



UNIVERSITI PUTRA MALAYSIA

***ENHANCEMENT OF EFFICIENCY OF WATER ELECTROLYSIS FOR
HYDROGEN PRODUCTION***

SEYEDKAVEH MAZLOOMI

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BERILMU BERBAKTI

**ENHANCEMENT OF EFFICIENCY OF WATER ELECTROLYSIS FOR
HYDROGEN PRODUCTION**

By

SEYEDKAVEH MAZLOOMI

**Thesis Submitted to the School of Graduate Studies, Universiti Putra
Malaysia, in Fulfilment of the Requirements for the Degree of Master of
Science**

March 2013

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Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the requirement for the degree of Master of Science

ENHANCEMENT OF EFFICIENCY OF WATER ELECTROLYSIS FOR HYDROGEN PRODUCTION

By

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March 2013

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In this thesis, after analyzing the properties of hydrogen in analogy to a number of common fuels, its productions methods and advantages of water electrolysis are introduced briefly. The frequency response of a water electrolysis cell is analyzed in order to examine the possibilities of reaching higher production efficiencies in other methods of power application than steady DC mode. Conductance of such cells was observed to reach a maximum value at a certain frequency depending on the physical cell characteristics. As a result, a series of experiments were continued in order to examine the possibility of enhancing the process efficiency.

Laboratory tests were performed on a cell in different voltages ranging between 2 V and 10 V, frequencies ranging between DC and 20 MHz and duty cycles varying in the range of 10% to 100%. The efficiency value was recorded for each case and the results were compared with those of DC mode. The

gathered data shows water electrolysis process will be more efficient when the power is applied to the cell in the form of pulses. According to the experimental results, an efficiency enhancement of up to 14% can be achieved by applying short pulses to the system instead of a DC voltage. The result of the latter is prevention of the formation of retarding phenomena such as electrical double layers and diffusion layer in the vicinity of the electrodes. However, the production rate decreases in pulsating power application since the total amount of energy application to an electrolytic bath is remarkably reduced in pulsating voltage application mode. In other words, the time requirement of producing the same amount of hydrogen can be longer as much as near 3 times of that of low efficiency process.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia
sebagai memenuhi keperluan untuk ijazah Master Sains

**PENINGKATAN KECEKAPAN ELEKTROLISIS AIR UNTUK
PENGELUARAN HIDROGEN**

Oleh

SEYEDKAVEH MAZLOOMI

Mac 2013

Pengerusi: Nasri Bin Sulaiman, PhD

Fakulti: Kejuruteraan

Dalam tesis ini, selepas menganalisis ciri-ciri hidrogen dalam analogi kepada beberapa bahan api biasa, kaedah pengeluaran dan kelebihan elektrolisis air diperkenalkan secara ringkas. Sambutan frekuensi sel elektrolisis air dianalisis untuk mengkaji kemungkinan mencapai kecekapan pengeluaran yang lebih tinggi dalam kaedah lain yang diterima pakai kuasa daripada mod DC yang stabil. Kealiran sel-sel itu telah diperhatikan untuk mencapai nilai maksimum pada frekuensi tertentu bergantung kepada ciri-ciri sel fizikal. Hasilnya, satu siri eksperimen diteruskan untuk mengkaji kemungkinan meningkatkan kecekapan proses.

Ujian makmal telah dijalankan pada sel voltan yang berbeza antara 2 V dan 10 V, kekerapan antara DC dan 20 MHz dan kitaran tugas yang berbeza-beza dalam lingkungan 10% hingga 100%. Nilai kecekapan dicatatkan bagi setiap

