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## Performance Assessment of Electric Energy Storage (EES) systems based on reversible solid oxide cell

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### Abstract

This paper focuses on the performance assessment of a novel and efficient EES (electric energy storage) system based on ReSOC (reversible solid oxide cell) technology. The ReSOC is an electrochemical energy conversion device working at high temperature (600-1000°C) that can operate reversibly either as a fuel cell (SOFC) or as an electrolyzer (SOEC). In this study, a ReSOC unit fed by mixtures of CH<sub>4</sub>, CO, H<sub>2</sub>O and H<sub>2</sub> is proposed and analyzed. In particular, in the SOFC mode, where electricity is generated, the reactant gas, mainly formed by CH<sub>4</sub> and H<sub>2</sub>, is converted into a mixture of H<sub>2</sub>O and CO<sub>2</sub>. The exhausts from the SOFC are used as the reactant gas for the SOEC operation. During the electrolysis process, CH<sub>4</sub> can be also produced thanks to the methanation reaction that, under proper operating conditions, occurs at the cathode of the solid oxide cell.

The ReSOC unit behavior is investigated by developing a thermo-electrochemical model, able to predict its performance (i.e. roundtrip efficiency, polarization curve, thermally self-sustaining conditions) under different operating conditions. The ReSOC model, built with a modular architecture, is performed through thermodynamic, thermochemical and electrochemical sub-models taking into account mass and energy balances, chemical reactions (reforming, shifting reactions and methanation) and electrochemical relationships. Available literature data have been used for the model validation and a calibration procedure has been performed in order to evaluate the best fitting values for the model parameters. Furthermore, in order to estimate the thermoneutral conditions in SOEC operating mode, the ReSOC thermal behavior has been analyzed under different operating temperatures.

Results pointed out that, by feeding the cell with a syngas mixture, the reforming reaction (in the SOFC mode) and the methanation reaction (in the SOEC mode) allow to simplify the cell thermal management. Moreover, the best performance in terms of stack roundtrip efficiency (about 70%), can be reached operating the ReSOC at low temperature (700°C).

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**Keywords:** SOEC, SOFC, Electric Energy Storage, roundtrip efficiency, thermal neutrality

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## 1. Introduction

The most viable option for sustainable economic development and reduction of polluting emissions in electric power generation is the realization of very high energy-penetration of intermittent renewable. As a matter of fact, renewable energy technologies have the potential to undercut the cost of electricity generation to control the global warming. A crucial issue, associated with renewable energy sources (i.e. solar and wind), is the intermittence and their stochastic feature, so make more difficult to manage them. Moreover, the fluctuating nature of renewable energies causes an extensive strain on the grid infrastructure.

Electrical Energy Storage (EES) has been recognized as a solution to increase the penetration of renewable energy technologies because providing time varying energy management, meeting peak energy demands, meeting remote loads, and improving system reliability [1].

Some recent studies demonstrate that an EES system based on reversible solid oxide cell (ReSOC) technology (HES, hydrogen energy storage) offers a potentially high efficiency, low cost, and scalable distributed energy resource [2-6]. Depending on the cell polarity, the ReSOC can operate either as a fuel cell (SOFC mode) to electrochemically oxidize fuel species (i.e.,  $H_2$ , CO) and generate electricity, or as an electrolysis cell (SOEC mode) to electrochemically reduce reactant species (i.e.  $H_2O$  and/or  $CO_2$ ) while consuming electrical energy.

There is a great deal of literature based on individual mode operation (i.e., either fuel cell or electrolysis operation only) addressing experimental and mathematical modeling; however, motivated by more recent interest in using ReSOCs for energy storage, some studies describe fuel cell energy storage processes, meaning the coupling of fuel cells and electrolysis cells with hydrogen and oxygen storage.

Experimental results on cyclic operation of a ReSOC present results from 10 charge/discharge cycles of 10 minutes each which show stable operation in both modes (limiting the operating current density to  $50 \text{ mA/cm}^2$ ) and indicate a high roundtrip efficiency of 91.5% [7]. Bi et al. [8] discussed the application of proton-conducting oxides in both solid oxide fuel cells (SOFCs) and solid oxide electrolysis cells (SOECs) focusing on discussing the importance of adopting chemically stable materials in both fuel cell and electrolysis modes.

