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Fuel Cells and solid oxide fuel cells Technology

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ABSTRACT

Fuel cells are energy conversion devices that produce electricity via a chemical reaction in the cell and this happens as long as fuels are supplied. There are traditional hydrogen fuel cells,, liquid-fueled fuel cells and now solid-oxide fuel cells. The liquid-fueled fuel cells are an attractive alternative to the traditional hydrogen fuel cells since they eliminate the need to transport and store hydrogen gas, which is an expensive exercise. Whereas the solid oxide fuel cells (SOFCs) are quite clean and efficient power sources for the generation of electricity from different types of fuels such as hydrogen, natural gas, and biogas, and also do not have corrosive components. The SOFCs do not require costly metal electro-catalysts due to high operation temperatures in the range of $800℃ - 1000℃$. Initially fuel cells, batteries, etc. were conceived with liquid electrolytes that serve to transfer charge between the anode and the cathode. But a solid-oxide fuel-cell is a type of fuel cell in which solid oxide material is used as an electrolyte to conduct negative oxygen ions from the cathode to the anode. The electrochemical oxidation of the oxygen ions with hydrogen or carbon monoxide thus takes place on the anode side. New reactions have been proposed for SOFC, and the calculations have been done for the change in reaction enthalpy (ΔH) and harvested energy. The reaction enthalpy is more than the reaction enthalpy obtained from the standard reaction in the SOFC. This means that we get more power from the cell and a large current flows in the load, resulting in a higher voltage across the load.

Keywords: Fuel Cell, SOFC, Electrochemical Oxidation, Reaction Enthalpy, Harvested Energy

1. INTRODUCTION

Solid oxide fuel cells systems are flexible and highly efficient and this makes them a perfect technology for taking care of fluctuations in energy generation from photovoltaic power plants and wind energy power systems. They can be simultaneously used with fossil fuel sources and renewable fuel sources. Thus, they can be used to support the transition to a sustainable energy system [1]. They can lead to a great reduction in greenhouse gases and this can assist in improving the quality of life.

The solid oxide ceramic high-temperature fuel cell systems (SOFC) provide flexible, ultra-efficient electricity and heat generation using renewable and conventional energy sources. The SOFC's are a key technology that can accelerate the transition to renewable energies for our future energy systems [1]

Improved energy efficiency and reduced greenhouse emissions are the most important goals for future energy conservation and clean energy. Solid oxide fuel cells are a class of fuel cells characterized by the use of a solid oxide material as the electrolyte. They use solid oxide electrolyte to conduct negative oxygen ions from the cathode to the anode. The requirement for zero-emission power generation has treated the solid oxide fuel cells (SOFC) a possible solution of the problem of zero emissions [2]. An important point is the capacity of SOFCs to run on hydrogen and at higher efficiency than alternative fuel cells when providing combined heat and power (CHP), and fuel flexibility allows for a transition from hydrocarbons towards zero-emission power generation as part of the hydrogen economy.

