Developing an Integrated Solar Powered System to Generate Hydrogen from Sea Water

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This paper highlights efforts to design and fabricate a lab-scale integrated module to generate and purify hydrogen from sea water obtained from Gulf of Aqaba in a totally green process. The electrochemical reactor performance was controlled and monitored online. Multiple regression analysis was used to produce a model to show the effect of panel temperature (Tc), solar irradiation (Ir) and current (I) on the production of hydrogen (Q):

$$Q = 158.0047 - 0.37203 \times Tc^{1.6} + 0.02923 \times Ir^{1.4} + 0.006901 \times I^2$$

This model was selected according to the highest R^2 and R^2adj and model significant according to ANOVA statistical analysis based on F- and P-tests. The produced hydrogen was further purified using a Pd-Cu membrane and the results revealed that the membrane was able to separate and purify hydrogen from the feed mixture with infinite selectivity in the studied range of variables.

1. INTRODUCTION

Solar Hydrogen Energy Systems (SHESs) are receiving an increasing attention worldwide due to the huge energy potential of solar radiation and the effective conversion to chemical energy carriers like hydrogen [1,2]. Hydrogen produced from water using solar energy is a sustainable source for home heat and/or electric energy supply [3,4]. There has been much recent development of SHESs for hydrogen production, however, cost and efficiency can be the main shortcomings for proper

utilization, therefore, a great attention has been given to reduce the system cost and to maximize the overall system efficiency. Different researches and studies have been made to investigate and model all the factors that affect the efficiency of SHESs to aid in designing a more efficient system with optimum operating conditions, for example Clarke *et.al.*[5], Shaltout [6], and Djafour et.al. [7]. There has been also some demonstrations of renewable energy hydrogen systems where hydrogen is generated using solar radiation, e.g., Negrou [8] and Gibson et.al. [9].

A solar PV plant utilizing hydrogen as the energy storage media and a fuel cell for generating power was installed in 1991 at Humboldt State University-Telonicher Marine Laboratory [10]. In this system, a 9.2 kWe PV array was directly coupled to a 6 kWe alkaline electrolyser capable of producing 20 L of hydrogen/min. Another hydrogen-based renewable energy demonstration system (PHOEBUS) was installed at Forschungszen trum Julich, Germany and supplied part of the energy to the Central Library [11]. Agbossou *et al.* [12] investigated hydrogen renewable energy system that comprised mainly of a 10 kWe wind generator, a 1 kWe PV array, a 5 kW alkaline electrolyser (capable of delivering 1 normal m3/h hydrogen compressed at 7 bar), a 5 kW PEM fuel-cell stack, 48 V deep discharge batteries for voltage stabilization, a DC bus controller that included batteries for energy transfer and a DC–AC inverter. The system was installed and investigated at Hydrogen Research Institute of Quebec University. The voltage produced by a wind generator and solar PV array was regulated and converted to suit the electrolyser.

Most of the previous researches have used interfacing controls and power electronics such as maximum power point tracker (MPPT) and/or a DC–DC converter in order to interface the electrolyser to intermittent energy sources such as solar PV and also to allow for different current–voltage characteristics of the electrolyser and the solar PV array. It would be particularly advantageous if an electrolyser can be simply and efficiently coupled directly to a source of renewable electrical energy, thus reducing the need for intervening electronics [13]. Troncoso *et.al.*[14] investigated the implementation of electrolysis plant in combination with wind power plant. The economic rationale was investigated for an isolated power system as a function of wind penetration, wind curtailment target, electrolyser cost, hydrogen system efficiency and hydrogen sales price. Derbal-Mokrane *et.al.*[15] studied the feasibility of hydrogen production at high temperature electrolyser, using a hybrid solar resource, thermal energy to produce high temperature, steam water and photovoltaic energy for electricity requirements.

Hydrogen can be generally purified through one of three major processes: (1) pressure swing adsorption (PSA), (2) fractional/cryogenic distillation, or (3) membrane separation [16]. While PSA and fractional/cryogenic distillation systems are in commercial operation, they are not cost effective and are quite energetically demanding for the separation and purification of H_2. In addition, neither of these methods provides sufficient purity for the targeted applications in the H_2 economy.

