INTRODUCTION

Renewable energy sources (RES) will play a crucial role in the world’s future energy supply. However, prior to their large-scale integration into the existing infrastructure, certain challenges have to be addressed.

One of the main challenges is associated with the somewhat unpredictable and fluctuating nature of wind and solar energy sources, which can cause imbalances between the production and consumption of electrical energy in the grid, [1] and [2]. These inconsistencies in supply (due to the stochastic nature of renewable energy sources) and forecasting difficulties can be reduced to a certain extent by different (electrical) energy storage systems, [3] and [4]. Energy storage systems enable power-supply reliability and quality as well as system stability. Pumped-storage hydroelectric power plants maintain the balance between the supply and demand of electricity in the transmission network, while renewable energy sources (except large hydroelectric power plants) are usually connected to the distribution network, [4]. Energy storage based upon converting electrical energy to chemical (internal) energy of hydrogen and back is foreseen as one possible solution to this problem, [3] and [5].

Hydrogen is proposed as an energy-efficient pathway. Therefore, it is recognised as one of the energy carriers of the future, [5]. An electrolyser using electricity to generate hydrogen from water, a hydrogen storage tank and a fuel cell that recombines hydrogen with oxygen to generate electricity would be the main components of the so-called hydrogen infrastructure, which would balance the production and consumption of electrical energy in the distribution network (Fig. 1).

A demonstration laboratory for the simulation of advanced energy systems has been constructed at the hydrogen production site on the location of Šoštanj Thermal Power Plant (TEŠ). The primary aim of this project is to use technologically-advanced hydrogen production and logistics solutions in the design and demonstration of an advanced energy supply system that uses renewable energy sources and enables the optimisation of the performance of existing energy sources, [6]. This paper summarises the first stage of this project, whose main objective was to experimentally investigate the operational and energy characteristics of a commercial electrolyser and evaluate its role in advanced energy supply systems.

THEORETICAL PRINCIPLES OF WATER ELECTROLYSIS

The splitting of water into gaseous hydrogen and oxygen by the action of electricity can be expressed as, [7]:

\[ \text{H}_2\text{O}(l) + 2F \rightarrow \text{H}_2(g) + 1/2 \text{O}_2(g) \] (1)

where \( F \) is the Faraday constant representing the magnitude of electric charge per mole of electrons (96487 As/mol, [8]). Eq. (1) shows that water electrolysis is an extremely clean process, since no polluting by-products are formed. However, it should not be forgotten that a technology cannot be cleaner than the energy source used to power it, [7]. A definite advantage of electrochemical technology is its reversibility. The reverse of the above reaction (Eq. (1)) occurs in an \( \text{H}_2-\text{O}_2 \) fuel cell, [7];
The quantity of a substance altered at an electrode during electrolysis can be calculated according to Faraday’s (first) law of electrolysis, [9]:

$$m = \frac{dm_p}{dt} = \frac{M_p}{\nu_e \cdot F} \cdot I.$$  

The mass flow of the product ($m_p$) is directly proportional to the electric current ($I$) running through the electrolytic cell. Other terms in the expression correspond to the Faraday constant ($F$), stoichiometric coefficient (required number of moles of electrons for the formation of one mole of product; oxygen: $\nu_e = 4$; hydrogen: $\nu_e = 2$) and the molar mass of the product (oxygen: $M_p = 32.0$ g mol$^{-1}$; hydrogen: $M_p = 2.02$ g mol$^{-1}$, [8]).

An electrochemical system is in equilibrium when the following condition is met, [10]:

$$\Delta G = z \cdot F \cdot U_{rev},$$  

where $\Delta G$ represents the change in Gibbs energy, $z$ is the number of moles of electrons transferred in the reaction (for water electrolysis $z = 2$) and $U_{rev}$ is the reversible (cell) voltage, which is the minimum voltage needed to drive the water-splitting reaction (and also the maximum amount of useful work that can be derived from the system when driving the reaction in the opposite direction; Eq. (2)), [11]. At standard conditions ($T = 25 ^\circ C$ and $p = 101.3$ kPa, [8]), the reversible voltage is equal to 1.23 V ($\Delta G^0 = 237.0$ kJ mol$^{-1}$, [8]).

