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FUEL CELLS: CONTAMINATION AND RECENT ADVANCEMENTS FOR ITS STABILIZATION

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Abstract

A Fuel cell will soon be a very robust weapon to replace current highly carbonized fossil fuel generation of electricity by a more cleaner and efficient source of generation. The kind of electricity generation that is being used nowadays impacts environment badly and contributes to Global Warming. In this context, Fuel Cell can prove to be significant in generation of cleaner energy. In addition to this, fuel cell is more efficient as it doesn't operate as any heat engine; so not bound up to Carnot efficiency. Certain impurities either from the fuel that is being supplied or from the environment might result into undesirable reactions that cause fuel cell contamination. This becomes the reason for the dramatic drop in the performance of a fuel cell that has been observed from our working "Solar Hydrogen Plant"^{Fig A} model. For testing and extending the life of a fuel cell by detecting and mitigating the cause of degradation, the researchers has passed the sample of distilled water and Ultra-pure water (Type I) after electrolysis through Ion Chromatographer, Total Dissolved

Solid test, Electrical Conductivity Test, and pH test. The cell has been allowed to run in the lush green campus of Banaras Hindu University whose environmental conditions are considered to be less polluted in the Varanasi. Henceforth, the researchers also tested atmospheric situations and its effect on fuel cell. The results of this paper will be useful in designing a device which can mitigate contamination and will ensure successful domestication of Fuel cell. In this paper, various methods of obtaining pure hydrogen gas like Pressure Swing Adsorption (PSA), High & Low Temperature Diffusion, Solvent absorption of CO and CO₂ are also discussed. A new model of fuel cell installation is also discussed with an additional stage of hydrogen fuel filtration using PSA, TSA to supply hydrogen fuel with purity up to 99.8%.

Keywords

Fuel Poisoning, PSA, Reformation, Contamination Mitigation

1. Introduction

With incipiently developed technology which uses electrical input, we cannot deny the over exploitation of fossil fuel to meet the growing electricity demand. A Proton-Exchange Membrane or Polymer electrolyte membrane fuel cells, has its properties of being able to produce a cleaner electrical energy with a very good efficiency. Platinum is a relatively inactive metal but the unusual property of being able to adsorb hydrogen gas at moderate temperature creates its great significance in fuel cells. Hydrogen Fuel cell has withal its great paramountcy in space shuttle operation as it engenders di-hydrogen monoxide i.e. water as a byproduct which makes astronaut be able to survive in space. Fuel cells do not operate utilizing a thermal cycle, precisely not be working as a heat engine so it's not bound to Carnot efficiency and hence can show very high conversion efficiency in the range of 40%-60%.

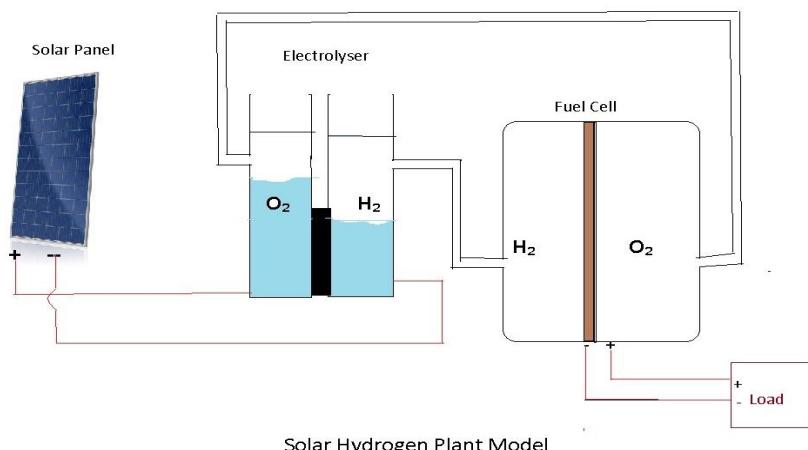


Figure 1: A Source: Self

However, for successful commercial and domestic use the discussion must not be only limited to catalytic reactions only. Instead, more effort is required to improve its stability and to decrease its cost in the market (Cao, M., Wu, D. and Cao, R. (2014)). The contamination of fuel cell causes a dramatic drop in its performance. So, we'll be fixating on the techniques to amend its stability when exposed to work for long-time or when installed as a commercial source of electricity generation.

