

Performance of Reversible Solid Oxide Cells: A Review

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Abstract

It was shown about 25 years ago that solid oxide fuel cells (SOFC) could also work in the solid oxide electrolyser cell (SOEC) mode. Electrolysis of both water (steam) and CO₂ was demonstrated. During the 1990'ies very little electrolysis work was reported in the open literature, whereas an enormous amount of results on the SOFC R&D appeared.

Recently, the increasing interest in hydrogen production has revived the interest in the solid oxide cell (SOC) as electrolyser. During the time since the work of Isenberg the power density of the SOC has increased significantly, or in other words, the area specific internal cell resistance has been decreased substantially at least for fresh cells.

This paper describes briefly the development and status of the SOC performance over the latest 25 years, and problems in obtaining high performance, reliability and durability are outlined. Examples of results obtained in the SOC research at Risø are given as illustrations of obtainable results for the SOFC technology together with a short review of the international state of the art for SOC. H₂O electrolysis with a new SOEC resulted in a record breaking current density of -3.6 A/cm² at a cell voltage of 1.48 V, which is the overall thermoneutral voltage. Assuming an electricity cost of 3.6 US\$/GJ, the H₂ production price is estimated to 4.8 US\$/GJ equivalent to 30 US\$/barrel crude oil.

The technical requirements to the SOC technology imposed by the economic situation of today are discussed. It is indicated that the SOC holds a considerable potential as an energy converter in a society with a high fraction of renewable electricity supply. Production of synthesis gas by electrolysis of a mixture of steam and CO₂ may be used as basis for production of CO₂-neutral hydrocarbon fuels. The type of R&D that is necessary in order to obtain a commercially viable SOC converter is finally discussed.

Introduction

A strongly increased interest in hydrogen and CO₂ neutral energy production has aroused during the recent years, see e.g. [1,2], and very enthusiastic but not necessarily realistic visions have been published, e.g. [3]. The hydrogen economy vision has been rejected by Bossel et al. [4], and the rejection is convincing. From Bossel's reports one may get the impression that it does not make sense at all to use electrical energy for electrolysis for hydrogen production. Bossel's main argument is that there is a big loss of the order of 75 - 80 % in converting electricity into hydrogen and back to electricity again, whereas there is only about a 10 % loss in transporting electricity by the grid. This argument is also correct in its essence even though the exact loss number in the electrolysis may be lower for future electrolyser generations than the usually assumed 30 %. Another feature pointed out by Bossel is that molecular hydrogen is troublesome and expensive to handle.

We think that in spite of these arguments there might be a future market for efficient electrolysers, which can split not only water but also carbon dioxide and produce synthesis gas, a mixture of hydrogen and carbon monoxide. All types of hydrocarbon based fuel may be produced from H₂ and CO. Especially it is inexpensive to produce the simplest synthetic fuels, namely methane, CH₄, the main constituent in natural gas, and methanol, CH₃OH. Also synthetic petrol and diesel may be (and have been in Germany during Second World War) produced from synthesis gas in large quantities using the Fischer-Tropsch method [5]. Today new large plants for manufacturing of synthetic diesel from synthesis gas using Fischer-Tropsch are being built due to the high oil price. The synthesis gas is made by steam reforming of cheap natural gas, which is available in certain places, e.g. the Near East. Synthesis gas may also be produced from coal. It is generally believed that production of synthetic fuels will be profitable, if the price of crude oil will be stable above 50 US\$ per barrel.

The question "is there a possible market for efficient electrolysis?" can only be answered by economic assessments. It is clear that the market will only be there if one of two conditions will be fulfilled: 1) the price per unit energy of fossil fuel is significantly higher than the price for alternative energy like renewable energy (wind, solar, hydropower) or nuclear energy; or 2) fossil fuel consumption is restricted by political means. As a first step in answering if it is possible to fulfil condition 1), we will here estimate at which price hydrogen may be produced by electrolysis. The price of synthesis gas produced by electrolysis in terms of price per kJ may be similar to that of hydrogen, but the price of CO₂ of a reasonable purity is a complicated story, which we will deal with in another publication at a later stage. We will make a brief review of electrolysis in general and reversible solid oxide cells (SOC) in particular before we present the economic estimate and an outlook over the future work to be done.

