

Research Article

Characterization and Performance Evaluation of Dye Sensitized Solar Cell Using Nanostructured TiO₂ Electrode

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Metal-free organic sensitizer consisting of donor, electron conducting, and anchoring anhydride groups was engineered at molecular level and synthesized. Dye sensitized solar cells based on conjugated naphthalene dye were fabricated using nanoporous electrode. Photoelectrodes with a 7 μm thick nanoporous layer and a 5 μm thick light-scattering layer were used to fabricate dye sensitized solar cells. DSSCs were fabricated in a FTO/nc-TiO₂/organic dye/I⁻/I₃⁻/Pt/FTO device geometry. Dye sensitized solar cell was characterized by current density-voltage (*J-V*) measurement. All current-voltage (*I-V*) measurements were done under 100 mW/cm² light intensity and AM 1.5 conditions. The photovoltaic data revealed a short circuit photocurrent density of 1.86 mA/cm², an open circuit voltage of 430 mV, and a fill factor of 0.63, corresponding to an overall conversion efficiency of 0.53%.

1. Introduction

The demand for clean and environmentally friendly energy sources continues to increase due to depletion of conventional energy sources. One of the promising candidates of these new energy sources is solar energy, which is clean, renewable, and limitless. Dye sensitized solar cells have gained much more attention as one of the most promising energy harvesting devices since the report of ruthenium based photosensitizers in 1991 by O'Regan and Grätzel [1]. Ruthenium photosensitizers have shown high photoelectric conversion efficiencies of over 11% under AM 1.5 conditions [2]. Compared with them, metal-free organic dyes have advantages including lower cost, easier modification and purification, and environmental friendliness [1–3].

Generation of electrical energy from solar light is a long-term research interest and dye sensitized solar cells with a mesoporous TiO₂ have been regarded as one of the most promising candidates among these regenerative energy sources [4–12]. Although the research activities in this field are spanning the range from devising novel semiconductor electrodes and solid state hole conductors [13], particular importance is the development of well performing dyes. Organometallic and metal-free organic dyes are requested to

display broad light absorption capability, which can cover the whole solar light spectrum along with fast electron injection of electrons from photoexcited dyes to the conduction band (CB) of semiconductor electrodes and slow recombination between the injected electrons and resulting dye cations and/or I⁻/I₃⁻ redox couple. It is also crucial to design suitable binding groups as well as the spacer between the groups and dye core. Relative rate of electron injection is managed by the anchoring groups which are chemically adsorbed and attached to TiO₂ electrode. The controllability reaches cell performance including short circuit current (*J*_{sc}), open circuit voltage (*V*_{oc}), and overall conversion efficiency [14–22].

Thin films of organic dyes with conjugated π -electron systems are widely used in electronic and optoelectronic devices such as organic light-emitting diodes or solar cells. Hybrid organic/inorganic materials are promising candidates in photovoltaic (PV) cells. There are several basic requirements guiding the molecular engineering of an efficient sensitizer. The excited state should match the energy of the conduction band edge of the oxide. Light excitation should be associated with vectorial electron flow from the light harvesting moiety of the dye towards the surface of the semiconductor surface providing for efficient electron

transfer from the excited dye to the TiO₂ conduction band, a strong conjugation across the donor and anchoring groups, and good electronic coupling between the lowest unoccupied orbital (LUMO) of the dye and the TiO₂ conduction band. A major factor for the low conversion efficiency of many organic dyes in the DSSC is the formation of dye aggregates on the semiconductor surface. Such an aggregation phenomenon would affect the light absorption by filtering effect [23, 24]. Therefore, for obtaining optimal performance, aggregation of organic dyes needs to be avoided through appropriate structural modification [24, 25].

Anhydride groups are easily attached to TiO₂ electrode and it is very well known in the literature that these groups bound to TiO₂ surface over dicarboxy forms with the opening anhydride ring. Monoanhydride groups are immobilized into TiO₂ electrode to realize electron injection under illumination. Perylene monoanhydride derivatives are proven to present photovoltaic efficiencies in dye sensitized solar cells [4]. These results confirm the importance of anhydride structure.