2. Hydrogen Fuel Cell

Like the battery, the fuel cell is a simple device, consisting of only four fundamental components that support electrochemical reactions. They are as follows:

1. An electrically negative electrode called the anode to which a fuel (Hydrogen gas) is supplied;
2. An electrically positive electrode called the cathode to which an oxidant (oxygen) is supplied.
3. Polymer Electrolyte Membrane i.e. PEM (a substance that sanctions movements of electrically conductive ions) that dissects the two electrodes;
4. An external electric circuit connecting two electrodes

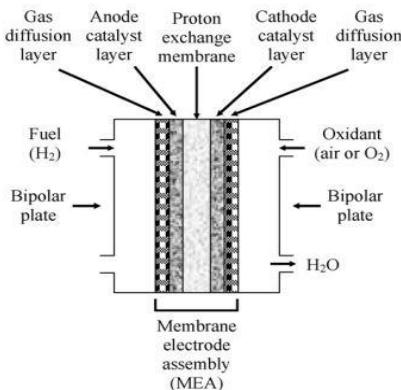


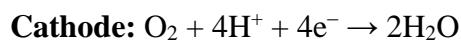
Fig. Basic components of proton exchange membrane fuel cell.

Figure 2: Fuel Cell

Source: *Journal of Power Sources 165 (2007) 739–756*

2.1 Working

The electricity is transmitted to the external circuit because of certain electrochemical reactions. In this reaction, the electron is generated at anode and then it peregrinates through an external circuit producing current and then consumed at the cathode. Within the cell, ions flow utilizing semi-permeable membrane i.e. PEM. Following reactions undergo during the operation of hydrogen fuel cell:



Anode: $2\text{H}_2 \rightarrow 4\text{H}^+ + 4\text{e}^-$

The hydrogen ions -- protons-- are transported by an ion-conducting membrane (the proton-exchange membrane) to opposite electrode, where they combine with oxygen from atmosphere and the electrons returning from the load to form water (DeLuchi, M. A. Ogden, J.M.).

3. Production of Hydrogen Gas

It is well known that there is abundance of hydrogen element in Earth's crust therefore; hydrogen gas can be produced from various sources such as natural gas, coal, hydrocarbons and by many other chemical reactions. Hydrogen gas can also be produced by splitting water by electricity which is called as electrolysis. Here, in our experiment researchers are using electrolyser for electrolysis of distilled water. The commercial production of hydrogen by electrolysis of water achieves an efficiency of 70–75%. However, the cost of hydrogen produced by this route is several times higher than that produced from fossil fuels (J P.P. Edwards, V.L. Kuznetsov, W.I.F. David). At current stage of technology, reformation of hydrocarbons including methane from natural gas is the most significant sources for the production of hydrogen gas. This method bounds us to install a purifying stage consisting of PSA, High & Low temperature diffusion since, reformation produces impurities like CO, CO₂ and H₂S along with the production of hydrogen gas.

Steam reforming and partial oxidation or auto thermal reforming are usually used to produce hydrogen-rich gas called “reformate,” which typically contains 40 to 70% H₂, 15 to 25% CO₂, 1 to 2% CO, small quantities of inert gases (water vapor and nitrogen), and sulfur impurities (Xuan Cheng, Zheng Shi, et al).

4. Methods to Obtain Pure H₂ Fuel

Because of wide range of use of hydrogen scientists have developed many techniques which can be used to obtain pure hydrogen gas. Here, we'll discuss methods to draw pure hydrogen gas which can be used to supply pure hydrogen gas to the fuel cell. Henceforth pure H₂ supply will cause minimal poisoning of fuel cell.

4.1 Pressure Swing Adsorption

Pressure swing adsorption (PSA) processes are used for the production of high purity hydrogen from steam methane reforming off-gas (SMROG) and refinery off gases (ROG)(S. SIRCAR & T. C. GOLDEN (2000)). Pressure swing adsorption systems for oxygen production were first used

in the 1970s (N.M. Prosser, M.M. Shah, 10). This technology for the separation of gasses utilizes the fact that under high pressure, gases tend to be attracted to solid surfaces, or "adsorbed". The higher the pressure, the more gas is adsorbed. When the pressure is reduced, the gas is released, or desorbed.