However, at this voltage (and conditions), the water-splitting reaction is endothermic; hence, for isothermal operation heat must be absorbed from the surrounding environment. The total amount of energy needed in water electrolysis is equivalent to the change in enthalpy ($\Delta H$), which differs from $\Delta G$ by the entropic term $T \cdot \Delta S$, [9]:

$$H_2(g) + 1/2 O_2(g) \rightarrow H_2O(l) + 2F.$$  

Fig. 1. The idea of hydrogen infrastructure integration into conventional power system
\[ \Delta G = \Delta H - T \cdot \Delta S. \] (5)

The entropic term \( T \cdot \Delta S \) represents thermal irreversibility, which for a reversible process is equal to the heat demand. The voltage corresponding to the total energy demand (\( \Delta H \)), the thermo-neutral (cell) voltage (\( U_{\text{tn}} \)) is given by the equation, [10]:

\[ \Delta H = z \cdot F \cdot U_{\text{tn}}. \] (6)

At standard conditions, the thermoneutral voltage is equal to 1.48 V (\( \Delta H^0 = 285.5 \) kJ mol\(^{-1} \), [8]). When the electrolytic cell is operated above \( U_{\text{tn}} \), the reaction becomes exothermic, and heat must be removed from the cell for isothermal operation (Fig. 2).

Due to the energy losses associated with reaction kinetics as well as charge transport through electrical leads and the electrolyte, all practical devices for water electrolysis operate in a voltage regime that is above the thermoneutral voltage. The operating or actual voltage (\( U_{\text{op}} \)) of an electrolytic cell can be expressed as, [7], [11]:

\[ U_{\text{op}} = U_{\text{rev}} + \sum \Delta U_{\text{el}} + \Delta U_{\text{ohm}} + \Delta U_t. \] (7)

The overvoltage (the voltage in excess of the \( U_{\text{rev}} \)) can be divided into three categories. The term \( \Sigma \Delta U_{\text{el}} \) represents the sum of the anodic and cathodic overpotentials. It arises as a result of several polarisation effects, including low activity of the electrodes in the electrolyte (known as ‘activation overpotential’). The electrode overpotential increases logarithmically (Fig. 3) with current density (\( j \)) as given by the Tafel relation, [11]:

\[ \Delta U_a = a + b \cdot \ln(j), \] (8)

where \( a \) and \( b \) are characteristic constants for the electrode system. The electrode overpotential can be minimised by selecting electrode materials with high electro-catalytic activity, and maximum real-to-apparent surface area as well as by operating the process at elevated temperatures (enhances the reaction rate) and pressures. In contrast, managing the process at higher current densities, which are associated with higher production rates (Eq. (3)), increases overvoltages (Eq. (8)).

The term \( \Delta U_{\text{ohm}} \) represents the energy dissipation related to ohmic drops in the electrolytic cell that occur mainly at the electrodes, electrical lead wires, metal-metal joints, and inside the electrolyte. Optimisation of the cell design, i.e. minimising the distance between the electrodes and reducing the electrolyte-resistance, lowers the ohmic overvoltage. An increase in operating temperature also helps to reduce \( \Delta U_{\text{ohm}} \) as it decreases the electrolyte resistance, [7] and [11]. This category of overvoltage changes according to Ohm’s law (Fig. 3):

\[ \Delta U_{\text{ohm}} = R \cdot I. \] (9)

The last term in Eq. (7) is \( \Delta U_t \), which expresses a phenomenological observation that the operating voltage applied to an electrolytic cell (at constant operating conditions; \( T, p \) and \( I \)) tends to increase with time as a consequence of performance degradation. The latter can be due to a loss of activity of electrode materials (surface wear) as well as an increase in ohmic drops (decrease in electrolyte concentration, loosening of electrical connections), [7]. Overvoltage minimisation is essential for the high efficiency operation of the electrolytic cells.
The contribution of mass concentration overvoltage is not shown in Fig. 3, since it would become apparent only at some point outside the voltages tested here.

Three process versions have been developed for water electrolysis: alkaline water electrolysis, membrane water electrolysis (also SPE water electrolysis or PEM water electrolysis) and high-temperature (steam) electrolysis, [9] and [14].

