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Numerical prediction of off-design performance for a Power-to-Gas system coupled with renewables

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9 Abstract

10 In this study, the off-design performance of a Power-to-Gas process are predicted by means of a developed calculation model, 11 implemented in commercial tool environments. With the aim to evaluate the behaviour of the several components in off-design 12 conditions, specific calculation models have been integrated in the whole system model. Then, starting from a real wind 13 production profile, four configurations of the Power-to-Gas system have been analysed, evaluating the annual operating time 14 of the integrated Power-to-Gas/wind systems. In addition, in order to assess the cost effectiveness of the technology, a 15 preliminary economic analysis has been performed. The results highlight that the most performant configuration is the one at 16 ambient pressure, with the co-electrolyzer and the high temperature methanation section operating at the same temperature, 17 showing a methane production of 184 ton/year and an overall efficiency of about 75 %. At the current technology readiness 18 level, the economic competitiveness of the process is strongly affected by the synthetic natural gas sell price.

19

20 Keywords: Power-to-Gas; co-electrolysis; methanation; storage system; off-design performance; renewables.

21

22 **1. Introduction**

One of the main targets in many Countries is the decarbonization of the energy sector, as also defined in the European Roadmap 2050 [1]. In order to achieve this goal, a possible solution is to increase the penetration of Renewable Energy Sources (RES) into the electrical system. However, RES such as wind and solar supply electrical power in an intermittent way. This volatility results in issues related to the management of local and regional electric networks, due to the several hours and even days of electricity surplus and deficit. Indeed, as reported by Karimi et al. [2], despite its promising success, photovoltaic

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(PV) penetration presents various issues, such as voltage fluctuation, voltage rise and voltage balance, and its impact on the distribution system has to address for seamless integration in the power system. Moreover, Eltawil et al. [3] have evaluated the operation of grid-connected PV generators, demonstrating that control problems can be registered as a consequence of the variable power generation. Similar problems arise in case of wind generators, which are affected by a short-term nonprogrammable and intermittent power production and also by a long-term variability [4].

33 As the share of these sources in power generation increases, long term and even seasonal storage capacities are required to 34 ensure a reliable energy supply. In this context, the needed flexibility to balance the stochastic energy production and then to 35 make easier the RES integration in the electric network can be offered by the emerging technology of Power-to-Gas (P2G) [5], 36 i.e. the process of converting the surplus of renewable energy into a gas fuel. As reported by Yang et al. [6], the P2G energy 37 conversion method can provide an effective solution to the energy dilemma; indeed, this technology can be seen as a flexible 38 energy-use mechanism to absorb redundant renewable energy and to mitigate the natural gas supply shortage for power plants 39 in the meantime. Sayedin et al. [7] claim that although most of the renewable energy technologies are well known, the 40 integration of these technologies has still some issues; the mismatch between the output specification of photovoltaic modules 41 and electrolyzers load at different irradiance conditions indicates the importance of the optimal matching of the photovoltaic 42 and electrolyzers. For this reason, the Author's investigation is based on the evaluation of the behaviour of an innovative P2G 43 process at various loading conditions, considering the variability of the electric energy supply. Indeed, the P2G process can be 44 considered a viable flexible technology [8]; as presented by Kupecki et al. [9], the solution of the grid imbalances can be 45 represented by an electrolyzer, which mitigates the problem by coupling the electric and gas grids. Petipas et al. [10] show that 46 with the addition of control strategies, electrolyzers should operate across a wider load range, even below 60 % of the design 47 load. As a consequence, the loading power of a P2G system can follow the fluctuating demand and then balance the 48 intermittent RES production [11]. According to Frank et al. [12], reversible solid oxide cells represent a promising approach in 49 order to guarantee a constant power supply. With this purpose, the renewable energy that might be wasted (peak production) 50 can be exploited to produce useful fuel gases. Following this way, several P2G pilot plants have been realized worldwide, as 51 shown by Gahleitner [13].

The Solid Oxide Electrolyte Cell (SOEC) technology is currently on research and development level and there are not many publications related to the modelling of its operations, especially in off-design conditions (i.e. when the input power differs from the design value). One of the few studies about the off-design characteristics of a SOEC is that presented by Motylinski et al [14], in which a SOEC operated in electrolysis mode has been modelled and analysed; the used methodology derives from a solution used to predict the performance of fuel cell power units with solid oxide technology, both in steady-state [15] and in transient operations [16]. Furthermore, as reported by Safari et al. [17], the Synthetic Natural Gas (SNG) generated by the P2G plant can be fed into the existing gas grid and marketed as an emission-free option for SNG-fuelled users; since the capacity for SNG is high around the world and the methanation process captures CO₂, the SNG production can be the most promising P2G technology.

In this framework, the innovative step of the Authors' study is the numerical prediction of off-design performance for a P2G process including a SOEC operated in co-electrolysis [18] mode. Indeed, the aim of this study mainly stands in the development of a calculation model for the off-design operations of the P2G process, focusing on a particular and innovative P2G system based on a high temperature co-electrolyzer (simultaneous electrolysis of H₂O and CO₂) of SOEC technology coupled with an advanced experimental methanator, which allows to operate at relatively high temperatures.

66 This work is a prosecution of Author's previous study [19], whose novelty stood in the development and analysis of the P2G 67 system, considering the possibility to thermally integrate the electrolysis with the methanation process, as a first step towards a 68 physical integration between the two components. In this previous study, the analysis has been carried-out in design condition, 69 considering all the sub-sections working at their set-points of operating temperature and pressure. In order to evaluate the 70 behaviour of the several components in off-design conditions a numerical code has been developed in the study here reported. 71 In more detail, the code has been developed in commercial tool environments and it is able to predict the operating point shift 72 of the several components during the off-design operations. The analysed key parameters are the operating temperature and 73 pressure. The pressurization of the system has been investigated since it can [20]:

74 - increase the SOEC power density;

75 - improve the produced SNG quality;

76 - reduce the size of the auxiliary components.

Furthermore, different temperature settings have been evaluated in order to explore possible thermal synergies among the subsections; the thermal synergy can be achieved operating both the co-electrolyzer and the methanation section within relatively high temperature ranges (in particular, the co-electrolysis operation at intermediate temperature has been demonstrated in a work by Lo Faro et al. [21]). In addition, in order to predict co-electrolysis behaviour under several conditions, a specific calculation sub-model has been integrated in the whole system model. Then, in order to evaluate the off-design performance of the P2G process under different operating conditions, four system variants have been analysed, starting from a real wind production profile and evaluating the annual operating time of the integrated P2G-wind systems.