kes dan keputusan telah dibandingkan dengan mod DC. Data yang dikumpul menunjukkan proses elektrolisis air akan menjadi lebih cekap apabila kuasa yang digunakan untuk sel dalam bentuk denyutan. Menurut hasil kajian, peningkatan kecekapan sehingga 14% boleh dicapai dengan menggunakan denyutan pendek kepada sistem dan bukannya voltan DC. Keputusan kedua adalah pencegahan pembentukan fenomena memperlakan seperti lapisan berganda elektrik dan lapisan penyebaran di sekitar elektrod. Walau bagaimanapun, kadar pengeluaran berkurangan dalam permohonan kuasa denyut kerana jumlah permohonan tenaga kepada mandi elektrolisis adalah amat berkurangan dalam mod permohonan voltan denyut. Dalam erti kata lain, keperluan masa untuk menghasilkan jumlah yang sama hidrogen boleh lagi sebanyak berhampiran 3 kali bahawa proses kecekapan rendah.

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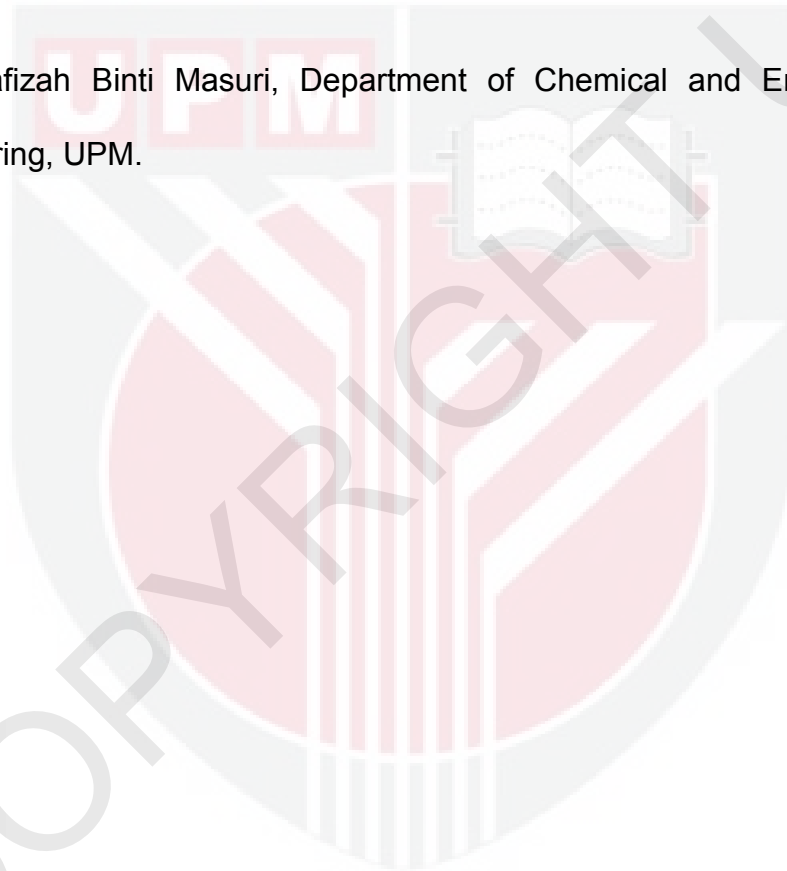
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DECLARATION

I declare that the thesis is my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously, and is not concurrently, submitted for any other degree at Universiti Putra Malaysia or at any other institution.



SEYEDKAVEH MAZLOOMI

Date: 14 March 2013

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LIST OF ABBREVIATIONS

AC	Alternative Current
ADC	Analog to Digital Converter
Al	Aluminum
atm	Atmosphere
BJT	Bipolar Junction Transistor
BMI.BF4	1-butyl-3-methyl-imidazolium-tetrafluoroborate
CAES	Compressed Air Energy Storage
CCM	Cubic Centimeter per minute
Co	Cobalt
Cox	Carbon Oxides
CS	Carbon Steel
D	Duty Cycle
DC	Direct Current
e	Electron
η	Efficiency
ϵ_0	Vacuum Permittivity
ϵ_r	Relative Static Permittivity
EMI	Electro Magnetic Interference
ESL	Equivalent Series Inductance
ESR	Equivalent Series Resistance
FC	Fuel Cell
FFT	Fast Fourier Transform
GC	Gas Chromatograph

