

Understanding the History of Fuel Cells

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Abstract-- Fuel cells are one of the key enabling technologies for future hydrogen economy. For the last 20 years applications for the fuel cells are mostly replacing internal combustions engines, and providing power in stationary and portable power applications. But the history of the fuel cells is more than the last 20 years; actually it has more than 150 years! It is the purpose of this paper to present the development of the fuel cells across the time. The paper discuss the typical characteristics and electrochemical reactions a fuel cell.

Additionally, the paper presents the basic concepts, applications for the six main types of fuel cell technologies. Finally, it is intended with the paper to present the concepts and history related to the fuel cells in a basic way. This paper will be very helpful for undergraduate researches, history and professional engineers without previous knowledge of the technical fields related to the fuel cells.

Index Terms-- Fuel Cells, History, Power Engineering Education, Product Development

I. INTRODUCTION

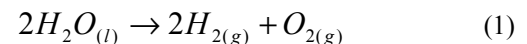
APPROXIMATELY, 46% of the electricity generated worldwide comes from the combustion of fossil fuels which has an environmental impact due to the production of the fossil fuels as well as from their combustion. Also, these resources won't last forever as the increasing population eats up the reserves of fossil fuels faster than ever. Our future quality of life demands alternatives solutions to meet the market needs as well as the production of reliable, high quality and cheap energy, contemplating a reduction of environmentally harmful emissions, to include green house gases and toxic waste. People must rely on the renewable energy resources in order to achieve this goal [1].

Fuel cells are appealing because they offer high efficiency, excellent part load performance, lower emissions of regulated pollutants, and a wide size range [2]. In the past, fuel cell systems were thought to be attractive for utilities, on-site cogeneration, and specialized transportation applications such as trucks and buses. However, at the time, most demonstration projects were limited to utility power applications [3]. But right now, many researchers, manufacturers, energy companies, and regulatory agencies are currently working to develop a variety of fuel cell types and to plan the infrastructure that will support these new technologies. These efforts involve development of new materials, economical manufacturing processes, advanced equipment for supplying air and fuel, advanced power electronics for controlling the cell output, and comprehensive

approaches for systems analysis and optimization. The future of the fuel cells looks very bright with new initiatives and collaborations of many nations based on the mutual need of energy and the replacement of the fossil fuels.

II. HISTORY OF FUEL CELL

Despite their modern high-tech aura, fuel cells actually have been known to science for more than 150 years! Though generally considered a curiosity in the 1800s, fuel cells became the subject of intense research and development during the 1900s. In 1800, British scientists William Nicholson and Anthony Carlisle had described the process of using electricity to decompose water into hydrogen and oxygen [4]. This process is named electrolysis.

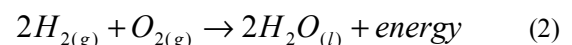


In 1832, Michael Faraday reported that the quantity of elements separated by passing an electrical through a dissolved salt was proportional to the quantity of electric charge passed through the circuit. From these experiments the two fundamental Laws of Electrolysis were derived. Without former advance mathematical background, Faraday is considered the best experimentalist!

TABLE I
FARADAY'S LAWS OF ELECTROLYSIS

<i>Faraday's 1st Law of Electrolysis</i>
The mass of a substance produced at an electrode during electrolysis is proportional to the number of moles of electrons (the quantity of electricity) transferred at that electrode.
<i>Faraday's 2nd Law of Electrolysis</i>
The number of Faradays of electric charge required to discharge one mole of substance at an electrode is equal to the number of "excess" elementary charges on that ion.

William Robert Grove, however took this idea one step further or, more accurately, one step in reverse in 1838. Grove discovered that by arranging two platinum electrodes with one end of each immersed in a container of sulfuric acid and the other ends separately sealed in containers of oxygen and hydrogen, a constant current would flow between the electrodes. The sealed containers held water as well as the gases, and he noted that the water level rose in both tubes as the current flowed. By combining several sets of these electrodes in a series circuit, he created what he called a "gas battery"- the first fuel cell [4]. Grove is considered to be the father of the fuel cell.



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In 1889, Ludwig Mond and assistant Carl Langer described their experiments with a gas-powered battery using coal-derived "Mond-gas" that attained 6 amps per square foot (measuring the surface area of the electrode) at 0.73 volts with electrodes of thin, perforated platinum. They called their system a fuel cell. Interestingly, Ludwig Mond was very successful in the business of sodas with the commercial use of the Solvay process.