Electrolysis in General

Electrolysis is a 200-year-old method for hydrogen production, and still electrolysis is presently, and for the foreseeable future, the only method of practical importance for hydrogen production by splitting of water. The chlorine-alkaline electrolysis, which is worldwide the largest source of electrolytic produced hydrogen, has been in commercial use for about 100 years. In this process hydrogen is regarded as a by-product and chlorine is the main product, and therefore the chlorine-alkaline electrolysis is not dealt further with here, as this is not really interesting in the present context. Only a vanishing small portion (of the order of 0.1 %) of the world production of hydrogen is produced directly by water electrolysis. Even this small quantity has been declining during the recent years since the

electrolytic production of hydrogen for fertilizer manufacture is not competitive with production from natural gas [6].

Low Temperature Electrolysis of Liquid Water

Conventional alkaline water electrolysis works with an aqueous alkaline electrolyte. The cathode and anode areas are separated by a micro-porous diaphragm to prevent mixing of the product gasses. With output pressures of 0.2 - 0.5 MPa these processes can reach efficiencies of around 65%. Instead of the alkaline electrolyte in an inert diaphragm an acid solid proton conductor of the Nafion type (the same as in a PEMFC) may be used as a combined electrolyte and diaphragm. Further technical details may be found elsewhere [7]. Conventional water electrolyser units with capacities from 1 kW_e to 125 MW_e are commercially available. The Electrolyser Corporation Ltd. (Canada) and Norsk Hydro Electrolysers AS (Norway) and DeNora (Italy) are well-established manufacturers of electrolysers. Other manufacturers have also established themselves in Europe, e.g. Hydrogenics Corporation. Low temperature (below 100 °C) electrolysers are being developed for high-pressure water electrolysis allowing the direct generation of hydrogen at pressures up to 12 MPa based on alkaline technology. A 5 kW_e prototype was constructed and tested at Forschungszentrum Jülich in Germany [8]. As the volumetric energy density of gaseous H₂ is rather low, it is an advantage to produce pressurised hydrogen directly. Furthermore, the internal electric resistance is lower in case of high pressures resulting in overall increased energy efficiency in the production of pressurised hydrogen.

High Temperature Electrolysis

High temperature electrolysers were under development during the 1980'es [9 -12]. One advantage of the high temperature that a part of the energy required for water splitting is obtained in the form of high temperature heat, and thus the electrolysis is performed with lower electricity consumption. The discussions focussed on the use of heat from solar concentrators or waste heat from power stations for this purpose [12]. Due to a low energy price this development was stopped around 1990. The high temperature solid oxide electrolyser cell (SOEC) has the advantage that it can also split CO₂ into CO and O₂. Further, the high temperature is speeding up the reaction kinetics, which in turn decreases the internal cell resistance and, thereby, increases the energy efficiency. These features open up new potential possibilities for a broader application of renewable or nuclear energy in the future, if fossil fuels become scarce, and therefore several R&D-projects on SOEC are now being started again both in Europe, USA and Japan [13 - 20].

A system consisting of a heat exchanger and a reversible SOC system has clear advantages compared to low temperature electrolysis. Because water electrolysis is increasingly endothermic with temperature, electricity demand can be significantly reduced, if the formation of hydrogen is taking place at high temperatures (600-1000 °C) as illustrated in Fig. 1. The electric energy need is reduced because the unavoidable joule heat of an electrolysis cell is utilized in the water (steam) splitting process at high temperature. If heat is available from sources such as heat of geothermal (e.g. on Island), solar or nuclear origin, this will further reduce the electric energy demand for hydrogen production by steam electrolysis. Even where such high temperature heat is not available SOEC may be of interest. All heat sources with temperatures above 100 °C (the boiling point of water) are extremely beneficial since electric energy for steam rising will be saved.