A typical solid oxide fuel cell (SOFC) consists of a solid electrolyte, generally a ceramic, sandwiched between the anode and the cathode. Hydrogen as fuel is delivered to the anode, and air as the oxidant is delivered to the cathode [Fig.1]. The diagram in [Fig.1] shows the operating principles of the SOFC. In this, the electrodes are solid porous structures that allow the fuel and air (O_2) to diffuse to the electrode, and the products of the electrochemical reaction on the anode side to diffuse away from the electrolyte.
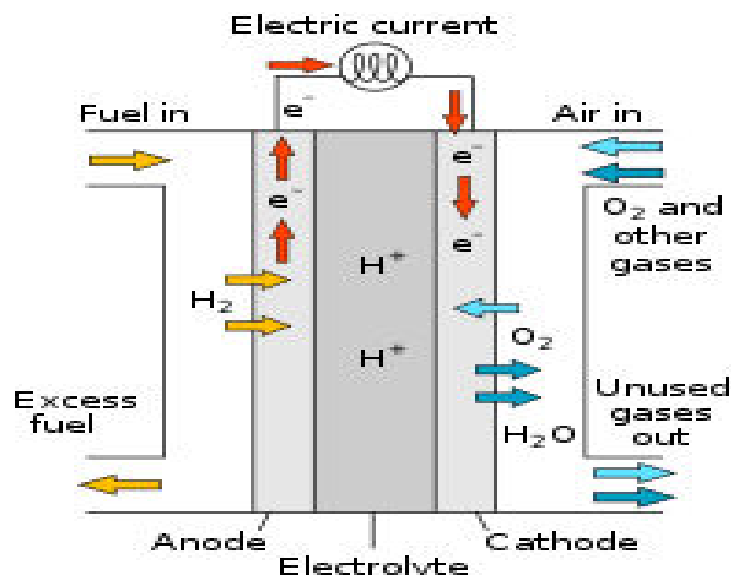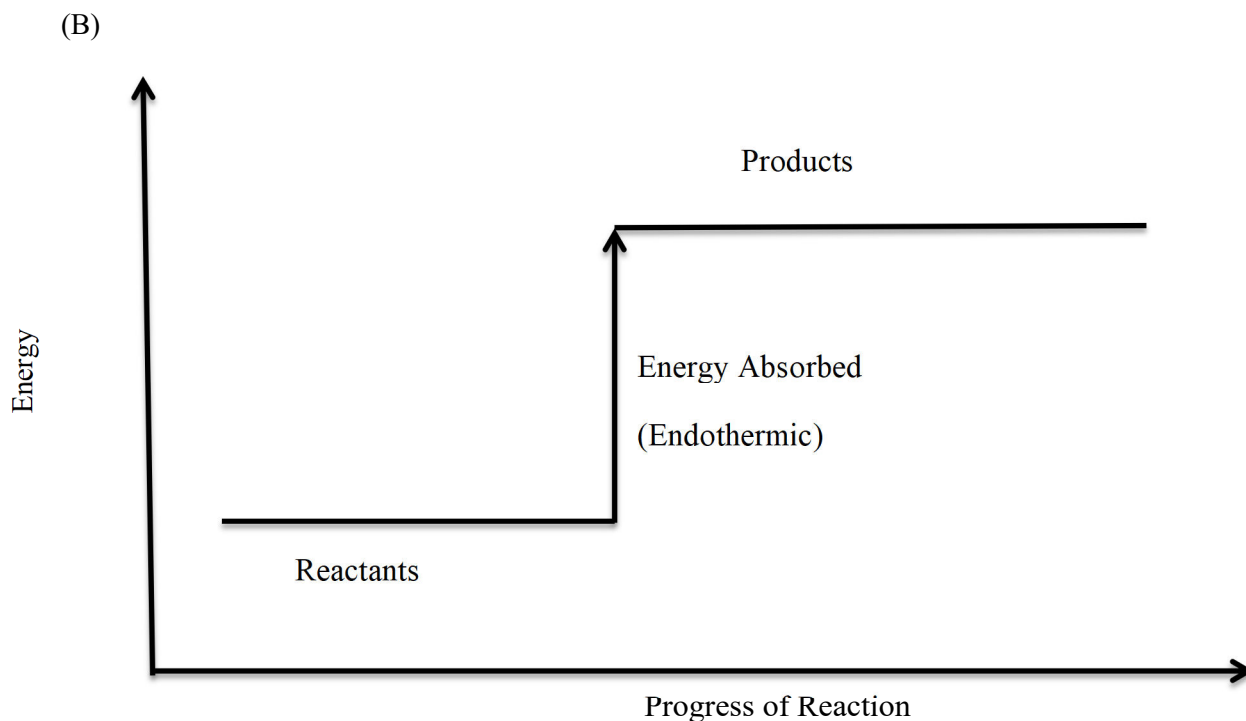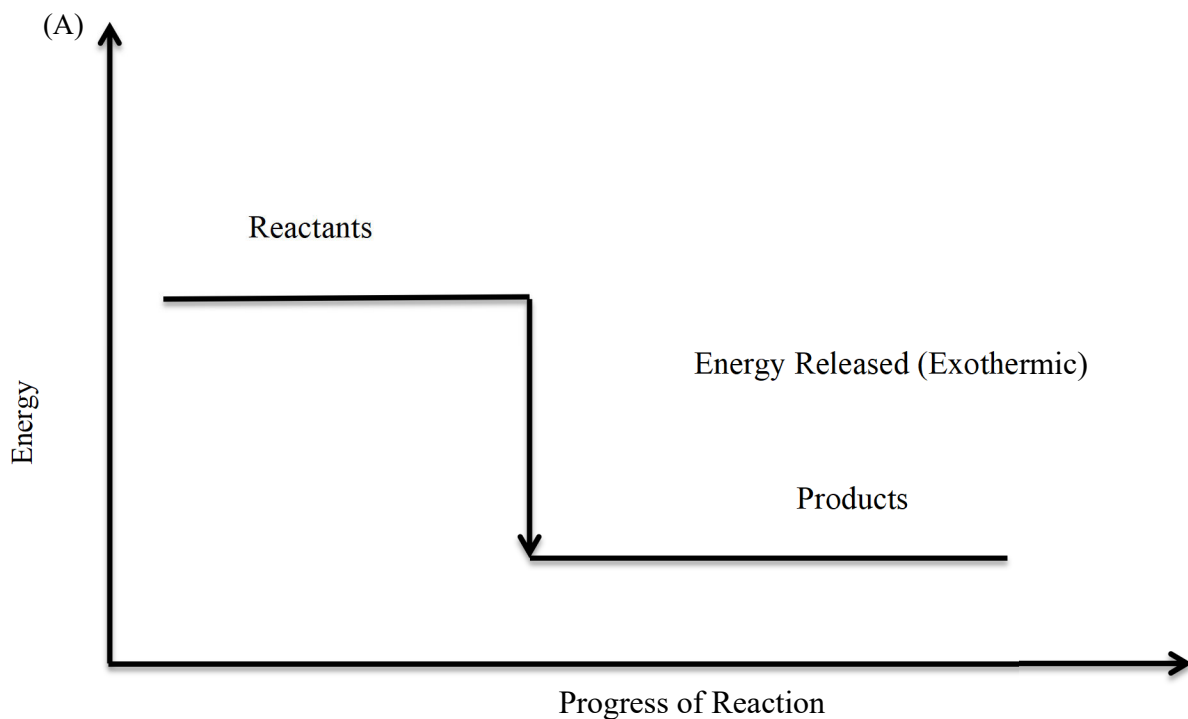
Figure 1. Solid Oxide Fuel Cell