In this paper, N,N'-diphenylbenzene-1,4-diamino-1,8-naphthalic anhydride (EE-1) has been synthesized to fabricate dye sensitized solar cells. E_{HOMO} and E_{LUMO} energy levels of conjugated naphthalene dye were calculated using cyclic voltammogram. E_{LUMO} level of EE-1 was found to be 3.23 eV and the E_{LUMO} level is suitable for efficient electron injection to conduction band of (E_{LUMO} (TiO₂) = 4.2 eV). EE-1 dye comprising anchoring groups can inject electrons to the conduction band of TiO₂ in organic dye sensitized solar cells. The photovoltaic data revealed a short circuit photocurrent density of 1.86 mA/cm², an open circuit voltage of 430 mV, and a fill factor of 0.63, corresponding to an overall conversion efficiency of 0.53%. Ground state optimization and HOMO and LUMO frontier orbitals of EE-1 were presented in Figure 1.

2. Experimental Section

2.1. Materials. Diphenylamine, 4-bromo-1,8-naphthalic anhydride, t-BuOK, and palladium acetate were purchased from Aldrich. P(t-Bu)₃ was supplied from Acros Organics. All solvents were of spectroscopic grade and were used without any further purification.

2.2. Materials Characterization. The UV-Vis absorption spectrum of synthesized dyes was recorded in an Analytic JENA S 600. Cyclic voltammetry measurements of synthesized dye were taken by using CH-Instrument 660 B Model Potentiostat equipment. Dye sensitized solar cells were characterized by current-voltage (*I-V*) measurement. All current-voltages (*I-V*) were done under 100 mW/cm² light intensity and AM 1.5 conditions. 450 W Xenon light source (Oriel) was used to give an irradiance of various intensities. *J-V* data collection was made using Keithley 2400 Source-Meter and LabView data acquisition software.

2.3. Procedure for Synthesis of N,N'-Diphenylbenzene-1,4-diamino-1,8-naphthalic Anhydride Derivative. N,N'-Diphenylbenzene-1,4-diamino-1,8-naphthalic anhydride is

synthesized according to the literature with some modifications [4]. The mixture of diphenylamine (1.6 g, 6.4 mmol) and 4-bromo-1,8-naphthalic anhydride (3.3 g, 12 mmol) in dry toluene (30 mL) was added to t-BuOK (1.6 g, 15 mmol), P(t-Bu)₃ (0.12 g, 0.5 mmol), and palladium acetate (0.056 g, 0.25 mmol). The resulting mixture was heated to 120°C for 28 h and then cooled to room temperature. Then water (40 mL) and ethyl acetate (40 mL) were added to the mixture. The organic layer was separated from the aqueous layer, washed successively with water and brine solution, and dried over anhydrous magnesium sulfate. The crude product was further separated by column chromatography (Silicagel, CH₂Cl₂/Hexane a red solid 74%).

N,N'-Diphenylbenzene-1,4-diamino-1,8-naphthalic anhydride. Yield: 74%. IR (KBr): cm⁻¹, 3051, 1761, 1708, 1580, 1564, 1504, 1485, 1458, 1391, 1334, 1305, 1262, 1221, 1147, 1167, 1114, 1078, 1018, 998, 898, 876, 852, 828, 772, 722, 743, 694.

¹H NMR (400 MHz, CDCl₃, TMS): δ 8.51 (dd, 2H), 8.48 (dd, 2H), 8.32 (dd, 2H), 7.6 (t, 2H), 7.34 (d, 2H), 7.28 (t, 2H), 7.08 (m, 4H), 7.03 (t, 2H), 6.97 (m, 4H), 6.93 (t, 2H).

2.4. Fabrication of Dye Sensitized Solar Cell. The dye sensitized solar cell device fabrication starts with cleaning of the fluorine doped tin oxide (FTO) coated glass substrates in acetone and isopropanol by using an ultrasonic bath. FTO (SnO₂: F, Pilkington TEC-15; R_{sheet} : 150/Ω), electrically conductive oxide-coated glass, was used as transparent electrode. TiO₂ electrode consists of an adsorbent mesoporous layer with 20 nm anatase TiO₂ particle size of 7 nm thickness and a second light scattering layer with 400 nm anatase TiO₂ particle size of 5 nm thickness. Preparation and characterization of double-layer TiO₂ electrode were previously described by Burfeindt et al. [25].

