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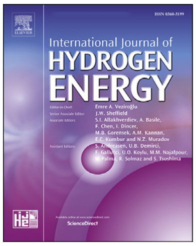

Cost-optimized design point and operating strategy of polymer electrolyte membrane electrolyzers

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HIGHLIGHTS

- Implementing electrochemical parameters into a techno-economic model of PEM-WE.
- Simulation of the operating strategy that leads to the lowest cost of hydrogen.
- Dependency of the strategy on the electricity price and profit margin.

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ABSTRACT

Green hydrogen is a key solution for reducing CO_2 emissions in various industrial applications, but high production costs continue to hinder its market penetration today. Better competitiveness is linked to lower investment costs and higher efficiency of the conversion technologies, among which polymer electrolyte membrane electrolysis seems to be attractive. Although new manufacturing techniques and materials can help achieve these goals, a less frequently investigated approach is the optimization of the design point and operating strategy of electrolyzers. This means in particular that the questions of how often a system should be operated and which cell voltage should be applied must be answered. As existing techno-economic models feature gaps, which means that these questions cannot be adequately answered, a modified model is introduced here. In this model, different technical parameters are implemented and correlated to each other in order to simulate the lowest possible levelized cost of hydrogen and extract the required designs and strategies from this. In each case investigated, the recommended cost-based cell voltage that should be applied to the system is surprisingly low compared to the assumptions made in previous publications. Depending on the case, the cell voltage is in a range between 1.6 V and 1.8 V, with an annual operation of 2000–8000 h. The wide range of results clearly indicate how individual the design and operation must be, but with efficiency gains of several percent, the effect of optimization will be indispensable in the future.

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Introduction

Hydrogen is one of the most important chemical compounds, with an annual production of about 70 million tonnes [1]. It plays a fundamental role in the production of commodity chemicals like ammonia today, and is often referred to as a green energy carrier that could potentially reduce CO_2 emission in various industrial sections in the future. In steel production, which contributes about 8% to global CO_2 emissions [2], it can replace coke as the reducing agent [3]. Additional applications “were identified where hydrogen can become a cost-competitive low-carbon solution before 2030,” which “account for up to 15% of global energy consumption.” [4] Today, however, around 830 million tonnes of CO_2 are emitted annually from hydrogen production, which account for about 2.4% of global emissions [1]. Less than 4% of hydrogen generation utilizes CO_2 emission-free technologies [5]. Therefore, it is necessary to improve clean technologies such as polymer electrolyte membrane (PEM) water electrolyzers [6]. The urgency of bringing such technologies into application requires an interdisciplinary view of a broad level of technology readiness. Although basic research is still required and carried out on the one hand, market launches are being prepared on the other [7]. The challenge here is that technical developments are often not sufficiently linked to economic perspectives and, conversely, that economic considerations do not include a detailed perspectives on technical conditions. However, this is important, as the technology requires both cost reductions and increased efficiency [8]. Finally, efficiency and costs are the two sides of the coin that make up the levelized cost of hydrogen.

Just as there have been many different studies on the levelized cost of hydrogen, their results vary by region and assumption. For the United States, a current cost of 5 $/kg, excluding taxes, is stated by the Department of Energy (DOE) for centrally-produced hydrogen [9]. The International Energy Agency (IEA) assumes production costs of 3–5 $/kg in China depending on whether grid or renewable electricity is used [1]. For Europe, WaterstofNet states costs of 5.20 €/kg without fees in Flanders [10] and according to the National Organisation Hydrogen and Fuel Technology (NOW), the costs total about 10 €/kg, including taxes and fees, in Germany [11]. In the global view, the Hydrogen Council assumes 6 $/kg hydrogen [4]. Aside from country-specific studies, cost analysis with respect to specific technologies exist. By using solar energy, the levelized cost of green hydrogen is 5.50 $/kg [12]. In the case of wind energy, a price of 4.40 €/kg can be achieved, including storage and transport [13]. All of these studies exhibit an impressive level of detail in terms of their technical and economic contexts. They are based on even more detailed studies regarding the precise investment costs of PEM-electrolyzers [14] and their overall system efficiency [15], as well as power supply distributions that result from natural fluctuations associated with renewable energy [16] or fluctuations in the price of grid electricity [5]. However, the principle of calculating the levelized cost of hydrogen is the same in all cases with more or less small differences in detail and contains investment, operation, maintenance, and electricity costs. Here, various studies clearly demonstrate that the electricity price dominates the price of hydrogen [17]; the reduction in investment and maintenance costs only has a minor effect [18]. However, together with electricity costs and system efficiency, these parameters are frequently used for sensitivity analysis. From this point of view, the economic aspects are effectively taken into account, but from the technical point of view, important parameters such as current density or cell voltage are not sufficiently considered. It can be argued that a variation in the efficiency means consideration of the technical side, but its implementation is incomplete in detail. The reason for this is the modeling approach taken in most studies. The technical side starts with the question of how much power the modeled electrolysis system should have, how this power can be technically achieved, and how much it costs. Although values are defined in some cases, a more professional and popular approach is to assume specific costs for stack and system components expressed in prices per stack area or in terms of electrical power. In fact, the values can be converted into each other if the power density is known, which results from the system's design point. This is defined by the current density and cell voltage, and also determines the efficiency of the plant, which ultimately allows the cost of hydrogen to be calculated [12].

Such approaches constitute essentially a good compromise between looking at the fundamental technical concept of electrolyzers and simplifying the model, as the focus is on calculating the levelized cost of hydrogen. The technical side is therefore handled as a requirement to achieve the actual goal of the study. However, the definition of the design point is apparently arbitrary, even if the selection is based on values according to state-of-the-art systems. Therefore, studies are an individual consideration of an operating point. This is not a bad thing though, because a system requires boundary conditions in order to simplify its complexity. Nevertheless, as the levelized cost of hydrogen should be calculated precisely, the boundaries are set on the technical side, which causes an imbalance between economic and technical considerations.

In this study, a path out of this imbalance is demonstrated. Although the operating point of the electrolyzer is normally selected in order to precisely calculate the cost of hydrogen, herein a recommended operating point is calculated under the boundary condition of lowest levelized cost of hydrogen for a defined economic scenario. Therefore, the tradeoffs between current density and cell voltage as well as operating time and electricity price are analyzed. The first tradeoff stems from the system characteristics that the plant efficiency, which is mainly influenced by the cell voltage, decreases with increasing hydrogen production rate, which is proportional to current density. A high production rate means that the contribution of the investment costs to the cost of hydrogen is lower as more hydrogen is produced over the lifetime of the system. In contrast, the operating cost increases because more electricity is required in order to produce the same amount of hydrogen. The second tradeoff stems from a frequently mentioned approach that hydrogen should be produced when the electricity price is low [19]. In this case, the operating costs are low, but same is the annual hydrogen production. Consequently, the contribution of the investment costs to the cost of hydrogen increases as also mentioned for the first tradeoff. In total, all parameters set up a more-dimensional tradeoff that requires an optimization.

This study aims to figure out the best tradeoff in order to help technically-oriented R&D-groups to refocus their work. In contrast to other studies, design and cost of the electrolyzer are not predetermined in order to optimize the operating strategy depending on the electricity price. Instead, the electric power available to operate the electrolyzer is fixed. The conversion of this power can be achieved through a variety of different electrolyzer setups with different operating points and specific investment cost. The arising question is which design point should be chosen if the electricity price distribution is known. To answer this question, it is assumed that design and operating point are the same, while the electrolyzer operates at this point or not at all. For this purpose, current and future electrolyzer systems that are connected to the electricity grid are used for evaluation. The cases originate from the DOE and relate to polarization curves and costs which are derived from their specification table. This work does not claim to offer a precise calculation of the cost of hydrogen, but shows how the technical side can be introduced into existing economic considerations.

