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Solid Oxide Electrolysis Cell co–methanation supported by Molten Carbonate Fuel Cell—a concept

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Abstract

The paper presents a concept of coupling a Solid Oxide Electrolysis Cell with a Molten Carbonate Fuel Cell for co–electrolysis of H_2O with CO_2 for generating synthetic fuel (methane based) for an electricity storage application on a larger scale. The concept is focused on coal/natural gas fired power plants for upgrade as peak energy storage. MCFC anode and SOEC cathode are exposed to the same flow, SOEC produces hydrogen for MCFC and MCFC delivers CO_2 for methanation processes. Both electrodes have compatible polarity, thus they can be directly connected by the current collector and there is no need to apply bipolar plates. On the other side, SOEC will release oxygen to the flue gases and MCFC will capture oxygen and carbon monoxide, thus at the outlet will be a flow with increased oxygen content and decreased carbon dioxide concentration. The concept requires detailed electrochemical, chemical, and thermal simulations.

Keywords: Solid Oxide Electrolysis Cell, Molten Carbonate Fuel Cell, Methanation, energy storage, power–to–gas

1. Introduction

Environmental protection and sustainability are becoming entrenched parts of our daily life. Cars are equipped with catalysts, waste is reused, and CO_2 penalties are a fixture in EU regulations. New ways are being sought to reduce CO_2emissions.

From some time, new tendencies have emerged in the search for clean and efficient power production, in particular the generation of electricity [1–5]. There is an upsurge in the use of renewable energy sources, such as wind, solar [6] and geothermal power. Much is being done to improve the cleanliness of energy production based on the burning of fossil fuels [7, 8], but this seems to be insufficient to attain the goal set by the European Union of continuous reduction of CO_2emissions. Work to reduce CO_2 emissions is ongoing, e.g.: [9]. There are also other options to reduce emissions, such as raising the conversion efficiency of primary energy into power by direct conversion devices (fuel cells) [10], which generate electricity directly from the fuel supplied. Ideally, in this case the fuel is hydrogen [11, 12], but unfortunately it must be produced (eg. by electrolysis [12]) as it does not occur in nature in an unconnected form.

In contrast to classical power plants, fuel cells are not limited by Carnot or Rankine cycle efficiency like other technologies based on the heat cycle [13]. Thus they are a very promising technology for power generation in the future, due to the direct conversion of fuel into electricity. In contrast to low temperature cells [14–16], high temperature fuel cells can be coupled with other devices, such as gas turbines [17, 18] (making a hybrid system), and can be used in poly-generation [19, 20]. Two types of fuel cell can be coupled to a gas turbine system: (i) Molten Carbonate Fuel Cell (MCFC) [21–26] and (ii) Solid Oxide Fuel Cell (SOFC) [27–34]. Coupling the SOFC with a gas turbine system requires one to op-

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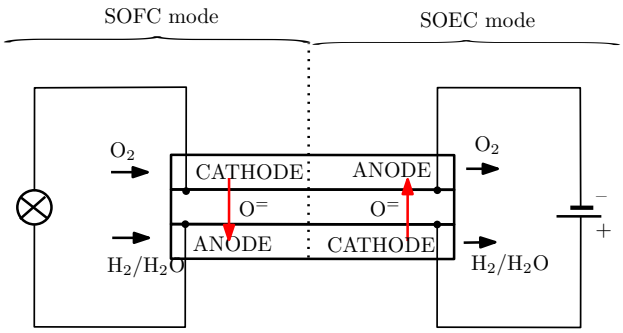

Figure 1: Configuration example of SOFC and SOEC with the variable polarization at the electrodes

erate the fuel cell at increased pressure, which can be quite easily realized in the case of SOFC but is much more difficult in the case of MCFC. On the other hand, seeing as MCFC can be used as a separator of CO_2 from flue gases emitted by classical power plants, it is difficult to choose which of the two is the most promising technology. Their high working temperatures enable the use of fuels other than pure hydrogen—like biofuels [35–37]. SOFCs are characterized by electrolyte in solid form, allowing a certain flexibility of design.