2 EXPERIMENTS

Experiments have been conducted on a commercial alkaline water electrolysis system, which was initially acquired (and designed) for the needs of the Šoštanj thermal power plant for hydrogen (cooling of electric generators) and oxygen (welding) production. The alkaline electrolyser went into operation in 2006. A programmable logic controller (PLC), a computer and a fuel cell were added to the existing water electrolysis system and hydrogen storage tank. The primary aim of the first stage of the project was to investigate operational and energy characteristics, as well as the limitations of the water electrolyser before connecting all the elements into a system of advanced energy supply.

The analysed commercial alkaline electrolyser presented in Fig. 4 operates at a pressure range up to 25 bar g and has a maximum production capacity of 15 Nm³ hydrogen per hour. The cell stack consists of a series of 90 interconnected, circular electrolysis cells (each cell has an electrode area of 0.2463 m²) arranged in a bipolar configuration. The basic parts are assembled and compressed in a unique and patented way, following the filter press system (zero-gap geometry). A 30% aqueous solution of potassium hydroxide (30% KOH) is used as the electrolyte. The produced hydrogen has a nominal purity of 99.998%, and the hydrogen storage tank has a volume of 20 m³, [12]. In comparison to a typical alkaline water electrolysis system, it has two distinctive features, [12]:

• demineralised water is fed to the system by gravity via an intermediate buffer (called ‘Fill Up Volume’ – Fig. 4), which is cyclically pressurised by the pressure of the system (valves are switched...
in a certain sequence), so there is no need for a demineralised water feed pump, and

• the special design of the cell stack enables the circulation of the electrolyte without the use of an electrolyte (circulation) pump.

An advantage of this configuration is that the system has lower electrical load, thereby contributing to a lower consumption of electricity. However, the operation of the Fill Up Volume buffer involves hydrogen losses, which have a negative impact on the overall energy efficiency of the system.

2.1 Experimental Procedure

Signals from twenty-one (21) sensors installed in the system provide data regarding temperature (cell stack, deoxo drier, cooling system, process room, electrical room), gauge pressure (hydrogen storage tank line, cooling system, gas separator), electrolyte level (oxygen and hydrogen gas separator), electrical voltage (cell stack, UPS), electric current (cell stack), electrical conductivity (feed water), volume percentage of hydrogen in oxygen (coalescing filter for oxygen) and electricity consumption (hydrogen production site). During the experiments, the ambient temperature and pressure, the temperature in hydrogen storage tank and the experimental time were also monitored.

During electrolyser operation, two parameters can be varied: electric current (in our case between 180 and 400 A, where 400 A corresponds to the nominal production of hydrogen; 15 Nm³ per hour), which is proportional to the production of hydrogen (Eq. (3)) and the position of the back pressure regulator (BPR in Fig. 4), which regulates gauge pressure in the system and was between 12 and 21 bar g in our case. The operating temperature of the process (electrolyte temperature) is predefined and locked by the manufacturer. During the experiments, it varied in a range from 59 and 65 °C.

Two main sets of measurements were concluded, and are presented in experimental matrix in Fig. 5. In the first set (experiments BPRi), the position of the BPR was fixed (BPR1, BPR2, BPR3 in Fig. 5) and the electric current was varied. At a single position of the BPR, four measurements at different electric currents were made. The measurement at an experimental point lasted 20 minutes and the entire experiment lasted 5 hours. The second set of measurements (experiments pCONST) were made with a variation of both the position of the BPR and the electric current. The position of the BPR was varied to achieve constant pressure in the system (p16, p18, p20 in Fig. 5).

According to the analysis of the BPRi experiments, the duration of a single measurement point was shortened to 5 or 10 minutes. The entire experiment lasted 4 hours and 40 minutes.

The data from 20 sensors were automatically recorded every 30 seconds. In the case of electricity consumption, ambient temperature and pressure and temperature in hydrogen storage tank measurement data were collected manually at the beginning and at the end of individual sets of measurements to obtain the integral value of each observed parameter.

