



Solid Oxide Fuel Cell systems for electricity generation: An optimization prospect

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ABSTRACT

The present study concerns with an optimization method of Solid Oxide Fuel Cell (SOFC) systems for electricity generation, directly fed by CH₄ (methane) or C₂H₅OH (ethanol). By considering a detailed thermodynamic analysis, an innovative simulation model, namely THERMAS (THERmodynamic MATHematical Simulation), was designed and implemented. A specific SOFC-based system was selected to be the reference simulation, which is assumed to be equipped with heat exchangers (preheaters), a reformer, a SOFC-stack system and an afterburner. THERMAS allows for an extended parametric analysis in terms of energy and exergy and offers the opportunity to investigate all the operational characteristics that affect system's efficiency. The optimization process relies on the difference between the energy and exergy efficiency, where an OPF (OPTimization Factor) has been introduced and particularly estimated for each simulated scenario, based on several operational parameters, such as fuel composition, extension of chemical reactions and temperatures. The introduction of this OPF is actually an innovative improvement towards an easy-to-use optimization of SOFC systems. It is found that the design of a SOFC-based power plant fueled by pure hydrocarbons, has to be carefully simulated before its, otherwise the purification of fuels sounds meaningless.

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1. Introduction

It is widely known that fuel cells are devices that directly convert chemical energy of the feeding fuel to electricity without Carnot limitations [1–3]. CH₄ (methane) and C₂H₅OH (ethanol) are hydrocarbons rich fuels while they stimulate a reliable alternative to fuel options. The increased flexibility on fuel choice that SOFC (Solid Oxide Fuel Cells) advantageously present [4,5], strengthen further the investigation of several fuels utilization. The direct comparison among these scenarios will reveal the most efficient case study under similar operational conditions.

Experimental results on SOFC based projects fed with pure CH₄ or C₂H₅OH to produce electric energy reveal theoretical efficiencies up to 60%–90%, either when electricity is combined with heat production or not [6–11]. Although impressive, these findings do not refer to optimized systems, suffering by a lot of roughly attained or actually non-realistic assumptions, such as 100% pure hydrogen, poor consideration of water gas-shift reaction, poor description of

the reactions take place in the burner, 100% insulation of the system, etc. [6,8]. To improve electricity production processes, it is of great importance to thermodynamically identify the portion of the total energy that is able to produce useful work (i.e. exergy), excluding the irreversibilities (thermal and other energy losses) [12–14].

Under this prospect the basic aim of the presented study is the use of an innovative detailed thermodynamic model, named THERMAS (THERmodynamic MATHematical Simulation), in order to simulate and optimize a SOFC-based system by comparing its operation through the use of different fuels, methane and ethanol. The innovation of this process involves real life operational conditions without theoretical restrictions on the values of the several parameters, extensive parametric analysis based on fundamental energy and exergy theory, as well as the use of an innovative parameter named OPF (OPTimization Factor). This scientific approach offers the opportunity to choose a great variety of different values for each operational parameter individually, thus allowing for studies within unexplored and experimentally impossible operational ranges, while innovatively introduces exergy efficiency to identify the optimal scenario per system. It is important to note that a specific effort has been put here to

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generalize the presented approach in order to be applicable in any SOFC-based system for electricity production.

2. Theory

Energy balance theory supports several theoretical studies on different engineering processes, as well as for the needs of the current modelling this will constitute one of the main criterion which has to be satisfied for each individual process during system's operation. The general energy balance which has to be followed over the devices can be expressed as [15]:

$$\left\{ \sum_i \left[(\Delta H_f^0)_i + \int_{T_0}^{T_{in}} m_i (\tilde{C}_p)_i dT \right] + Q + \sum_r |\Delta H_{lex}|_{in} \right\} = \left\{ \sum_i \left[(\Delta H_f^0)_i + \int_{T_0}^{T_{out}} m_i (\tilde{C}_p)_i dT \right] + Q + \sum_r \Delta H_{end} \right\}_{out} \quad (1)$$

where ΔH_f^0 is the enthalpy of formation at reference state, \tilde{C}_p is the mean molar specific heat capacity, ΔH refers to the enthalpy changes of the reactions which are characterized by positive or negative values due to the exothermic and endothermic character according to each process. Also in the above equation, the magnitude Q describes either the essential thermal energy for the finalization of a specific process or the thermal losses to the environment while the reference state for the calculations is considered the environmental standard conditions ($T_0 = 298K$ and $P_0 = 1atm$). By assuming ideal gases, the mean molar specific capacity can be calculated through NASA Polynomials with the appropriate specific coefficients for each chemical element [16].