HHV	Higher Heat Value
HFL	Higher Flammability Value
HPE-ALWR	High Pressure Electrolysis-Advance Light Water Reactor
HTGR	High Temperature Gas cooled Reactor
HTSE	High Temperature Steam Electrolyzer
Hz	Hertz
ICE	Internal Combustion Engine
IEA	International Energy Agency
IGBT	Insulated Gate Bipolar Transistor
Ir	Iridium
K	Kelvin
kHz	Kilohertz
KOH	Potassium Hydroxide
LCD	Liquid Crystal Display
LHV	Lower Heat Value
LFL	Lower Flammability Level
M	Mole
MHz	Megahertz
Mo	Molybdenum
MOSFET	Metal Oxide Semiconductor Field Effect Transistor

CHAPTER 1

INTRODUCTION

Global warming and energy crisis are among the most important issues that threaten the peaceful existence of the man-kind. They have been showing their faces much more clearly in the past century and no concrete solution is introduced in order to curb their ill-effects on the planet. Many different approaches are under experimental investigations or being utilized in this regard. However, adopting clean and emission-free energy cycle is known to be a major break-through in this regard. Utilization of renewable and sustainable energy sources is a promising solution for the mentioned problems. However, one of the major deficiencies of the use of such resources is the method of energy storage.

Utilization of powerful batteries, heat storage, compressed air, pumped storage and similar technologies have been studied in this regard. Some of the available methods are absolutely practical in order to store the surplus energy production of renewable plants. Yet, one of the most challenging subjects according to the usage of such sources is how man is able to transport or transmit the energy. At the same time, transportation is known to have a large share in polluting the environment. Developing a new fuel economy seems to be one of the most important sectors in accordance with achieving an absolutely clean and “green” energy cycle.

1.1 Reasons of looking for alternative energy carriers

The idea of using alternative fuels was strengthened noticeably after the global energy crisis of 1974 [1, 2]. According to the published statistics by the International Energy Agency (IEA) currently, the world consumes fossil fuels in very large scales (over 89 million barrels per day). A wide and really enhanced global scientific, social and political infrastructure supports this popularity. A wide and really enhanced global scientific, social and political infrastructure supports this popularity. This consumption level does not come without problems indeed. Pollutant emission of harmful materials [3-5] and greenhouse gasses [6] accompanied with current global warming issues [7] are only few of their disadvantages. These fuels have limited exhaustive resources [8] and they can be found in certain parts of the planet.

Furthermore, political conflicts, mainly caused by their highly volatile price [9, 10] is a distinct drawback that definitely threatening the existence of the human-race. In addition, these fuels are oil derivatives with a wide range of formulations where each can be fed to a certain and limited group of consumer machinery. These fuels are basically being “burnt” in order to release their energy content, which causes a large fraction of it to release to the atmosphere as heat-waste in the process of combustion [11, 12].

1.2 Available solutions

Since the age of water mills, man was trying to control the energy content of the environment. Many methods have been and are being used in this

relevance. Figure 1.1 is an illustration of some of the modern methods of energy storage. Many of the mentioned approaches are practical when a certain scale of energy storage is the subject of debate. Some require a vast area for being developed where others are economical if and only if mega scale storage is required. A few have a very low energy storage capacity or limited life spans.