The Faradayic efficiency of SOEC has been shown to be 100 % over a period of 1000 h [12], i.e. there are no parasitic reactions. This taken together with the endothermic nature of the water splitting means that the hydrogen efficiency, defined as the total chemical energy (enthalpy of reaction, ΔH) in the hydrogen divided by the electric energy consumed, will be 100 % minus the heat loss from the electrolyser to the surroundings. Thus, for well-insulated SOEC stacks in systems in the range of 1 MW or above (ca. 1 m³ stack volume) the thermal loss can most probably be well below 10 %. Also, some electric energy will be consumed in the system for inverters and pumps, but again for reasonably sized systems this may be few % only. This means that the SOEC technique has a potential of a hydrogen efficiency of ca. 90 % for a system.

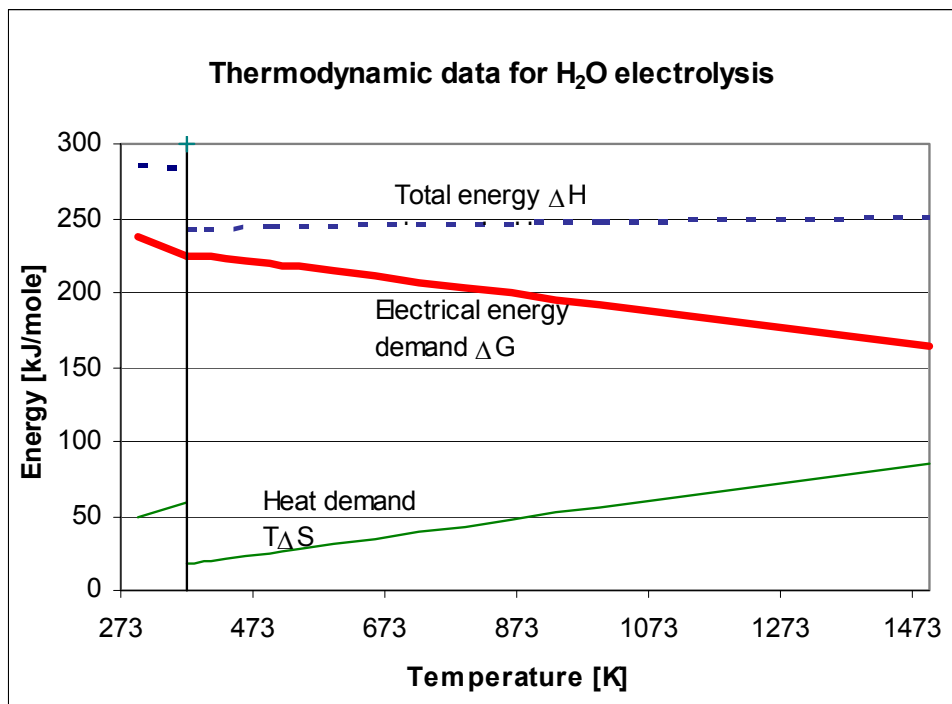


Figure 1 Thermodynamics of water electrolysis. Data are taken from [21]

