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MSc THESIS REPORT

FUEL-ASSISTED ELECTROLYSIS
COMPARISON TO
CONVENTIONAL HIGH-TEMPERATURE ELECTROLYSIS

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Fuel-assisted electrolysis comparison to conventional high-temperature electrolysis

MASTER OF SCIENCE THESIS

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Abstract

Solid Oxide Fuel-assisted Electrolysis Cells (SOFEC) are a relatively recent development as far as fuel cell research is concerned. Laboratory tests predict that they have the ability of efficiently producing hydrogen and, most importantly, at a lower cost compared to conventional high temperature electrolysis. The SOFEC mode of operation is related to the conventional SOFC and SOEC. The available theoretical knowledge for those modes is presented and then applied to SOFEC. In this report the operation of SOFEC is analyzed and explained, while the experimental part aims at validating the theory through laboratory experiments, inspired by previous tests in literature.

The conducted experiments focus on symmetrical button cells, with platinum being the electrode material for both the anode and the cathode. The initial tests are carried out on custom-made cells, with platinum pasted on an electrolyte substrate by hand. The equipment used, as well as the procedure of preparing the test setup are detailed. This is followed by proposed setup modifications for future application. Afterwards, the results are presented. These focus on the polarization curves for different operating modes, while comparisons between SOFEC and SOEC modes illustrate the positive impact of the assisting fuel on the performance of the electrolyzer. Hydrogen and carbon monoxide are used as fuels, while carbon dioxide is utilized for co-electrolysis with steam. Comparisons of the electrolyzer performance between different fuels are presented and explained. Finally, the results are summarized and recommendations for future research are given.
Table of Contents

Abstract ........................................................................................................... v

Table of Figures ............................................................................................. vii

List of Tables ................................................................................................. x

1 Solid Oxide Fuel-Assisted Fuel Cells ......................................................... 1
   1.1 Introduction ....................................................................................... 1
   1.2 Solid Oxide Fuel Cell (SOFC) .......................................................... 2
   1.3 Solid Oxide Electrolyzer Cell (SOEC) ............................................. 4
   1.4 Solid Oxide Fuel-Assisted Electrolyzer Cell (SOFEC) ..................... 5
   1.5 Solid Oxide Cell Voltage and Power ............................................... 7
   1.6 Solid Oxide Cell Efficiency .............................................................. 11
   1.7 Previous experiments on SOFEC .................................................... 13
   1.8 Co-electrolysis of carbon dioxide and steam .................................. 15

2 Experimental setup .................................................................................. 16
   2.1 Setup outline .................................................................................. 16
      2.1.1 Gases Inputs/Output ................................................................ 16
      2.1.2 Electrical Connections ............................................................ 17
   2.2 Setup Preparation ........................................................................... 18
      2.2.1 Nickel mesh ........................................................................... 18
      2.2.2 Mica sealing ........................................................................... 19
      2.2.3 Cell Accommodation .............................................................. 19
      2.2.4 Aluminum-silicate felt ............................................................ 20
      2.2.5 Gold mesh and wires ............................................................. 20
   2.3 Ceramic furnace .............................................................................. 23
      2.3.1 Temperature control ................................................................. 23
   2.4 Analysis equipment ........................................................................ 23
   2.5 Future setup modifications ............................................................. 24
      2.5.1 Flaxell Open-Flanges Set-Up modifications ............................. 24
      2.5.2 10-by-10 ceramic test setup modifications ............................. 26

3 Experimental Results .............................................................................. 27
   3.1 Introduction to the experiments ....................................................... 27
   3.2 Platinum pasted electrolyte substrate ............................................. 27
      3.2.1 SOFEC and SOEC modes experimental results comparison ...... 28
      3.2.2 Carbon dioxide as an extra oxygen source for electrolysis ........ 31
      3.2.3 Carbon monoxide as a fuel in SOFEC mode ......................... 33
   3.3 Cermet cathode symmetrical button cell ....................................... 34
      3.3.1 Comparison between SOEC and SOFEC modes .................... 35
      3.3.2 Carbon monoxide as assisting fuel for electrolysis .................. 36
      3.3.3 Steam and carbon dioxide co-electrolysis .............................. 37
      3.3.4 Cell degradation after 3 days ................................................. 38

4 Conclusions and discussion .................................................................. 40

5 Bibliography ......................................................................................... 42

Appendix ....................................................................................................... a
Table of Figures

Figure 1. Illustration of a SOFC. [5] .......................................................... 2
Figure 2. Illustration of a SOEC. [5] .......................................................... 4
Figure 3. Simplified illustration of SOFC. [5] .......................................... 5
Figure 4. Illustration of SOFC. [5] .......................................................... 7
Figure 5. The energy transfer processes of SOEC and SOFC. [8] ................. 7
Figure 6. Vrev for changing H2O flow at the cathode. [5] ............................ 9
Figure 7. Different polarizations within the SOFC. [5] ............................. 9
Figure 8. Cell voltage (based on theoretical V-I curves of SOFC and SOEC) and power density of all three modes. [5] .......................................................... 10
Figure 9. A schematic of cell operation in four quadrants. Quadrant I: SOFC, Quadrant II: SOEC, Quadrant III: SOFC, and Quadrant IV: SOFEC. The schematic also shows equivalent circuits and the directions of current flow. [4] .......................................................... 10
Figure 10. Experimental data on SOFC, SOEC, and SOFEC modes of operation. The SOFEC data, which are in quadrants I and IV, are also plotted in quadrants III and II by a 180° rotation for comparison with the SOEC mode. [4] .......................................................... 14
Figure 11. The i-V and i-P of SOEC, 12% CO-SOFEC and 12% CH4-SOFEC. .......................................................... 15
Figure 12. The efficiency ignoring heat of SOEC, CO-SOFEC and CH4-SOFEC. .......................................................... 15
Figure 13. Diagram of co-electrolysis of CO2 and H2O in a solid oxide cell, as part of a renewable fuel cycle. The electrode reactions are also given. [12] .......................................................... 15
Figure 14. Bottom view of the Open Flanges Test Set-Up: Tubes and flanges (a, o, e & l) are made with Inconel 600 & 601 that gives a full protection against high temperature corrosion. ............................. 16
Figure 15. Top view of the setup in the ceramic furnace during an experiment. .......................................................... 17
Figure 16. Setup preparation: The nickel foam sits on the bottom plate, with a hole in the center and paths for better gas distribution. Mica sealing around the cell is also present in the photograph. .......................................................... 18
Figure 17. Setup preparation: Three gold meshes were used instead of nickel, one on top of the other. .......................................................... 19
Figure 18. Setup preparation: The cell sits on top of the nickel mesh and mica sealing. It is kept in place by four thin pieces of transparent tape. .......................................................... 20
Figure 19. Setup preparation: Bottom aluminum-silicate felt on top of the cell. The hole is approximately the same diameter as the cell's electrode (anode in this case). The hole in the
center is present in order to allow the gases to pass through to the cell. .................21

Figure 20. Setup preparation: Gold mesh with two gold wires welded to it. The part of the aluminum felt that was removed is placed on top of the gold mesh to provide extra stability and sealing. The hole in the center is present in order to allow the gases to pass through to the cell. .........21

Figure 21. Setup preparation: The top aluminum felt is place on top of the cell and the bottom felt. The hole in the center is present in order to allow the gases to pass through to the cell. .......22

Figure 22. Setup closed, with the cell inside, and ready to be put in the furnace, with the anode thermocouple wire being also visible in the picture. The gold wires used for the anode current collection can be seen as well, running through two of the four holes of the white ceramic pipe. ........................................................................................................................................22

Figure 23. Kittec furnace - Left: Test rig is inserted / Right: Furnace interior after an experiment ....23

Figure 24. Reference 600+ potentiostat - Left: Front view / Right: Back view. [14] .......................24

Figure 25. Proposed changes for the Fiaxell Open Flanges Test Set-Up upper plate in order to incorporate outlet gases pipe on the anode side. Left: Design with external outlet pipe. Right: Design with outlet pipe around the inlet pipe. .................................................................25

Figure 26. Design with external outlet pipe. Left: Bottom view. Right: Side view. ........................25

Figure 27. Design with outlet pipe around the inlet pipe. Left: Bottom view. Right: Side view........26

Figure 28. Illustration of the modified top part of the 10x10 ceramic setup. Left: 3D view of the model. Right: Top view with the internal openings being visible.................................................................26

Figure 29. Cell 1 - SOFC mode: Polarization curve comparison between 100% H$_2$ and 50% H$_2$......29

Figure 30. Cell 1 - SOFEC mode: Polarization curve comparison between 100% H$_2$ and 50% H$_2$ at the anode. 20% H$_2$O was used at the cathode. ........................................................................................................30

Figure 31. SOFEC vs SOEC mode polarization curve comparison between Cell 1 and Cell 4. 100% H$_2$ was used at the anode in SOFEC mode and 50% H$_2$O at the cathode in both modes.................30

Figure 32. Cell 4 – SOEC mode: Polarization curve comparison between 0% CO$_2$ - 50% H$_2$O (steam electrolysis) and 50% CO$_2$ - 50% H$_2$O (CO$_2$/H$_2$O co-electrolysis) at the cathode side of the cell. The anode side was fed with N$_2$, .................................................................32

Figure 33. Cell 4 – SOFEC mode: Polarization curve comparison between 0% CO$_2$ - 50% H$_2$O (fuel assisted steam electrolysis) and 50% CO$_2$ - 50% H$_2$O (fuel assisted CO$_2$/H$_2$O co-electrolysis) at the cathode side of the cell. In both cases the anode side was fed with 100% H$_2$ as a fuel...........32

Figure 34. Cell 4: Polarization curve comparison between 100% CO, 50% H$_2$ - 50% CO and 100% H$_2$ at the anode side in Fuel Assisted Electrolysis mode. For all three cases 50% H$_2$O was fed to the cell’s cathode side. ..................................................................................34

Figure 35. Cermet cell: SOFEC vs SOEC mode polarization curve comparison. 100% H$_2$ was used at the anode in SOFEC mode and 50% H$_2$O at the cathode in both modes........................................35

Figure 36. Cambridge cell: Polarization curve comparison between 100% CO and 100% H$_2$ at the anode
side in Fuel Assisted Electrolysis mode. For both cases 50% H₂O was fed to the cell’s cathode side.

Figure 37. Cermet cell – SOEC mode: Polarization curve comparison between 0% CO₂ - 50% H₂O (steam electrolysis) and 50% CO₂ - 50% H₂O (CO₂/H₂O co-electrolysis) at the cathode side of the cell. The anode side was fed with N₂. .......................... 36

Figure 38. Cermet cell – SOFEC mode: Polarization curve comparison between 50% H₂O - 0% CO₂ (fuel assisted steam electrolysis) and 100% CO₂ - 0% H₂O (fuel assisted CO₂/H₂O co-electrolysis) at the cathode side of the cell. In both cases the anode side was fed with 100% H₂ as a fuel. .......... 38

Figure 39. Cermet cell: Polarization curve comparison after 3 days of operation. On both cases 100% H₂ was used at the anode, while the cathode was fed with 50% H₂O. ......................... 39

Figure 40. Cell 4: Polarization curve comparison between SOFEC mode for 100% H₂ at the anode before and 2 minutes after an impedance spectroscopy experiment using 100% CO at the anode. 50% of H₂O was used at the cathode side of the cell. ............................................. a

Figure 41. Polarization curve comparison between SOFEC mode for 100% H₂ at the anode before and 2 minutes after an impedance spectroscopy experiment using 100% CO at the anode. 50% of H₂O was used at the cathode side of the cell. ............................................. b

Figure 42. Proposed changes for the Fiaxell Open Flanges Test Set-Up upper plate in order to incorporate outlet gases pipe on the anode side. Left: Design with external outlet pipe. All values are in mm. ................................................................. c

Figure 43. Proposed changes for the Fiaxell Open Flanges Test Set-Up upper plate in order to incorporate outlet gases pipe on the anode side. Right: Design with outlet pipe around the inlet pipe. All values are in mm. ................................................................. d

Figure 44. Illustration of the modified top part of the 10x10 ceramic setup. All values are in mm. ....... e

Figure 45. Illustration of the modified top part of the 10x10 ceramic setup. All values are in mm. ....... f
List of Tables

