Chem. Notes 2020, 1(1), 69-81

The prospect of liquid-based dye sensitized solar cells (DSSCs): A Comparative Review

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Article Received: 26 June 2020

Article Accepted: 30 June 2020

Abstract

Energy demands including electricity consumptions have been increasing rapidly in recent years. Silicon solar cells, the most highly used cells dominating global markets, contribute significantly in providing sustainable form of energy. More importantly, it promisingly enables people especially in rural and remote areas to have access to electricity that could potentially stimulate rural development. However, this energy platform still suffers for high production and maintenance costs and the use of hazardous materials. Soft interface solar cells seems to take advantage over silicon solar cell design but review articles explaining the advantages of this new approach are rarely found. This article provides a general overview about an emerging solar cell based on liquid│liquid junction interface from chemistry perspective, highlights its benefits over silicon based solar cells and argues that this technology is a potential substitute for the existing ones by focusing specifically on the issues of costs, environmental impacts, electron transfer and fabrication techniques.

Keywords: liquid│liquid interface, DSSC, solar cell, renewable energy

Introduction

Electricity (mostly using fossil fuels as the generating power) is inevitably important for humankind. Lack of electricity in developing countries including Indonesia has become one of the significant challenges that limit rapid development especially in rural and remote areas. In the broader context, the challenge of non-renewable form of energy source depleting significantly as a consequence of high consumption requires serious responses. In addition, the negative impacts of this carbon based form of energy to the environment leading to global warming and climate change need to be taken into account in providing solutions to overcome or at least reduce its alarming effects on our society.

Silicon-based solar cells have been proposed by the Indonesian government to provide an adequate supply of green electricity in remote areas in this archipelago country[1]. It is an ideal approach but the issues related to production and maintenance costs require viable solutions. Research about this type of device has been extended to the development of DSSC platforms using dyes attached to solid surfaces of semiconducting materials[2]. The

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efficiency of these solid-based DSSCs are approaching the theoretical maximum estimations[3] but are suffering for some practical reasons such as cost for producing and maintaining the device and the use of harmful materials[4]. This paper will review current research work on liquid-based DSSCs in comparison to silicon based technology and argue that this could be a suitable approach to substitute existing cells in the near future owing to their cost-effectiveness, environmental benefits, high rate of electron transfers and simple fabrication.

Basic Principles of Liquid-Based DSSCs

Dye-sensitized solar cells (DSSCs) have been well reviewed by many scholars[2,5]. As shown in Figure 1, it take advantage of molecular materials as photosensitizer (S) to harvest sunlight in the form of photo-generated excited states (S*), which subsequently inject charges into alternative semiconducting materials such as TiO_2. The excitation from the valence band to the conduction band of semiconductors allows the electron to be collected at the electrode which then flows to the counter electrodes and with the aid of the electrolyte (I^-/I_3^-) undergoing reduction and oxidation, the excited dye can be re-oxidised back to its stable form. This cyclic photoelectrochemical reaction is one of the key factors that enable continuous flow of electrons to ensure stable electricity generation. DSSCs have advantages over silicon solar cells in terms of manufacturing and being feasible to assemble through reel-to-reel printing methods on flexible substrates. However it suffers from challenges associated with the contact and connection of the dye due to trapping and recombination issues to the mesoporous semiconductor to ensure efficient charge injection and transfer through the device, and the use of corrosive redox mediators (I^-/I_3^-)[6].