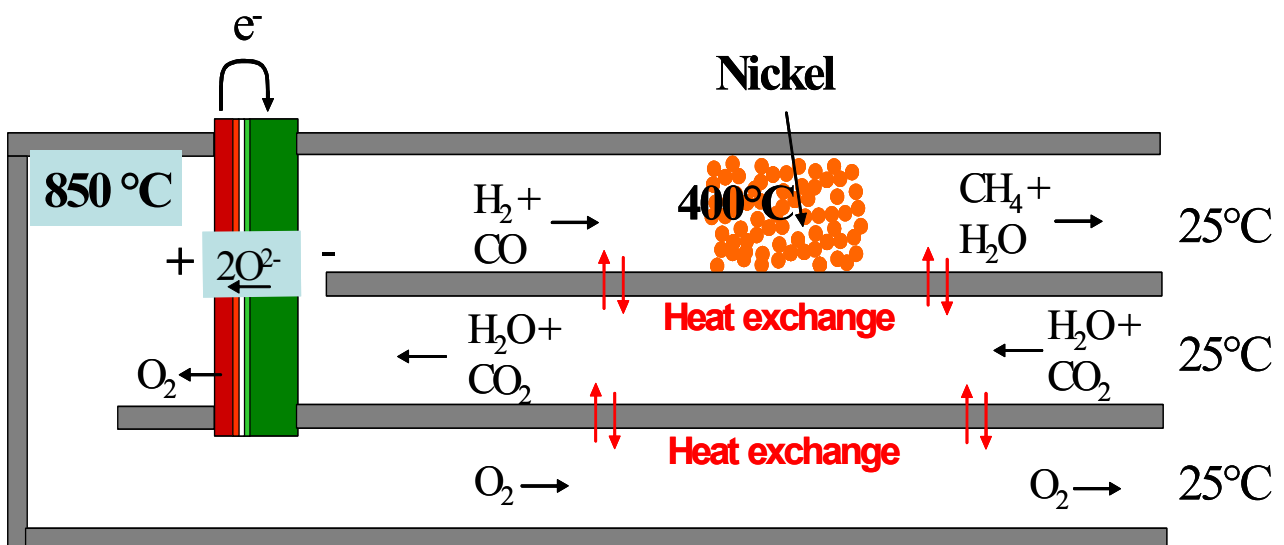


Figure 2 Sketch of an SOEC system for CH₄ production by electrolysis of steam and CO₂. CO₂ and H₂O are fed through the heat exchanger to the cell. Here it is split into H₂ and CO (syngas) and O₂. On the way out, the synthesis gas is catalysed into CH₄ and H₂O using a Ni-catalyst

As mentioned SOEC can split carbon dioxide into carbon monoxide and oxygen, and the CO₂ splitting has an endothermicity similar to that of water splitting. This means that electrolysis of a mixture of steam and carbon dioxide results in a mixture of hydrogen and carbon monoxide called syngas. By catalytic reactions a number of other energy carriers may be produced from syngas. The two simplest are methanol and methane. The preferred catalyst for CH₄ formation is Ni. Since the negative electrode of a SOC is partly made of Ni it is in principle possible to produce CH₄ within the cell (at high pressure and low temperature [15]). The entropy change for CH₄ production from CO₂ and H₂O is nearly zero. This means that the overall efficiency for a conversion of electricity to CH₄ and back again can be very high if the reaction kinetics is high, since only small reaction entropy losses occur. The catalytic reaction to form CH₄ or CH₃OH from syngas can also be done in the heat exchanger after the cell as sketched in Figure 2. This means that the energy for H₂O vaporization can be produced within the system. A combination of the two ways to produce CH₄ may prove to be the best production method, since it seems to optimise efficiency and production rate.

Status of SOC

The SOC of all types are basically reversible cells and can be operated as solid oxide fuel cells (SOFC) for electricity production and as well as solid oxide electrolysis cells (SOEC) for production of hydrogen and synthesis gas. Figure 3 presents the kinetics for SOCs fabricated and tested at Risø in both fuel cell mode (SOFC) and electrolyser mode (SOEC) at different temperature and steam partial pressure [21]. It illustrates that the cell is genuinely reversible as the I-V curves go smoothly through the zero-current-density-point.