Theoretical models for the simulation of electrode supported Ni/YSZ–YSZ–LSM/YSZ cell in direct and inverse mode have been developed, calibrated and validated on experimental data by Moyer et al. [9], Garcia-Camprubi et al. [10], Ni et al. [11–14]. A general ReSOC model is developed by Kazemipoor et al. [2, 4], Ferrero et al. [15]. The validation results show that the fitting parameters, extracted from the calibration study, can precisely simulate current-voltage curves of a Ni/YSZ–YSZ–LSM/YSZ planar cell tested with  $H_2/H_2O/CO/CO_2$  mixtures.

In this study, a ReSOC unit fed by different mixtures of  $CH_4$ , CO,  $H_2O$  and  $H_2$  either in the SOFC and SOEC operation modes, is proposed and analyzed. In particular, in the SOFC mode, where electricity is generated, the reactant gas (mainly formed by  $CH_4$  and  $H_2$ ) is converted into a mixture of  $H_2O$  and  $CO_2$  that is the reactant gas feeding of the ReSOC in the SOEC operation. During the electrolysis process,  $CH_4$  can be also produced thanks to both the reverse water gas shift (RWGS) reaction and the methanation reaction that, under proper operating conditions, occur in the cathode of the solid oxide cell.

The EES is investigated by developing, in Aspen Plus [16] environment, a thermo-electrochemical model that can readily switch between the operational modes, able to predict the performance (i.e. roundtrip efficiency, polarization curve, thermal balance, thermally self-sustaining conditions) of the ReSOC unit under different operating conditions and cell design. The ReSOC model, built with a modular architecture, is performed through thermodynamic, thermochemical and electrochemical sub-models taking into account mass and energy balances, reforming and shifting reactions, electro-chemical relationships.

The model validation has been carried out by means of available literature data and a calibration procedure has been applied to evaluate the best fitting values for the calibration parameters.

Furthermore, in order to investigate the thermal behavior of the ReSOC unit and to define the better operating conditions in terms of roundtrip efficiency and reactant compositions, a sensitivity analysis on the cell operating temperature has been performed and the overall heat fluxes have been estimated.

## 2. Model description

The numerical model of the ReSOC has been developed by following the approach detailed in ref. [17] in which the model of a solid oxide fuel cell is presented.

The basic model assumptions are: i) steady-state, ii) isothermal conditions, iii) ideal gas mixtures, iv) negligible pressure drop. The unit cell is discretized in N-elements along the flow direction and each J-element consists of a fuel electrode, an oxidant electrode and an electrolyte.

Figures 1a and 1b show the main flowsheet of the ReSOC unit (the energy storage unit) and the flowsheet of the J-element of the SOFC and the SOEC sub-models that have been implemented in the main flowsheet as hierarchy block that allows a sub-flowsheet of greater detail to be encapsulated in a single high-level block. Thus, as shown in Fig. 1a, the ReSOC unit consists of two hierarchy blocks (SOFC and SOEC), heat exchangers and gas compressors.