In particular, in Section 2 of the paper the studied P2G system is introduced and the analysed configurations are presented. In Section 3, the P2G numerical model for off-design operations is described. In Section 4 and in Section 5, the analysis of the P2G system coupled with a wind generator and a preliminary economic analysis are carried-out. Finally, the results of the performed analyses are provided and discussed in Section 6.

88 2. The Power-to-Gas system: description and analysed configurations

89 As previously mentioned, the considered P2G process refers to a previous study of the Authors [19], where this innovative 90 P2G system has been proposed. In particular, the innovative feature of the P2G system is the coupling of a high temperature 91 co-electrolyzer (600 - 850 °C) with an experimental advanced methanation reactor, consisting in a fixed-bed reactor with a new 92 formulation of structured catalyst, which allows to operate at relatively high temperatures (450 - 600 °C).

93 Fig. 1 shows a simplified scheme of the P2G system in study. Briefly, the system is characterized by water and carbon dioxide 94 as main material input streams, while the energy input consists in electric power, from NP-RES (Non-Programmable 95 Renewable Energy Sources), and thermal power, from an external heat source. The main output of the system is a SNG stream, 96 to be introduced into the natural gas (NG) network. The P2G system includes also a sweep-air inlet stream shown in figure, for 97 the SOEC anodic-side products removal (not shown in figure for sake of simplicity), and a tail-end output water stream, 98 separated from the generated SNG.

99



- 101
- 102

Fig. 1. Simplified block diagram of the P2G system [19].

103

104 In more detail, the system is composed by three major sections: a reactants pre-heating section including a HRS (Heat 105 Recovery Section) for the P2G internal heat recovering (used for the inlet water partial preheating, as shown in Fig. 1), a high-106 temperature (HT) section (highlighted in red in Fig. 1) and a downstream low-temperature (LT) section (in blue in figure). In 107 particular, the HT section includes the following components:

- 108 a high temperature co-electrolyzer (SOEC in figure), where H_2O and CO_2 are converted into hydrogen and carbon 109 oxide as main species, by means of the electric power input to the P2G storage system;
- 110 a high temperature methanation (HTM) sub-section, where the methanation reactions occur and the reactants are 111 converted into a rough SNG. This sub-section has been modelled on the basis of an experimental structured catalyst.

112 The downstream LT section has been introduced in order to improve the quality of the rough SNG produced by the HTM. In

113 particular, the LT section is composed by:

- a low temperature methanation (LTM) sub-section, based on conventional catalytic methanation technology;
- an additional SNG processing section, where the SNG is mainly compressed, cooled and separated from residual
 water, before the introduction into the NG distribution network.
- Regarding the operating temperature, the pressure levels and the heat recovery section arrangement, all of these design settings
 have been the subject of an in-depth comparative analysis among several configurations reported in [19].
- 119

120 **2.1 Power-to-Gas system configurations**

121 In order to evaluate the off-design performance of the P2G system under different operating conditions, four configurations 122 have been considered in this study. The key analyzed parameters are the operating pressure and the operating temperature of 123 the several sub-sections of the P2G system; in particular, the pressurization of the P2G system has been investigated since it 124 can significantly increase the SOEC power density, improve the produced SNG quality and reduce the size of the auxiliary 125 components; furthermore, different temperature settings have been evaluated, in order to explore possible thermal synergies 126 among the sub-sections. The choice of the SOEC (and then of the downstream sub-sections) pressure level has generated two 127 different layouts of the P2G system. The first one (Fig. 2) is a P2G system with the SOEC and the methanation sections (HTM 128 and LTM) at ambient pressure and with a tail end pressurization, before the introduction into the NG network. The final SNG 129 storage pressure has been set equal to 60 bar, consistent with a high-pressure NG pipeline.

The second one (**Fig. 3**) is a P2G system with a pressurized SOEC. Thus, in this system, the HTM and the LTM sections operate at the same pressure of the SOEC; also in this case, at the end of the process the produced SNG is pressurized up to the same final storage pressure (60 bar). As shown in **Fig. 3**, the pressurized layout requires at the inlet a pump for the inlet water stream and a compressor for the inlet carbon dioxide stream. Moreover, the pressurization of the Sweep Air stream has also been considered.

- Regarding the pressurized layout, for the SOEC (and then for the downstream sub-sections) an operating pressure equal to 8 bar has been chosen; this value is in line with the current state-of-the-art of pressurized SOEC technology (even if higher values are also expected in the future – see the HELMETH project [22]) and it corresponds to the experimental data provided in the study by Mehran et al. [23] on a prototypal co-electrolyzer SOEC.
- 139 Then, for both layouts, two different temperature settings have been considered. Indeed, the studied configurations can be 140 summed up in:
- Ambient 1: P2G system at ambient pressure, with the SOEC at 850 °C, the HTM at 450 °C and the LTM at 200 °C;

- Ambient 2: P2G system at ambient pressure, with the SOEC at 600 °C, the HTM at 600 °C and the LTM at 200 °C;

- Pressurized 1: pressurized P2G system, with the SOEC at 850 °C, the HTM at 450 °C and the LTM at 200 °C;

- Pressurized 2: pressurized P2G system, with the SOEC at 600 °C, the HTM at 600 °C and the LTM at 200 °C.

145 In the configurations Ambient 1 and Pressurized 1, the SOEC operating temperature has been set equal to 850 °C, in line with 146 high performance SOEC operating conditions, in accordance to the available studies on high temperature SOEC [24]. The 147 HTM operating temperature has been set equal to 450 °C, in order to exploit the HTM highest conversion rate value, according 148 to [19]. The LTM operating temperature has been set after a parametric study of its effect, considering the typical temperature 149 range of operation of this technology [25]. On the other hand, in configurations Ambient 2 and Pressurized 2, in order to 150 explore possible thermal synergies among the sub-sections, different temperature levels have been investigated in the Authors' 151 previous study [19]; in particular, the thermal synergy can be achieved operating both the co-electrolyzer and the methanation 152 section within relatively high temperature ranges (the co-electrolysis operation at intermediate temperature has been 153 demonstrated in a work by Lo Faro et al. [21]).



