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Ethylene epoxidation on Ag/YSZ electrochemical catalysts: Understanding of oxygen electrode reactions

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Abstract

In this study, we have investigated, for the very first time, the oxygen electrode reactions on Ag/YSZ electrochemical catalysts both in oxygen and under reaction conditions compatible with the ethylene epoxidation reaction. Electrochemical Impedance Spectroscopy (EIS) in combination with *in-situ* Raman spectroscopy and catalytic activity measurements were used to identify and understand the main oxygen reaction pathways. The results obtained suggested that the rate limiting step under an O₂ reaction atmosphere (at 300 °C) is the O₂ adsorption/dissociation process on the Ag catalyst-electrode. In addition, the polarization resistance increased with time under the presence of O₂. This was attributed to the formation of Ag₂O on the catalyst surface or near surface, which limits the oxygen electrode reactions. Finally, we observed that the addition of ethylene in the feed stream hinders the electrode reaction, due to its competitive chemisorption with oxygen on Ag. These results give new insights into the design of selective Ag/YSZ catalyst for ethylene epoxidation.

1. Introduction

Ag-based electrodes interfaced with O²⁻ conductor solid electrolytes (e.g. Ytria Stabilized Zirconia, YSZ) have been extensively studied in literature for electro-catalytic processes [1–3]. This is mostly due to the fact that Ag exhibits excellent properties as an effective cathode for solid oxide fuel cells (SOFC), including its excellent electrical conductivity and high performance for the oxygen reduction reaction.

The electrochemical properties of these electrochemical cells have been therefore mostly studied at temperatures compatible with SOFC operation (600-1000 °C). According to literature, the oxygen electrode reactions include the following pathways (see Fig. S1) [4,5]: A) Transport of molecular oxygen into the pores of the electrode (O₂ gas diffusion), B) Dissociative chemisorption of oxygen on the metal active sites (O_{2 gas} → 2 O_{ads}), C) Diffusion of atomic oxygen to the electrode/electrolyte interface, D) Electron transfer reaction (O_{ads} + 2e⁻ → O²⁻). It is well understood that steps C and D should take place at the so-called triple phase boundaries (tpb): gas phase/catalyst-electrode (Ag)/solid electrolyte (YSZ).

However, Ag/YSZ materials could be also used for other applications at lower reaction temperatures. In this sense, since the discovery of the phenomenon of Electrochemical Promotion of Catalysis (EPOC), this kind of electrochemical cells could be considered as “electrochemical catalysts” [6–8]. The particularity of these cells is based on the fact that the electrode behaves as a catalyst (catalyst-electrode) for a given reaction. This way, upon the application of an electrical current (or potential), a controlled amount of O²⁻ ions migrates through the solid electrolyte to the catalyst-electrode. From a catalytic point of view, the O²⁻ ions might behave as “electronic promoters”, modifying the electronic density of the catalyst-electrode, hence allowing to promote its activity and/or selectivity [9–11].

The EPOC phenomenon, discovered more than 30 years ago, can boost the catalytic activity and selectivity for a wide variety of environmental (e.g. reduction of NO_x from mobile sources [12,13]) and industrial processes (e.g. ammonia synthesis [14], hydrogen production [15]).

The promotional effect of O²⁻ ions on a catalyst-electrode was reported for the first time for the ethylene epoxidation reaction on a Ag/YSZ electrochemical catalyst [9,16,17] for the production of ethylene oxide (EO). EO is an essential building block for chemical industry as a precursor of added-value molecules such as plastics and polyester fibers. EO is