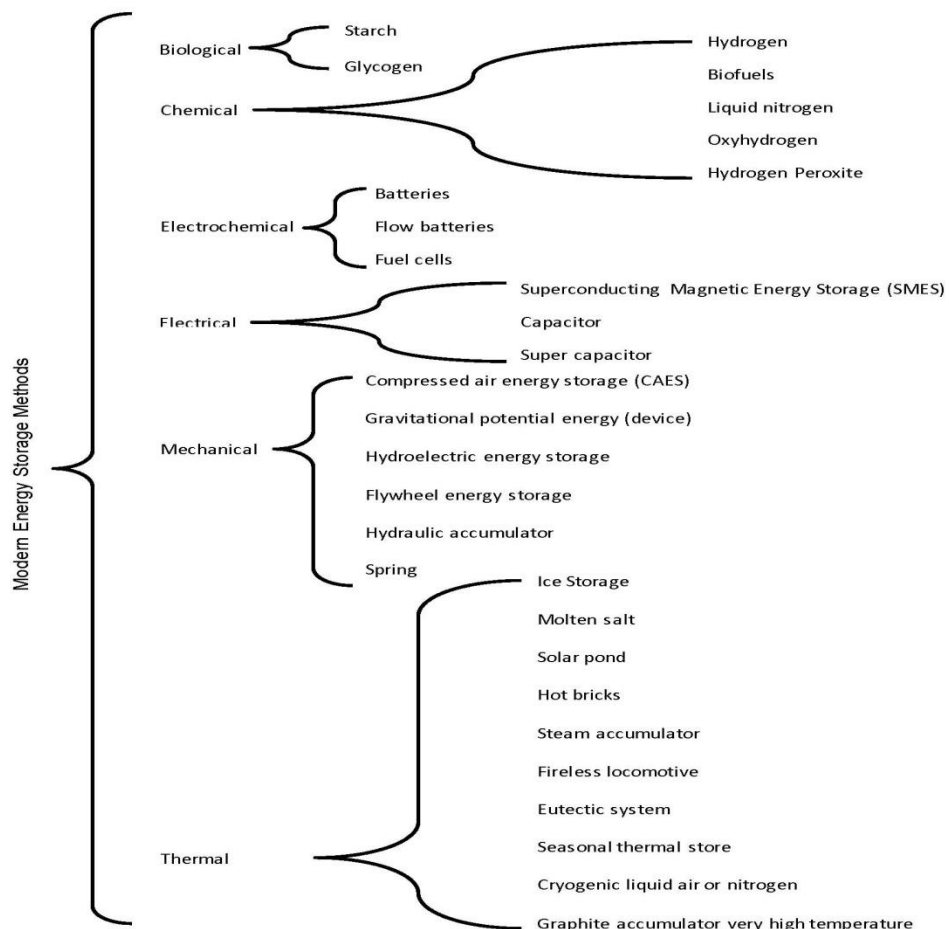


Figure 1.1: Modern methods of energy storage

Here are a few instances of the above-mentioned:

1- Hydroelectric energy storage requires building of dams and as a result artificial lakes.

2- Twisting a spring makes us able to store enough power to energize small scale consumers such as toys or watches.

3- Even the best quality batteries have a limited life span. Their efficiency decrease as they age and they need a remarkable time for being charged. Storing large amounts of energy into batteries usually require industrial sophisticated electronically-controlled chargers, voltage regulators and current limiters.

4- Biological energy storage in industrial scales requires large pieces of land.

In addition, most of the mentioned methods in Figure 1.1 are unable to be utilized for producing fuels or transportable energy carriers. A promising fuel or energy carrier has to have reasonable price, the ability of being produced and consumed in an absolutely clean and emission free cycle, transportable nature, high energy per mass/volume content and acceptable safety features.

1.3 Benefits of using hydrogen

Outstanding properties and features of hydrogen make it a very promising energy carrier or fuel, although it is not naturally available as a ready to use substance. Different methods are being used in order to mass produce hydrogen. According to its abundance, hydrogen can be extracted from a

variety of materials and compounds or be produced by utilizing a wide range of methods including some clean and “green” approaches. More importantly, hydrogen can be produced anywhere across the planet. Exceptional energy per mass content, storage and transportation possibilities, safety features and reduced harmful emissions are few advantages of this substance as an energy carrier.

