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Performance analysis of solid oxide fuel cells

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Abstract

Fuel Cells are the only source of electric energy which do not pollute the environment. The fuel cells require Hydrogen and Oxygen, which combine electrochemically to provide electric current, heat energy and water. Fuel Cells are the green energy source, leading to zero polluting emissions. A fuel cell, as compared to ICE or BEV, has much higher performance and well-to-wheel efficiency. A fuel cell when integrated with a powertrain provides all these benefits which are required in an automotive application. Hydrogen being the fuel for energy generation, an FCEV can be refueled in a short time and provides a higher range than a BEV whose charging time is relatively longer. There are many fuel cells which have been the source of power in automotive application, however, SOFC scores many positives over others. Despite Solid Oxide Fuel Cells having a high operating temperature, they can operate with a variety of fuels containing Hydrogen inside. The fuels for an SOFC may include syngas, biogas, coal gas, propane, or natural gas. Unlike PEMFC, SOFC is not sensitive to fuel impurities. This leads to higher SOFC performance and greater efficiency. An SOFC operating at a high temperature is not likely to use expensive catalysts for necessary ionic reactions required inside. However, the heat up time or start up time must be relatively low in an automobile integrated with SOFC powertrain. This paper evaluates heat up or start up time in an SOFC, coupled with the powertrain in automobile integrated with SOFC powertrain.

Keywords: Fuel Cell, SOFC, Overpotential, Power, Performance, Thermodynamics, Efficiency.

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1. Introduction

In the year 1839, Sir William Robert Grove proposed the concept of a fuel cell for the first time. By using electrolysis as the basis, he designed a reverse process that combined hydrogen and oxygen to produce electricity. The first ever patent on fuel cells was filed by Fritz Haber, using a solid electrolyte in 1905. In his patent, glass and porcelain were used as electrolyte materials and platinum and gold as electrode materials. Between 1933 and 1959, Francis Thomas Bacon worked on and demonstrated a fully

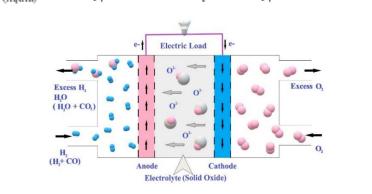
functional alkaline electrolyte fuel cell, AFC. In 1960, NASA used the same AFC technology developed in its Apollo space program. In 1990, the first direct methanol fuel cell, DMFC, was developed by NASA jet propulsion.

Nernst was the first to develop Solid oxide fuel cell (SOFC) in 1899 by incorporating zirconia (ZrO₂) as an ion conductor for Oxygen. The output of a fuel cell is three folds, electricity, heat and water or vapor, operating at a very high temperature 600 – 1000 deg C. In late 1930s, the Swiss scientists, E. Baur and H. Preis worked extensively on Solid Oxide electrolysis by using yttrium, zirconium, cerium, lanthanum, and tungsten oxide. And, in 1937, they came out with the first operational ceramic fuel cell at 1000 deg C. In 1940s, a Russian scientist O.K. Davtyan added monazite sand to the mixture of sodium carbonate, soda glass and tungsten trioxide for increasing the conductivity and mechanical strength of fuel cell but resulted in chemical reactions with shorter life. In 1950s, the Netherlands Central Technical Institute in Hague, Consolidation Coal Company in Pennsylvania and General Electric in New York continued their research on Solid oxide fuel cell technology to achieve stable solid electrolytes. This could resolve the issues of high internal electrical resistance and short circuiting. In view of the high pollution and energy conservation, fuel cells are efficient sources of power without emitting any harmful gases in the environment. This property of fuel cells makes them a strong contender for automotive application. However, the start-up times of Solid Oxide fuel cells may be quite high due to high operational temperatures.

A Solid Oxide Fuel cell may operate indefinitely if it is supplied with an uninterrupted source of hydrogen and oxygen, which is present in the air. Hydrogen atoms, which get disintegrated from a hydrocarbon gas, react with oxygen atoms electrochemically during oxidation to create water. Electrons are released within the process and flow as an electrical current through an external circuit (Ahuja et al., 2022). The fuel which is suitable to be used in an SOFC includes Syngas or Biogas. As a result, the chemical energy in SOFC is directly converted into electricity by fuel cells, the only by-products being pure water and heat, which can also be used as by-products. Solid Oxide Fuel cell systems can be up to 60 percent efficient and even higher when heat is also used for energy regeneration (Fernandes et al., 2018). The major advantage of using Solid Oxide fuel cells is their high efficiency of energy conversion. This is because they have the ability to use even impure fuel as input. Fuel cells and Solid Oxide Fuel Cells in particular, do not make any noise during working due to absence of any moving parts. Since direct combustion doesn't take place inside a fuel cell, there is no emission in terms of products like NOx, SOx and particulate matter. Fuel cells are modular and can be scaled to desired sizes and can meet very high-power requirements as well. All this makes SOFC an obvious choice of fuel cell in automotive applications.

