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Performance analysis of a water splitting reactor with hybrid photochemical conversion of solar energy

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ABSTRACT

In this paper, a new hybrid system for hydrogen production via solar energy is developed and analyzed. In order to decompose water into hydrogen and oxygen without the net consumption of additional reactants, a steady stream of reacting materials must be maintained in consecutive reaction processes, to avoid reactant replenishment or additional energy input to facilitate the reaction. The system comprises two reactors, which are connected through a proton conducting membrane. Oxidative and reductive quenching pathways are developed for the water reduction and oxidation reactions. Supramolecular complexes $[[(\text{bpy})_2\text{Ru}(\text{dpp})_2\text{RhBr}_2](\text{PF}_6)_5$ are employed as the photo-catalysts, and an external electric power supply is used to enhance the photochemical reaction. A light driven proton pump is used to increase the photochemical efficiency of both O_2 and H_2 production reactions. The energy and exergy efficiencies at a system level are analyzed and discussed. The maximum energy conversion of the system can be improved up to 14% by incorporating design modification that yield a corresponding 25% improvement in the exergy efficiency.

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1. Introduction

Solar driven water splitting combines several attractive features for sustainable energy utilization. The energy source of the sun and the reactive media of water are readily available and renewable, and the resultant fuel of hydrogen and product of water are each environmentally clean. Fujishima and Honda [1] reported on hydrogen production from photocatalytic water decomposition by using a TiO_2 single crystal electrode. The authors examined the processes of directly transforming solar energy into chemical energy. Numerous other past studies have been conducted on the photodecomposition of water as a clean solar energy conversion process leading to a non-polluting fuel.

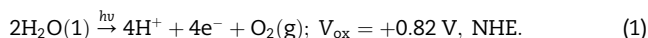
Solar energy cannot be directly stored or continuously supplied. Therefore, the conversion of solar energy to a type of storable energy has crucial importance. To utilize a low density solar flux as effectively as possible, all wavelengths of light should be used, and the efficiency of each step of the energy conversion steps should be improved. Another alternative for hydrogen production by solar energy without consumption of additional reactants is a hybrid system. A hybrid system which combines photochemical, thermochemical and electrochemical reactions has been reported previously [2].

Compared to hydrogen production methods based on fossil fuels, the high investment cost of solar hydrogen generation is a challenging issue. Production of hydrogen in a regenerative

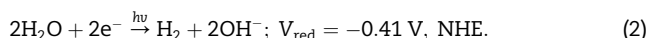
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fashion such as photochemical splitting of water has been studied [3]. The reductive side of this process requires the development of catalysts that promote the reduction of protons to molecular hydrogen, facilitated by direct excitation by a photosensitizer. Recently, molecular platinum- and palladium-based systems have been developed as heterogeneous catalysts [4,5]. Supramolecular complexes in a photocatalytic hydrogen production scheme that results in high turnover rates and numbers are another area of interest [6]. The long-lived metal-to-ligand charge-transfer (MLCT) excited state of $[\text{Ru}(\text{bpy})_3]^{2+}$ has motivated photochemical and photophysical studies leading to light to energy conversion processes [7,8]. The MLCT excited state of $[\text{Ru}(\text{bpy})_3]^{2+}$ and its analogs have the required energy to split water into hydrogen and oxygen, but it requires complicated multi component systems for operation.

One of the most promising areas is development of photochemical water splitting using supramolecular devices, which are able to capture the incident solar radiation, and generate electrons or holes at the active center where water reduction or oxidation occurs, respectively. Such systems mimic natural photosynthesis and mainly consist of supramolecular complexes of organic molecules which possess active metallic centers [9]. The water photo-oxidizing reaction occurs as follows:



Following this reaction under the influence of photonic radiation, the active center absorbs two electrons from one water molecule. The water reduction proceeds according to:



For a complete cycle, two water molecules are required. Thus, the total cell potential, E , is written as $E = E_1 + E_2 = -0.83 \text{ V to } 0.40 \text{ V} = -1.23 \text{ V}$.

