



Performance of zinc chlorophyll based molecules for dye sensitized solar cell



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ABSTRACT

In this study, we investigated the influence of long carbon chains in ester group of the zinc chlorophyll derivatives, zinc hexyl 3-devinyl-3-hydroxymethyl-pyropheophorbide-*a* (ZnChl-C6), zinc octadecyl 3-devinyl-3-hydroxymethyl-pyropheophorbide-*a* (ZnChl-C18), zinc dodecyl 3-devinyl-3-hydroxymethyl-pyropheophorbide-*a* (ZnChl-C12) and zinc methyl 3-devinyl-3-methylhydroxymethylpyropheophorbide-*a* (ZnChl-OH), on the DSSC performance. Photovoltaic performances were determined in the order of ZnChlC12 > ZnChlC18 > ZnChlC6 > ZnChlOH from *J*–*V* curves. The higher *J*_{sc} obtained from the ZnChl-C12 sensitizer could result from the presence of long alkyl chain which reduce the charge recombination rate and facilitate electron injection. However, it was observed to decrease in the DSSC performance with the presence of longer alkyl chains (>C12). It was understood from the obtained results, the length of the alkyl chains is directly affect the DSSC performance.

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1. Introduction

The research on dye sensitized solar cells (DSSCs) have quickly attracted more attention, due to their low cost production and energy conversion efficiencies compared to silicon based solar cells, after publishing the remarkable paper in 1991 by Michael Grätzel [1,2]. In dye sensitized solar cells, a dye sensitizer plays the key role in light harvesting and energy conversion [3]. Key aspects for DSSCs reside in increasing the photon collection efficiency over a broad spectrum of wavelengths in the visible regime and in retarding the charge recombination process [4–7]. The mechanism of DSSCs resembles photosynthesis. In the fascinating process of photosynthesis, water and carbon dioxide are converted into oxygen and sugar by the assistance of sunlight. In both photosynthesis and in DSSCs dyes are utilized to capture light energy. Chlorophylls are amazing compounds as a photosensitizer in visible region. They play an important role not only for light harvesting and also energy and electron transfer processes in natural photosynthesis [8–22].

Chlorophyll derivatives, which absorb at the near infrared region of solar energy have always led to high current generation and act as a driving force in developing chlorophyll sensitizers in DSSCs [22–26].

In this study, we aim to investigate the influence of long carbon chains in ester group of the zinc chlorophyll derivatives on the DSSC performance. To the best of our knowledge, there is no information regarding on this issue for zinc chlorophyll derivatives in the literature up to now.

2. Experimental section

All materials were reagent grade and were used as received unless otherwise noted.

2.1. Materials characterization

The UV–Vis absorption spectra of synthesized dyes were recorded in a 1 cm path length quartz cell by using Analytic JENA S 600. ¹H NMR spectra were measured on a Bruker 400 MHz spectrometer. The mass spectra were obtained via electrospray ionization (ESI-MS), and matrix-assisted laser desorption/ionization

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(MALDI). 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 (J – V) measurement. All current–voltage (J – 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 by using Keithley 2400 Source-Meter and LabView data acquisition software.

2.2. Synthesis and chemical characterization of zinc chlorophyll-*a* derivatives

Anhydrous solvents were either distilled from appropriate drying agents or purchased from Merck and degassed prior to use by purging with dry argon and kept over molecular sieves. All chemicals were purchased from commercial sources, and used as received. Synthetic details and characterization of the compounds are described below. Molecular structures of the complexes were shown in Fig. 1.