Friedrich Wilhelm Ostwald, a founder of the field of physical chemistry, provided much of the theoretical understanding of how fuel cells operate [4], [5]. In 1893, he experimentally determined the interconnected roles of the various components of the fuel cell: electrodes, electrolyte, oxidizing and reducing agents, anions, and cations. Grove had speculated that the action in his gas battery occurred at the point of contact between electrode, gas, and electrolyte, but was at a loss to explain further. Ostwald, drawing on his pioneering work in relating physical properties and chemical reactions, solved the puzzle of Grove's gas battery. His exploration of the underlying chemistry of fuel cells laid the groundwork for later fuel cell researchers [5].

Francis Thomas Bacon (1904-1992) developed the first practical hydrogen-oxygen fuel cells, which convert air and fuel directly into electricity through electrochemical processes. He began researching alkali electrolyte fuel cells in the late 1930s. In 1939, he built a cell that used nickel gauze electrodes and operated under pressure as high as 3000 psi. During World War II, Bacon worked on developing a fuel cell that could be used in Royal Navy submarines, and in 1958 demonstrated an alkali cell using a stack of 10-inch diameter electrodes for Britain's National Research Development Corporation. Though expensive, Bacon's fuel cells proved reliable enough to attract the attention of Pratt & Whitney. The company licensed Bacon's work for the Apollo spacecraft fuel cells [5]. Interestingly, these key works were revolutionary at their time and putting today the hydrogen in good position as the next carrier of fuel useful for the economy of the world.

As an interesting note, Allis Chalmers Fuel Cell Tractor was an experimental tractor developed during 1959. This 20 horse power tractor has 1008 individual fuel cells, fueled by a mixture of gases (mainly propane) which in turn created a current flow. The 1008 fuel cells made an output of about 15kW and one volt of output per fuel cell. This experimental tractor is at the Smithsonian Institute.

III. WHY HYDROGEN?

Hydrogen is an energy carrier, not an energy source, meaning it can store and deliver energy in a usable form. As an energy carrier, hydrogen offers several advantages. It can be produced using abundant and diverse domestic energy resources, including fossil fuels, such as natural gas and coal, renewable energy resources, such as solar wind and biomass, and nuclear energy. This diversity of energy supply would mean we do not need to rely on any single energy resource or on foreign source of energy [6]. Producing hydrogen from renewable and nuclear sources, and from fossil fuel-based systems with carbon sequestration, yields near zero greenhouse gas or criteria emissions. Hydrogen can power all sectors of the economy: transportation, energy, industrial, and buildings. But to achieve this task, a fuel cell is needed.

IV. WHAT IS A FUEL CELL?

A fuel cell is an electrochemical device that dynamically converts the energy of a chemical reaction between hydrogen and an oxidant into electrical energy for our consumption [7]. Fuel cells work just like a battery in its basic principles: two electrodes separated by an electrolyte. They differ from batteries in that they are designed for continuous replenishment of the reactants consumed. They produce electricity from an external supply of fuel and oxidant (typically oxygen or air, although chlorine and chlorine dioxide have also been used) as opposed to the limited internal energy storage capacity of a battery. The physical structure of a fuel cell consists of an electrolyte layer separating two electrodes. At the present time fuel cells are in research and are constantly being improved for better performance and application purposes [7].

V. HOW DO FUEL CELLS WORK?

In principle, a fuel cell operates like a battery, consisting of an electrolyte placed between two electrodes: an anode and a cathode. Unlike the battery, a fuel cell does not run down or require recharging [7]. It will produce energy in the form of electricity and heat as long as fuel is supplied. Oxygen passes over one electrode and hydrogen over the other, generating electricity, water and heat. Hydrogen fuel is fed into the "anode" of the fuel cell. Oxygen (or air) enters the fuel cell through the cathode. This reaction occurs along a catalyst, the hydrogen atom splits into a proton and an electron, which take different paths to the cathode. The proton passes through the electrolyte. The electrons create a separate current that can be utilized before they return to the cathode, to be reunited with the hydrogen and oxygen in a molecule of water. Figure 1 shows the typical reaction for a fuel cell to produce electricity. Figure 2 shows a methanol fuel cell. Figure 3 shows the typical polarization curve for a fuel cell.