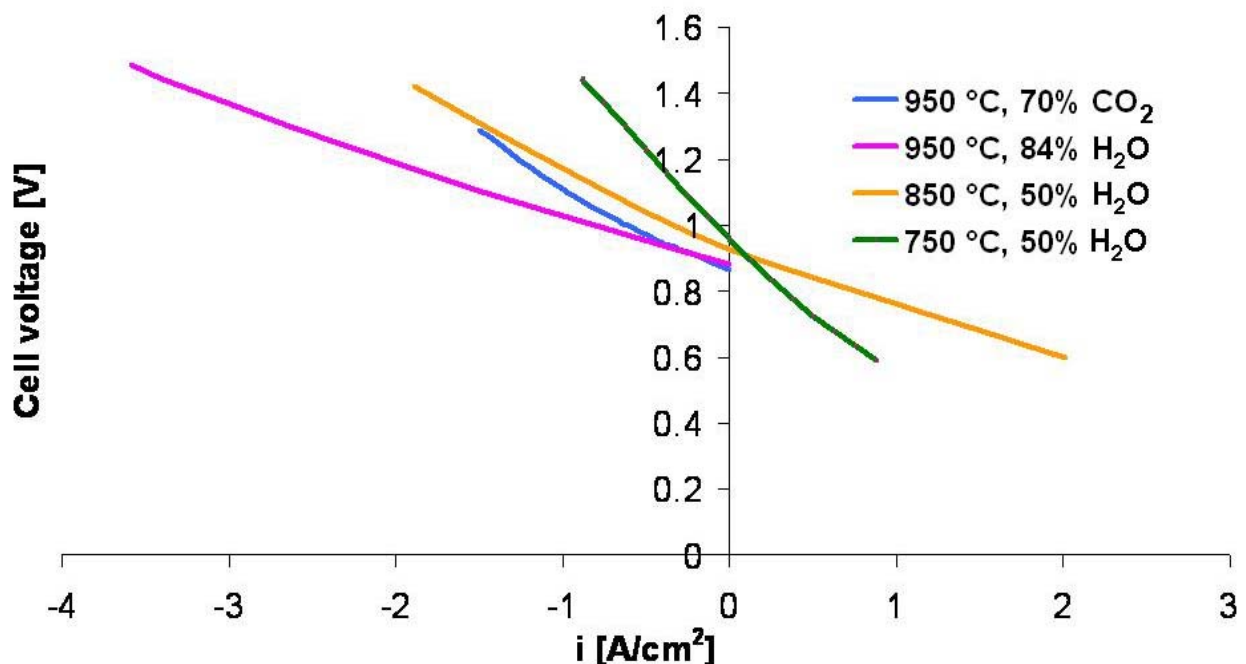


Figure 3 Kinetics of a Risø SOC working as an electrolyser cell (negative current densities i) and as a fuel cell (positive current densities i) at different temperatures and steam or CO₂ partial pressures in the inlet gas to the cell [21]

At a cell voltage of 1.48 V the produced joule heat within the cell equals the consumed heat in the steam generation plus the steam electrolysis process. 1.48 V is therefore called thermoneutral potential (E_{tn}). At E_{tn} and 950 °C with 84% H₂O + 26% H₂ in the inlet gas a

current density of -3.6 A/cm^2 was measured with 30% steam utilization. To the authors best knowledge this is the highest current density reported in the literature for SOEC operation. Also included in Figure 3 is an i-V curve at $950 \text{ }^\circ\text{C}$ with 70% CO_2 + 30% CO in the inlet gas. At -1.5 A/cm^2 the cell voltage was 1.29 V and the CO_2 utilization was 21 %. The internal resistance (the slope of the i-V curve) is almost as good in electrolyser mode as in fuel cell mode.

A low internal resistance of the cell both at start-up and during thousands of hours of electrolysis operation is important for SOCs to become interesting from a commercial point of view as the hydrogen production price is dependent on the resistance of the cell. So far only few results on durability of high performing SOECs have been reported in the literature. Even though the operation of the SOCs is reversible and even have comparable initial performance in electrolysis and fuel cell mode, the degree of passivation of the cells during long-term testing in fuel cell and electrolysis operation mode can be dramatically different as seen by comparison of test of the same types of cell in the two modes [22,23]. Whereas the cell in fuel cell mode is reasonably stable over years with high current density $1 - 2 \text{ Acm}^{-2}$, it is in the electrolyser is only stable at relatively low current density at 0.25 Acm^{-2} . In the electrolyser mode a special phenomenon is seen. Figure 4 shows how the cell voltage first increase slightly and then decrease slowly again.