While the share of renewable energy sources in the electricity production mix is still rising, it does have some negative effects, including destabilization of the power system—through sudden changes in power. Thus, the issue of electricity storage is becoming an increasingly major issue. The classic way of storing energy—pumped storage—has been more or less fully utilized in practical terms owing to limited locations. Other solutions (eg. Compressed Air Energy Storage power plants) were discontinued after pilot installations, mainly due to relatively low efficiency and location restrictions. With electricity generation the most promising technology is fuel cells, which generate power in electrochemical reactions with potentially ultra–high efficiency. An alternative idea is a hydrogen power station consisting of a ceramic electrolysis/fuel cell, which could be used to store electricity daily in the form of chemical power. For those kinds of systems, determining an adequate control strategy is a very important task and can be realized by classic algorithms as well as artificial intelligence tools.

The solid oxide fuel cell and solid oxide electrolysis cell are often one device operating at different polarities depending on the current operating mode (charging/discharging). The SOFC or SOEC mode can be chosen by changing the operating mode by reversing the polarity (anode flow then becomes the cathode and *vice versa*)—see Fig. 1. This solution will change the direction of the flow of $O^=$ ions through the solid electrolyte, and the electrode which was supposed to "capture" oxygen will now give oxygen to the main flow.

In fact, hydrogen in a pure form is very difficult to compress and store, and a more favorable option would be to generate methane from power–to–gas plants [38] and feed it into the gas distribution system. This has several advantages, since it would link the power grid with the gas distribution system [39]. Storage of excess electricity in the form of methane would become possible, since the gas infrastructure has a very large energy storage capacity. The feeding–in of synthetic methane is unproblematic, whereas hydrogen feed–in involves several uncertainties. It is not clear to what extent hydrogen (currently there is a 2% limit) can be fed into the gas distribution system and the information available about the impacts and risks of doing so is very contradictory.

Synthetic methane is produced from hydrogen and carbon monoxide or carbon dioxide in the Sabatier process. The chemical reactions are strongly exothermic and require catalysts such as Ni or Ru. Ni is thereby optimal in respect to its activity, selectivity and costs, but requires input gases that are very pure [39]. In fact, methane is the only hydrocarbon fuel that has already been synthesized in power–to–gas pilot plants [39].

Co-electrolysis of CO_2/H_2O mixtures in SOECs is much more complex than electrolysis of H_2O or CO_2 alone, respectively. This is because the reversible shift reaction (Eq. (1)) and methanation reactions or reversed direct internal reforming (DIR) reactions (Eqs (2)..(4)) may occur in the porous cathode (fuel electrode) [40].

$$CO_2 + H_2 \leftrightarrow CO + H_2O \quad (1)$$

$$CO + 3H_2 \leftrightarrow CH_4 + H_2O \quad (2)$$

$$2CO + 2H_2 \leftrightarrow CH_4 + CO_2 \quad (3)$$

$$CO_2 + 4H_2 \leftrightarrow CH_4 + 2H_2O \quad (4)$$

The methanation reactions are highly exothermic, so it is not thermodynamically favored at high temperatures, but high pressures promote this reaction. The significant increase in methane formation at 20 bar operation lowers both the hydrogen and carbon monoxide

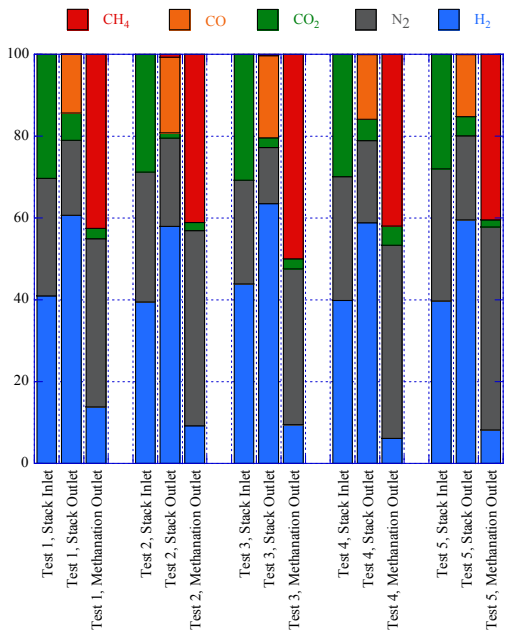

Figure 2: Coelectrolysis with subsequent methanation [43]

