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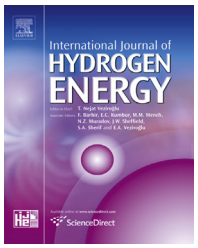

A review and evaluation of photoelectrode coating materials and methods for photoelectrochemical hydrogen production

Canan Acar[*], *Ibrahim Dincer*

Clean Energy Research Laboratory (CERL), Faculty of Engineering and Applied Science, University of Ontario Institute of Technology, 2000 Simcoe Street North, Oshawa, Ontario, L1H 7K4, Canada

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ABSTRACT

In this study, CdS, TiO_2, CdSe, WO_3, Fe_2O_3, and CuO/Cu_2O based photoelectrode coating materials are considered for investigation under some significant selected coating methods, namely, chemical vapor deposition (CVD), electrochemical deposition (ECD), electrodeposition (ED), sol–gel (SG), spin coating (SC), and spray pyrolysis (SP). Their performance evaluations are carried out comparatively for photoelectrochemical hydrogen production. The photocurrent generation and voltage/light requirements of these photoelectrodes are also compared to evaluate the impact of material and method selection on photoelectrochemical hydrogen generation. The results show that among selected photoelectrode coating materials, CdS based photoelectrodes generate the highest photocurrent (3715.58 $\mu A/cm^2$), followed by CdSe (2963.43 $\mu A/cm^2$), CuO/Cu_2O (1873.33 $\mu A/cm^2$), TiO_2 (1500.60 $\mu A/cm^2$), WO_3 (1435.28 $\mu A/cm^2$), and Fe_2O_3 (443.3 $\mu A/cm^2$). Average photocurrent densities of selected coating methods show that photocathodes processed by spin coating produce the highest photocurrent (2343.57 $\mu A/cm^2$), followed by electrochemical deposition (1623.36 $\mu A/cm^2$), electrodeposition (1359.77 $\mu A/cm^2$), spray pyrolysis (1217.50 $\mu A/cm^2$), chemical vapor deposition (619.44 $\mu A/cm^2$), and sol–gel (335.06 $\mu A/cm^2$).

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Introduction

Due to population growth and rise in standards of living, global energy demand keeps increasing. This demand is expected to continue even more in the future, due particularly to some highly-populated developing countries, e.g., China and India. With their non-homogeneous distribution, limited nature, slow formation, and rapid depletion, fossil fuels are not considered to be able to fully meet future generations' energy needs. Environmental damage caused during processing, transfer, and utilization of fossil fuels is another issue. The emissions caused by fossil fuel combustion, especially CO_2, are known as greenhouse gases and considered to be the main cause of climate change. In the recent decades, there has been an ongoing research on clean and renewable energy systems to meet world's increasing energy demand without exhausting existing sources and damaging the environment. Renewable energy resources are treated as promising candidates compared to fossil fuels since they are replenished also their end use emits very low or zero

* *Corresponding author.*
E-mail address: Canan.Acar@uoit.ca (C. Acar).
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pollutants. However, renewables have intermittent and fluctuating nature, they require a medium to be stored. Hydrogen, as an abundant and clean energy storage medium, has a potential to meet world's energy demand. When produced from renewables and water, production and end use of hydrogen does not emit any pollutant.

Solar energy has a great potential to produce hydrogen in a clean and efficient way since it is considered as inexhaustible and nonpolluting. There are two major solar energy conversion technologies, such as (i) solar thermal conversion: uses infrared (IR) part (about 49%) of the solar spectrum (e.g., solar heat collectors) and (ii) solar photon conversion: uses visible/near–ultraviolet (UV) part (51%) of the solar spectrum. Furthermore, the solar photon conversion devices, namely, solar cells, can be classified into three main categories [1], such as (a) solid–state photovoltaic (p/n junction) solar cells; PV, (b) metal–semiconductor based Schottky barrier (M–S) solar cells and (c) semiconductor liquid junction based photoelectrochemical (PEC) solar cells.