Fig. 2. Layout of the P2G system at ambient pressure.



- 156
- 157

Fig. 3. Layout of the pressurized P2G system.

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159 3. The Power-to-Gas system numerical model

160 The P2G system thermodynamic model has been developed in ASPEN HysysTM environment [26], a commercial tool for 161 numerical lumped-parameter modelling of complex energy systems. Standard units from ASPEN HysysTM library have been 162 employed to model common components, like separators, heat exchangers, pumps and compressors, while specific sub-models

163 have been implemented for the key components of the P2G system. In particular, the thermodynamic model includes:

164 - a preheating HRS, made by several heat exchanging segments;

- three different reactors for the SOEC, to simulate the electrolytic reactions, the reverse water-gas shift reaction and
 the methane formation reaction:
- 167 $H_2 O \to H_2 + \frac{1}{2} O_2$ (1)

168
$$CO_2 \to CO + \frac{1}{2}O_2$$
 (2)

- $169 \qquad \qquad CO_2 + H_2 \leftrightarrow CO + H_2O \tag{3}$
- $170 \qquad \qquad CO + 3H_2 \leftrightarrow CH_4 + H_2O \tag{4}$
- 171 a conversion reactor for the HTM section;
- 172 a multi-step equilibrium reactor for the LTM section;
- 173 a two-step intercooled final compression, with water separation.

The reactions occurring in the SOEC are mainly influenced by the operating temperature and pressure; in addition, also the mass stream at the inlet of the reactors and the chemical composition of the stream can condition the SOEC behaviour. More details on the thermochemical model, on the reactors arrangements and on the chemical settings, can be found in the previous 177 study of the Authors [19], where a parametric design analysis of the P2G system has been carried out, taking into account an 178 electric power supply design point for the SOEC set at 1 MW. Furthermore, in the Authors' study [19], the sensitivity analysis 179 of the ASPEN HysysTM model on boundary conditions and inlet power condition has been performed.

The main set point parameters in design conditions implemented in the P2G system are summarized in **Table 1**. In particular, the conversion rate target of electrolysis reactions is assumed equal or higher than the 80 % in [27]: in this study, a conservative value equal to the 80 % is set. Furthermore, the O_2 molar fraction in the anode outlet is set at 0.5, as recommended in [28]. Finally, in order to allow the introduction of the produced SNG into the NG network, the pressure at the outlet of the systems has been set on the basis of typical values for high pressure lines of the Italian natural gas network [29].

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Table 1. Main set point parameters of the P2G system in design conditions for the analysed cases.

Parameter	Ambient 1	Ambient 2	Pressurized 1	Pressurized 2
P2G inlet reactants temperature [°C]	25	25	25	25
P2G inlet reactants pressure [bar]	1	1	1	1
SOEC input electric power [MW]	1	1	1	1
SOEC operating temperature [°C]	850	600	850	600
SOEC operating pressure [bar]	1	1	8	8
SOEC inlet H ₂ O fraction [% vol.]	80	80	80	80
SOEC inlet CO ₂ fraction [% vol.]	20	20	20	20
H ₂ O electrolysis reaction Conversion Rate [%]	80	80	80	80
CO2 electrolysis reaction Conversion Rate [%]	80	80	80	80
O2 molar fraction in the anode stream [-]	0.5	0.5	0.5	0.5
HTM operating temperature [°C]	450	600	450	600
HTM operating pressure [bar]	1	1	8	8
LTM operating temperature [°C]	200	200	200	200
LTM operating pressure [bar]	1	1	8	8
NG distribution network pressure [bar]	60	60	60	60

187

As previously mentioned, the aim of this study is the performance prediction of the P2G system both in design and off-design conditions; in particular, the system operates in off-design when the input electric power differs from the design value. In order to take into account the off-design operations, the thermodynamic model developed in ASPEN HysysTM has been integrated with a

191 specific calculation sub-model for the SOEC, developed in MATLABTM environment [30], as shown in Fig. 4. The carried-out 192 analysis is of quasi-static type; this assumption implies that the considered system has a time-dependent response, but the 193 inertia effects have been neglected. As a consequence, the proposed model does not consider process dynamics. However, 194 since process dynamics (accumulation of mass and energy in the several components of the plant) can play an important role in 195 the estimation of the system performance, this aspect will be considered in future studies.

196



197 198

Fig. 4. Simplified block diagram of the numerical procedure for design and off-design analysis of the P2G system.

199

In more detail, the calculation procedure starts with the P2G system layout definition and the thermodynamic design analysis, performed within the ASPEN HysysTM environment, based on the set point design power input and on the basis of the other system boundary conditions of **Table 1**. This calculation provides as output all the internal and outlet stream flows and enthalpy values, corresponding to the reference inlet power condition. Then, off-design calculation of the same system performance under different electric power inputs can be performed, by interrogating a specific SOEC sub-model (MATLABTM sub-routine); the SOEC submodel is based on external experimental data for a reference SOEC stack (see **Fig. 4**). The used experimental data derive from an experimental study [23], in which a co-electrolyzer of SOEC technology has been tested in a limited set of operating conditions. The empirical SOEC sub-model calculation provides, as output, data on the SOEC off-design operations (mainly the reaction products outlet temperature, T_{oUT} , and the processed mass flow, \dot{m}_{SOEC}) which affect the behaviour of the other P2G subsections. Thus, these data are used as input by the P2G system overall thermodynamic model (in ASPEN HysysTM), to obtain all the stream flows and enthalpy values in the off-design operating point. Finally, the values of the several quantities in the off-design operating point are transferred to MATLABTM environment, where performance parameters are calculated.

The HRS is necessary in order to pre-heat the water stream at the inlet of the system, using heat available at different downstream sections of the P2G system. In more detail, heat is recovered from the SOEC outlet cathode and anode streams and from all the methanation reactors cooling sections. The HRS heat-exchangers arrangement in the heat recovery line has been optimized in the Authors' previous work [19] evaluating the temperature levels of the available heat flows. In particular, it has been decided to place the HRS in the more heat demanding water line. Despite this, in order to feed the SOEC with reactants at its operating temperature, an external heat source has also been considered.

When in off-design conditions, the values of the temperatures and of the mass flows of the streams involved in the HRS differ from the values in design condition, mainly due to the different SOEC input electric power and outlet temperature.