content of the cathode effluent [41]. Pressurized operation decreases the cell internal resistance and enables improved system efficiency, potentially lowering the fuel production cost significantly. In [42], a thermodynamic analysis of synthetic methane and dimethyl ether (DME) production using pressurized SOECs is presented. The main parameters of cell operating temperature, pressure, inlet gas composition and reactant utilization are varied to examine how they influence cell thermoneutral and reversible potentials, in situ formation of methane and carbon at the NieYSZ electrode, and outlet gas composition. For methane production, low temperature and high pressure operation could improve system efficiency. For DME production, high pressure SOEC operation necessitates higher operating temperature in order to avoid carbon formation at higher reactant utilization.

The CO_2 methanation process is currently being tested on a laboratory scale and the technical challenges include: heat dissipation, providing an optimal reaction temperature and storing hydrogen in a manner that precludes fluctuations [39]. A methanation reactor [44] installed downstream of a 10-cell stack SOEC stack was tested in [43, 45]. The methanation reactor consisted of an 18 mm inner diameter stainless–steel tube, approximately 1.5 m in length. A commercial steam reforming catalyst (R-67R from Haldor Topsoe) was placed in this tube . The catalyst was a nickel on magnesium aluminate. The reactor was maintained

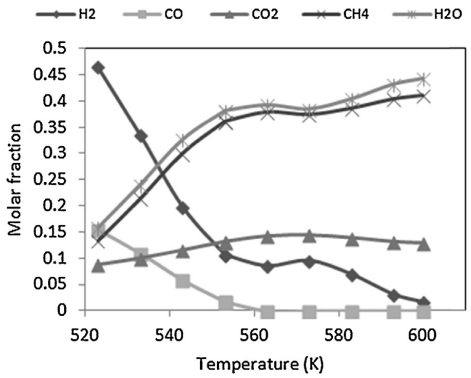

Figure 3: Effect of temperature on outlet gas composition at 0.4 MPa and H_2/ CO = 3 [44]

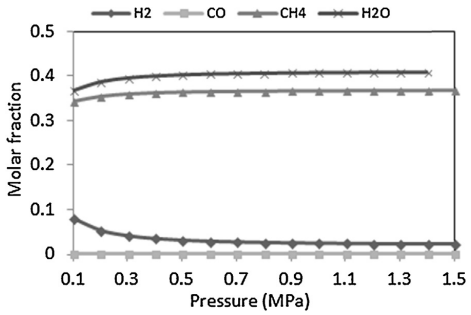

Figure 4: Effect of pressure on outlet gas composition at 593 K and H_2/ CO = 3 [44]

at approximately 300°C (573 K) for testing, giving approximately 40..50% of methane at the reactor outlet. In [40], a two-cell stack was mounted into the oven and equipped with a DC power supply. The stack was tested between fuel cell and electrolysis modes—steam electrolysis (H2O el.) or co-electrolysis of steam and CO2 (co-el.)]—for nearly 8,100 h. The stack was operated during the first 4,000 h under SOFC mode, and for the rest of time was operated under SOEC mode in the new test bench. The co-electrolysis of mixture H_2O and CO_2 was operated with different current densities (–0.3, –0.5 and –0.8 A/cm^2) and different compositions of the inlet-gas. The temperature of the furnace was kept at 760°C.

The methanization reactions are exothermic and therefore thermodynamically favored at low temperatures: Fig. 3 shows that CO conversion is low below 550 K. This is due to the fact that the catalyst used is not active enough at low temperatures. However, at higher temperatures, CO and H_2 conversions increase [44].

Increased operating pressure favors the formation of methane in the SOEC unit [41], see Fig. 4. The process modeling fidelity of both the integrated SOEC syngas generation and Fisher–Tropsch synthesis plants are mutually comparable and allow for the quantifica-

tion of energy consumption for all components in the system and the identification of synergies between the two major subsystems.

The preliminary testing results on electrolysis and co–electrolysis show that the voltage degradations are higher in co–electrolysis mode than in steam electrolysis alone [40].