Note that semiconductor liquid junction photoelectrochemical solar cells are in between solid–state photovoltaics and natural photosynthetic systems. The solid state solar cells are advanced, efficient and reliable devices with solar to electricity conversion efficiencies of greater than 10%. In some special multilayer solid–state cells the efficiencies can go up to 25–26% [2]. One of the most important factors affecting solar conversion efficiencies is the type of solar absorbing material.

Solid–state photovoltaic cells are suitable for special applications such as providing power to remote locations. Photoelectrochemical cells are in early research and development phase, but they have a high potential to utilize a broader range of solar spectrum in an efficient manner. In Table 1, the aspects material synthesis and processing, junction formation, conversion efficiency, *in situ* storage, stability and economic viability for PV and PEC cells are compared. It is clear that PEC has significant advantages over PV cells such as convenient and economically viable material synthesis and processing. PEC cells are also easy to manufacture, the process involves immersing the photoactive semiconductor into the electrolyte containing a suitable redox couple. Possibility of *in situ* storage is another major advantage of PEC cells. Main components of PEC cells are (i) photoactive semiconductor (SC) electrode immersed in a (ii) suitable redox couple electrolyte and a (iii) metal or semiconductor metal as counter electrode. When irritated with light of $h\nu \geq E_g$, the semiconductor generates and separates electron (e^-)–hole (h^+) charge carriers.

There are several available solar hydrogen routes in the open literature. By taking advantage of direct use of sunlight, PEC systems offer low cost, environmentally benign, and efficient ways to produce hydrogen. PEC systems integrates solar energy collection and water electrolysis in a single photocell [3–5]. PEC systems are classified based on their Gibbs free-energy change as:

i. Regenerative PEC solar cells ($\Delta G = 0$) (Direct conversion of solar to electrical energy).

ii. Photoelectrosynthetic cells ($\Delta G \neq 0$) (Conversion of solar to chemical energy).

a. Photoelectrolysis cells ($\Delta G > 0$) (Endoergic conversion of solar to chemical energy).

e.g., $2H_2O \rightarrow 2H_2 + O_2$

b. Photocatalytic cells ($\Delta G < 0$) (Exoergic conversion of solar to chemical energy)

e.g., $N_2 + 3H_2 \rightarrow 2NH_3$

In the literature, there are many studies on water splitting by using semiconductor photoelectrodes. First photoelectrochemical (PEC) water splitting activity was reported by Fujishima and Honda [3] by using TiO_2 electrodes. The photon energy is converted into chemical energy in these systems and as a result, there is a significant increase in Gibbs free energy. This reaction can be written as

$$H_2O + h\nu \rightarrow H_2 + \frac{1}{2}O_2 \quad \Delta G^\circ = 238 \text{ kJ/mol} \tag{1}$$

There are over 100 photocatalytic metal oxide based systems in the open literature which reported to achieve overall water splitting (simultaneous H_2 and O_2 production). Yet most of these photocatalysts have large band gaps and they require ultraviolet (UV) light ($\lambda < 400$ nm) irradiation. Fig. 1 shows that almost half of the solar energy on Earth's surface is within the visible light region (400 nm $< \lambda <$ 800 nm). Therefore, large scale photocatalytic water splitting based hydrogen production requires the efficient utilization of visible light. By using standard solar spectrum, maximum solar conversion efficiency for photocatalytic water splitting is calculated with a quantum efficiency of 100%. This amount would be only 2% even if the entire UV region up to 400 nm were exploited, which goes up to 16% when the light spectrum up to 600 nm is used and further to 32% when light up to 800 nm is utilized [6]. For that reason, there are numerous studies in the literature on efficient water splitting by using visible light.

The aim of this study is to summarize the current state of photoelectrode coating methods and materials for photoelectrochemical hydrogen production. In this regard,

Table 1 – Comparison of PV cells and PEC (Modified from Ref. [2]).

	PV cells	PEC
Material synthesis	Costly	Relatively cheap
Material processing	Costly	Relatively cheap
Junction formation	Difficult	Relatively easy
Routine conversion efficiency	>10% (>%20 in special devices)	>7% in special cells
In situ storage	Not possible	Possible
Cell stability	Stable	Stable with proper electrolytes
Economic viability	Viable but costly	Viable, cheaper than PV

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