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Mechanism of Water Splitting and Hydrogen-Oxygen Production

A research presented to the Board of College Education-Al-Qadisiyah University as a Part of Requirements of Bachelor's Degree in Physics

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﴿أَوَلَمْ يَرَ الَّذِينَ كَفَرُوا أَنَّ السَّمَاوَاتِ وَالْأَرْضَ كَانَتَا رَتْقاً فَفَتَقْنَاهُمَا وَجَعَلْنَا مِنَ الْمَاء كُلَّ شَيْءٍ حَيٍّ أَفَلَا يُؤْمِنُونَ ﴾ صدق الله العظيم [الأنبياء : 30]

In the name of Allah, the Beneficent, the Merciful

(Have not those who disbelieve known that the heavens and the earth were of one piece, then We parted them, and We made every living thing of water ? Will they not then believe)

Sura Al-Anbiya : 30

Dedication

To :

Our parents (fathers and mothers).

Who devoted their whole life (times and sources) to prepare all our needs and to make our way clear .

Abstract

A short report consists three chapters: the first chapter is an introduction which shows and explains the importance of the hydrogen energy as a renewable and clean energy source in scientific community and government organizations. Also, we (the researchers) gave a short account of historic facts about the previous experiments of water splitting and attached them with the modern ones. At last of chapter one, the researchers show the problems that lead to the necessity of producing clean energy. Photolysis or photo splitting of water into gaseous O_2 and H_2 represents one of the most challenging and promising ways of solar energy accumulation.

Chapter two concerns about the Mechanism of Water Splitting by studying reviews and summarizes the very recent progress (mainly in the last 2–3 years) on three major types of solar hydrogen production systems: particulate photocatalysis (PC) systems, photoelectrochemical (PEC) systems.

The type (Photoelectrochemical PEC) water splitting of cell electrolyzes water to hydrogen and oxygen gas by irradiating the anode with electromagnetic radiation. This has been referred to as artificial photosynthesis and has been suggested as a way of storing solar energy in hydrogen for use as fuel . Also, we explained the chemical interactions of these operations and added some figures to show expanded dimensions of the subject.

Chapter three is a short conclusion about the important results and statistics that the researchers reached and reviewed through the research. At the end, we listed the references which we used in the research.

Acknowledgements

We would like to express our deep gratitude to all our teachers at university, although busy with their jobs, they did their best to help us during our study.

We specially thank the faculty of our college and department (College of Education - Department of Physics) .

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Chapter One
Introduction

In recent years, hydrogen energy has found increased attention as a renewable and clean energy source in scientific community and government organizations[1–3]. Besides, it has enormous potential to be developed as a new substitutive energy resource for solving energy crisis in the future. Among many methods for the generation of hydrogen, solar water splitting is a particularly attractive one because of the environmental friendliness and the abundance of water source [4-7]. Fujishima and Honda were the first to demonstrate the concept of water splitting in a series of experiments using titanium dioxide as a Photoanode [8]. Up to date, many efforts have been done on the development of photoelectrodes to improve the efficiency of hydrogen generation.

Due to the rapid acceleration of technological advances across the globe, each year more and more products are produced to make our lives more comfortable and convenient. Most of these products originate from natural sources such as petroleum, coal, natural gas and mineral sources; meanwhile, many environmental problems occur during the various steps of the extraction, transportation, and transformation of raw materials into final products. These problems include CO_2 emission, which

contributes greatly to global warming, ozone depletion, and air and water pollution, which increase health risks to living things. Although these problems may only appear to impact local areas, they actually pose cumulative hazards on a global level. For example, it is reported that pesticides and heavy metals have been detected in remote Antarctic areas, far from their points- of-use. As a result, each country around the world is facing challenges to look for better strategies to solve such energy and environmental problems. Nanomaterials are novel material forms that emerged in the 1980s and have been studied intensely in recent years [9-12].