Dye solution was prepared in chlorobenzene (1:1) mixture with a concentration of 0.5 mM N,N'-diphenylbenzene-1,4-diamino-1,8-naphthalic anhydride (EE-1). TiO₂-coated electrode, after sintering at 450°C for 30 min and cooling to 100°C, was kept overnight in EE-1 solutions for adsorption. EE-1 adsorbed TiO₂-coated glass was washed with pure chlorobenzene and dried in vacuum. Platinized FTO glass was used as a counter electrode. Platinization of counter electrode was done by coating of FTO glass with 1% solution of hydrogen hexachloroplatinate (Aldrich) in 2-propanol and annealing at 400°C for 30 min. Cells were prepared in sandwich geometry. Surlyn-1702 (DuPont) frame was used as a spacer and a thermoplastic sealant between the two electrodes. Prepared cells were sealed by heating at 100°C. Electrolyte was filled into the space created by Surlyn-1702 between the electrodes under vacuum using a small hole predrilled on the counter electrode with the help of a diamond drill. After filling the electrolyte, a small hole was sealed again using a piece of Surlyn-1702 and a piece of cover glass. The electrolyte consisted of 0.6 M N-methyl-N-butyl imidazolium iodide (BMII) + 0.1 M LiI + 0.05 M I₂ + 0.5 M 4-tert-butyl pyridine (TBP) in acetonitrile. Active areas of the cells were adjusted to 0.159 cm² with a special mask.

Dye sensitized solar cells were characterized by current-voltage (*I-V*) measurement. All current-voltages (*I-V*) were

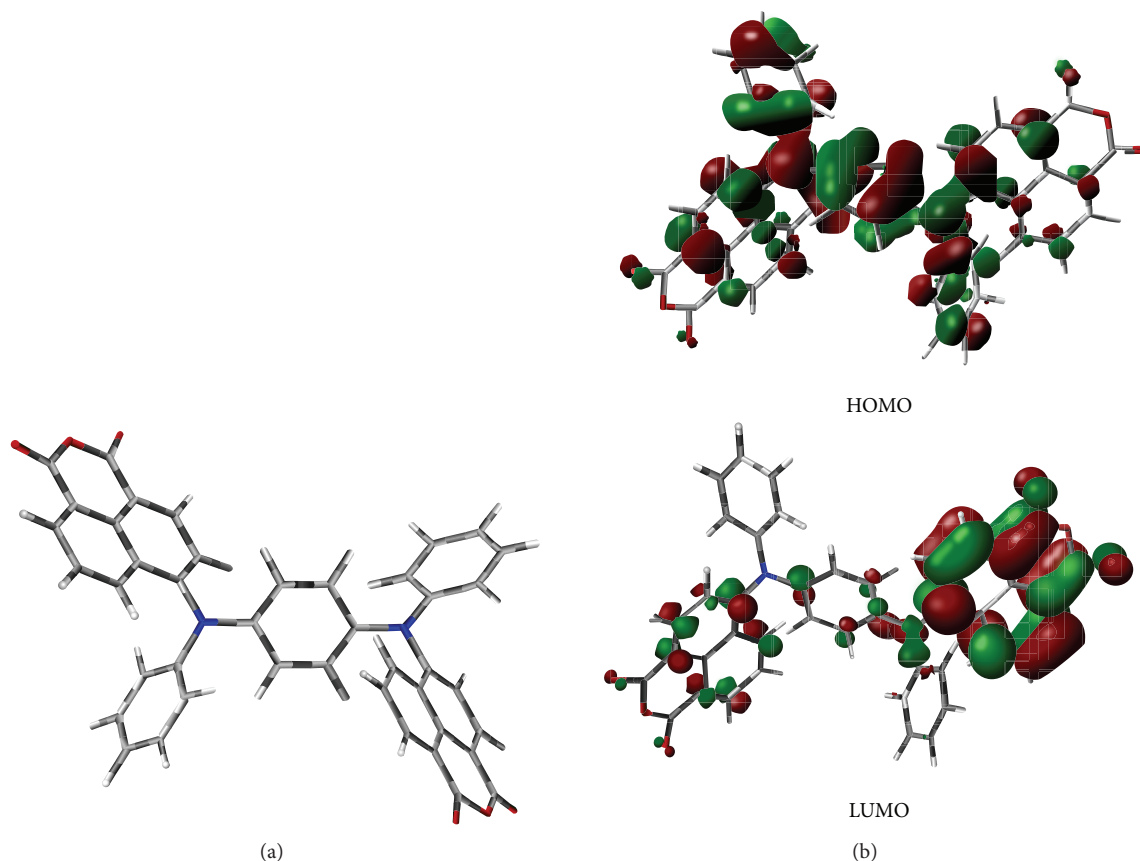


FIGURE 1: Ground state optimization of EE-1 (a) and HOMO and LUMO frontier orbital of EE-1 (b).

done under 100 mW/cm^2 light intensity and AM 1.5 conditions. 450 W Xenon light source (Oriol) was used to give an irradiance of various intensities.