For many researchers that investigate the applications of hydrogen as an energy carrier or fuel require readily extractable broad-spectrum knowledge on various processes involved in this regard including their pros and cons and possible modifications that make the processes suitable for future development. Such literature is rare to be found as many research papers address narrowly focused aspects of the subject.

1.4 Methods of production

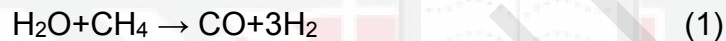
A range of methods is in use to generate hydrogen from different resources. Unfortunately, fossil based fuels are still the main recourse for industrial mass scale hydrogen production probably due to their low costs and easy usage in machines that designed for fossil fuels. This fact is absolutely in contradiction with policies towards a green and sustainable energy cycle.

1.4.1 Hydrogen from fossil fuels

Fossil fuels have large and heavy hydro-carbon based molecular structure. Extracting hydrogen by breaking the bonds between hydrogen and carbon

content is one of the most popular methods of hydrogen production [13]. This substance can be extracted from biomass [14], coal [15], gasoline, oil (heavy and light), methanol and methane [16].

Nowadays, Steam-Methane Reforming (SMR) is known as the most economical method [17] and has the largest share in global hydrogen production (almost 48%) [18]. The reaction of this highly endothermic process is given by Equation 1:



Coal and oil have the second and third place in this ranking with 30% and 18% relative share [17]. Hydrogen production by the means of water electrolysis has the smallest share of 4% among the available methods of large scale hydrogen production [19] where other resources are not being used in mass and industrial scales. Figure 1.2 shows the global share of each method of industrial-scale hydrogen production.

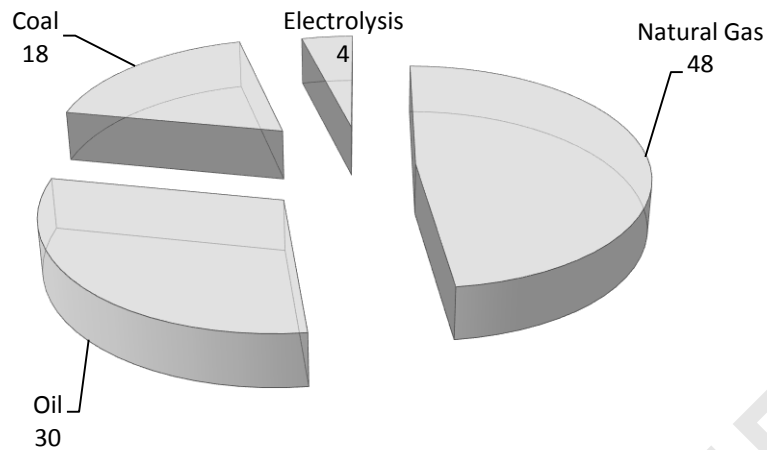


Figure 1.2: Common methods of hydrogen mass production

Hydrogen is a byproduct of other oil refinement in many cases. Reasonable production price [20] and possibility of mass production [16] are other Advantages of fossil based hydrogen production. However, this approach of hydrogen production suffers from problems which are mainly based on their pollution ratings and limited resources. These methods of hydrogen production usually emit CO or CO₂ and other greenhouse gasses. The resources are not renewable [13] and the production is not known as “green”.

1.4.2 Hydrogen from water

Splitting of water molecule by means of electrolysis has been studied for a long time [21, 22]. Water is subjected to an electric current in order to force its molecules to decompose [19]. A typical schematic of an electrolysis cell is illustrated in Figure 1.3.