Photolysis or photo splitting of water into gaseous O_2 and H_2 represents one of the most challenging and promising ways of solar energy accumulation, as solar hydrogen can then be used as an alternative fuel. Numerous comprehensive studies [13-14] have been performed since the first report of photocatalytic water splitting on TiO_2 surface was published in 1972 [15]. The band energy position of TiO_2 relative to the electrochemical potentials of the H_2/H_2O and O_2 /H_2O redox couples [16-17] demonstrates that due to the presence of a large over potential for the evolution of H_2 and O_2 on the TiO_2 surface, TiO_2 alone cannot photo decompose H_2 O; rather the photo assisted oxidation of the oxygen vacancy sites on a reduced TiO_2 surface is responsible for H_2 evolution

[18-19] TiO_2 -assisted photo electrolysis of water is achieved using a close circuit photo electrochemical cell that consists of a TiO_2 anode and a Pt counter electrode and is exposed to near-UV light [20-21]

Generation of the photo electrochemical e^-_ h+ pair in TiO_2 is followed by the transfer of electron to the Pt electrode and reduction of water molecule to evolve H_2 according to the following reaction:

$$2H^+ + 2e^- \rightarrow H_2 \uparrow \quad (1)$$

whereas at the anode, water oxidation takes place according to following reaction:

$$H_2O + 2h^+ \rightarrow 1/2\ O_2\uparrow + +2H^+ \quad (2)$$

Therefore, the overall photo splitting of water will be given as follows:

$$H_2O\ (hv + TiO\text{-}Pt) \rightarrow 1/2\ O_2\uparrow + H_2 \uparrow \quad (3)$$

Various measures have been taken to increase the rate of hydrogen evolution over TiO_2 surfaces such as using artificial high-power UV light sources and/or nanostructured materials. However, the energy conversion efficiency on TiO_2 was rather low and the reasons for this are [22]:

1) fast recombination of photo generated electrons and holes.

2) fast backward reaction.

3) inability to harvest visible and IR light at longer wavelengths than ~400 nm.

4) less effective surface area.

The new technologies need to be developed to efficiently and economically capture CO_2 from air. Therefore, environment purification (i.e., CO_2 reduction) and energy production (i.e., liquid fuel) can be achieved simultaneously, as shown schematically in Figure 1.1 Because of their higher volumetric energy densities, hydrocarbon fuels can alleviate hydrogen storage issues. Further, hydrocarbon fuels can be used in existing gasoline infrastructure with limited modifications. Hence, building expensive liquid H_2 infrastructure can be avoided. Therefore, there is an immense potential in solar water splitting both for carbon-free energy and environment. Research and development in this area are under advancement. Figure 1.1 shows schematic diagram of solar water splitting for energy production and environment purification.