High temperature co–electrolysis of H_2O and CO_2 seems very attractive due to low cost catalyst needed and high methane content gas as a product. The co–electrolysis requires electricity (e.g. unstable renewable sources), water, and carbon dioxide. Paradoxically, the most challenging here is delivering carbon dioxide as it needs to be generated (by burning coal) or captured from other sources (biogas, flue gas, air, etc).

2. The concept of SOEC based co–methanization supported by MCFC

Apart from hydrogen, CO_2 is the second reactant for methanation and must be provided with low economic and energy effort, ideally with high purity and an adaptable flow rate to suit the fluctuating demand [46]. Carbon dioxide may be obtained from fossil sources by carbon capture in coal–fired power plants, can be generated directly [47] or as a by–product in industrial processes like cement or lime production. Regenerative carbon dioxide is delivered as a by–product in the fermentation process of biogas plants and in biomass gasification and it can also be extracted from the ambient air. Problems related to carbon dioxide sources include the low efficiency of the absorption process from air, the limited capacity of biogas plants and the higher energy demand of power plants that capture fossil carbon dioxide. The overall efficiency of CO_2 methanation is largely dependent on the purity of the carbon dioxide [39]. Thus, this paper proposes to use Molten Carbonate Fuel Cell for supplying CO_2 captured from coal/natural gas fired power plants. This solution gives pure CO_2 delivered directly to the reaction zone.

MCFC can be used to concentrate carbon dioxide, eg. from coal [48] or gas fired power plants, and might form part of a Carbon Capture and Storage system. To function, an MCFC requires a flow of CO_3^{2-} as the electron carrier through the electrolyte. This is achieved by feeding CO_2 to the cathode, where it reacts and flows as CO_3^{2-} to the anode (Fig. 5). There, it becomes carbon dioxide again and, after removing water vapor, may be transported as pure gas to the storage point. In effect, MCFCs work as a filter, allowing exclusive flow of CO_2.

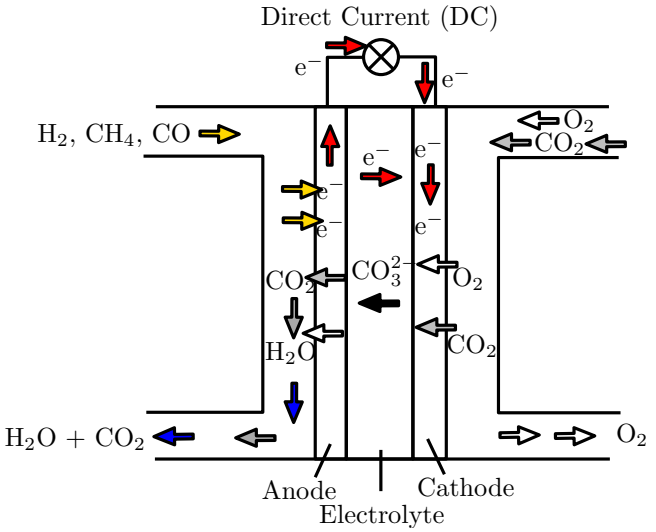

Figure 5: Working principles of MCFC

Thus, MCFC can be used as one of the options for carbon dioxide capture from flue gas and can deliver a mixture of H_2O and CO_2 directly to the SOEC. On the other hand, SOEC can deliver hydrogen as fuel for MCFC. This tandem looks very attractive, as pure gases are delivered for both devices (pure hydrogen/steam to MCFC and pure carbon dioxide/steam to SOEC). The degradation of the two electrochemical devices is expected to be reduced considerably. Additionally, MCFC produces Direct Current (DC) which can support SOEC directly, giving lower terminal voltage than co–electrolysis alone.

By having two high temperature electrochemical devices connected each other by flows and electric circuits on site, theoretically it is possible to construct a stack sandwiched by both types of power sources, as shown in Fig. 6. The stack needs to be fed by flue gas, water, and electricity; and will produce methane rich gas, capturing CO_2 at the same time. With regard to fluctuating and intermittent renewable power sources, the methanation process is more critical than the electrolysis process, since its operating temperatures are higher. To be able to operate the methanation reactor continuously and thus be able to maintain the reaction temperature at a constant level, hydrogen storage has to be installed as a buffer. By installing the SOEC–MCFC tandem in a power plant, the hydrogen storage buffer can be omitted, due to electricity being available on site to keep the device on stand–by mode and/or internal hydrogen infrastructure usually being available