The fundamental requirement is to calculate the enthalpy of the reaction (ΔH) in the cell, and this is called the energy that can be harvested. The value of ΔH will depend on the chemicals involved in the reaction [3-7]

There are a few major developments that have led to the increase in performance of the SOFC. One is the fabrication of thin electrolyte ($10 \mu m$) with high density to minimize the ohmic loss of the electrolyte. The second point is the reduction of excess overpotentials observed for O_2 reduction reaction at the cathode in the single SOFC. But, to develop the state of the-art IT-SOFC (Intermediate Temperature Solid Oxide Fuel Cell), a new strategy for the design of key interfaces in SOFC cells is required. The SOFC's contain a number of interface regimes which play key roles in the performance of SOFC's. The gas-solid interfaces on both cathode and anode systems control the charge transfer phenomena in the fuel cell reaction. Then the three–phase boundary (TPB) at the electrode-electrolyte interface is an important active area site where both the electron exchange and formation of conduction ions takes place. Hence key microstructures at the interface of SOFC devices are of great importance, and the control of the interface structures at cathode/electrolyte, and in the electrolyte and in the current anode layer will affect the performance of the IT-SOFC devices. The third important point for the development of IT-SOFC's is microstructure control in the anode layer so as to improve the anode performance and chemical stability of the anode to small amounts of impurity H_2S present in the fuel [8].

In this paper, we are not concerned with the experimental aspects of SOFC and or IT-SOFC [8]. The main objective is to propose new chemical reactions and calculate the reaction energy or reaction enthalpy changeΔH. Some features of the process of reaction and the calculation of enthalpy reaction,ΔH, are described as an example of how to go about to understand some of the important features and functions of SOFC.

(A)

Reactants

Energy Released (Exothermic)

Products

Energy

Progress of Reaction

(B)

Products

Energy Absorbed

(Endothermic)

Energy

Reactants

Progress of Reaction

In the diagram it is shown that in the process shown in (B) that the reactant absorbs the energy (endothermic process) and break the bond to create the products. But to form a new bond, energy is released (exothermic process) and we get new products,. In the exothermic reactions, reactants are in the higher energy state than the products and the energy difference between them is called change in enthalpy of the reaction (ΔH). The enthalpy change (ΔH) is always negative. Whereas in endothermic reactions, the products are in higher energy state than the reactants, and the energy difference or reaction energy or the change in the enthalpy of reaction (ΔH) between the reactants and the products is always positive (+ΔH). Hence, we get;

I.   That the bond enthalpy describes how much energy is required to form or break the bond

II. Combined bond enthalpy for all formed bonds and broken bonds during the process of reaction gives an idea about the total change in the energy of the system, and this is known as the reaction enthalpy or the change in enthalpy (ΔH). If ΔH is positive, the reaction is endothermic and if the ΔH is negative, the reaction is exothermic. To break the bonds, energy is absorbed (endothermic), to form the new bonds, energy is released (exothermic). The enthalpy reaction (ΔH) is the difference (exothermic-endothermic).

As an example we consider the following reaction and calculate (ΔH). Thus we write a reaction, $CH_4 + Cl_2 \rightarrow CH_3Cl + HCl$........................ 1

We need the energies to make and break different bonds. For instance, for different types of bonds,, energy required to make or break one molecule of a particular bond is generally expressed in KJ/mol, and for different bonds the values are,

$$\left. \begin{array}{l} C - H \rightarrow 412 \dfrac{KJ}{mol} \\ Cl - Cl \rightarrow 242 \dfrac{KJ}{mol} \\ C - Cl \rightarrow 331 \dfrac{KJ}{mol} \\ H - Cl \rightarrow 432 \dfrac{KJ}{mol} \end{array} \right\} .... 2$$

Now we calculate the total energy required to break one $C - H$ bond and on $Cl - Cl$ bond; and this is $412 \dfrac{KJ}{mol} + 242 \dfrac{KJ}{mol} = 654 \dfrac{KJ}{mol}$.

