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THERMODYNAMIC AND ENVIRONMENTAL EVALUATION OF SOLID OXIDE ELECTROLYSIS CELLS FOR GREEN HYDROGEN PRODUCTION

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Abstract. *Among the technological routes for green hydrogen production, high-temperature electrolysis (HTE) is more efficient than conventional electrolysis because it operates with reduced electrical potentials, resulting in a reduction in irreversibilities. However, since HTE is still under development, studies are necessary to evaluate its thermodynamic behavior and environmental impacts in different operating modes and with the use of different renewable energy sources to meet the electrical and thermal demands of the process. Therefore, this study aims to perform a thermodynamic and environmental evaluation of the process under different operating conditions. For that, a SOEC (solid oxide electrolysis cell) system model is developed and validated using experimental data from the literature. Subsequently, a methodology is applied for exergy and environmental assessment of the SOEC system to evaluate its performance and behavior under different temperatures, pressures, and steam molar conversion ratios (SC). For operation at the thermoneutral voltage at 800°C, 1.013 bar, and SC of 0.7, the exergy efficiency obtained for the system is 90%. For the same conditions, the renewability exergy index was 8.99, assuming that all energy demand is met by renewable sources. Overall, this study demonstrates that SOEC systems can be a viable and sustainable technology to produce green hydrogen and the results can be applied in other analyses to determine the viability of the system for a wide range of applications.*

Keywords: *environmental evaluation, exergy analysis, high-temperature electrolysis, green hydrogen, SOEC.*

1. INTRODUCTION

Currently, hydrogen plays a vital role in the chemical industry, with applications ranging from ammonia and fertilizer production to petrochemical refining, food processing, power plant generator cooling, metallurgical processes, space industry fuel, and semiconductor manufacturing (Dawood, Anda, and Shafiullah, 2020). The primary uses of hydrogen are currently found in oil refining and ammonia production (Hanley, Deane, and Gallachóir, 2017). In most of these industrial applications, hydrogen is produced using fossil fuels. Coal gasification and the reforming of natural gas are the most mature processes applied for this objective. However, these conventional technologies are highly polluting in terms of greenhouse gas emissions, mainly carbon dioxide (Foit et al., 2017).

Considering technologies for clean and efficient energy conversion, electrochemical processes play an essential role. High-temperature electrolysis (HTE) using solid oxide electrolysis cells (SOEC) is a promising method to produce hydrogen. HTE is more efficient than conventional electrolysis because it operates with reduced electrical potentials, which results in reduced irreversibilities. Despite being a process under development, it is approaching maturity, and there are currently some demonstration plants in operation in Europe. These solid oxide cells have great operating flexibility. They can be used for the direct electrochemical conversion of steam (H₂O), carbon dioxide (CO₂), or both to hydrogen (H₂) or syngas (H₂ + CO), respectively. They can be thermally integrated with various chemical synthesis processes, allowing for the recycling of captured CO₂ and H₂O into synthetic methane, methanol, ammonia, and other synthetic fuels. And, when operated in reverse, the electrolyzer cell functions as a solid oxide fuel cell (SOFC) (Hauch et al., 2020).

The need for decarbonization and the growing energy demand encourage research and development of new energy conversion systems, especially routes for producing renewable hydrogen. Technical, thermodynamic, and environmental aspects must be analyzed to obtain the highest efficiencies and lowest environmental impacts in the conversion processes. Thus, in this paper, a methodology for exergy and environmental assessment is applied to the SOEC system to evaluate its performance and behavior for operation at different temperatures, pressures, and steam molar conversion ratios.

2. SOLID OXIDE ELECTROLYSIS CELLS (SOEC)

The principle of an electrolyzer is to convert water and electricity into hydrogen and oxygen. The overall reaction of water electrolysis can be represented by Eq. (1):



The minimum electrical power (\dot{W}_{min}) required for electrolysis (reversible process) is equal to the change in Gibbs free energy flow rate ($\Delta\dot{G}$), shown in Eq. (2), where \dot{H} represents the enthalpy flow rate, T temperature and \dot{S} the entropy flow rate of the reactants and products of the chemical reaction. With increasing temperature, the electrolysis process becomes progressively endothermic.

$$\dot{W}_{min} = \Delta\dot{G} = \dot{G}_{prod} - \dot{G}_{react} = \dot{H}_{prod} - \dot{H}_{react} - T(\dot{S}_{prod} - \dot{S}_{react}) \quad (2)$$

An electrolysis plant is composed of electrolyzer stacks (where water is split into hydrogen and oxygen) and the balance of plant (BoP), which includes energy supply, water supply and purification, compression, processing, and storage of gases. Fig. 2.1 illustrates a simplified schematic of an electrolysis system. High-temperature electrolysis, also called steam electrolysis is an electrochemical process for producing hydrogen from superheated steam. The technology used is the solid oxide electrolyzer cell (SOEC). Operating temperatures are usually in the range of 600°C to 850°C. The high operating temperature is an important feature of SOEC technology, giving rise to its two main advantages over alkaline and polymeric membrane electrolyzers: higher efficiencies and faster chemical kinetics (O'Brien, 2012; Laguna-Bercero 2012).