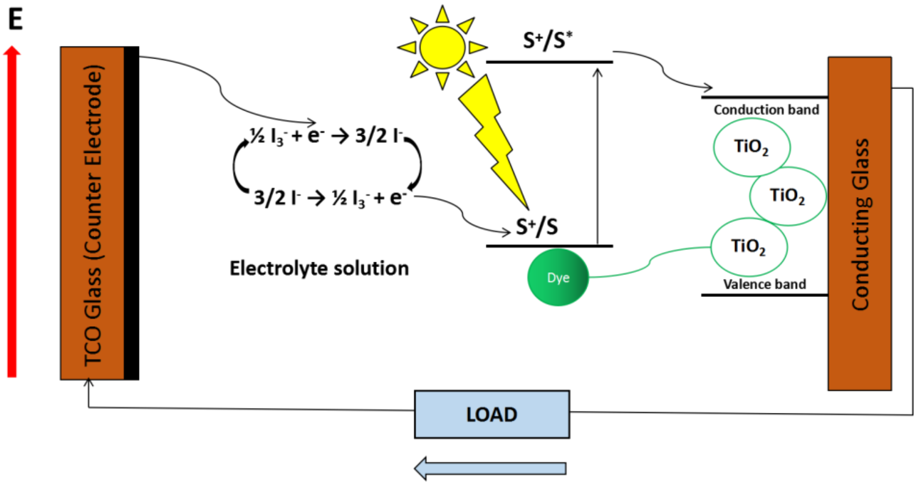

Figure 1. A schematic representation of a dye-sensitised solar cell (adopted with modification from (7))

Liquid-based dye sensitized solar cells, however, offer a new concept in DSSC design that alleviates the need for complex solid-state structures and avoids or reduce the issues of charge traps and charge injection barriers at dye-mesoporous interfaces[8]. The use of a soft liquid|liquid interface formed between water and organic liquids such as 1,2-dichloroethane[9-11] or α, α, α -trifluorotoluene ($C_6H_5CF_3$)[12] allows the two redox 'half cells' to be separated by a simple liquid|liquid junction formed between two immiscible liquids where dye molecules take place to perform light harvesting processes (Figure 2). The resulting interfaces are defect-free and self-healing, allowing solar cell to be manufactured in a simple, robust device that offers all of the advantages of the DSSC platform without the inherent of physical, chemical and engineering challenges of the dye-semiconductor interface while oil phase act to dissolve complex compounds as redox mediators such as acetylferrocene and 1,1 dimethylferrocene[9,13], in order to regenerate the excited states of the photosensitizer back to its stable form.

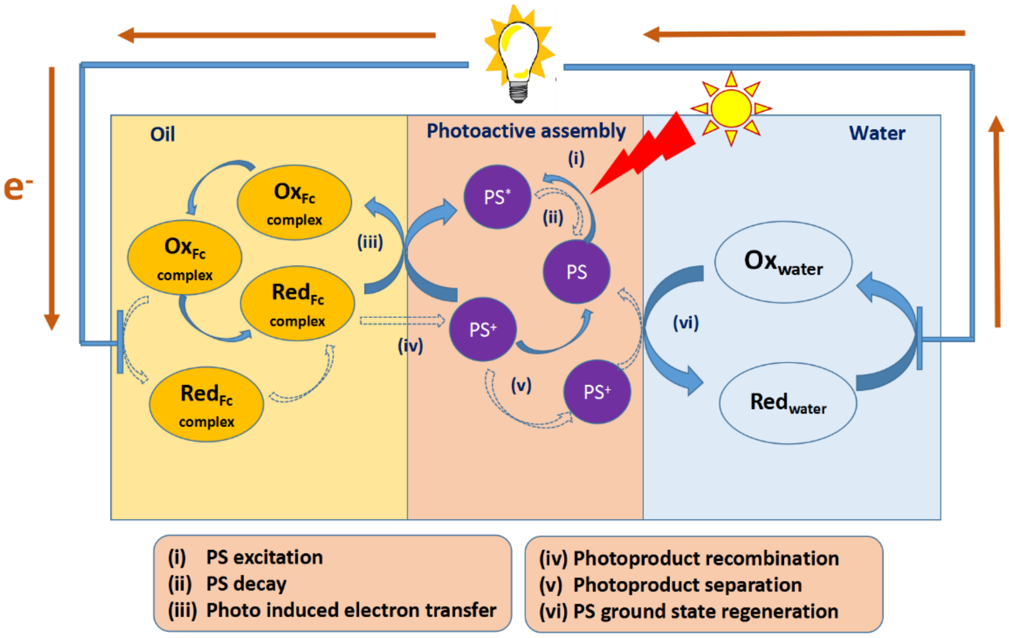

Figure 2. General scheme of liquid|liquid junction solar cell illustrating the elementary steps (PS = dye photosensitizer, Fc = ferrocene). Adopted with modification from (8) and (14)

There are three main parts of the liquid|liquid solar cell system (Figure 2). The middle part (pink) is the interface where confined photosensitizers are situated. It is an ideal environment in which to assemble free-standing films of semi-conducting metal oxides, metal-organic frameworks and nanosheets such as metal bipyridiyl complexes and porphyrins[15-17] to perform light harvesting process (Figure 3). In the oil system (yellow), hydrophobic redox mediators with, in principle, low propensity to cross the liquid|liquid interface plays a critical role together with water system (blue) undergoing redox processes to complete the cyclic mechanism in generating electricity.