commercially produced at low temperatures (around 300°C) on Ag/ α -Al₂O₃ catalysts using chlorinated hydrocarbons (1,2-dichloroethene (1,2-DCE) or vinyl chloride) in ppm quantities in the gas phase as moderators and alkali promoters (Cs ~300 ppm). However, the competition with the direct oxidation process (leading to the production of CO₂ instead of EO) leads to low EO yield and/or selectivity. Therefore, the industrial process to produce EO is neither efficient, nor eco-friendly (due to the use of 1,2-DCE). EPOC could be an alternative for a direct and environmentally-friendly new route for epoxidation of ethylene in order to produce EO with high selectivity and alkene conversion. For instance, in ref. [9] it was found that pumping O²⁻ ions towards the Ag electrode via applied polarization allowed to dramatically enhance the selectivity and yield of EO production at 400 °C and atmospheric pressure. It was demonstrated that the rate of EO production can exceed the rate of O²⁻ electrochemically supplied by a factor of 400. In other words, upon polarization, the enhancement of the EO production due to the catalytic process on the Ag catalyst-electrode (C₂H₄ + 0.5 O₂ → C₂H₄O) was much more pronounced than that for the electro-catalytic oxidation of C₂H₄ with the O²⁻ ions (C₂H₄ + O²⁻ → C₂H₄O + 2 e⁻). The magnitude of this electrochemical activation strongly depends on the kinetics of the oxygen electrode reaction at the Ag/YSZ interface. A slow electrochemical process will increase the lifetime of ionic species. In that case, intermediate products such as partially discharged O^{δ-} ionic species can migrate over the entire gas-exposed Ag surface and substitute Cl⁻ anions. Furthermore, the lifetime of these O^{δ-} ionic species is long enough if the coverage of “normal” chemisorbed oxygen species coming from the gas phase is high. Therefore, to predict the potential impact of EPOC for boosting the EO selectivity, it is crucial to unravel the oxygen electrode mechanism at the Ag/YSZ interface. Unfortunately, to the best of our knowledge, this has only been scarcely investigated below 400 °C, and never under ethylene epoxidation reaction conditions. Therefore, the aim of this work is, for the very first time in literature, to study and thoroughly understand the oxygen electrode mechanism at the Ag/YSZ interface at 300°C and under reaction conditions characteristic of the ethylene epoxidation catalytic process.

2. Experimental

In order to study the overall impedance of the electrochemical catalyst, a symmetric electrochemical cell was used with the following configuration: Ag//Ytria-Stabilized Zirconia (YSZ)//Ag, where Ag catalyst-electrodes with a geometrical area of 2.7 cm², were placed on the opposite sides of a YSZ solid electrolyte disk. Electrochemical properties were

investigated in the same atmosphere for both electrodes and without any polarization. Therefore, both Ag electrodes have the same electrochemical properties and the impedance spectra represents the overall contribution of the two electrodes. First, a YSZ disk of 18.7 mm diameter and 1 mm thick was prepared by sintering a commercial YSZ powder (Tosoh, ZrO_2 -8 mol % Y_2O_3) at 1350 °C for 2 h (densification higher than 98%). We decided to prepare Ag coatings with similar morphology and porosity than those implemented in early studies on EPOC for ethylene epoxidation [9,16,17]. Commercial silver-organic ink (Metalor ® HPS-FG32) was deposited on both faces via the screen printing method, followed by calcination at 600°C for 2 h in air (heating ramp of 2 °C min⁻¹). The final Ag loading was ~20 mg on each electrode. Fig. S2 a) and b) show SEM images of the “fresh” electrochemical catalyst. The thickness of the Ag catalyst-layer is in the range 10–25 µm. The sample exhibits low porosity with Ag agglomerates in the micrometer range, which could be attributed to the high calcination temperature, as previously observed [11].

For electrochemical measurements, the Ag//YSZ//Ag sample was placed between two Au current collectors in an alumina holder inside a continuous-flow quartz reactor. The current collectors were attached to Au wires and then connected to a potentiostat/galvanostat VersaSTAT 3 (Princeton Applied Research). The impedance measurements were recorded by the Frequency Response Analyzer (FRA) Module of the potentiostat/galvanostat over a frequency range from 1 MHz to 1 mHz with an amplitude of the AC signal of 20 mV. All the tests were performed at 300 °C under an overall gas flow of 10 L h⁻¹. This temperature is compatible with the ethylene epoxidation reaction. Two different reaction atmospheres were investigated: O₂ in He (0.8 to 8 % volume) and the ethylene epoxidation reaction mixture (4 % C₂H₄ / 1 % O₂) at atmospheric pressure. The impedance data analysis was performed by the Z-view Software.