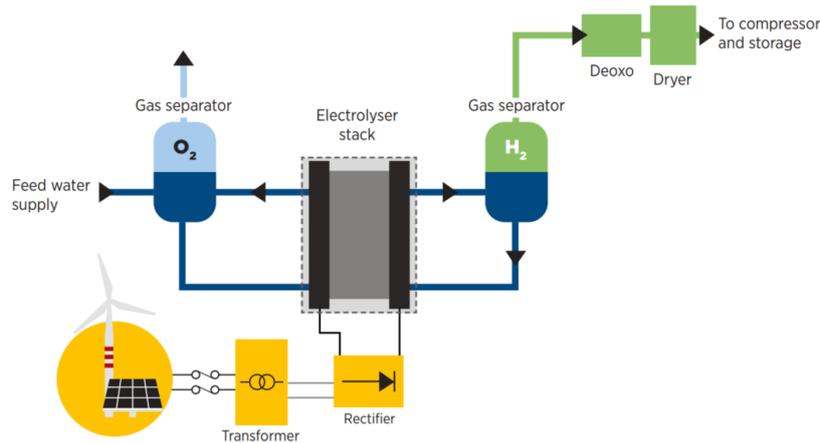


Figure 1. Basic components of an electrolysis system (IRENA, 2020)

SOECs can be used for the direct electrochemical conversion of steam (H_2O), carbon dioxide (CO_2), or both to hydrogen (H_2), carbon monoxide (CO), or syngas ($H_2 + CO$), respectively. Fig. 2.2 shows a schematic of a flat plate cell stack. Electrochemical reduction of H_2O or CO_2 takes place in the negatively charged fuel electrode, and the oxide ions are conducted through the electrolyte to the positively charged oxygen electrode, where they recombine to O_2 in the gas phase (Mogensen et al., 2019).

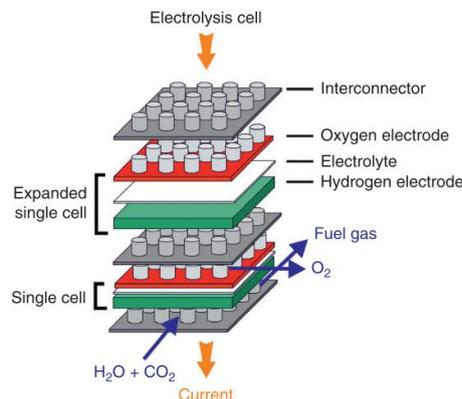


Figure 2. Schematic of a solid oxide electrolyzer cell (Mogensen et al., 2019)

3. METHOD

3.1 SOEC Model

In this study, a semi-empirical SOEC (zero dimension) system model was developed according to the works of O'Brien (2012), Petipas, Brisse, and Bouallou (2013), and Hansen (2015). The modeling and simulation of the electrolysis process were performed via Python using the Coolprop library to obtain the thermophysical properties of the substances (Bell et al., 2014). The system operation was evaluated for a pressure range from 1 to 10 bar, temperatures from 600 to 1000°C, and steam molar conversion (SC) ratios from 0.6 to 0.8. The SC ratio is defined as the molar quantity of steam that reacts to form hydrogen. The injection of hydrogen along with water to improve the reaction conditions at the electrolyzer inlet was analyzed for a molar fraction ($y_{H_2,in}$) range between 0 and 0.1. Finally, a pressure loss of 40 mbar was also considered.

For the electrolysis process to take place, the minimum voltage required is the standard state open-cell voltage or reversible voltage (V_0) given by Eq. (3), where ΔG is the Gibbs free energy change of the reaction, and F is the Faraday constant (96486 C/mol).

$$V_0 = \frac{\Delta G}{2F} \quad (3)$$

The reversible voltage (V_0) applies to the case where reactants and pure products are separated (O'Brien, 2009). To consider the range of gas compositions that occur in real electrolyzers, the Nerst open cell potential (V_N), given by Eq. (4), must be considered. \bar{R} is the universal gas constant, p represents the partial pressures of the species and T is the temperature during the reaction (Wang et al., 2017).

$$V_N = \frac{\Delta G}{2F} + \frac{\bar{R}T}{2F} \ln \left(\frac{p_{H_2} p_{O_2}^{0.5}}{p_{H_2O}} \right) \quad (4)$$

The Area Specific Resistance (ASR) represents the irreversible losses in the electrolyzer. It depends on the operating temperature and can be estimated using the empirical correlation given by Eq. (5), where the coefficient ε is used to adjust the curve and achieve the correct ASR for the electrolyzer. The temperature must be entered in Kelvin. For a stack, ε can be assumed to be $0.25 \Omega \cdot \text{cm}^2$, while for a single cell, ε is close to zero (Petipas, Brisse, and Bouallou 2013). Figure 3 illustrates the variation of ASR for the temperature range of 600 to 1000°C, considering different values of ε .

