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Experimental study on direct solar photocatalytic water splitting for hydrogen production using surface uniform concentrators

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ABSTRACT

In this paper, a direct solar photocatalytic water splitting system with surface uniform concentrators (SUCs) is designed. Parameters influencing hydrogen production rates and energy conversion, viz. sacrificial agent concentration, catalyst concentration and circulation speed, are analyzed under typical days. It is found that the system with SUCs has better performances with higher sacrificial agent, higher catalyst concentration and lower speed: double and triple concentration of the sacrificial agent will improve the energy conversion efficiency by 4.52% and 19.35%, respectively; double and triple the photocatalyst concentration will improve the energy conversion efficiency by 81.32% and 200.00%, respectively; energy conversion efficiency under valve-half-closed and the valve-closed conditions are improve by 21.82% and 118.18% comparing with the valve-open condition.

Introduction

Fossil fuels have become the primary energy source of the world energy consumption for decades. Environmental problems, i.e. acid rain, ozone depletion, air pollution, greenhouse effect, are terrible consequences by the usage of fossil fuels [1,2]. Renewable energy, such as solar, wind and hydrogen, will play an increasingly crucial role in the future [3]. This is particularly true to solar energy, which may be converted into chemical energy by natural solar photo-assisted reactions [4]. On the other hand, more and more researchers believe that hydrogen is another alternative solution to solve the environmental problems [5]. With the potential to develop a non-carbon energy system, hydrogen can be generated from clean and renewable sources [6–8]. In addition, hydrogen is considered as a clean carrier since byproduct of its combustion is only water [9]. Some efforts had been carried out by adding hydrogen to the experimental system and the hydrogen contribution to the system was also evaluated by establishing one model of the energy system [10]. Among the regenerative ways, photochemical splitting of water generates considerable interests, in which solar photons are absorbed and drive water is split directly into H_2 and O_2 [11,12]. There