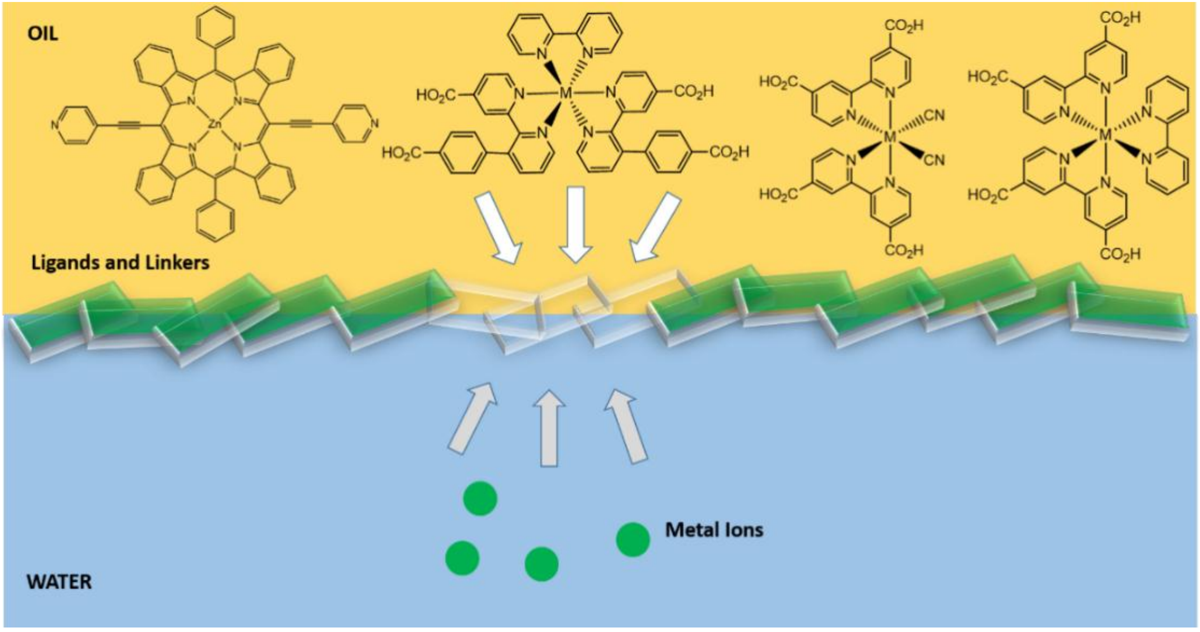

Figure 3. Model of the interfacial assembly of a metal-organic framework at a liquid|liquid interface, and illustrative molecular structures of linkers and ligands (adopted with modification from (15).

Discussions

Cost effectiveness

Liquid-based DSSCs have fewer manufacturing processes that could lead to lower production costs. Even though solid-state silicon based solar cells dominate 93 per cent of the global photovoltaic (PV) markets due to their high efficiencies[5], the existing technology has numerous disadvantages. There are several cost ineffective techniques that have been employed to break Si-O bonds in SiO_2 to produce high purity of silicon for solar cells and other microelectronic material purposes (Figure 4). The first technique is a process that contains two main steps. Initially, metallurgical grade silicon (MG-Si) is reacted with hydrochloric acid to form chlorosilanes including trichlorosilane (TCS) after distilled at 31.8°C. Finally, poly-silicon is obtained by using pyrolysis approach at 1150°C in Bell or Siemens reactors[18].