$$ASR = \exp \left(\frac{4900}{T} - 5.95 \right) + \varepsilon \quad (5)$$

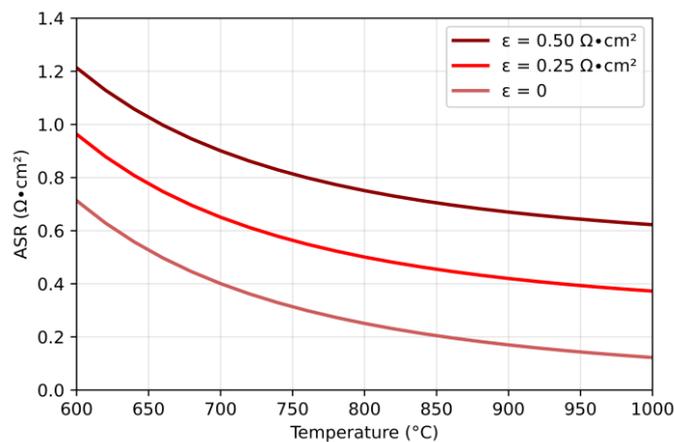


Figure 3. ASR as a function of the cell operating temperature

The electrolyzer current intensity (j) in A/cm^2 is obtained from Eq. (6), where V_{op} is the operating voltage, in V. And the total area of the electrolyzer stacks is calculated using the Faraday Law in Eq. (7), according to the required hydrogen molar flow rate (\dot{n}_{H_2}).

$$j = \frac{V_{op} - V_N}{ASR} \quad (6)$$

$$A_{SOEC} = \dot{n}_{H_2} \frac{2F}{j} \quad (7)$$

Electrolysis is an endothermic process. Therefore, the heat flux of the reaction (q_r''), in W/cm^2 , is negative, and it is given by Eq. (8). ΔS is the entropy change, and ΔH is the enthalpy of the reaction. The ohmic heat flux (q_{ohm}'') is determined using Eq (9).

$$q_r'' = \frac{j}{2F} T \Delta S = \frac{j}{2F} (\Delta G - \Delta H) \quad (8)$$

$$q_{ohm}'' = j^2 ASR = j(V_{op} - V_N) \quad (9)$$

Depending on the operating voltage, the net heat flux in the stack may be negative, zero, or positive. This is illustrated in Fig. 4. For a zero net heat flux, the operation is at a thermoneutral voltage (V_{TN}), which is a function of the enthalpy of reaction (ΔH), according to Eq. (10).

$$V_{TN} = \frac{\Delta H}{2F} \quad (10)$$

Since the enthalpy of the reaction is strictly a function of temperature (ideal gas approximation), the thermoneutral voltage is also strictly a function of temperature, independent of cell ASR and gas compositions. Operation at or near the thermoneutral voltage simplifies the thermal management of the stack by minimizing the need for significant excess gas flow for cooling or heating purposes, while also reducing thermal stresses on the components (O'Brien, 2009).

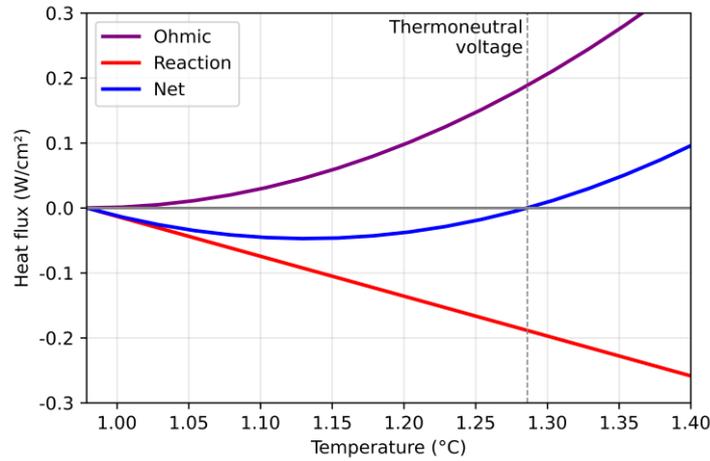


Figure 4. Heat flux in a stack for $T = 800^\circ C$ and $ASR = 0.5 \Omega \cdot cm^2$

The electrical power (\dot{W}_{SOEC}) required for the electrolysis can be calculated using Eq. (11). Finally, the efficiency of electrolysis (η_{SOEC}) can be defined according to Eq. (12). For operation at thermoneutral voltage, this equation leads to an efficiency of 100%.

$$\dot{W}_{SOEC} = V_{op} \cdot j \cdot A_{SOEC} \quad (11)$$

$$\eta_{SOEC} = \frac{\dot{n}_{H_2} \Delta H}{\dot{W}} \quad (12)$$

3.2 Exergy and Environmental Analysis

Exergy efficiency is an important parameter to evaluate the real performance of a component based on the limits imposed by the First and Second Laws of Thermodynamics. Fig. 5 illustrates the control volume for an electrolyzer. Pure steam (or with a small concentration of H_2) enters the electrolyzer. At the outlets, oxygen and hydrogen mixed with residual steam exits. To carry out the process, electricity is supplied to the system.