2. SOFC – Solid Oxide Fuel Cell

The working temperature of SOFCs varies from 800 °C to 1000 °C. It works on methane gas or Syngas as fuel and generates energy from direct fuel oxidation and from reforming of fuel to H_2 and CO_2 . The H_2 is then split into Hydrogen ions (H^+) and electrons at Anode. Air is supplied to the Cathode. The oxygen molecules at the Cathode are split into oxygen ions (O^{2-}) and four electrons. When they reach the anode, the oxygen ions re-combine with the hydrogen ions, and heated water is produced. The electrons released at the anode generate the electrical current. For Anodes, a porous layer of composite Ni catalyst and yttria-stabilized zirconia (YSZ) are the most used materials. This is a composite of ceramic and metal and is an ionic conducting material. Due to its porosity, gas phase species can also be passed through it. For Cathodes, a porous composite mixture of LSM (lanthanum strontium manganite) and YSZ (yttria-stabilized zirconia) is the most used materials. For Electrolytes, ceramic mixed metal oxides are the most used materials. The most popular SOFC electrolyte, YSZ is exclusively used in fuel cells operating above 750 deg C. The combustion of hydrogen has an HHV of 285.8 kJ/mole. However, the Gibbs free energy (GFE) for the reaction is only 237.2 kJ/mole, which is the maximum electricity produced by a fuel cell. Therefore, the difference, 48.6 kJ/mole, is released as heat energy, which can be used outside the fuel cell. In a Solid Oxide fuel cell, the overall electrochemical reaction may be expressed as:



$$H_2 + \frac{1}{2}O_2 \rightarrow H_2O_{(\text{tianuid})} + 237.2 \text{ kJ/mole electricity} + 48.6 \text{ kJ/mole heat}$$

(1)

Figure 1. Sold Oxide Fuel Cell (SOFC) (Wachsman and Singhal)

3. SOFC Flow Equations

In an SOFC, the electrochemical reaction converts the fuel gas supplied into Hydrogen, which then disintegrates into Hydrogen ions and electrons, which produce electrical energy. The coal syngas is primarily a mixture of hydrogen and CO. It has significant water vapor and some levels of CO_2 and other species in very small quantities. When H_2 oxidation takes place at Anode, it contributes to electrochemical power generation. At the same time, Methane (CH_4) is reformed to CO and H_2 , conforming to the steam reforming reaction. CO, when reacts to steam, releases CO_2 and H_2 . Consequently, there exists three simultaneous reactions in a fuel cell - steam reforming reaction for methane, the water-gas shift reaction, and the electrochemical reaction. These reactions in an SOFC are enumerated as follows:

$$CH_4 + H_2O \to CO + 3H_2 \text{ (steam reforming)} \tag{2}$$

$$CO + H_2O \rightarrow CO_2 + H_2 (water gas shift)$$

$$H_2 + 1/2O_2 \rightarrow H_2O (overall cell reaction)$$

$$(2)$$

$$(3)$$

$$(4)$$

$$H_2 + 1/2O_2 \rightarrow H_2O$$
 (overall cell reaction) (4)

The amount of hydrogen consumed in the fuel cell reactions, $\dot{n}_{H_{2-n}}$ (mol/s), as per Faraday's law is enumerated as:

$$\dot{n}_{H_{2-\sigma}} = \frac{tA}{n_{\sigma}F} \tag{5}$$

The amount of Hydrogen supplied for a known Fuel utilization factor U_f is given as:

$$\dot{n}_{H_{2-\sigma}} = \frac{\dot{n}_{H_{2-\sigma}}}{U_f} = \frac{iA}{n_\sigma F U_f} \tag{6}$$

The air stream molar flow rate is then calculated as:

. .

$$\dot{n}_{a-in}(O_2) = \dot{n}_{H_{2-s}} \frac{v_{f/2}}{v_a}$$
(7)

The fuel stream molar flow rate required to produce the supplied amount of hydrogen is then enumerated as:

$$\dot{n}_{f-in} = \frac{\dot{n}_{H_{2-s}}}{x_{fc}} = \frac{iA}{n_{e}FU_{f}x_{fc}} \quad \dots \quad [\frac{mol}{s}]$$
(8)

$$x_{fc} = x_{H_z} + x_{c0} + 4x_{CH_z}$$
(9)