VI. CHARACTERISTICS OF FUEL CELLS

The major component of a fuel cell system, the fuel cell stack, is composed of individual fuel cells assembled in repetition. Thus, the fuel cell stack is modular, and can be constructed in sizes ranging from a few watts up to over a megawatt [8]. Other components of the fuel cell system, particularly the fuel processor, do not scale as well as the stack. However, even fuel cell systems incorporating fuel processors can be constructed to meet a variety of applications with power needs as small as 10 kW. Across the entire range of applicable sizes, fuel cell systems offer attractive electrical conversion efficiencies. A fuel cell system remains efficient even at off-design conditions. The efficiency for various fuel cell systems ranges from 40%–50% for simple systems in a broad range of sizes. The smaller the system is created the more the efficiency compares to those provided by fuel cell systems at design conditions [8]. Furthermore, no conventional system can maintain efficiencies comparable to fuel cell systems at part-load operation. More complex fuel cell systems can yield even higher efficiencies. For a

combined system consisting of a pressurized solid oxide fuel cell (SOFC) with the exhaust gas driving a gas turbine, the overall electrical conversion efficiency can be as high as 60%. The combined gas turbine/steam cycle is the only conventional cycle that can approach this level of efficiency at least at design load. Since fuel cells can operate at high efficiency even in relatively small sizes, they are attractive in small-scale cogeneration applications such as buildings [8]. Fuel cells can offer cogeneration efficiencies as high as 80% by producing electricity and thermal energy for applications such as water heating or space heating. Fuel cells are also attractive because of their low environmental impact relative to conventional systems. The fuel cell stack itself operates on hydrogen, thus, water is the only product from the stack reaction. However, if the hydrogen is produced from a hydrocarbon fuel, then carbon dioxide is also produced. Any reaction that completely oxidizes a hydrocarbon fuel must produce carbon dioxide in proportion to the quantity of carbon in the primary fuel source. Thus, the overall system including the fuel processor produces both water and carbon dioxide.

Fuel cells minimize emissions of regulated pollutants. The emissions of currently regulated pollutants such as carbon monoxide, nitrous oxides, sulfur oxides, and particulates are well below current air quality regulations and typically nearly nonexistent [9]. Even carbon dioxide emissions are lower with a fuel cell system because the system efficiency is higher and the amount of fuel used is lower. It is good to mention that carbon dioxide emissions are not currently regulated. Besides, fuel cell systems are also relatively quiet and unobtrusive so that the overall impact on the environment is small. This permits fuel cells to be located in a variety of places that would not be acceptable for conventional power plants. For example, fuel cell systems have been sited near hospitals, near housing facilities, and even in New York's Central Park. Locating fuel cell power systems near the need for power eliminates electrical transmission losses, facilitates cogeneration, and in some cases improves the reliability of the power systems [10].

VII. TYPES OF FUEL CELLS

A. Proton Exchange Membrane

PEM technology was invented at General Electric in the early 1960s, through the work of Thomas Grubb and Leonard Niedrach. GE announced an initial success in mid-1960 when the company developed a small fuel cell for a program with the U.S. Navy's Bureau of Ships (Electronics Division) and the U.S. Army Signal Corps [4]. The unit was fueled by hydrogen generated by mixing water and lithium hydride. This fuel mixture was contained in disposable canisters that could be easily supplied to personnel in the field. The cell was compact and portable, but its platinum catalysts were expensive. These fuel cells operate at relatively low temperatures (about 175°F), have high power density, can vary their output quickly to meet shifts in power demand, and are suited for applications, such as in automobiles, where quick startup is required. According to the U.S. Department of Energy (DOE), "they are the primary candidates for light-duty

vehicles, for buildings, and potentially for much smaller applications such as replacements for rechargeable batteries." This type of fuel cell is sensitive to fuel impurities. Cell outputs generally range from 50 watts to 75 kW [11][12].

B. Molten Carbonate (MCFC)

In the 1930s, Emil Baur and H. Preis in Switzerland experimented with high-temperature, solid oxide electrolytes. They encountered problems with electrical conductivity and unwanted chemical reactions between the electrolytes and various gases (including carbon monoxide). The following decade, O. K. Davtyan of Russia explored this area further, but with little success. By the late 1950s, Dutch scientists G. H. J. Broers and J. A. A. Ketelaar began building on this previous work and decided that limitations on solid oxides at that time made short-term progress unlikely. They focused instead on electrolytes of fused (molten) carbonate salts [5].