Table 1. Cell 1: Gas flow rates used during the experiments in SOFC mode, both for the anode and the cathode of the cell ........................................................................................................... 28

Table 2. Cell 1: Gas flow rates used during the experiments in SOEC mode, both for the anode and the cathode of the cell ........................................................................................................... 28

Table 3. Cell 1: Gas flow rates used during the experiments in SOFEC mode, both for the anode and the cathode of the cell ........................................................................................................... 29

Table 4. Cell 4: Gas flow rates used during the experiments in SOEC mode, both for the anode and the cathode of the cell ........................................................................................................... 31

Table 5. Cell 4: Gas flow rates used during the experiments in SOFEC mode, both for the anode and the cathode of the cell ........................................................................................................... 31

Table 6. Cell 4: Gas flow rates used during the experiments in SOFEC mode, both for the anode and the cathode of the cell ........................................................................................................... 33

Table 7. Cambridge cell: Gas flow rates used during the experiments in SOEC mode, both for the anode and the cathode ........................................................................................................... 35

Table 8. Cambridge cell: Gas flow rates used during the experiments in SOFEC mode, both for the anode and the cathode ........................................................................................................... 35

Table 9. Cambridge cell: Gas flow rates used during the experiments in SOEC mode, both for the anode and the cathode ........................................................................................................... 37

Table 10. Cambridge cell: Gas flow rates used during the experiments in SOFEC mode, both for the anode and the cathode ........................................................................................................... 37

Table 11. Cambridge cell: Gas flow rates used during the experiments in SOFEC mode, both for the anode and the cathode ........................................................................................................... 36
1 Solid Oxide Fuel-Assisted Fuel Cells

1.1 Introduction

The use of fossil fuels as the main source of energy throughout the years poses many threats to the environment, which include global warming due to the release of greenhouse gases (GHG), atmospheric pollution and acid rain. In an effort to cope with the environmental issues which have arisen and in order to ensure the decarbonization of the energy sector, the European Climate Foundation (ECF) has issued a series of recommendations and solutions for each sector, such as transportation, buildings, industry and electricity. These recommendations are proposed in order for each sector, and especially the electricity one, to comply with the GHG reduction targets set by the European Union and which should be achieved by 2050 [1]. Under this scope, hydrogen can potentially be used in many sectors, as it could be an efficient transportation fuel, as well as a means of producing electricity without GHG emissions. Still, most of the hydrogen needs are covered by hydrogen production through fossil fuels, such as coal gasification or steam reforming from natural gas [2].

Electrolysis of water is a method of producing hydrogen by consuming electricity, while also reducing the local environmental pollution, as it is an emission-free procedure. In the reverse mode, hydrogen can be used to produce electricity by being oxidized back to water.

The above procedure can be take place in electrochemical devices known as Solid Oxide Cells (SOC). When SOC are used in order to produce electricity they are called a Solid Oxide Fuel Cells (SOFC), which can operate with various gaseous fuels such as hydrogen, natural gas, syngas, biogas, carbon monoxide and many reformed hydrocarbons. These devices can also work in reverse, which means that they can be used to perform water vapor electrolysis, therefore called Solid Oxide Electrolyzer Cells (SOEC). In order for SOEC to work, an external voltage needs to be applied, which will break the bonds between the hydrogen and oxygen molecules of water at the steam electrode, transporting oxygen ions through the electrolyte to the oxygen electrode.

Many European governments are already promoting the implementation of renewable energies, such as wind and solar power. However, hydrogen production is not yet considered in the market due to the high costs that the available hydrogen technologies encompass, more notably electrolyzers. Although producing hydrogen through fuel cell electrolysis has many advantages, the price of doing so is still 2-3 times higher than that of the conventional steam reforming [3]. A potential solution to this problem could be a third mode for SOC which operates by having fuel such as hydrogen, reformed hydrocarbons and natural gas at the fuel electrode, while steam is used as an oxidant at the oxygen supplying electrode. A mix of carbon dioxide and steam are present at the fuel electrode, while water vapor at the oxygen electrode is converted to hydrogen. In order to help the chemical driving force, a small voltage should be applied to the cell. The fuel cells used as described above are called Solid Oxide Fuel-Assisted Electrolyzer Cells (SOFEC) [4]. The main reason for introducing the SOFEC mode is to increase the efficiency of electrolysis. This is achieved by replacing a part of the electricity needed, which is rather expensive to produce, with a low value fuel such as methane.

Most SOC consist of two electrodes and a layer of electrolyte between them. One of the electrodes is the oxygen supplying one and the other one is the fuel electrode. The oxygen electrode, which is the anode in SOEC, typically consists of Sr-doped LaMnO$_3$ (LSM), while for the fuel electrode Ni and YSZ (yttria-stabilized zirconia) is used. A layer of electrolyte is placed between the electrodes, while in most cases it is made of YSZ. The typical operating temperature of a SOFC is between 700°C and 1000°C, while the fuel mix that is fed to the fuel electrode is H$_2$ and CO that results from reforming a carbonaceous fuel. In order to convert the fuel chemical free energy of oxidation into electricity, a load needs to be connected to the fuel cell, between the anode and the cathode.
A SOFEC therefore takes advantages of the characteristics of SOFC and SOEC, as it can electrolyze water as a SOEC, but also oxidize hydrogen as a SOFC, under certain conditions. A further examination of both operating modes is performed in Sections 1.2 and 1.3 before moving on to explaining how exactly a SOFEC works in Section 1.4.

Taking all of the above into account the main objectives of the experiments performed during this thesis are:

- Comparison of the performance between the SOFEC and SOEC modes
- Verifying the decrease in electrical power demand between the SOFEC and SOEC modes
- Comparison of the SOFEC performance between different gaseous fuels

A detailed description of the experimental setup used is given in Chapter 2, while the experiments and the results obtained are presented in Chapter 3. Finally, the conclusions of this report are drawn in Chapter 4, while also recommendations for future research are given.

1.2 Solid Oxide Fuel Cell (SOFC)

Figure 1 shows a simplified illustration of how a SOFC works as well as the parts it consists of. Fuel and oxygen is consumed by the fuel cell, while water, electricity and heat is produced. The anode is the electrode where the oxidation reaction takes place, while the cathode is the electrode where the reduction reaction occurs. In particular, fuel is added at the anode (in this case H₂) which reacts with the oxygen ions traveling through the electrolyte from the cathode to the anode. During the oxidation reaction that happens at the anode, O₂⁻ react with H₂ releasing electrons and creating water. The electrons released travel through an external circuit, passing through the load and are then consumed at the cell cathode. In a usual operation of a SOFC air would be used instead of pure oxygen at the cathode, which reacts with the electrons that arrive from the anode, producing oxygen ions that travel through the electrolyte to the anode, and thus completing the circle.

The half-cell reactions that govern the process during SOFC mode can be seen below. Notice that for each hydrogen molecule that gets oxidized, two electrons are released.

At the oxygen electrode (cathode) of a SOFC:

\[
\frac{1}{2} O_2 + 2e^- \rightarrow O^{2-}
\]

Oxygen ions (O²⁻) travel from the oxygen electrode to the fuel electrode through the electrolyte, while at the fuel electrode (anode) the half-cell reactions are:

\[
H_2 + O^{2-} \rightarrow H_2O + 2e^-
\]
Solid Oxide Fuel-Assisted Fuel Cells

\[ CO + O^{2-} \rightarrow CO_2 + 2e^- \]

The resulting electrons travel through the external electrical circuit to the connected load and then to the oxygen electrode. If the above equations are combined, the following overall equations can be written:

\[ 2H_2 + O_2 \rightarrow 2H_2O \]

\[ 2CO + O_2 \rightarrow 2CO_2 \]

For the SOFC at standard conditions (SATP: temperature of 25°C and pressure of 1 bar) the reversible voltage can be obtained using the Gibbs free energy. The sign of the Gibbs free energy is negative, denoting that the reaction is exothermic, thus the system is releasing heat to the environment. Therefore, the cell voltage at standard conditions is given by:

\[ E_0 = -\frac{\Delta G}{zF} = 1.184 \, V \text{ for water vapor and } E_0 = 1.229 \, V \text{ for liquid water} \]

where

\[ \Delta G = -228.5 \, kJ: \text{Gibbs free energy for gaseous H}_2\text{O and }\]

\[ \Delta G = -237.2 \, kJ: \text{for liquid H}_2\text{O at standard conditions} \]

\[ F: \text{Faraday’s constant} = 96485 \, \text{C/mol e}^- \]

\[ z: \text{electrons freed during reaction/atom} = 2 \]

However, the cell in this case will not be operating under standard conditions, but at temperatures higher than the boiling temperature of water. Thus, the Gibbs free energy of water vapor should be considered, and thus the cell voltage at standard conditions is:

\[ E_0 = +1.18 \, V \] \hspace{1cm} (1)

However, in the case of different temperatures and pressures being used, the Nernst equation is used. The Nernst potential combined with Eq. (1), gives the reversible cell voltage of the fuel cell:

\[ E_{rev} = E_0 + \frac{RT}{zF} \cdot \ln(Q) \] \hspace{1cm} (2)

where

\[ R: \text{Gas constant} = 8.314 \, J/K \cdot mol \]

\[ T: \text{Temperature in K} \]

\[ Q: \text{Thermodynamic reaction quotient} \]

\[ F: \text{Faraday’s constant} = 96485 \, \text{C/mol e}^- \]

\[ z: \text{electrons freed during reaction/atom} \]

The above equation takes into account the temperature of the fuel cell, the number of electrons being released in one reaction, as well as the thermodynamic reaction quotient \( Q \) and adjusts the standard cell voltage for different temperatures and pressures. The reversible cell voltage represents the minimum of electrical demand for water splitting and determines the open-circuit voltage (OCV) of the operating fuel cell. The OCV is the voltage measured at zero current and it is a little lower than the reversible cell voltage. Also, the Nernst potential is the maximum voltage the fuel cell can deliver in theory, without taking into account voltage drops caused by internal resistances, for certain operation parameters.
The amount of gases that are used during the cell operation are denoted by the factor $Q$, the reaction quotient. The thermodynamic reaction quotient can be calculated by taking into account the partial pressures of the fuel cell:

$$Q = \left( \frac{y_{H_2} \cdot y_{O_2}^{1/2}}{y_{H_2O}} \cdot \left( \frac{P_{\text{cell}}}{P_0} \right)^{1/2} \right)$$  \hfill (3)

where

- $y_{H_2O}$: partial pressure of H$_2$O
- $y_{H_2}$: partial pressure of H$_2$
- $y_{O_2}$: partial pressure of O$_2$
- $P_{\text{cell}}$: operating cell pressure
- $P_0$: standard pressure

To summarize, as a result the Nernst equation, the standard cell potential will change depending on the partial pressures of the products and the reactants. Also, it is clear from Equation (2) that the cell voltage will decrease with rising temperature. However, increasing the pressure affects the electrochemical kinetics positively, resulting in higher cell voltage.

1.3 Solid Oxide Electrolyzer Cell (SOEC)

A Solid Oxide Electrolyzer Cell closely related to a SOFC, but instead of generating electricity, it produces hydrogen by using electric energy to split water. A SOEC consists of the same parts as a SOFC, with oxygen ions travelling through the electrolyte, as it is illustrated in Figure 2. In this case though the SOFC cathode is now the SOEC anode (oxidation is taking place), while the SOFC anode is now the SOEC cathode (reduction is taking place).

While the cell is acting as a SOEC, steam is supplied to the cathode. External voltage should be applied between the electrodes, with the oxygen electrode connected to the positive terminal, which should be higher in comparison to the water decomposition potential. The half-cell reaction at the cathode forms as follows:

$$H_2O + 2e^- \rightarrow H_2 + O^{2-}$$

At the oxygen electrode of a SOEC, which now acts as the anode, the half-reaction is:

$$O^{2-} \rightarrow \frac{1}{2} O_2 + 2e^-$$
Solid Oxide Fuel-Assisted Fuel Cells

The overall reaction can be written as:

\[ 2H_2O \rightarrow O_2 + 2H_2 \]

The reversible voltage for the SOEC at standard test conditions can be obtained through the Gibbs free energy. For SOEC operation the value will be the same as in SOFC mode, but with a positive sign, meaning that energy needs to be given to the system in order for the reaction to take place. By taking Faraday’s law into account and assuming water vapor as the reactant, the cell voltage at standard conditions is:

\[ E_0 = -\frac{\Delta G}{2F} = -1.18 \, V \]  \( (4) \)

where

\[ \Delta G = +228.5 \, kJ \]: Gibbs free energy for gaseous H\(_2\)O

The above values mean that in order to electrolyze liquid water in an electrolyzer cell at 25°C, a voltage of at least 1.18 \( V \) is needed to drive the reaction, disregarding irreversible losses.