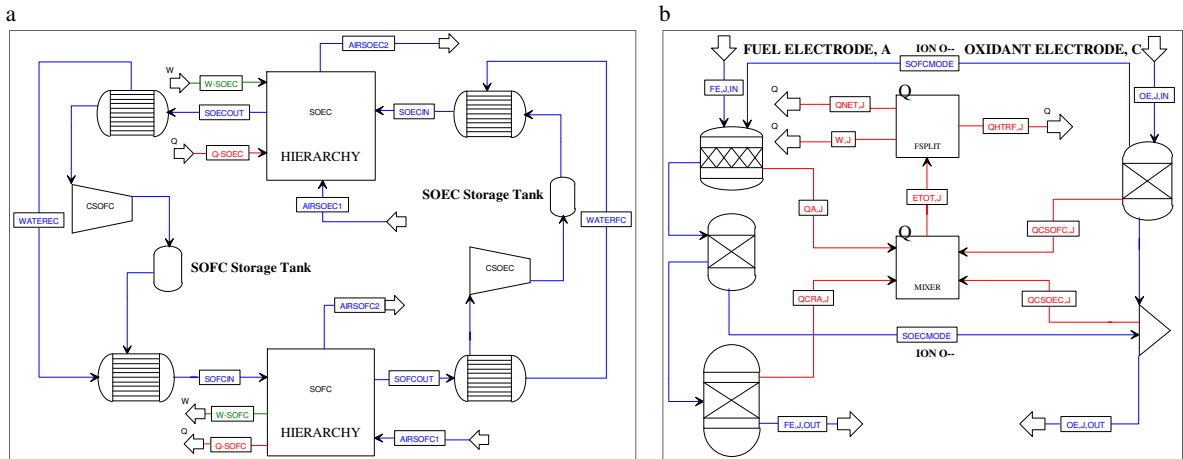


Fig. 1. (a) ReSOC unit flowsheet, (b)SOFC/SOEC Hierarchy flowsheet (J-element)

The J-element model comprises an oxidant electrode (labelled C) and a fuel electrode A (labelled A). The oxidant electrode is modeled as a separator block (in the SOFC mode) in which the oxygen is separated from nitrogen and sent to the fuel electrode or a mixer block (in the SOEC mode) in which the oxygen from the fuel electrode is mixed with the air that feeds the oxidant electrode. The fuel electrode consists of: i) a stoichiometric reactor in which the electro-oxidation reaction (in the SOFC mode) or the electro-reduction reaction (in the SOEC mode) occur; ii) a separator block (in the SOEC mode) to separate the oxygen generated from the electrolysis; iii) a Gibbs reactor in which chemical reactions (i.e. reforming reaction and water gas shift reaction in the SOFC mode, methanation reaction and reverse water gas shift in the SOEC mode), can take place. This last block allows to calculate the actual reactant composition that varies along the fuel electrode side due to electrochemical and chemical reactions. According with several scientific papers, the electrochemical oxidation of CO at the fuel electrode as well as the electrochemical reduction of CO<sub>2</sub> are neglected because of the dominating of H<sub>2</sub> over CO and H<sub>2</sub>O over CO<sub>2</sub>. Laboratory data show that the kinetics of the CO<sub>2</sub> electrochemical reduction reaction at the SOEC cathode are generally slow compared with the steam reduction, and CO<sub>2</sub> consumption mostly depends on the RWGS reaction when steam is present [4,17].

The energy balance is solved by using two specific block calculators:

- A Thermal Mixer block (QMIXER), used to perform the energy balance of the J-element. The term  $E_{TOT,J}$  takes into account all energy fluxes:

$$E_{TOT,J} = \Delta H_{A,J} + \Delta H_{C,J} = Q_{A,J} + Q_{C,J} + Q_{CRA,J} \quad (1)$$

where  $\Delta H_{A,J}$  e  $\Delta H_{C,J}$  are the enthalpies change of fuel electrode and oxidant electrode streams which are the sum of  $Q_{A,J}$  ( $\Delta H$  of reaction and sensible heat change) and  $Q_{CRA,J}$  (the net thermal energy due to the steam reforming e water gas shift reactions) in the fuel electrode side and the  $Q_{C,J}$  flux in the oxidant electrode side.

• A Thermal Splitter block (QFSPLIT), used to separate the energy output streams of the J-element, in terms of  $W_J$ ,  $Q_{NET,J}$  and  $Q_{HTRF,J}$  (the convective and radiative fluxes to the surrounding). In order to estimate each of these energy fluxes, a Fortran block calculator is implemented in the model flowsheet.  $E_{TOT,J}$  can be written as:

$$E_{TOT,J} = W_J + Q_{NET,J} + Q_{HTRF,J} \quad (2)$$

The term  $W_J$ , the actual electric power, is:

$$W_J = V_J \cdot I_J = W_{rev,J} \mp Q_{LOSS,J} \quad (3)$$

where  $V_J$  is the voltage and  $I_J$  is the current intensity, calculated by applying the Fick's law. The voltage of the J-element at different current values is calculated by subtracting (in SOFC operation mode) or adding (in SOEC operation mode) the potential losses to the  $OCV$  (open circuit voltage):

$$V_J = OCV \mp (\eta_{act,A} + \eta_{act,C} + \eta_{ohmic} + \eta_{conc,A} + \eta_{conc,C})_J = OCV \mp (\sum_i \eta_{i,k})_J \quad k=A,C \quad (4)$$

where  $OCV$  is the open circuit voltage (calculated at the cell inlet for the first J-element),  $\eta_{act,A}$  and  $\eta_{act,C}$  are the electrodes activation overpotentials,  $\eta_{ohmic}$  is the ohmic overpotential, and  $\eta_{conc,A}$  and  $\eta_{conc,C}$  denote the electrodes concentration overpotentials due to the mass transfer limitations.