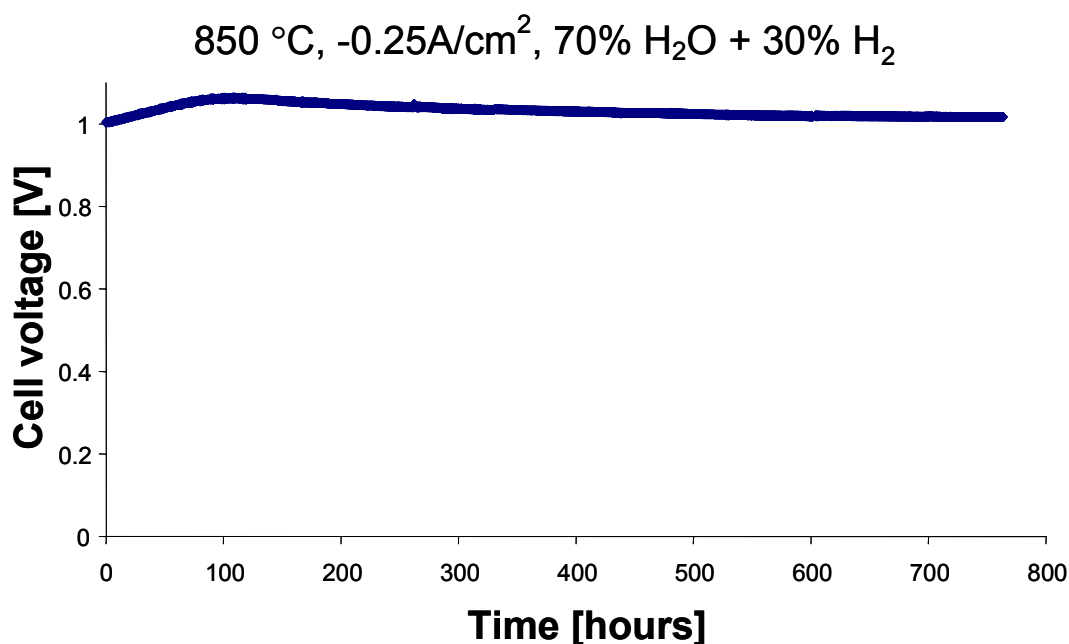


Figure 4 Cell voltage as a function of time for an electrolysis test -0.25 A/cm^2 , 850°C , $p(\text{H}_2\text{O}) = 0.7 \text{ atm}$ and $p(\text{H}_2) = 0.3 \text{ atm}$ at the hydrogen electrode

This passivation (the increase of the cell voltage over time) becomes more pronounced with higher current densities, higher water partial pressure and lower temperature. At some conditions the cell does not activate again unless the current is reversed, i.e. the cell is loaded in fuel cell mode. Thus, the passivation/activation is at least partially reversible. Strong indications have been found that the passivation is due to an accumulation of impurities at the three phase boundaries [23,25]. The phenomenon is under further study at Risø. We believe strongly that this passivation can be handled by proper handling of the trace impurities in the hydrogen electrode.

Table 1 summarizes the literature results of area specific internal resistances (ASR) of SOECs at similar conditions. The variation is large, but by examination of the references it seems that there is some correlation to the year of publication. In other words, the SOCs have been improved very significantly over time due to the large international R&D efforts. There are good reasons to believe that these improvements will continue to happen during the coming years.

Table 1 Some reported initial performances of electrolysis cells. Comparison of ASRs obtained from i-V-curves. The ASRs are taken as the slopes in the linear regions of the electrolysis i-V-curves presented in the references cited. For each reference the ASR on full cells for systems and with the experimental conditions closest to the ones applied in this work is given

Ref.	T [°C]	p(H ₂ O) [atm]	p(H ₂) [atm]	ASR [Ωcm ²]	Specifications
[23]	850	0.50	0.50	0.27	Ni/YSZ-YSZ-LSM planar
[23]	950	0.50	0.50	0.15	Ni/YSZ-YSZ-LSM planar
[12]	1000	0.67	0.33	1.17	Ni/YSZ-YSZ-LSM tubular
[24]	908	0.67	0.33	2.7	Ni/YSZ-YSZ-LSM tubular
[25]	1000	0.91	0.09	2	Ni/YSZ-YSZ-LSM
[26]	1000	0.50	0.50	0.7	Ni/YSZ-YSZ-LSM
[17]	850	0.50	0.50	0.45	Ni/YSZ-ScSZ(175 μm)LSM
[19]	900	0.50	0.50	1.8	Ni/SDC-YSZ-LSC
[18]	850	0.11	0.89	0.35	Ni/YSZ-ScSZ(125 μm)-LSM

Assessment of the SOEC Economy

This section gives some cautious economic estimates of the economic potential of the SOEC. The operation temperature of SOC is in the range of 700 to 1000 °C. This implies that a significant part of the heat required for the electrolysis process can be obtained as heat, see Figure 1. This provides an opportunity to utilise the inevitably produced Joule heat due to the passage of electrical current through the cell and reduces the overall electricity consumption, and, thereby, the H₂ production price. Also the kinetics of a SOC gets increasingly better with increasing temperature. The internal polarisation resistance follows an Arrhenius expression [27]. This results in higher current densities at given cell voltage and steam partial pressure as the temperature is increased.