3 RESULTS

On the basis of experimental data and the alkaline water electrolysis system specifications and limitations, the following calculations have been performed:

• energy efficiency of an electrolytic cell, \( \eta_c \),
• constants of the empirical \( I - U \) model,
• energy efficiency of the entire system, \( \eta \), and
• hydrogen losses within the boundaries of the system, \( \xi \).

3.1 Energy Efficiency of an Electrolytic Cell

The energy efficiency of an electrolytic cell can be calculated as:

\[
\eta_c = \frac{\dot{m}_{H_2,c}}{U_{op} \cdot I} = \eta_F \cdot \frac{\dot{m}_{H_2,c} \cdot H_{S,H_2}}{U_{op} \cdot I},
\]

where \( \eta_F \) is the faradaic efficiency (also known as current efficiency) defined as the ratio between the actual and theoretical maximum amount of hydrogen produced in an electrolytic cell, \( \dot{m}_{H_2,c} \) is the theoretical mass flow of hydrogen produced in an electrolytic cell (Eq. (3)), \( H_{S,H_2} \) is the higher heating value (HHV)
of hydrogen, and \( I \) is the electric current. Further derivation of Eq. (10) shows that \( \eta_c \) can also be written as, [12]:

\[
\eta_c = \eta_F \cdot \frac{U_{\text{in}}}{U_{\text{op}}}. \tag{11}
\]

Since there is no sensor for hydrogen mass flow measurement installed in the system, it was assumed that the hydrogen mass flow from an electrolytic cell is equal to \( m_{\text{H}_2,c} \) calculated from Eq. (3). This means that the Faraday efficiency, \( \eta_F \), in our calculations is equal to 1. Experimental data from [10] show that the Faraday efficiency is close to 1 at higher current densities. These conditions were also confirmed during experiments.

3.2 Empirical \( I - U \) Model

For the evaluation of different operating pressures on the performance of the electrolyser cell stack, the following empirical current–voltage (\( I - U \)) relationship was used, [10]:

\[
U_{\text{op}} = U_{\text{rev}} \cdot \frac{I}{A} + r_2 \cdot \ln \left( r_3 \cdot \frac{I}{A} + 1 \right), \tag{12}
\]

where \( r_1 \), \( r_2 \) and \( r_3 \) are parameters, in general depended on operating temperature and pressure. The ratio \( I / A \) represents current density (\( j \)):

\[
j = \frac{I}{A}, \tag{13}
\]

where \( A \) is an electrode area.

The results presented in Fig. 7 and Table 1 show that the operating pressure between 16 and 20 bar \( g \) has no significant influence on the electrolyser cell stack performance. Experimental data for this analysis were collected during the pCONST experiment.

The results from both experiments (BPRi and pCONST) show that the energy efficiency of an electrolytic cell at typical operating conditions is between 73 and 83%, where higher electric current corresponds to lower energy efficiency (Fig. 6).

Because of the system and experimental setup limitations, the energy efficiency of an electrolytic cell is, in our case, equal to the energy efficiency of the cell stack, [12].

![Fig. 6. Energy efficiency of an electrolytic cell at typical operating conditions](image)

![Fig. 7. \( I - U \) curves for different operating pressures](image)

Table 1. Operating conditions and values of parameters in the empirical \( I - U \) model

<table>
<thead>
<tr>
<th>Operating condition Parameter</th>
<th>( p ) [bar ( g )]</th>
<th>( T ) [°C]</th>
<th>( r_1 \cdot 10^{-3} ) [V m²/A]</th>
<th>( r_2 ) [V]</th>
<th>( r_3 ) [m²/A]</th>
</tr>
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<tr>
<td>16.0</td>
<td>61.4</td>
<td>0.17893</td>
<td>0.10175</td>
<td>0.10164</td>
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</tr>
<tr>
<td>18.0</td>
<td>62.1</td>
<td>0.16484</td>
<td>0.10485</td>
<td>0.10431</td>
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<tr>
<td>20.1</td>
<td>62.5</td>
<td>0.17795</td>
<td>0.10216</td>
<td>0.10235</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 8 also presents results from the BPRi experiments, including experimental data in the pressure range between 15 and 21 bar \( g \), which further confirm that the performance of the electrolyser cell stack is not influenced by the operating pressure in this (extended) pressure range.