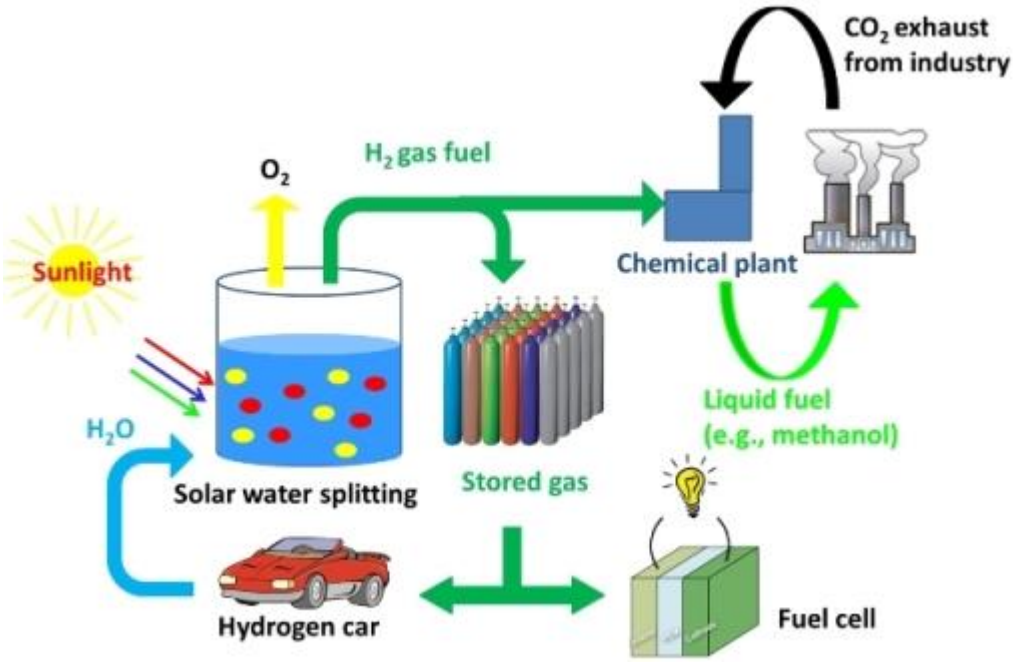

Figure 1.1: Schematic diagram of solar water splitting for energy production and environment purification.

Chapter Two

Mechanism of Water Splitting

2.1 Introduction

Water splitting is the general term for a chemical reaction in which water is separated into oxygen and hydrogen. Efficient and economical water splitting would be a key technology component of a hydrogen economy. Various techniques for water splitting have been issued in water splitting patents in the United States. In photosynthesis, water splitting donates electrons to power the electron transport chain in photosystem.

Widely used solutions for solar hydrogen production mainly fall into three categories (Figure 2.1):

1. Photocatalyst (PC) systems.
2. Photoelectrochemical (PEC) systems.
3. Photovoltaic photoelectrochemical (PV- PEC) hybrid systems.

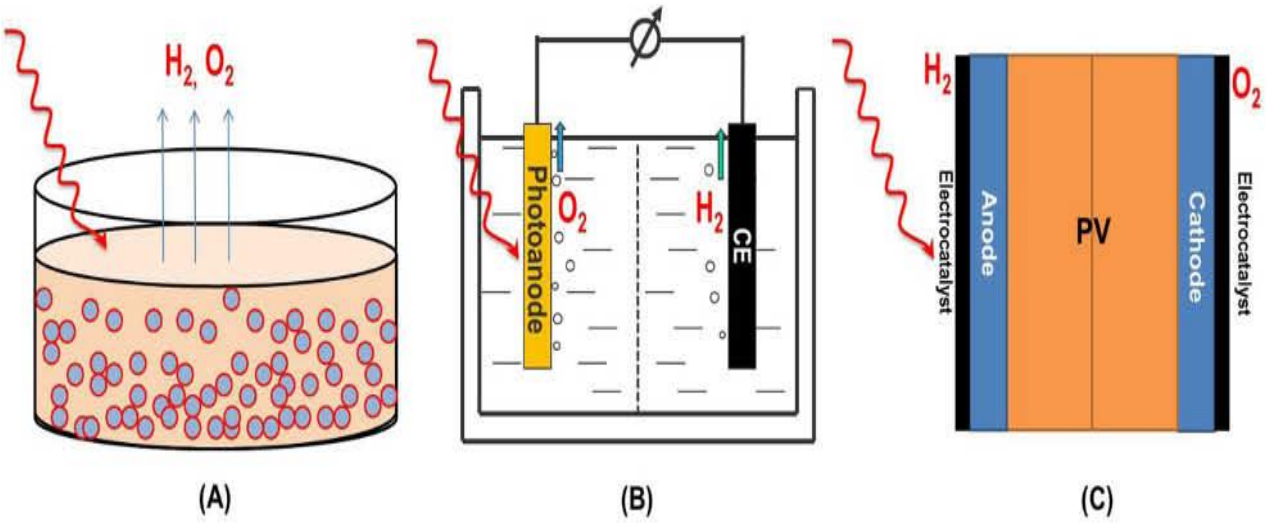

Figure 2.1: The solutions for solar hydrogen via water splitting. (A) Particulate photocatalytic (PC) water splitting system, (B) photoelectrochemical (PEC) water splitting system and (C) photovoltaic photoelectrochemical hybrid (PV/PEC) system.