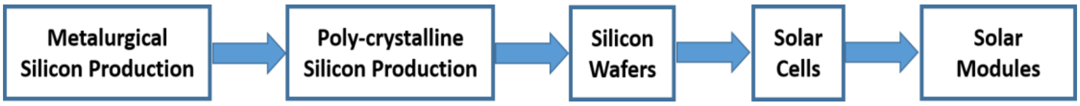

Figure 4. Supply chain of PV production in solar cell technology

Czochralski (CZ) method is another technique that is normally used to produce single crystalline silicon. The process using this CZ procedure similarly requires high temperature maintenance for a wide range of time. These two approaches are apparently capital extensive and consume high amount of energy. Another technique is Bridgman process which results multi-crystalline instead of single crystalline silicon. As a consequence, electron recombination leading to the decrease in conversion efficiency become a concern of this process. Besides, both CZ and Bridgman methods have drawbacks in terms of silicon wafer production which have to be cut from silicon blocks causing 50% silicon material lost as dust during the processes and so requires additional significant cost for recycling the dust. Carella and co-workers[5] also agree that silicon materials have to be highly purified in order to be successfully doped during cell production. They emphasize that purification procedures involving extreme temperatures and vacuum conditions typically require long and complicated fabrication techniques. Thus, advanced and time-consuming approaches may result in high production costs that lead to an increase in cell device prices.

In contrast, liquid based DSSCs use silicon-free materials without inheriting high-cost manufacturing processes associated with the traditional technology. This is because liquid materials used in the proposed design, in principle, can be straightforwardly mixed to form an interface between two immiscible liquid-liquid junctions due to their different solubilities. Samec, Eugster, Fermiín, and Girault[19], for instance, report that by employing water│1,2-dichloroethane as the liquid-liquid junction with porphyrins as selected dyes, the photo electrochemical reactions are likely to occur indicated by rapid regeneration of porphyrins as photosensitisers. They also highlight that the surface concentration of the photosensitiser shows constant conditions for various experimental systems both for shorter to longer illumination times and lower to higher photon intensities. The experimental results also fit well with a computational model designed to confirm the validity of the chemical reaction mechanism occurring at this type of soft interface. In addition, Scanlon and co-workers currently studied nanostructured arrangement of zinc(II) *meso*-tetrakis(4-carboxyphenyl) porphyrin assembled at interface between water and organic liquid for light harvesting. They found that the formation of nanostructure seems to uniquely occur with regards to the formation of hydrogen bonding as a bridging template connecting with the adsorbed porphyrin moieties. The hydrophobicity nature of neutral protonated and tetra-anionic protonated types allows an independent self-assembly process[20]. It is important to note that this cutting edge research significantly improves the formation of porphyrin nanostructure and therefore avoids complicated experimental designs required for previous research on the topic such as acidic pH control to avoid metal ion expulsion[21], complex amphiphilic porphyrin molecule design[22], and additive introduction that normally leads to uncontrollable route of reaction[23]. The interface formation occurs independently according to their polarities without outside forces is promising when it comes to the applications as it avoids extended and complex fabrication methods associated with classical solar cells. Thus, it would be affordable, for people to have access to these potentially cheaper device.

Environmental Benefits

Liquid based DSSC technology uses environmentally friendly materials. It is claimed that silicon PV technologies are more viable than other carbon based fuels or other means of green energy technologies in producing electricity. This argument is based on the calculation by using Levelized Cost of Electricity (LCOE) method[24]. However, manufacturing silicon solar panel is similar to other advanced technology production which only differs in types of toxic waste generated such as poisonous gases, reactive chemical solutions and heavy metals. Especially when producing in larger scales, enormous quantities of harmful materials are released to the environment[25]. For instance, silicon material production for solar cell application is highly dependent on fossil fuels, the significant contributor to greenhouse gas emissions and global warming, as generating power to run the manufacturing processes. The initial step of the whole process of PV manufacture alone involves pure silicon metal production obtained by reducing quartz using a carbothermic process in smelters with around 45 MW power of electricity generated mainly using coal, oil coke, charcoal and woodchips[26-27]. The study shows that the combustion processes to generate electricity for producing each ton of metallurgical silicon (Mg-Si) using this carbon based fuels releases about 5-6 tons of CO$_2$. This excludes gaseous waste resulted during gathering, and transporting the raw materials required to the smelting centers which may release even more toxic gases polluting the environment (Table 1). Megatons of carbon monoxide and carbon dioxide have been emitted even more in total for electricity generation for the rest of solar panel production including polysilicon, ignots, solar cell dan PV module productions.