According to experimental data from [10], in addition to electric current, operating (electrolyte) temperature has a significant influence on the alkaline water electrolysis process. However, in our case, electrolyte temperature could not be changed, since it is locked by the manufacturer. Nevertheless, its influence can be evaluated indirectly from experimental data obtained at a constant electric current before the beginning of the BPRi experiment (Fig. 9).
In Fig. 9, it can be observed that by operating the electrolyser at constant electric current for a longer period of time the electrolyte temperature tends to follow the sinusoidal behaviour of the coolant temperature as a result of the operating characteristics of the cooling system. Furthermore, it can be seen that operating (cell) voltage varies with the same frequency as the electrolyte temperature, but with some phase shift. The phase shift is approximately equal to π, which means that the operating voltage reaches its minimum value when the electrolyte temperature is approximately at the maximum value. This observation corresponds to the experimental data from [10] that indicates that higher operating temperatures have a favourable effect on the process of water electrolysis, since a lower operating voltage results in higher energy efficiency (Eq. (11)). The results presented in Fig. 10 confirm that the variation (sinusoidal oscillation) in voltage and consequently in the energy efficiency of an electrolytic cell is a result of the variation of electrolyte temperature and not of the electric current.

The electrolyte temperature and consequent energy efficiency of the cell could be more constant if the difference between regulation temperatures that turn on/shut off the cooling system of the electrolyte would be smaller. Because the manufacturer locked those settings, we could not test them. Another possibility to obtain more constant electrolyte temperature is to implement a different coolant system with evaporation cooling in which temperature of the coolant is constant.

### 3.3 Energy Efficiency of the Entire System

The energy efficiency of the entire system can be defined as:

\[
\eta = \frac{\dot{H}_{\text{H}_2} \cdot P_{\text{ele}}}{P_{\text{ele}}} = \frac{\delta \cdot \dot{m}_{\text{H}_2}(t_m) \cdot H_{\text{H}_2}}{P_{\text{ele}}},
\]

where \( \delta \) is the purity of hydrogen leaving the system, \( \dot{m}_{\text{H}_2}(t_m) \) represents the average mass flow of hydrogen leaving the system and \( P_{\text{ele}} \) is the electric power input of the entire system. \( P_{\text{ele}} \) is determined as the amount of electric energy consumed during the time of the experiment:

\[
P_{\text{ele}} = \frac{W_{\text{ele}}(t_m) - W_{\text{ele}}(0)}{t_m},
\]

and the average mass flow of hydrogen leaving the system is calculated according to this gas law:

\[
\dot{m}_{\text{H}_2}(t_m) = \frac{V_{\text{tank}}}{t_m \cdot R_{\text{H}_2}} \cdot \frac{P_{\text{tank}}(t_m)}{T_{\text{tank}}(t_m)} - \frac{P_{\text{tank}}(0)}{T_{\text{tank}}(0)}.
\]
is no direct measurement of hydrogen mass flow in the system. On the basis of the measured data and project documentation (volume of the hydrogen storage tank), the calculation was done for a longer period of hydrogen production to minimise the error of the calculated mass flow of hydrogen leaving the system. The results of the measurements show that the (average) energy efficiency of the entire system during the BPRi measurement set was 56.5±4.2%; during the $p_{\text{CONST}}$ measurement set, it was 54.6±3.9% (Table 2). Differences in efficiency are in principle due to higher losses of hydrogen in the second set of measurements, which corresponds to the higher operating pressure in the system, although during this experiment the average electric current was lower (Table 2), resulting in the higher energy efficiency of an electrolytic cell. The relatively high measuring uncertainty (±4%) is due to the mass flow calculation on the basis of gas law that was (in this case) the only possibility of indirectly measuring the mass flow of hydrogen leaving the system.

### 3.4 Hydrogen Losses within the Boundaries of the System

Because of the specific design of the analysed alkaline water electrolysis system, also presented in Section 2, a portion of the produced hydrogen is used for system conditioning, resulting in hydrogen losses in the atmosphere. The hydrogen losses ($\xi$) are calculated on the basis of the following equation:

$$\xi = 1 - \frac{\delta \cdot m_{\text{H}_2} \left( t_m \right)}{\eta_f \cdot N \cdot m_{\text{H}_2, c} \left( t_m \right) n}$$  \hspace{1cm} (17)

where $N$ is the number of electrolytic cells in the cell stack. Since the average mass flow of hydrogen leaving the system is calculated according to gas law, calculations are performed for a longer period of hydrogen production.