By 1960, they reported making a cell that ran for six months using an electrolyte "mixture of lithium-, sodium- and / or potassium carbonate, impregnated in a porous sintered disk of magnesium oxide." However, they found that the molten electrolyte was slowly lost, partly through reactions with gasket materials. About the same time, Francis T. Bacon was working with a molten cell using two-layer electrodes on either side of a "free molten" electrolyte. At least two groups were working with semisolid or "paste" electrolytes and most groups were investigating "diffusion" electrodes rather than solid ones.

In the mid-1960s, the U.S. Army's Mobility Equipment Research and Development Center (MERDC) at Ft. Belvoir tested several molten carbonate cells made by Texas Instruments. These ranged in size from 100 watts to 1,000 watts output and were designed to run on "combat gasoline" using an external reformer to extract hydrogen. The Army especially wanted to use fuels already available, rather than a special fuel that might be difficult to supply to field units.

Molten carbonate fuel cells use an electrolyte composed of a molten carbonate salt mixture suspended in a porous, chemically inert matrix, and operate at high temperatures, approximately 1,200°F. They require carbon dioxide and oxygen to be delivered to the cathode. To date, MCFCs have been operated on hydrogen, carbon monoxide, natural gas, propane, landfill gas, marine diesel, and simulated coal gasification products. 10 kW to 2 MW MCFCs have been tested on a variety of fuels and are primarily targeted to electric utility applications [12].

C. Solid Oxide (SOFC)

Swiss scientist Emil Baur and his colleague H. Preis experimented with solid oxide electrolytes in the late 1930s, using such materials as zirconium, yttrium, cerium, lanthanum, and tungsten. Their designs were not as electrically conductive as hoped and reportedly experienced unwanted chemical reactions between the electrolytes and various gases, including carbon monoxide.

By the late 1950s, research into solid oxide technology began to accelerate at the Central Technical Institute in The Hague, Netherlands, Consolidation Coal Company, in Pennsylvania, and General Electric, in Schenectady, New York. A 1959 discussion of fuel cells noted that problems with

solid electrolytes included relatively high internal electrical resistance, melting, and short-circuiting due to semi conductivity. It seems that many researchers began to believe that molten carbonate fuel cells showed more short-term promise.

Solid oxide fuel cells use a hard, non-porous ceramic compound as the electrolyte, and operate at very high temperatures – around 1800°F. One type of SOFC uses an array of meter-long tubes, and other variations include a compressed disc that resembles the top of a soup can. Tubular SOFC designs are closer to commercialization and are being produced by several companies around the world. SOFCs are suitable for stationary applications as well as for auxiliary power units (APUs) used in vehicles to power electronics [5], [12].

D. Alkaline (AFC)

Francis Thomas Bacon (1904-1992) of Britain began experimenting with alkali electrolytes in the late 1930s, settling on potassium hydroxide (or KOH) instead of using the acid electrolytes known since Grove's early discoveries. KOH performed as well as acid electrolytes and was not as corrosive to the electrodes. Bacon's cell also used porous "gas-diffusion electrodes" rather than solid electrodes as Grove had used. Gas-diffusion electrodes increased the surface area in which the reaction between the electrode, the electrolyte and the fuel occurs. Also, Bacon used pressurized gases to keep the electrolyte from "flooding" the tiny pores in the electrodes. Over the course of the following twenty years, Bacon made enough progress with the alkali cell to present large scale demonstrations.

In the early 1960s, aircraft engine manufacturer Pratt & Whitney licensed the Bacon patents and won the National Aeronautics and Space Administration (NASA) contract to power the Apollo spacecraft on space missions. These cells can achieve power generating efficiencies of up to 70 percent. They were used on the Apollo spacecraft to provide both electricity and drinking water. Alkaline fuel cells mainly use potassium hydroxide as the electrolyte and operate at 160°F. However, they are very susceptible to carbon contamination, so require pure hydrogen and oxygen [11].