For catalytic measurements, the sample was placed in a quartz reactor described elsewhere [11]. The reactants and products were analyzed by an on-line micro gas chromatograph (µGC-R3000, SRA), composed of a BF PPU 8m/PPQ 1m column (CO₂ analysis) and a VAR PLOT Q 8m column (Ethylene Oxide analysis). The detection limit for Ethylene oxide analysis was ~ 2 ppm. A light-off experiment was performed from 100 °C to 300 °C under ethylene epoxidation reaction conditions (4 % C₂H₄, 1 % O₂, 3 L h⁻¹, He balance).

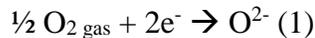
Raman spectroscopy was performed in-situ at 300 °C under an 8 % O₂ (in N₂, 1.5 L h⁻¹) reaction atmosphere in the region of 200-1600 cm⁻¹ using a LabRam HR (Horiba) spectrometer equipped with a CCD detector cooled at -75 °C. Measurements were carried out

under microscope with a laser wavelength of 514.53 nm. A low dispersion grating of 1800 grooves/mm was chosen to get a maximum signal (to improve the resolution). The spectra were recorded and treated using the Labspec software (Horiba). Prior to the analysis, the instrument was calibrated using a silicon wafer.

3. Results and discussion

3.1. Electrochemical properties in oxygen

The oxygen electrode reaction on the Ag electrode was carried out under an oxygen-based reaction atmosphere (in the absence of ethylene). Figure 1 shows a typical impedance spectrum of the Ag/YSZ electrochemical catalyst under 8 % O₂ at 300°C. The spectrum could be separated into two arcs located in the high frequency zone (1 MHz to 1 KHz) and two arcs in the low frequency zone (1 KHz to 1 mHz). To determine the contribution of each process of the oxygen electrode reaction, a typical equivalent circuit [18] composed of a resistance and a Constant Phase Element (CPE) in parallel for every depressed-arc was used to fit the data (see inset on Fig 1). Usually, the YSZ solid electrolyte resistance produces at least two different arcs in the high frequency region, where the first one (highest frequency range) could be attributed to the charge transport through the bulk of the electrolyte, and the second arc is generally due to the transport through the grain boundaries [4,19]. The low frequency contributions observed in the spectra could be therefore attributed to the oxygen electrochemical reaction over the electrodes, which seems to be composed of at least two different electrochemical processes. In the proposed equivalent circuit, R1 and R2 correspond to the resistance of the solid electrolyte, while the sum of R3 and R4 represents the total polarization resistance for the overall electrode reaction (1).



CPE are constant phase elements, which in general could be related to the double-layer capacitance [4]. Further experiments were performed in order to associate the main steps for the oxygen electrode reactions with the R3 and R4 contributions observed on Fig. 1. To this purpose, the effect of the oxygen partial pressure on the polarization resistance was explored at the same reaction temperature (300 °C). The oxygen concentration was varied from 0.8 % to 8 %. Fig. 2a shows the impedance spectra of the low frequency region (1 KHz to 1 mHz, electrode reactions). The experiments were performed in the chronological order indicated by the arrow in the legend. Hence, the spectra under 4 % and 8 % O₂ were taken twice. It could

be observed (Fig. 2a) a very important influence of the O₂ concentration on the impedance spectra, increasing both R3 and R4 resistances as the oxygen partial pressure decreases. Previous studies [3,20] have reported that the variation of the polarization resistance with the O₂ partial pressure obeys the equation (2).

$$R = k P_{O_2}^{-n} \quad (2)$$

Fig. 2b shows the log-log variation of R3 and R4 polarization resistances (obtained by treatment of Fig. 2a spectra) with oxygen partial pressure. The value of the slope “n” has been found to provide important insights regarding the species involved in the electrode reactions. For instance, Siebert et al. [20] found that $n = 0.5$ could be attributed to a concentration variation of adsorbed dissociated oxygen. In other words, it could be related with the dissociative chemisorption of O₂ and the further diffusion of atomic oxygen to the “tpb” (Paths B and C in Fig. S1). On the other hand, n values equal to 0.25 and 1 have been attributed to the charge transfer step (Path D in Fig. S1) and the O₂ gas diffusion in the electrode (Path A in Fig. S1), respectively [3]. Based on the results shown on Fig. 2b, a $P_{O_2}^{-0.22}$ dependency has been observed for R3 (higher frequency part of the polarization resistance), which could be attributed to the charge transfer process at the Ag/YSZ interface. However, a $P_{O_2}^{-0.72}$ dependency was observed for R4 (lower frequency). According to the previous discussion, this resistance could be attributed to either the oxygen dissociative chemisorption ($n = 0.5$) or to the gaseous diffusion of O₂ in the porosity of the Ag electrode ($n = 1$).