The above theory based on the first law of thermodynamics constituted the major tool on the simulation and optimization of several engineering systems for numerous decades until the second law of thermodynamics, known as exergy analysis, acquired practical significance in the optimization of energy systems [17].

Exergy is actually a thermodynamic property that describes the quality of the produced energy corresponding to the maximum useful work provided by a system during to its reversible transition to a thermodynamic state in equilibrium with its environment [18]. The exergy analysis (availability analysis) determines in general the energy losses (i.e. energy) due to the existence of several irreversible mechanisms during operation, like combustion process [19]. Exergy depends on both the states of the system and its environment while exergy calculation considers processes of thermal, mechanical and chemical character and it is convenient, however, to be separated into two terms. More precisely, physical exergy, e_{ph} , expresses the useful work that a chemical element can produce if it is brought reversibly from the state of the system to the "restricted dead state", which is a state in thermal and mechanical equilibrium with the environment and can be generally expressed as [20]:

$$e_{ph} = m \left\{ \int_{T_0}^T \tilde{C}_p dT - T_0 \left[\int_{T_0}^T \frac{\tilde{C}_p}{T} dT - R \ln \left(\frac{P}{P_0} \right) \right] \right\} \quad (2)$$

On the other hand, chemical exergy e_{ch} , expresses the useful work that the chemical elements, per stream in each specific case study, can produce if it is brought reversibly in chemical equilibrium with the environment. It is essential to be used an appropriate "exergy reference environment" in order to be estimated the standard chemical exergy e_0 according to the following expression [20]:

$$e_{ch} = m_{tot} \left(\sum_i x_i (e_0)_i + RT_0 \sum_i x_i \ln x_i \right) \quad (3)$$

Even if the characteristics of each device are unique, a general approach on the exergy balance can be mathematically modelled in general for all the processes in each operational step. The expression which has to be satisfied in a SOFC based system for each device separately, while ignoring kinetic and potential energies during the present simulations (THERMAS modelling), can be described as [20]:

$$\left[\sum_k \left(e_{ch} + m_{tot} \Delta T \sum_i x_i (\tilde{C}_p^e)_i \right) \right]_{in} + e^Q = \left[\sum_k \left(e_{ch} + m_{tot} \Delta T \sum_i x_i (\tilde{C}_p^e)_i \right) \right]_{out} + I + W_{el} \quad (4)$$

where e^Q is an exergy term associated with heat transfer when a device interchange energy with its environment, I is the irreversibility rate associated with heat losses which describes the amount of exergy destruction, while \tilde{C}_p^e is the mean isobaric exergy capacity calculated for each chemical element through the expression [21]:

$$\tilde{C}_p^e = \frac{1}{T - T_0} \left(\int_{T_0}^T \tilde{C}_p dT - T_0 \int_{T_0}^T \frac{\tilde{C}_p}{T} dT \right) \quad (5)$$

Finally, the term W_{el} in Eq. (4) represents the amount of chemical energy which transforms into useful energy (electric load) and can be directly calculated, in order to be incorporated in THERMAS as follows [20]:

$$W_{el} = \sum_r \left[- (H_{prod} - TS_{prod}) + (H_{react} - TS_{react}) \right] \quad (6)$$

where H is the enthalpy and S the entropy terms, both calculated by NASA polynomials [16].

3. The SOFC-based systems

The above presented theory was incorporated in a computational basis and constitutes an innovative software simulation model, potentially applicable in any SOFC-based system. As a specific case study, we consider here a biogas-fed SOFC power plant. In this context, an extensive parametric analysis on each dynamic operational variable has to be performed. For the sake of comparison between CH_4 and C_2H_5OH , a biogas fed system based on a SOFC-stack was designed under real life operational scenarios without theoretical restriction. This leads us to present a system which is capable to thermally interact with its environment and its uniqueness is enforced by the fact that its operation can entirely be adjusted under the user's needs. For the sake of completeness, it has been assumed that the theoretical open circuit voltage is 1.23 V at 298 K (in practice is approx. 1 V) while, under load conditions, the cell voltage is between 0.5 and 0.8 V. Obviously, these values do not affect the results.

