCBI. In the case of H_2Pc on PTCBI, more positive potential was found at the boundary region than that of the bilayer region. The peak was indicated for localization of positive charge at the boundary region. Since the measured area only involve the surface of the electrode, then investigation for the bulk output is necessary. Hence, photoelectrochemical study was conducted for the electrode with having more boundary region, named as dot terrace bilayer electrode, where the area ratio: PTCBI area = 80 % and H_2Pc area =20 %. The dot terrace bilayer electrode exhibited more negative threshold potential (~ -0.2 V) for photoanodic current. The external quantum efficiency of the dot terrace bilayer electrode was eight times higher (at λ =590 nm) than that of the bilayer electrode. Moreover, as photocatalyst film (*i.e.* H_2Pc dot/PTCBI/PTFE [PTFE: Polytetrafluoroethylene] membrane filter), the dot terrace structure showed higher quantum efficiency (5.1%) than that of the bilayer (3.2%) for the decomposition of acetic acid. The present design and method was utilized for efficient photocatalyst by mitigating the potential loss from photon energy to redox powers without changing molecular component.

Chapter 3 is "Photoelectrochemistry of Thiol Oxidation with Bilayer and Mixture of Perylene Derivative/Phthalocyanine Heterojunction Electrodes" and it describes about photoelectrochemistry of thiol oxidation by charge transfer complex in bilayer and bulk heterojunction (BHJ) of PTCBI and H_2Pc electrodes. Bulk heterojunction system can facilitate charge separation due to large area of donor/acceptor interface compared to the bilayer system. Charge transfer complex is expected to form in the bulk heterojunction and could enhance the light absorption in the longer wavelength. It will be great merits to utilize natural sun light efficiency. In this study, I investigate the absorption spectrum and the photoelectrochemistry of thiol oxidation for the bilayer and co-deposited layer electrodes composed of metal-free Pc (H_2Pc) and PTCBI having electrode-electrolyte interface. The contribution of charge transfer complex towards the generation of photocurrent was investigated in the action spectrum data at the longer wavelength by comparing the action spectrum for monolayer electrode with that of the bilayer and co-deposited electrodes. Based on the analysis in their absorption spectra, a new absorption band in the longer wavelength (λ > 800 nm) for both bilayer and co-deposited photoelectrode suggested a formation of charge transfer complex. A photoanodic current was observed at λ ~ 880 nm for the both bilayer and co-deposited electrodes, while no absorption and photocurrent for single layers of PTCBI and H_2Pc. By assuming the Langmuir adsorption equilibrium at the solid/water interface, the kinetic parameters for the photoanodic current of thiol was analyzed for the longer wavelength of irradiation (λ ~ 900 nm), and it was indicated that the rate of oxidation in the co-deposited was higher than that of the bilayer due efficient charge separation in the charge transfer complex.

Finally, Chapter 4 is "Overall Summary" and it summarizes that the study about the micrometer p/n structure (as terrace bilayer) showed that the p/n boundary region can exhibit the potential characteristic differently based on their arrangement and the study about the potential at the boundary could be useful to understand the bulk heterojunction photocatalyst which has more varieties of p/n structure arrangement that affect its performance. The study about the p/n nanostructure photocatalyst showed that the bulk heterojunction photoelectrode can exhibit photoanodic current at the longer wavelength with the threshold of wavelength much wider into the infrared region than that the case of bilayer. The photoelectrocatalysis study of on those structured heterojunction electrodes emphasized the effect of the electronic properties towards the photocatalyst performance.