PSA method uses different adsorbents which have the ability to differentiate mixture of gasses. Examples of few adsorbents used in PSA are activated carbon, silica gel, alumina, resin and zeolite. As the result of the experiments done by Frederico Relvas, Roger D. Whitley, Carlos Manuel Silva, and Adélio Mendes characterizes a copper loading of 2 mmol·gads⁻¹ as adsorbent for the effective removal of CO as well as other impurities. A product stream with +99.97% hydrogen was obtained, with 76.19 % recovery and a carbon monoxide concentration of 0.17 ppm (Frederico Relvas, Roger D. Whitley, et al).

4.2 High & Low Temperature Diffusion

In this technique, the raw gas is diffused through a metal membrane such as palladium. In the 1980's Johnson-Matthey sold hydrogen generators based on steam-reforming methanol which used a palladium diffuser to provide very high purity hydrogen for various specialized uses (J. E. Philpott(1985)). High cost and limited supply of palladium are the major disadvantages of this system. Nowadays, nickel membranes have been tested as a possible substitute for palladium (Y. Sakamoto, H. Tanaka, et al).

In case of low temperature diffusion the gas has to diffuse through polymer membrane. The setup is too much expensive for commercial use. For its successful installation for domestic purpose many ongoing projects are going to reduce its cost. Membrane diffusion units which should be suitable for H₂/CO separations are available from Monsanto Co. (U.S.A.), DuPont Co. (U.S.A.) and Ube Industries Ltd. (Japan) (J.C. Amphlett, R.F. Mann, et al).

4.3 Solvent Absorption of CO₂ and CO

This method is mainly used in chemical industries to remove impurities from mixture of gasses. The basic step is to dissolve unwanted impurities to solvent and hence high purity hydrogen gas can be extracted as a product. Chemical absorption systems have been in use since the 1930s for the capture of CO₂ from ammonia plants for use in food applications and hence, are a commercially realized technology, though not at the scale required for power plants (Anusha Kothandaraman(2006)).

5. Electrode Contamination & its Mitigation

The contamination can be because of fuel impurities such as CO, CO₂ or NH₃; air pollutants (NO_x, SO_x, CO, and CO₂); and few cationic ions like Fe³⁺ and Cu²⁺ (Xuan Cheng, Zheng Shi, et al).

5.1 CO Contamination

CO binds with the Platinum surface intensely which causes decrement in surface area available for hydrogen adsorption and oxidation. So, it's obvious to reach out to conclusion that CO poisoning on Pt catalyst will increase, with increase in exposure time. Nowadays, various techniques such as current–voltage polarization and EIS (Electrochemical Impedance Spectroscopy) (Prudenziati, M., & Hormadaly, J. (2016)) can be used to quantify the effects of CO poisoning on H₂ oxidation. Benesch and Jacksier (R. Benesch and T. Jacksier) reported that the time it took for cell voltages to decay to a threshold value of 0.3 V were 1 and 9 h, respectively, when the cells were exposed to 50 and 10 ppm of CO. However, utmost care should be taken while using EIS techniques as stated in recently published paper (J.-M. Le Canut, R.M. Abouatallah, et al) that one can misinterpret fuel cell flooding with CO poisoning.

Moreover, temperature has also its effects on performance of a fuel cell. When the anode was fed with H₂ containing 250 ppm of CO, the performance at temperatures below 80°C was much lower than that at 100°C (Divisek, J., Oetjen, et al (1998)).

5.1.1 Mitigating CO Poisoning

There exist three methods to mitigate the effect of CO poisoning: (i) the use of a platinum alloy catalyst, (ii) higher cell operating temperature and (iii) introduction of oxygen into the fuel gas flow (A. Rodrigues, J. C. Amphlett, et al). Analysis of cited publication shows that a pure Pt catalyst experiences more CO poisoning than a Pt-alloy catalyst. Ralph and Hogarth confirmed that a PtRu alloy catalyst is more tolerant to CO poisoning than a pure Pt Electrocatalysts by experiments using a fuel cell operated at 80°C with hydrogen containing 10, 40, and 100 ppm CO (T.R. Ralph, M.P. Hogarth).

Air-bleed on the anode side of the stack is a well-known technique for mitigating CO poisoning (Gottesfeld 1988) (Johannes Tjønnås, Federico Zenith, et al). Unconverted carbon monoxide from the reformer can significantly reduce the activity of the fuel cell's anodic catalyst. For this, corrective action is to increase the air bleed to oxidise CO to CO₂ (Johannes Tjønnås, Federico Zenith, et al).