The entire fuel feeding system of the ongoing simulated project consisting of methane or ethanol (almost purified fuel with low carbon dioxide and moisture content) while air and water keeps separate inlet streams, each passing throughout a heat exchanger to reach the desirable functional temperature. Furthermore, an internal reforming device gets in contact (back-to-back) with an afterburner and combined with a fuel cell stack finalize the entire power plant (Fig. 1).

From chemistry point of view, in order to be evaluated the realistic operational approach of the presented CH_4 -fed system, it is

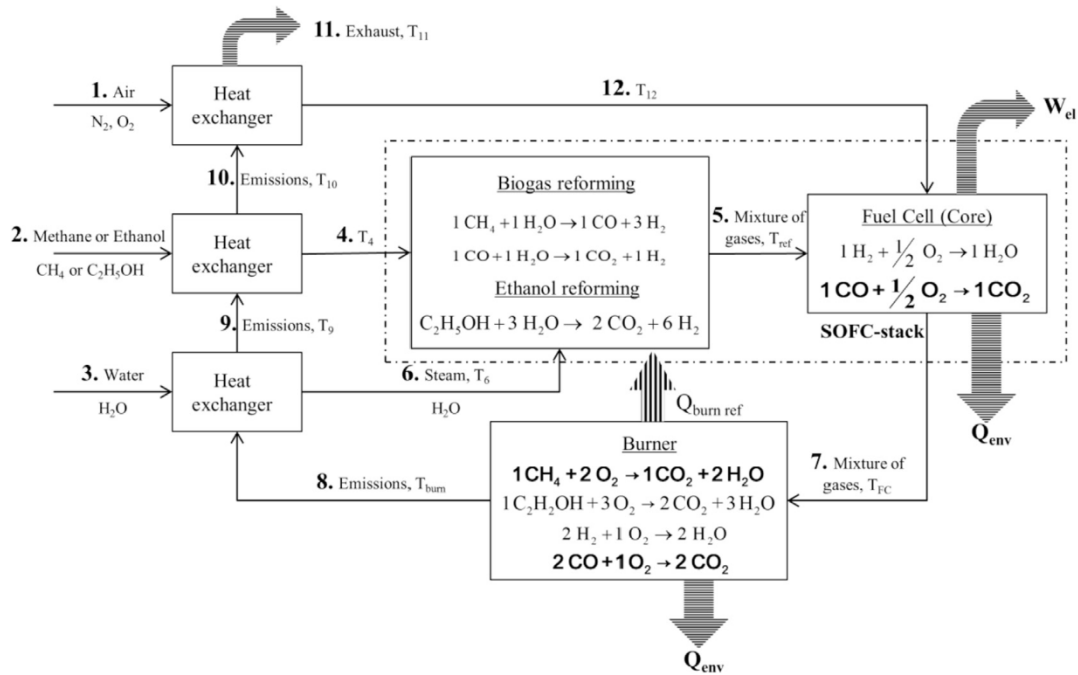


Fig. 1. Flow chart of the power plant.

crucial to be mentioned that the present simulated project is additionally characterized by the WGS (water gas-shift) reaction in the reformer's bulk phase. Due to this fact the bold chemical reactions, as presented in Fig. 1, have to be completely omitted from the system simulation when C₂H₅OH is used as the fuel source. Also, it is important to be highlighted that while mathematically programming the system operation all the reactions' extensions, the operational temperatures per stream and of all individual units can dynamical change their values according to the user's needs and the architecture of each device. These numerous options per variable can lead the research to unexplored pathways which experimentally is hard to be investigated due to the arisen restrictions during the real life design of such a system.

4. Results & discussion

Since previous research attempts incorporate specific computational models based on exergy theory under several theoretical operational assumptions the current research study, apart from incorporating an innovative simulation software tool under a more realistic prospect, aims at describing an optimization process in terms of OPF values. This value represents the absolute difference between the energetic and exergetic efficiency of a given system, as the following Eq. (7) describes:

$$OPF = (n_{en} - n_{ex})100 \quad \text{with} \quad -100 < OPF \leq 100 \quad \text{and} \quad n_{en} \neq 0 \quad (7)$$

where n_{en} is the power produced by fuel cell divided by the total internal energy of input fuel and n_{ex} is the electric power produced by fuel cell divided by the amount of internal exergy of the input fuel.