220 In order to simulate the off-design behaviour of the HRS, first of all, the (U·A) values (where U $[W/m^2K]$ is the global heat 221 transfer coefficient and A [m²] is the heat exchanger surface) of each heat exchanger of this section have been calculated in 222 design conditions (these values have been obtained through the simulation of the P2G model in ASPEN HysysTM). In **Table 2** 223 the (U·A) values of the HRS in design condition for the four analysed configurations are reported. When the SOEC and the 224 HTM work at the same temperature value of 600 °C (Ambient 2 and Pressurized 2), the heat exchanger (HE-6) used to 225 recovery the heat from the stream at the outlet of the SOEC (and then at the inlet of the HTM) is not necessary. In these two 226 configurations, the heat exchanger with the highest value of UA is HE-1 (2115 W/K for Ambient 2 and 6047 W/K for 227 Pressurized 2). On the other hand, when the SOEC and the HTM work at different temperatures (Ambient 1 and Pressurized 228 1), an additional heat exchanger is required (HE-6) and it is the one with the highest value of UA.

- 229 After having obtained the (U·A) values in design condition, they have been kept constant also in off-design conditions.
- Regarding the methanation sections (HTM and LTM), the chemical reactors work at the temperature set-point (see **Table 1**) and the
- chemical reactions are able to follow the change of the mass flow value in off-design conditions.
- Finally, the compression and separation section (C & Sep) considers the electrical energy consumption variation of the compressors
- 233 due to the different processed mass flow of the produced SNG in off-design conditions.
- 234 In the following paragraph, the co-electrolyzer off-design sub-model will be described.

235	Table 2. UA values of the HRS heat exchangers for the four analysed configurations in design condition (values obtained)
236	through the simulation of the P2G model in ASPEN Hysys TM).

	UA [W/K]				
	Ambient 1	Ambient 2	Pressurized 1	Pressurized 2	
HE-1	2124	2115	4075	6047	
HE-2	623.9	539.2	713.1	657.5	
HE-3	117.9	120	221.9	134.7	
HE-4	209	164.1	757.8	240.1	
HE-5	2775	589.2	773.1	725.8	
HE-6	5100	-	17290	-	

238 **3.1 Co-electrolyzer semi-empirical sub-model**

The developed calculation sub-model (**Fig. 5**) allows to evaluate the outlet temperature and the electrical efficiency of the SOEC in off-design conditions starting from input parameters. The knowledge of these two output parameters is essential in order to predict the behaviour of the whole P2G system in off-design conditions; indeed, the SOEC efficiency strongly affects the P2G system efficiency due to the high electrical energy consumption of this sub-section, while the outlet temperature value determines the operating point of the other sub-sections.

The model inputs can be divided into two main categories: experimental data and actual operating conditions. In particular, the empirical model requires, as experimental data, a limited number of information derived from an existing SOEC stack, used as reference; the reference experimental data are: i) a set of polarization curves (i.e. voltage-current density of the experimental stack) at different operating conditions, ii) the stack surface (A_{SOEC}) and iii) the stack inlet mass flow (\dot{m}_{SOEC}). The used experimental data (polarization curves and stack data) derive from an experimental study [23], where a SOEC co-electrolyzer has been tested under different pressure and temperature conditions.

250 The actual operating temperature (T_{SOEC}) and pressure (p_{SOEC}) are additional input of the off-design model and they can be

- 251 different from the temperature and pressure tested in the experiments.
- 252 The process flow of this sub-model can be divided into three steps:
- 253 i) calculation of generalized polarization curves;
- 254 ii) design condition definition;
- 255 iii) off-design performance evaluation.
- Each step of the sub-model will be described below.





Fig. 5. Flow chart of the SOEC off-design semi-empirical sub-model.

258

260 <u>i) Calculation of generalized polarization curves</u>

The first step of the sub-model consists in the calculation of generalized polarization curves, i.e. first-order polynomials that describe the evolution of the cell voltage as a function of the current density. Starting from a limited set of experimental polarization curves for the reference stack, the model is able to obtain the polarization curves of the SOEC under extended operating conditions (inlet temperature and operating pressure) through the interpolation and extrapolation of the experimental data with first-order polynomials (**Fig. 6a** and **Fig. 6b**); the slope of the polynomials has been obtained taking into account the temperature and pressure variations.

The used experimental polarization curve points derive from an experimental study [23], in which a co-electrolyzer of SOEC technology has been tested in a limited set of operating conditions (the investigated temperature range is 750-850 °C, while the pressure range is 1-8 bar). Regarding the operating temperature, the experimental polarization curves have been extrapolated in a wider range, between 600 °C and 850 °C, i.e. the extended operating range considered in this study (see **Fig. 6a**), typical for a co-electrolyzer of SOEC technology [31]. Moreover, generalized polarization curves have been obtained also for different pressure values, by interpolating the available experimental points (**Fig. 6b**).

273





Fig. 6a. Generalized (extrapolated) polarization curves plotted for various operating temperature values, at $p_{SOEC} = 1$ bar (dots: experimental data points [23]; continuous lines: polynomial curves).

Fig. 6b. Generalized (interpolated) polarization curves plotted for various operating pressure values, at $T_{SOEC} = 850$ °C (dots: experimental data points [23]; continuous lines: polynomial curves).

274

275 <u>ii) Design condition definition</u>

- 276 The SOEC design operating point i.e. the cell voltage and current density in design condition has been identified on the
- 277 polarization curve for the actual operating conditions, by referring to the generalised experimental polarization curves.
- 278 In particular, in this study, the SOEC design point has been assumed coinciding with the "thermoneutral" condition, where the
- thermoneutral voltage V_{tn} is defined as the potential at which the cell is thermally stable with respect to its equilibrium state at
- 280 V_{OCV} (Open Circuit Voltage) [32]. In thermoneutral condition, the heat released by the electric losses in the cell is equal to the

energy required by the co-electrolysis reactions. This operating condition is desirable for SOEC technology, in order to avoid any additional device to provide heat or to extract it from the stack and this condition leads to have the same temperature value for the inlet reactants and the outlet products.