1.4 Solid Oxide Fuel-Assisted Electrolyzer Cell (SOFEC)

![Figure 3. Simplified illustration of SOFC. [5]](image)

The SOFEC is a device that can be described as a combination of both SOFC and SOEC, as it can oxidize hydrogen as a SOFC and can also electrolyze water as a SOEC. A simplified illustration of a SOFEC can be seen in Figure 3.

As mentioned before, during SOFEC mode the cell operates as an electrolyzer. However, steam is fed to the cathode side, which is the oxygen supplying electrode for this mode. From the anode side, fuel is fed to the cell. Oxygen ions travel through the electrolyte to the fuel electrode, while electrons travel from the fuel to the steam electrode through the external circuit.

The reduction reaction taking place at the SOFEC cathode is:

\[ 4H_2O + 8e^- \rightarrow 4H_2 + 4O^{2-} \]

However, on the fuel side the reactions change depending on the assisting fuel used at the anode.

CO–SOFEC

At the anode where CO gets oxidized, the half-reaction is:

\[ 4CO + 4O^{2-} \rightarrow 4CO_2 + 8e^- \]
Therefore, the total cell reaction is:

**CO–SOFEC:**

\[ CO + H_2O \rightarrow CO_2 + H_2 \]

**CH4–SOFEC**

In the case of CH₄ being used as a fuel, depending on the absence or presence of steam, the reaction can take place in one or more steps [7].

**Direct oxidation**

When SOFEC operates in this mode, methane reacts directly with oxygen ions, having the following half-reaction:

\[ CH_4 + 4O^{2−} \rightarrow 2CO_2 + 2H_2O + 8e^− \]

The total cell reaction in the case of total oxidation of methane is:

\[ CH_4 + 2H_2O \rightarrow CO_2 + 4H_2 \]

**Indirect oxidation**

The partial oxidation of CH₄ is a combination of different reactions:

Step 1: Steam reforming reaction:

\[ CH_4 + H_2O \rightarrow CO + 3H_2 \]

Step 2: Water gas shift reaction:

\[ CO + H_2O \rightarrow CO_2 + H_2 \]

Combining steps 1 and 2 results in the same reaction as in the case of total oxidation of methane:

\[ CH_4 + 2H_2O \rightarrow CO_2 + 4H_2 \]

Step 3: H₂ oxidation:

\[ 4H_2 + 4O^{2−} \rightarrow 4H_2O + 8e^− \]

Combining all three steps of the partial oxidation of CH₄:

\[ CH_4 + 4O^{2−} \rightarrow CO_2 + 2H_2O + 8e^− \]

Therefore, taking into account the half-reaction of the SOFEC cathode, the total cell reaction for a CH₄–SOFEC is exactly the same as if total oxidation occurred:

**CH₄–SOFEC:**

\[ CH_4 + 2H_2O \rightarrow CO_2 + 4H_2 \]

A simple schematic of the reactions taking place in a SOFEC is shown in Figure 4. However, in the case of just hydrogen used as a fuel, the only reaction that occurs at the anode is the hydrogen oxidation \((4H_2 + 4O^{2−} \rightarrow 4H_2O + 8e^−)\), which is illustrated in Figure 3.
As it can be seen, in both of the above reactions (either having just CO as fuel or CH$_4$), H$_2$ is produced at the cathode side, which in the case of SOFEC is the oxygen supplying electrode (steam/H$_2$ electrode), while CO and CH$_4$ are consumed at the anode side, which is the fuel electrode. The main difference between the two fuels is the amount of steam and fuel needed to produce the same amount of hydrogen. In order to do that, four times the amount of CO is needed and double the amount of steam, in comparison to using CH$_4$ as assisting fuel. Also, steam is not necessary for the total oxidation, but it is recommended in order for carbon deposition not to occur. Especially when having reactions with CO$_2$ as a product, there is always a chance of carbon being present on the electrode, which would result in lower cell efficiency. In order to counteract the carbon deposition, steam is added to the anode side, but once that is done it is safer to assume that the partial oxidation of CH$_4$ is going to take place. This is because as steam is added the reformation reaction is more likely to happen [7].

![Figure 4. Illustration of SOFEC. [5]](image)

When methane enters the anode, one can assume that all the reactions happen instantly, which means that the whole amount of CH$_4$ entering turns into CO and H$_2$, and right after that the water gas shift reaction occurs. This is exactly the same reaction that happens in a SOFC when methane and steam are added to the anode, which is called direct internal reforming [5].

### 1.5 Solid Oxide Cell Voltage and Power

![Figure 5. The energy transfer processes of SOEC and SOFEC. [8]](image)

It was mentioned above that SOFEC are similar to SOFC in operational terms, as the current for both modes flows in the same direction. Thus they can be combined in series with the anodes of both cells fed with fuel, while the cathode of the SOFEC being exposed to steam and the cathode of the SOFC to air [8]. On the other hand, SOFEC can be compared to SOEC in terms of results, as they both electrolyze...
water and produce H₂, while a SOFEC consumes much less electricity to produce the same amount of H₂. Hydrogen production in the case of SOFEC is, therefore, less dependent on the electrical power input.

The lowest voltage at which water electrolysis might occur in a Solid Oxide Cell is 1.18 V, as mentioned above. But as this is the minimum value at which that can be achieved, the process will be very slow. In order to increase the reaction speed, thus having more O₂ and H₂ gases produced per unit of time, the voltage should be increased. However, by increasing the applied voltage, a lower amount of H₂ per amount of electrical energy used is expected, lowering the efficiency of the process [5]. Due to the high amount of voltage needed in order to reach the decomposition voltage of water, nearly 80% of the cost of electrolysis comes from the electricity cost [9]. By assisting the electrolysis process using natural gas, a voltage reduction of as much as 1 V (from 1.2 V to 0.2 V) can be achieved compared to conventional steam electrolyzers [3]. By managing to lower the needed voltage, the electrical energy to hydrogen conversion ratio would consequently increase [5], making the price of hydrogen produced in this way closely comparable to the conventional way of industrially reforming methane.

Assuming the assisting fuel in the SOFEC is CH₄, there are three main possible reactions that occur at the cell’s anode, simultaneously, considering that partial oxidation of CH₄ takes place [7]:

Steam reformation: \( CH_4 + H_2O \rightarrow 3H_2 + CO \)

Water gas shift: \( CO + H_2O \rightarrow H_2 + CO_2 \)

H₂ oxidation: \( 4H_2 + 2O_2 \rightarrow 4H_2O \)

At the cathode side only water electrolysis occurs:

\( H_2O \) electrolysis: \( 4H_2O \rightarrow 4H_2 + 2O_2 \)

Because the reformation of methane happens at the anode entrance, all reactions occur instantaneously. Thus, the only reactions that could play a role in calculating the cell voltage are hydrogen oxidation at the anode and water electrolysis at the cathode. However, these two reactions have an equal, but opposite enthalpy and Gibbs free energy, meaning that they cancel each other out energetically. Therefore, the only thing that factors in determining the Nernst potential is the concentration difference between the reactants and products at the anode and cathode. [5]

During the operation of a hydrogen fuel cell, two electrons are released from each H₂ molecule, as explained in Section 1.2, and travel through the external circuit. This means that for one mole of H₂ used, the charge of the electrons that pass through the external circuit is \(-2F\), where \( F \) is the Faraday constant, which indicates the charge of one mole of electrons. Therefore, in a reversible fuel cell, the electrical work that can be obtained by oxidizing 1 mole of fuel is:

\[
W_{electr,rev} = charge \cdot voltage = -2F \cdot E_{rev}
\]

By studying Figure 3 it is possible to assume that in order to calculate the reversible work for SOFEC, the reversible work of SOFC and SOEC could be combined, with the difference that in this case the oxygen partial pressure can be ignored, as only oxygen ions travelling from the anode to the cathode through the electrolyte are present. Thus, the following equation can be derived, which calculates the reversible work done by a SOFEC operating on pure hydrogen [5]:

\[
W_{FC,rev,SOFC} = R \cdot T \cdot \ln \left( \frac{Y_{H_2}(a) \cdot Y_{H_2O}(c)}{Y_{H_2O}(c) \cdot Y_{H_2}(c)} \right)
\]
Solid Oxide Fuel-Assisted Fuel Cells

It is obvious that the reversible work of SOFEC depends only on the partial pressures of hydrogen and steam at the anode and cathode, while the pressure under which the fuel cell operates does not influence the calculation at all [5]. Thus, the reversible cell voltage is influenced by the change of the ratio of \( \text{H}_2\text{O} \) fed to the cathode. There is a certain fuel utilization for each water flow at which the logarithm, and the cell voltage as a result, become zero. The fuel utilization of a fuel cell is defined as the ratio between the fuel flow that reacts and the fuel flow injected to the cell, and shows the amount of fuel that actually reacts. Figure 6 shows how the reversible cell voltage changes for different \( \text{H}_2\text{O} \) molar flows at the cathode, depending on the fuel utilization at \( T = 1273 \text{ K} \).

![Figure 6. \( V_{\text{rev}} \) for changing \( \text{H}_2\text{O} \) flow at the cathode. [5]](image)

By changing the molar flow of water, its utilization will alter as well, so the only way that the utilization of hydrogen at the anode and cathode can be the same is when the molar flows at both sides are equal. For example, for equal molar flows of \( \text{H}_2 \) and \( \text{H}_2\text{O} \) and fuel utilization \( U_f < 0.5 \) the SOFEC will be able to produce electricity, as well as consume electricity. On the other hand, for \( U_f > 0.5 \) the SOFEC is capable of just consuming power. The graph happens to be quite similar to the SOFC, with the addition of the SOFEC being able to produce hydrogen as well.

![Figure 7. Different polarizations within the SOFC. [5]](image)

The polarization curve of a SOFC can be seen in Figure 7. The voltage drop at the middle linear part of the voltage curve is caused by ohmic losses, while activation and concentration losses are responsible for the voltage drops at the beginning and the end respectively.

The next step will be to analyze what happens to the voltages for each cell mode depending on the current density. In Figure 8 the cell voltage of each fuel cell mode can be seen, depending on the current density. The “\( E_{\text{rev}} \) SOFC/SOEC” horizontal line indicates the reversible cell voltage of SOFC and SOEC. It is clear that with increasing current density SOFC has a lower voltage than the reversible cell voltage. On the other hand, SOEC has a higher cell voltage, as it consumes electrical power in order to operate. The case of SOFEC is presented by the dotted line in Figure 8. The reversible cell voltage is positive at low current densities. Increasing the current density results in a lower cell voltage, as fuel...
is being used. At higher current densities the cell voltage becomes negative. A detailed explanation of this is given below, while focusing on Figure 9.

![Figure 8. Cell voltage (based on theoretical V-I curves of SOFC and SOEC) and power density of all three modes. [5]](image)

In the second part of Figure 8 the power consumption or production is shown. The power density of a fuel cell can be calculated by:

\[
P \left( \frac{W}{m^2} \right) = i \left( \frac{W}{m^2} \right) \cdot V
\]

(7)

In order to have the same reference points for voltage and current calculations as in literature, the negative of the voltage source should be the SOEC fuel electrode, while the positive to the SOEC oxygen electrode. In Figure 9 a simple schematic of operation is shown, along with simplified electrical equivalent circuits in order to show the current flow. \( E_A \) is the external voltage applied to the cell, while \( R_a \) is the external resistance of the load.