Membrane separation is currently considered to be the most promising for hydrogen separation. An optimum membrane should separate H_2 from gas mixtures with high flux (or permeance) and purity (selectivity). The most important H_2-selective membranes are: (1) dense polymeric membranes, (2) microporous ceramic membranes (e.g., silica, alumina, zirconia), (3) dense metallic membranes (Palladium alloys), (4) porous carbon, and (5) dense proton conducting ceramic membranes. Dense membranes separates H_2 based on the solution/diffusion mechanism where gas molecules are adsorbed

on one side of the membrane, dissolve in the membrane material, diffuse through the membrane, and desorbs on the other side of the membrane [17].

If diffusion through the membrane takes place in the form of ions and electrons (= proton exchange transport) or as atoms (e.g. for hydrogen transport through dense metal), the molecule needs to split up after adsorption and recombine after diffusing through the membrane. Palladium (Pd) and Pd alloy dense membranes have attracted considerable attention owing to their perfect permeability and selectivity towards H_2 [18−20]. Significant improvement in membrane permeance has been achieved using composite membranes, i.e., thin layer of Pd alloys supported on porous materials such as ceramics or stainless steel [21,22]. Pd alloys (e.g. addition of rare earth elements such as silver, platinum, copper, or gold to palladium) are preferred over pure Pd to prevent H_2 embrittelment that takes place below or near 300°C for pure palladium and to increase the H_2 flux. Zhang et al.[23], found that air oxidation does not change the H_2 solubility in Pd-Ag membranes, but enhances the H_2 sorption kinetics significantly. Separation properties of the H_2-selective membranes have been tested using lab-made separation setups by different groups, e.g., Xomeritakis *et.al.*[24], Pizzi *et.al.*[25], and Catalano *et.al.*[26].

In this work an integrated module was developed comprising of a water electrochemical reactor and a membrane-based hydrogen purifier. The module was utilized to generate and purify hydrogen from sea water obtained from Gulf of Aqaba of Jordan in a totally green process, i.e., using renewable energy resources. The system was used to develop an empirical relation to correlate major electrolysis variables with the hydrogen production rate.
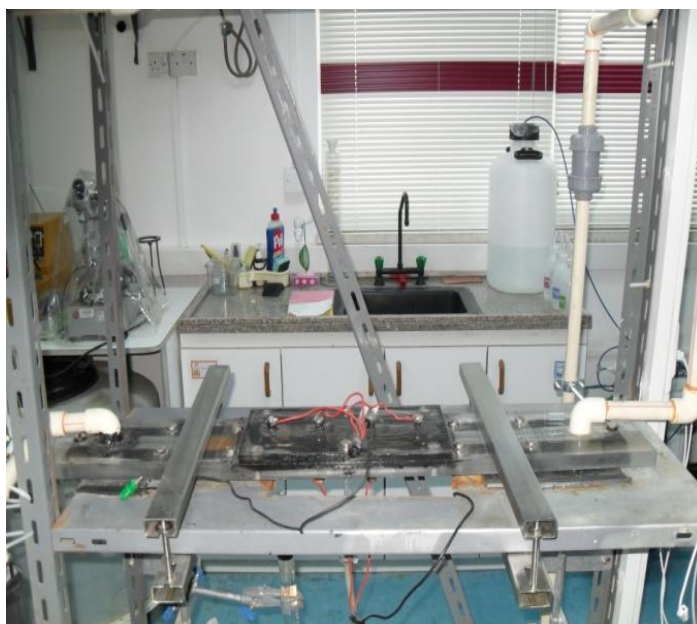
2. EXPERIMENTAL WORK