A pure unlimited O_{2 gas} diffusion process (usually represented by the so-called Warburg element) manifests itself in the impedance spectra as a straight line making a 45 ° angle with the Z_{real} axis [1]. Based on the shape of these spectra, it seems that the O₂ gas diffusion resistance is negligible, and therefore the low frequency resistance R4 could be mostly attributed to the oxygen adsorption process. Similar results were obtained by Li et al. [3] with a La_{1.6}Sr_{0.4}NiO₄-Ag/YSZ electrochemical cell. At 700 °C, they observed two different contributions to the electrode polarization resistance, which were attributed to the adsorption/desorption process (low frequency) and the charge transfer (high frequency).

The EIS technique could be also used to clarify the rate limiting step regarding the O₂ electrode reaction. Based on the results shown on Figs 2 a and b, it is evident that R4 is larger than R3. It seems to indicate that the oxygen adsorption/dissociation process is the major rate determining step for the Ag electrode in the whole range of O₂ partial pressure studied, at the

reaction temperature of 300 °C, in good agreement with a previous study performed by Huang et al. [21] for low temperatures SOFC (300-500 °C) on Ag-based electrodes.

The second spectra recorded under O₂ 4 % and 8 % exhibit an increased overall polarization resistance (R3 + R4) with respect to the first analysis. It seems to indicate that the electrode suffers some modification with time in the presence of oxygen. In order to verify this effect, Fig. 3a shows the evolution of the impedance spectra with time under an 8 % O₂ reaction atmosphere. An important increase of the electrode resistance is observed, with a more pronounced effect on the R4 resistance (previously attributed to the oxygen dissociative adsorption process). In addition, Fig. 3b shows the comparison of the impedance spectra recorded under 8 % O₂ for the Ag/YSZ electrochemical catalyst before and after a reducing treatment under a 5 % H₂ reaction atmosphere (He balance, 300 °C, 10 L h⁻¹) at 300°C. Comparing the two spectra, it could be clearly observed that the R4 resistance strongly decreased (it almost disappeared) after the H₂ treatment. A small shift was observed in the spectrum after H₂ treatment, which could be attributed to a slightly lower reaction temperature (~ 5 °C). These results suggest that the interaction of the Ag catalyst-electrode with O₂ leads to a significant modification of the electrode performance, mainly affecting the O₂ dissociative adsorption path.