2.2 Particulate Photocatalyst (PC) systems:

In PC systems, which are the simplest and lowest cost for potential scalable solar hydrogen production, photocatalyst powders are dispersed in water for hydrogen production under light irradiation. However, the necessity of H_2/O_2 gas separation and an enclosed reaction system on a large- scale are disadvantages in PC water splitting processes.

The molecular sieving effect of microporous membranes has shown promise for the safe separation of the mixture of H_2 and O_2 gas. Great progress has been achieved in gas separation by zeolite membranes, and some reviews have highlighted the recent advances in both fundamental science and potential industrial applications [23–25].

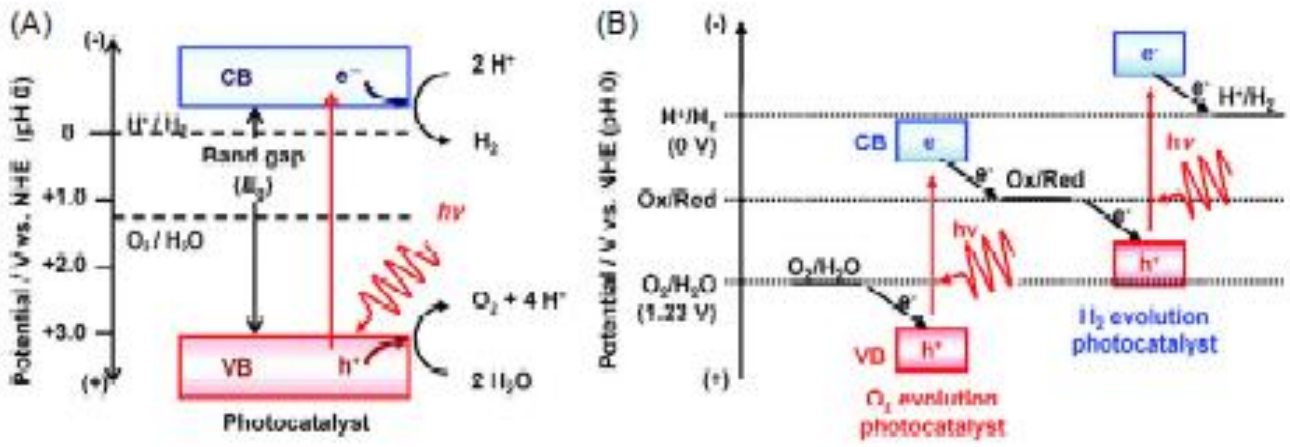

Figure 2.2: Energy diagrams of photocatalytic water splitting based on one-step excitation (A) and two-step excitation (Z-scheme) (B). Adapted with permission from reference [16]. Copyright 2014 Royal Society of Chemistry.

The H_2/O_2 mixture produced in PC systems may be separated by molecular sieving based on the different kinetic diameters of the two gases (H_2, 2.9 Å; O_2, 3.5 Å). In photoelectrochemical systems, the photocatalysts must first be prepared on conductive substrates as electrodes and a small additional bias applied for water splitting.

To achieve overall water splitting with particulate photocatalyst systems, single-step excitation and two-step excitation (Z- scheme) photocatalysts have been investigated for solar hydrogen production, Figure 2.2.

In the case of single-step excitation photocatalysts , the band gap of the semiconductor must at least straddle the chemical potentials for proton reduction and water oxidation thermodynamically, however, this is not necessary for a two-step Z-scheme photocatalyst system.

In Z-scheme water splitting systems, an electronic mediator is used to connect the hydrogen-evolution and oxygen-evolution redox reactions even though H_2 and O_2 production takes place on the surface of different photocatalysts.

Many one-step excitation photocatalysts have been investigated in the past few decades for overall water splitting in PC systems. Kato et al. [33] reported that NiO-loaded $NaTaO_3$ doped with lanthanum showed a high photocatalytic activity for water splitting into stoichiometric amounts of H_2 and O_2 under UV irradiation and an apparent quantum efficiency (AQE) of 56% at 270 nm. The representative visible-light-responsive photocatalyst for overall water splitting is Rh@Cr_2O_3/GaN:ZnO, which was first reported by Maeda et al. [34,35] with an AQE of up to 5.9% at a wavelength of 420 nm.