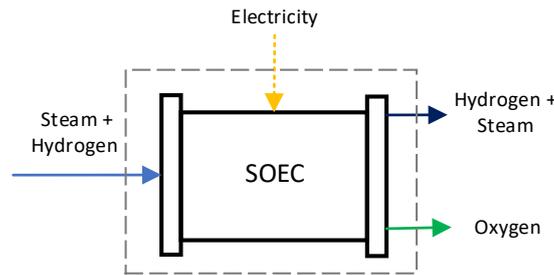


Figure 5. SOEC control volume

For operation at a thermoneutral voltage and assuming the electrolyzer is well insulated, the process is isothermal. The exergy efficiency (η_B) for the SOEC can be defined according to Eq. (13), called the degree of perfection, being the ratio between the total exergy flow rate of the products (\dot{B}_{prod}) and the total supplied exergy (\dot{B}_{sup}). Therefore, the rate of destroyed exergy (\dot{B}_{dest}) is defined according to Eq. (14):

$$\eta_B = \frac{\dot{B}_{prod}}{\dot{B}_{sup}} = \frac{\dot{B}_{H_2+H_2O,out} + \dot{B}_{O_2,out}}{\dot{B}_{H_2O+H_2,in} + \dot{W}_{SOEC}} \quad (13)$$

$$\dot{B}_{dest} = \dot{B}_{sup} - \dot{B}_{prod} = (\dot{B}_{H_2O+H_2,in} + \dot{W}_{SOEC}) - (\dot{B}_{H_2+H_2O,out} + \dot{B}_{O_2,out}) \quad (14)$$

Regarding the environmental assessment, the analysis will be based on the renewability exergy index (λ) proposed by Velásquez et al. (2008). This indicator is based on the concept of a reversible process to quantify the renewability of an energy conversion process, taking into account the relationship between the rates of the exergy rate of the products (\dot{B}_{prod}), the exergy rate from non-renewable sources consumed (\dot{B}_{fossil}), the destroyed exergy rate (\dot{B}_{dest}), the exergy rate for deactivation of treating wastes (\dot{B}_{deact}), the exergy rate related to waste disposal (\dot{B}_{disp}) and the exergy rate of emissions (\dot{B}_{emis}), according to Eq. (15).

The concept of renewability, in this case, is associated with the process and not with the product or an energy source, which provides an objective way to quantify the environmental quality of processes. The deactivation exergy is defined as the amount of exergy required to treat process wastes to equilibrium conditions with the environment. This exergy appears in the index if the effluent treatment system is included in the analyzed control volume. Otherwise, the exergies of emissions and residues must be used.

$$\lambda = \frac{\dot{B}_{prod}}{\dot{B}_{fossil} + \dot{B}_{dest} + \dot{B}_{deact} + \dot{B}_{disp} + \dot{B}_{emis}} \quad (15)$$

The analysis based on the renewability exergy index implies processes:

- Environmentally unfavorable: $0 < \lambda < 1$;
- Internally and externally reversible and use of non-renewable inputs only: $\lambda = 1$;
- Environmentally favorable: $\lambda > 1$;
- Internally and externally reversible, using only renewable inputs: $\lambda \rightarrow \infty$.

In this analysis, it was considered that for system operation there are no net CO₂ emissions ($\dot{B}_{emis} = 0$) or generation of wastes ($\dot{B}_{deact} = \dot{B}_{disp} = 0$) since only hydrogen and oxygen are produced. Regarding the amount of exergy derived from fossil fuels, three scenarios were considered: (i) electricity is generated 100% from renewable sources ($B_{fossil} = 0$); (ii) electricity is obtained from the grid, which, considering the Brazilian energy mix, consists of approximately 20% generation from fossil fuels (iii) electricity is 100% obtained from fossil fuels.

4. RESULTS AND DISCUSSION

The validation of the system model has been fulfilled against the data presented by Hauch et al. (2020) from electrolyzers fabricated in 2006 and 2020, as indicated in Fig. 6. This figure represents the variation of electrolyzer voltage for different current densities. Model accuracy increases as the voltage approaches the thermoneutral value.