![Figure 9. A schematic of cell operation in four quadrants. Quadrant I: SOFC, Quadrant II: SOEC, Quadrant III: SOFC, and Quadrant IV: SOFEC. The schematic also shows equivalent circuits and the directions of current flow. [4]](image)
The maximum voltage for a SOFC can be achieved at zero current. This voltage is the open-circuit voltage, which would be the reversible cell voltage if one neglected the internal electronic leakage. On the other hand, the maximum current for SOFC would be achieved when short-circuited, under no load. Voltage and current for SOFC are both positive, as depicted in the first quadrant of Figure 9. Thus, the power density of SOFC is always positive and electricity is produced by the fuel’s chemical energy of oxidation. During SOEC mode the current changes direction, as electrical work is done to the system and hydrogen is produced. The cell voltage is positive and greater than the OCV, while the current is negative, as seen in the second quadrant. It is important to notice that the third quadrant does not correspond to any real operating situation, but is shown in the shake of continuation.

As seen in Figure 9, both SOFEC and SOFC operate in the positive current density region, which is placed on the x-axis of the graph. The operation of SOFEC in particular is depicted in the fourth quadrant of the graph. Concentrating on the line highlighted by the red arrow in the SOFEC mode of Figure 9, when the current density starts rising (fuel is being used), the cell voltage (y-axis of the graph) is positive. At the point where the voltage is zero, the hydrogen and water concentrations at the anode and cathode, respectively, are equal. As the current density increases, the cell voltage becomes negative.

During the SOFEC mode the cell operates as a combination of both SOFC and SOEC, as mentioned before. As a result, its operation is not as straightforward as the first two modes, mainly due to the fact that the voltage can take both positive and negative values depending on the current density region of operation, as seen in Figure 9. The sign of the voltage in combination with the always positive current density of the SOFEC seen in the fourth quadrant of Figure 9, means that both a power input (negative power density) and a power output (positive power density) can be observed during the SOFEC mode.

Three different operating cases of a SOFEC operation can distinguished, depending on the cell voltage:

- $V > 0$
  For low current densities the reversible cell voltage of the SOFEC is positive, which means that the power density is also positive, exactly as in the case of the SOFC. As a result, the SOFEC can generate electricity for positive cell voltage values, as well as produce hydrogen. This indicates that the assisting fuel’s chemical energy is higher than the electrical demand, due to the irreversible losses of the SOFEC not being too large in this current density range. [8]

- $V < 0$
  For higher current densities the reversible cell voltage of the SOFEC is negative, which means that for $V_{OCV} < V < 0$ the power density is negative, while the chemical energy from the assisting fuel becomes lower than the irreversible losses of the SOFEC. As a consequence of the negative power, the cell demands electrical energy in order to split water and in this current density range. Therefore, the SOFEC works as an electrolyzer, producing hydrogen while electrical power is consumed. [8]

- $V = 0$
  At the current density point where the reversible cell voltage of the SOFEC is zero ($V = 0$), the power density is equal to zero as well. This means that, at least theoretically, the SOFEC produces hydrogen while there is no electrical power input or output. [8]

1.6 Solid Oxide Cell Efficiency

In order to define the efficiency of a fuel cell, it would be logical to compare the electrical energy produced by the cell with the heat that would be produced if the fuel was burnt instead. Thus, the
efficiency of a fuel cell can be calculated by [10]:

$$\eta_{\text{fuel cell}} = \frac{\text{electrical energy produced}}{-\Delta \tilde{h}_f}$$ \hspace{1cm} (8)

where $\Delta \tilde{h}_f$ is the calorific value of the fuel, being negative when energy is released. In the case of hydrogen there are two values for $\Delta \tilde{h}_f$, one for the $H_2O$ being steam (lower heating value):

$$H_2 + \frac{1}{2} O_2 \rightarrow H_2O \text{ (steam)}$$

$$\Delta \tilde{h}_f = -241.83 \text{ (LHV)}$$

and one for condensing the resulting $H_2O$ back into liquid (higher heating value):

$$H_2 + \frac{1}{2} O_2 \rightarrow H_2O \text{ (liquid)}$$

$$\Delta \tilde{h}_f = -241.83 \text{ (HHV)}$$

In a totally efficient system the operating cell voltage could be calculated by:

$$E_{HHV} = \frac{-\Delta \tilde{h}_f}{2F} = \frac{-HHV}{2F} = 1.48 \text{ V}$$ \hspace{1cm} (9)

or

$$E_{LHV} = \frac{-\Delta \tilde{h}_f}{2F} = \frac{-LHV}{2F} = 1.25 \text{ V}$$ \hspace{1cm} (10)

Thus, considering the LHV of hydrogen, and with $V_c$ being the actual cell voltage measured, the cell efficiency can be written as:

$$\eta_{\text{FC,LHV}} = \frac{V_c}{E_{LHV}} \cdot 100\%$$ \hspace{1cm} (11)

In practice though, some amount of the fuel fed to the cell does not react. The fuel utilization coefficient is defined by:

$$U_f = \frac{\text{mass of fuel reacted in cell}}{\text{mass of fuel input to cell}}$$ \hspace{1cm} (12)

A common value for $U_f$ is 0.95, which allows for an accurate estimate of the efficiency just by measuring the operating voltage of the cell:

$$\eta_{\text{FC,LHV}} = U_f \cdot \frac{V_c}{E_{LHV}} \cdot 100\%$$ \hspace{1cm} (13)

Waste heat is a common derivative in industrial applications, but it is generally considered a low-quality type of energy. Ignoring the heat demand of the fuel cell system, only chemical energy present in the fuel as well as electrical energy are taken into account in order to calculate the efficiency of the system. Thus, the efficiency of the system during SOEC mode is given by [8]:

Solid Oxide Fuel-Assisted Fuel Cells

D. Papalexis
T.U. Delft

12
Solid Oxide Fuel-Assisted Fuel Cells

\[ \eta_{SOEC} = \frac{Q_{H_2}}{Q_e} = \frac{LHV_{H_2}}{2F \cdot V_c} \]  \hspace{1cm} (14)

where

- \( Q_{H_2} [W/m^2] \): chemical energy in hydrogen,
- \( Q_e [W/m^2] \): electricity,
- \( LHV_{H_2} \): lower heating value of hydrogen,
- \( LHV_{H_2} = -248.3 \text{ kJ/mol} \) for SOFC operating temperatures

By substituting \( E_{LHV} = \frac{LHV_{H_2}}{2F} \), the SOEC efficiency can be re-written as:

\[ \eta_{SOEC} = \frac{E_{LHV}}{V_c} \]  \hspace{1cm} (15)

where

\[ E_{LHV} = \frac{-LHV_{H_2}}{2 \cdot 96485 C/mol} \approx 1.29 V \]

During SOFC mode the voltage of the cell can be either positive or negative, depending on the current supplied to the cell. Thus, the efficiency of the cell is given by a different formula, depending on the sign of the voltage, because for negative voltages the cell produces both hydrogen and electricity, while for positive voltages it produces hydrogen while consuming electricity, as explained in Section 1.5. Therefore, the efficiency of the cell in SOFC mode is [8]:

\[ \eta_{SOFC} = \begin{cases} \frac{Q_e + Q_{H_2}}{Q_{fuel}} = \frac{LHV_{H_2} - 2F \cdot V_c}{LHV_{fuel}}, & V_c < 0 \\ \frac{Q_{H_2}}{Q_{fuel} + Q_e} = \frac{LHV_{H_2}}{LHV_{fuel} + 2F \cdot V_c}, & V_c \geq 0 \end{cases} \]

where

- \( Q_{H_2} [W/m^2] \): chemical energy in the fuel,
- \( Q_{fuel} = \frac{i \cdot LHV_{fuel}}{2F} \)

1.7 Previous experiments on SOFC

Continuing the discussion from Section 1.5, the operation of a SOFC can be shown using experimental data in Figure 10. Tao et al. [4] studied the behavior of a button cell (2 cm² active area) fuel cell under all three modes of operation at 800°C. Hydrogen, humidified methane and syngas were separately used as fuel, with air at the cathode side for SOFC mode and steam at the cathode for SOFC mode. During SOEC mode, humidified H₂ was used at the fuel electrode. For low current densities during SOFC mode the cell voltage is positive, while at some point it becomes negative with increasing current density (quadrants I and IV).

When the SOFC is in the first quadrant (positive power output), it operates as a SOFC, producing electricity and hydrogen, as well, at the steam electrode. The OCV is low (~0.1 V to ~0.2 V), while a current density of about 2 A/cm² was achieved at about ~0.45 V. The same data is also shown mirrored and flipped in the second and third quadrant in order to compare the gains in the external energy demands to the SOEC mode. The experimental results that are going to be presented in later chapters are going to be presented in that way, as well, in order for the comparison between the two modes to be more clear.
In their report Tao et al. [4] also point out that the difference in voltage between the two electrolyzer modes for the same amount of current is about 1.2 V, exactly in line with the initial study from Pham et al [7]. Comparing the SOEC and SOFEC modes, one can notice a difference in slopes. Since the ohmic losses are the same for the two modes, this means that the difference is caused mainly by the lower concentration polarization of the SOFEC mode. In this mode, both the anode and the cathode of the cell are exposed to gases containing H₂ (H₂ at the anode and H₂O at the cathode). Compared to SOFC mode, where the anode is expected to have a lower concentration polarization than the cathode as a result of the low molecular weight H₂ present at the anode, the SOFEC mode has a lower total concentration polarization [4].

![Figure 10. Experimental data on SOFC, SOEC, and SOFEC modes of operation. The SOFEC data, which are in quadrants I and IV, are also plotted in quadrants III and II by a 180° rotation for comparison with the SOEC mode. [4]!](image)

In another study, Luo et al. [8] experimented on a button cell using different anode gases (H₂, CH₄ and CO) at 800°C, in order to compare the energy demands between SOFEC and SOEC modes of operation. An important finding was that for a current density of 0.3 A/cm², SOFEC uses about 80% less electricity, while CO- or CH₄-assisted SOFEC performed better than SOEC, especially when using methane. Particularly, 12% CO-SOFEC and 12% CH₄-SOFEC are about 7% and 30% more efficient than a SOEC at 800°C and current density below 0.25 A/cm². Also, for the same fuel concentration, CH₄-SOFEC boasts higher performance than CO-SOFEC, due to the latter’s higher anode polarization. The above can be seen in Figure 11 and Figure 12.

In tests performed by Wang et al. [11], Pd-CeO₂-YSZ was found to be the material with the highest catalytic activity of the ones tested, while it also showcased high reductions in OCV for the SOEC mode compared to other anode materials, such as Cu-CeO₂ and Cu-Co-CeO₂. Furthermore, a possible choice for the oxygen electrode in the case of SOFC and SOEC would be Sr-doped LaMnO₃ (LSM) + YSZ, but during SOFEC mode the material is unstable under reducing environment, which is the oxygen supplying electrode that acts as the cathode. A possible solution proposed by Tao and Irvine [12] is to replace Mn with Cr (LaSrCrMnO₃, LSCM), which would increase the stability in both reducing and oxidizing environment. In this way LSCM could be used as the SOFEC steam/H₂ electrode and as the oxygen one during SOFC mode.
1.8 Co-electrolysis of carbon dioxide and steam

Co-electrolysis of steam and carbon dioxide can be performed in a Solid Oxide Cell in order to produce hydrocarbon fuels from recycling CO$_2$. If the power supplied to the cell during co-electrolysis derives from renewable sources, the whole process can be sustainable. High temperature electrolysis of CO$_2$ and/or H$_2$O in a SOC can lead to a syngas product (high-energy mixture of CO and H$_2$ gases), which can be later used to synthesize liquid fuel, through the Fischer-Tropsch procedure for example. The whole process is summarized in Figure 13, which illustrates the reactions taking place at the SOC electrodes.

The process of syngas preparation through a WGS reactor with the gas ratio needed for catalytic fuel production can be eliminated by implementing co-electrolysis in the SOEC or SOFEC system. [13]
2 Experimental setup

2.1 Setup outline

The experiments, during which the button solid oxide cells (SOC) were tested, were conducted using the Fiaxell Open Flanges Test Set-Up, which can be seen in Figure 14. The cell is shown as (h) in the picture. Under conventional operation, the bottom tube (o) is used to feed the cell with fuel, which is indicated by the red arrows, while air (yellow arrows) comes from the top pipe (a). For the majority of the experiments done on cells in the duration of this thesis, pipe (a) was used in order to feed the cell with either hydrogen, carbon-monoxide and/or nitrogen, while flange (o) was used to feed the other side with steam, carbon-dioxide, oxygen and/or nitrogen, depending on the kind of experiment conducted. Thus, gases are flowing on each side of the cell, while the excess fuel at the edges of the cell is burnt in the alumina felts (f). A nickel mesh (k) was used for current collection purposes on one side of the cell, which was in contact with the bottom flange (l), as seen in Figure 16, while on the other side the current collection was possible through a gold mesh (g) with two wires welded on it. The nickel mesh was replaced by a gold mesh just for the last part of the experiments, in order to avoid nickel oxidation. A thermocouple was inserted through the small tube (b) till the upper alumina felt (f) in order to have accurate temperature readings near the cell.