A heat exchanger is used in order to save expenses for heating the feed gas to working temperature of the SOC stack. The typical operating temperature for catalysis of H₂ + CO is well above the boiling temperature of water and the heat generated by the catalysis process may therefore be used for steam rising in the inlet part of the heat exchanger. Only 30% of the steam was utilized at the maximum current density of -3.6 Acm⁻². The remaining steam in the outlet gas can be separated from the hydrogen by condensation and recycled in the heat exchanger.

The high current density result is used below in an estimation of the H₂ production price. The other economical input parameters are given in Table 2. Using the input given in Table 2, the H₂ production prices is estimated to be 4.9 US\$/GJ, taking the higher heating value (HHV) of H₂. This corresponds to a break even crude oil price of 30 US\$/barrel again using the HHV, which is significantly cheaper than the current crude oil stock market price which is above 50 US\$/barrel crude oil.

Table 2 Input for calculation of H₂ production cost

SOC stack	2100 US\$/m ² cell area
Investment cost	6300 US\$/m ² cell area*
Interest rate	5%
Depreciation time	10 years.
Operation time	5 years
Demineralised Water cost	2.3 US\$/m ³
Electricity price	1.3 US¢/kWh (3.6 US\$/GJ)
Cell temperature	950 °C
Cell voltage	1.48 V (E_{tn})
H ₂ O utilization in the SOC stack	30%
Energy loss in heat exchanger	5%

*A 5kW plant based on SOFC technology is predicted to cost 350-550 US\$/kWe [28]. Assuming a power output of 1W/cm² this corresponds to an investment cost of 3500-5500US\$/m² cell area.

Figure 5 shows the parts of the H₂ production price given the assumptions in Table 2. It is seen that electricity costs for evaporation and steam rising dominates the investment cost. Note that the production price is calculated at E_{tn} , which means that the electricity to hydrogen efficiency is 95%. Heat losses to the surroundings are not included in this calculation, but these can be minimised by thermal insulation using cheap materials such as mineral wool.

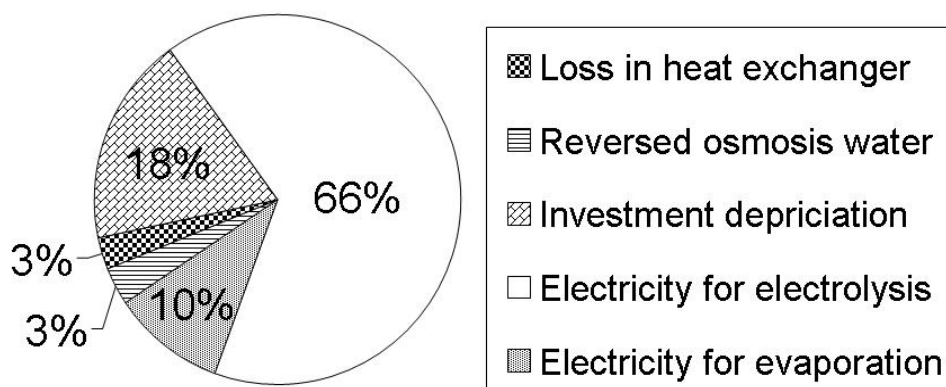


Figure 5 Parts of H₂ production price. It is seen that electricity costs takes up 3/4 of the total production price

The electricity for evaporation can be omitted in a synthetic fuel production where the heat from the catalytic reaction is used for steam rising. However, investment costs for the catalyser as well as lower higher heating value (HHV) of the synthetic fuel compared (on a molar basis) to the synthesis gas, will result in a higher production price of the synthetic fuel than of hydrogen in terms of US\$/GJ (HHV).