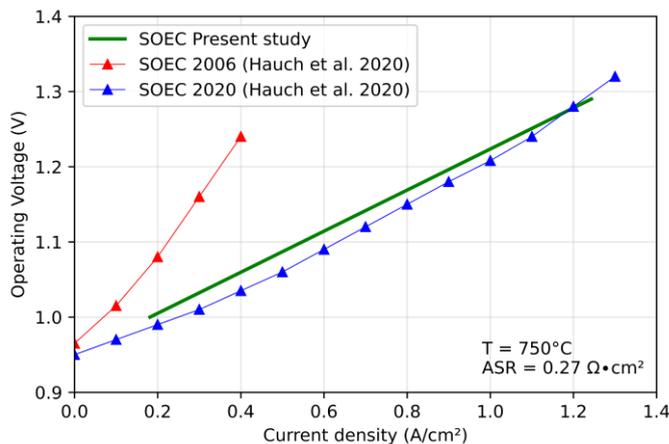


Figure 6. Data for model validation

The electrical power required for the process to produce 1.0 kg of hydrogen at thermoneutral voltage is shown in Fig. 7(a) as a function of the temperature. Despite the considered interval being 400°C, a 0.9% increase in electrical power is observed. For 800°C, $\dot{W}_{SOEC} = 123.1$ MJ/kg_{H2}. Fig. 7(b) presents the \dot{W}_{SOEC} as a function of the operating voltage at 800°C. In this case, the variation is significant. Below V_{TN} , the cell operates in an endothermic mode, requiring the addition of heat to the process, which leads to a decrease in \dot{W}_{SOEC} . However, operating under these conditions is complex and involves thermal stress on the cells.

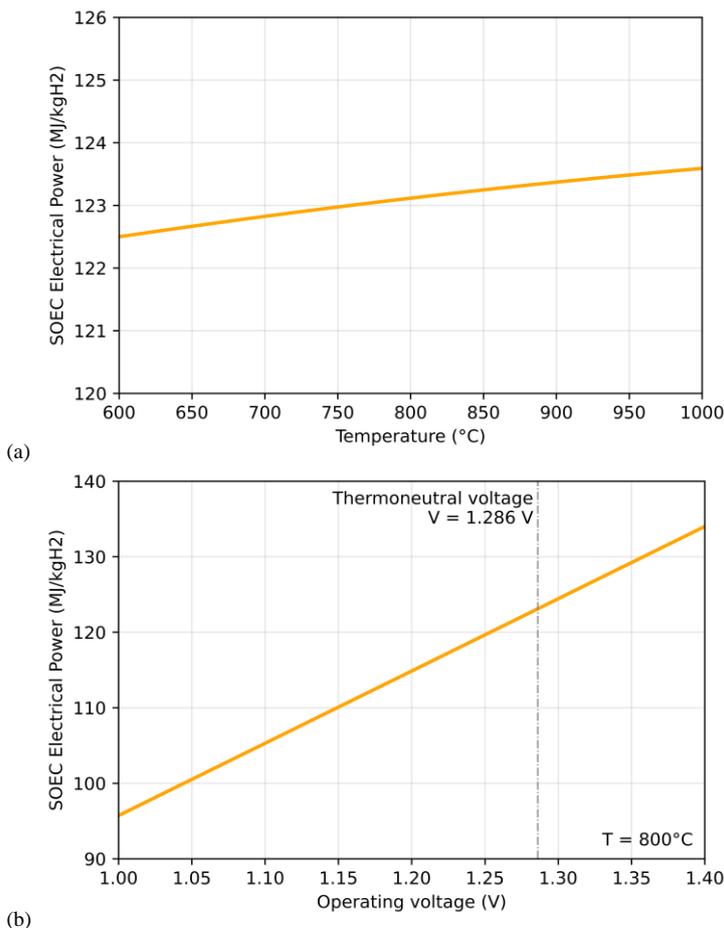


Figure 7. The electrical power required for the electrolysis (a) vs. temperature (b) vs. operating voltage.

The heat demand required for steam production is shown in Fig. 8 for different values of SC and temperature, considering a pressure of 1.013 bar. More heat is required as the operating temperature of the cell increases and as the conversion rate to hydrogen decreases. For a temperature of 800°C and an SC of 0.7, the thermal demand is 51.8 MJ/kg_{H2}, which represents 42% of the electrical demand for the process in the thermoneutral voltage.

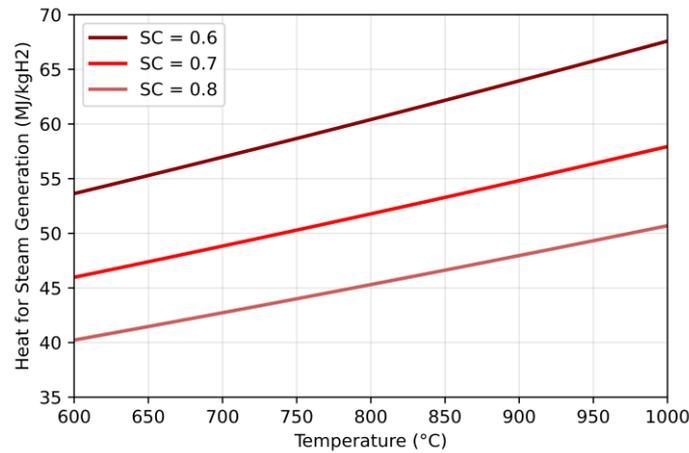


Figure 8. Thermal demand for steam generation

The exergy rates are detailed in Fig. 9 for $SC = 0.7$, $p = 1.013$ bar, $y_{H_2,in} = 0$, and operation at thermoneutral voltage. The values also refer to the production of 1 kg of hydrogen. The supplied exergy refers to the sum of the exergy associated with electricity and the exergy of the steam at the inlet of the electrolyzer. The exergy of the products comprises the sum of the exergy of hydrogen, oxygen, and steam remaining from the process. Since there is no heat transfer to the environment, the difference between the supplied exergy and the exergy of the products is equal to the exergy destroyed. It is observed a small decrease in the exergy destroyed as the temperature of increases. For 800°C , $B_{sup} = 146.6$ MJ/kg_{H₂}, $B_{prod} = 131.9$ MJ/kg_{H₂}, and $B_{dest} = 14.7$ MJ/kg_{H₂}.