5.2 Air Contaminants

Fuel cell is also highly sensitive towards poisoning caused by air impurities. Few effects caused by air impurities (SO₂, CO₂, NO₂ etc) are reversible, and some are non-reversible. Though, reversible poisoning can be eliminated but certainly it has been observed that long time exposure to

it degrades the fuel cell permanently. In the presence of SO₂ in the air stream, the fuel cell current density dropped by over 50% (R. Mohtadi, W. Lee, J.W. Van Zee).

The performance degradation with NOx present in the air stream was also observed by a recent electrochemical impedance measurement (D. Yang, J. Ma, et al). However, the performance loss induced by the presence of NO₂ could be recovered by reintroducing neat air into the contaminated cathode (ZhengShi, D.S., HuiLi, KhalidF., et al).

6. Experiments

For testing and extending the life of a fuel cell, the researchers carried out electrolysis by both Distilled water and Ultrapure water (Type I).

6.1 Ion Chromatography using Ultrapure Water

Ion chromatography is a process that separates ions and polar molecules based on their affinity to the ion exchanger. Solar Hydrogen Plant^a is allowed to operate in the open-environment of Banaras Hindu University. Volume of fuel supplied and time is noted, and then sample is tested for ions by chromatography. The samples are taken after electrolysis of 24mins.

Instrument Specification

Instrument name: Ion chromatography Metrohm (930 compact IC, Switzerland)

Column used: Metrosep C4 (for cation) Metrosep A Supp.5 (for anion)

Table 1: Sample Specification

Sample Name	Volume of Sample Taken	Time of Electrolysis	Environment of Electrolysis
O ₂	45ml	24mins	Open Atmosphere (BHU)
H ₂	45ml	24mins	Open Atmosphere (BHU)

In this experiment, the samples are taken after an electrolysis of ultrapure water for 24mins. After electrolysis, the sample of both H₂ and O₂ chamber are taken separately and stored in a closed tube of 45ml and then sent to CIF BHU for Ion chromatography.

Sample O₂ for Anions

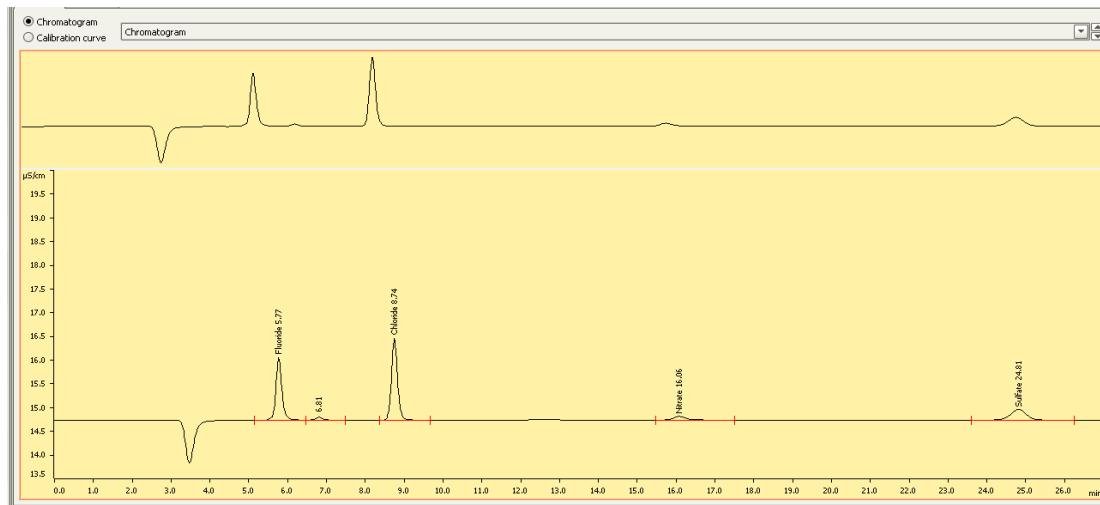


Figure 3: Ion Chromatography Result
Source: Central Instrument Faculty (BHU)