The above factor actually describes the optimization level of a system: the lower the OPF value, the better the optimization. In other words, OPF tends to zero for optimal system's operation, since exergy efficiency (i.e. useful work divided by useful energy entered the system) approximates energy efficiency in that case. Obviously

a system which is out of order is also described by a zero OPF value, not characterized as optimized due to zero energy production, which corresponds to an optimization process that is actually meaningless. Additionally, the upper limit of the above presented factor characterizes a heating system (i.e. a wood burning stove) where the entire internal energy of the initial fuel transforms into heat, contrary to the lower limit which represents a system with low energy potential, transformed almost totally into useful work. Under this respect, OPF could be considered as a powerful engineering tool since it adequately describes both the energetic and exergetic behavior of the system, allowing therefore for an ease control of all the operational parameters.

4.1. Validation

For validation purposes, a pure CH₄-fed case study presented by Douvatzides et al. [8] which is similar to the present project (see Fig. 1) was simulated under the same theoretical restrictions. The specific system's operational characteristics analytically presented

Table 1

Validation of THERMAS model compared with already existing theoretical results (Douvatzides, 2004).

	CH ₄ -Scenario		C ₂ H ₅ OH-Scenario	
	Previous Model	THERMAS	Previous Model	THERMAS
T ₄ (K)	1165		1140	
T ₅ (K)	1150		1112	
T ₆ (K)	1165		1140	
T ₇ (K)	1200		1200	
T ₈ (K)	1200		1200	
T ₉ (K)	-	1050	-	1070
T ₁₀ (K)	947	915	934	905
T ₁₁ (K)	617	614	729	727
T ₁₂ (K)	800		625	
Q _{env.fc} (%)	0	22.69	0	17.96
Q _{env.burn} (%)	0	0.92	0	0.81
n _{en} (%)	69.20	78.17	73.10	79.82
n _{ex} (%)	66.90	56.07	66.50	55.64

in Table 1, as well as the calculated efficiencies through both models while extensions of H₂ (hydrogen) reforming and oxidization are equal approx. to 95% for each simulated scenario. For the needs of this validation, the inlet temperatures of CH₄-steam and air flows was considered at 298 K, while WGS and CO (carbon monoxide) electrochemical oxidization reactions during CH₄ usage were neglected, as in Ref. [8].

It is worth noticing that both approaches are characterized by an almost identical temperature T₁₁ (see Fig. 1) for the emitted gas mixture, equal to 617 K (previous model [8]) versus 614 K (THERMAS) and 729 K (previous model [8]) versus 727 K (THERMAS) for CH₄ and C₂H₅OH scenarios, respectively. It has to be mentioned that the thermal losses tent to zero for both fuels fed the system in Ref. [8], on the contrary to THERMAS model which confirms that this cannot constitute a realistic prospect. By incorporating an atomizer supplied by the entire amount of the extra produced thermal energy by the afterburner, Douvatzides et al. [8] apparently have been reduced the environmental thermal losses and the irreversibility rate, which influences the exergy destruction and energy efficiencies. Due to this fact a more realistic modelling for such a system adopts a rate of correction on these values, as Table 1 shows.

More precisely the results on the energetic efficiency, as they arisen from the above presented theoretical approaches, vary from 6.72% (C₂H₅OH-Scenario) up to 8.97% (CH₄-Scenario), while this difference on the exergetic efficiency is at approximately 10% for both scenarios. Also, the successful validation process reveals that the temperatures of the flue gases calculated through both computational tools are almost identical, with a negligible difference between 2 K and 3 K, while the energetic exergetic efficiencies follows the same behavior. THERMAS modelling presents a slightly improved energetic behavior, while a low increase of exergy destruction has been reported, mainly due to the reconsideration of the adiabatic processes assumed in Ref. [8].

4.2. The optimization context

By considering the above analysis, the presented simulation tool seems to have the ability to predict realistic operational results by incorporating the entire energy and exergy theory as it is presented previously. Also it is able to reveal the optimization level for each case study and how important is for several operating parameters to be investigated and examined from scratch before an ongoing establishment in order to totally evaluate the entire behavior of such a system under real life.