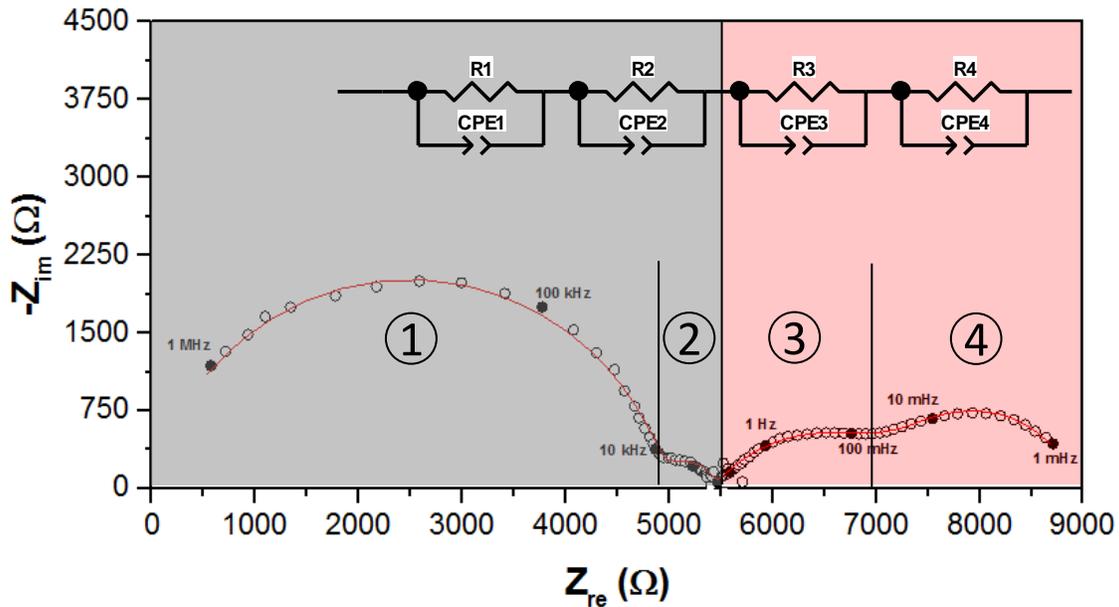


Figure 1. Impedance spectrum of the Ag/YSZ/Ag symmetric electrochemical catalyst. Reaction conditions: 8 % O₂ in He, overall flow = 10 L h⁻¹, Temperature = 300 °C