The spectral output of the lamp was matched in the region of 350–700 nm with the aid of Schott K113 Tempax sunlight filter. I - V data collection was made by using Keithley 2400 Source-Meter and LabView data acquisition software. I - V characteristics of dye sensitized solar cell in dark and under illumination are shown in Figure 5.

3. Results and Discussion

3.1. Absorption Spectrum of N,N' -Diphenylbenzene-1,4-diamino-1,8-naphthalic Anhydride Derivative. The main importance of organic dye in dye sensitized solar cell is to make absorption in visible region. 1,8-Naphthalic anhydride group is only absorbed in the UV region. If diphenyl amine group is condensed onto naphthalic anhydride group from 4 positions, a pronounced red shift is observed in the absorption spectrum because of bathochromic effect. Absorption is observed at 500 nm region and shifted up to 620 nm in Figure 1 for N,N' -diphenylbenzene-1,4-diamino-1,8-naphthalic anhydride. N,N' -Diphenyl benzene-1,4-diamino-1,8-naphthalic anhydride is absorbed in the visible

region and is a promising material for dye sensitized solar cell application.

3.2. E_{HOMO} and E_{LUMO} Values of N,N' -Diphenylbenzene-1,4-diamino-1,8-naphthalic Anhydride Derivative. E_{HOMO} and E_{LUMO} values of N,N' -diphenylbenzene-1,4-diamino-1,8-naphthalic anhydride (EE-1) were calculated by using cyclic voltammograms (Figure 3). Solutions of EE-1 derivative were prepared in chloroform (10^{-3} M). A three-electrode cell was used consisting of glassy carbon working electrode, Pt wire counter electrode, and Ag/AgCl reference electrode, all placed in a glass vessel. Tetrabutylammonium hexafluorophosphate (TBAPF_6), 0.1 M, was used as supporting electrolyte. Ferrocene was used as internal reference electrode. EE-1 derivative shows both reversible reduction and oxidation potential in Figure 2. In calculation, the zero vacuum level of ferrocene was taken as 4.8 eV [19]. We used the following equation to calculate E_{HOMO} and E_{LUMO} energy levels of EE-1: $E_{\text{LUMO}} = -e(E_{1/2(\text{redox})} - E_{\text{fer}^-}) + 4.8$ [19]. E_{HOMO} and E_{LUMO} energy levels of EE-1 derivative were 5.06 eV and 3.23 eV, respectively. E_{HOMO} and E_{LUMO} values of EE-1 are presented in Table 1. E_{LUMO} value of EE-1 is higher than the conduction band of TiO_2 , $E_{\text{CB}} \text{ TiO}_2 = 4.2 \text{ eV}$. Schematic energy diagram of the device fabricated using EE-1 dye was

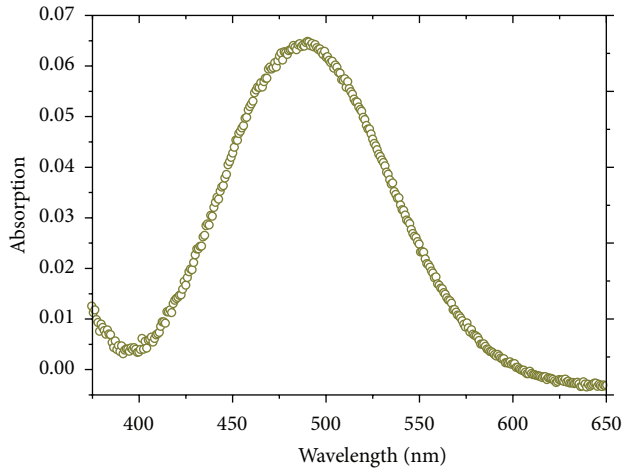


FIGURE 2: Absorption spectrum of N,N' -diphenylbenzene-1,4-diamino-1,8-naphthalic anhydride derivative.