Figure 1. The electrochemical reactor used to conduct the experiments

The electrochemical reactor (ECR), shown in Figure (1), was connected to the electrochemical flow and data acquisition system as schematically illustrated in Figure (2). Four solar cells (i.e. PV) with an area of 3.3445 m^2 were connected with two batteries (24V D.C.) utilized to provide the ECR with the needed power. In each experiment, raw sea water was pumped into the flow system at a constant turbulent flow measured using a flow meter. A current limiter was used to set the voltage and current at the required values. A data acquisition system consisting of a data logger connected to a PC was used to measure different various parameters (currents, voltages, solar irradiance, temperature at the PV cells and ambient temperature) online every minute via the measurement devices. A bubble flow meter was utilized to measure the flow of pure Hydrogen gas at each voltage and current supplied by the current limiter. In order to obtain pure hydrogen free of oxygen, an oxygen trap containing a mixture of Pyrogalol and KOH was also used downstream the ECR followed by silica gel to dry the produced gas. The purity of hydrogen for each run was measured using gas chromatography (GC).
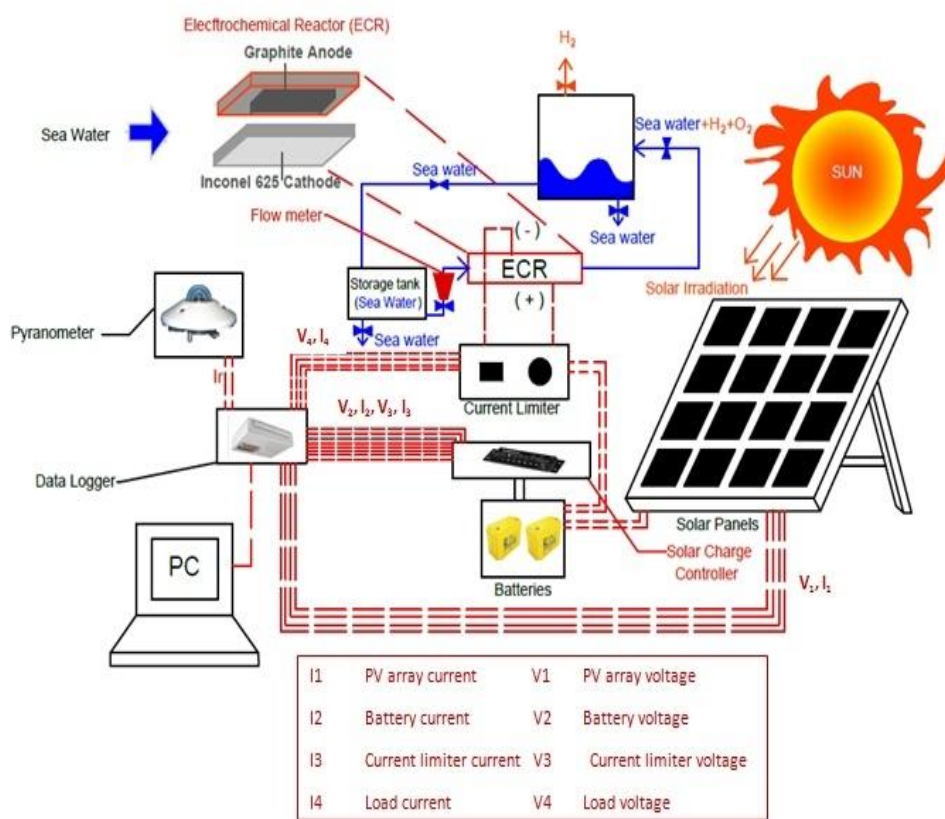

Figure 2. Schematic diagram shows the solar panel- ECR -electrochemical flow and data acquisition system

Further purification of Hydrogen close to 100% was done using a membrane-based Hydrogen Purification Unit (HPU) employing a Pd-Cu alloy metallic membrane which is known to be highly selective to hydrogen, see Figure (3). The HPU consists primarily of an upstream gas delivery section, a hydrogen-selective membrane, and a down-stream gas analysis section. Separation was conducted at

a high trans-membrane differential pressure (up to 250 psi) and high temperature (< 300°C). At these conditions, hydrogen is exclusively separated by the membrane according to the widely accepted solution-diffusion mechanism illustrated in Figure (4) in which the following steps take place:

1. Chemisorption of hydrogen molecules (H_2) from the gas phase onto the membrane surface.
2. Dissociation of the hydrogen molecules (H_2) into two hydrogen atoms (H).
3. Dissolution of the hydrogen atoms into the metal lattice.
4. Diffusion of the hydrogen atoms through the bulk metal.
5. Re-association of the hydrogen atoms into hydrogen molecules on the surface.
6. Desorption of the hydrogen molecules from the surface into the gas phase.