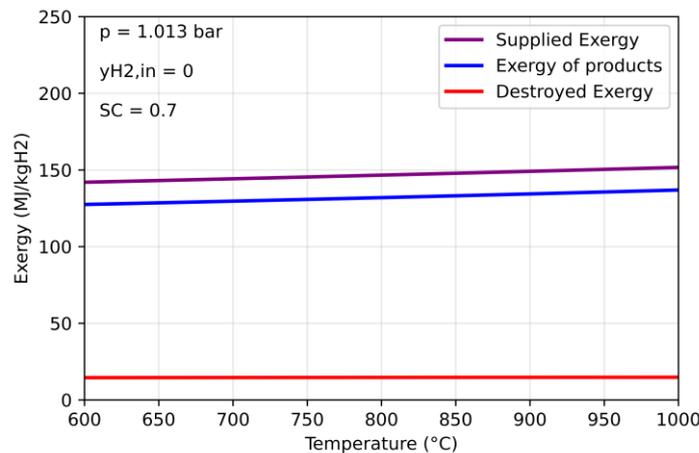


Figure 9. Supplied exergy, the exergy of products, and destroyed exergy

The exergy efficiencies values for the electrolyzer are presented in Fig. 10 as a function of temperature, pressure (Fig.10a), steam conversion rate (Fig.10b), and hydrogen inlet molar fraction (Fig.10c) considering operation at the thermoneutral voltage. It is observed that the electrolysis process presents high efficiencies. The increase in efficiency level with increasing temperature should reduce ohmic losses in the process and is an effect of operation at thermoneutral voltage. Furthermore, there is a slight increase in efficiency as the pressure and hydrogen conversion rate increase. This occurs due to the properties of hydrogen, which become more prominent in the mixture with steam as its concentration and partial pressure increase. Pressure is a parameter that can be controlled and increased considering the limitations of the electrolyzer. In turn, the steam molar conversion rate can be increased by optimizing the geometry and operating conditions of the equipment.

The injection of H₂ at the inlet also generates an increase in exergy efficiency considering the other parameters fixed. This occurs because there will be an increase in exergy rates at input and output, causing an increase in the ratio of exergy of products to that of reactants. This fraction of hydrogen is important to be included in the inlet stream to maintain reduced conditions at the cathode. However, this increase in exergy efficiency does not generate any other significant advantage. For the analyzed scenarios, the exergy efficiency ranges from 89.58% ($T = 600^\circ\text{C}$, $p = 1.013$ bar, $SC = 0.6$, and $y_{H_2,in} = 0$) to 93.11% ($T = 1000^\circ\text{C}$, $p = 10$ bar, $SC = 0.8$, and $y_{H_2,in} = 0.1$).