![Diagram of experimental setup](image)

*Figure 14. Bottom view of the Open Flanges Test Set-Up: Tubes and flanges (a, o, e & i) are made with Inconel 600 & 601 that gives a full protection against high temperature corrosion.*

2.1.1 Gases Inputs/Output

The gases used during the experiments were fed to the cell through the lab’s gas supply system, which uses mass flow controllers for specific gases such as hydrogen, nitrogen, oxygen, carbon-monoxide and carbon-dioxide. The input value to the controller’s panel is the gas flow in ml/min, which was used
Experimental setup

to calculate the gas percentages fed to the cell’s anode and cathode. The gases were connected to a mutual pipe, which was fed to the anode and cathode of the cell, respectively, though rubber tubes. The output gases from the cell are led to the lab’s ventilation system via rubber pipes connected to the cell’s cathode, as seen in Figure 15.

The steam needed for the electrolysis experiments was provided by a water pump through the inlet seen in Figure 15. The flow of water could be set in ml/min through pump control panel, while the amount of steam that was present at the cell cathode was calculated for 800°C, taking into account the expansion rate of steam at that temperature.

The temperature control of the furnace where the Fiaxell setup rests during the experiments cannot give an accurate reading of the temperature near the cell, thus a thermocouple needs to be used in order to get a more precise measurement. Therefore, before putting the Fiaxell setup in the furnace, a thermocouple has to be connected to the cell’s anode, through pipe (b) seen in Figure 14, in order to get accurate temperature readings from near the core of the setup. There is another thermocouple present in order to get temperature readings from the other side of the cell, as seen in Figure 15.

2.1.2 Electrical Connections

In order for the cell to work both in fuel cell and in electrolysis mode, an external circuit needs to be
connected to it. The cathode of the cell is in electrical contact with the nickel (or gold in later experiments) mesh, which, in turn, is electrically connected to the bottom plate (l) as seen in Figure 14. The voltage is then measured from the thick wire extending from the bottom plate, which is shown in Figure 15, while the current is measured from the blue-coated wire. On the other side of the cell, a gold mesh with two gold wires welded on it is used to get the current and voltage from the cell, each from a different gold wire. Wires with clamps are used to connect all of the above wires to the Gamry Instruments device, which is used to calculate the response of the cell and create the polarization curve for each experiment scenario.

2.2 Setup Preparation

While the Fiaxell Open Flanges Test Set-Up provides proper sealing for most cases with the aid of the aluminum-silicate felts above and below the cell, the circumstances of the experiments that took place during this thesis created the need for some necessary alterations to how the cell is accommodated in the setup.

2.2.1 Nickel mesh

In the configuration used during the experiments, the electrical connection between the cathode and the lower metallic plate of the Fiaxell setup was ensured by the presence of a nickel mesh that was placed in the center of the plate, while the cell was placed on top of the mesh. A photograph of the mesh placement is shown in Figure 16. In this manner, the electrode on this side of the cell is electrically connected to the metallic plate, which is in turn connected with the wires on the top of the setup, as seen in Figure 15 for current collection purposes.

The nickel foam was kept in place using a small amount of liquid glue, which helped make the preparation of the setup easier and when the temperature rises it is evaporated, thus not hindering the gas flow through the mesh. Also a hole of the same diameter as the gas feeding pipe in the center of the plate was created, as well as an X-shaped pattern was cut out to ensure better gas distribution over the entire active area of the cell.

For the latest parts of the experiments, where a smaller diameter cell was used, a gold mesh replaced
Experimental setup

the nickel foam. Because the thickness of the gold mesh used was 1mm and that of the mica layer around it was 3mm, three identical gold meshes, one on top of the other, needed to be used in order for the mesh to have the same thickness as the surrounding mica sealing, as seen in Figure 17. Again, the gold mesh was kept stable on the metallic plate by the use of liquid glue.

2.2.2 Mica sealing

Except for the aluminum felt used as sealing, an extra mica layer is used around the nickel foam, as seen in Figure 16. The layer is cut in a circular ring fashion and placed around the nickel mesh in order to reduce the amount of gas that is lost along the surface of the metallic plate. During the preparation of the cells, it was found that the ideal width of the difference between the inner and outer circular ring diameters should be at least 5 mm in order for the cell to be properly sealed.

![Figure 17. Setup preparation: Three gold meshes were used instead of nickel, one on top of the other.](image)

As for the thickness of the mica layer itself, it should be the same as the nickel mesh that sits inside it. This is because the cell is going to be placed on top of both the nickel mesh and the mica sealing, so their thickness should be similar in order for the cell not to bend, and eventually break, when pressure is applied to it. During these sets of experiments, a mica layer of about 3mm thickness was used.

After each experiment was conducted the thickness of the mica layer was measured again in order to observe how it may change after being under pressure and high temperature. However, no notable change was observed, except for the fact that the layer became more fragile upon removing.

2.2.3 Cell Accommodation

The button solid oxide cell itself sits on the nickel mesh and the mica layer. Both the thickness of the mesh and mica are the same, so that when the cell rests on top of them it does not bend and eventually break when pressure is applied on it. The electrode of the cell (cathode in this case) is electrically connected to the nickel mesh, while the other side (anode) is where the gold mesh is going to be placed on for current collection purposes.
The cell is kept in place with four transparent tapes, to ensure that it will not move during the setup preparation, as seen in Figure 18. The tape is not touching the electrode, so the gold mesh on top is in full contact with the anode.

### 2.2.4 Aluminum-silicate felt

Usually the aluminum felts are hosted above and below the cell, between the plates of the Fiaxell setup. In the setup configuration used during these experiments, the nickel mesh was placed directly on the bottom plate (l), shown in Figure 14. Therefore, in order to achieve the electrical connection between the plate and the nickel mesh, a circular hole in the lower aluminum felt was created, approximately the same diameter as the cell’s top electrode (anode in this case) to ensure that the fold mesh is in contact with the electrode and that the gold wires are in contact just with the aluminum felt, as seen in Figure 19. Thus, an extra set of sealing around the cell is needed, in order to ensure that there as little gas leaking as possible, shown in Figure 16. After placing the gold mesh on top of the electrode, the circular part removed is put back again on top of the gold mesh, in order to provide extra stability and sealing. A small hole is cut through in order for gases to reach the cell, as seen in Figure 20. On top of these, the second, upper aluminum felt is placed, providing extra sealing and electrical insulation for the gold wires. The result can be seen in Figure 21.

### 2.2.5 Gold mesh and wires

A gold mesh was used throughout the experiments in order to collect the current from the anode of the cell. Two gold wires were welded to it using the lab’s welding machine. The wires were led through two different holes of a ceramic tube that extended to the upper part of the setup, when the latter rests in the ceramic furnace, clearly seen in Figure 21. The gold mesh and wires were placed over the aluminum felt, with the mesh being electrically connected to the electrode though the circular hole cut in the felt. It was then pressed down with the remaining part of the felt to ensure that it would stay in place during the setup preparation, as seen in Figure 20.
Experimental setup

Figure 19. Setup preparation: Bottom aluminum-silicate felt on top of the cell. The hole is approximately the same diameter as the cell’s electrode (anode in this case). The hole in the center is present in order to allow the gases to pass through to the cell.

Figure 20. Setup preparation: Gold mesh with two gold wires welded to it. The part of the aluminum felt that was removed is placed on top of the gold mesh to provide extra stability and sealing. The hole in the center is present in order to allow the gases to pass through to the cell.
Figure 21. Setup preparation: The top aluminum felt is placed on top of the cell and the bottom felt. The hole in the center is present in order to allow the gases to pass through to the cell.

Figure 22. Setup closed, with the cell inside, and ready to be put in the furnace, with the anode thermocouple wire being also visible in the picture. The gold wires used for the anode current collection can be seen as well, running through two of the four holes of the white ceramic pipe.
Experimental setup

2.3 Ceramic furnace

The ceramic furnace used during the experiments is the Kittec Furnace for the Open-Flange setup, which can reach temperatures up to 1320°C. The Kittec furnace is designed by Fiaxell specifically to host the Open Flanges Test Set-Up [14]. In Figure 23 two pictures of the furnace can be seen, one of the exterior, with the test rig inserted during an experiment and one showing the interior after the test rig has been removed.

![Figure 23. Kittec furnace - Left: Test rig is inserted / Right: Furnace interior after an experiment.](image)

2.3.1 Temperature control

In order to control the temperature of the furnace, a Bentrup 505 controller connected to the oven was used. The controller provided programmable temperature control with two heating ramps. Thus, the desired temperature could be reached in two steps, providing the temperature rise rate, the end temperature and the duration that the furnace maintains the desired temperature for each step.

The furnace is capable of reaching the operation temperature in about 2 to 3 hours, but when the initial experiments were conducted the cell most of the times broke, probably due to the high temperature rise rate, meaning that the cell could no longer be used for experiments. Therefore, and in order to assure that the cell would remain intact while heating, the heating up procedure took place overnight, with a rise of 50°C per hour. In this manner no problems occurred with the cells tested and experiments could be conducted on them as usual.

2.4 Analysis equipment

The purpose of the experiments conducted during this thesis was to compare the performance of fuel-assisted electrolysis to conventional high temperature electrolysis using a solid oxide cell. The main way to achieve that was to compare the polarization curves for each cell derived from each mode, thus electrical equipment was a major part of the whole process. In order to collect the voltage and current from the cell under operating conditions, a combination of both hardware and software equipment needs to be used. For the polarization curves of all three SOC modes to be drawn, an
Experimental setup

An instrument that acts both as a power supply and a load should be used. The ideal solution for the kinds of cells tested during this thesis proved to be a potentiostat from Gamry Instruments, as far as hardware is concerned, while the accompanying software was used for monitoring.

![Figure 24. Reference 600+ potentiostat - Left: Front view / Right: Back view. [15]](image)

Due to the fact that the cell sizes tested were between 20mm to 25mm in diameter, the amount of current that could be drawn from the cell was a few hundred mA, meaning that the power supply equipment of the lab could not be used, as they could only sense higher currents. The potentiostat used during these experiments was the Reference 600+ from Gamry Instruments, shown in Figure 24. This particular instrument is a high-performance, research-grade potentiostat/galvanostat/ZRA designed for fast, low-current measurements. The current flow between the working and the counter electrode is the measured variable by the potentiostat, with the controlled variable being the cell potential [15].

During the experiments, the recording of the majority of the data was accomplished through the accompanying software made by Gamry Instruments which worked in conjunction with the Reference 600+ potentiostat. Cell polarization could be initialized through the dedicated software, which offers real-time monitoring of the process. The Echem Analyst software, also accompanying the potentiostat, was used for comparison purposes between the polarization curves recorded and it provided an easy way of identifying if the cell was stabilized under new gas input flows.

### 2.5 Future setup modifications

#### 2.5.1 Fiaxell Open-Flanges Set-Up modifications

While the Fiaxell setup proved to be reliable and easy to use for the kinds of tests that were performed during this thesis, there are some parts of it that could be improved for future experiments. Specifically, the setup provided two outlets for gases coming through one side of the cell (cathode electrode in this particular case), but lacked a proper outlet pipe for exiting anode gases. The two small pipes next to the inlet pipe, as seen in Figure 22 (one of them hosts the anode thermocouple), can be used as an exhaust for anode gases, but the pipes themselves are short and there is no access to them during the cell operation, as this part of the setup rests inside the furnace. Therefore, there is no way of collecting the gases coming out of these pipes, meaning that a composition analysis of the outlet gases is not possible for this side of the cell.
Experimental setup

For the above reasons two distinct ways of incorporating a pipe for the exiting anode gases were created in SolidWorks. Both ways were designed with the current setup in mind, therefore only small adjustments were done to the upper plate seen in Figure 22, as the rest of the setup and connections should remain the same. The main idea behind the design of the two 3D models was to modify the already present inlet pipe for the cathode gases, which is the thick one in the figure, in order to incorporate the outlet pipe next to it, in the first model, or around it, in the second one.