Table 1. The raw materials for production of each ton of mg-Si (Kato et al and Globe) and annual rate (Thorsil) from (27)

Raw material	Kato et.al	Globe	Thorsil
Quartz	2.4 t	2.8 t	310,000 tpy
Coal	550 kg	1,4 t	195,000 tpy
Oil coke	200 kg	-	
Charcoal	600 kg	-	
Woodchip	300 kg	2.4t	185,000 tpy

Moreover, Mulvaney[28] reports that during silicon-based device production processes, quartz, the raw material containing silicon metals, needs to be transformed into a purely structured polysilicon. The targeted product is obtained by adding hydrochloric acid to the raw materials to produce trichlorosilanes. It is then reacted with hydrogen to result in desired polysilicon and liquid silicon tetrachloride ($SiCl_4$) as a by-product. $SiCl_4$ compounds can re-produce hydrochloric acid if exposed to water used in washing and cooling systems which cause water acidification and toxic fume emissions. For example, waste generated from solar panel productions by Jinko Solar Holding Company polluted water in a nearby river ecosystem[28-29]. They also mention that each kilogram of polysilicon product obtained generates three to four kilograms of silicon tetrachloride as waste with schematic reactions as shown below.

$$5(MG\text{-})Si + 16\,HCl \longrightarrow 4\,HSiCl_3 + 6H_2 + SiCl_4$$
$$2\,SiHCl_3 \longrightarrow SiH_2Cl_2 + SiCl_4$$
$$2\,SiH_2Cl_2 \longrightarrow SiHCl_3 + SiH_3Cl$$
$$2\,SiH_3Cl \longrightarrow SiH_2Cl_2 + SiH_4$$
$$HSiCl_3 + H_2 \longrightarrow Si + 3HCl$$

Furthermore, other substances such as, lead and cadmium metals are used in the development of silicon based devices, which are proven scientifically to have hazardous impacts on human health and the environment[4]. Research reported by the International Renewable Energy Agency (IRENA) shows that in 2016 the panel waste was around 250,000 metric tonnes and it could increase significantly to 78 million tonnes by 2050 which equals to 2,000 times the weight of the Eiffel Tower[30]. Their study also shows that 1.8 million panels contain 1000,000 pounds of cadmium metals. They suggest that by 2016 about 11,000 tonnes of lead and 800 tonnes of cadmium could be spread out along with PV installations. These imply that the higher the number of silicon solar cells produced, the greater the hazardous wastes generated which could seriously threaten the environment. This clearly emphasizes that silicon solar cell is not essentially green as claimed

On the other hand, promising research findings using organic and inorganic liquids such as water and 1,2-dichloroethane could be further developed to be utilized during the fabrication processes because those are harmless by their nature[19]. Mixing these two forms of liquid enables the formation of an interface where dyes acting as photosensitisers can be dissolved between the two liquids to perform reduction and oxidation (redox) reactions. The reactions which characteristically occur in a cyclic manner initiates electron transfers. The process then generates electricity that could appropriately replace the specific role of reaction in silicon PV technology with green sources and processes. Furthermore, Plana et al.[11] discuss that by employing the same soft interface it enables them to achieve a significant value of incident-photon-to- current-efficiency (IPCE) of more than 75% with only a small amount of photoactive materials layered between the two immiscible liquids. They also argue that this system is unique as it requires not only nanostructured but also green materials (oil and water) without any resistant related issue which is normally inherited by the existing silicon solar cell technology.

Higher Rates of Electron Transfer

Liquid-liquid interfaces in DSSCs could promote a higher rate of electron transfers. Some experts state that silicon based solar cells demonstrate durable qualities where it is estimated that their lifetimes could reach around 25 years on average[4]. Nevertheless, Bozyigit, Lin, Yazdani, Yarema, and Wood[31] state that solid-state interfaces of the existing cells are likely to trap electrons inside their mesoporous structures of solid structure before reaching the semiconducting materials. The film thickness also plays an important role in the light diffusion across the materials to allow generated electrons to pass through and reach the electrodes to be collected as electricity. This shows that silicon based cells have lower electron interactions because the material thicknesses are normally greater that limit rapid penetration of sunlight into the system to generate electrons. They also point out that solid materials prohibit electrons produced to be transferred in a simple manner resulting in charge recombination by passing back the electrons to photoactive materials. This slow rate of light penetration and generated electrons causes energy losses that influence device performance.