More and more photocatalysts for overall water splitting under visible light irradiation have been explored recently (e.g., $In_{1-x}Ni_xTaO_4$

[36], $LaMg_xTa_{1-x}O_{1+3x}N_{2-3x}$ [37], nitrogen-doped graphene oxide quantum dots [38], In GaN/GaN nanowires [39].

Very recently, using a particle transfer method, Wang et al. [40] prepared a photocatalyst sheet for overall water splitting composed of a hydrogen- evolution photocatalyst ($SrTiO_3$: La,Rh) and oxygen-evolution photocatalyst ($BiVO_4$:Mo) with Au acting as an electronic shuttle . The photocatalyst sheet exhibited an extremely high overall water splitting activity with an AQE of 30% at 419 nm and STH efficiency exceeding 1%, which is the highest reported for a particulate photocatalyst system. The STH of this system is to expected be further improved by employing photocatalysts with narrower bandgap energies than the short absorption edge wavelengths of $SrTiO_3$: La,Rh and $BiVO_4$:Mo (520 and 540 nm, respectively).

Although the STH efficiency is still far below the estimated industrial requirement of 10%, the work leads us to believe that scalable water splitting using particulate semiconductors is not far from unreachable in the future. One of the greatest obstacles to the future application of PC systems is the cogeneration of H_2 and O_2 gas, which results in a combustible mixture; suitable engineering controls will be strongly needed to mitigate this safety risk [41,42]. Gas separation in PC systems is a very costly process.

It has been estimated that the energy required for gas separation and the dilution of the gas mixture below the explosion limit will use 60% of the energy stored in the produced hydrogen [42]. Therefore, efficient photocatalysts responsive to visible light, especially to wavelengths larger than 600 nm, are desired because the maximum peak of the solar spectrum is located near this range.

Furthermore, the investigation of new gas separation strategies and new materials for H_2/O_2 separation (e.g., highly selective molecular sieve membranes for oxygen/hydrogen permeation) has become more and more important for future applications. Two-step excitation water splitting (Z-scheme) systems that couple a hydrogen-evolution photocatalyst (HEP) and oxygen- evolution photocatalyst via an electronic shuttle (e.g., IO^3/I, Fe^{3+}/Fe^{2+}) have also attracted increasing attention since they were first reported in 2002 [43–46].

The highest AQE for a Z-scheme water splitting system was recently reported by Chen et al. [47], who constructed a heterojunction between $MgTa_2O_{6-x}Ny$ and TaON for efficient charge separation, and it could act as a HEP for Z-scheme water splitting to achieve an AQE of 6.8%@420 nm. Although the H_2 and O_2 gases are physically separated on the different sides of Z-scheme

systems, their AQE and STH efficiencies are still very low and\ the highest AQE has only been improved from 6.3% (Pt/ZrO2/TaON as HEP) [48] to 6.8% in more than 5 years.

The visible light responsive photocatalyst, especially for the hydrogen- evolution reaction in the presence of the electronic shuttle,

largely determines the efficiency of Z-scheme water splitting.

2.3 Photoelectrochemical (PEC) systems:

To make a PEC cell work, one or both of the electrodes should be a photoactive semiconductor, in which a space- charge layer forms at the semiconductor/liquid junction. Upon irradiation, photogenerated carriers are separated by the space-charge field and the minority carriers (holes for an n-type photoanode and electrons for a p-type photocathode) travel to the semiconductor-liquid interface for reaction [26].

There is no need for gas separation in PEC water splitting systems because the production of H_2 and O_2 is spatially separated at different electrode sides. When semiconductor-based photocatalysts are prepared on an electronic substrate to form photoanodes- or photocathodes for PEC water splitting in the presence of an electrolyte solution, electron transfer takes place at the semiconductor-solution interface. This causes the Fermi level to equilibrate with the redox potential of the electrolyte solution and

the band bending is consequently produced at the space-charge-layer. Three different configurations of PEC water splitting system have been explored: single photoanode (n-type semiconductor), as shown in figure 2.3.