In off-design conditions instead, the heat released by the electric losses in the cell differs from the energy required by coelectrolysis reactions; in general, the electric energy demand due to the co-electrolysis reactions can be expressed as the variation of the Gibbs free energy Δg :

$$287 \qquad \Delta g = \Delta h - T \cdot \Delta s \tag{5}$$

where, Δh [kJ/kmol] is the enthalpy variation of the co-electrolysis reactions, *T* [K] is the SOEC operating temperature equal to the reactants inlet temperature and Δs [kJ/kmolK] is the entropy variation of the reactions; it must be highlighted that the enthalpy variation and the entropy variation are influenced by the operating temperature and pressure of the SOEC.

If $V_{cell} < V_{tn}$ (endothermic mode), the electric energy Δg is lower than the enthalpy variation Δh and then additional heat is required to maintain the operating temperature.

If instead $V_{cell} > V_{tn}$, the cell operates in the exothermic mode and this corresponds to an increase in the cell temperature, because the electric energy supply Δg exceeds the enthalpy variation.

295 Thus, in order to evaluate the thermoneutral voltage, the following thermal balance equation has to be solved:

$$296 \qquad I \cdot \Delta V_{tn} = \dot{n}_{syn} \cdot T \Delta s \tag{6}$$

where, *I* [A] is the electric current of the SOEC (*I*=*J*·*A*_{SOEC}), ΔV_{tn} [V] is the voltage related to the losses in thermoneutral condition and \dot{n}_{syn} [kmol/s] is the molar flow of produced syngas. According to the Faraday's Law (7), \dot{n}_{syn} in the electrochemical reactions can be expressed as:

$$300 \qquad \dot{n}_{syn} = \frac{1}{ZF} \tag{7}$$

301 where, *Z* is the valency number of ions of the substance and *F* [C/kmol] is the Faraday constant.

302 Then, eq. (6) can be reformulated as:

$$303 \qquad \Delta V_{tn} = \frac{T\Delta s}{ZF} \tag{8}$$

304 Finally, V_{tn} can be calculated by eq. (9):

$$305 \qquad V_{tn} = V_{OCV} + \Delta V_{tn} \tag{9}$$

306 Using the calculated V_{tn} value at given temperature and pressure, the corresponding design current density can be obtained 307 directly from the generalized polarization curve.

- 308
- 309

310 iii) Off-design performance evaluation

Once the design (thermoneutral) point is identified on the polarization curve, it is possible to identify the off-design point, i.e. when the input electric power value ($P_{el,off-design}$ [W]) and the related current ($I_{off-design}$ [A]) and voltage ($V_{off-design}$ [V]) values differ from the design values; then, the new SOEC performance and in particular the updated SOEC outlet temperature (T_{out} [K]) can be calculated in off-design operations. This temperature value coincides with the value of the inlet temperature (T_{soEC} [K]) only in design conditions. The off-design T_{out} estimation can be performed using the heat released by electric losses in off-design conditions ($\dot{Q}_{off-design}$ [W]) expressed according to eq. (10):

$$317 \qquad \dot{Q}_{off-design} = \left(V_{off-design} - V_{OCV}\right) \cdot I_{off-design} \tag{10}$$

In this equation, in order to quantify the heat released by electric losses, the heat flux has been expressed as the product of the electric current in off-design conditions and the difference between the voltage and the open circuit voltage (i.e. the voltage given by the electric current flow in the circuit). Combining this equation with the expression of the heat released by electric losses in thermoneutral condition (\dot{Q}_{design} [W]), the outlet temperature in off-design conditions can be estimated as:

$$322 T_{out} = T_{SOEC} + \frac{\dot{q}_{off-design} - \dot{q}_{design}}{\dot{m}_{SOEC} \cdot c_p} (11)$$

323 where \dot{m}_{SOEC} is the actual SOEC inlet reactants mass flow [kg/s] and c_p [J/kgK] is the related specific heat.

324 Finally, the SOEC electrical efficiency [22] can be calculated according to:

325
$$\eta_{SOEC} = \frac{\dot{n}_{syn} \cdot HHV_{syn}}{P_{el}}$$
(12)

where, HHV_{syn} [kJ/kmol] is the higher heating value of the produced syngas and P_{el} [kW] is the electrical power input of the SOEC related to the electrical current consumption of the cell ($P_{el}=V \cdot I$). According to (7), the SOEC efficiency is proportional to a constant factor per applied voltage:

329
$$\eta_{SOEC} = \frac{HHV_{syn}}{ZF} \cdot \frac{1}{V} \propto \frac{constant}{V}$$
 (13)

330 Then, the SOEC efficiency in off-design conditions can be calculated through the evaluation of the off-design voltage value.

331

4. Analysis of the system coupled with renewables

With the aim to evaluate the P2G system performance in off-design conditions, the coupling with a wind generator has been considered. In this study, only a specific type of renewable generator (wind) has been evaluated, even if other renewable sources can be exploited for this application; although a photovoltaic module, for example, presents an electricity production profile quite different with respect to the wind, the proposed methodology and model are general and can be applied also to other electrical energy sources. The SOEC design electric power size for each P2G configuration has been set at 1 MW and, in the calculation of the overall input power, also the compression and auxiliaries' consumption has been taken into account. The P2G off-design operating range has been assumed between -50 % and +50 % of the design inlet power.

Regarding the coupled wind source, a year of production has been analyzed, taking into account the annual wind generation duration curve data (see **Fig. 7**), referring to the Italian wind production (TERNA [33], [34]) available with a 5 min time step. The wind annual production has a peak value equal to 1700 kW, while the mean annual wind power is equal to 750 kW, corresponding to a nameplate power equal to 3 MW and 2200 equivalent hours of operation per year.

As already mentioned, the carried-out analysis is of quasi-static type. As a consequence, since the proposed model does not consider process dynamics, the considered system has a time-dependent response, but the inertia effects have been neglected. However, the process dynamics (accumulation of mass and energy in the several components of the plant) can play an important role in the estimation of the system performance and this aspect will be considered in future studies.

The analysis has been carried out with a dedicated code, developed in MATLABTM and integrated in the ASPEN HysysTM environment. This code allows to evaluate the annual operating time of the P2G system, the surplus of electric energy not used by the system – and then introduced into the electric grid or eventually wasted – and the values of the performance parameters. In more detail, the code analyzes each time step of the year:

if, in the given time step, the electric power produced by the wind plant is lower than the lower operating limit of the P2G
system, then all the electric energy is introduced into the electric grid;

if, in the given time step, the electric power produced by the wind plant is within the operating range of the P2G plant, then
all the electric energy is employed within the process;

if, in the given time step, the electric power produced by the wind plant is greater than the upper operating limit of the P2G
 system, then the difference between the wind production and the maximum electric load of the P2G system is introduced
 into the electric grid.