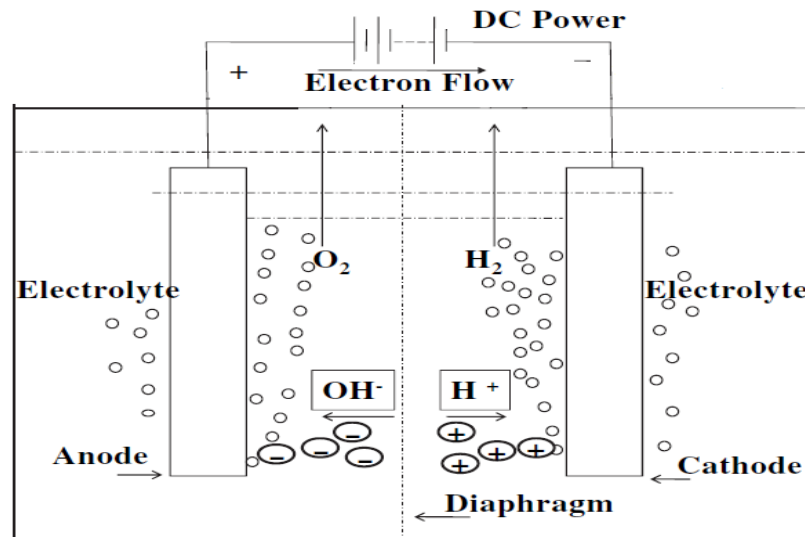


Figure 1.3: Structure of an electrolysis cell

A simple water electrolysis cell consists of a container, at least two electrodes, an electrolyte material and in some cases a separator plate which is also called the diaphragm. Most common electrolyte materials are ionized aqueous solutions. Since pure water is not an electricity conductor, an ionizing material is usually being added to the water in order to make it conduct the electrical current.

The metallic electrode plates are placed in the electrolyte where a power supply provides the required energy for the current to pass through the cell. Some water electrolysis cells have more than two electrodes. In this case, the voltage is applied to the first and last plates, where each side of each plate will perform as either anode or cathode for the facing sides of the neighbor electrodes (refer Figure 4.12). These cells require higher voltage levels indeed since the voltage will divide between compartments.

As it was mentioned earlier, this method does not have a large share in global hydrogen production. High production costs [23] due to low conversion efficiency and electrical power expenses [24-26] can be named as the main drawbacks of electrochemical hydrogen production. Hence, water electrolysis is not a method of choice for large-scale production of this substance, in the present context. As a result, electrolytic hydrogen was not able to find its way as a competitive alternative for traditional fuels.

Water electrolysis process requires a minimum energy of 39.39 kWh kg⁻¹ of hydrogen generation at full conversion efficiency. However, the energy demand per kg of hydrogen production of a typical high capacity industrial scale electrolyzer is much larger than the mentioned Figure [27, 28]. Many efforts are made in order to increase the efficiency of water electrolysis [23, 27, 29-31]. Higher efficiencies were obtained in extreme pressure and temperature conditions. At the same time, increased investment is required to build more complex and sophisticated electrolyzers which are able to perform under intense temperature and pressure conditions [32-34]. In these cases, higher production efficiency comes with dramatically increased corrosion, operation and maintenance (O&M) costs and reduced life span [1, 23]. On the other hand, estimations show that the monetary investment per production capacity unit reduces as the capacity increases [16]. Therefore, most of the available electrolyzers work at temperatures lower than the boiling point of water and do not exceed the pressure barrier of 50 bar.

Despite the mentioned cost disadvantages, water electrolysis has some unique qualities. Electrolysis could be used for hydrogen production at any place around the globe. The only requirements of this production are electricity and water where the production rate/capacity could be tuned for a certain demand at any place [13, 16]. With regard to the characteristics of water electrolysis, this method is capable of producing absolutely sustainable and clean hydrogen. This goal can be achieved if and only if the required electricity is obtained from an emission-free method such as wind, solar, geothermal systems, ocean wave or other renewable and green sources. The latter is further supported by the fact that such energy generating systems can be developed 8 times faster than those with oil-base fuels [12]. Whereas their net energy profile shows very close overall values for both methods over time, there are still some lifespan advantages for the case of renewable approaches.