This refers to the left-hand side of $Eq.1$ where bonds are to be broken to create or form new bonds for the right-hand side materials to be formed.

Now the energy released during the formation of new $C - Cl$ and $H - Cl$ bond is $331\frac{KJ}{mol} + 432\frac{KJ}{mol} = 763\frac{KJ}{mol}$.

New change in energy (change in enthalpy ΔH) = Energy absorbed while breaking the bond – Energy released while forming the bond i.e;

$$\Delta H = 654\frac{KJ}{mol} - 763\frac{KJ}{mol} = -109\frac{KJ}{mol} \dots\dots\dots\dots\dots 3$$

Hence, there is a net release of energy in this reaction (exothermic). The important point to understand in SOFC is that there is an oxidation reaction which is caused by contact between substances (metals or non-metals) and oxygen molecules. Oxidation refers to the loss of electrons; when, say, carbon is oxidized, it means that carbonations in the fuel lose electrons as the carbon atoms are converted into carbon dioxide. The electrons they lose are in hydrogen atoms, which are made up of proton and an electron. More technically, we can say that oxidation can be defined as the loss of one electron during the phase where two or more elements interact [9,10,11]. A simple understanding will mean that oxidation is loss of electrons, and reduction is gain of electrons; or reduction is the opposite of oxidation. Oxygen- reduction reaction is a chemical reduction in which the oxidation number of participating chemical species changes. It involves transfer of electrons between two species. When an atom gains an electron, it reduces or decreases its oxidation number. In fact, in a reduction-oxidation reaction, the oxidation state changes in electron donor and electron acceptor species. A solid oxide fuel cell (SOFC) uses the movement of electrons and generates electricity in a few basic steps. Here, natural gas goes through a steam reforming process. The chemical reaction produces hydrogen (H_2), the carbon monoxide (CO), carbon dioxide (CO_2) and steam (H_2O). .[12,13,14]. There are two important reactions in SOFC [12] that represent the overall electrochemical reaction.

I. Anode Reaction; $\quad H_2 \rightarrow 2H^+ + 2e^- \dots\dots\dots\dots\dots\dots 4$

II. Cathode Reaction; $\quad 2H^+ + 2e^- + \frac{1}{2}O_2 \rightarrow H_2O \dots\dots\dots 5$

III. Overall Chemical Reaction; $H_2 + \frac{1}{2}O_2 \rightarrow H_2O(steam) \dots\dots 6$

Now the electromotive force or reversible open circuit voltage is given by the Nernst equation for $E.M.F, E,$[12]

$$E = E^0 + \frac{RT}{2F}\log\left(\frac{P_{H_2}P_{O_2}^{\frac{1}{2}}}{P_{H_2O}}\right) \ldots\ldots\ldots 7$$

Where;

$E = The\ electromotive\ force\ or\ reversible\ open\ circuit\ voltage\ V$

$E^0 = The\ E.M.F\ at\ standard\ pressure\ and\ voltage\ V$

$R = Universal\ gas\ constant = 8.314JK^{-1}mol^{-1}$

$F = Faraday\ constant\ (charge\ of\ one\ mole\ of\ electrons) = Avagadro's\ number\ (6.023x10^{23})x\ charge\ on\ the\ electron\ (1.6x10^{-19}c) = 96368\ Coulombs$

$P_i = Partial\ pressure\ (P_{H_2} - Hydrogen;$

$P_{O_2}^{\frac{1}{2}} - Oxygen\ and\ P_{H_2O}steam\ measured\ in\ terms\ of'bar'atmospheric\ pressure)$

In general, we have to look for a new reaction that can lead to higher voltage and the cell should have longer life span. The heating of the cell should also be small. New materials may have to be proposed for the cathode and the anode.

To increase the fuel cell efficiency and its life span, one can use an anode off-gas circulation fan driven by a steam turbine that runs on steam lubricated bearings. This is a novel way to improve the efficiency of domestic solid oxide fuel cells and these cells are rated at six Kwe (Killowat-electric). The steam turbine-driven fan which recycles gases through the system, likewise builds cell life efficiency and expectancy. The excess fuel is generally utilized in a burner thereby saving the electrical system effectiveness. In such systems, the wear and tear of the parts is low [15].

Finally, we do not plan to propose any new experimental structure of SOFCs, and have rather proposed some new reactions in which energy can be harvested (Energy harvesting is defined as the conversion of ambient-energy into electrical energy; as the collection and storage of ambient energy for on-demand, off-grid use). Widely used energy harvesting devices are solar, thermal, RF, piezoelectric, and fuel cells.