Sample O₂ for Cations

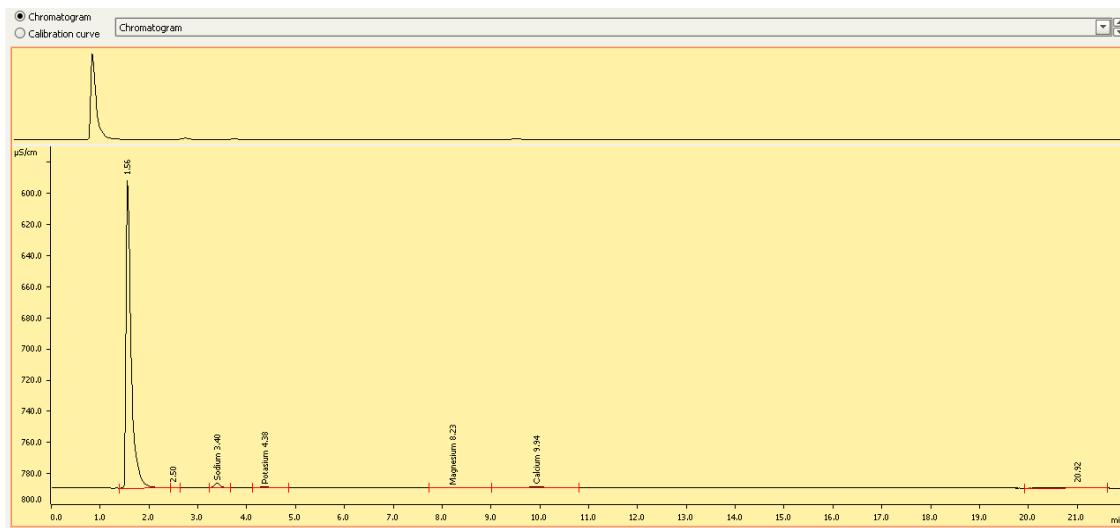


Figure 4: Ion Chromatography Result
Source: Central Instrument Faculty (BHU)

Quantitative Measurement of Ions (ppm) in O₂ Sample:

Results				
Results				
Anions				
Component name	Retention time [min]	Height [$\mu\text{S}/\text{cm}$]	Area [$(\mu\text{S}/\text{cm}) \times \text{min}$]	Concentration [ppm]
Fluoride	5.77	1.329	0.238	1.215
Chloride	8.74	1.737	0.311	2.137
Nitrate	16.06	0.077	0.029	0.822
Sulfate	24.81	0.225	0.109	1.067
Cations				
Component name	Retention time [min]	Height [$\mu\text{S}/\text{cm}$]	Area [$(\mu\text{S}/\text{cm}) \times \text{min}$]	Concentration [ppm]
Sodium	3.40	2.889	0.378	1.376
Potassium	4.38	0.667	0.144	1.067
Magnesium	8.23	0.230	0.083	0.294
Calcium	9.94	0.696	0.304	2.108

Figure 5: Ion Chromatography Result

Source: Central Instrument Faculty (BHU)

Sample H₂ for Cations

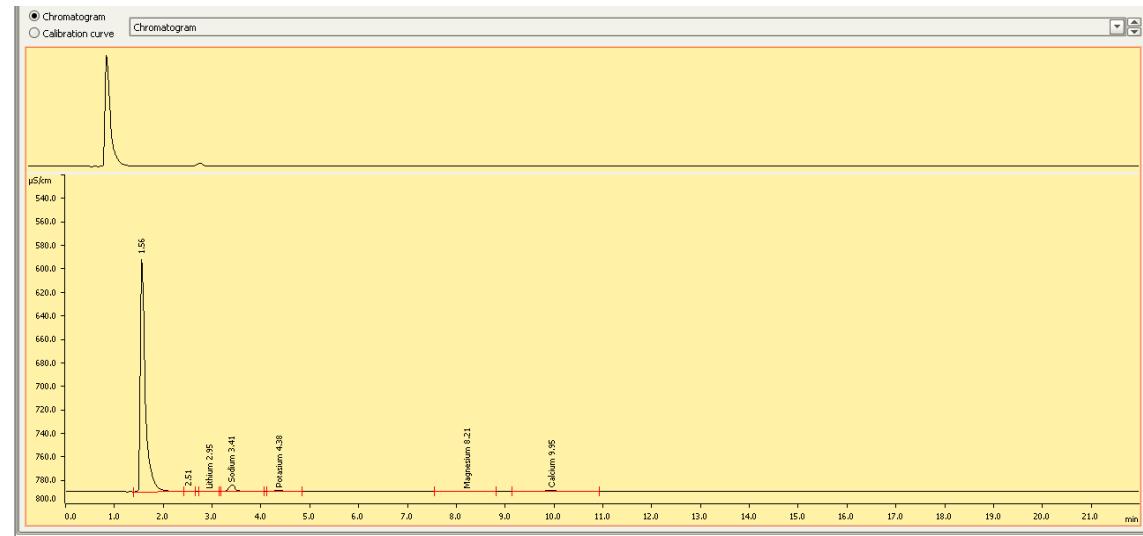


Figure 6: Ion Chromatography Result

Source: Central Instrument Faculty (BHU)