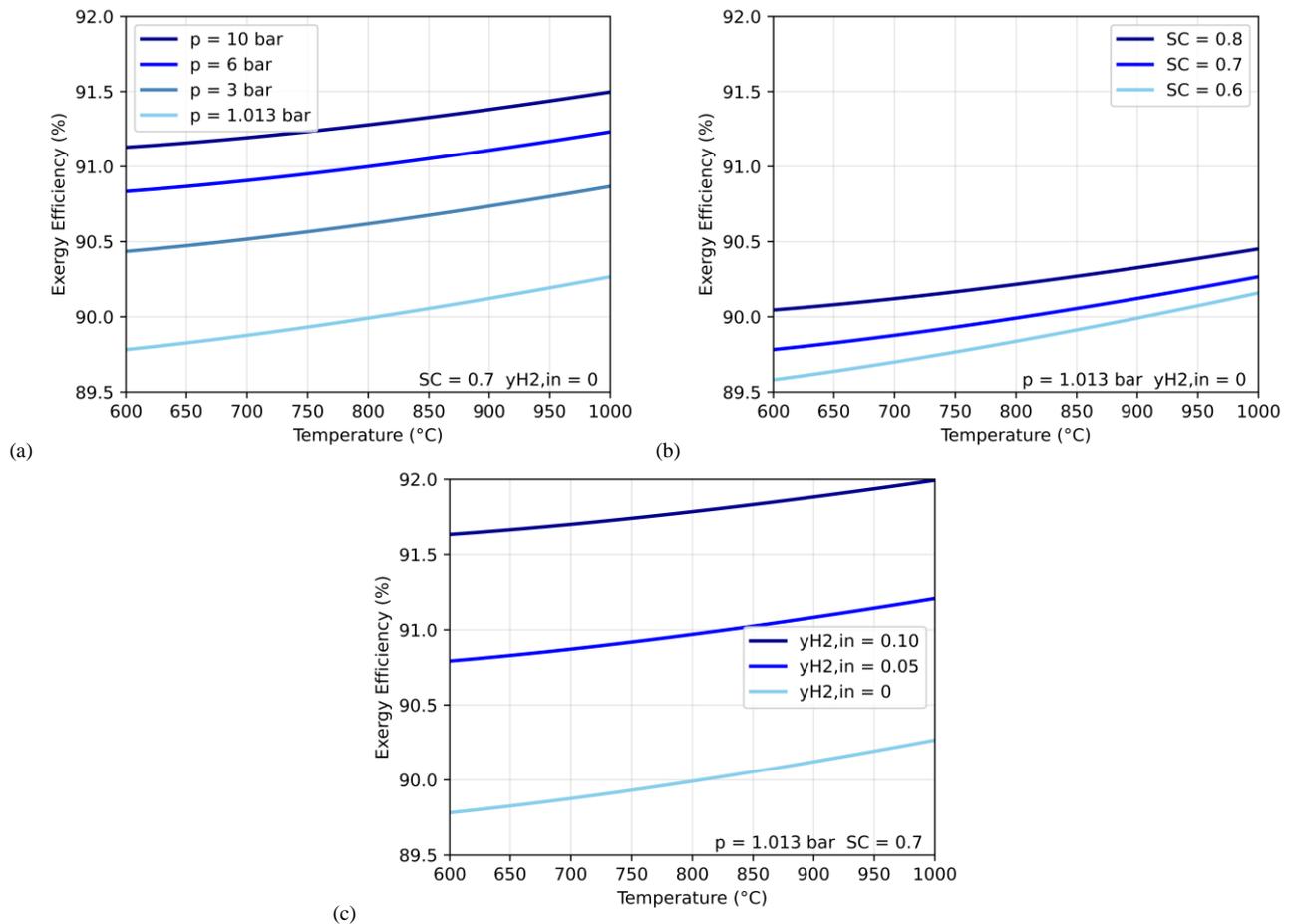


Figure 10. Exergy efficiency (a) vs. T, and p (b) vs. T, and SC (c) vs. T, and $y_{H_2,in}$

To better evaluate the influence on the exergy efficiency of the electrolyzer, Fig. 11 presents a sensitivity analysis. The average values in the ranges of the analyzed parameters were considered as a reference ($T = 800^{\circ}\text{C}$, $p = 5.5$ bar, $SC = 0.7$, and $y_{H_2,in} = 0.05$). Temperature is the main parameter that affects efficiency. However, a 10% increase in T (in Kelvins) increases exergy efficiency by only 0.11%. For a 10% higher pressure, the increase is even smaller, just 0.05%. Therefore, variations in the exergy efficiency of the high-temperature electrolysis process are very small, considering the operation at thermoneutral voltage.

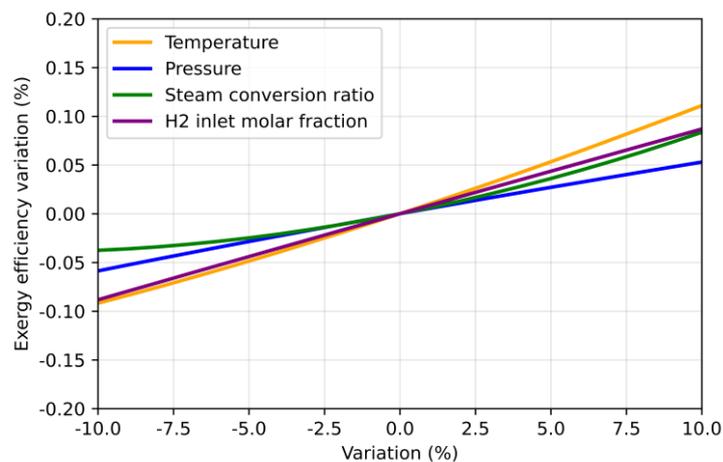


Figure 11. Sensitivity analysis for exergy efficiency

The renewability exergy index (λ) is presented in Figs. 11a, for $SC = 0.7$, $p = 1.013$ bar, $y_{H_2,in} = 0$, and operation at thermoneutral voltage. The behavior of this indicator is similar to that of exergy efficiency but with some specificities. In addition to the thermodynamic parameters that influence exergy efficiency, this index also evaluates the ratio between renewable and fossil energy used in the electrolysis. For the scenario where electricity is obtained 100% from renewable

sources and therefore, green hydrogen is produced, $\lambda = 8.99$, for $T = 800^\circ\text{C}$. When considering grid electricity, even if predominantly renewable sources, the value decreases to 3.36. Finally, if 100% non-renewable electricity is considered, $\lambda = 0.96$, that is, in this case, as expected there is an environmentally unfavorable process. In Fig. 11b, it is presented the variation of the index about the percentage of renewable electricity in the electrolysis process. The value of λ increases significantly when the percentage of renewable energy is above 60%. Considering all analyzed scenarios, the renewability exergy index ranges from 0.95 ($T = 600^\circ\text{C}$, $p = 1.013$ bar, $SC = 0.6$, $y_{\text{H}_2,\text{in}} = 0$, and 100% non-renewable electricity) to 13.52 ($T = 1000^\circ\text{C}$, $p = 10$ bar, $SC = 0.8$, $y_{\text{H}_2,\text{in}} = 0.1$, and 100% renewable electricity).