The resulting models are illustrated in Figure 25. On the left side of the image the solution featuring an external outlet pipe next to the gas inlet can be seen. A closer look at this design’s modifications is visible in Figure 26. In order for both to fit through the already present hole that hosts the gas inlet, the latter was redesigned so that it is smaller in diameter, while the exhaust pipe for the anode gases was placed alongside it, all the way to the upper part of the setup when it rests in the oven. The gases are fed to the anode through the now narrower inlet pipe and the exiting gases are led out of the closed setup through the second pipe.

The second design can be seen on the right side of Figure 25 and in a close-up view in Figure 28. The 3D model consists of two concentric pipes, with the inner one being the anode gas inlet, while the outer one is the gas outlet. The inner pipe has a smaller diameter compared to the original design, while the external one is of the same diameter as the current inlet pipe. In this way, the inlet gasses are fed to the anode through the inner pipe, with the exhaust being the outer one, that leads the exiting gasses out of the closed setup. A more detailed illustration of both designs can be found in the Appendix.
2.5.2 10-by-10 ceramic test setup modifications

One more modification was designed for another setup which, while not tested during this thesis, can be used in order to conduct experiments on bigger diameter cells in the future. The 3D model shown in Figure 28 was also created in SolidWorks and depicts the modified top part of the 10x10 ceramic setup. The purpose, this time as well, was to create an outlet opening for gas collection. Due to the ceramic material used for the part, as well as for the other gas input or output components connected to it, the place where this connection happens was created so that the ceramic pipes stay in place, screwed to the main part. In this manner the risk of the components being broken during installation or removal is lower. A more detailed illustration of the modified part is presented in the Appendix.

Figure 27. Design with outlet pipe around the inlet pipe. Left: Bottom view. Right: Side view.

Figure 28. Illustration of the modified top part of the 10x10 ceramic setup. Left: 3D view of the model. Right: Top view with the internal openings being visible.
3 Experimental Results

3.1 Introduction to the experiments

The operation of Solid Oxide Cells (SOC) was tested under different operating conditions and modes throughout the duration of this thesis. The main incentive behind the experiments conducted in a lab environment, though, was to compare the two electrolyzer modes of a high temperature SOC, namely the Solid Oxide Electrolyzer Cell (SOEC) mode and the Solid Oxide Fuel-assisted Electrolyzer Cell (SOFEC) mode. As explained in Section 1.4, the use of assisting fuels at the anode lowers the electrochemical cell potential, resulting in increased overall efficiency of the high temperature electrolysis process.

The principal behind the SOFEC mode of operation is to use a cheap fuel in order to replace part of the electricity demand for electrolysis, which is a high quality and expensive energy. Fuels such as carbon monoxide or methane could be used as reducing gases at the SOFEC anode for that purpose. During the experiments in SOFEC mode two gases were used as fuels, hydrogen and carbon monoxide, while the chemical reactions that take place on both anode and cathode were presented in Section 1.4. Obviously, using hydrogen as a fuel in order to produce hydrogen through electrolysis is not practical, but it is used as a base for the comparison between different operating modes, as well as to compare with data obtained from the use of CO as a fuel.

The setup used for the entirety of the experiments was the Fiaxell Open Flanges Test Set-Up, which was introduced in Section 2.1. While many cells where tested throughout the duration of this thesis, it was decided that the most presentable results would occur when symmetrical cells were used, as both sides could act as anode and cathode, depending on the operating mode. Therefore, two kinds of cells, codenamed custom and cermet cells, respectively, and the results obtained from them are illustrated in Sections 3.2 and 3.3, while all of the experiments presented in this thesis were conducted at a temperature of 800°C.

3.2 Platinum pasted electrolyte substrate

The next set of experiments was carried out in the Fiaxel setup, using a custom-made 3cm$^2$ button cell. The cell consisted of an electrolyte substrate, while a thin layer of platinum was pasted on both sides of the cell. The advantage of having a symmetrical cell is that there is no need to swap the electrodes each time there is a change in operating mode.

During the course of these experiments, a total of four cells where used, each consisting of an identical electrolyte substrate. The thickness, and thus the amount, of the platinum paste used was mainly kept at the same level across all four cells, a procedure that proved to be quite challenging, taking into consideration that the material was pasted using a small paintbrush. This particular set of experiments was carried out at a temperature of 800°C.

The purpose of these experiments was to test the efficiency of these custom-made cells under different operating modes, assess the durability and their consequent degradation after the end of the experiments. In order to achieve the above, the cells were tested under three operating modes: Fuel Cell (SOFC mode), Electrolysis (SOEC mode) and Fuel-Assisted Electrolysis (SOFEC mode). The main gases used as a fuel were H$_2$ and CO, while different compositions of each were fed to the cell. Other gases used during the experiment were H$_2$O, N$_2$ and CO$_2$.

During the experiments the Gamry hardware was used to set the voltage or current, while the Gamry software was used in order to monitor the results in real-time, for different operating modes and fuel compositions.
Specifically, when operating in SOFC mode, the cathode was always fed with air (20% O₂ and 80% N₂). On the other side, that of the anode, different compositions of H₂ where used. During SOEC or SOFEC mode, 20% and 50% water compositions where tested at the cathode side, while nitrogen was supplied as well in order to keep the gas compositions at the desired levels. CO₂ was also fed to the cathode for some of the experiments during SOEC and SOFEC mode, to act as an additional oxygen supplier. Furthermore, on the anode side of SOFEC mode different amounts of either H₂, CO or a combination of the two were used, adding N₂ in cases where certain gas composition ratios were needed. During SOEC mode just nitrogen was used at the anode.

The graphs that are going to be presented below are polarization curves (I – V curves) derived from different operating modes of the cells. In order for the comparison between the graphs to be easier, some clarification is needed beforehand. Due to the conventions adopted for easier polarization curve comparison reasons, the voltage signs are swapped from what was originally explained in Section 1.5. For all three modes the values of the current are presented as positive in the graphs, while in reality the current flow is opposite between electrolysis and fuel cell modes. This means that for positive voltage values the cell is producing electricity in SOFC mode, while it is consuming electricity in SOEC mode in order to electrolyze steam. Also during SOFEC mode, positive voltage values suggest that the cell is consuming electricity to electrolyze water, while negative values mean that the cell is generating electricity, as well as electrolyzing steam and, thus, producing hydrogen.

3.2.1 SOFEC and SOEC modes experimental results comparison

Taking into consideration that the cells were custom-made, stability could not be guaranteed for the whole set of cells used. Thus, while all four cells tested gave encouraging results, only graphs derived from Cell 1 and Cell 4 are going to be presented below. This choice was made solely on the stability that those two cells showcased, making them the most suitable candidates for presenting results from.

During the experiments with Cell 1, only hydrogen was used as a fuel at the anode, while just steam was fed to the cathode in SOFEC mode. During SOFC mode air was used at the cathode side. In Table 2 and Table 3 the exact gas flow rates used in SOEC and SOFEC modes are shown respectively, while the gas flow rates used during SOFC mode can be seen in Table 1.

<table>
<thead>
<tr>
<th>Anode (mL/min)</th>
<th>H₂</th>
<th>N₂</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>100% H₂</td>
<td>100</td>
<td>-</td>
<td>100</td>
</tr>
<tr>
<td>50% H₂</td>
<td>100</td>
<td>100</td>
<td>200</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Cathode (mL/min)</th>
<th>O₂</th>
<th>N₂</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>100% air</td>
<td>40</td>
<td>160</td>
<td>200</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Anode (mL/min)</th>
<th>H₂</th>
<th>N₂</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>100% N₂</td>
<td>-</td>
<td>200</td>
<td>200</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Cathode (mL/min)</th>
<th>H₂O</th>
<th>N₂</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>20% H₂O</td>
<td>40</td>
<td>160</td>
<td>200</td>
</tr>
<tr>
<td>50% H₂O</td>
<td>160</td>
<td>160</td>
<td>320</td>
</tr>
</tbody>
</table>
**Table 3.** Cell 1: Gas flow rates used during the experiments in SOFEC mode, both for the anode and the cathode of the cell

<table>
<thead>
<tr>
<th></th>
<th>Anode (mL/min)</th>
<th>Cathode (mL/min)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>H₂</td>
<td>N₂</td>
</tr>
<tr>
<td>100% H₂</td>
<td>100</td>
<td>-</td>
</tr>
<tr>
<td>50% H₂</td>
<td>100</td>
<td>100</td>
</tr>
</tbody>
</table>

The polarization curves of 100% and 50% of H₂ gas fed to the cathode can be seen in Figure 29. Using pure hydrogen gas as a fuel at the anode in SOFC mode results in higher voltage output at the same current density, compared to feeding the cathode with 50% H₂. Therefore, this suggests that when the hydrogen content in the gas fed to the anode rises, the cell produces more electrical power, which translates into higher cell performance.

The same compositions were used at the anode in SOFEC mode, using 20% of steam at the cathode side. A comparison between the two hydrogen compositions is shown in Figure 30. In the case of SOFEC, a lower voltage for the same current density means that the power needed in order for the cell to electrolyze steam is lower as well. Therefore, when a gas with a richer hydrogen content is used during SOFEC mode a higher cell performance is achieved. Interestingly, for low current densities the voltage is negative, which means that the cell is electrolyzing steam, thus producing hydrogen as well as generating energy, as explained in Section 1.5. So in both SOFC and SOFEC modes, higher hydrogen content in the anode gas results in better cell performance.
Finally, a direct comparison between SOEC and SOFEC modes is shown in Figure 31. In this graph the comparison includes data from Cell 4 as well, in order to observe the difference in performance, not only between different modes, but also between different cells. Mind the difference between the polarization curves of each cell, which is a result of them being custom-made and, thus, not having the exact same characteristics, e.g. the resistance, which would result in a different I-V curve shape and also limit the current that could be drawn.
Experimental Results

The exact gas compositions used on Cell 4 can be seen in Table 4 and Table 5. The same polarization pattern can be observed for both cells, as the voltage needed in order to electrolyze steam is less during SOFEC mode.

Specifically, the highest difference in voltage observed for the same current density was 1.11 V, and was recorded for Cell 4. The drop in voltage observed between the two modes for Cell 4 is in line with the values reported in literature \(1.0\ V < \Delta V < 1.2\ V\) [7]. This translates in about 66% drop in electrical power consumption:

\[
power\ consumption\ saving\ (%) = \frac{P_{SOEC} - P_{SOFEC}}{P_{SOEC}} \cdot 100% = \frac{V_{SOEC} \cdot I - V_{SOFEC} \cdot I}{V_{SOEC} \cdot I} \cdot 100% = \frac{V_{SOEC} - V_{SOFEC}}{V_{SOEC}} \cdot 100% = \frac{1.69\ V - 0.58\ V}{1.69\ V} \cdot 100% = 65.68%\]

The drop in power consumption appears to be lower than the 80% reduction that other previous experiments showcased [8], but considering the cells used for this part of the experiments were custom-made, the results can be considered as quite encouraging.

3.2.2 Carbon dioxide as an extra oxygen source for electrolysis

The main focus for the experiments done on Cell 4 was to implement CO\(_2\) alongside steam at the cathode for both SOEC and SOFEC modes. The basic principal behind that was to drive more oxygen ions through the electrolyte, not only by splitting H\(_2\)O into H\(_2\) and O\(^2\)\(^-\), but also CO\(_2\) into CO and O\(^2\)\(^-\). The process is known as CO\(_2\)/H\(_2\)O co-electrolysis, as explained in 1.8, and is mainly a technique used in order to convert CO\(_2\) into hydrocarbon fuels [13].

Except for using CO\(_2\) at the cathode during the course of the experiments on Cell 4, CO gas was also used as a fuel at the anode, both pure and mixed with hydrogen gas at various compositions in SOFEC mode, though the results are presented in the Section 3.2.3. Furthermore, measurements were also done on Cell 4 in order to verify the results that derived from the experiments conducted on Cell 2 and Cell 3.

Table 4. Cell 4: Gas flow rates used during the experiments in SOEC mode, both for the anode and the cathode of the cell.