2. PROPOSED NEW REACTIONS FOR SOFC

For comparison, we briefly describe the standard reactions in the SOFC's i.e.,

$$H_2 + \frac{1}{2}O_2 \rightarrow H_2O \dots \dots 8$$
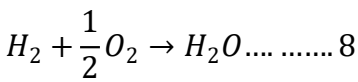
To calculate the energy released, we need the standard bond enthalpies that denote the enthalpy absorbed when bonds are broken at ambient (atmospheric) pressure and temperature ($= 300K$). Numerical values of bond enthalpies are:
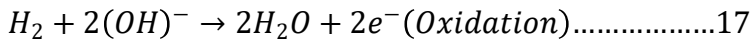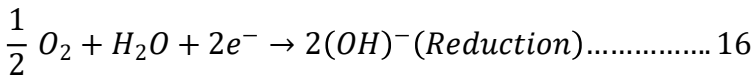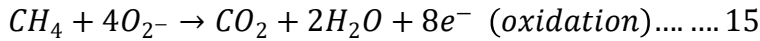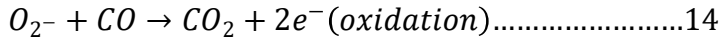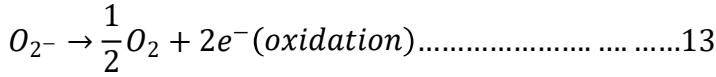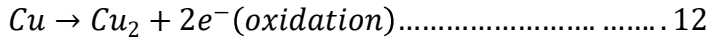
$$H - H = 432 \frac{KJ}{mol} \dots \dots 9$$

$$O - O = 494 \frac{KJ}{mol} \dots \dots 10$$

$$H - O = 460 \frac{KJ}{mol} \dots \dots 11$$

Now, $H_2O \rightarrow H - O - H$, and it has two $H - O$, bonds, and thus the bond enthalpy (energy) for H_2O is
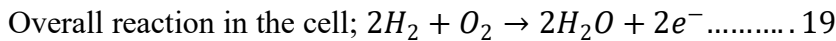
$$= 2 \times 460 \frac{KJ}{mol} = 920 \frac{KJ}{mol}$$

Whereas for $H_2 \rightarrow H - H$, the bond enthalpy is equal to $432 \frac{KJ}{mol}$; and for $O_2 \rightarrow O - O$, it is 494KJ/mol and $\frac{1}{2}O_2$ then has a bond enthalpy $= \frac{1}{2} \times 494 \frac{KJ}{mol} = 247 \, KJ/mol$. In $Eq.\,8$, the left side of reaction has bond enthalpy $= 432 \frac{KJ}{mol} + 247 \frac{KJ}{mol} = 679 \frac{KJ}{mol}$

The right side of $Eq.\,8$ has bond enthalpy $= 920 \frac{KJ}{mol}$. Hence the energy that can be harvested $= -920 \frac{KJ}{mol} + 679 \frac{KJ}{mol} = -241 \frac{KJ}{mol}$, which is negative. There are a number of other chemical reactions that have been used in SOFC systems are;

$$Cu \rightarrow Cu_2 + 2e^- (oxidation)\text{............................}12$$

$$O_{2^-} \rightarrow \frac{1}{2}O_2 + 2e^- (oxidation)\text{............................}13$$

$$O_{2^-} + CO \rightarrow CO_2 + 2e^- (oxidation)\text{........................}14$$

$$CH_4 + 4O_{2^-} \rightarrow CO_2 + 2H_2O + 8e^- \ (oxidation)\text{........}15$$

$$\frac{1}{2}O_2 + H_2O + 2e^- \rightarrow 2(OH)^- (Reduction)\text{...............}16$$

$$H_2 + 2(OH)^- \rightarrow 2H_2O + 2e^- (Oxidation)\text{.................}17$$

From these reactions, we can calculate the energy that can be harvested (conversion of ambient energy). Energy harvesting is the process in which energy is captured from the system's environment and converted into usable electric power.

We will now describe the reactions that are proposed for the fuel cell. The first is;

Cathode reaction; $O_2 + 4e^- \rightarrow 2O$ $\text{...............................}18$

Overall reaction in the cell; $2H_2 + O_2 \rightarrow 2H_2O + 2e^-$ $\text{..........}19$
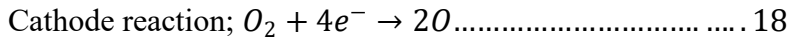
Now we calculate the energy that can be harvested for such a reaction;

$$2H_2 \rightarrow 2(H-H) = 2 \times 432 \frac{KJ}{mol} = 864 \frac{KJ}{mol}$$

$$O_2 \rightarrow O-O = 494 \frac{KJ}{mol}$$

$$\text{Total} = (864 + 494) \frac{KJ}{mol} = 1358 \frac{KJ}{mol} \text{........................}20$$

$$2H_2O = 2(H-O-H) = 2(2 \times 460)\frac{KJ}{mol} = 1840 \frac{KJ}{mol} \text{.........}21$$

$Eq.\,20$ Gives the energy absorbed (or energy required) to break the bonds (endothermic) and is positive (+ve)