Fig. 7. The considered annual wind power production monotonic duration curve.

362

363 5. Preliminary economic analysis

In order to evaluate the cost effectiveness of the P2G technology, a preliminary economic analysis has been performed. The aim of this analysis is the estimation of the maximum investment cost of the P2G plant in order to have a competitive technology. In more detail, the economic feasibility of the whole project is evaluated through a cash flow analysis, based on the Net Present Value (NPV) defined as:

$$368 \qquad NPV = -I_0 + \sum_{i=1}^{t} \frac{CF_i}{(1+r)^i} \tag{14}$$

where, I_0 [\in] represents the total initial investment (P2G system), r is the discount rate (assumed equal to 7 %), CF_i [\in] is the net cash inflow at the i-th year and t [year] is the useful time horizon. The cash inflow for the general year i can be calculated as it follows:

$$372 CF_i = R_{SNG} - C_{O\&M} (15)$$

where, R_{SNG} [€] is the revenue due to the SNG sell and $C_{O\&M}$ [€] is the operation and maintenance cost of the P2G plant. It should be highlighted that the electric power cost has not been taken into account due to the renewable generation. The $C_{O\&M}$ of the P2G plant has been assumed equal to 45 €/h according to [20]; the operation and maintenance cost has been related to the P2G operating time. In this economic analysis, a time horizon equal to 10 years has been assumed.

377 The aim of this analysis is to find the I_0 in order to have a return on the investment in the considered time horizon, varying the

378 SNG cost. As a consequence, the economic problem can be expressed as:

379
$$NPV = 0, I_0 = \sum_{i=1}^n \frac{CF_i}{(1+r)^i}$$
 (16)

380 In the framework of a parametric analysis, the cost of the SNG has been varied from 0 to $100 \notin$ /MWh in order to evaluate the 381 P2G investment cost behaviour. It must be highlighted that in the SNG sell, only the methane content has been taken into 382 account.

In addition, in order to consider in the analysis a form of economic incentive for this emerging technology, a financial bonus in the SNG sell has been considered. In more detail, an economic incentive equal to $32 \notin$ /MWh has been assumed, according to the Italian legislation for biomethane injection into the natural gas grid [35].

In this economic analysis, the generated electricity that is introduced into the electric grid has not been accounted within the cash flows. However, in order to evaluate the P2G process as an energy storage system, this quantity has been calculated through the proposed model; indeed, with the aim to reduce over-generation (when the power production from renewables exceeds the demand), the electric energy discharged to the grid can be a critical parameter to assess the performance of an energy storage system. In addition, since the generated electricity introduced into the grid can be a marginal value of the natural gas price, it will be analyzed in future studies.

392

393 6. Results and discussion

In this section, the results of the carried-out analysis are shown. In more detail, at first the results of the SOEC and of the HRS sub-models are presented and, then, the results of the comparative analysis among the several P2G system configurations are discussed. Finally, the results of the preliminary economic analysis are reported.

397

398 6.1 Co-electrolyzer off-design performance

399 In this section, the SOEC off-design non-dimensional curves, which represent the component behavior depending on the 400 operational load, are shown. In particular, in Fig. 8a, the normalized polarization curves (evolution of the cell voltage as a 401 function of the cell current density) for several operating temperatures are shown. All of these quantities are expressed in a 402 dimensionless form with respect to the design condition. The results show that, with the increase in the temperature, a lower 403 voltage is required for current densities lower than the design point - corresponding to the point (1;1) in the graph - while for 404 greater current densities the situation is the opposite. Furthermore, the pressurized SOEC behavior has been evaluated (Fig. 8b), considering a pressure range between 1 bar and 8 bar (experimental SOEC operating range) with a step equal to 1 bar. The 405 406 pressurization of the SOEC appears to be penalized due to the higher voltage requested for current densities lower than the 407 design point current density; for current densities higher than the design point current density, the situation is the opposite.





Fig. 8a. Effect of T_{SOEC} on the normalised polarization curve for $p_{SOEC} = 1$ bar.

Fig. 8b. Effect of p_{SOEC} on the normalised polarization curve for T_{SOEC} = 850 °C.

In **Fig. 9a**, the outlet temperature of the SOEC as a function of the electric load for several temperatures is presented. As it can be noted, the outlet temperature varies from a minimum of about -15 % (for an electric load equal to 50 % of the design point) to a maximum of about +13 % (for an electric load equal to 150 % of the design point), considering an operating temperature of 850 °C. For lower operating temperatures the slope of the curve is lower. Indeed, for electrical loads lower than the design point, the value of the voltage at set current density is higher for lower operating temperatures (see **Fig. 8a**) and, then, the outlet temperature is higher, according to (10)-(11); for electrical loads higher than the design point, the situation is the opposite: this leads to a slope of the curve lesser degree.

In **Fig. 9b**, the effect of the operating pressure on the outlet temperature of the SOEC as a function of the electric load is shown. The outlet temperature varies from a minimum of about -11 % (for an electric load equal to 50 % of the design point) to a maximum of about +10 % (for an electric load equal to 150 % of the design point), considering an operating pressure of 8 bar. In this case, for lower operating pressures the slope of the curve is higher. This behaviour can be explained through the evaluation of the voltage (see **Fig. 8b**) for the several operating pressures, according to (10)-(11).



Non-dimensional outlet temperature [-] 1 bar 2 bar 3 bai 4 ba 1.1 5 bar 6 bar 7 bai 8 bar Design point 0 T_{SOEC} = 850 °C 0.8 ∟ 0.5 0.75 1.25 1.5 Electric load [-]

Fig. 9a. Normalised outlet temperature as a function of the electric load for several operating temperatures (p=1 bar).

Fig. 9b. Normalised outlet temperature as a function of the electric load for several operating pressures (T= 850 °C).

The SOEC efficiency gain is shown as a function of the electrical load for several temperatures in **Fig. 10a.** In this figure, the design point is represented by the point (0;1), with a null efficiency gain in correspondence of the design electric load. The efficiency gain varies from a maximum of about + 14% (for an electric load equal to 50 % of the design point) to a minimum of about -10 % (for an electric load equal to 150 % of the design point) for an operating temperature of 850 °C. Even in this case, for lower operating temperatures the slope of the curve is lower.