Electricity cost is the major constituent of the production price. Figure 6 shows the H₂ production price dependency on the electricity price. Note that the potential H₂ production price using SOEC technology is almost half of that of using ordinary alkaline electrolysis at an electricity price of 2 US¢/kWh.

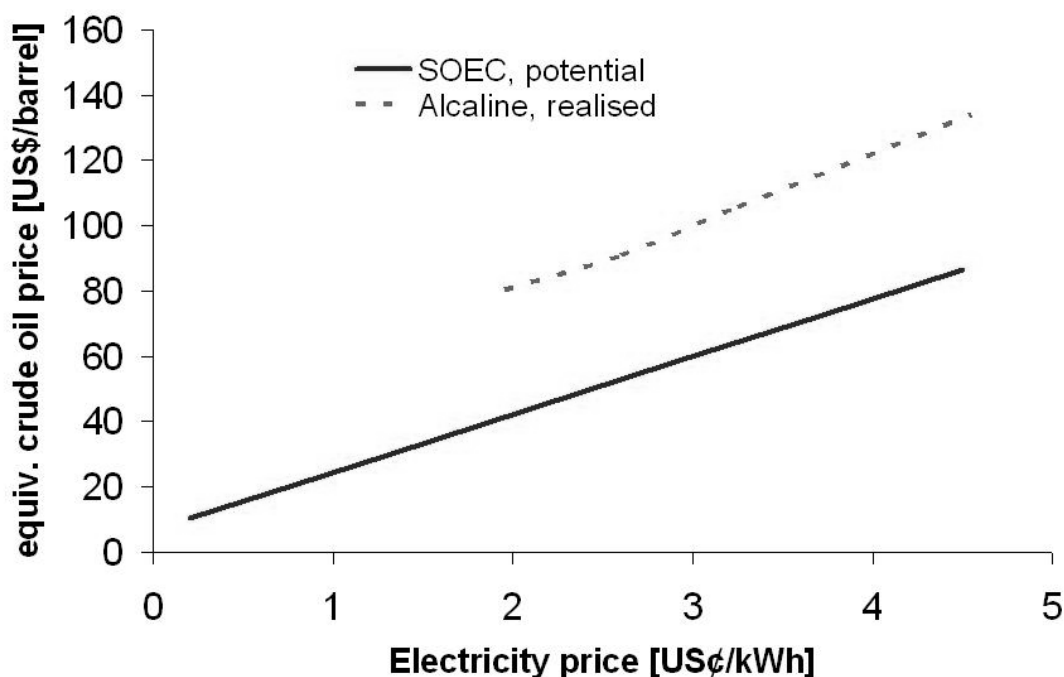


Figure 6 H₂ production price given as equivalent crude oil price vs. electricity price using HHV. The price of H₂ production from alkaline electrolysis is presented for comparison [29]

Outlook

A comparison between the calculated price for hydrogen production based on our experimental results and the world market price of crude oil indicates that the SOEC technology has a potential for the production of synthetic fuel from renewable energy sources in places where electricity can be extremely cheaply generated, i.e. with large scale hydro power systems (e.g. Egypt, Brazil, Iceland, Canada, Norway, Congo), or with excess energy from the primary and secondary control of existing power station capacity with significant nuclear component (e.g. France, Belgium, Switzerland).

However, before this potential can be realized more R&D is required. It is therefore necessary to produce a high performing SOEC as well as a long-term stable cell with a continuous high performance. A massive R&D effort is probably necessary in order to obtain inexpensive electrolyzers with a sufficient high durability and efficiency for a broad commercial application. The total need is estimated to be hundreds of man-years over many years. Mainly the following research areas are important in this context: 1) materials research in order to identify improved materials and fabricate effective structures, 2) surface science in order to understand the nature of the interfaces between the electrodes and electrolyte, and 3) solid state electrochemistry in order to understand the processes and the losses involved. It is of major importance that these areas are researched in an intimate interplay. Furthermore, we believed that there is a special application of electrolysis in large integrated future systems, which produce both electricity and synthetic fuel as described in the RETrol vision by the Danish company, Elsam [30]

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