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were so many works based on semiconductor particles in hydrogen production using solar energy [13]. Also in some researches of producing hydrogen, during the early days, using solar energy had been acted out in different countries [14]. In order to improve the efficiency of hydrogen production and reduce the cost of hydrogen generation, some researchers pay special attention on discovering efficient photocatalysts, viz. d^0 metal oxide photocatalysts (Ti^{4+}, Zr^{4+}, Nb^{5+}, Ta^{5+}, W^{6+}, and Mo^{6+}), d^{10} metal oxide photocatalysts (In^{3+}, Ga^{3+}, Ge^{4+}, Sn^{4+}, and Sb^{5+}), f^0 metal oxide photocatalysts (Ce^{4+}) and non-oxide photocatalysts [15–21]. In order to test the efficiency and stability of photocatalysts, some other researchers concentrated on the design of photoreactors [22]. In general, photoreactors of the commercial-pilot-scale reported in some critical reviews can be classified into several series: Parabolic Trough Collectors (PTCs), Non-Concentrating Collectors (NCCs) and Compound Parabolic Collectors (CPCs) [4]. Huang et al. found that main energy loss was the reflection from photoreactor window and proposed a passive self-mixing hydrogen generation system [22]. A significant number of pilot-scale photocatalytic technology demonstrations were based on PTCs. Sandia National Laboratories had done one such research in 1989, USA. The system was single-axis tracked with a total area of 465 m^2 [23]. Twelve “Helioman” units were installed in series at the Plataforma Solar de Almeria (PSA), Spain. Four parabolic trough aluminized mirrors were involved in each “Helioman” with a total area of 29.2 m^2 and they were two-axis tracked [24]. A set of single-axis tracked PTCs conducted on two drive trains were used in The National Renewable Energy Laboratory (NREL) [25]. Each drive train with concentration ratio being 20 had a total area of 78 m^2. Also, there was one project used NCC reactors for the wastewater's decoloration at Menzel Temime. Combined with two bioreactors, NCCs (2.5 m $\times$ 10 m) with a total reaction area of 50 m^2 were south-oriented and their incidence were 20° [26]. Comparing with the PTCs, performance improvement of CPCs was reported to be in the range of 30%–200% [27,28]. PSA had conducted a series of six CPCs modules with a total area of 8.9 m^2 in the early time [29]. This series could degrade dichlorophenol solutions as high as 200 mg/L within the reactor. The degradation of a wide variety of water contaminants, such as bacteria [30], dye effluents [31–33], pharmaceuticals [34], municipal waste water [35], pesticides [36] and compounds resistant to other treatments [37,38] have been successfully studied based on such CPC series at the PSA. Sarria et al. installed a collector with area of 3.08 m^2 to dig out biorecalcitrant compounds' degradation [39]. A plant, equipped with 14 CPC reactor modules with a total area of 150 m^2, was set up for the detoxification of pesticides [40]. Besides that, the strategies of power management for a power system had been developed [41]. As mentioned above, most of the NCCs, PTCs and CPCs were designed for photocatalytic detoxification or biological purposes. Few efforts have been carried out on the pilot demonstration of photocatalytic hydrogen production. One experimental system was built at PSA and hydrogen was collected by CPC reactors using Pt/(TiO_2–N) and Pt/(CdS–ZnS) as photocatalysts, respectively [42]. In order to realize the economic viable of the solar photocatalytic hydrogen generation technology, some efforts have been carried out at the State Key Laboratory of Multiphase Flow in Powering Engineering (SKLMFPE). Jing et al. [43] designed a CPC photoreactor with the maximum half incident angle being 14°. This system achieved the hydrogen productivity of 1.88 L/h and the apparent energy conversion efficiency of 0.47. Based on Jing's work, we [44] built one pilot-scale hydrogen production experimental system with the developed CPC (concentration ratio being 4.22) and total reaction area being 103.7 m^2. After that, Yang et al. [45] developed a mathematical model which was recognized as the surface uniform concentrator (SUC). As we know, two lines were left on the receiving surface of the circular pipe after using CPC. The SUC was applied to avoid non-uniform concentration of solar intensity on the receiving surface of CPC, because after using SUC, the light can be distributed uniformly around the circular tube. Based on our previous study, an outdoor direct solar photocatalytic water splitting system for hydrogen generation is designed and analyzed in this study. The SUC is used as the concentrator instead of the previous CPC ones. Parameters influencing hydrogen production and energy conversion rates, viz. sacrificial agent concentration, catalyst concentration and circulation speed, are analyzed under typical days.

Experimental setup

Experimental materials

The NiS-$Cd_xZn_{1-x}S$ is the photocatalyst because of its high energy conversion efficiency being 6.0% [17]. The sacrificial agent is the mixture solution of Na_2SO_3 and Na_2S with being substitution of sulphide pollutant, such as hydrogen sulfide aqueous solution, which is a dangerous waste which would seriously pollute the environment [46], furthermore, it would also cause the chronic poisoning of workmen [47]. The other basic conditions of the experiments are shown in Table 1.

Experimental system

A new direct solar concentrating hydrogen production system is designed on the basis of previous research as shown in Fig. 1 [45]. The solution is pumped into the reaction tube. Solar radiation is focused on the reaction tube with the SUCs, which actives the solar photocatalytic water splitting process in the pipe with the material being PVC. With SUCs, the energy of the incident light can be focused on the surface of the circular pipes within higher energy density. The mixture of hydrogen and solution is pumped back into the storage tank through the

Table 1 – Basic conditions of the experiments.

Items	Numerical Value
Concentrator	SUC
Volume of reaction solution (including the tank and flow pipes)	70 L
Sacrificial agent	Mixture solution of Na_2SO_3 and Na_2S
Photocatalyst	NiS-$Cd_xZn_{1-x}S$
Temperatures in the photoreactor	30-60 °C

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