Moreover,  $W_{rev,J}$  is the reversible electric power that is defined as:

$$W_{rev,J} = OCV \cdot I_J \quad (5)$$

The term  $Q_{LOSS,J}$  is the heat production associated with the overvoltage losses:

$$Q_{LOSS,J} = |OCV - V_J| \cdot I_J \quad (6)$$

The net thermal power, introduced in eq.2, results:

$$Q_{NET,J} = Q_{LOSS,J} + Q_{REACT,J} + Q_{CRA,J} - Q_{HTRF,J} \quad (7)$$

where  $Q_{REACT,J}$ , that is the heat flux due to the electrochemical reaction, is calculated as:

$$Q_{REACT,J} = E_{TOT,J} - W_{J,rev} \quad (8)$$

The term  $Q_{HTRF,J}$  is evaluated as follows:

$$Q_{HTRF,J} = h \cdot A_{cell,J} \cdot (T_{cell} - T_{room}) + \sigma \cdot \epsilon \cdot A_{cell,J} \cdot (T_{cell})^4 \quad (9)$$

where  $A_{cell,J}$  is the surface no insulated,  $T_{cell}$  is the cell temperature (K),  $T_{room}$  (K) is the ambient temperature,  $h$  is the convective heat transfer coefficient,  $\sigma$  the Stefan-Boltzmann constant and  $\epsilon$  the emissivity of material [17].

Finally, all these equations are used to evaluate the overall cell performance in terms of cell voltage ( $V$ ), current intensity and density ( $I, j$ ), electric power ( $W$ ) and net thermal power ( $Q_{NET}$ ).

$$V = OCV + \frac{1}{N} \sum_{J=1}^N \left( \sum_i \eta_{i,k} \right)_J \quad (10)$$

$$I = \sum_{J=1}^N I_J \quad (11)$$

$$j = I/A_{cell} \quad (12)$$

$$W = \sum_{J=1}^N W_J \quad (13)$$

$$Q_{NET} = \sum_{J=1}^N Q_{NET,J} \quad (14)$$

### 2.1 Validation procedure

The model validation requires the evaluation of the fitting parameter according with the experimental data and the analysis of the fuel electrode streams compositions coming from the ReSOC unit both in the SOFC and SOEC operating mode. Therefore, the validation procedure has been carried out in two steps: in the first, the numerical polarization curves have been compared with the data reported in [3]; in the second one, the comparison between the input/output streams compositions, in the SOFC and SOEC operating mode, have been performed taking into account the results reported in [3].

The solid oxide cell used in this study is a planar Ni/YSZ-supported type (10-15 mm thick Ni/YSZ cermet electrode supported by a 300 mm thick porous Ni/YSZ layer, a 10-15 mm thick YSZ electrolyte, and 15-20 mm thick LSM/YSZ) described in [2,3]. Figure 2 shows the comparison between the polarization curves obtained from the simulation results and the data reported in [3], whereas in table 1 the fitting parameters, used for the model calibration, are summarized.

