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# Long-term tests of a Jülich planar short stack with reversible solid oxide cells in both fuel cell and electrolysis modes

Van Nhu Nguyen\*, Qingping Fang, Ute Packbier, Ludger Blum

Forschungszentrum Jülich GmbH, Institute of Energy and Climate Research, Wilhelm-Johnen-Straße, D-52428 Jülich, Germany

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## ABSTRACT

A two-cell planar stack in the Jülich F-design with solid oxide cells has been built and the reversible operation between fuel cell and electrolysis modes has been demonstrated. The cells were anode supported cells (ASC) with yttria-stabilized zirconia (YSZ) electrolytes, Ni/YSZ hydrogen electrodes and perovskite oxygen electrodes with lanthanum strontium cobalt ferrite (LSCF). This paper summarizes and discusses the preliminary experimental results on the long-term aging tests of the reversible solid oxide planar short stack for fuel cell operation (4000 h) at a current density of  $0.5 \text{ A cm}^{-2}$  which shows a degradation of 0.6% per 1000 h and for steam electrolysis operation (3450 h) and co-electrolysis operation of  $\text{CO}_2$  and  $\text{H}_2\text{O}$  (640 h) under different current densities from  $-0.3$  to  $-0.875 \text{ A cm}^{-2}$  which show different degradation rates depending on current density and on steam or co-electrolysis. Copyright © 2013, Hydrogen Energy Publications, LLC. Published by Elsevier Ltd. All rights reserved.

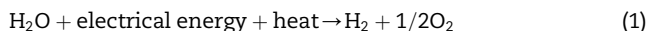
## 1. Introduction

In recent years there has been an increased focus on hydrogen as an alternative energy carrier because of limited fossil fuel sources, increasing oil prices and environmental considerations.

Proton exchange membrane (PEM) electrolyzer cells for  $\text{H}_2$  production are commercially available, but are expensive because of the catalyst used in the cell [1–3]. It is thermodynamically advantageous to operate endothermic reactions such as  $\text{H}_2\text{O}$  electrolysis at high temperatures because a larger part of the energy required for the reaction can then be supplied in the form of high temperature heat. The solid oxide electrolyzers show great potential since the use of high-temperature heat reduces their electricity need [2].

High-temperature solid oxide cells (SOCs) can be reversibly operated under dual modes: power generation mode and electrolysis mode. In the first mode, the SOC acts as a fuel cell (SOFC) and generates electricity by electrochemically combining fuel and oxidant. In electrolysis mode the SOC acts

as an electrolyzer (SOEC) and produces chemicals such as hydrogen by water (steam) electrolysis:



SOECs allow high electrical-to-chemical energy conversion efficiencies even exceeding 100% [4–7]. High temperature electrolysis seems to be very attractive for the production of hydrogen or syngas in the context of renewable energy conversion and storage. Reversible SOECs–SOFCs in an ideal configuration can produce hydrogen from steam (SOE mode) which is then stored and subsequently used to generate electricity and heat (SOFC mode).

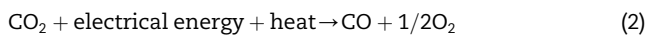
SOEC technology has the advantage that it can be built on the available solid oxide fuel cell (SOFC) technology, which has seen a significant research and development effort [8–12].

Beside steam electrolysis, SOECs are also capable of electrolyzing carbon dioxide to carbon monoxide and oxygen [13–15]:

\* Corresponding author. Tel.: +49 2461 615393; fax: +49 2461 616695.

E-mail address: [va.nguyen@fz-juelich.de](mailto:va.nguyen@fz-juelich.de) (V.N. Nguyen).

Abbreviations			
ASC	anode supported cell	FU	fuel utilization
YSZ	yttria-stabilized zirconia	RSR	reversible shift reaction
LSF	lanthanum strontium ferrite	GC	gas-chromatographic method
LSM	La <sub>0.8</sub> Sr <sub>0.2</sub> MnO <sub>3</sub>	<i>j</i>	current density
LSCF	lanthanum strontium cobalt ferrite	<i>U</i> <sub>1</sub> , <i>U</i> <sub>2</sub>	voltage of cell 1, cell 2
PEM	proton exchange membrane	<i>U</i>	voltage of stack
SOE	solid oxide electrolysis	Δ <i>U</i>	voltage changes of stack
SOC	solid oxide cell	Δ <i>t</i>	difference of time
SOFC	solid oxide fuel cell	slm	standard liters per minute
SOEC	solid oxide electrolyzer cell	kh	1000 h
DC	direct current	EOL	end of life
ASR	area-specific resistance		



At high temperatures (600–900 °C), higher efficiencies are achievable for endothermic reactions, such as H<sub>2</sub>O and/or CO<sub>2</sub> electrolysis, owing to favorable thermodynamic conditions and also because of the improved kinetics for electrode reactions. The thermodynamic reason for higher electrical efficiencies is a decrease in the molar Gibbs energy of the reaction with increasing temperature while the molar enthalpy remains essentially unchanged.