<table>
<thead>
<tr>
<th>Anode (mL/min)</th>
<th>H(_2)</th>
<th>CO</th>
<th>N(_2)</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>100% N(_2)</td>
<td>-</td>
<td>-</td>
<td>200</td>
<td>200</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Cathode (mL/min)</th>
<th>H(_2)O</th>
<th>CO(_2)</th>
<th>N(_2)</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>50% H(_2)O - 0% CO(_2)</td>
<td>125</td>
<td>-</td>
<td>125</td>
<td>250</td>
</tr>
<tr>
<td>50% H(_2)O - 50% CO(_2)</td>
<td>125</td>
<td>125</td>
<td>-</td>
<td>250</td>
</tr>
</tbody>
</table>

Table 5. Cell 4: Gas flow rates used during the experiments in SOFEC mode, both for the anode and the cathode of the cell.

<table>
<thead>
<tr>
<th>Anode (mL/min)</th>
<th>H(_2)</th>
<th>CO</th>
<th>N(_2)</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>100% H(_2)</td>
<td>100</td>
<td>-</td>
<td>-</td>
<td>100</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Cathode (mL/min)</th>
<th>H(_2)O</th>
<th>CO(_2)</th>
<th>N(_2)</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>50% H(_2)O - 0% CO(_2)</td>
<td>125</td>
<td>-</td>
<td>125</td>
<td>250</td>
</tr>
<tr>
<td>50% H(_2)O - 50% CO(_2)</td>
<td>125</td>
<td>125</td>
<td>-</td>
<td>250</td>
</tr>
</tbody>
</table>
Experimental Results

Table 4 shows the compositions of the gases used at the anode and the cathode of Cell 4 during the SOEC experiments, while Table 5 shows the compositions for SOFEC mode. Nitrogen was used in order to keep the gas composition ratio at the desired level.

In Figure 32 a comparison between steam electrolysis (50% H₂O at the cathode) and CO₂/H₂O co-electrolysis (50% H₂O and 50% CO₂ at the cathode) is shown. A similar comparison is presented in Figure 33, but in this case between fuel assisted steam electrolysis (100% H₂ at the anode and 50% H₂O at the cathode) and fuel assisted CO₂/H₂O co-electrolysis (100% H₂ at the anode, 50% H₂O and 50% CO₂ at the cathode).

**Figure 32.** Cell 4 – SOEC mode: Polarization curve comparison between 0% CO₂ - 50% H₂O (steam electrolysis) and 50% CO₂ - 50% H₂O (CO₂/H₂O co-electrolysis) at the cathode side of the cell. The anode side was fed with N₂.

**Figure 33.** Cell 4 – SOFEC mode: Polarization curve comparison between 0% CO₂ - 50% H₂O (fuel assisted steam electrolysis) and 50% CO₂ - 50% H₂O (fuel assisted CO₂/H₂O co-electrolysis) at the cathode side of the cell. In both cases the anode side was fed with 100% H₂ as a fuel.
In both cases the results follow the same trend, as when CO₂ was used during SOEC and SOFEC modes the voltage needed for electrolysis was higher. At the point shown in Figure 32, this translates into a mere 6% rise in electricity consumption when CO₂ is being used in combination with steam for co-electrolysis purposes during SOEC mode. On the other hand, during SOFEC mode seen in Figure 33, the rise in power consumption was higher, measuring a 13% increase in electricity demand when CO₂ was used.

Therefore, more power was consumed when CO₂ was present in the cathode gas. In other words, the cell showcased a slightly better performance with just steam at the cathode, rather than with the inclusion of CO₂ gas, meaning that the results are in line with reported data from similar co-electrolysis experiments [13].

Interestingly for both modes, the open-circuit voltage (OCV) is nearly equivalent when either just steam or a mixture of steam and carbon dioxide was used at the cathode, as the disassociation energy of both CO₂ and H₂O (to produce CO and H₂, respectively) is almost identical [13].

3.2.3 Carbon monoxide as a fuel in SOFEC mode

During the experiments done in this section, CO was used as a fuel at the anode, either pure or in combination with hydrogen, while on the cathode side just steam was used. The main objective for these set of experiments was to compare the performance of the cell under different fuel compositions, varying from pure hydrogen to pure carbon monoxide. Due to the chance of carbon passivating the anode, experiments using 100% H₂ gas were conducted before using CO as well, with the exception of the comparison in Figure 40, where the temporal passivation of the cell was tested. The exact gas compositions used during this set of experiments can be seen in Table 6.

| Table 6. Cell 4: Gas flow rates used during the experiments in SOFEC mode, both for the anode and the cathode of the cell. |
|-----------------|-----------------|-----------------|-----------------|-----------------|
| Anode (mL/min)  | H₂              | CO              | N₂              | Total           |
| 100% H₂         | 100             | -               | -               | 100             |
| 100% CO         | -               | 100             | -               | 100             |
| 50% H₂ - 50% CO | 50              | 50              | -               | 100             |

<table>
<thead>
<tr>
<th>Cathode (mL/min)</th>
<th>H₂O</th>
<th>CO₂</th>
<th>N₂</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>50% H₂O</td>
<td>125</td>
<td>-</td>
<td>125</td>
<td>250</td>
</tr>
</tbody>
</table>

In Figure 34 a comparison between using pure CO and H₂, as well as a mixture of the two can be seen, as far as the resulting polarization curve is concerned. The first notable point is that when CO is present in the fuel gas the OCV is more negative. This observation is also documented in the results of experiments performed in literature [8]. At the same region the resistance of the cell is the same for all cases, as the slopes of the I – V curves are identical. For high current densities though, the performance of the cell is hindered by the amount of CO added to the gas fed to the anode, as seen in Figure 34. At the high current density section of the graph, increasing the ratio of CO/H₂ in the gas results in higher voltage difference in respect to pure H₂, for the same current density. This suggests that the performance of the cell declines as the CO content in the gas fed to the anode rises.
Experimental Results

Figure 34. Cell 4: Polarization curve comparison between 100% CO, 50% H2 - 50% CO and 100% H2 at the anode side in Fuel Assisted Electrolysis mode. For all three cases 50% H2O was fed to the cell’s cathode side.

Furthermore, by comparing the power demand difference between the two gases, the SOFC which uses carbon monoxide as a fuel appears to demand more power when compared to the SOFC operation that uses pure hydrogen at the anode. Specifically, at the point shown in Figure 34, the CO-SOFC exhibits a nearly 30% increase in power demand:

\[
\text{power demand increase (\%) } = \frac{P_{\text{CO-SOFC}} - P_{\text{H2-SOFC}}}{P_{\text{CO-SOFC}}} \cdot 100% = \frac{V_{\text{CO-SOFC}} - V_{\text{H2-SOFC}}}{V_{\text{CO-SOFC}}} \cdot 100% = \frac{0.98 V - 0.70 V}{0.98 V} \cdot 100% = 28.5% 
\]

3.3 Cermet cathode symmetrical button cell

The final part of the experiments was conducted on a cermet cell provided by Cambridge University. The cermet cell used was made specifically for the present experiments and it is not commercially available. This cell is actually a LSM symmetrical cell with a scandia stabilized ceria electrolyte (ScCeSZ), with the main difference with the ones used before being that it was manufactured using machinery for electrode printing on the electrolyte. LSM cathode cells can be well used for SOFCs that operate in the range of 600°C to 850°C [16]. In a reducing atmosphere during SOFC mode, such an electrode can exhibit instability [4]. However, partial substitution of Mn with Cr (LSCM) increases the stability in both reducing and oxidizing environments, meaning that such a material can be used as the cathode for both SOFC (oxygen electrode) and SOFC (steam/H₂ electrode) modes [4]. However, due to time constraints it was not possible to acquire such a cell. Therefore, the choice of using the already available cermet cell (LSM) was made, despite the admitted limitations. The main experiments performed with the cermet cell were for the SOFC mode. Nonetheless, and primarily out of curiosity, additional experiments were conducted in order to examine how the cell performed in SOEC mode, even though the limitations were known.

The purpose of using such a cell was to validate the results obtained during the experiments conducted on the custom-made cells, but also investigate how the cell degrades after 3 days of usage. The cermet cell used was smaller in size, as it had a diameter of 20mm compared to the 25mm of the custom-made cells. Another variation was that the electrode surface was of square shape, in contrast
Experimental Results

to the circular shape of the custom-made cell. As a result, the active area of the cell was also smaller, measured at 1.32cm². On the other hand, the operating conditions during the experiments were the same as before, including the gas flow compositions and the temperature of 800°C, in order for the results to be as comparable with the previous ones as possible. Overall, the cell displays a similar behavior under the same gas compositions for both modes, as analyzed throughout Section 3.3.

3.3.1 Comparison between SOEC and SOFEC modes

Initially, a comparison between the SOEC and SOFEC mode is illustrated in Figure 35. The gas flows used during this particular set of experiments is shown in Table 7 and Table 8 for SOEC and SOFEC mode, respectively. For the SOFEC mode in particular, 100% of hydrogen gas was fed to the anode side of the cell.

Table 7. Cermet cell: Gas flow rates used during the experiments in SOEC mode, both for the anode and the cathode

<table>
<thead>
<tr>
<th>Anode (mL/min)</th>
<th>H₂</th>
<th>N₂</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>100% N₂</td>
<td>-</td>
<td>200</td>
<td>200</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Cathode (mL/min)</th>
<th>H₂O</th>
<th>N₂</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>50% H₂O</td>
<td>160</td>
<td>160</td>
<td>320</td>
</tr>
</tbody>
</table>

Table 8. Cermet cell: Gas flow rates used during the experiments in SOFEC mode, both for the anode and the cathode

<table>
<thead>
<tr>
<th>Anode (mL/min)</th>
<th>H₂</th>
<th>N₂</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>100% H₂</td>
<td>100</td>
<td>-</td>
<td>100</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Cathode (mL/min)</th>
<th>H₂O</th>
<th>N₂</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>50% H₂O</td>
<td>160</td>
<td>160</td>
<td>320</td>
</tr>
</tbody>
</table>

Figure 35. Cermet cell: SOFEC vs SOEC mode polarization curve comparison. 100% H₂ was used at the anode in SOFEC mode and 50% H₂O at the cathode in both modes.

In the same manner as with the custom-made button cells, the SOFEC mode appears to perform better...
than the SOEC mode, as the voltage for the same current density is significantly lower when fuel is assisting the electrolysis process. For lower current densities the voltage is negative, as the cell is producing electricity as well as hydrogen. The main difference between the previous results is that the maximum voltage difference between the two modes for the same current density is 0.89 V, while for rising current density the difference is decreasing, with it being about 0.69 V at the point shown on the graph in Figure 35.

Another important note is that more current was able to be drawn from this cell, in some cases even 10 times more, when compared with the previously tested custom cells. This was despite the smaller active cell area of the cermet cell, showcasing the importance of a more technical manufacturing procedure for a solid oxide cell, which allows for higher currents to be drawn.

3.3.2 Carbon monoxide as assisting fuel for electrolysis

The next set of experiments was performed during SOFEC mode with CO used as a fuel, while the results are presented in comparison with those obtained when pure hydrogen was used at the anode side of the cell, illustrated in Figure 36. The exact gas compositions used are presented in Table 9.

| Table 9. Cermet cell: Gas flow rates used during the experiments in SOFEC mode, both for the anode and the cathode. |
|---|---|---|---|---|
| | Anode (mL/min) | | | |
| | H₂ | CO | N₂ | Total |
| 100% H₂ | 100 | - | - | 100 |
| 100% CO | - | 100 | - | 100 |
| | Cathode (mL/min) | | | |
| | H₂O | CO₂ | N₂ | Total |
| 50% H₂O | 160 | - | 160 | 320 |

In a similar fashion to the tests conducted on the custom cells, the cermet one underperforms when 100% CO is used instead of 100% H₂, with the voltage for almost the whole range of current densities being higher when carbon monoxide is used. The exception is again observed for lower current densities, where the OCV of the CO-SOFEC is more negative than the one using H₂ as a fuel, with the first showcasing a better initial performance, but as the current density increases, the power demand rises higher than that of the H₂-SOFEC. The same trend was recorded for the custom cell, with the
Experimental Results

results being in line with findings from previous experiments in literature [8] [13].