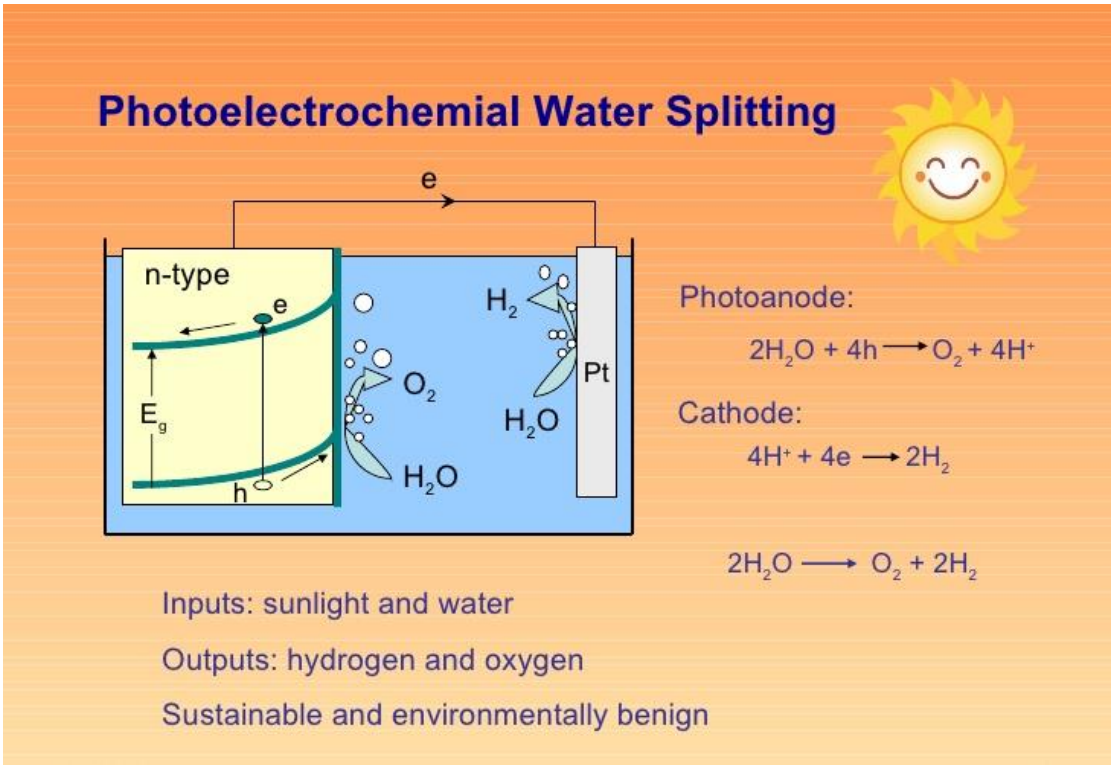

Figure 2.3: Shows photoelectrochemical water splitting cell and explains it's inputs and outputs.

2.4 Photovoltaic-photoelectrochemical hybrid systems:

PV-PEC hybrid system for hydrogen production is based on the coupling of highly efficient photovoltaic solar cells and with water electrolysis. PV-PEC systems have many advantages for hydrogen

production compared with PEC systems if cost is not a major consideration.

For example, for PEC systems the difficulties are a lack of efficient light absorber (for reasonable solar conversion efficiency, the band gap must be less than 2.0 eV), the corrosion of the semiconductor (thermodynamically, the most useful semiconductors are photochemically unstable in water), and the energetics (the difficulty of matching the semiconductor band-edge energies to the H_2 and O_2 evolution reactions) [27,28], all of which are not present for PV-PEC water splitting system.

For the hydrogen production via solar water splitting, some latest reviews focusing on materials designing, engineering and energy evaluation have been comprehensively summarized [29–32]. For photovoltaic-photoelectrochemical hybrid systems, three approaches have been proposed for coupling the photovoltaic material with the electrolytic water splitting components: integrated PEC devices, partially integrated PEC devices, and non-integrated PEC devices (Figure 2.4).