Methodology

Model

Levelized cost of hydrogen

The levelized cost of hydrogen, the LCOH, is defined as the costs of electrolysis referring to the mass of hydrogen produced, $m_{H_2}^{p}$, over its lifespan. The total costs consist of investment, C_{inv}, the cost of capital, C_{cap}, and operating costs that are separated into fixed, C_{fix}, and variable costs, C_{var} [20]:

$$LCOH = \frac{C_{inv} + C_{cap} + C_{fix} + C_{var}}{m_{H_2}^{p}} \tag{1}$$

Based on this expression, the parameters can be specified in depth. In this work, the power-specific investment costs, γ_{inv}, are defined as the investment costs per plant input power, which includes the power demand of the balance-of-plant. They are annualized by the capital recovery factor, which depends on the effective interest rate, i, and the plant life, t_{plant}. The term that includes the interest rate is called the capital recovery factor and is the cost of capital. The relative fixed costs are expressed as relative values, μ_{fix}, and refer to the annualized specific investment costs. The variable costs are defined as being equal to the electricity costs, σ_{el}, while the water costs are neglected. With the efficiency of the plant, η_{plant}, and the annual full load operating time, t_{op}, the levelized cost of hydrogen is given by the following expression according to NOW [11]:

$$LCOH = \frac{\Delta H_{LHV}}{\eta_{plant}} \left\{ \left[\frac{i(1+i)^{t_{plant}}}{(1+i)^{t_{plant}} - 1} + \mu_{fix} \right] \cdot \frac{\gamma_{inv}}{t_{op}} + \sigma_{el} \right\} \tag{2}$$

In this equation, ΔH_{LHV}, depicts the lower heating value of hydrogen. This kind of presentation is interesting because it allows the discussion of some important technical relationships between parameters. These are outlined in the following sub-chapters.

Specific investment costs and plant efficiency

The specific investment costs, γ_{inv}, are expressed in units of currency per input power. These diminish if the total costs decrease or the input power increases. Here, a challenge arises, as the input power of electrolysis plants depends on the operating point. An increase in the stack voltage means an increase in the current and input power consumption. Therefore, it is possible to reduce the specific investment costs by choosing a high operating voltage. Of course, this is an incomplete perspective due to decreasing plant efficiency when increasing the voltage. For this reason, a more consistent parameter would be to refer the investment costs relative to the output power, as this would take into consideration both the input power and plant efficiency. Saba et al. made a collection of these values, which were published in the last 30 years [21]. Although most studies refer to the specific costs of the input power, the authors had to transform the values into specific investments cost referred to as the output power. Unfortunately, many studies did not contain information regarding plant efficiency, and consequently it must be assumed by Saba et al. which poses an important question: if neither the efficiency nor the operating point is known, what is the informational content of this study? The given values for specific investment costs based on the input power are not comparable because the technical boundary conditions are missing. According to the literature, the voltage range of PEM electrolyzers is between 1.8 and 2.2 V [22], which is equivalent to a voltage efficiency of between 57% and 68% relative to the lower heating value of hydrogen. Therefore, the specification massively affects the calculation of the levelized cost of hydrogen and, even if a value was given, the issue remains as to why it was chosen in that way. Ideally, the choice of the operating point should not be specified in advance but should itself be an optimized value resulting from the calculation of the lowest levelized cost of hydrogen achievable [23].

Qualitatively, the situation can be expressed as follows. A low voltage equates to a high efficiency and low current density, which itself means a low specific hydrogen production rate. A high voltage, meanwhile, is associated with a high production rate and low efficiency. Although a high production rate reduces the specific investment costs, the variable costs decrease when the production rate is low because less electricity is required due to a high plant efficiency. It becomes obvious that an optimization is reasonable in which the specific investment costs are directly related to the plant efficiency. Therefore, the specific investment costs are broken down into basic parameters. The supply of the balance-of-plant reduces the available input power, P_{in}, that can be used for the electrochemical process in the stack. Therefore, the power that is available for the stack, P_{stack}, is:

$$P_{stack} = P_{in} \cdot \eta_{BOP} \tag{3}$$

Here, η_{BOP} is the efficiency of the balance-of-plant. The input power of the stack can also be described by its operating point thus:

$$P_{stack} = A_{cell} \cdot j_{op} \cdot U_{op} \cdot n_{cell} = A_{stack} \cdot j_{op} \cdot U_{op} \tag{4}$$

In this equation, A_{cell} denotes the active cell area, j_{op} the operating current density, U_{op} the operating cell voltage, and n_{cell} the number of cells that the stack consists of. The number of cells and active area per single cell can be combined into the total active area of the stack, A_{stack}. The equation reveals that different setups are possible for converting the same input power. The question remains as to whether a system should be operated at high power density and low stack area or low power density and a large active area. For high manufacturing capacities, the cost of the stack is proportional to its total active area [14]. Then, the specific investment costs of the stack area, α_{stack}, are calculated in units of total stack costs per square meter of active stack area. In principle, this leads to the same specific investment costs independent on the active cell area. Although this is only an estimation, a more precise description cannot be considered adequately because a separate study would be necessary. As will be shown later, the results presented are only marginally affected by this inaccuracy. Finally, the specific investment costs of the stack, γ_{stack}, result from the linking of eqs. (3) and (4):

$$\gamma_{stack} = \frac{C_{stack}}{P_{in}} = \frac{\eta_{BOP}}{j_{op} \cdot U_{op}} \cdot \frac{C_{stack}}{A_{stack}} = \frac{\eta_{BOP}}{j_{op} \cdot U_{op}} \cdot \alpha_{stack} \tag{5}$$

This equation's denominator contains the operating mode of the electrolyzer. The correlation of the current density and cell voltage results from the polarization curve and is not explicitly stated in this equation for reasons of clarity, but should be noted at this point. The product of these parameters is the power density of the stack. Adding specific investment costs of the balance-of-plant, γ_{BOP}, to those of the stack and taking into account the installation cost relative to the investment costs, $r_{install}$, the specific investment costs of the plant result (see eq. (2)):

$$\gamma_{inv} = \left(\frac{\eta_{BOP}}{j_{op} \cdot U_{op}} \cdot \alpha_{stack} + \gamma_{BOP} \right) \cdot (1 + r_{install}) \tag{6}$$

The specific cost of the balance-of-plant are further specified in the next sub-chapter.

The efficiency of an electrolysis plant is affected by several factors. The most dominant contribution to the overall efficiency is the voltage efficiency, η_{volt} [24]. This is determined by the voltage equivalence of the lower heating value of hydrogen referred to as the applied cell voltage. Therefore, the voltage efficiency decreases with an increasing hydrogen production rate, as previously stated. In addition, the efficiency of the plant is also affected by the loss of produced gas and is called the Faraday efficiency, $\eta_{Faraday}$. Typically, this term decreases with increasing hydrogen production rates. The product of Faraday and voltage efficiency is the stack efficiency of the plant, η_{stack}. The overall plant efficiency is the product of the efficiencies of the stack and the balance-of-plant. The balance-of-plant comprises a power converter, heating, gas drying, and compression. These contributions are strongly correlated because the Faraday and gas compressor efficiency are linked through the operating stack pressure, as well as the heat demand and stack efficiency through the operating stack temperature. The loss of product gas is primarily based on gas permeation across the membrane. It is relevant to note that this permeation process is dominated by the diffusion of hydrogen through the polymer membrane, which leads to a proportional increase in stack operating pressure [25]. However, the operating pressure should be as high as possible in order to reduce the use of external gas compressors. From a technical perspective, pressurized electrolysis is always an energetic tradeoff between product gas loss and required compression work. Several studies have investigated this topic in the past, whereas different optimal pressure levels have been determined depending on the model and operating conditions that were used. The pressure range extends from low [26], [[,27] to medium [15,28], to high pressure [29]. In this study, all of the above aspects were considered by using a model published by the authorship to simulate the efficiency of a system [24]. This approach determined the efficiency of the electrolysis plant, η_{plant}, in eq. (2) as a function of the stack operation and properties. It is $\eta_{plant} = \eta_{stack}\eta_{BOP}$ with $\eta_{stack} = \eta_{stack}(U_{op}, j_{op}, T, p, \ldots)$ and $\eta_{BOP} = \eta_{BOP}(U_{op}, j_{op}, T, p, \ldots)$ to specify that the efficiencies are complex functions that must be modeled. Finally, the equation to calculate the levelized cost of hydrogen contains the relationship between efficiency, stack operation, and specific investment costs. Although the link between these parameters has been established, the fixed and variable costs should also be adjusted next.