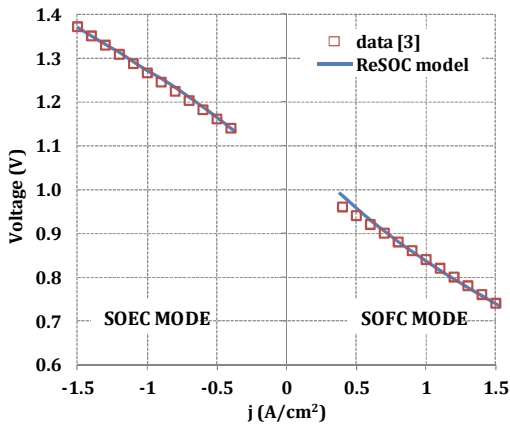


Table 1. Input fitting parameters for ReSOC model

		SOEC MODE	SOFC MODE
$T_{cell}/p_{cell}$	°C/bar	650/20	650/20
$\sigma_{el}$	S/m	3.47[18]	3.47[18]
$\sigma_{\lambda}$	S/m	11.24 [19]	11.24[19]
$\sigma_c$	S/m	$7.67 \cdot 10^3$ [19]	$7.67 \cdot 10^3$ [19]
$\sigma_{int}$	S/m	$8.5 \cdot 10^4$ [2]	$8.5 \cdot 10^4$ [2]
$\alpha_A/\alpha_C$	-	0.2/0.46	0.9/0.24
$E_{act,A}/E_{act,C}$	kJ/mol	120 [15]	120 [15]
$\gamma_A/\gamma_C$	A/m <sup>2</sup>	$3.6 \cdot 10^9/3.9 \cdot 10^9$	$8.0 \cdot 10^8/2.3 \cdot 10^9$
$\xi/\tau$	-	0.3/3 [2]	0.3/3 [2]
$a,b,c$	-	1,1,0.25[17]	1,1,0.25 [17]

Fig. 2. Model calibration results for a ReSOC operating with different reactant compositions and constant fuel utilization equal to 0.7. SOFC reactant composition (vol%): 35.3 H<sub>2</sub>, 1.0 CO, 51.1 CH<sub>4</sub>, 12.0 H<sub>2</sub>O, 0.7 CO<sub>2</sub>; SOEC reactant composition (vol%): 17.4 H<sub>2</sub>, 3.2 CO, 4.6 CH<sub>4</sub>, 54.2 H<sub>2</sub>O, 20.6 CO<sub>2</sub>. The oxidant electrode is fed by 99.99 vol% pure oxygen.

It is worth noting that a very good fitting has been obtained in either SOEC and SOFC operation modes.

The reactant utilization factor at the fuel electrode is expressed in terms of the consumption of electrochemical species:

$$U_{FE,SOFC} = \frac{n_{H2,consumed}}{n_{H2,in} + n_{CO,in} + 4 \cdot n_{CH4,in}} \tag{15}$$

$$U_{FE,SOEC} = \frac{n_{O2,generated}}{n_{H2O,in} + n_{CO,in} + 2n_{CO2,in}} \tag{16}$$

For the oxidant electrode, in the SOFC mode, the reactant utilization factor is calculated as:

$$U_{OE,SOFC} = \frac{n_{O2,consumed}}{n_{O2,in}} \tag{17}$$

In the SOEC mode, a generation factor can be defined as follows :

$$G_{OE,SOEC} = \frac{n_{O2,generated}}{n_{O2,in}} \tag{18}$$

The comparison of the fuel electrode reactant/product compositions in each mode and the reference data are illustrated in table 2. It can be noted that the stream exiting the fuel electrode in SOFC operation is the stream entering the fuel electrode in the SOEC operation and viceversa (as in fig.1).The fuel electrode utilization factors,

$U_{FE,SOF C}$  and  $U_{FE,SOEC}$ , are assumed equal to 0.9 and 0.45, respectively, according with [3]. As a consequence, the number of J-element in the SOFC sub-model is equal to 5, while in the SOEC sub-model it is 3.

The model results show a good accordance with reference data.

Table 2. Streams composition at 650°C: comparison between numerical results of the ReSOC model and reference data

Stream Composition (vol%)	SOECIN/SOFCOUT		SOF CIN/SOECOUT	
	ReSOC Model	Ref [3]	ReSOC Model	Ref [3]
H <sub>2</sub>	9.1	8.2	30.2	33.1
H <sub>2</sub> O	63.8	65.4	14.1	13.3
CO	1.8	1.2	1.7	0.4
CO <sub>2</sub>	25.3	24.6	1.6	0.4
CH <sub>4</sub>	-	0.7	52.4	52.8

### 3. ReSOC operating analysis

In the designing of an EES based on SOC technology, the most significant challenge is the thermal management of the ReSOC stack. In the case of mixture reactants feeding, the thermal management can be simplified because both the endothermic reforming reaction (SOFC mode) and the exothermic methanation reaction (SOEC mode) can favor the thermal balance of this EES system. Thus, in order to investigate the thermal behavior of the ReSOC unit, a sensitivity analysis on the cell operating temperature has been performed.