2.2.1. Synthesis of zinc hexyl 3-devinyl-3-hydroxymethylpyropheophorbide-*a* (ZnChl-C6)

ZnChl-C6 was synthesized according to the procedures reported in the literature [27,9]. UV–VIS (MeOH) λ_{\max} 652 (0.835), 605 (0.154), 575 (0.080), 528 (0.028), 426 (0.920), 407 (0.721), 324 (0.320). ¹H NMR (DMSO) 9.58, 9.34, 8.46 (each 1H, s, 5, 10, 20-H), 5.61 (2H, d, J = 5 Hz, 3-CH₂), 5.54 (1H, t, J = 5.5 Hz, 3¹-OH), 5.09, 5.01 (each 1H, d, J = 19.5 Hz, 13¹-CH₂), 4.54–4.48 (1H, m, 18-H), 4.27 (1H, m, 17-H), 3.95–3.90, 3.87–3.83 (each 1H, m, 17²-COOCH₂), 3.76 (2H, q, J = 7 Hz, 8-CH₂), 3.58 (3H, s, 12-CH₃), 3.26 (3H, s, 2-CH₃), 3.24 (3H, s, 7-CH₃), 2.61–2.52, 2.25–2.17 (each 2H, m, 17-CH₂CH₂), 1.74 (3H, d, J = 7.5 Hz, 18-CH₃), 1.66 (3H, t, J = 7.5 Hz, 8¹-CH₃), 1.19–1.15 (8H, m, 17²-COOC(CH₂)₄), 0.79 (3H, t, J = 7 Hz, 17²-COOC₅CH₃). MS (ESI) found: m/z 684.3. Calcd. for C₃₈H₄₄N₄O₄Zn: M⁺, 684.3.

2.2.2. Synthesis of zinc dodecyl 3-devinyl-3-hydroxymethylpyropheophorbide-*a* (ZnChl-C12)

ZnChl-C12 was synthesized according to the procedures reported in the literature [27,9]. UV–VIS (MeOH) λ_{\max} 649 (0.606), 602 (0.116), 554 (0.061), 513 (0.050), 423 (0.957), 403 (0.611), 313 (0.221). ¹H NMR (DMSO) 9.59, 9.36, 8.46 (each 1H, s, 5, 15, 20-H), 5.62 (2H, s, 3-CH₂), 5.59 (1H, br-s, 3¹-OH), 5.11, 5.02 (each 1H, d, J = 19.5 Hz, 13¹-CH₂), 4.51 (1H, m, 18-H), 4.25 (1H, m, 17-H), 3.92, 3.84 (each 1H, m, 17²-COOCH₂), 3.77 (2H, q, J = 7.5 Hz, 8-CH₂), 3.59 (3H, s, 12-CH₃), 3.27 (3H, s, 2-CH₃), 3.25 (3H, s, 7-CH₃), 2.64–2.53, 2.27–2.18, (each 2H, m, 17-CH₂CH₂), 1.75 (3H, d, J = 7.5 Hz, 18-CH₃),

1.67 (3H, t, J = 7.5 Hz, 8¹-CH₃), 1.36 (2H, m, 17²-COOCH₂), 1.22–1.04 (18H, m, 17²-COOC₂(CH₂)₈), 0.80 (3H, t, J = 7 Hz, 17²-OOC₁₀CH₃). MS (MALDI) found: m/z 768.4. Calcd. for C₄₄H₅₆N₄O₄Zn: M⁺, 768.4.

2.2.3. Synthesis of zinc octadecyl 3-devinyl-3-hydroxymethylpyropheophorbide-*a* (ZnChl-C18)

ZnChl-C18 was synthesized according to the procedures reported in the literature [27,9]. UV–VIS (MeOH) λ_{\max} 652 (0.623), 607 (0.112), 575 (0.057), 528 (0.018), 426 (0.676), 408 (0.524), 324 (0.222). ¹H NMR (DMSO) 9.57, 9.34, 8.45 (each 1H, s, 5, 10, 20-H), 5.61 (2H, s, 3-CH₂), 5.54 (1H, br-s, 3¹-OH), 5.09, 5.00 (each 1H, d, J = 19.5 Hz, 13¹-CH₂), 4.55–4.46 (1H, m, 18-H), 4.27–4.21 (1H, m, 17-H), 3.94–3.3.88, 3.86–3.79 (each 1H, m, 17²-COOCH₂), 3.75 (2H, q, J = 7 Hz, 8-CH₂), 3.57 (3H, s, 12-CH₃), 3.25 (3H, s, 2-CH₃), 3.24 (3H, s, 7-CH₃), 2.65–2.50, 2.30–2.15, (2H, 1H, m, 17-CH₂CH₂), 1.74 (3H, d, J = 7 Hz, 18-CH₃), 1.66 (3H, t, J = 7.5 Hz, 8¹-CH₃), 1.30–1.04 (32H, m, 17²-COOC₂(CH₂)₁₆), 0.82 (3H, t, J = 7 Hz, 17²-COOC₁₇CH₃). MS (MALDI) found: m/z 852. Calcd. for C₅₀H₆₈N₄O₄Zn: M⁺, 852.