The H_2-rich stream product is called *Permeate*, and feed leftover is called *Retentate*. The downstream gas analysis section was used to analyze the gas content of the feed, permeate and retentate via a gas chromatography unit (GC).
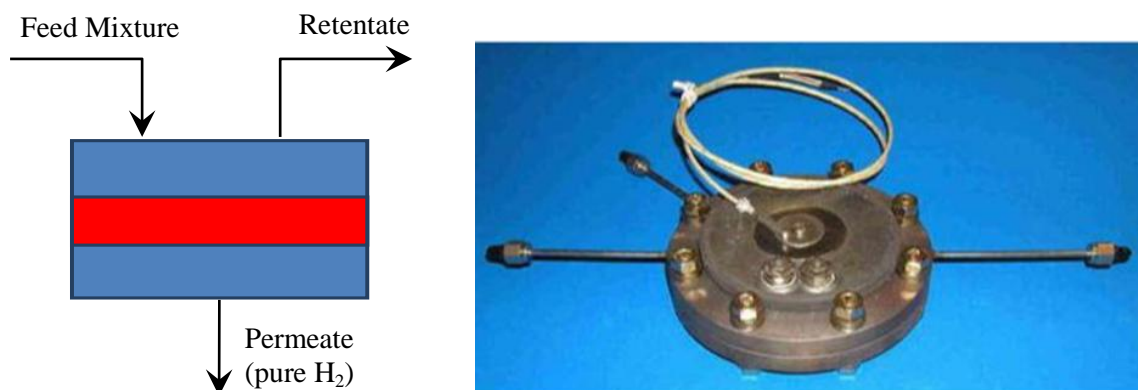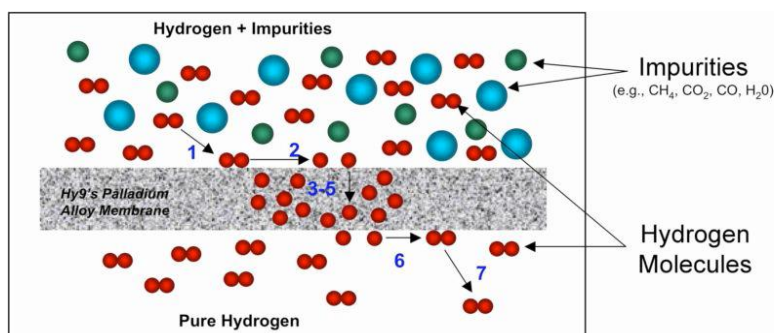

Figure 3. The hydrogen purification membrane

Figure 4. Hydrogen purification mechanism within Pd-Cu membranes

3. RESULTS AND DISCUSSION

Several parameters play a vital role in the production of hydrogen using the electrolyzer powered by solar energy, therefore a model correlating this effect would be very useful. A correlation representing the relationship between the production of hydrogen (Q) and current of the ECR, solar irradiation and solar panel temperature was obtained based on ANOVA multiple-regression analysis as follows:

$$Q = 158.0047 - 0.37203 \times Tc^{1.6} + 0.02923 \times Ir^{1.4} + 0.006901 \times I^2 \qquad \ldots \ldots (1)$$

with $R^2 = 99.9\%$ and $R^2_{adj} = 99.9\%$. This model was significant based on F and P tests. It was obvious that the production rate of hydrogen is proportional to the increase in the ECR power as shown in Figure (5).
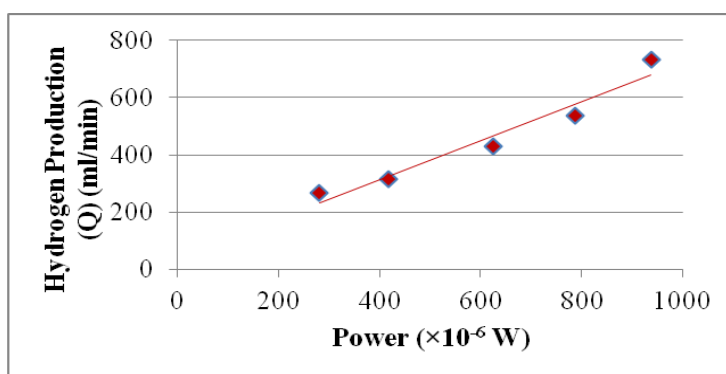