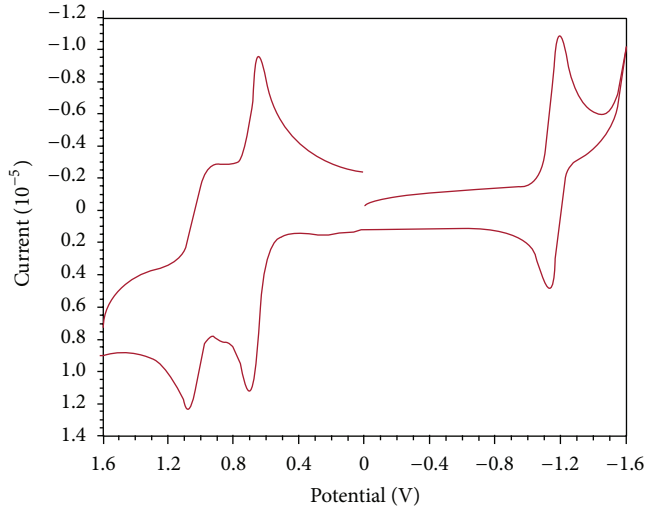


FIGURE 3: Cyclic voltammogram of N,N' -diphenylbenzene-1,4-diamino-1,8-naphthalic anhydride (EE-1).

TABLE 1: E_{LUMO} and E_{HOMO} energy levels of synthesized dye.

	E_{red}	E_{ox}	E_{HOMO}	E_{LUMO}
EE-1	-1.18	0.64	5.06	3.22

$E_{ferrocene} = 0.39$ V (ferrocene is internal reference electrode).

E_{red} = reduction potentials of synthesized derivative.

E_{ox} = oxidation potentials of synthesized derivative.

TABLE 2: Photovoltaic performances of DSSC.

	J_{sc} (mAcm^{-2})	V_{oc} (mV)	FF	η (%)
EE-1	1.86	430	0.63	0.53

shown in Figure 4. EE-1 derivative was absorbed in the visible region and EE-1 dye can inject the electrons to the conduction band of TiO_2 . EE-1 is promising material in the field of DSSC application. Schematic diagram was presented in Figure 4.

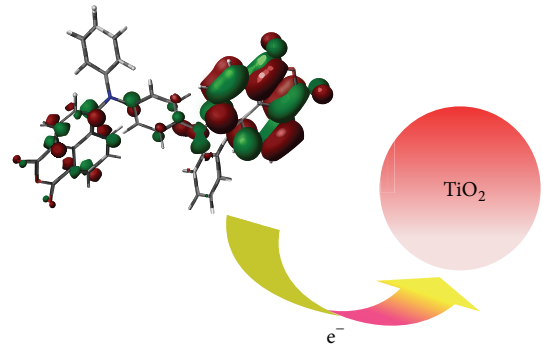


FIGURE 4: Schematic diagram of electron transfer from dye to TiO_2 .

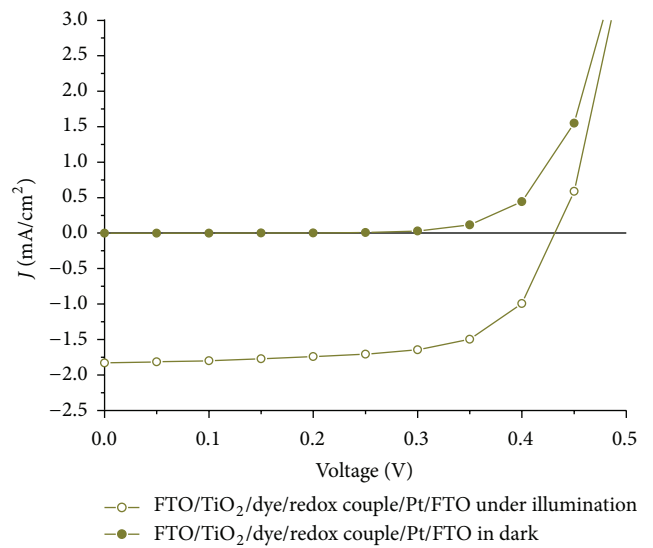


FIGURE 5: I - V curve of the device "FTO/ nc-TiO_2 /conjugated naphthalene dye (EE-1)/ Γ^-/I_3^- /Pt/FTO."