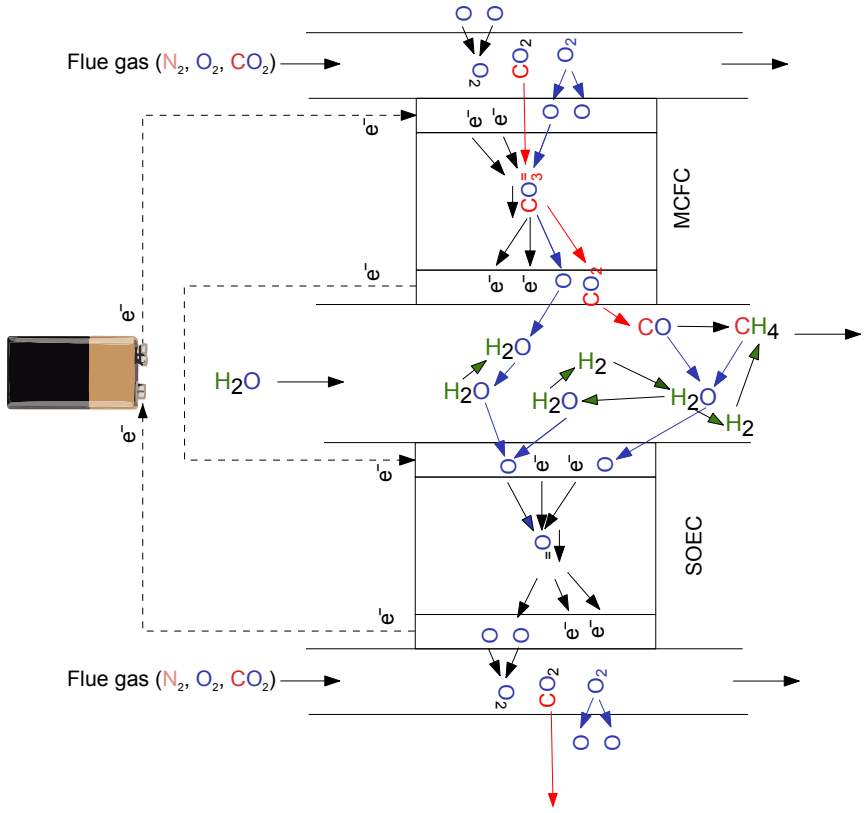

Figure 6: The idea of coupling SOEC with MCFC for direct generation of syngas based on flue gas and water

(hydrogen is used for cooling electric generators).

MCFC anode and SOEC cathode are exposed to the same flow, SOEC produces hydrogen for MCFC and MCFC delivers CO_2 for methanation processes. Both electrodes have compatible polarity, thus can be directly connected by current collector—MCFC anode "–|+" cathode of SOEC and there is no need to apply bipolar plates. On the other side, SOEC will release oxygen to the flue gases and MCFC will capture oxygen and carbon monoxide, thus at the outlet there will be a flow with increased oxygen content and decreased carbon dioxide concentration.

The concept requires detailed electrochemical, chemical and thermodynamic calculations. Important roles will be played by (i) catalysts used on "fuel" electrodes, and (ii) temperature, as MCFC works at around 650° C, whereas SOEC requires 800°C+. In fact, MCFC could work at elevated temperatures if adequate non–metallic materials are chosen.

3. Conclusions

The paper presents a concept of coupling a Solid Oxide Electrolysis Cell with a Molten Carbonate Fuel Cell for co–electrolysis of H_2O with CO_2 to generate synthetic fuel (methane based) for an electricity storage application on a larger scale. The concept is focused on coal/natural gas fired power plants for upgrade as peak energy storage.

The concept seems very attractive from a few viewpoints:

1. Lower voltage required for co–electrolysis
2. Compact device with potential of integrating with a coal fired boiler
3. No need for hydrogen and carbon dioxide storage
4. Material compatibility (nickel based catalyst) with methanation reactions
5. Temperature compatibility with methanation reactions
6. Renewable sources energy storage and carbon dioxide capture at the same time

Detailed investigations based on mathematical modeling and experiments will conducted in the next step of research.

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