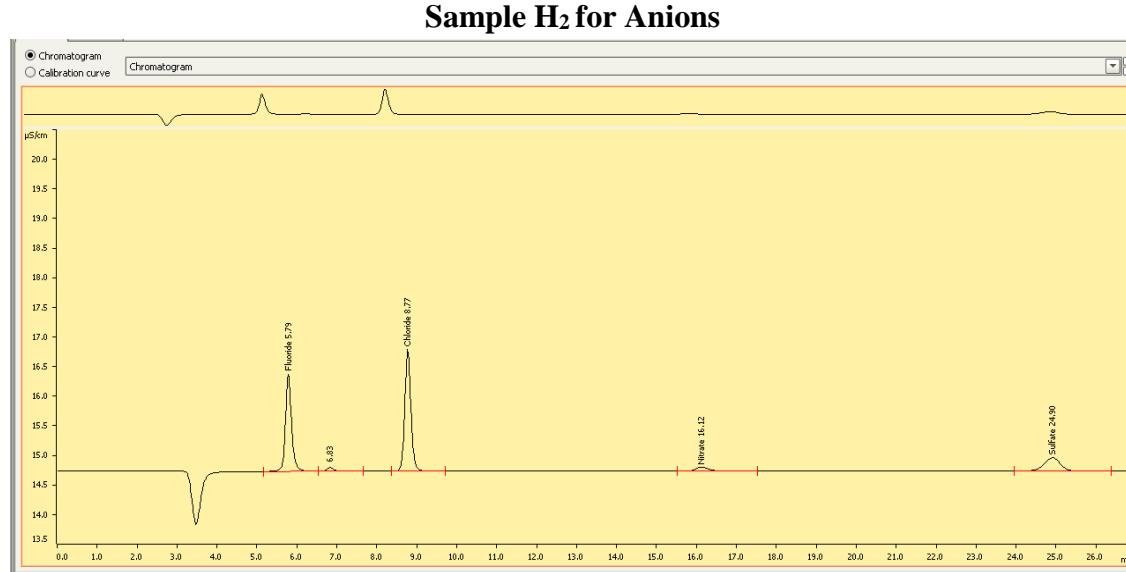


Figure 7: Ion Chromatography Result

Source: Central Instrument Faculty (BHU)

Quantitative Measurement of Ions (ppm) in H₂ Sample

Results				
Results				
Anions				
Component name	Retention time [min]	Height [μS/cm]	Area [(μS/cm) x min]	Concentration [ppm]
Fluoride	5.79	1.649	0.291	1.366
Chloride	8.77	2.058	0.368	2.372
Nitrate	16.12	0.071	0.027	0.803
Sulfate	24.90	0.226	0.107	1.057
Cations				
Component name	Retention time [min]	Height [μS/cm]	Area [(μS/cm) x min]	Concentration [ppm]
Lithium	2.95	0.013	0.003	0.031
Sodium	3.41	5.638	0.840	2.869
Potassium	4.38	0.859	0.184	1.255
Magnesium	8.21	0.214	0.077	0.282
Calcium	9.95	0.730	0.312	2.133

Figure 8: Ion Chromatography Result

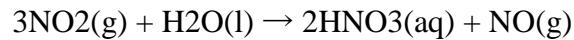
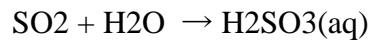
Source: Central Instrument Faculty (BHU)

The sample obtained after the solar-hydrogen plan model is exposed in an open environment for electrolysis and generation of hydrogen gas for the operation of the fuel cell is then passed through Ion chromatography test whose result detected some ions indicating contamination of fuel cell.