Fixed operating costs and replacement interval of the stack

The fixed operating costs were introduced as a relative value referring to the specific investment costs in Eq. (2). They include the costs of maintenance of the electrolysis plant, the replacement of the stack, and others. Instead of referring the value to the overall specific investment costs, each part of the plant is assigned to one relative fixed cost value. Therefore, the separation of the specific investment costs into stack and balance-of-plant factors in the previous sub-chapter can also be transferred to the fixed cost description.

Starting with the relative costs of the maintenance of the balance-of-plant, μ_{BOP}, it is assumed that this operational expenditure is dominated by maintenance of the compressor and that the operating time does not affect it [30]. As it refers to the investment costs of the balance-of-plant, it is calculated as follows:

$$\mu_{BOP} = \mu_{comp} \cdot \frac{C_{comp}}{C_{BOP}} \tag{7}$$

In Eq. (7), μ_{comp} symbolizes the relative maintenance costs of the external gas compressors relative to its investment cost, C_{comp}, whereas C_{BOP} is the investment cost of the balance-of-plant. The compressor size and its corresponding costs depend on the required specific compression work, w_{comp}. This compression work depends on the pressure level before and after the compression and the number of compressor stages that are used [31]. The actual power demand of the compressor is higher than this thermodynamic value due to the mechanical efficiency of the compressor, η_{comp}. The cost also depends on the hydrogen flow rate, which can essentially be expressed by the operating current, I_{op}, of the electrolyzer.

By implementing the investment cost per compressor power, $\gamma_{comp,inv}$, the nominal cost of the compressor, C_{comp}, results:

$$C_{comp} = \frac{w_{comp}}{\eta_{comp}} \cdot \frac{I_{op}}{2F} \cdot \gamma_{comp,inv} \tag{8}$$

The investment costs of the balance-of-plant are calculated accordingly:

$$C_{BOP} = \frac{I_{op} \cdot U_{op}}{\eta_{BOP}} \cdot \gamma_{BOP} \tag{9}$$

Here, it must be noted that the compressor cost is already included in the balance-of-plant costs. It is advisable to separate these to consider the significant impact of the compressor on the overall cost, which strongly depends on the ratio of the stack and storage pressure [32]. This approach enables a higher flexibility when rating different systems:

$$\gamma_{BOP} = \underbrace{\frac{C_{comp} \cdot \eta_{BOP}}{I_{op} \cdot U_{op}}}_{=\gamma_{comp}} + \gamma_{BOP,rest} \tag{10}$$

In this equation, γ_{comp} is the investment cost of the compressor referring to the plant power and $\gamma_{BOP,rest}$ describes the power-specific investment costs of the balance-of-plant, excluding the compressors (see also supplementary information A).

In addition to the balance-of-plant, the relative maintenance costs of the stack, μ_{stack}, must be estimated. This value is assumed to depend on the operating time and assumes that the replacement interval of the stack, $t_{replace}$, is shorter than the lifespan of the rest of the plant. Therefore, a replacement interval exists and its inverse is the annual amount of relative replacement costs. In addition to that, the residual value of the replaced stack must be considered [9], and therefore a value, ρ_{stack}, is introduced that describes the net replacement costs relative to the original investment costs:

$$\mu_{stack} = \frac{t_{op}}{t_{replace}} \cdot \rho_{stack} \tag{11}$$

The replacement interval of the stack is not equal to the lifespan, which is indeterminate as typically no sudden death but rather a continuous degradation in stack performance tends to occur. There are different reasons for this degradation, and it is still under discussion which effect most strongly affects the development of decreasing stack performance, as the setup appears to play an important role [33]. In principle, the degradation of the stack shows an increase in cell voltage while keeping the current density constant. Consequently, the power demand increases over time, which is why the power electronics must be oversized, and it also means that the voltage efficiency decreases continuously (see also Supplementary Information B) [34].

Studies that investigate the degradation rate often show a family of polarization curves [35]. These curves indicate that the degradation rate measured in μV/h is not constant over the entire polarization curve. If degradation rates were a constant, low cell voltages would be affected much more strongly relative to high cell voltages. This is in contrast to investigations that show that the higher the cell voltage or current density, the higher the degradation [36]. Therefore, a degradation rate, $\dot{U}_{deg,ref}$, belongs to a given cell voltage, which is depicted as a reference cell voltage, $U_{deg,ref}$. In this work, it is assumed that the ratio of these values is a constant, meaning that the relative degradation rate, r_{deg}, is constant for all cell voltages. In addition to this, it is also assumed that the degradation rate does not change over time. It is clear that these assumptions are very idealistic, but when accepting them, it is:

$$r_{deg} = \frac{\dot{U}_{deg,ref}}{U_{deg,ref}} = \frac{\dot{U}_{deg,op,0}}{U_{op,0}} \Leftrightarrow \dot{U}_{deg,op,0} = U_{op,0} \cdot r_{deg} \tag{12}$$

By means of this, the average operating cell voltage is given by Eq. (13):

$$\overline{U}_{op} = U_{op,0} + \dot{U}_{deg,op,0} \cdot \frac{t_{replace}}{2} \tag{13}$$

As previously noted, this negatively affects the voltage efficiency of the electrolyzer, η_{Volt}. It is the ratio of the potential of the heating value of hydrogen, $U_{\Delta H}$, and the applied cell voltage:

$$\overline{\eta}_{Volt} = \frac{U_{\Delta H}}{\overline{U}_{op}} = \frac{U_{\Delta H}}{U_{op,0} + U_{op,0} \cdot r_{deg} \cdot \frac{t_{replace}}{2}} = \underbrace{\frac{U_{\Delta H}}{U_{op,0}}}_{=\eta_{Volt,0}} \cdot \frac{1}{1 + \underbrace{r_{deg} \cdot \frac{t_{replace}}{2}}_{=\lambda_{deg}}} \tag{14}$$

Eq. (14) indicates that the average voltage efficiency is the product of the initial voltage efficiency at the beginning of the stack operating interval, $\eta_{Volt,0}$, and a factor that contains a degradation function, $\lambda_{deg} = \lambda_{deg}(t_{replace}, \ldots)$. This function can be modified in terms of different descriptions of the degradation.

Similar to Eq. (13), the costs of the power electronics assigned to the power specific balance-of-plant costs excluding compressors, $\gamma_{BOP,rest}$, increase. Separating the rest into electrical, $\gamma_{BOP,el}$, and mechanical parts, $\gamma_{BOP,mech}$, and considering the increasing power consumption, the cost of the balance-of-plant is described by Eq. (15) thus:

$$\gamma_{BOP,rest} = \gamma_{BOP,el} \cdot (1 + r_{deg} \cdot t_{replace}) + \gamma_{BOP,mech} \tag{15}$$

These equations show how the replacement interval of the stack contributes to the levelized cost of hydrogen. In the case of Eq. (11), it becomes clear that the annual operating time of the system is also an important parameter and its dependence will be discussed next.