2.2.4. Synthesis of zinc methyl 3-devinyl-3-methylhydroxymethylpyropheophorbide-*a* (ZnChl-OH) (3¹R/S = 1/1) (ZnChl-OH)

Zinc methyl 3-devinyl-3-methylhydroxymethylpyropheophorbide-*a* (ZnChl-OH) (3¹R/S) was synthesized according to the slightly modified procedure [22]. Methyl pyropheophorbide-*a* (106 mg, 0.19 mmol) was dissolved in 30% hydrogen bromide in acetic acid (40 ml) and was heated at 45 °C under argon atmosphere for 4 h. Subsequently, solution was poured into ice-water, extracted with CH₂Cl₂, twice washed with H₂O, dried over Na₂SO₄ and concentrated *in vacuo*. After evaporation residue was dissolved in MeOH (50 ml), 5 ml of H₂SO₄(conc) was added dropwise to the solution at 0 °C and left stirring overnight at ambient temperature. Afterwards, solution was poured into ice-water, extracted with CH₂Cl₂ twice washed with H₂O, dried over Na₂SO₄ and concentrated *in vacuo*. Methyl 3-devinyl-3-methylhydroxymethylpyropheophorbide-*a* (3¹R/S) was obtained with yield 49.6 mg (54%) after purification by column chromatography (0.2% MeOH/CH₂Cl₂) and recrystallization from CH₂Cl₂/MeOH and was used for the next step.

Methyl 3-devinyl-3-methylhydroxymethylpyropheophorbide-*a* (3¹R/S, 49.6 mg, 87.6 μmol) was dissolved in the mixture of MeOH (3 ml) and CH₂Cl₂ (12 ml). Subsequently, Zn(OAc)₂ (anhydrous) (19.3, mg, 87.6 μmol) was added to the resulted solution and mixture was refluxed for 3.5 h. After reaction completion, 4% NaHCO₃(aq) was added and stirred for additional 15 min. Reaction mixture then was washed with water, dried over Na₂SO₄ and

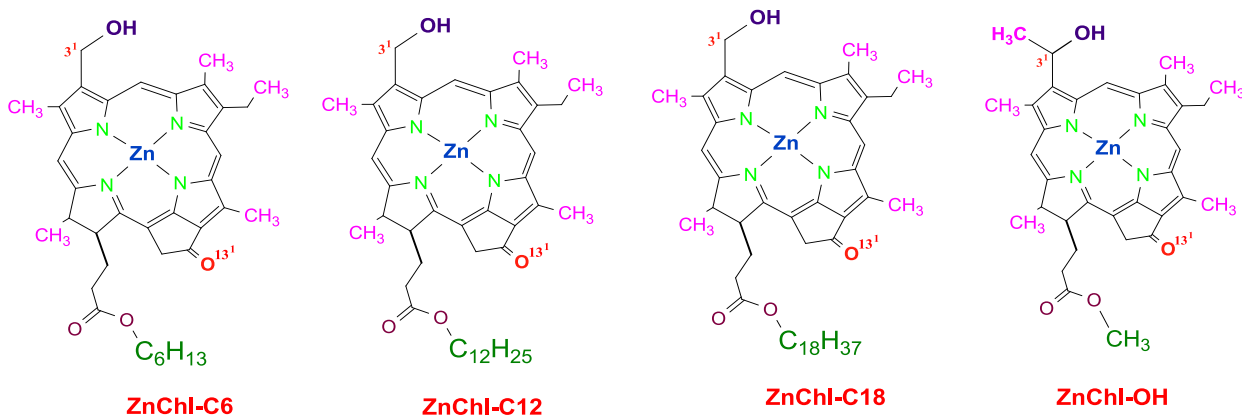


Fig. 1. Molecular structures of the zinc chlorophyll derivatives.

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