The molar flow rate for each component j for an identified fuel gas composition x_j , in the fuel stream is calculated as:

$$\dot{n}_{f-in}(j) = \dot{n}_{f-in} x_j = \frac{iAx_j}{n_e F U_f x_{fe}} \qquad \dots \quad \left[\frac{mol}{s}\right]$$
(10)

where:

• $j = \{H_2, CO, CH_4, CO_2, H_2O, N_2\}$

To avoid carbon deposition in an SOFC, more amount of steam is required. The quantity of steam required is nearly twice the amount needed for the reforming and water-gas shift reactions. The required molar flow rate of steam is thus enumerated as:

$$\dot{n}_{H_2 0} = (\dot{n}_{f-in}(CO) + \dot{n}_{f-in}(CH_4) * 2) * 2 \quad \dots \quad [\frac{mal}{s}]$$
(11)

The additional steam molar flow rate supplied is hence given by:

$$\dot{n}_{ADD} = \dot{n}_{H_2O} - \dot{n}_{f-in}(H_2O) = (\dot{n}_{f-in}(CO) + \dot{n}_{f-in}(CH_4) * 2) * 2 - \dot{n}_{f-in}(H_2O)$$
(12)

Therefore, the total molar flow rate of the fuel stream entering the fuel cell is given by:

$$\dot{n}_f = \dot{n}_{f-in} + \dot{n}_{ADD} \quad \dots \quad \left[\frac{mal}{s}\right] \tag{13}$$

4. SOFC Power Equations

In an SOFC stack, the open-circuit voltage is the maximum operating voltage when no current is flowing through the external circuit. The Nernst equation is defined as the relationship between the Standard potential E^{a} and the open-circuit voltage, which can be determined at partial pressures of reactants and products at temperature T_{c} :

$$V_{id} = E^{o} + \frac{RT_{c}}{nF} \ln \frac{[H_{2}][Q_{2}]^{1/2}}{[H_{2}O]}$$
(14)

where:

$$E^{o} = 1.272 - \frac{2.764T_{c}}{10^{4}} \tag{15}$$

The Nernst potential has losses, which are irreversible, when the electrical cell circuit is closed. The losses include the ohmic resistance losses of the cell elements, the activation losses at the electrodes and the concentration polarization losses. Thus, the cell voltage is calculated as:

$$V_{Cell} = V_{id} - \Delta V_{act} - \Delta V_{ohm} - \Delta V_{conc}$$
(16)

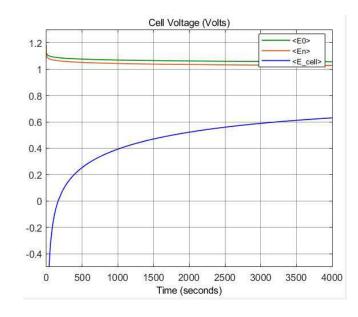


Figure 2. Sold Oxide Fuel Cell (3.5 kW) Voltage vs Time

Activation overpotential is calculated according to the general Butler–Volmer equation. It depends on the kinetics of the electrochemical reactions occurring at the anode and cathode. The respective activation overpotentials of the anode and cathode can be calculated as:

$$V_{act.a} = \frac{2RT_{\sigma}}{n_e F} \sinh^{-1}\left(\frac{i}{2t_{0.a}}\right) \tag{17}$$

$$V_{act,c} = \frac{2RT_c}{n_c F} \sinh^{-1}\left(\frac{i}{2i_{0,c}}\right) \tag{18}$$

$$i_{0,j} = k_j \cdot exp \left(-\frac{E_j}{R.T_c}\right) \quad \dots \cdot \left[\frac{A}{m^2}\right]$$
 (19)

where:

$$k_{an} = 6.54. \ 10^{13} \cdot \left(\frac{R.T_c}{2F}\right) \quad \dots \cdot \left[\frac{A}{m^2}\right] \tag{20}$$

$$E_{an} = 1.4.10^5 \dots \left[\frac{j}{mal}\right]$$
 (21)

$$k_{car} = 2.35.10^{13} \cdot \left(\frac{RT_c}{2.F}\right) \quad \dots \cdot \left[\frac{A}{m^2}\right]$$
 (22)

$$E_{cat} = 1.37.10^5 \dots \left[\frac{I}{mal}\right]$$
 (23)

$$\Delta V_{act} = V_{act,a} + V_{act,c} \tag{24}$$

The *ohmic over-voltages* are expressed by the Ohms law:

$$\Delta V_{ohm} = \frac{l}{A} \left(\frac{\tau_e}{\sigma_s} + \frac{\tau_a}{\sigma_a} + \frac{\tau_c}{\sigma_c} + \frac{\tau_{int}}{\sigma_{int}} \right) \tag{25}$$

where:

• σ_i = Material Conductivity, calculated with a temperature-dependent relation

$$\sigma_e = C_{1e} \exp\left(\frac{C_{2e}}{r}\right) \tag{26}$$

$$\sigma_a = \frac{c_{1\alpha}}{r} \exp\left(\frac{c_{2\alpha}}{r}\right) \tag{27}$$

$$\sigma_c = \frac{c_{1c}}{T} \exp\left(\frac{c_{2c}}{T}\right) \tag{28}$$

$$\sigma_{int} = \frac{c_{1int}}{r} \exp\left(\frac{c_{2int}}{r}\right) \tag{29}$$

The concentration overpotential at the anode and cathode have been included in the evaluation of are enumerated as:

$$\Delta V_{conc_{An}} = \frac{RT_c}{2F} \ln(1 - U_f) \tag{30}$$

$$\Delta V_{conc_{Cat}} = \frac{RT_c}{2F} \ln \left(1 - U_f \cdot U_a\right) \tag{31}$$

$$\Delta V_{conc} = \Delta V_{conc_{An}} + \Delta V_{conc_{Car}}$$
(32)

While evaluating the performance of a fuel cell, the fuel utilization factor is defined as:

$$U_{f} = \frac{x}{(H_{z}^{i} + CQ^{i} + 4CH_{z}^{i})}$$
(33)

$$U_a = \frac{z}{2.\theta_2^l} \tag{34}$$

where, each mole of CH_4 generates 4 moles of H_2 (3 by reforming and 1 by shift), as per equations (2) and (3).

The Utilization Factor, U_f is pre-assigned for evaluation of fuel cell performance. By using above equation, it is, therefore, possible to calculate z and to obtain the electrical current of the cell as:

$$I_{TOT} = z.2.F \tag{35}$$

The FC output power is then calculated as:

20

$$P_{sofc} = V_{Cell} . I_{TOT}$$
(36)

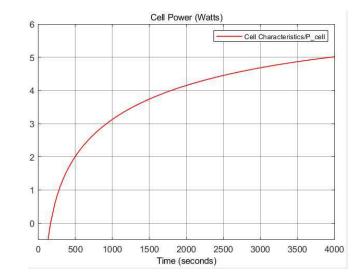


Figure 3. Sold Oxide Fuel Cell (3.5 kW) Power vs Time

The SOFC electrical efficiency is then calculated as:

$$\bigcap_{E,sofc} = \frac{P_{sofc}}{\dot{m}_{f-in}(CH_4).LHV_{CH_4} + \dot{m}_{f-in}(CO).LHV_{CO} + \dot{m}_{f-in}(H_2).LHV_{H_2}}$$
(37)

5. Fuel Cell Thermodynamics

The specific enthalpy and entropy for H_2/O_2 fuel cell are given by:

$$\Delta H = [h_{f,H_20}^0 + C_{p,H_20} (T_c - 298.15)]_{H_20} - \frac{1}{2} [h_{f,0_2}^0 + C_{p,0_2} (T_c - 298.15)]_{O_2} - [h_{f,H_2}^0 + C_{p,H_2} (T_c - 298.15)]_{H_20}$$
(38)

$$\Delta S = \left[s_{f,H_20}^0 + C_{p,H_20} \ln\left(\frac{T_o}{298.15}\right) \right]_{H_20} - \frac{1}{2} \left[s_{f,0_2}^0 + C_{p,0_2} \ln\left(\frac{T_o}{298.15}\right) \right]_{\theta_2} - \left[s_{f,H_2}^0 + C_{p,H_2} \ln\left(\frac{T_o}{298.15}\right) \right]_{H_2}$$
(39)

For Hydrogen, H_2

$$C_{pR_2}(T) = 3.057 + 2.677 * 10^{-3}(T) - 5.810 * 10^{-6}(T)^2 + 5.521 * 10^{-9}(T)^3 - 1.812 * 10^{-12}(T)^4$$
(40)

For Oxygen, O_2

$$C_{p,0_2}(T) = 3.626 - 1.878 * 10^{-3}(T) + 7.055 * 10^{-6}(T)^2 - 6.764 * 10^{-9}(T)^3 + 2.156 * 10^{-12}(T)^4$$
(41)