Variable costs and operating time

As previously noted, only electricity costs are considered variable costs in this study. In addition, the assumption has been made that the analyzed electrolysis system is connected to the electricity grid, which is typical in applications today. Therefore, questions arise regarding how part load operating can be considered and how often it would be viable to transform electricity into hydrogen. To answer the first question, we assume that the electrolyzer is operated with a constant current density, since load change leads to durability issues [37]. However, it is worth mentioning that a recently published study indicates that the cost of hydrogen can be slightly reduced with dynamic operation [38].

To answer the second question of how often the electrolyzer should be operated, an approach published by Zauner et al. was used [39]. This was based on the spot market price used to evaluate an average electricity price as a function of operating under full load hours. The authors showed that the cost of hydrogen is minimal when the annual operating time is higher than 5000 h (see also Supplementary Information C). In this study, the same approach is used to combine electricity costs and operating time, which is briefly described as follows: it considers a specific electricity price distribution and converts it into the average electricity price as a function of the annual availability. The average electricity price results from considering only the days of the lowest electricity prices over the course of the year until the desired operating time is reached. If past price data are used, it is a retrospective approach. It means that a forecast is only practicable if the market price is stable.

Simulation

Electricity price

The average electricity price as a function of the annual operating time, $\sigma_{el} = \widehat{\sigma_{el}}(t_{op})$, is determined according to the method described in sub-chapter 2.1.4. For this approach, the daily average price data from the spot market of the European Energy Exchange (EEX) are selected for 2019 (see Fig. 1a) [40]. These data are converted to answer the question of what the lowest possible average electricity price was that had to be paid in 2019 in order to draw electricity for a certain number of hours at the EEX. The resulting data are then fitted (see Fig. 1b). In addition to this price, there are grid fees and taxes. According to Eurostat, the total average electricity price for non-households in the EU-28 countries was 82.55 €/MWh in the first half of 2019 [41]. This makes a difference of $\Delta\sigma_{el} = 44.83$ €/MWh between the non-household consumer price and the average spot market price. Incorporating this difference, Eq. (16) results as an estimation of the electricity price as a function of the annual operating time. The parameter $\Delta\sigma_{el}$ can also be used to systematically adjust the electricity price function, as will also be performed later in this work. This means that an offset parameter is added, which does not affect the distribution properties. It is:

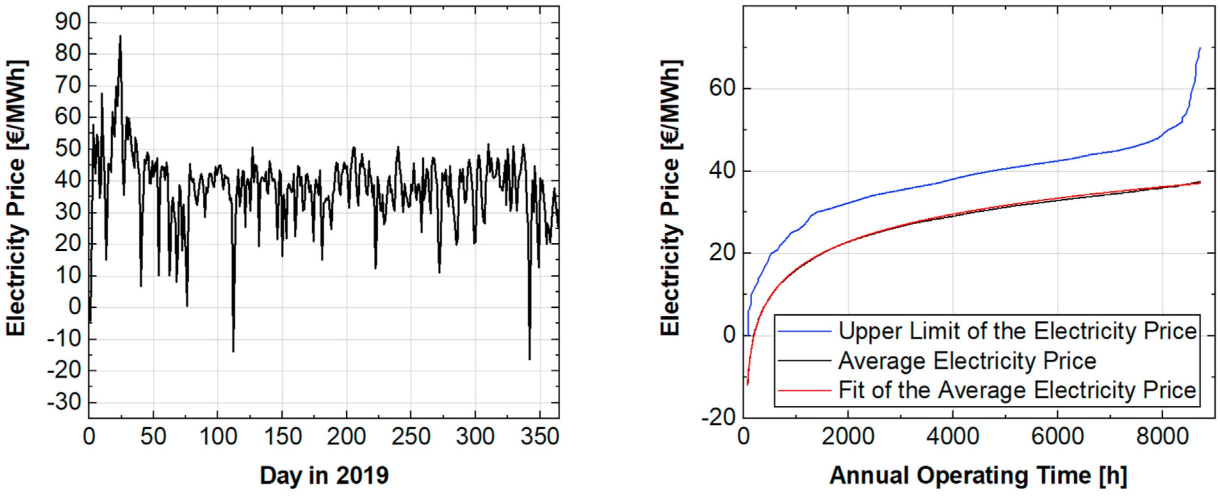
$$\widehat{\sigma_{el}}(t_{op}) = \left[9.51\cdot\ln(t_{op} - 21.28) - 49.28 + \Delta\sigma_{el}\right]\frac{€}{MWh} \qquad (16)$$

Basic electrolyzer data

The presented model is applied to four case studies that were reported in 2020 by the DOE [9]. They differed between current (2019) and future (2035) cases, whereas for both so-called distributed and central cases were introduced. The assumed electrolyzers deliver 1500 kg (distributed) and 50,000 kg H_2 per day (central), and have different parameter values. It is important to note that the reported costs are not the current ones of commercially-available electrolyzers but a mass production projection at a production capacity of 700 MW/year. Table 1 summarizes the values from the above report and the available supplementary information.

Data conversion

The values listed in Table 1 must be converted to make them applicable in the presented model. Therefore, the specific costs relate to the overall plant instead of the stack, whereby costs are converted from \$/kW into €/kW. The currency exchange rate is assumed to be 1.14 \$/€. In addition to that, the capacity factor and replacement interval are multiplied in order to obtain the replacement interval in units of operating hours. The area-specific costs result from Eq. (5) and the relative degradation rate from Eq. (12). The efficiency of the balance-of-plant is the quotient of the plant and electrical efficiency, both of which are listed in Table 1.



(a) (b)

Fig. 1 – (a) Spot market price at the EEX in 2019; and (b) the upper limit of the electricity price that occurs when the system is operated for the given annual operating time. Deviating from the upper limit, the average electricity price as well as its fit are illustrated and describe the average price that must be paid when the system should be operated for the given annual operating time. Latter is important for the simulation.

Table 1 – Original parameters from the DOE for central (50,000 kg H_2/day) and distributed (1500 kg H_2/day) production of hydrogen in 2019 (current) and 2035 (future) [9]. In the current cases, the specific costs refer to costs that would have been incurred if mass production had been achieved in 2019. They differ from the actual cost of the electrolyzers, but were used to maintain consistency within the data set.

Parameter	Current Distributed	Future Distributed	Current Central	Future Central	Unit
$j_{op,DOE}$	2.0	3.0	2.0	3.0	A/cm^2
$U_{op,DOE}$	1.9	1.8	1.9	1.8	V
t_{plant}	20	20	40	40	y
γ_{stack}	342	143	342	143	$\$/kW_{Stack}$
$\gamma_{BOP,el}$	121	97	82	68	$\$/kW_{Stack}$
$\gamma_{BOP,mech}$	136	140	36	23	$\$/kW_{Stack}$
ρ_{stack}	15	15	15	15	%
$r_{install}$	12	10	12	10	%
η_{plant}	59.7	64.8	60.1	65.0	%
$\eta_{electrical}$	66.1	69.8	66.1	69.8	%
T [42]	353	353	353	353	K
Capacity factor	97	97	97	97	%
$t_{replace}$	7	10	7	10	y
$\dot{U}_{deg,ref}$	1.5	1	1.5	1	mV/(1000 h)
Electrolyzer outlet pressure [43]	2	4.8	2	4.8	MPa
Effective interest rate, I [43]	10	10	10	10	%

Table 2 – Values that can be obtained by means of the information in Table 1 and/or given equations.