Furthermore, at the point marked on Figure 36 the power consumption when pure CO is used as a fuel is 32% higher compared to the H₂-SOFEC, which is close to the 29% recorded between the same compositions used on the custom cells.

3.3.3 Steam and carbon dioxide co-electrolysis

For the next set of experiments carbon-dioxide was used as an extra oxygen source for electrolysis. The exact gas flows used for this part can be seen in Table 10 and Table 11 for SOEC and SOFEC modes, respectively. The differences between the different compositions were subtle for this particular cell, therefore an extra composition of 100% CO was used as well and the results were examined.

Table 10. Cermet cell: Gas flow rates used during the experiments in SOEC mode, both for the anode and the cathode.

<table>
<thead>
<tr>
<th></th>
<th>Anode (mL/min)</th>
<th>Cathode (mL/min)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>H₂</td>
<td>CO</td>
</tr>
<tr>
<td>100% N₂</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th>H₂O</th>
<th>CO₂</th>
<th>N₂</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>50% H₂O</td>
<td>160</td>
<td>-</td>
<td>160</td>
<td>320</td>
</tr>
<tr>
<td>50% H₂O - 50% CO₂</td>
<td>125</td>
<td>125</td>
<td>-</td>
<td>250</td>
</tr>
<tr>
<td>100% CO₂</td>
<td>-</td>
<td>160</td>
<td>-</td>
<td>160</td>
</tr>
</tbody>
</table>

Table 11. Cermet cell: Gas flow rates used during the experiments in SOFEC mode, both for the anode and the cathode.

<table>
<thead>
<tr>
<th></th>
<th>Anode (mL/min)</th>
<th>Cathode (mL/min)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>H₂</td>
<td>CO</td>
</tr>
<tr>
<td>100% H₂</td>
<td>100</td>
<td>-</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th>H₂O</th>
<th>CO₂</th>
<th>N₂</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>50% H₂O - 0% CO₂</td>
<td>125</td>
<td>-</td>
<td>125</td>
<td>250</td>
</tr>
<tr>
<td>50% H₂O - 50% CO₂</td>
<td>125</td>
<td>125</td>
<td>-</td>
<td>250</td>
</tr>
</tbody>
</table>

Figure 37 depicts the polarization curve difference as the result of three different CO₂ and steam compositions. As was the case with the custom cells, the performance degrades with rising carbon dioxide content in the cathode gas, but the difference in voltage is subtle. In this case though the OCV are not the as similar as before. This should be attributed to this particular experiment’s timing, as the co-electrolysis was tested on the third and final day of the cell’s operation. This claim is further supported by the next results shown in Figure 38.

The comparison between the custom and cermet cells was completed with the CO₂/H₂O co-electrolysis experiments conducted during SOFEC mode. The results are inconclusive though, as the trend of the polarization curves observed for the custom cell experiments does not appear during the tests on the cermet cell, as seen in Figure 38. There is a high chance that these results diverge from the norm due to the cell degradation that occurred over the passage of three days running experiments on the cell, as stated already. Still, these particular results are presented for the sake of homogeneity.
3.3.4 Cell degradation after 3 days

The final measurements recorded for the cell before removing the setup from the oven aimed at comparing its performance between the first and final day of operation. Previous cells tested during this thesis were used for a maximum of 3 days, as the degradation was hindering the comparison process with the results of the first and the third day showcasing a significant difference. This is the main reason why four custom cells were used in order to get results worthy of presentation. In the case of the cermet cell, time constraints only allowed for one cell to be used and degradation was already high at the third day of operation, as described in Section 3.3.3.
Experimental Results

The comparison between the polarization curves during SOFEC mode with 100% hydrogen at the anode between the first and the third day of operation is illustrated in Figure 39. Both curves were measured before any other experiments were conducted on each day. After three days there is a significant drop in cell performance, which is indicated by the voltage rise throughout the current density range. At the point marked in Figure 39 there is a 62% rise in electricity demand compared to the first day of operation and this is probably the reason why in Section 3.3.3 the experiments performed on the third day gave inconclusive results. The high cell degradation could also be explained by the fact that the LSM material used for both electrodes showcases instability in reducing atmosphere during SOFEC mode of operation [4].

Typically, the degradation of a solid oxide cell is higher when it operates solely as a SOEC, rather than as a SOFC in power production mode [17]. Generally, the ohmic losses are connected to the electrolyte, but the electrode can have a contribution as well. During previous studies in literature [18], it was observed that the ohmic losses of the cell exhibit a small increase as the anode electrode slowly sinters over time. In some cases, when the conditions allow it, high pressures could develop at the electrolyte-oxygen electrode, causing delamination of the oxygen electrode [17].

![Figure 39. Cermet cell: Polarization curve comparison after 3 days of operation. On both cases 100% H2 was used at the anode, while the cathode was fed with 50% H2O.](image-url)
Conclusions and discussion

The Solid Oxide Fuel-assisted Electrolyzer Cell technology is relatively new, as it was patented in 2000. However, via this technique, it is possible to lower the cost of producing hydrogen through electrolysis. While this solution is not totally free of CO₂ emissions, it is a fair compromise between the present and the renewable, emission-free future of energy applications.

The principal behind the operation of a SOFEC is simple: the added fuel gets oxidized at the anode, releasing hydrogen (and CO₂). During this thesis, this step is skipped for most of the experiments, as hydrogen is used directly as a fuel. Nevertheless, the resulting H₂ gets oxidized at the anode of the cell, as is the case with SOFC, a process which is associated with negative Gibb’s free energy. On the other side of the cell, steam is fed to the cathode. In the same way with a SOEC, water gets electrolyzed, providing oxygen ions that travel through the electrolyte and take part in the oxidation reaction at the anode. The latter reaction provides a part of the energy needed for the electrolysis to occur, while the rest is provided by electricity. This results in a lower electrical energy demand for electrolysis compared to a common high-temperature water electrolyzer cell.

The purpose of this report is to reproduce the theory behind the SOFEC operation in a laboratory environment, taking into account the results and suggestions of experiments conducted in the past. Firstly, in order to get acquainted with the experimental procedures, the theory of the SOFC and SOEC is explained and then applied to the SOFEC, as a result of their shared characteristics. Furthermore, the setup used in the laboratory is analyzed, describing both the equipment used and the steps followed for the setup preparation. Also, a number of modifications are suggested, which could be applied to the setup in the future. These changes are made in order to gain access to the exhaust gases of both electrodes, instead of just the SOFC cathode one that is provided by the current setup. Finally, the experimental results are presented and analyzed.

Experiments. The experiments are mainly conducted on custom platinum-pasted electrolyte substrate symmetrical cells. In order to validate the results, the same tests are repeated on a cermet cathode symmetrical cell. The platinum pasted cells tested are symmetrical in order to be able to cycle between SOC modes without having limitations due to the electrode materials used for the anode and cathode. In fact, the platinum electrode cells proved to be suitable for all three modes. On the other hand, the cermet cell tested is probably decent for use in SOFEC mode under certain conditions.

All of the tests were conducted at a temperature of 800°C with the aid of the Fiaxell Open Flanges Test Set-Up. In order to simplify the experimental process, pure hydrogen is used as a fuel for most of the experiments, as mentioned before. Apart from hydrogen, also carbon monoxide was used as a fuel, while carbon dioxide was used for co-electrolysis with steam.

Results. The majority of the results are comparable for the two kinds of cells tested during the experiments. The main difference that the cermet cell showcased compared to the custom-made one was that a higher amount of current could be drawn, even though the active cell area was smaller. The results and the comparisons between SOC operation modes are presented through the polarization curves derived from the experiments.

The Solid Oxide Cells that were tested in SOFEC mode operated within three distinct operating cases, depending on the reversible cell voltage:

- When V>0, the SOFEC produces electricity as a SOFC, but it also generates hydrogen.
- When V<0, the SOFEC electrolyzes water as SOEC, generating hydrogen.
- When V=0, the SOFEC produces hydrogen without any electricity input or output.

During SOFEC mode the voltage of the cell is up to 1.11 V lower compared to a SOEC with the same...
Conclusions and discussion

steam content at the cathode. This translates in about 66% percent drop in electrical power consumption.

In the case of carbon monoxide being used as a fuel, the performance of the cell drops when compared to pure hydrogen fed to the anode, exhibiting almost 30% increase in electric power demand. Nonetheless, a CO-SOFEC still demands less electricity than a SOEC operating with the same steam content at the cathode.

Furthermore, when carbon dioxide is used for co-electrolysis with steam, the increase in power demand is just 6% for the case of SOEC and 13% for SOFEC mode compared to just steam being fed to the cell cathode.

Finally, both kinds of cells exhibit high and rapid long-term degradation. In the most evident case, just after 3 days of continuous operation the electric power demand for SOFEC mode is up to 62% higher than that on the first day of testing.

Recommendations. SOFEC operation is still in its infant phase, as far as experimental research is concerned. As a result, the first recommendation would be to utilize the proposed 3D models in this thesis in order to improve the Open Flanges Test Set-up. These models are designed in order to provide more flexibility, as they provide access to exhaust gases for both electrodes, as explained before.

Moreover, considering that the main advantage of SOFEC operation is the power consumption reduction, other kinds of inexpensive fuels and fuel combinations could be used for future research. Methane would be an ideal candidate, because it is associated with acceptable performance, as described in previous experiments cited in this thesis. Combinations of methane and carbon monoxide would also be a possibility, with economic and CO₂ emission criteria being the main factors for determining an acceptable ratio of the fuel content.

An experimental technique which is not covered in this thesis is the electrochemical impedance spectroscopy (EIS). EIS provides an insight at the reaction kinetics on the electrodes, as well as further analysis of the processes occurring in a fuel cell, being able to distinguish between the different types of polarization that occur within the fuel cell. Therefore, it is highly recommended that EIS is also implemented in future experiments.
5 Bibliography


Bibliography

22-apu-22h-interconnect-test-setup-furnace-mass-flow-controller.


Appendix

Cell Passivation Experiments

A test to observe the response of the custom cell before and after a CO fuel-assisted electrolysis experiment was performed. In Figure 40 the polarization curves of 100% H₂ gas at the anode are shown. The first curve depicts the response of the cell before the CO experiment, which lasted about 40 minutes and contained 100% CO at the anode. Two minutes after the CO experiment was completed, another polarization curve was measured. The results show that the performance of the cell temporarily degraded (passivated) after the experiment with CO, but after 10 minutes the cell recovered (activated), suggesting that only a short-term degradation occurred [13]. The main reason for this occurrence is that CO is still present at the anode, partly preventing the newly fed H₂ gas from being oxidized at the electrode. After several minutes the flow of hydrogen causes carbon monoxide to exit the anode and the cell exhibits the previously recorded performance once again.

![Figure 40. Cell 4: Polarization curve comparison between SOFEC mode for 100% H₂ at the anode before and 2 minutes after an impedance spectroscopy experiment using 100% CO at the anode. 50% of H₂O was used at the cathode side of the cell.](image)

Following, the temporary passivation of the cermet cell was tested, with measurements before and after an experiment with CO at the anode. The polarization curves shown in Figure 41 resulted under SOFEC operation with 100% H₂ used as fuel. The performance of the cell dropped temporarily, but after several minutes of just hydrogen fed to the anode the cell regained its previous status, as was the case with the custom cells. The passivation of the custom cell was attributed to CO still being present at the anode and this seems to be the case for the cerment cell as well.
Figure 41. Polarization curve comparison between SOFEC mode for 100% H₂ at the anode before and 2 minutes after an impedance spectroscopy experiment using 100% CO at the anode. 50% of H₂O was used at the cathode side of the cell.
Appendix

Fiaxell Open Flanges Test Set-Up Modifications

Figure 42. Proposed changes for the Fiaxell Open Flanges Test Set-Up upper plate in order to incorporate outlet gases pipe on the anode side. Left: Design with external outlet pipe. All values are in mm.
Figure 43. Proposed changes for the Fiaxell Open Flanges Test Set-Up upper plate in order to incorporate outlet gases pipe on the anode side. Right: Design with outlet pipe around the inlet pipe. All values are in mm.
Figure 44. Illustration of the modified top part of the 10x10 ceramic setup. All values are in mm.
Figure 45. Illustration of the modified top part of the 10x10 ceramic setup. All values are in mm.