$Eq.21$ Gives the energy released to form the bonds (exothermic) and is negative. Thus the energy that can be harvested is: $\frac{KJ}{mol} = -482\frac{KJ}{mol}$ 22

Hence the energy harvested for such a reaction is large, and thus we can get more power from the cell. It will lead to larger velocity of charge carriers resulting in larger current. $I = \eta \varepsilon \upsilon$. Increase in current will lead to higher voltage across the load resistance R

Another reaction could be;

$$2H_2 + 4(OH^-) \rightarrow 4H_2O + 4e^- \dots\dots\dots\dots\dots 23$$

Now we calculate the energy that can be harvested. The energy required to break the bonds (endothermic, +ve) on the left side of $Eq.23$ is;

$$2H_2 = 2(H - H) = 2 \times 432\frac{KJ}{mol} = 864\frac{KJ}{mol}$$

$$4(OH^-) = 4(O - H) = 4 \times 460\frac{KJ}{mol} = 1840\frac{KJ}{mol} \dots\dots\dots\dots 24$$

The energy released (exothermic, -ve) to form the bonds on the right-hand side of $Eq.23$ is:

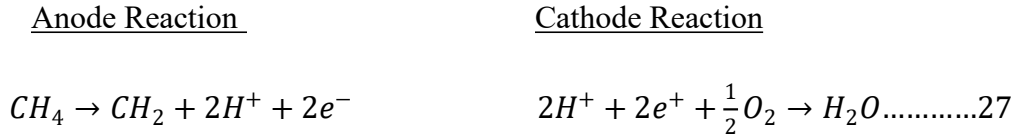$$4H_2O = 4(H - O - H) = 4(2 \times 460)\frac{KJ}{mol} = 3680\frac{KJ}{mol} \dots\dots\dots\dots 25$$

Hence, the energy that can be harvested is: $(2704 - 3680)\frac{KJ}{mol} = -976\frac{KJ}{mol}$ 26

The energy is quite large and thus we can get more power from the cell. In this way more reactions can be designed to get larger harvested energy such that the chemical and thermodynamic equilibrium of the cell is not disturbed. More reactions can be proposed keeping in mind the chemical and thermodynamic properties, the electrical parameters, power output and life time of the cell

Recently, new types of solid oxides are being developed for the fuel cells [16]. These are called perovskite-type oxides (PTO), and they have been researched for the last some 10 years. The cell functions on the basis of deformations that occur due to variations in their ionic radii and electronegativity, which are due to the production of oxygen or cation deficiencies or changes in their bonding angles. The defects and the order-disorder in the crystal structure can lead to large changes in functional properties.

The performance parameters like specific capacity, power conversion efficiency, cycle life and energy density will determine the future scope of applications of these materials [3][16].
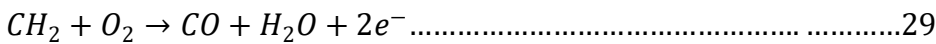
Yet another chemical reaction that is proposed is,

<u>Anode Reaction</u>   <u>Cathode Reaction</u>

$$CH_4 \rightarrow CH_2 + 2H^+ + 2e^-$$ $$2H^+ + 2e^+ + \frac{1}{2}O_2 \rightarrow H_2O \ldots\ldots\ldots 27$$

Thus, the overall (chemical) electrochemical reaction can be;

$$CH_4 + \frac{1}{2}O_2 \rightarrow CH_2 + H_2O + 2e^- \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots 28$$
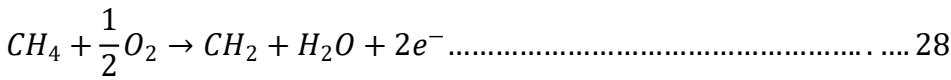
The CH_2 that comes out can be used, and the overall electrochemical reaction can be written as,

$$CH_2 + O_2 \rightarrow CO + H_2O + 2e^- \ldots\ldots\ldots\ldots\ldots\ldots\ldots 29$$

Bond enthalpy for $CH_2 = 2 \times 412 = 824 \frac{KJ}{mol}$ and the bond enthalpy for $O_2 = 494 \frac{KJ}{mol}$.

Thus the energy required to break the bonds (endothermic, +ve) is $1318 \frac{KJ}{mol}$

Bond enthalpy for $CO = 1072 \frac{KJ}{mol}$

Bond enthalpy for $H_2O \ (H - O - H) = 920 \frac{KJ}{mol}$. Thus, the energy released to form the bonds is $1992 \frac{KJ}{mol}$.

Hence,   the   energy   that   can   be   harvested   is $(1318 - 1992) \frac{KJ}{mol} = -$

$674 \frac{KJ}{mol}$ $\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots 30$

The energy is quite large, and thus we can get larger current using $Eq. 7$ for E and the graphs are as shown below in the results section.