Parameter	Current Distributed	Future Distributed	Current Central	Future Central	Unit
γ_{stack}	271	116	273	11,760,	€/kW_{plant}
$\gamma_{BOP,el}$	96	79	65	56	€/kW_{plant}
$\gamma_{BOP,mech}$	107	114	29	19	€/kW_{plant}
t_{op}	8500	8500	8500	8500	h
$t_{replace}$	59,480	84.972	59,480	84,972	h
α_{stack}	12,555	7347	12,555	7347	€/m^2
r_{deg}	$7.89 \cdot 10^{-7}$	$5.56 \cdot 10^{-7}$	$7.89 \cdot 10^{-7}$	$5.56 \cdot 10^{-7}$	1/h
$\eta_{BOP,rest,DOE}$	90.3	92.8	90.9	93.1	%

Adjustments

Unfortunately, information on hydrogen permeability in future polymer membranes is not presented in the DOE report. If the current permeability of hydrogen in PFSA is assumed, an unacceptably low Faraday efficiency would result, especially at low current densities. Therefore, it was decided that electrochemical compression be limited to 2 MPa, and the compression from 2 MPa to 4.8 MPa be performed by mechanical gas compressors in future. According to the DOE, the impact on the cost of hydrogen is less than 10 c/kg. Drawing on literature values and simulations, important parameter values are determined and listed in Table 3.

Construction of curves

Although Table 1 only contains one current density and cell voltage per case study, it is necessary to construct full polarization curves in order to obtain all available operating points. Therefore, a model that has already been published is used to fit the initial polarization curves (Fig. 2a) (see Supplementary Information D for more details) [46]. Contrary to this, the plant efficiencies (Fig. 2b) are not fitted, but are a result from the applied model, which was fed with information from Tables 1–3. The result indicates a precise match between the DOE-given plant efficiency and the simulation result in the current case at the DOE operating point. In future cases, a slight but acceptably small deviation occurs (see Supplementary Information E for an explanation). The cell voltage increase of the stacks over lifetime is in a range of 70–120 mV in both cases (see Supplementary Information F for full polarization curves).

The simulation indicates that the maximum plant efficiencies are at about 70% in both cases and decrease to lower values due to the decrease in voltage efficiency. It turns out that the efficiency at the current reference point of 2 A/cm^2 is assumed to increase in future by approximately 6.4% absolutely or 10% relatively compared to the current case (see Supplementary Information G for more details).

Protocol

As most of the parameters have been previously fixed, only a few open parameters remain variable. These are the operating current density and the annual operating time. In the simulation, the specified operating current density is between 0.02 A/cm^2 and 10 A/cm^2 and the step size is 0.01 A/cm^2. The cell voltage is limited to 2.5 V. At any point, the annual operating time is varied between 500 h and 8750 h in steps of 50 h in order to obtain the annual operating time at which the levelized cost of hydrogen is reduced to a local minimum. Instead of using the current density, the results are displayed as functions of the applied cell voltage, which enables a better comparison of different systems. The local minima of the operating time variation at different operating points finally

Table 3 – Assumption made regarding the compression of hydrogen. (*) indicates that a value was determined via simulation.

Parameter	Current Distributed	Future Distributed	Current Central	Future Central	Unit
Stack outlet pressure [43]	2	2	2	2	MPa
Gas compressor outlet pressure [43]	2	4.8	2	4.8	MPa
Compressor efficiency, η_{comp}	–	75	–	75	%
w_{comp} *	–	2565	–	2565	J/mol
$\gamma_{comp,inv}$ [44]	–	2100	–	2100	€/kW_{fluid}
μ_{comp} [45]	–	5	–	5	%
$\gamma_{comp,ref}$ (from eq. (10))	–	19	–	19	€/kW_{plant}

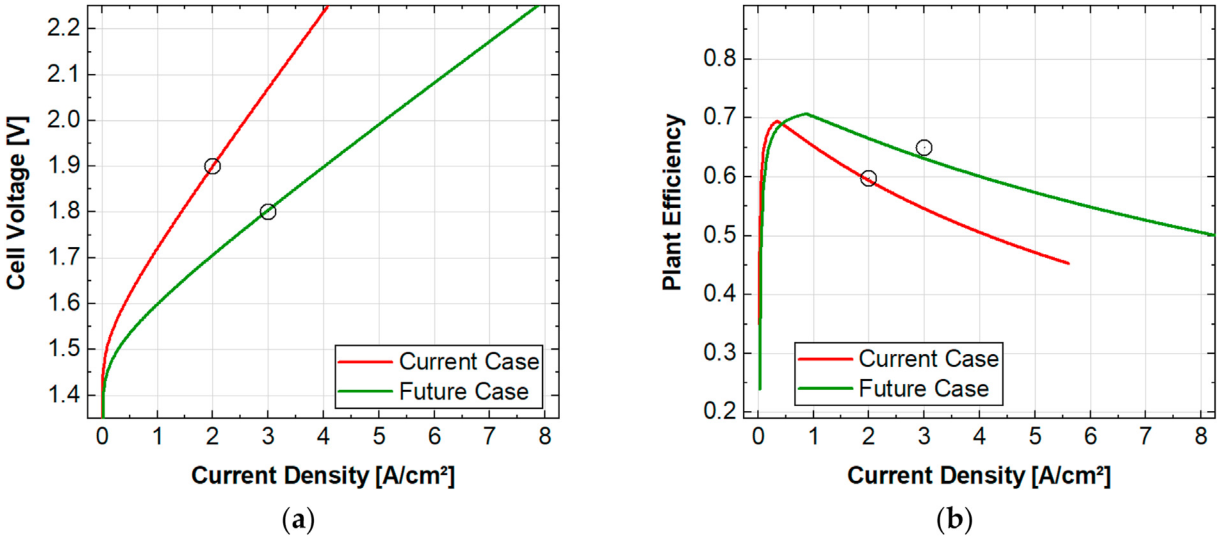


Fig. 2 – (a) Simulation of the initial polarization curve; (b) plant efficiency as a function of the current density for the current and future central case. The distributed cases are not illustrated in order to maintain visibility. The DOE values are circled.

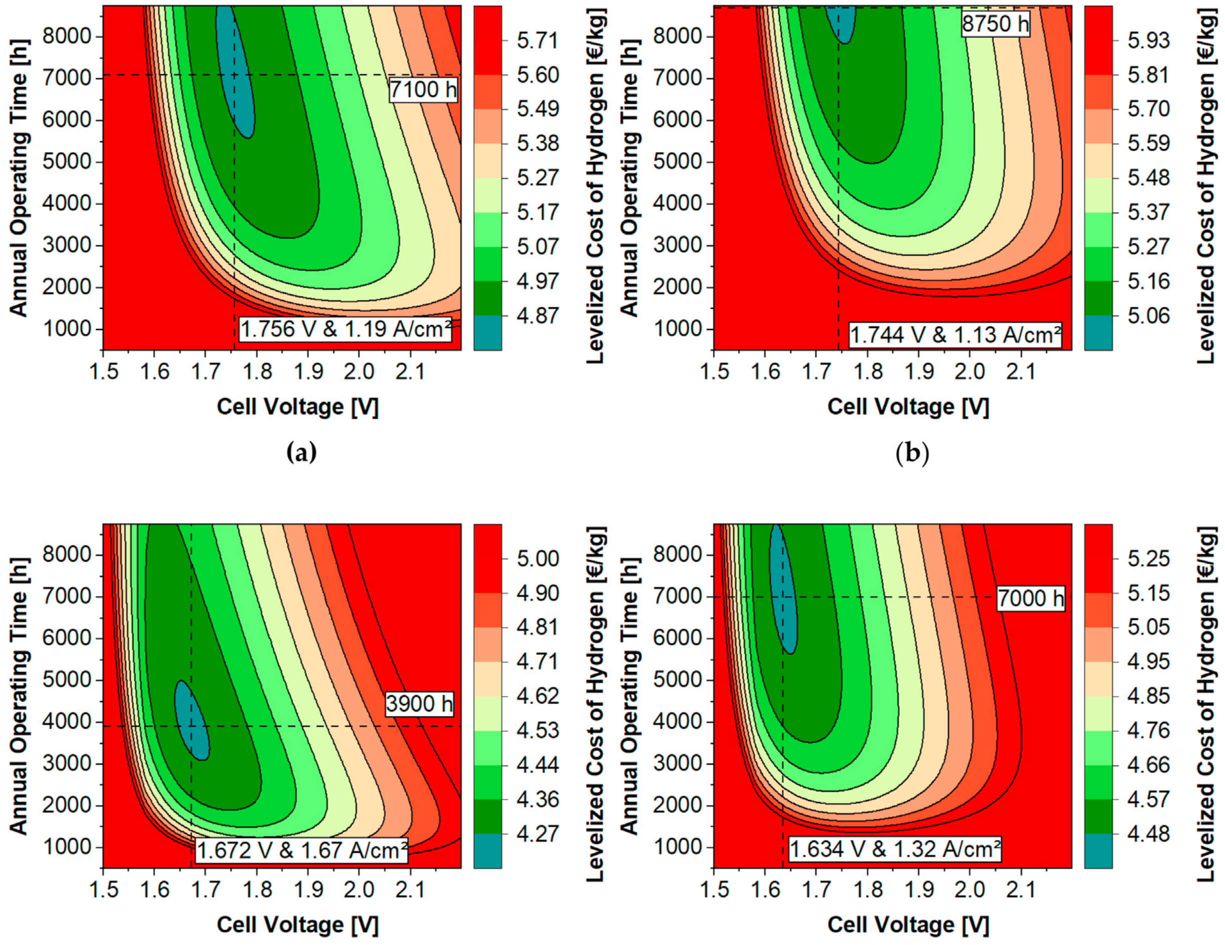
allows the determination of the global minimum of the levelized cost of hydrogen.