3.3. Photovoltaic Characterization. The current-voltage curve for DSSC based on conjugated naphthalene dye is shown in Figure 5 and Table 2. Under standard global AM 1.5 solar irradiation, the TiO_2 solar cell sensitized with EE-1 gave a short circuit current density (J_{sc}) of 1.86 mA cm^{-2} , open circuit voltage (V_{oc}) of 430 mV, and a fill factor of 0.63, corresponding to an overall conversion efficiency (η) of 0.53%.

The entire energy conversion efficiency, η , is calculated by means of the following equations:

$$\eta = \frac{V_{oc} I_{sc} FF}{P_{light}} \quad (1)$$

Here, V_{oc} is open circuit voltage (V), I_{sc} is short circuit current (mA/cm^2), and FF is fill factor and P_{light} (mW/cm^2):

$$FF = \frac{V_{max} I_{max}}{V_{oc} I_{sc}} \quad (2)$$

where V_{\max} and I_{\max} are voltage and current at the point of maximum power output of cell.

Binding of the organic dye onto the TiO_2 surface can take place via anhydride groups. According to the reports in the literature, six different adsorption modes can be seen between dye and metal oxide surfaces: (i) covalent bonding of dye onto the semiconductor surface, (ii) electrostatic interactions, (iii) hydrogen bonding, (iv) self-assembly, (v) physisorption of dye on the surface via van der Waals interactions, and (vi) physical entrapment [26, 27]. For the EE-1 dye, covalent binding occurs between anhydride anchoring group and semiconductor surface would be more dominant compared to the others. And in the literature, it is proven that anhydride groups are attached to TiO_2 surface in the dicarboxylic form. Carboxylic groups are the most important anchoring groups to attach to the surface of titanium dioxide. Anhydride groups are attached to titanium dioxide easily. In this context, the covalent binding taking place at the dye-semiconductor interface is very important. Theoretical calculations also showed that, in the LUMO level, electron densities were localised on the anhydride group. As a result of this effect, the dye binding onto the TiO_2 surface can contribute to electron injection efficiently, and this leads to electron injection yield for TiO_2 based DSSC.

4. Conclusion

We have molecularly engineered a novel type of organic sensitizer. By increasing the π -conjugation of the dye, the E_{HOMO} and E_{LUMO} energy levels were tuned, which was further supported by electrochemistry. Theoretical calculations showed that electron densities were localised on the anhydride group. When the dye excited to the LUMO level, efficient electron transfer will be to the conduction band of TiO_2 . Conjugated organic dye, which has aryl amine moiety as the electron donor and anhydride moiety as electron acceptor, showed good performance as photosensitizers in DSSCs. We obtained an overall conversion efficiency of 0.53% under AM 1.5 irradiation (100 mW/cm^2) with a DSSC based on EE-1 (short circuit photocurrent density of 1.86 mA/cm^2 , an open circuit voltage of 430 mV, and a fill factor of 0.63).

Conflict of Interests

The author declares that there is no conflict of interests regarding the publication of this paper.


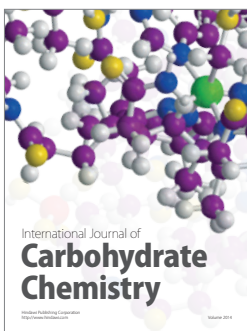
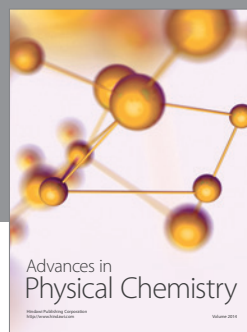
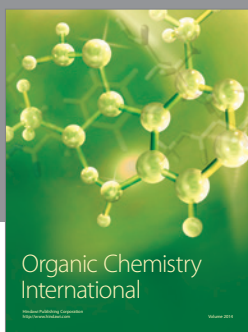
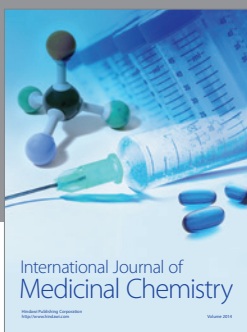
Acknowledgments

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