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### ► To cite this version:

Vanessa Bezerra Villela, Elizabeth Vera, Franck Fournet Fayard, Laurent Dessemond, Steil M.C., et al.. Catalytic versus Electrocatalytic Layer for Oxidative Coupling of Methane on Solid Oxide Fuel Cells. 18th International Congress on Catalysis, Jul 2024, Lyon, France. <hal-04889262>

**HAL Id: hal-04889262**

**<https://hal.science/hal-04889262v1>**

Submitted on 15 Jan 2025

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## Catalytic versus Electrocatalytic Layer for Oxidative Coupling of Methane on Solid Oxide Fuel Cells

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### Introduction

Light olefins such as ethylene (C<sub>2</sub>) serve as important building blocks for a large range of applications like polymers, lubricants, cosmetics, etc, and their production represent a crucial challenge for the fabrication of more sustainable petrochemicals.<sup>1</sup> Methane, a widely available feedstock, can be converted to C<sub>2</sub>+ hydrocarbons indirectly via syngas, however it is energetically and economically expensive.<sup>2</sup> In this sense, oxidative coupling of methane (OCM) represents a more viable route for the direct conversion of methane into higher value hydrocarbons, such as ethylene.

The use of solid oxide fuel cells (SOFC) for the performance of electrochemically aided catalytic reactions has gained interest because of the larger availability of green electrons from renewable sources. Therefore, the electrocatalytic oxidative coupling of methane (EOCM) can allow to selectively tune the active oxygen for the reaction and thus increase the selectivity of the desired products.<sup>3</sup> Here we study the OCM on two different anode architectures of a SOFC: a classical perovskite-MIEC electrocatalytic anode (La<sub>0.7</sub>Sr<sub>0.2</sub>Fe<sub>0.8</sub>Co<sub>0.2</sub>O<sub>3</sub>) and a double-layered anode composed of NiO/YSZ coupled with a catalytic layer of La<sub>0.5</sub>Ce<sub>0.5</sub>O<sub>1.75</sub> (Figure 1).

### Materials and Methods

La<sub>0.5</sub>Ce<sub>0.5</sub>O<sub>1.75</sub> (LCO) was prepared by the combustion method (with excess urea) while La<sub>0.7</sub>Sr<sub>0.2</sub>Fe<sub>0.8</sub>Co<sub>0.2</sub>O<sub>3</sub> (LSFO) was prepared by an EDTA-citrate method. The powders were characterized by X-ray diffraction (XRD), Raman spectroscopy and scanning electron microscope (SEM). The electrolyte-supported single cells were prepared using a YSZ-8 substrate. The electrode layers were deposited by screen printing on each side of the electrolyte. The cell with LSFO was symmetric. For the double-layered anode cell, a La<sub>0.65</sub>Sr<sub>0.3</sub>MnO<sub>3</sub> (LSM)/YSZ cermet was used as cathode and the anode was based on a NiO/YSZ cermet, upon which a LCO catalytic layer was spray-coated.

The single cell was set up in a three-atmosphere test bench comprised of two alumina tubes (anode and cathode side) using a gold ring and glass for sealing. The EOCM tests were conducted at 800 °C under different reaction conditions. The outlet gases were analyzed by gas chromatography and the electrochemical measurements were performed with a impedancemeter/potentiostat.

### Results and Discussion

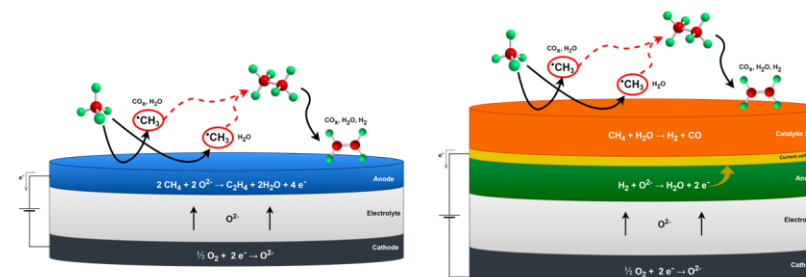
XRD diffractograms of the powders showed single-phase crystalline structures. For LCO, Raman spectroscopy showed the presence of bands associated with the formation of

oxygen vacancies while SEM images evidenced a porous microstructure. The catalytic tests on powder LCO demonstrated that it is active for C<sub>2</sub> hydrocarbons production, with a higher production of CO<sub>x</sub> but a fair selectivity for C<sub>2</sub>H<sub>4</sub>. Additionally, no coke formation was observed even after 20h of reaction. Then, the EOCM was tested on the different cells by varying the O<sup>2-</sup>/CH<sub>4</sub> ratio. The CH<sub>4</sub> conversion (X) presented a slight increase when increasing the ratio. Regarding the C<sub>2</sub> selectivity (S) and yield (Y), a maximum was observed at 0.12. The formation rate of carbon oxides (CO + CO<sub>2</sub>) increased with the ratio, which was attributed to an increase on the amount of O<sup>2-</sup> species that favored the CO<sub>x</sub> products.

**Table 1. Summary of the EOCM performance of the LCO/NiO-YSZ/YSZ/LSM cell.**

| $O^{2-}/CH_4$ | $XCH_4$ | $SC_2$ | $YC_2$ | $SCO_x$ | $YCO_x$ | $SH_2$ | $YH_2$ | $C_2H_4/C_2H_6$ | $CO/CO_2$ |
|---------------|---------|--------|--------|---------|---------|--------|--------|-----------------|-----------|
| <b>0.06</b>   | 65%     | 3%     | 2%     | 8%      | 5%      | 81%    | 54%    | 0.7             | 0.18      |
| <b>0.12</b>   | 67%     | 7%     | 5%     | 16%     | 10%     | 74%    | 60%    | 0.9             | 0.03      |
| <b>0.30</b>   | 68%     | 4%     | 3%     | 23%     | 16%     | 70%    | 63%    | 0.7             | 0.04      |

Finally, the EOCM was also tested on the symmetric LSFO/YSZ/LSFO cell. The differences found were attributed to the different secondary reactions controlling the CO<sub>x</sub> formation.



**Figure 1. EOCM process on the different types of architectures: electrocatalytic perovskite-MIEC (left) and double-layered (right) anode cells.**

### Significance

The influence of different anode-architectures was evaluated for the enhancement of electrocatalytic oxidative coupling of methane. An innovative double-layered anode showed fair performances for the EOCM.

### References

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