The SOEC efficiency gain is presented as a function of the electrical load for several pressures in **Fig. 10b**. The efficiency gain varies from a maximum of about +10 % (for an electric load equal to 50 % of the design point) to a minimum of about -7 % (for an electric load equal to 150 % of the design point) for an operating pressure of 1 bar. For lower operating pressures the slope of the curve is higher.





Fig. 10a. SOEC efficiency gain as a function of the electric load for several operating temperatures (p= 1 bar).

Fig. 10b. SOEC efficiency gain as a function of the electric load for several operating pressures (T= 850 °C).

432 **6.2 Heat recovery section off-design performance**

433 In this section, the off-design performance of the HRS are shown. In particular, the HRS heat exchanged in design and off-434 design conditions for the four analyzed configurations is presented in Fig. 11. In all the considered configurations, the heat 435 exchanger with the highest value of heat exchanged is HE-1, with a value between 105 kW and 109 kW in design condition. 436 This situation is also confirmed in off-design conditions: HE-1 shows a value of the exchanged thermal power higher than 150 437 kW for all the analyzed configurations at 150 % of the load and a value of about 50 kW at 50 % of the load. Comparing 438 configurations with the same temperature setting, the results are similar: indeed, for Ambient 1 (Fig. 11a) and Pressurized 1 439 (Fig. 11c), the thermal power exchanged in the whole HRS in design condition is equal to about 295 kW for both the 440 configurations; Ambient 2 (Fig. 11b) shows a value of the thermal power exchanged in design condition slightly higher with 441 respect to the configuration Pressurized 2 (Fig. 11d), respectively 242 kW and 233 kW.

442



Fig. 11. HRS heat exchanged for the four analysed configurations in design and off-design conditions: a) Ambient 1; b) Ambient 2;c) Pressurized 1; d) Pressurized 2.

444 **6.3 Power-to-Gas system performance**

445 In Table 3, the results of the comparative analysis among the several considered variants are shown. The system Ambient 2 446 shows the highest number of operating hours, equal to 3896 h/year, due to the lowest electrical consumption with respect to the 447 other variants. Indeed, while the SOEC input power is the same for all the variants, the overall electrical consumption is 448 different and, in particular, higher for the pressurized systems. The operating time of the system Ambient 2 corresponds to 449 about 45 % of the whole length of the year. Due to the lower operating time, the pressurized systems (Pressurized 1 and 450 Pressurized 2) present a higher amount of electric energy introduced into the electric grid (1231 MWh/year for Pressurized 1 451 and 1224 MWh/year for Pressurized 2), and a lower energy utilization factor, equal to about 74 % for both the systems; on the 452 contrary, the systems at ambient pressure (Ambient 1 and Ambient 2) show an energy utilization factor equal to about 75 %.

Regarding the SNG production, the systems at ambient pressure show higher values (216 ton/year for Ambient 2) with respect to the pressurized systems (188 ton/year for Pressurized 2). As a consequence, also the methane production follows the results of the SNG production; indeed, even if the SNG quality is higher for the pressurized systems, the systems at ambient pressure show a higher operating time and then a higher SNG production, that leads to a slightly higher methane production (184 ton/year for Ambient 2).

458 In order to evaluate the efficiency of the P2G variants throughout the year, the utilization efficiency (η_U) has been defined:

$$459 \qquad \eta_U = \frac{\sum_i m_{SNG,i} \cdot LHV_{SNG,i}}{E_{WIND}} \tag{17}$$

460 where, $\dot{m}_{SNG,i}$ [kg/s] is the mass flow of produced SNG at the i-th instant of the year, $LHV_{SNG,i}$ [kJ/kg] is the lower heating 461 value of the produced SNG at the i-th instant of the year and E_{WIND} [kWh] is the wind energy available in the whole year.

462 From the point of view of this efficiency, Ambient 2 shows better performance with respect to the other variants: indeed, for463 Ambient 2 the utilization efficiency is equal to 59 %.

464 In order to take into account also the thermal energy consumption, the overall first law efficiency has been introduced:

$$465 \qquad \eta_{I,TOT} = \frac{\sum_{i} \dot{m}_{SNG,i} \cdot LHV_{SNG,i}}{\sum_{i} P_{in,i} + \sum_{i} \dot{Q}_{in,i}}$$
(18)

466 where, $P_{in,i}$ [kW] is the P2G system inlet electric power at the i-th instant of the year and $\dot{Q}_{in,i}$ [kW] is the total amount of 467 input heat required by the P2G system at the i-th instant of the year.

The system Ambient 1 shows better performance here too, with a value of about 75 %, even if the thermal consumption is higher for this system, with a value of about 175 MWh/year. This result can be explained through the evaluation of the SNG production, which results higher for this system.

471

Parameter	Ambient 1	Ambient 2
P2G operating time [h/year]	3895	3896
Utilization factor [%]	44.4	44.5
Wind energy available [MWh/year]	4644	4644
Energy to the grid [MWh/year]	1175	1174
Energy utilization factor [%]	74.7	74.7

Total thermal energy externally requested [MWh/year]

SNG production [ton/year]

CH₄ production [ton/year]

Utilization efficiency [%]

Overall first law efficiency [%]

 Table 3. Annual results of the comparative analysis among the several P2G variants.

215

183

160

58.3

74.6

216

184

175

59.0

75.1

Pressurized 1

3784

43.2

4644

1231

73.5

189

181

117

55.1

72.5

Pressurized 2

3797

43.3

4644

1224

73.6

188

183

154

55.3

71.9

474

473

475 In order to analyse the off-design performance of the studied variants, the instantaneous values of the key calculated quantities 476 are presented in Fig. 12-14. In Fig. 12, the instantaneous SNG production throughout the year is shown. The system Ambient 2 477 presents a production slightly higher than the system Ambient 1 in the times of the year with the highest wind power 478 production (with a peak of about 0.028 kg/s), while when the wind production decreases the system Ambient 1 shows better 479 performance from this point of view. The pressurized systems present nearly the same SNG production, lower with respect to 480 the systems at ambient pressure. In Fig. 13, the instantaneous CH₄ production is shown. For all variants, the methane 481 production is about the same, with the system Pressurized 2 showing a slightly higher production, due to the higher quality of 482 the produced SNG. In Fig. 14, the instantaneous first law efficiency values are shown as a function of the wind production. 483 The trend of the first law efficiency highlights that the lower is the electric load, the higher is the efficiency due to the trend of 484 the SOEC efficiency (Fig. 10a and Fig. 10b). In particular, the system Ambient 1 shows a peak of about 0.82.