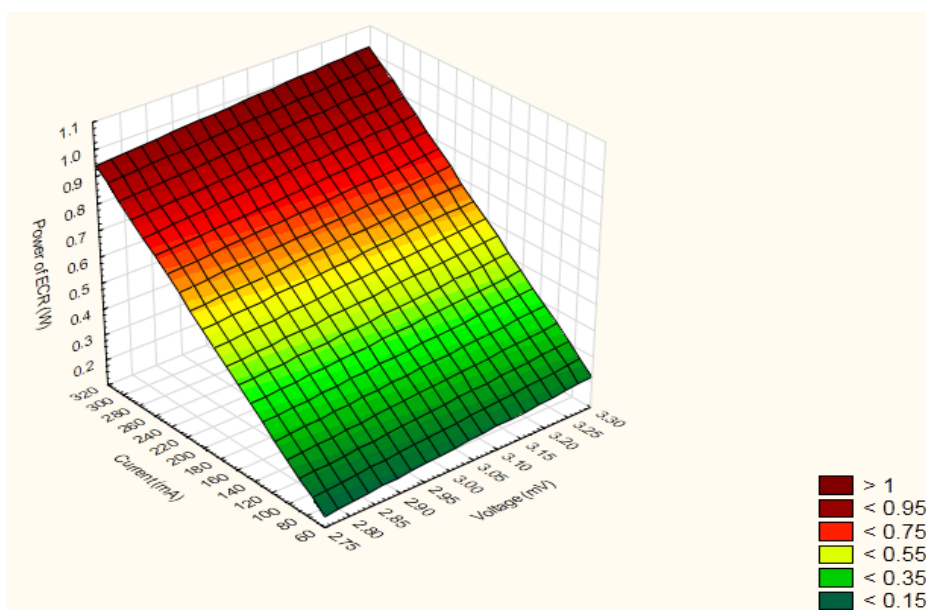**Figure 5.** Hydrogen production rate as a function of ECR power

Figure 6. Three dimensional mapping of ECR Power Vs. ECR current and potential

A three dimensional mapping using MATLAB shown in Figure (6) summarizing the effects of both the ECR current and voltage supplied by the batteries on the power consumption during electrolysis. It is obvious that ECR power is proportional to ECR current and potential which is in agreement with the studies of Balabel *et al*.[27] and Badea *et al.* [28].

Therefore it is obvious from Eq. (1) and Figure (6) that current and potential of the ECR, solar irradiation and panel temperature play an important role in the production of hydrogen. On the other hand, GC analysis showed that the purity of hydrogen in the gas produced by the electrolyzer is 99.913% as presented in Table 1.

Table 1. GC analysis of the hydrogen gas produced by the ECR using the solar cells (i.e.PV)

Dates	Gases %				
	O_2	N_2	CH_4	CO_2	H_2
10/May./2011	-	0.087	-	-	99.913

Table (2) summarizes experiments conducted to verify the effectiveness of the hydrogen purification unit using various mixtures of hydrogen and nitrogen.

Table 2. Details of the purified sample using Pd-Cu membrane

	Feed	Retentate	Permeate
Flowrate time	1.84	28.42	1.69
(sec/10 mL)	1.91	28.5	1.69
	1.85	28.58	1.69
Average time (sec)	1.867	28.50	1.69
Flow rate (mL/min)	321.43	21.05	355.03
Pressure (barg)	1.39	1.39	0
H_2%	99.909	57.146	100
Temperature(^{o}C)	300		

Stream compositions were tested by GC. The feed streams contain N_2 and H_2, thus the GC chromatogram yielded a peak for N_2. H_2 can be calculated by subtracting N_2 percentage from 100%. It was observed for all tests that N_2 content in the retentate is higher than that in the feed as shown in Figure(7). This is logical because H_2 in the feed is the only permeating component to the permeate side leaving most of N_2 in the retentate and thus its composition increase. In all cases the permeate stream was empty of any N_2 peaks indicating that it is composed entirely of H_2. A small N_2 peak can, however, is observed in the permeate chromatogram in Figure (7), this small peak is caused by the left-