6.1.1 Observation & Result

The major hurdle in the domestication of a fuel cell is its delegate nature and high sensitivity towards impurities when exposed to work. Though utmost care has been taken by researchers while taking samples from both the chambers still some traces of ions like Sodium, Lithium, Chloride, Fluoride, Magnesium is detected by our chromatographer. The source of these foreign ions might be impurities in tube, chamber and sweat by skin contact (reason for the detection of Na^+ , Cl^-). Retention time and peak in the curve has been seen in case of few ions for which the chromatographer is not calibrated. The name of these unknown ions can be drawn by using reverse engineering.

During electrolysis environmental impurities like Nitrogen Dioxide (NO_2) and Sulfur Dioxide (SO_2) initiates the following reactions:



Above reactions are the reasons for the detection of Nitrate and sulfate ions in the samples. Fortunately, these reactions serve in a good way by increasing the rate of electrolysis as it add ups to the ions of water which increases its conductivity. From the experimental results, it has been found that graphite is a good choice for the production of maximum hydrogen compared to various other electrodes (Yuvaraj, A.L. AU - Daniel, et al). But ensure that we must use platinum as electrodes in electrolyser to avoid further reactions of produced acids with C which would result into the production of NO_2 and CO_2 .

6.2 pH Test

Researchers have used distilled water for electrolysis and then the sample is tested for it pH. The samples are taken after electrolysis of 30mins.

Table 2: Results of pH Test

S.No.	H ₂ Sample	O ₂ Sample	Distilled water
1	7.87	7.25	7.28
2	8.04	7.28	7.37
3	8.13	7.23	7.40
Average	8.01	7.25	7.35

From above test the shift in pH has been noted which clearly indicates the change in nature of distilled water after electrolysis. It can be observed that the distilled water in H₂ chamber turned more basic in nature whereas the distilled water in O₂ chamber tends to decrease in pH which indicates formation of some acidic component inside the chamber.

6.3 Electrical Conductivity Test

Researchers have used distilled water for electrolysis and then the sample is tested for it pH. The samples are taken after electrolysis of 30mins.

Table 3: Results of EC Test

S.No.	H ₂ Sample (μs)	O ₂ Sample(μs)	Distilled Water(μs)
1	35.8	30.9	28.7
2	35.9	31.3	28.9
3	36.1	31.7	29.1
Average	35.93	31.30	28.9

There is significant increase in the value of electrical conductivity from the value of distilled water. This increase in EC signifies formation of some ionic components during the process of electrolysis. The reason for this increment in the value of EC is the reaction that occurred inside the chamber due to gasses like SO₂ and NO₂.

6.4 Total Dissolved Solid Test

Researchers have used distilled water for electrolysis and then the sample is tested for it pH. The samples are taken after electrolysis of 30mins.

Table 4: Results of TDS Test

S.No.	H ₂ Sample (ppm)	O ₂ Sample(ppm)	Distilled water(ppm)
1	17.1	15.5	13.3
2	17.0	15.7	13.5
3	17.1	15.5	13.6
Average	17.06	15.56	13.46

Total Dissolve Solids (TDS) is a parameter that count all dissolved minerals in the water (M. R. Islam, M. K. Islam, et al). There is increase in the TDS of both H₂ and O₂ chambers which indicates addition and formation of foreign component.

Above carried out tests resulted into the conclusion that change (either increase or decrease) in pH or electrical conductivity of sample indicates addition of impurities in to the fuel supplies to the fuel cell. Also, the increase in TDS of sample implies the addition of dissolved foreign particles that are causing the contamination in fuel cell.

7. Proposed Model

The new model has been proposed with an aim of achieving long term utilization of fuel cells as a domestic source of electricity generation. The political-economic analysis and payback period of this model is a very interesting future research topic. Researchers are trying to analyze every aspect of this model before giving it a physical significance.

7.1 Hydrogen Production Technologies

Table 5: Efficiencies of Different Sources of Hydrogen Gas Production

Technology	Feedstock	Efficiency	Maturity
Steam Reforming	Hydrocarbons	70–85%	Commercial
Electrolysis	H ₂ O + electricity	50–70%	Commercial
Partial oxidation	Hydrocarbons	60–75%	Commercial
Biomass gasification	Biomass	35–50%	Commercial

Source: Hydrogen Production Technologies: Current State and Future Developments Christos M. Kalamaras and Angelos M. Efstathiou Chemistry Department, University of Cyprus, 1678 Nicosia, Cyprus Received 9 January 2013; Accepted 31 March 2013