E. Phosphoric Acid (PAFC)

Experimenters have used acids as electrolytes since the time of William Grove's first gas battery in 1842 – he used sulfuric acid. But phosphoric acid, a poor conductor of electricity, was not as attractive, and PAFCs were slower to develop than other types of fuel cells. In 1961, G. V. Elmore and H. A. Tanner revealed new promise in phosphoric acid electrolytes in their paper "Intermediate Temperature Fuel Cells." They described their experiments using an electrolyte that was 35 percent phosphoric acid and 65 percent silica powder pasted into a Teflon gasket. "Unlike sulfuric," they noted "phosphoric acid is not reduced electrochemically under cell operating conditions." Also, their PAFC ran on air, rather than pure oxygen. "An acid cell was operated for six months at a current density of 90 [milliamps per square centimeter] and 0.25 v. with no apparent deterioration." In the mid 1960s, the U.S. Army explored the potential for PAFCs that ran on "logistic

fuels," meaning fuels commonly available to units in the field. For the Army's tests, a cell was produced by Allis-Chalmers and used an Engelhard Industries steam reformer and an electrical inverter from Varo, Inc. Engelhard's O. J. Adlhart developed a "plastic-bonded electrolyte" during this program [4], [5].

Phosphoric acid fuel cells are one of the few that are commercially available today. Hundreds of fuel cell systems have been installed in 19 nations – in hospitals, nursing homes, hotels, office buildings, schools, utility power plants, landfills and waste water treatment plants. PAFCs generate electricity at more than 40% efficiency – and nearly 85% if the steam this fuel cell produces is used for cogeneration – this compares to about 35% for the utility power grid in the United States. Phosphoric acid fuel cells use liquid phosphoric acid as the electrolyte and operate at about 450°F. One of the main advantages to this type of fuel cell, besides the nearly 85% cogeneration efficiency, is that it can use impure hydrogen as fuel. PAFCs can tolerate a CO concentration of about 1.5 percent, which broadens the choice of fuels they can use. If gasoline is used, the sulfur must be removed [13][14].

F. Direct Methanol (DMFC)

Initially developed in the early 1990s, the direct methanol fuel cell was invented and developed at the NASA Jet Propulsion Laboratory and the University of Southern California (USC) and is protected by 56 issued and over 62 pending patents worldwide.

Methanol fuel cells are replacements for traditional batteries and are expected to gain a substantial market share because they offer longer operating time as compared to current lithium ion batteries and may be instantaneously recharged by simply replacing the disposable fuel cartridge. DMFC products are being developed for these applications by companies such as Samsung in Korea, and by Toshiba, NEC, Hitachi and Sanyo in Japan.

These cells are similar to the PEM cells in that they both use a polymer membrane as the electrolyte. However, in the DMFC, the anode catalyst itself draws the hydrogen from the liquid methanol, eliminating the need for a fuel reformer. Efficiencies of about 40% are expected with this type of fuel cell, which would typically operate at a temperature between 120-190°F. This is a relatively low range, making this fuel cell attractive for tiny to mid-sized applications, to power cellular phones and laptops. Higher efficiencies are achieved at higher temperatures. Companies are also working on DMFC prototypes to be used by the military for powering electronic equipment in the field [15]. Figures 2 show a DMFC..

VIII. TYPES OF FUEL CELLS

Figure 4 summarizes the last sections showing the evolution of the fuel cells across the time. Now, it is time to discuss the advantages and applications related to the fuel cells. Some of the advantages related to fuel cell systems are the promise to provide compared to conventional power systems include modularity, high efficiency across a broad range of load conditions, and low environmental impact. These advantages coupled with projected cost reductions will make fuel cells attractive in a variety of applications. Fuel cells are very useful

as power sources in remote locations, such as spacecraft, remote weather stations, large parks, rural locations, and in certain military applications [16].

A fuel cell system running on hydrogen can be compact, lightweight and has no major moving parts. Because fuel cells have no moving parts, and do not involve combustion, in ideal conditions they can achieve up to 99.9999% reliability. This equates to less than one minute of down time in a six year period [16], [17]. A new application is combined heat and power (CHP) for family home, office buildings and factories. This type of system generates constant electric power (selling excess power back to the grid when it is not consumed), and at the same time produces hot air and water from the waste heat. A lower fuel-to-electricity conversion efficiency is tolerated (typically 15-20%), because most of the energy not converted into electricity is utilized as heat. Some heat is lost with the exhaust gas just as in a normal furnace, so the combined heat and power efficiency is still lower than 100%, typically around 80%. In terms of energy however, the process is inefficient, and one could do better by maximizing the electricity generated and then using the electricity to drive a heat pump.