For direct comparison purposes, the input mass flow rate has been assumed constant (1 kg s⁻¹) in any feed-stream case and the pressure has been set to 1 Atm. As concern the steam and air flow, they have been adjusted so as to totally satisfy the main reactions (reforming and H₂ electrochemical oxidization) under stoichiometric conditions. Also for all the simulated scenarios in order to be comparable methane fuel usage over ethanol the temperatures T_{ref}, T_{FC}, T_{burn} and T₁₂ will be kept at 1200 K, 1100 K, 1000 K and 1000 K respectively.

The use of THERMAS model offers the opportunity to dynamically change the extensions of each chemical reaction individually per simulated scenario. An accurate correlation between real life operation and theoretical simulation of a methane case study is characterized by: a) the almost constant extension of the WGS reaction, over 90% (bulk phase of the reformer) and b) the extension of the electrochemical oxidization reaction of CO (anode of SOFC), at approx. 15%, with a validation up to 1/4 of the extension of the hydrogen reaction [22].

By considering several CO electrochemical oxidization reaction's values, in Fig. 2, the energetic efficiencies (cross-signed line) and

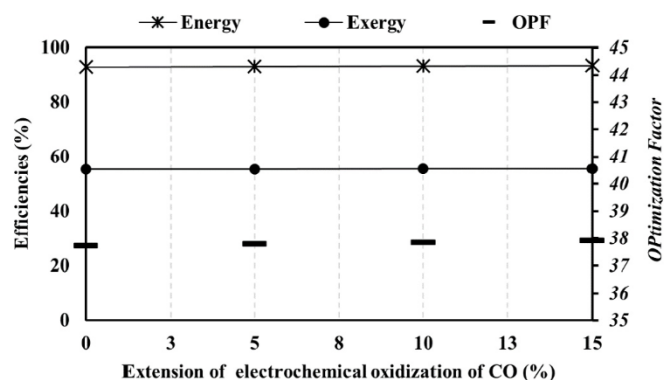


Fig. 2. The effect of electrochemical oxidization of CO on system's behavior.

the exergetic one (circle-signed line), as depicted through the primary y-axis, seems to be not influenced by the extension of electrochemical reaction, since WGS reaction consumes almost all the available toxic carbon monoxide. Almost the same constant behavior is followed by the system's optimization level (OPF in secondary y-axis of Fig. 2, reported with thick dashes). Note that the above notation is followed for all the next presented.

By considering WGS reaction during its operation the energetic and exergetic efficiency can be improved by 17.32% and 10.30% respectively while the OPF was destructed only by 7% as it was graphically presented at the secondary y-axis through Fig. 3. Also, as the same figure presents, it is worth noticing that the lines which represents energy and exergy efficiency, upper and lower respectively, are not parallel. This means that the extension of WGS reaction has less impact on the exergetic efficiency rather than on the energetic one. This chemical reaction is actually an internal process during system's operation and does not generate additional irreversibilities (e.g. thermal exchanges) among several devices which drastically influence the exergy destruction.

Fig. 4 depicts the effect of extension of reforming and H₂ oxidization reactions on the overall exergetic efficiencies. Independently of the fuel choice by lowering the reforming extension a consequent decrease of overall efficiency is observed as far as the amount of H₂ available for utilization in fuel cell decreases accordingly. It is worth noticing that the results presented in Fig. 4 refer only to reforming and H₂ oxidization while the WGS and the CO oxidization reactions are of constant extensions (approx. 90% and 15% respectively), values that are actually imposed by real life scenarios.

The dashed lines in Fig. 4 represents the simulation process of C₂H₅OH fed scenario which reveals limited energy efficiency over CH₄ scenario besides the fact that the exergetic does not present a remarkable difference. As concern the level of the optimization

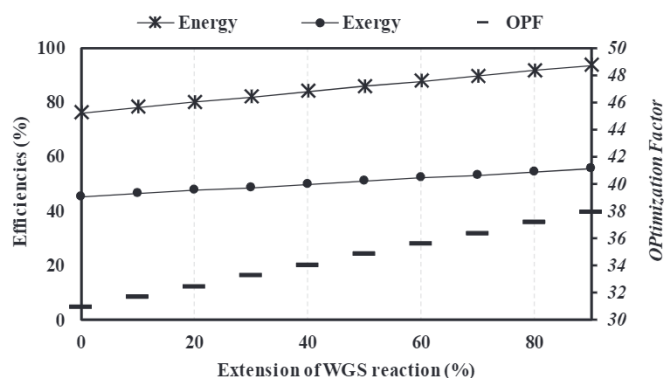


Fig. 3. The effect of WGS reaction on system's behavior.