The solar to hydrogen efficiency of photovoltaic photoelectrochemical (PV-PEC) systems are the highest among all three kinds of solar hydrogen production system. The first monolithic PV-PEC device for

hydrogen production via water splitting was constructed by Khaselev et al. [27] using a $GaInP_2$/GaAs tandem cell in 1998, which showed an STH efficiency more than 10%. Combining a WO_3/$BiVO_4$ photoanode with a double-junction GaAs/InGaAsP solar cell to construct a self-operating integrated photoelectrodes.

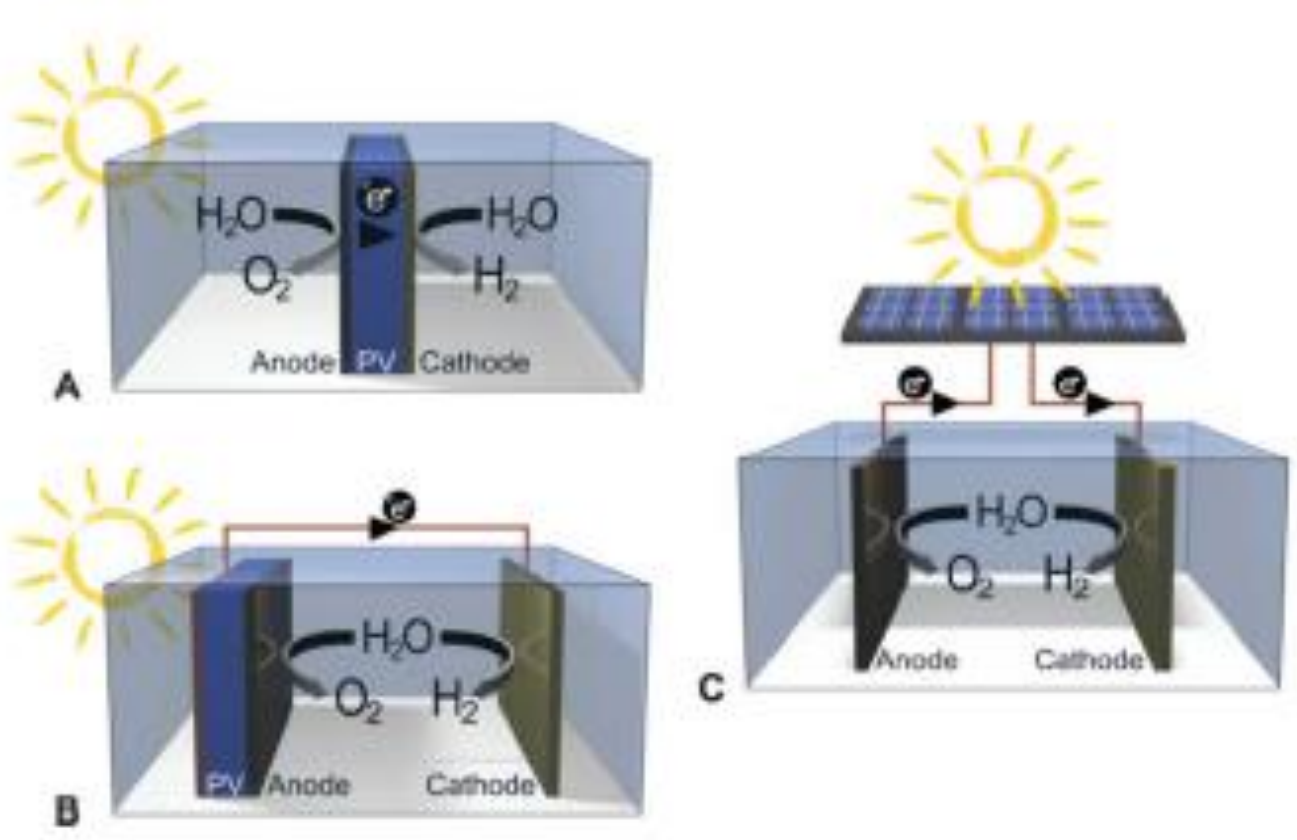

Figure 2.4. Three approaches for coupling the photovoltaic material and electrolytic water splitting components. (A) integrated PEC device; (B) partially integrated PEC device; (C) non-integrated PEC device.