Recently, there has been a surge of research activity for electrolysis of H<sub>2</sub>O and CO<sub>2</sub>/H<sub>2</sub>O mixtures [8,16–21] for the purpose of fuel production using SOC.

Co-electrolysis of CO<sub>2</sub>/H<sub>2</sub>O mixtures in SOECs is much more complex than electrolysis of H<sub>2</sub>O or CO<sub>2</sub> alone, respectively. This is because the reversible shift reaction (RSR, Eq. (3)) and methanation reactions or reversed direct internal reforming (DIR) reactions (Eqs. (4)–(6)) may occur in the porous cathode (fuel electrode) [6,17,19–21],



Hydrogen or the syngas (mixture of H<sub>2</sub> and CO) that is produced by Eqs. (1) and (2) can be used to produce synthetic fuels such as synthetic natural gas (methane) or Fischer Tropsch liquid fuels (methanol or hydrocarbons) or other chemicals [22]. Desired H<sub>2</sub>/CO ratios vary from ~1.4 to 2.1, and this can be varied by the H<sub>2</sub>O/CO<sub>2</sub> ratio input to the electrolyzer. The synthetic methane or liquid fuel is oxidized to produce electricity in a fuel cell again or mechanical energy in a heat engine.

The fundamentals of physics and electrochemistry dictate that the energy densities of molecular hydrogen are unlikely ever to be competitive with liquid fuels for transport applications. The production of carbon-neutral liquid fuels is proposed as a route to the continued provision of compatible, affordable, and sustainable transport [23]. Solid oxide fuel

cells utilizing endothermic internal reforming of liquid fuels may have efficiency and system advantages [24,25]. Because the operating temperatures of SOFCs are high enough to provide the high-temperature heat needed for reforming, SOFC can use internal reforming within the stack, which results in a simplified system (a separate reformer system is not needed) and in higher system efficiency, because energy consumption for air cooling is reduced.

The performed energy balance calculations by Blum et al. [25] for steady state operation with methane revealed that high temperature fuel cells can achieve at least 80% for the fuel utilization because of the possibility of internal reforming, or even over 90% in case of anode off-gas recycling. The highest electrical system efficiency with up to 63% can be achieved with planar SOFC systems, because this concept allows high fuel utilization together with high cell voltages.

A generic liquid-fuel energy cycle utilizing a renewable electrical source and stack with reversible SOCs is proposed by Zhan et al. [17]. Note that the cycle is CO<sub>2</sub> neutral, that is, the same amount of CO<sub>2</sub> is produced in oxidizing the hydrocarbon as is consumed in electrolysis. Sterner [26] described a concept of an integrated power, heat, and transport system combining renewable methane. The renewable-power-to-methane efficiency is predicted to be 48% [26].

A so far critical issue with respect to use of SOCs in the electrolysis mode is the long-term stability. The main bottleneck at present for industrial application of SOECs is the cell degradation. Whereas voltage degradation rates below 1%/1000 h under constant current operation are demonstrated for SOFCs (and stacks), degradation in electrolysis mode tends to be higher. Hauch and co-workers [27] tested solid oxide cells in the electrolysis mode at the Risø National Laboratory at temperatures in the range 650–950 °C. The cells were consisting of an electrochemically active fuel electrode with Ni/YSZ material (YSZ = 8 mol%, Y<sub>2</sub>O<sub>3</sub>-stabilized ZrO<sub>2</sub>), an electrolyte with YSZ and an oxygen electrode with LSM/YSZ composite (LSM = La<sub>0.8</sub>Sr<sub>0.2</sub>MnO<sub>3</sub>). Long-term degradation rate of 2%/1000 h was obtained at 850 °C and current density of –0.5 A cm<sup>–2</sup>, whereas the degradation rate increased to 6%/1000 h at 950 °C and current density of –1.0 A cm<sup>–2</sup>. These are significantly higher than those for similar cells tested in fuel cell mode (<2%/1000 h at 950 °C, and 1.7 A cm<sup>–2</sup>). Stack

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