over from a previous run. To overcome the leftover, all GC tests were done twice. The final permeate chromatogram of this permeate sample was completely clean of any N_2 peak.
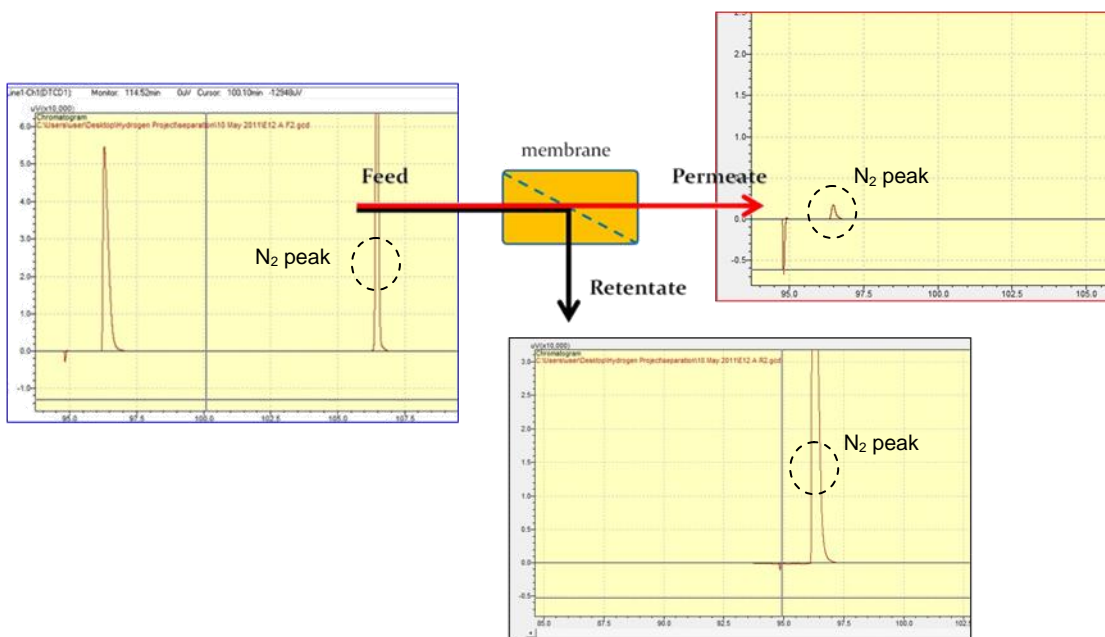

Figure 7. GC chromatograms for one of the samples

Percentage of hydrogen recovery from the feed mixture can vary roughly from 15 to 95% depending on the conditions as shown in table (3). Recovery values increases with the temperature of membrane and pressure difference. This is obvious because H_2 permeation through the membrane increases as the temperature and pressure difference increase. Corresponding recovery values for the sample name E12 are relatively low mainly because of the low content of H_2 in the feed and blockage of active sites of transport by the N_2 gas. This result is very near to the results obtained by Ryi et al. [29].

Table 3. H_2 recovery values from the feed

Sample Classification Name	Temperature (°C)	Feed H_2 content (%)	% Recovery
E2A	250	96.665	88.52
E2B	250	99.629	90.03
E2C	250	99.224	83.74
E3B	300	99.909	94.40
E3C	300	99.819	93.32
E12A	300	67.053	15.78
E12B	300	72.915	37.44
E12C	300	80.787	67.88

Based on the above results, the membrane efficiency in purifying hydrogen was verified and it is ready to receive actual feed from the reactor to run Proton Exchange Membrane Fuel Cell (PEM FC) as a last stage to retrieve the energy stored in hydrogen molecule and produce water which is considered as future work in this project.

4. CONCLUSIONS

The dependence of hydrogen production on the given variables (ECR current, solar irradiation and temperature of the solar panel) was obvious in the generated model. Three dimensional mapping showed that increasing electrolyzer voltage and current will increase the power consumption during electrolysis and increase the production rate of hydrogen. On the other hand the results revealed also that the membrane was able to separate and purify hydrogen from the feed mixture with infinite selectivity in the studied range of variables. The percentage of hydrogen recovery from the feed mixture can vary (15 to 95%) depending on the hydrogen in the feed and the temperature of the membrane.

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