Chapter Three
Conclusions

A great increase in research activity around solar hydrogen production via water splitting has been achieved in the past 2–3 years. The STH efficiency of particulate photocatalyst systems has now exceeded 1.0%, and this value has been improved to more than 2.5% and 22.4% for PEC and PV-PEC water splitting systems, respectively. Challenges and opportunities coexist in solar water splitting for hydrogen production like the two sides of a coin.

New semiconductor- based photocatalysts with wide range light absorption, new strategies for improving photogenerated charge separation, and new materials and techniques for gas separation must be explored urgently before scaled-up solar hydrogen production can be realized. Meanwhile, advanced characterization technologies, especially In-situ and ultra-fast spectroscopy methods, that can provide information at very fast time-scales are essential for understanding the mechanism of water splitting reactions.

The most exciting thing that has engaged more and more researchers in this field is that there are many important scientific problems that are still unclear, e.g., how charge separation takes place in condensed matter at very fast time- scale, how H–O chemical bond

breaking and H–H and O–O chemical bonds forming at the surface of a photocatalyst, and how to accurately simulate charge separation and surface reactions via theoretical calculations.

Such fundamental research will help us to deeply understand the mechanisms of and give further guidance for constructing Highly-efficient solar energy conversion systems.

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الخلاصة

بحث مصغر يحتوي على ثلاثة فصول :- الاول منها عبارة عن مقدمة تعرض وتوضح اهمية الطاقة الهيدروجينية كطاقة متجددة ونظيفة في المجتمعات العلمية والمنظمات الحكومية. كذلك قام الباحثين بإعطاء نبذة تاريخية حول التجارب السابقة لفصل جزيئة الماء وربطها بالتجارب المعاصرة . في نهاية الفصل الاول قام الباحثون بعرض المشاكل التي ادت الى الحاجة لإنتاج طاقة نظيفة . ان عملية فصل جزيئة الماء ضوئيا الى اوكسجين وهيدروجين تمثل واحدة من اهم التحديات والطرق الواعدة وذلك لوفرة الطاقة الشمسية .

في الفصل الثاني تناول الباحثون تقنيات فصل جزيئة الماء عن طريق استعراض الادبيات المنشورة وتلخيص العمل المستمر والمعاصر وخصوصاً في السنوات الثلاث الاخيرة لثلاث نظم لإنتاج الهيدروجين والاوكسجين باستخدام الطاقة الشمسية وهي:- نظام التحفيز الضوئي الخاص (PC) ، والنظام الضوئي الكيميائي الكهربائي (PEC) . ان النظام الضوئي الكيميائي الكهربائي PEC لفصل جزيئة الماء بأستخدام أقطاب كهربائية وخلايا الماء الى غازي الهيدروجين والاوكسجين عن طريق إضاءة قطب الانود بالإشعاع الكهرومغناطيسي . هذه العملية تم اقتراحها كطريقة لتوليد الهيدروجين لاستخدامه كوقود ، كذلك قام الباحثون بإيضاح التفاعلات الكيميائي لهذه العمليات ثم اضافة بعض الاشكال التوضيحية لعرض افاق اوسع من الموضوع .

ان الفصل الثالث هو عبارة عن استنتاج قصير حول اهمية النتائج التي قام الباحثون بالتوصل اليها واستعراضها خلال هذا البحث ، وفي النهاية قام الباحثون بإدراج المصادر التي تم استخدامها في البحث .

جمهورية العراق
وزارة التعليم العالي والبحث العلمي
جامعة القادسية / كلية التربية
قسم الفيزياء

ألية فصل الماء وانتاج الهيدروجين والاوكسجين

بحث مقدم إلى مجلس كلية التربية ـ جامعة القادسية كجزء من متطلبات الحصول على درجة البكالوريوس في الفيزياء من قبل

عباس خضير هادي الجبوري
عباس رحيم مطشر البديري

أشراف
أ.م.د. أراء مبدر حولي الشباني

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