Phosphoric-acid fuel cells (PAFC) comprise the largest segment of existing CHP products worldwide and can provide combined efficiencies close to 80% (45-50% electric + remainder as thermal). UTC Power is currently the world's largest manufacturer of PAFC [17].

Molten-carbonate fuel cells have also been installed in these applications, and solid-oxide fuel cell prototypes exist. However, since electrolyser systems do not store fuel in themselves, but rather rely on external storage units, they can be successfully applied in large-scale energy storage, rural areas being one example [8]. In this application, batteries would have to be largely oversized to meet the storage demand, but fuel cells only need a larger storage unit (typically cheaper than an electrochemical device). One such pilot program is operating on Stuart Island in Washington State. There, the Stuart Island Energy Initiative has built a complete, closed-loop system: Solar panels power an electrolyser which makes hydrogen. The hydrogen is stored in a 500 gallon tank at 200 PSI, and runs a relay on fuel cell to provide full electric back-up to the off-the-grid residence.

IX. CONCLUSION

Clearly, this paper shows aspects related to the fuel cells and its applications from the 19th century to the present. The paper presented the history and evolution of the fuel cells across the time. Also, it was discussed the characteristics for different types of fuel cells. Summarizing, the fuel cells use hydrogen to create electricity, with only water and heat as byproducts. Fuel cells can produce as long as fuel is supplied. Today, hydrogen fuel cells promise two to three times the efficiency of traditional combustion technologies; for vehicles, this can mean a 50% reduction in fuel consumption compared to a conventional vehicle with gasoline internal combustion engine; zero to near-zero levels of harmful emissions from vehicles and power plants also quiet operation, with fewer moving parts, scalability, and the use for a variety of applications including transportation, stability and portable power.

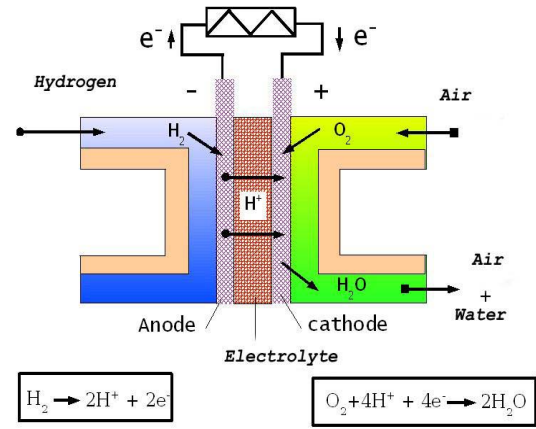


Fig. 1. A diagram showing the makeup of a fuel cell and the reaction it uses to produce electricity [18].

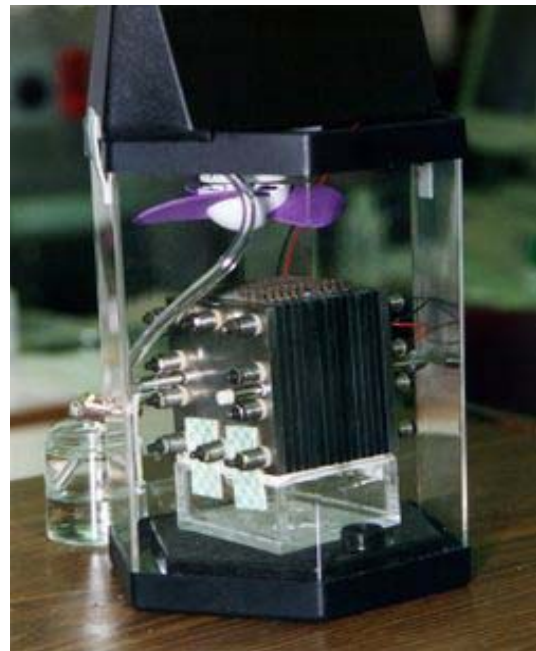


Fig. 2 Methanol fuel cell. The actual fuel cell stack is the layered cubic structure in the center of the image [19].