3. RESULTS AND DISCUSSIONS

3.1. Effects of Temperature

The dependence of SOFC performance on temperature is discussed in the graph below, this shows that voltage gain with respect to temperature is a strong function of temperature. The voltage gain is given by the equation $\Delta V_T (mV) = 1.3(T_2 - T_1)(^0C)$ for a cell operating at $1000^\circ c$;
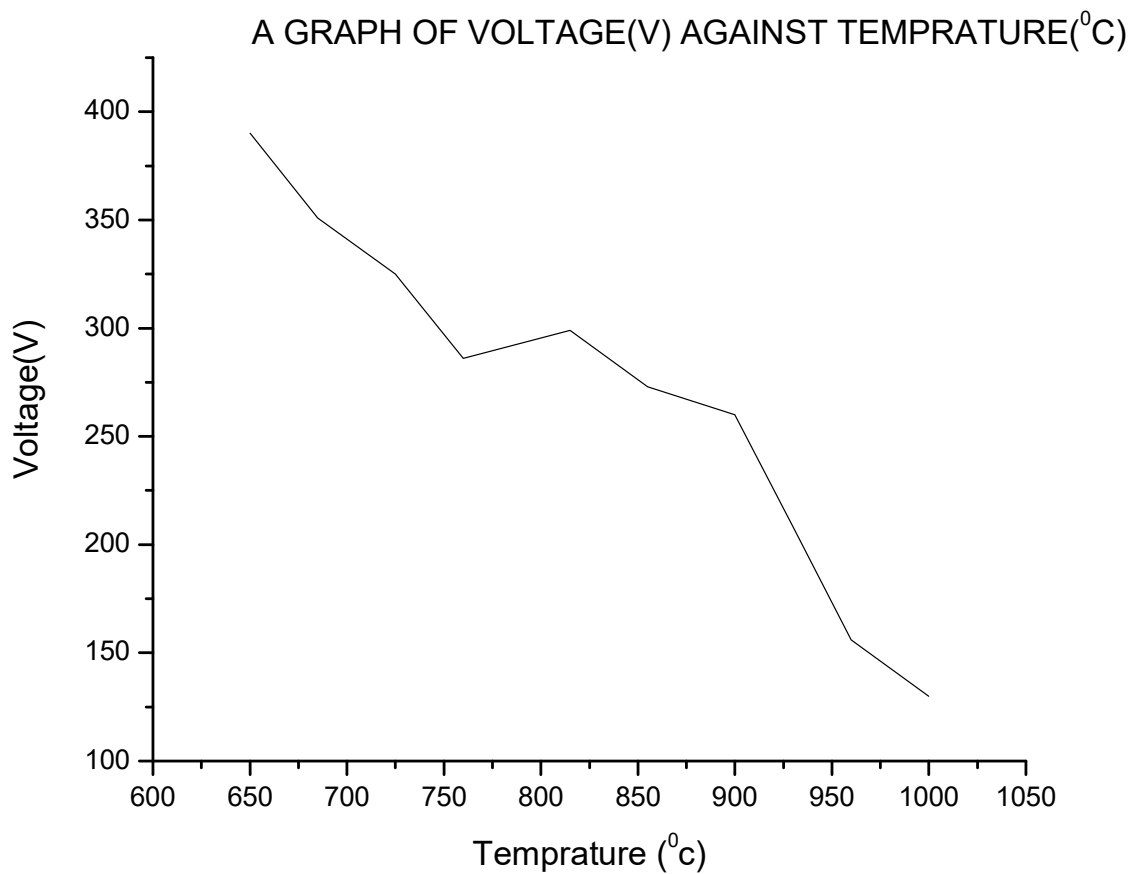

Figure 1. Variation of Voltage (V) with Temperature

3.2. Effects of Pressure

SOFC's show enhanced performance by increasing cell pressure. The following equation approximates the effect of pressure on cell performance at $1000°c$;