On the other hand, generated interfaces between two different liquid forms differing in their polarities could, theoretically, be a promising approach in terms of its high electron transfer capacity[11]. This is possible by incorporating soluble photosensitisers with high capacity to absorb light together with their compatible redox couples to regenerate excited state species back to their ground states in nanoscale levels. This can only be accomplished if the photoactive materials absorb enough light to perform the reaction. Liquid based cells seem to take advantages of this compared to the silicon based. Polman, Knight, Garnett, Ehrler, & Sinke[32] report that light penetration through liquid media is faster compared to solid media. This allows the light to be rapidly and directly absorbed by photoactive materials with less interactions with other mesoporous materials associated with the solid technology. The more light penetrates into dye materials the more it improves the electron excitation from valence bands to conducting bands to initiate quicker photo electrochemical reactions. The redox cycle in liquid media also promotes rapid self-healing processes stimulating a higher rate of electron transfers as the redox reactions occur simultaneously. As a consequence, the efficiency of the proposed cells are likely to be higher compared to solid silicon solar cells.

Simple Fabrication

Simple fabrication techniques are the other key advantage of liquid-based DSSC technology. It is important to note that silicon PV device production has to be based in factories equipped with advanced apparatus[4]. For example, silicon dioxides require high temperature (1150°C) to melt the compound using pyrolysis equipment known as Bell or Siemens reactor. This technique requires such a system to produce liquid silicon together with carbon monoxide and silicon dioxide gases. The liquid silicon obtained is then slowly removed and collected as desired products[18]. The approach clearly needs extensive electricity power to support the machinery work, advanced equipment to process the raw material and well-trained experts to operate the heating systems.

Conversely, there is no precise material preparation and fabrication required for the proposed technology as there is for the silicon based cells[18]. This is because the nature of the liquid-liquid system naturally creates an interface that allows the photo electrochemical reactions to occur between the two immiscible liquids[11]. This would not involve advanced assembling processes and would therefore be appropriate for people living in remote areas. In addition, the liquid-liquid junction is also self-healing due to the difference in densities of two liquids, which in turn increases liquid cells ability to independently adjusting themselves to proper states suitable for the photoactive materials and redox mediators to perform. Moreover, these liquid solutions are also replaceable which enables people, especially those in remote areas to self-maintain the devices without any difficult re-nanofabrication method employed. The aforementioned aspects discussed in this review is summarized in Table 2 below

Table 2. The advantages of liquid based DSSC

Concerning issues	**Silicon solar cells**	**Liquid DSSCs**
Cost	• Advanced manufacturing processes using extreme temperature and vacuum leading to high cost production • Require re-nanofabrication in factories for issues of contact and connections • Require periodic and consistent maintenance	• Self-assemble nanostructure processes that reduce manufacturing procedures • Require no extreme temperature and vacuum conditions • Potentially lower production costs
Environmental impacts	• Silicon purification requires more fossil fuels which corresponds to more toxic gases released to the environment • Silicon dust may be harmful for the workers during manufacturing processes • Rare metals used such as cadmium are harmful	• Water and oil as the main components are essentially green • Abundant resources • Less harmful waste
Fabrications	• Advanced nanofabrication techniques • Require expertise in the field • Factory based re-nanofabrication when need maintenance	• Simple fabrication • Self-healing and defect free • On the spot maintenance
Electron transfer	• Charge traps and recombination leading to greater energy lost	• Higher rate of electron transfer in liquid media that may increase cell efficiency

Conclusion and Outlook

Solar cells based on silicon materials for conversion of light into electrical energy have been well developed where the efficiencies of these devices are approaching the theoretical maximum calculations. However, this solar cell platform could be suitably replaced by liquid-based solar cells because the proposed technology potentially offers lower production costs, utilizes greener methods, produces higher electron transfer rates and requires simpler fabrication techniques. This promising field opens up possibilities for further research investigation across disciplines ranging from synthetic chemistry for designing appropriate dye photosensitizer and redox mediators, electrochemistry for redox properties analysis, spectroscopy for structural characterization, surface science for electron transfer studies in liquid interfaces, computational chemistry for molecular modelling and engineering experts for device assembling and testing. Bringing this broad collaboration all together could push this promising research forward into the application level. As a result, people in general and specifically those living in remote areas may be able to have access to electricity generated through this promisingly less expensive means of energy technology to meet their energy demands and promote rapid rural developments. Besides, it supports government agenda to reduce our dependence on fossil fuels by providing renewable energy for society.

Acknowledgments

The author would like to acknowledge Prof. Paul J. Low for his insightful discussions and supportive aids for image design. Peter Foggit is also acknowledged for proofreading the manuscript.

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