Results and discussion

Minimum cost strategies

The central production of hydrogen is performed by PEM-electrolyzers in the 100 MW class. The minimum costs are achieved when the cell voltage is 1.76 V and the current density is 1.19 A/cm^2 at a plant efficiency of 64.0%. The system should be operated for 7100 h per year (see Fig. 3a). This means that the electrolyzer can produce cheaper hydrogen when it is designed for a lower area-specific power consumption, which significantly increases the plant efficiency. Additionally, the annual operating time should be shortened accordingly. In this strategy, the simulation indicates that the minimum levelized cost of hydrogen would be 4.86 €/kg. In comparison, at the DOE value of 1.9 V and 8500 h per year, the levelized cost of hydrogen is 5.04 €/kg, whereas the reported value is 4.83 $/kg (4.24 €/kg). This actually appears to be quite different from the simulation result; however, the electricity price for non-households in the EU was used in this study instead of the value reported for the US. If this is taken into account, the difference is 0.15 €/kg, which is suspected to be a function of the different model descriptions. When assuming that the deviation is systematic, the cost difference between the operating points proposed by the DOE and the simulation is 0.18 €/kg or 3.6%. If such a cost reduction can be achieved without changing the operating strategy, an increase in the plant efficiency from 59.3% to 61.5% would be necessary. Although this increase actually seems to be unimpressive, its massive impact shows when the increase is expressed as a voltage reduction at same current density. In this case, the cell voltage must decrease by 70 mV, which is ambitious to achieve without the use of thinner membranes. Similar considerations apply to the current distributed case, where PEM-electrolyzers are in the 1 MW class (see Fig. 3b). The cost of hydrogen is 5.05 €/kg, which is slightly above the central case but lower than the 5.21 €/kg at the operating point proposed by the DOE. Again, this indicates a significantly lower cell voltage of 1.74 V at 1.13 A/cm^2, what is similar to the previous case, whereas the plant efficiency is also 64.0%. In contrast to this, the simulation recommends a full-time operation that approximately corresponds to the reference.

The future cases indicate a significant shift in both cell voltage and annual operating time to lower values compared to the previous data. For centrally-produced hydrogen, the cell voltage deceases to 1.67 V and the operating time to 3900 h. The corresponding current density increases by about 50% due to the thinner membrane and lower activation



(a) (b)

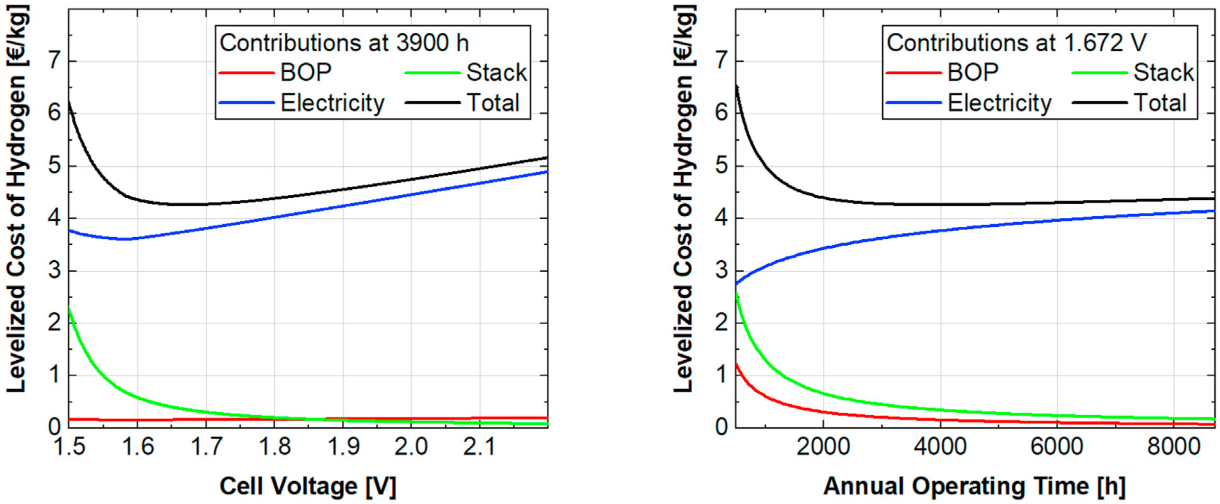
(c) (d)

Fig. 3 – Dependency of the levelized cost of hydrogen on the initially applied operating cell voltage and the annual operating time in the current (a) central and (b) distributed cases as well as in the future (c) central and (d) distributed cases. Each contour line represents an increase in the cost of hydrogen of 2% compared to the previous inner line. Operating points in the innermost field are within 0.01 €/kWh. The colors relate individually to the respective minimum price of the illustrated case. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

overpotential. The cost of hydrogen is 4.26 €/kg when applying the optimized operating strategy, which is 0.35 €/kg below the value that results from the DOE operating strategy. When converting this value into the plant efficiency, as has been done before, this improvement corresponds to a massive increase, from 63.2% to 68.2%. It indicates that an optimized operating strategy becomes even more important in future, as it is very difficult to achieve comparable efficiency gains via technical development. The results can be transferred to the distributed case. Here, the effect is even stronger, as the minimum cost occurs at a cell voltage of only 1.63 V at 1.32 A/cm^2. The annual operating time is 7100 h, and the cost of hydrogen is 4.48 €/kg.

With respect to the results that are illustrated in Fig. 3a–d, the question arises as to why this strong deviation from the reported operating strategy occurs and what the reason for it is. Eq. (2) with consideration of eqs. (6) and (16) helps to answer these questions, as the principle is visible in the structure of these equations. First, the position of the minimum cost in relation to the cell voltage and fixed operating time is discussed (see Fig. 4a). When applying a low cell voltage, a low power density results. However, if the total input power must be the same, it is obvious that the stack area will need to be larger. This means that the investment costs increase with decreasing cell voltage, as can be seen in Eq. (6). Consequently, it seems preferable to apply a high voltage. However, a high cell voltage means a low efficiency, which not only affects the contribution of the investment cost with the cost of hydrogen but also the conversion loss of electricity. This means that, according to Eq. (2) and Fig. 2b, the conversion of expensive electricity is less efficient. From this perspective, the voltage should be as low as possible in order to minimize efficiency losses. The balance of low investment costs and high conversion efficiency of electricity to hydrogen makes up the minimum.