Fig. 12. The instantaneous SNG production during the year.



Fig. 13. The instantaneous CH₄ production during the year.







Fig. 14. The instantaneous first law efficiency as a function of the wind production.

493 **6.4 Economic analysis**

494 In this section, the results of the economic analysis are shown. In particular, in Fig. 15 the investment cost per unit of P2G 495 plant size as a function of the SNG cost is presented. The difference in the trend among the configurations is almost negligible, 496 resulting in overlapping lines; thus, only configuration Ambient 2 has been considered in this section, since it leads to a higher 497 methane production and a higher number of operating hours. The natural gas market price range is highlighted: lower bound 498 corresponds to 2019 average NG price for industrial uses in Euro area (28.395 €/MWh); upper bound represents the 2019 499 average NG price for household consumers in Euro area (63.226 €/MWh). Both values refer to Eurostat [36]; NG prices for 500 industrial uses are defined as follows: average national price without taxes applicable for the first semester of each year for 501 medium size industrial consumers (annual consumption between 10000 GJ and 100000 GJ); on the other hand, NG prices for 502 household consumers are defined as follows: average national price including taxes and levies applicable for the first semester 503 of each year for medium size household consumers (annual consumption between 20 GJ and 200 GJ). With a SNG cost 504 comprised in the NG market price range, configuration Ambient 2 has no return in the investment in the considered time 505 horizon (10 years). Starting from a SNG cost of about 70 €/MWh, the P2G investment cost became positive coming up to a 506 value of about 1500 €/MW for a SNG cost of 100 €/MWh. On the other hand, if the SNG sell is incentivised, there's a range of 507 SNG cost (comprised between about 40 €/MWh and the household NG cost) in order to be competitive in the NG market.

508 In summary, the competitiveness of the studied technology (methanation coupled with high temperature co-electrolysis) is 509 strongly dependent on the investment cost of the plant. Governments' action still might play a role in order to incentivise the 510 SNG sell. Moreover, an increase in the renewable energy sources within the electricity production mix of a specific area could result in a great impact on the local energy system; this will imply a progressive cost reduction of the produced electric energy. Also the full development of the SOEC technology is desirable in order to improve the competitiveness of the analysed system. Indeed, as reported by Schmidt et al. [37], the cost uncertainty is one of the most remarkable barriers to invest in electrolysis technology. On the other hand, the P2G process exhibits characteristics that distinguish itself with respect to the other storage technologies and could make it competitive in the energy market: i) it shows a potentially limitless storage capacity, if the produced SNG is introduced into the NG network; ii) the produced energy in the form of SNG is transportable even at great distances; iii) it could exploit the already available NG infrastructures.

518



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- 520

Fig. 15. Investment cost per unit of P2G plant size as a function of the SNG cost.

521 7. Conclusions

In this study, an innovative P2G system, based on a high temperature co-electrolyzer of SOEC technology coupled with an advanced experimental reactor, has been analysed in order to predict its off-design performance. In more detail, a numerical model has been developed for the P2G system and, in order to simulate the behaviour of the SOEC and of the HRS, specific calculation sub-models have been integrated in the whole system model. With the purpose of determining the performance of the considered P2G system in real operating conditions when coupled with renewables, a wind plant has been considered. In particular, during the analysis also the effect of the pressure on the system has been taken into account, evaluating four different configurations of the P2G process.

529 The results show that the most performant configuration is the one at ambient pressure, with the SOEC and the HTM operating

at the same temperature (600 $^{\circ}$ C); indeed, this configuration presents higher values of the considered performance parameters

531 with respect to the other configurations (a methane production equal to 184 ton/year and an overall efficiency of about 75 %).

532 Finally, with the aim to assess the cost effectiveness of the P2G process, a preliminary economic analysis has been performed;

533 the results point out that, in order to have a competitive technology, an economic incentive in the synthetic natural gas sell

must be applied.

- 535 Process dynamics (accumulation of mass and energy in the several components of the plant) can play an important role in the 536 estimation of the system performance and, then, this aspect will be considered in future studies.
- 537

538 Nomenclature

539 Α Area [m²] 540 с Specific heat [J/kgK] 541 Е Energy [kWh] 542 F Faraday constant - 96485000 C/kmol 543 Specific Gibbs free energy [kJ/kmol] g 544 h Specific enthalpy [kJ/kmol] 545 HHV Higher heating value [kJ/kg] 546 I Electric current [A] 547 LHV Lower heating value [kJ/kg] 548 J Current density [A/m²] 549 Mass flow [kg/s] ṁ 550 'n Molar flow [kmol/s] 551 Р Power [kW] 552 Thermal power [W] Q 553 Discount rate r 554 Specific entropy [kJ/kmolK] s 555 Time horizon [year] t 556 Temperature [K] t 557 Global heat transfer coefficient [W/m²K] U 558 V Voltage [V] 559 Ζ Valency number of ions 560 **Greek symbols**

562	η	Efficiency	
563	Subscripts and Superscripts		
564	el	Electrical	
565	in	Inlet	
566	OCV	Open circuit voltage	
567	out	Outlet	
568	р	Pressure	
569	syn	Syngas	
570	tn	Thermoneutral	
571	Acronyms		
572	С	Compression	
573	CF	Net cash inflow	
574	el	Electrical	
575	HT	High temperature	
576	HTM	High temperature methanation	
577	i	i-th	
578	LT	Low temperature	
579	LTM	Low temperature methanation	
580	NG	Natural gas	
581	NP-RES	Non-programmable renewable energy sources	
582	NPV	Net present value	
583	O&M	Operation and maintenance	
584	P2G	Power-to-Gas	
585	PV	Photovoltaic	
586	R	Revenue	
587	RES	Renewable energy sources	
588	Sep	Separation	
589	SNG	Synthetic natural gas	
590	SOEC	Solid oxide electrolyte cell	
591	ТОТ	Total	

592UUtilization593volVolumetric594IFirst law

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