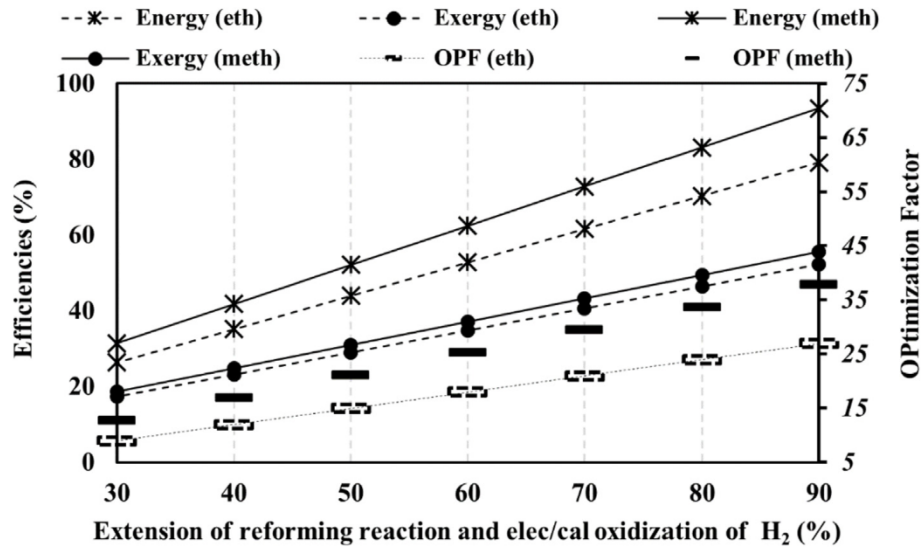


Fig. 4. The effect of reforming and electrochemical oxidization of hydrogen reactions on both system's behavior.

factor (see Fig. 4, secondary y-axis, underneath the linear efficiencies); the lower which corresponds to C_2H_5OH scenario reveals a more optimized system. Specifically, for extensions near 90%, the system for C_2H_5OH usage can efficiently manage the stored chemical energy in the initial fuel in order to produce useful work. In that case the efficiency of the useful energy (exergy) is 55.56% for CH_4 and 53.19% for C_2H_5OH while OPF is remarkably better for the second one (lower) at about 12%. Practically the above analysis shows that the use of rich hydrocarbons fuels seems to be meaningless without optimization and moreover the needs of the optimization in such systems seems to be inevitable in order to exploit the chemical energy from a rich pure fuel.

4.3. Optimal scenarios

The previous part of the present research study lead us to investigate the most optimal scenarios for both fuels as they can be designed through THERMAS modelling based on the innovative OPF which will be finally used as the unique indicator for the optimization level per case study. Also in order to be comparable these scenarios the operational temperatures of the main devices will be constant for both fuels. It has to be minded that the SOFC-stack temperature is influenced by the reformer's output stream (T_5 , see Fig. 1) and by its inlet air flow (T_{12} , see Fig. 1). So it is important to be mentioned that by lowering the SOFC-stack temperature both efficiencies are increased in absolute terms while this behavior simultaneously advances the destruction of the OPF during optimization process. Finally, it has to be underlined that afterburner's temperature plays an important role for the system's design as far as this device in particular offers the necessary thermic load for system's uninterrupted operation. Therefore, it is absolutely crucial to select the appropriate operational range where this device is not capable to drastically influence the system's efficiency due to its supportive character.

Under this prospect by lowering the overall operational temperature of such a system while increasing the extensions of all the chemical reactions in a high level and simultaneously by changing slightly the fuel-steam flow rate the feasible optimal scenarios under real life operational conditions as they were designed through THERMAS simulation tool are analytically presented through Table 2.

An extended study on this analysis reveals that the use of a rich hydrocarbon fuel under an accurate simulation tool, like THERMAS

Table 2
Optimal THERMAS modelling scenarios.