Table 3 summarizes the fuel electrode reactants compositions by varying the cell temperature in the range 700–850°C. The fuel electrode utilization factors,  $U_{FE,SOF C}$  and  $U_{FE,SOEC}$ , are assumed equal to 0.9 and 0.45, respectively, whereas the  $U_{OE,SOF C}$  and  $G_{OE,SOEC}$  are 0.7 and 0.085. In all cases the oxidant gas feeding is air.

Table3. Streams compositions at different cell operating temperature

Cell Temperature (°C)	700		750		800		850	
Composition (vol%)	SOECIN	SOF CIN	SOECIN	SOF CIN	SOECIN	SOF CIN	SOECIN	SOF CIN
H <sub>2</sub>	0.9	34.9	0.3	40.8	0.25	48.9	0.3	54.4
H <sub>2</sub> O	71.8	13.7	72.4	12.2	73.7	10.1	73.6	8.3
CO	0.2	4.3	0.1	7.6	0.08	10.0	0.1	13.9
CO <sub>2</sub>	27.1	2.7	27.2	3.0	25.9	2.24	26.0	1.9
CH <sub>4</sub>	-	44.4	-	36.4	-	28.7	-	21.4

As previously discussed, global reaction chemistry within ReSOC includes electrochemical fuel oxidation (or reduction), fuel reforming (or methanation), and water-gas shift (or reverse shift) processes.

With referring to the fuel electrode gas feeding of the SOFC (the SOFCIN stream in fig. 1a), the hydrogen content increases with the cell operating temperature because of the lower extent of the methanation reaction that is temperature depending (during the SOEC operation mode). Alternatively, in the fuel electrode gas feeding of the SOEC (the SOECIN stream in fig.1a), the water concentration increases due to the improvement of the electrochemical fuel oxidation, as well as the carbon dioxide content due to the water gas shift (WGS) reaction that is promoted by the H<sub>2</sub> depletion.

In figure 3a the calculated polarization curves are plotted. The increasing in the cell operating temperature allows to improve the ReSOC performance in both the operation mode thanks to the reduction of the overvoltage losses and the better characteristics in terms of compositions of reactant streams. However, for the stack and system thermal management, it could be desirable to operate with high concentrations of carbonaceous species (i.e. low hydrogen-to carbon ratio). Figure 3b shows the net thermal flux vs. the current density either in SOFC and SOEC operation. As expected, the thermal self-sustaining conditions are difficult to be achieved in the typical cell current density operating range, for temperatures higher than 750°C. As a matter of fact, the thermoneutrality conditions are reached at 1.6 A/cm<sup>2</sup> and 3.8 A/cm<sup>2</sup> at 700°C and 750°C, respectively.

As the cell temperature increases, the net thermal requirement is rising because less heat is generated from the methanation reaction and more heat is needed for the RWGS, so higher operating current densities are required in order to have greater resistivity losses allowing to reach the thermoneutrality.