A similar principle of balance applies to the annual operating time at a constant cell voltage (see Fig. 4b). At low operating times, electricity costs are cheapest, as is shown in Fig. 1b,



(a) (b)

Fig. 4 – (a) Hydrogen cost breakdown as a function of initial cell voltage at 3900 operating hours per year; and (b) as a function of the annual operating hours at a cell voltage of 1.672 V. The values correspond to the future central case. The values given for the balance-of-plant (BOP) and stack include the maintenance costs.

but hydrogen production is also low. This in turn means that investment and maintenance costs increase relative to the amount of production. When the operating time is instead high, the hydrogen production rate is high and the hardware costs decrease relatively, but the electricity costs remain high. Electricity and investment costs balance out at 3900 h according to the simulation. Both principles apply to each of the four cases discussed and beyond (see Supplementary Information H for the mathematical explanation).

Electricity price variation

As stated in the introduction, the cost of electricity is of major importance, and this can be confirmed even if the hydrogen production cost is minimized by an improved strategy. According to Fig. 4a and b, electricity costs contribute to about 90% of the future central case. For this reason, the electricity price is varied to demonstrate how it affects the operating conditions in order to achieve the lowest cost of hydrogen. The adjustment is performed by means of the electricity price offset parameter, $\Delta\sigma_{el}$, which adjusts the average annual electricity price (see Eq. (16)).

According to the simulation, the cost of hydrogen changes by approximately 0.50 €/kg per 10 €/MWh cost of electricity; the effect is almost linear. Furthermore, the simulation indicates that the cell voltage shifts from 1.67 V to higher values when the electricity price decreases. This tendency accelerates towards low prices, as it is just over 2.2 V at a low electricity price of 23 €/MWh. In the opposite direction, higher costs seem to only have a minor effect (Fig. 5a). This development also means a strong increase in the current density when the electricity price is less than it was initially. Accordingly, current densities of up to 10 A/cm^2 are linked to an applied cell voltage of 2.2 V, which yields power densities of more than 20 W/cm^2 compared to less than 3 W/cm^2 resulting from simulation with today's electricity price (see Fig. 5b). However, the plant efficiency tends to values of 50% and less, which is surprisingly low. At the other end of the scale, at 118 €/MWh, the current density is slightly above 1 A/cm^2 and the power density is less than 2 W/cm^2, whereas the efficiency is just below 70%. In summary, these values are remarkable, as they offer a high variance, at which point PEM electrolyzers operate the cheapest, as it depends on the boundary conditions. Additionally, the simulation also indicates that the annual operating time, where the cost of hydrogen is lowest, decreases, when the electricity cost does (Fig. 5a). Therefore, cheaper electricity is not used to operate more often, but to operate at higher loads. The reason for this is that a low electricity price means that its contribution to the cost of hydrogen is lower and losses due to efficiency are less relevant. As a result, investment costs become more relevant, making it attractive to reduce it when the electricity price would decrease. This can be achieved through lower overall investment costs and higher production rates. The operation at higher loads meets both requirements, as the power density increases and the cost decreases, because less stack area is required. However, this raises the question of why the operating time decreases rather than increases, as this would also increase the production rate. The reason for this is evaluated in Fig. 4b. Accordingly, the slopes of the curve are of special interest, and the illustrated cost breakdown indicates a high curvature for the electricity curve at low operating times. This means that its contribution to the overall cost changes most significantly towards lower values at the front end of the curve. In the initial case, it is not attractive to use this effect, as the impact of the investment costs is high due to the low operating point (compare with Fig. 4a). The situation changes when the operating load increases due to lower electricity costs and the impact from investment costs on the hydrogen cost become small (Fig. 4a). In this case, it is advantageous to use the high curvature at the front end of the electricity cost curve in Fig. 4b to optimize the operation.

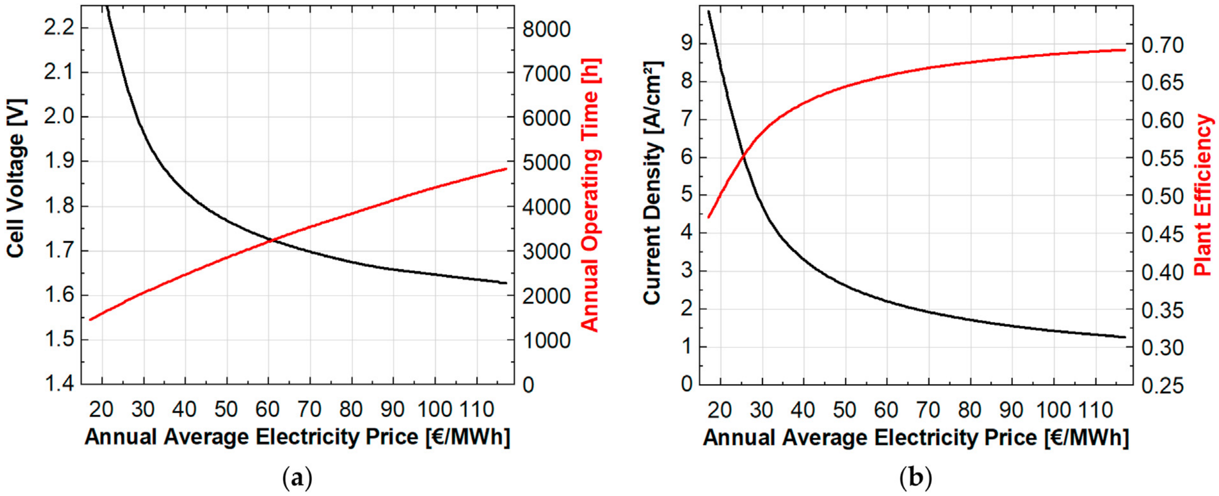


Fig. 5 – (a) Initial cell voltage and annual operating time where the cost of hydrogen are the lowest; and (b) the corresponding current density and plant efficiency as functions of annual average electricity price including taxes and fees. The values correspond to the future central case. The initial value is: 82.44 €/MWh.

Limitation of cost predictions

The above results and discussions are interesting, as they offer a new perspective when considering the operating strategy of PEM-electrolyzers. However, that is precisely why it is important to make some further remarks regarding the meaning of the results. The following sections are intended to specify the limitations that have not yet been taken into account. Here, limitation does not mean the limits of the mathematical description of the model, but those of a reliable quantification of the results.

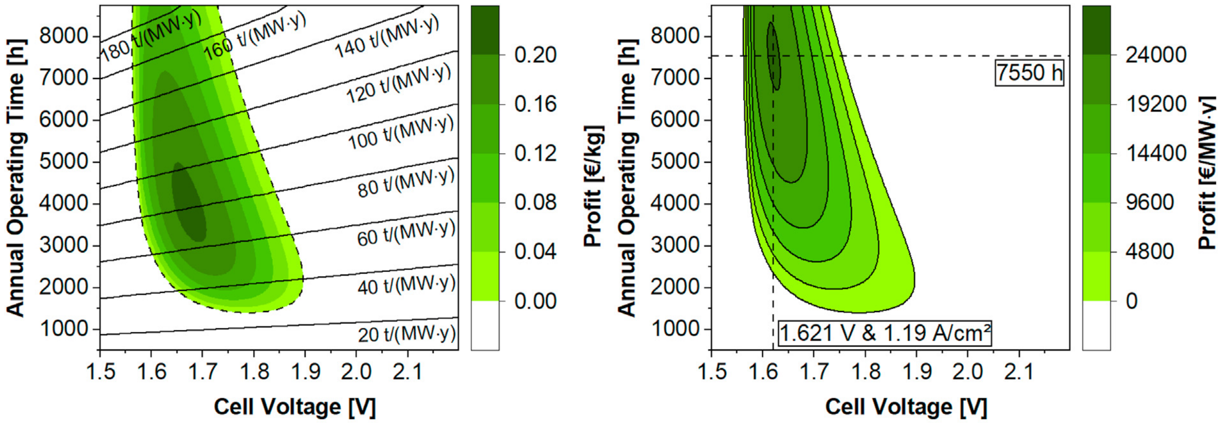
The results illustrated in Fig. 5a and b indicate that the operating strategy that leads to the lowest cost of hydrogen largely depends on the electricity price. This price is based on the cost of electricity, which itself depends on the electricity generation technique [47]. Moreover, geographical conditions, political factors, transport, and storage also affect the cost of electricity, thus impacting the production cost of hydrogen [48]. Although electricity from photovoltaics can be generated at 10 $/MWh in Saudi Arabia [49], 24 €/MWh in Spain, and 42 €/MWh in Finland [50], it is obvious that the operating strategy of PEM-electrolyzers must differ, as indicated in Fig. 5a. However, also within the same geographical region, differences appear due to the scale and application of the plant, which is obvious from a comparison between Fig. 3c and d. Additionally, the forecasting of production costs is not very reliable. With respect to the availability of iridium in particular, it seems that future demand for it due to PEM-electrolyzers will be significant compared to total iridium availability [51], which could affect its market price. Price changes can be sudden and sharp, as was seen in early 2021 [52]. Therefore, it is impossible to reliably predict the cost of the stack, as iridium accounts for one quarter of the cost [14]. Ultimately, this also weakens the predictability of operating strategies.