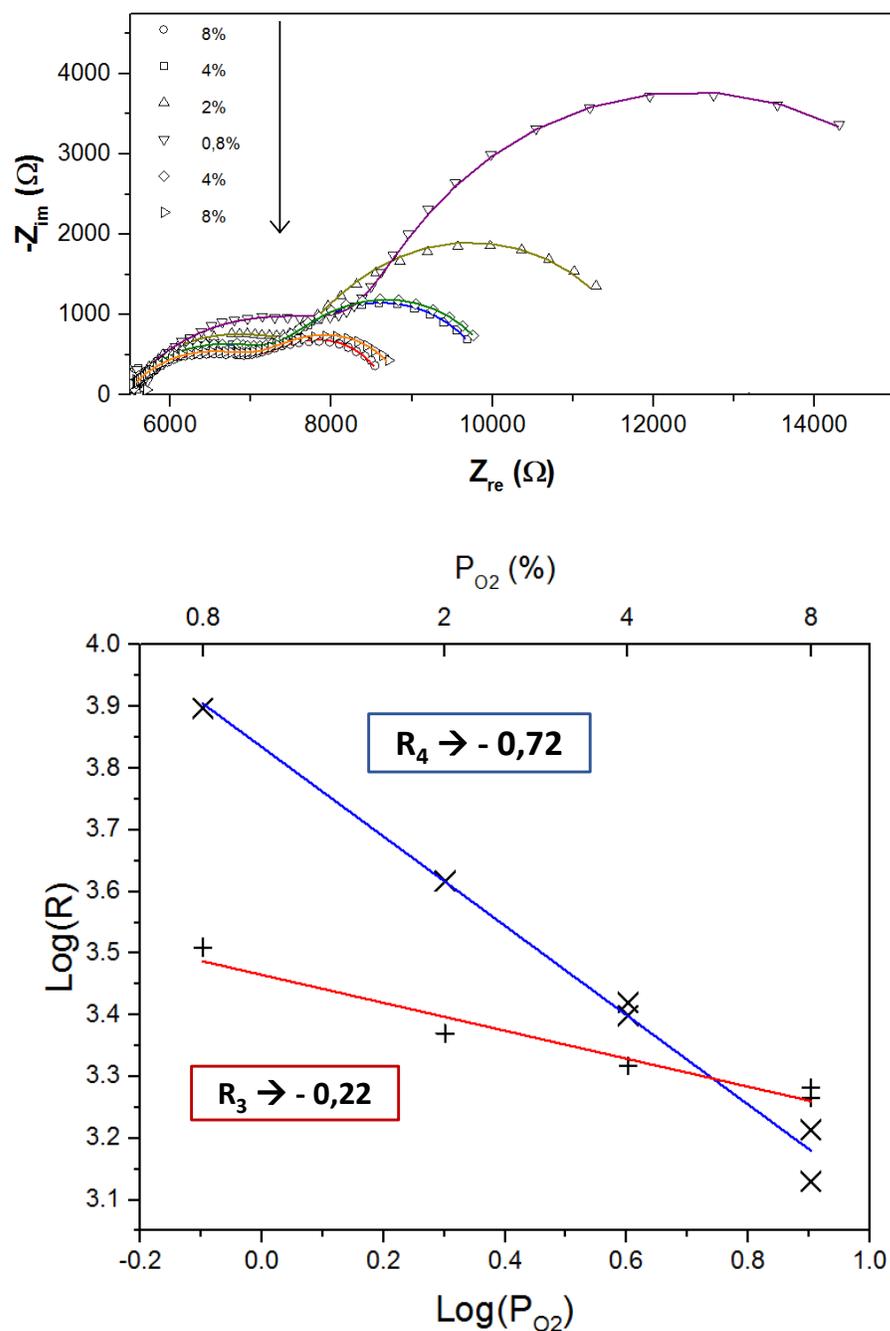


Figure 2. a) Impedance spectra of the Ag/YSZ/Ag symmetric electrochemical catalyst under different oxygen partial pressures. b) Variation of the polarization resistances (R_3 and R_4) with the oxygen partial pressure. Reaction conditions: 0.8 - 8 % O_2 in He, overall flow = 10 $L h^{-1}$, Temperature = 300 $^{\circ}C$

3.2. In-situ Raman spectroscopy

In-situ Raman spectroscopy was used to probe the adsorption behavior of oxygen on the silver catalyst-electrode at 300 $^{\circ}C$. Prior to the Raman measurements, the sample was in-situ pre-reduced at 300 $^{\circ}C$ in H_2 (6 % H_2 in N_2 , total flow 1.5 $L h^{-1}$) for 1 hour. Afterwards, the

reaction atmosphere was switched to O₂ (8 % in N₂, flow 1.5 L h⁻¹) and Raman spectra were obtained every 2 hours for 15.5 hours at a constant temperature of 300 °C. We observe similar bands in all the Raman spectra but the area and the intensity of the bands at 800 cm⁻¹ and 542 cm⁻¹ significantly change as a function of time. The band at ~ 800 cm⁻¹ was assigned by G. Waterhouse et al. [22] to an Ag-O stretching mode, due to surface chemisorbed oxygen formed by the segregation of atomic oxygen coming from the Ag subsurface layers. This atomic oxygen strongly adsorbed on Ag was demonstrated to be stable even at high temperatures (up to 923 K) in the presence of O₂ by *in-situ* Raman studies at different reaction temperatures. On the other hand, B. Pettinger et al. [23] performed Raman experiments on Ag-based materials at different reaction temperatures. They concluded that a band at approximately 530 cm⁻¹ could be representative of bulk Ag₂O species. The formation of Ag₂O species was also observed by M.Fee et al. [24] on Ag-based electrodes interfaced with a YSZ solid electrolyte under similar reaction conditions (O₂-based reaction atmosphere at 350 °C). Therefore, the bands observed at 800 cm⁻¹ and 542 cm⁻¹ in Fig. 4a could be attributed to strongly chemisorbed oxygen on the Ag catalyst-electrode and to the oxidation of silver to silver oxide, respectively. Fig. 4b shows the evolution of the ratio of the areas of the two bands (800 cm⁻¹/542 cm⁻¹) with time. The trend observed in this figure seems to indicate that the proportion of Ag₂O with respect to that of chemisorbed oxygen increased with time at 300 °C in an oxygen-based reaction atmosphere. Other bands observed at 425 cm⁻¹ and 910 cm⁻¹ on the Raman spectra could be attributed to different kind of adsorbed oxygen species on the Ag catalyst-electrode [25,26].

The results shown on Figs. 3 and 4 suggest that the formation of silver oxide species strongly modifies and limits the oxygen electrode reaction on Ag under the reaction conditions studied. It increased the polarization resistance, mainly concerning the oxygen dissociative adsorption process (R4), which has been identified as the rate determining step. A reduction step in H₂ (Fig. 3b) can reduce the silver oxide species, regenerating the Ag surface and promoting therefore the O₂ electrode reaction.