7.2. Flow Diagram

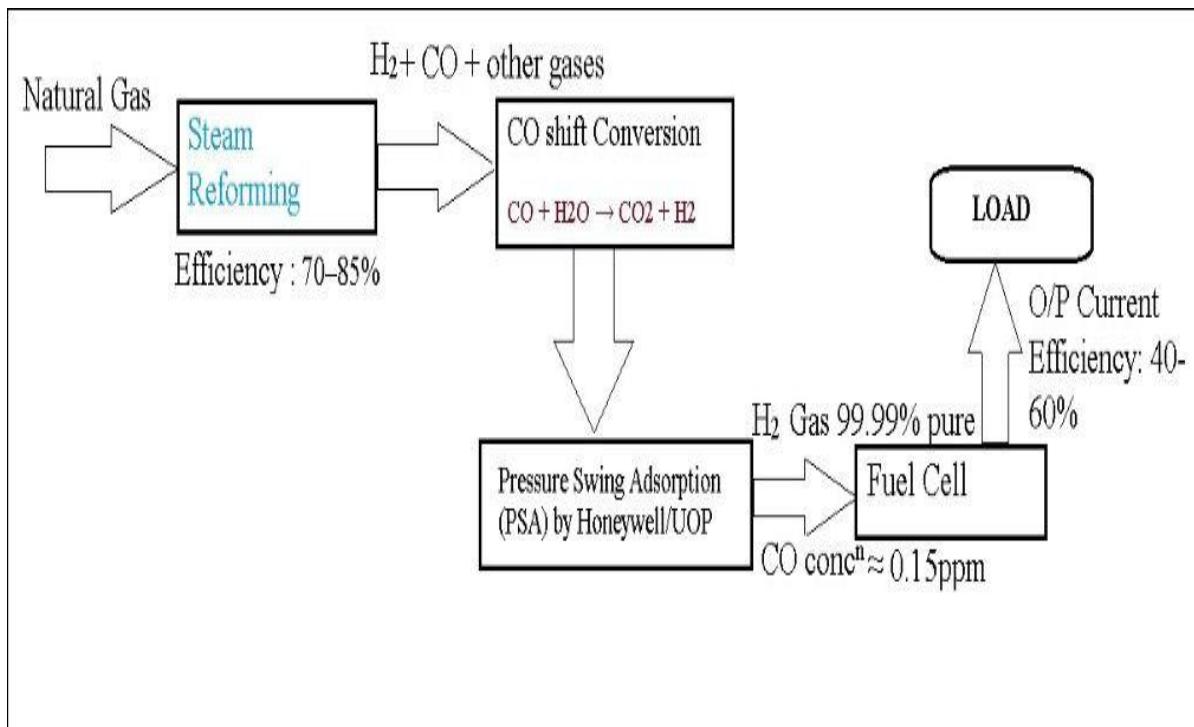


Figure 9: Flow Diagram for Proposed Model

Source: Self

Increased industrial and human activities are the main reasons for the higher level of metal ions present in waste water (Saha et al., 2017) (Nidhi, C., Sharma, B., & Singh, P. K. (2017)) and this is one of the reasons which bounded us to think for the contamination of fuel cell and its consequences. Electricity is playing a crucial role in everybody's life alternatively, energy usage index indicate the country's economy, growth and standard of living (Soppimath, V. M., & Hudedmani, M. G. (2017)). Henceforth, the above proposed model can play a crucial role in generating a clean and green energy for long time in a very efficient manner.

8. Conclusion

This paper is an introduction to the working of a fuel cell in electricity generation sector. According to the U.S. Department of Energy, the number of fuel cell systems shipped worldwide per year increased from 10 000 to more than 60 000 units, from 2008 to 2015 (Efficiency, U.S.D.o.E.O.o.E., et al). We have discussed the poisoning of fuel cell by different contamination sources such as CO and other components of air. Contamination prevention and mitigation (CO poisoning) techniques are also part of our discussion. Results of tests such as Ion chromatography, pH, TDS, and EC test has been analyzed along with the sources of impurities such as nitrate ion,

sulfate, NOx, COx, Sox etc. Sources of ionic impurities and its effect on electrolysis are also explained. Different ways of generating high purity hydrogen gas such as PSA, High & Low temperature diffusion, and absorption of CO and CO₂ in solvents are also explained. The solar hydrogen plant model proved to be helpful in detecting the impurities that can cause contamination. The proposed model would be able to supply high purity hydrogen which will cause negligible contamination in the fuel cell.

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