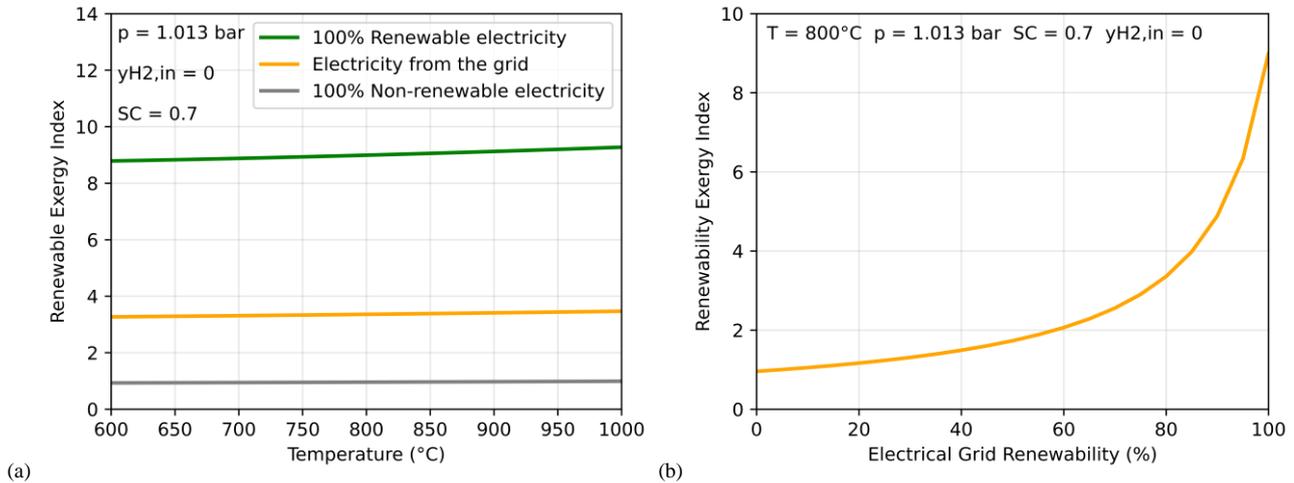


Figure 12. Renewability exergy index (a) as a function of T (b) as a function of the electrical grid renewability

Finally, a sensitivity analysis for the renewability exergy index is presented in Fig. 13. The influence of the parameters is similar to that obtained for exergy efficiency. However, for a temperature 10% higher, λ increases 1.38%. For a 10% higher pressure, the increase is 0.65%.

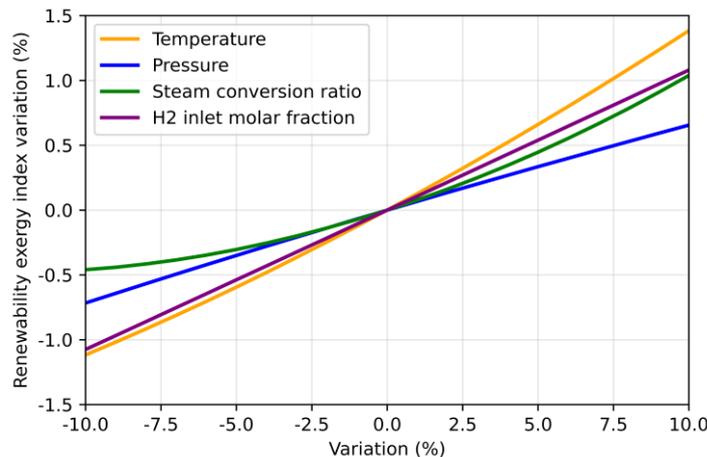


Figure 13. Sensitivity analysis for the renewability exergy index

5. CONCLUSIONS

In this study, a system model of a solid oxide electrolyzer cell (SOEC) for hydrogen production was developed and a thermodynamic and environmental evaluation was conducted considering operation at thermoneutral voltage. The obtained exergy efficiency for the considered conditions ranged from 89.58 to 93.11%, which represents only a 3.9% variation. Furthermore, the renewability exergy index ranged from 8.60 to 13.52 when considering 100% of the supplied exergy from renewable sources. These values unequivocally demonstrate that high-temperature electrolysis presents a high exergy efficiency, and it is an environmentally favorable process.

It is important to emphasize that these results exclusively pertain to the electrolysis process and do not account for other processes occurring in different components of a SOEC plant. Nonetheless, these findings serve as a crucial foundation for future analyses that encompass the overall plant balance and the utilization of thermal and electrical energy from diverse renewable sources such as solar and wind.

6. ACKNOWLEDGEMENTS

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