Every single renewable energy harvesting system has its own capital cost. Utilizing one, all or a combination of few of the new energy production systems is inevitable for future energy production demands [13, 24]. However, current concern is to analyze the possibilities of hydrogen production based on the available social, industrial and political infrastructures. Schoots *et al.* [20] calculated the required investment cost as 1000 US\$ kWh⁻¹ for nominal power of the hydrogen production plant. Referring to the mentioned power demand of hydrogen generation, the estimation is that a plant requires an investment of 50,000 US\$ for each 1kg h⁻¹ capacity of electrolytic hydrogen production.

On the other hand, evaluations show remarkable reductions of expense as the production capacity increases [16].

1.5 Problem Statement

Water electrolysis is not as efficient as widely used methods of hydrogen mass production. Therefore, production price by this method is relatively higher than other approaches. In addition, although water electrolysis is an electricity dependent process, literature about the effect of electric power application methods on the process efficiency are very rare to find.

Electrolytic hydrogen generation is mainly practiced under the influence of a DC voltage. Therefore, by knowing the electrical characteristics and behavior of a water electrolysis cell, possibility of the utilization of a pulsating power application method for enhanced process efficiency can be examined. The main goal is to conduct experiments with different methods of voltage application to minimize cell impedance and reach higher conductance levels in a water electrolysis bath. The latter points will lead to less power dissipation and higher production efficiency.

1.6 Aim and objectives

1- To introduce a simplified electrical equivalent circuit for a water electrolysis cell.

2- To propose a method of power application for enhancing the efficiency of hydrogen production by the means of water electrolysis.

3- To propose a container shape for electrically efficient water electrolysis.

1.7 Research scope and limitations

In order to introduce an electrical equivalent circuit for a water electrolysis cell, its frequency response is analyzed. Based on experimental work, the conductance bode diagrams of a series of electrolysis cells were plotted. By knowing the order of the transfer function of such cells over a certain frequency spectrum, and regarding the resemblance of their physical structure with electrolytic capacitors, a simplified equivalent circuit is introduced.

Regarding the introduced equivalent circuit, and based on the available published material, a method of voltage application to an electrolysis cell is proposed. Finally, a series of experiments were conducted to evaluate the effect of the proposed approach on the efficiency and hydrogen production rate of a number of electrolysis cell setups.

Maximum applied frequency to the experimental electrolysis cells was limited due to the spectral limitations of the utilized electrical function generator and maximum switching speed of the available power transistors.

Because of the lack of constant access to a gas chromatography unit, humidity, temperature, pressure and flow rate sensors were used to measure

the amount of produced hydrogen. Samples were taken for being tested by a gas chromatograph in order to validate the measurements and calculations. Hydrogen concentration was measured by the chromatograph unit for mixtures produced at calculated higher efficiencies.

1.8 Thesis organization

Section 1 of this thesis introduces the reasons of interest in hydrogen as an energy carrier. Characteristics of this substance are compared with those of a number of common fuels in this regards. In this section, different methods of hydrogen production are introduced as well as the benefits of electrolytic hydrogen production.

In the literature review section (section 2), the information gathered from several related published papers and textbooks. After the discussion about the influencing factors on the electrical efficiency of water electrolysis, different methods of power application to such systems are analyzed. In addition to the above-mentioned, common switching devices are compared in order to select a proper device of such kind for the experimental work.

The experimental cell setup, sensors, electronics and test conditions are introduced in section 3 to provide an insight into the experimental work.

The results are analyzed in section 4 in order to introduce an electrical equivalent circuit and evaluate the possibilities of energy saving by the utilized

method of voltage application. Finally, the concepts of the designed container are the subject of discussion in this section.

Research conclusions are briefly reported in section 5 in addition to the introduction of possible subjects for further research in the area of this thesis.



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