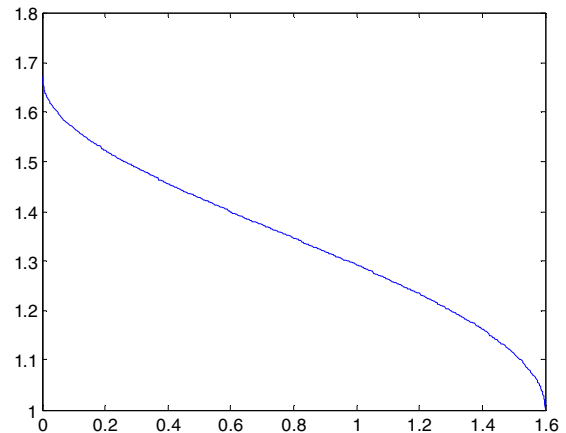


Fig. 3 Typical polarization current density vs voltage curve of a fuel cell.

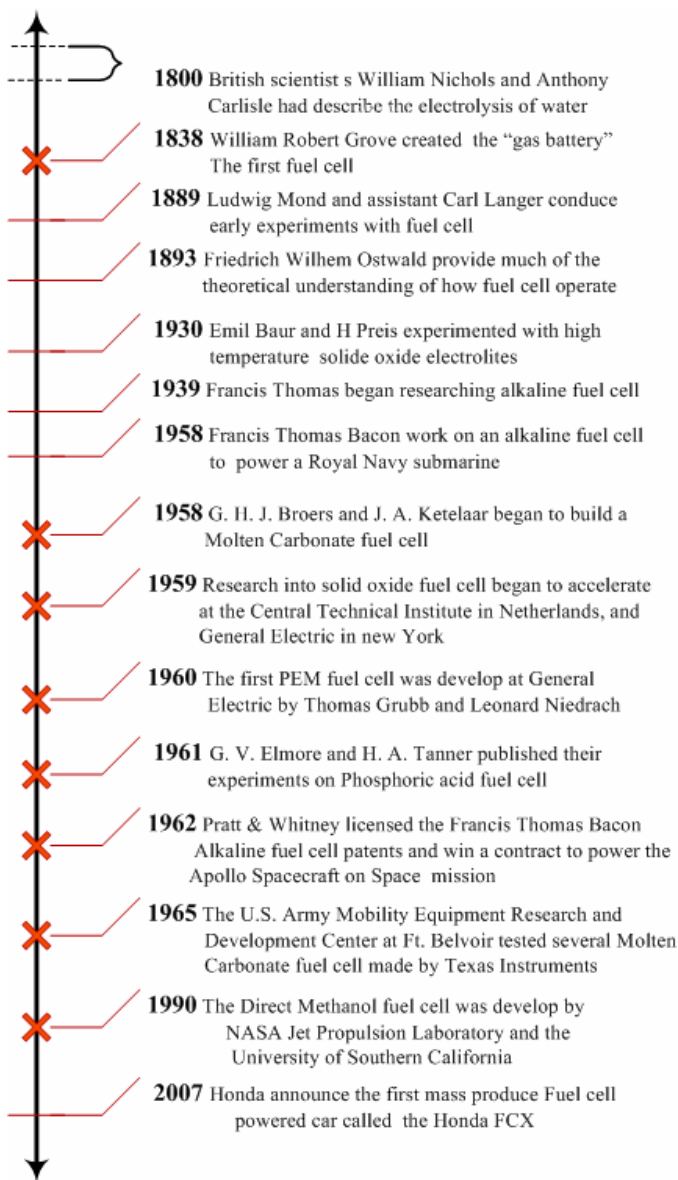


Fig. 4 Fuel Cell's history across time

Finally, it is expected that this paper will be used in the nearest future as supplementary material for undergraduate engineering education, historians and for professional engineers that want to expand their knowledge and learn more about the history and development of the fuel cells.

X. ACKNOWLEDGMENT

The authors gratefully acknowledge the contributions of all the members that are part of the Mathematical Modeling and Control of Renewable Energy Laboratory and Research Team (M-MCREL RT).

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- [18] Fig. 1. courtesy of U.S. department of energy E.E.R.E. http://www.eere.energy.gov/hydrogen_and_fuelcells/fuelcells/fc_types.html as on March 27, 2007
- [19] Fig. 2. Methanol fuel cell courtesy of NASA "<http://www2.jpl.nasa.gov/files/images/hi-res/p48600ac.tif>" as on March 27, 2007