	CH_4 -Scenario	C_2H_5OH -Scenario
$\epsilon_{ref}(\%)$	90	90
$\epsilon_{wcs}(\%)$	95	—
$\epsilon_{H_2}(\%)$	95	95
$\epsilon_{co}(\%)$	15	—
T_4 (K)	1000	1000
T_5 (K)	850	850
T_6 (K)	1000	1000
T_7 (K)	850	850
T_8 (K)	880	900
T_9 (K)	730	765
T_{10} (K)	608	610
T_{11} (K)	301	306
T_{12} (K)	850	850
$Q_{env,fc}$ (%)	23.21	21.90
$Q_{env,burn}$ (%)	0.18	5.35
n_{en} (%)	82.52	77.91
n_{ex} (%)	68.53	63.96
OPF	13.99	13.95

which incorporates an extended parametric analysis, can present an identical optimization level versus fuels characterized by a lower chemical potential. Obviously the preferable between these two systems simulated under the same operational conditions, as presented through Table 2, is the one which uses CH_4 as initial fuel. Even if both systems can manage under a proper way (identical OPF ~13.99 vs 13.95) their available chemical energy this which uses methane can produce more useful energy (exergy) over C_2H_5OH -fed system.

Moreover it is worth mentioning that for both scenarios through the parametric analysis the temperature of exhaust gas mixture approaches the environmental standard condition of 298 K. These adjustments which can be obtained by using a high precision optimization tool, such as THERMAS, can produce the really optimal scenario for such a system under any combination of operational parameters. Furthermore, THERMAS calculations are characterized by higher energetic and exergetic efficiencies (up to approx. 17% and 11%, respectively) compared with most of the existing theoretical studies (see Table 1 vs Table 2). It is important to note that this improvement has been achieved through the OPF value.

5. Conclusion

This research study incorporates an extensive parametric analysis through an innovative software tool, namely THERMAS, which incorporates fundamental energy and exergy theory analysis under more realistic approaches. Different fuels were used by a SOFC-stack based system in order to be revealed the most optimal under specific operational conditions towards an optimization process in terms of OPF. The present research work reveals that during theoretical simulations on CH₄-fed systems the WGS and electrochemical oxidization of CO have to be essentially incorporated in such studies, as actual operation indicates. The present research work reveals that rich hydrocarbons fuels in a SOFC-based system seems sounds meaningless without optimization due to the gap which is presented between its available chemical energy stored in the initial fuel and the useful one (exergy) which is produced as electric work. Also after severe adjustments on the operational parameters which are offered through an innovative simulation and optimization tool a CH₄-fed system is drastically improved, in terms of OPF, in order to be characterized as more profitable than C₂H₅OH-fed one under the same operational conditions. To conclude, the importance of an optimization process can be characterized as inevitable and such an innovative model indicates a potential solution for the optimal design of a SOFC-stack based system in the direction of the commercialization of systems which use hydrocarbon fuels.

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Nomenclature

Latin symbols

C_p	Molar Isobaric Specific Heat Capacity (J mol ⁻¹ K ⁻¹)
C_p^e	Mean Isobaric Exergy Capacity (kJ kmol ⁻¹ K ⁻¹)
E	Exergy (J)
e_Q	Exergy associated with heat transfer (J)
H	Enthalpy (J)
I	Irreversibility rate (J)
I^Q	Irreversibility rate due to heat losses (J)
M	Mass (moles)
\dot{m}	Mass flow rate (kg sec ⁻¹)
N	Efficiency (%)
OPF	Optimization factor
P	Pressure (atm)
Q	Thermal energy (J)
R	Gas constant, 8.1344 (J mol ⁻¹ K ⁻¹)
S	Entropy (J)
T	Temperature (K)
W_{el}	Electric energy (J)
X	Molar fraction

Greek symbols

ΔH	Enthalpy of formation (kJ mol ⁻¹)
ΔT	Temperature difference (K)
E	Extension of a reaction (%)

Subscripts

0	property at the state of the environment
accu	accumulation

burn	Afterburner
CO	carbon monoxide
ch	Chemical
cons	consumption
en	Energy
env	Environment
ex	Exergy
gen	generation
H ₂	Hydrogen
i	Chemical elements
in	input
k	Streams throughout a device
out	Output
ph	Physical
prod	Products (chemical elements)
r	Number of chemical reactions
react	Reactants (chemical elements)
ref	Reformer
SOFC	SOFC-stack system
tot	Total amount of components
WGS	water gas shift

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