Numerous studies on photochemical production of hydrogen have focused on the development of photocatalytic materials. Relatively few have examined thermodynamic studies of photon-to-electron conversion, theoretical power conversion efficiency, or energy and exergy efficiencies. This paper extends the analysis of a newly proposed photochemical water splitting system by C. Zamfirescu et al. [10] by implementing new modifications to improve the system performance. These modifications mainly consist of hybridization of a photocatalytic system by an external electric source, implementation of a light driven proton pump, and efficient photosensitizers. This paper investigates the performance of a photochemical hydrogen generation reactor through energy and exergy methods, and also comparisons with other water splitting methods.

2. System description

A photochemical reactor comprises a radiation source, a reactor transparent for the light, a gas product outlet and a photocatalytic reactor. As shown in Fig. 1, the vessel comprises two photochemical reactors separated by a light driven proton pump membrane. Similar to natural photo-

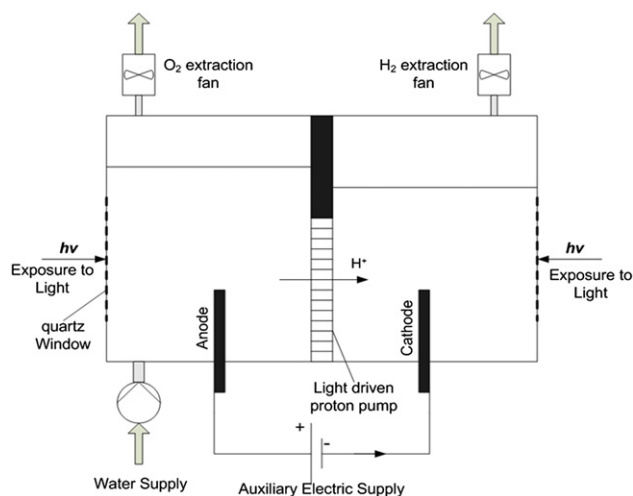


Fig. 1 – Schematic of water splitting photochemical reactor with external power source.

systems, an artificial water splitting photo-system will require a number of essential molecular components that must be organized in supramolecular complexes such that the photo-generated electron-hole pair is quickly and efficiently separated and their potential energy is delivered to functional co-catalysts. The system contains a minimum number of necessary components such as a photosensitizer, oxygen and hydrogen evolution reaction catalysts. An efficient and robust sensitizer absorbs photons and generates long-lived electron-hole pairs that are coupled to appropriate multi-electron co-catalysts for oxygen and hydrogen evolution.

Photolysis solutions consist of $\{[(\text{bpy})_2\text{Ru}(\text{dpp})_2\text{RhBr}_2](\text{PF}_6)_5$ (65 μM), water (0.62 M) acidified to pH = 2 with triflic acid, and dimethylaniline (1.5 M) in a solution of acetonitrile (4.46 mL). In the water photo-oxidation reactor, on the left, selected supramolecular photo-catalysts are used to capture light energy and generate electrical charges at reaction sites to oxidize water and produce oxygen gas and protons. Fresh water is continuously supplied to this reactor, and the flow rate of fresh water is adjusted such that the water level in the vessel remains constant. In the water reduction reactor, selected supramolecular complexes for photocatalytic reduction of water to hydrogen are dissolved in a proper concentration. These catalysts generate photoelectrons, which makes hydrogen evolution reactions occur. An auxiliary electric circuit has been considered to provide an external electric potential and improve the photocatalytic reaction performance. Negative charges, generated by a power supply, are donated at the electrode surface to the supramolecular devices, and these are transmitted to the reaction sites under the influence of photonic radiation. Two extraction fans are used above the liquid level, to extract oxygen and hydrogen, continuously.

As mentioned previously, this research will focus on the use of Ru(II)-bipyridyl complexes as sensitizers to drive the water splitting reaction. As seen in the modified Latimer diagram, shown in Fig. 2, each cycle starts with photoreactions to capture the light energy (at $\sim 450 \text{ nm}$ or 2.75 eV) to form the excited single molecule $[\text{Ru}(\text{bpy})_3]^{2+*}$. This molecule

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