For Water, $H_2 O$

$$C_{p,H_20}(T) = 4.070 - 1.808 * 10^{-3}(T) + 4.152 * 10^{-6}(T)^2 - 2.964 * 10^{-9}(T)^3 + 0.807 * 10^{-12}(T)^4$$
(42)

The heat is generated by chemical reaction during the water formation at anode side. It is calculated as:

$$Q_f = \frac{I}{2E} T_c \Delta S \tag{43}$$

Heat generation by ohmic losses due to ohmic resistance, at anode (Q_a) , cathode (Q_e) , the electrolyte (Q_e) and at the Interconnect (Q_{int}) is then enumerated as:

$$Q_a = R_a I^2 \tag{44}$$

$$Q_c = R_c I^2 \tag{45}$$

$$Q_e = R_e I^2 \tag{46}$$

$$Q_{int} = R_{int} I^2 \tag{47}$$

where:

$$R_i = \rho \frac{\tau}{A} \tag{48}$$

And,

$$Q_{TOT} = Q_f + (Q_a + Q_c + Q_e + Q_{int})$$
(49)

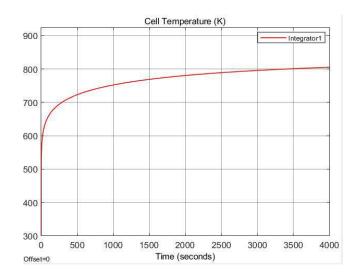


Figure 4. Sold Oxide Fuel Cell (3.5 kW) Temperature vs Time

And,

$$Q_{TOT} = M. C_s. \frac{dT_c}{ds}$$
(50)

By solving the differential equation, T_c can be plotted w.r.t. time.

The thermodynamic efficiency of the Fuel cell is then calculated as:

$$\bigcap_{Tsofc} = \frac{Actual \ electrical \ work}{Maximum \ available \ work}$$
(51)

$$\bigcap_{T,sofc} = \frac{\Delta G}{\Lambda H} = \frac{\Delta H - T\Delta S}{\Lambda H} = 1 - \frac{T\Delta S}{\Lambda H}$$
(52)

6. Conclusions

The performance of an SOFC is largely governed by its Cell temperature. As the Solid Oxide Fuel Cells work at high temperatures, their heat-up time becomes a challenging factor for many applications, particularly in automotives. Moreover, the rise of Cell temperature also depicts the thermal stresses in various components of fuel cell. All it is required is a shorter start-up time with less thermal stress. Automobiles have an obvious reason to have the minimum possible start-up time of a Fuel Cell. The start-up time may be minimized either by integration strategy on a vehicle or by changing the chemical composition of materials or electrolyte.

Nomenclature

- *i* Current density
- A Active area of element
- n_{p} No. of electrons transferred per mole of reactant
- *F* Faraday constant (96,485 Coulomb/mole)
- x_{fc} Number of moles of hydrogen produced by 1 mole of fuel
- T_e Cell Temperature (K)

- i_0 Exchange current density for each electrode 'j' (anode or cathode)
- k_i Pre-exponential factor represented by a first order polynomial temperature function
- $\boldsymbol{\mathcal{E}}_i$ Activation energy of each electrode 'j' (anode and cathode), represented by a constant
- T₁ Elements thickness
- σ_t Material Conductivity, calculated with a temperature-dependent relation
- C_{1a}, C_{2a} Anode Conductivity constants [95 x 10⁶; -1150]
- C_{1c}, C_{2c} Cathode Conductivity constants [42 x 10⁶; -1200]
- C1e, C2e Electrolyte Conductivity constants [3.34 x 10⁴; -10,300]
- C_{1int}, C_{2int} Interconnect Conductivity constants [9.3 x 10⁶; -1100]
- U_f Fuel Utilization Factor
- *U*_a Oxygen utilization factor
- \mathbb{Z} Number of H_2 moles reacting

 $\dot{m}_{f-in}(j)$ mass flow rate for each component j in the fuel stream

- *h*⁹ Molar enthalpy of formation at 298.15 K
- s_f^0 Molar entropy of formation at 298.15 K
- *I* Cell Current (A)
- △S Entropy of water formation reaction (J/mol-K)

 $R_{ar}R_{er}R_{er}R_{inc}$ Anode, Cathode, Electrolyte, and Interconnect ohmic resistance

- ρ Material resistivity = $1/\sigma$
- M Mass of the Cell (kg)
- C_s Equivalent average specific Heat coefficient (J/kg K)

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