Although with regard to the model, simulation and discussion have thus far been based on costs, it is clear that when selling hydrogen, a powerful parameter affects the model. This is the profit, which is the difference between revenue and costs. It is possible that economic actors are likely to try to increase their profits. This is to be shown in the following for the example of the future central case. According to Fig. 3c, the levelized cost of hydrogen will be 4.26 €/kg. In perfect competition, this value is equal to the selling price [53]. In reality, the selling price differs due to imperfections, and consequently offers a margin [54]. When assuming an arbitrary margin of 5%, the selling price is 4.47 €/kg and the specific profit is 0.21 €/kg. However, the overall profit should be more interesting for the operator. Therefore, the mathematical product of the specific profit and annual production should be maximized (Fig. 6a). This means that it may [55]be more interesting to produce more hydrogen, despite specific cost increases due to deviation from the cost minimum. This could lead to different operating strategies than these that result in minimal costs (see Supplementary Information I).

In the example, the operating cell voltage that maximizes the overall profit would be 1.62 V, with an annual operation of 7550 h compared to the case without the margin, in which it is 1.67 V and 3900 h (Fig. 6b). The levelized cost of hydrogen is 4.32 €/kg, which is 0.06 €/kg above the minimum cost, but the annual production increases by 105%–164 t/(MW·y) (compare with Fig. 6a). This result seems surprising, as higher production is linked to higher cell voltages. This thinking is valid for values relative to the stack area, but the opposite applies when these are related to the input power, as is performed here. In this case, a lower voltage means a higher stack area with higher efficiency.

This example demonstrates how a profit margin affects the position of the preferred operating point. Consequently, different margins lead to different strategies. However, the future selling price is unknown, which makes prediction of the operating strategy difficult.

Another aspect is that a grid connection is assumed, which allows use of the method described in chapter 2.1.4, which describes the correlation between the average electricity price and availability in the European grid. This approach works



(a) (b)

Fig. 6 – Dependence of (a) the annual power specific production quantity on the profit per quantity; and (b) the power-specific annual profit on the initially applied operating cell voltage and the annual operating time in the future central case.

retrospectively, but in fact the development at the energy exchange must be predicted for the future. This is very difficult, however, because not only the average price but also the distribution is needed. In addition, electrolyzers are not necessarily connected to the grid [56]. If hydrogen is used directly in an industrial process, it is rational to produce it on site from renewable energy sources without a grid connection. On the one hand, the electricity supply fluctuates in this case, otherwise the electrolyzer is easier to design economically, as the electricity price can be calculated precisely if the average wind and solar energy profiles are known. Without assuming a grid connection, the presented model changes in terms of the description of electricity availability and price. The function $t_{op}(\sigma_{el})$ changes to a function $t_{op}(P_{el})$, where the electricity price becomes a constant and the available power a variable. Additionally, the model requires minor modifications to consider occurring part load operation.

In summary, it is apparent that there are many uncertainties that prevent the precise specification of recommended operating conditions with general applicability. This means that the production of PEM electrolyzers might not be subject to classical mass production but that production must be individualized with regard to special boundary conditions. Even similar applications in the same region could require different approaches. Additional aspects are the fact that profit margins as well as material and data availability affect the method's reliability.

Conclusions

Techno-economic studies of PEM-electrolyzers have often lacked technical descriptions of the systems. This makes it difficult to understand the reason for the given operating strategy, which consists of operating times and conditions. Whereas specific costs, power consumption, and efficiency are usually given as fixed values, these values are broken down into more fundamental parameters that describe the system in terms of its technical basis. This makes it possible to compare different operating strategies with respect to cell voltage and operating times and also to determine a preferred strategy for the cheapest possible hydrogen production. The simulation results indicate that the recommended strategy strongly depends on the framework conditions under which the operation takes place. Here, the investment cost structure and cost of electricity are relevant. This suggests that PEM-electrolyzers must be specified in terms of their operation mode and region. Although in regions with low electricity costs, the applied cell voltage can be higher than 2 V, simulations indicate that in regions with temperate climate zones, the cell voltage can be much lower, between 1.6 V and 1.8 V. Typical operating times are between 2000 h and 8000 h depending on the environmental conditions and technical setup. These values partially deviate from existing suggestions from other studies, where higher cell voltages are assumed. In comparison, some portion of the efficiency can be gained by optimizing the strategy and the cost of hydrogen can be reduced by up to 10%. However, it must be noted that the result is sensitive to future development regarding the electricity price and material availability, as well as to profit margins, which can have an impact on it. Nevertheless, the presented model is a powerful tool for examining the impact of hardware parameters and operating strategies on the costs of hydrogen and thus for offering a new dimension of optimization.

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Declaration of competing interest

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Nomenclature

Parameter	Symbol	Unit
Latin Letters		
Area	A	m2
Nominal cost	C	€
Specific cost	c	€/kg
Faraday constant	F	C/mol
Current	I	A
Effective interest rate	i	–
Current Density	j	A/m^2
Levelized cost of hydrogen	$LCOH$	€/kg
Molar mass	M	kg/mol
Produced mass	m^p	kg
Number	n	–
Power	P	W
Pressure	p	Pa
Annual profit	p_{H_2}	€/y
Relative degradation rate	r	1/h
Annual operating time	t_{op}	h
Plant life	t_{plant}	y
Replacement interval	$t_{replace}$	y
Temperature	T	K
Voltage	U	V
Heating value voltage	$U_{\Delta H}$	V
Specific work	w	J/mol
Greek Latters		
Area specific cost	α	€/m^2
Power specific cost	γ	€/kW
Lower heating value	ΔH_{LHV}	kWh/kg
Electricity cost offset	$\Delta\sigma_{el}$	€/MWh
Efficiency	η	–
Degradation function	λ	–
Relative cost	μ	–
Power specific annual profit	Π	€/(y·MW)
Selling price	π	€/kg
Residual relative cost	ρ	–
Energy specific cost	σ	€/kWh
Indices		
Initial value	0	
Balance-of-plant	*BOP*	
Capital	*cap*	
Cell	*cell*	
Compressor	*comp*	
Degradation	*deg*	
DOE-value	*DOE*	
Electric	*el*	
Fixed	*fix*	
Hydrogen	H_2	
Input	*in*	
Installation	*install*	
Investment	*inv*	
Mechanical	*mech*	
Operating	*op*	
Plant	*plant*	
Reference	*ref*	
Rest	*rest*	
Stack	*stack*	
Variable	*var*	
Voltage specific	*volt*	

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ijhydene.2022.11.288.

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