### Results and Discussion

Results show that (average) hydrogen losses within the boundaries of the system were equal to 16.1±6.5% in the BPRi experiment setup and 18.1±6.2% in the $p_{\text{CONST}}$ experiment setup (Table 2). Higher hydrogen losses in the $p_{\text{CONST}}$ experiment (up to 2%) are due to higher operating pressure in the system. At higher operating pressures, more hydrogen is needed to fill the ‘Fill Up Volume’ (Fig. 4) for conditioning the system; consequently, losses of hydrogen are higher.

On the basis of performed calculations, the average energy flows within the system boundaries for both major sets of measurements (experiment BPRi and $p_{\text{CONST}}$) are presented in Fig. 11. It can be seen that hydrogen losses converted into energy flow represent a relatively high value; therefore, a question arises of whether a design of the system with a demineralised water feed pump would have a more favourable impact on the energy efficiency of the entire system.

### 4 CONCLUSIONS

The integration of systems based on renewable energy sources (mainly photovoltaics, wind and small hydropower plants) into the existing electricity infrastructure is necessarily associated with the
development of energy storage systems. One possible solution is to store electrical energy in the internal energy of hydrogen through the electrolysis of water.

A commercial alkaline electrolyser was analysed in detail regarding the energy efficiency of an electrolytic cell, the energy efficiency of the entire system and hydrogen losses within the boundaries of the system. In addition, the parameters of an empirical model that presents electrolyser characteristics, i.e. voltage vs. electric current (current density) dependency, were determined on the basis of experimental data. The results show that:

- The energy efficiency of an electrolytic cell at typical operating conditions is between 73 and 83%, where higher electric current corresponds to lower energy efficiency.
- The operating pressure between 16 and 20 bar g has no significant influence on the electrolyser cell stack performance. The electrolyser characteristics also show that higher hydrogen production is always linked with higher energy losses.
- In addition to electric current, operating (electrolyte) temperature has a significant influence on the alkaline water electrolysis process. The variation of temperature in the range between 59 and 65 °C at constant electric current (266 A) leads to a variation of the energy efficiency of an electrolytic cell in the range between 77 and 79%, where higher temperature corresponds to higher energy efficiency.
- The calculated overall (average) energy efficiency of the system on the basis of experimental data is in the range between 50 and 60%. The results suggest that the energy efficiency of the entire system depends on the operating pressure, where higher operating pressure corresponds to higher hydrogen losses within the boundaries of the system and consequently lower energy efficiency.
- During the experiments, (average) hydrogen losses within the boundaries of the system ranged between 10 and 25%. Hydrogen losses converted into energy flows represent a relatively high value (~7 kW); therefore, a question arises of whether the design of the system with a demineralised water feed pump would have a more favourable impact on the overall energy efficiency of the system.

The analysed commercial alkaline electrolyser is able to adapt relatively quickly to new operating conditions even if the electric current or/and system pressure changes significantly. However, operating the electrolyser at variable operating conditions has a negative impact on the purity of the produced gases and the energy efficiency of the entire system.

5 ACKNOWLEDGEMENTS

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6 NOMENCLATURE

<table>
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<th>Symbol</th>
<th>Description</th>
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<tr>
<td>A</td>
<td>electrode surface</td>
<td>[m²]</td>
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<td>a</td>
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<tr>
<td>b</td>
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<td>k</td>
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<tr>
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</tr>
<tr>
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<td>number of electrolytic cells</td>
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<td>z</td>
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GREEK SYMBOLS

Δ     | difference                  |
δ     | hydrogen purity             |
η     | energy efficiency           |
ν_e   | stoichiometric coefficient  |
ξ     | hydrogen losses             |
c     | electrolytic cell           |

SUBSCRIPTS

el | electrode                   |
ele | electric                    |
F   | Faraday                     |
H_2 | hydrogen                    |
7 REFERENCES