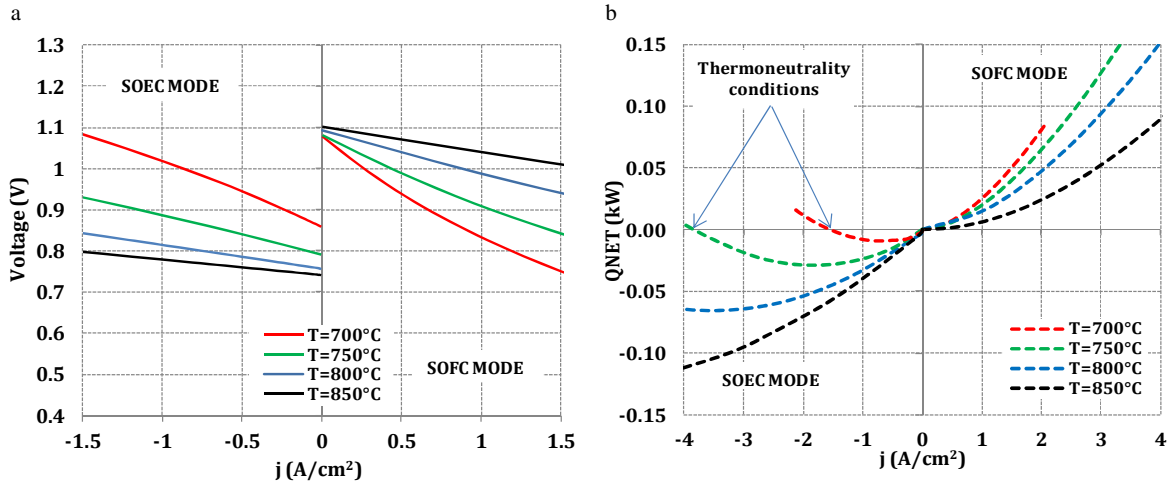


Fig. 3. (a) ReSOC polarization curves; (b) Thermal balance of the ReSOC under different operating temperature

#### 4. Performance assessment

The efficiency of a ReSOC as EES system depends on both the efficiency of the stack and the auxiliary power required by the balance of plant components (BOP) in the SOFC and SOEC operating modes [3]. The roundtrip system efficiency,  $\eta_{RT}$ , is defined as the ratio between the net energy generated in SOFC and the total energy supplied in SOEC mode. Thus, according to this definition, the BOP energies, that include the electric energy for compressors (Fig. 1 a) or energy requirements for feeding flows, are taken into account. This means that it is needed to know as the EES system works during the year in terms of produced or required energies.

In order to simplify this evaluation, but allowing anyway to estimate the ReSOC performance, a simplified equation, based on the SOFC power production and the SOEC power consumption can be applied. Therefore, it is possible to define the roundtrip stack efficiency,  $\eta_{RT,stack}$ , by neglecting the BOP energy required in both modes but considering the operation in SOFC and SOEC mode at the same current that corresponds to the current calculated at the SOEC thermoneutrality condition ( $I_{th,SOEC}$ ).

$$\eta_{RT,stack} = \frac{P_{SOFC}}{P_{SOEC}} = \frac{V_{SOFC} \cdot I_{th,SOEC}}{V_{SOEC} \cdot I_{th,SOEC}} \tag{19}$$

This efficiency has been calculated at different operating temperature, as shown in table 4.

Table 4. Round Trip stack efficiency

Cell Temperature (°C)	700	750	800	850
SOEC Voltage (V)	1.091	1.107	1.130	1.162
SOFC Voltage (V)	0.739	0.612	0.565	0.548
$\eta_{RT,stack}$ (%)	67.7	55.3	50.1	47.2

As it can be noted the temperature increasing implies lower round trip efficiencies. Thus, in the case of syngas feeding it is better to work at lower temperature.

## 5. Conclusion

In this study, a ReSOC unit, for the development of an EES system, fed by mixtures of CH<sub>4</sub>, CO, H<sub>2</sub>O and H<sub>2</sub> either in the SOFC and SOEC operation modes, is proposed and analyzed.

The system is investigated by a thermo-electrochemical model, developed by using the Aspen Plus code, that is able to predict the performance of the ReSOC unit under different operating conditions. The model, built with a modular architecture, is performed through thermodynamic, thermochemical and electrochemical sub-models taking into account mass and energy balances, reforming and shifting reactions, electro-chemical relationships. Available literature data have been used for the model calibration and validation.

The performance analysis, in terms of polarization curve, cell net thermal flux and stack roundtrip efficiency, has been carried out by varying the operating temperature in the range 700-850°C.

Results pointed out that by feeding the cell with a syngas mixture (CH<sub>4</sub>, CO, CO<sub>2</sub>, H<sub>2</sub>O, H<sub>2</sub>), the reforming reaction (in the SOFC mode) and the methanation reaction (in the SOEC mode) allow to simplify the cell thermal management. Moreover, the best performance in term of stack roundtrip efficiency (about 70%) can be reached operating at low cell temperature (700°C).

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