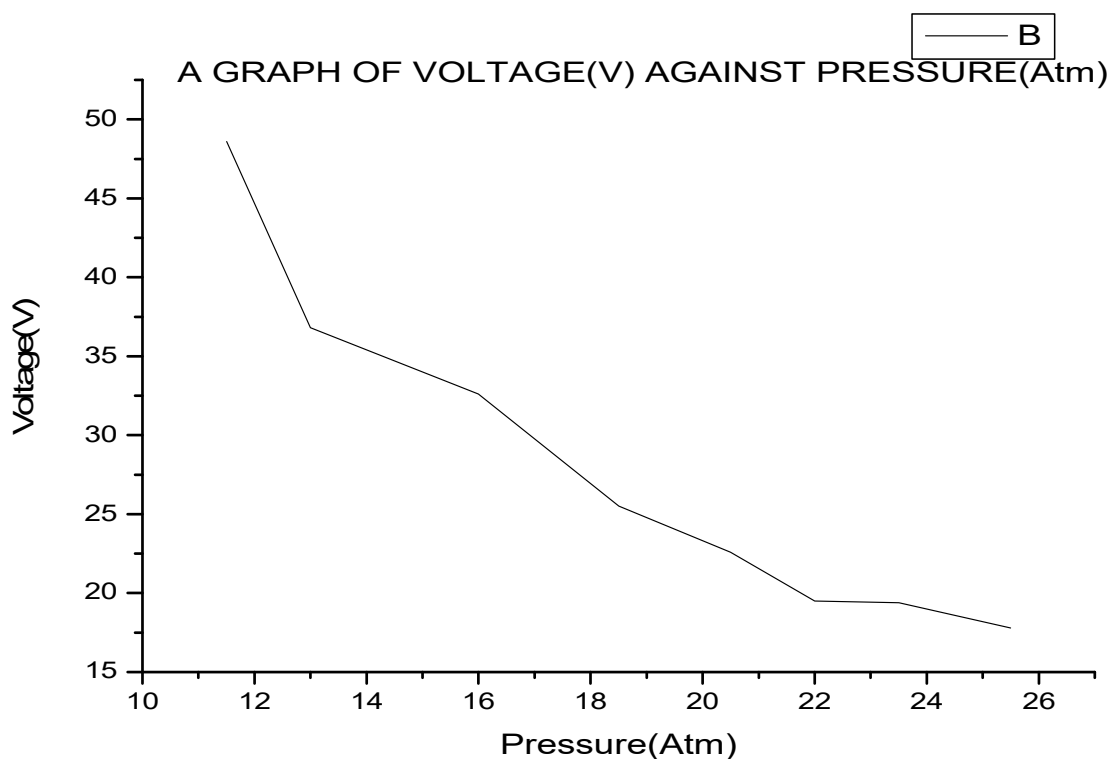$$\Delta V_P(mV) = 59\log\frac{P_2}{P_1}$$ Where P_1 and P_2 are different cell pressures.

Figure 2. Variation of Voltage (V) against Pressure

Pressure P depends on the density of gas and temperature. By varying density of gas and temperature, we can get different values for the pressures used in $Eq.\,7$. As a rule, pressure increases as the density and temperature increase.

The relation is based on the assumption that the potentials (electromotive) are predominantly affected by gas pressure and temperature, and that these potentials decrease with increase of pressure and temperature. (See graphs) Fig. I variation of voltage (V) with temperature, Fig. II Variation of voltage (V) against pressure.

The practical utility of the three proposed reactions can be appreciated only after such cells are fabricated and then its functioning is studied. It is important to understand that the basic reactions in a SOFC are $2H_2 + O_2 \rightarrow 2H_2O + 2e^-$. Thus whatever the proposed reactions, the products must contain H_2O (water or steam) and electrons (e^-) that when they flow through the load, give power, and this process confirms that a fuel cell generates electricity by a chemical reaction.

But the efficiency of SOFC's depends in the rate at which solids and gases interact at the surface of the electrodes constituting the SOFC's i.e the interactions that take place at the anode and cathode of the SOFC. Research will go on to explore ways to improve SOFC efficiency. One method of study is to expose different facet of its crystal structure to oxygen gas at operating pressures and temperatures. An important point of study is whether different reaction rates can be achieved from the same material just by changing which surface the oxygen reacts with [17]. In such cases, oxygen gas molecules adhere to the specially prepared electrode surface, and to examine how the atomic configuration at specific surfaces of these materials makes a difference when it comes to reacting with the oxygen gas at the cathode [18].

It is important to note that SOFC's do not have any corrosive components and do not require precious metal electrolytes since the operating temperatures are in the range of $800°C - 1000°C$ [3][4][20]. Recent research and future efforts are to focus on the design of the functional interfaces in SOFC's; and how to lower the operating temperature such that it lies in the range $400°C - 700°C$.

This can lead to increased lifespan of SOFC's, and or increase in operational durability and efficiency. This can also lead to increased thermal compatibility, thermal cycle capability, and reduce the materials and fabrication costs by using metallic interconnectors in the SOFC stack cell. [20, 21]. In this process, the important requirement is that the electrolyte should be thin, of the order of $10\mu m$ or less [3] with high density such that the ohmic loss of the electrolyte is small. Another requirement is to reduce the excess potentials observed for the O_2 reduction reaction at the cathode in the single cell [18].

4. CONCLUSIONS

Recent studies emphasize [18] that SOFC's can be fueled by externally reforming heavier hydrocarbons such as diesel, jet fuel, CO, CO_2 steam, and methane (CH_4) mixtures.

A reaction using CH_4 is proposed in this paper ($Eq.\,28$). These are formed by reacting the hydrocarbon fuels with steam or air in a device upstream of the SOFC anode. Such fuels can result in highly efficient electrochemical devices that convert the chemical energy of gaseous fuels directly into electrical power, which is the main activity of SOFC's.

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