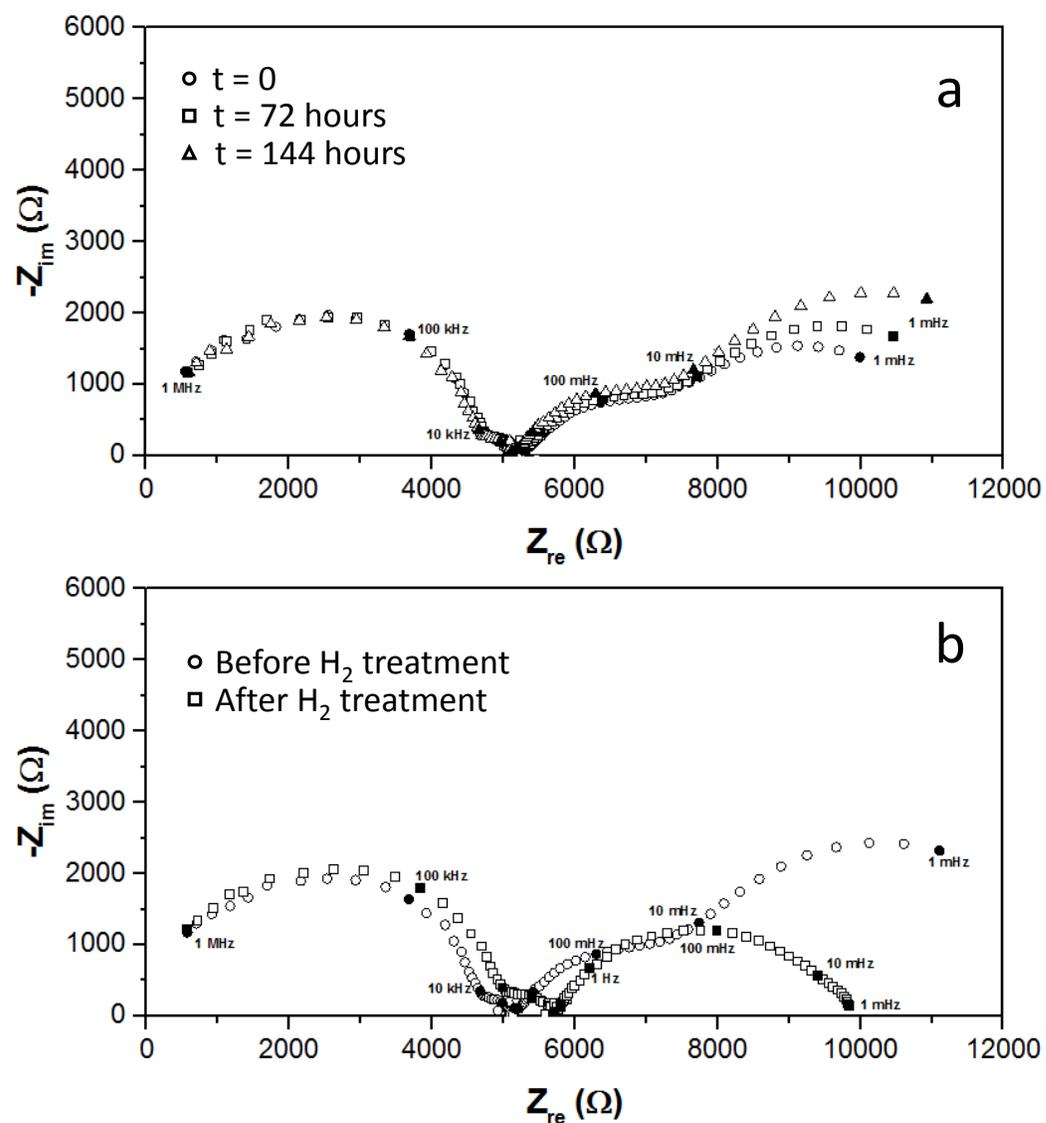


Figure 3. Impedance spectra of the Ag/YSZ/Ag symmetric electrochemical catalyst under 8% O_2 ($10 L h^{-1}$, Temperature = $300 ^\circ C$): a) Influence of time on stream, and b) Influence of a pretreatment under a 5 % H_2 reaction atmosphere (He balance, $10 L h^{-1}$, $300 ^\circ C$, 34 hours).

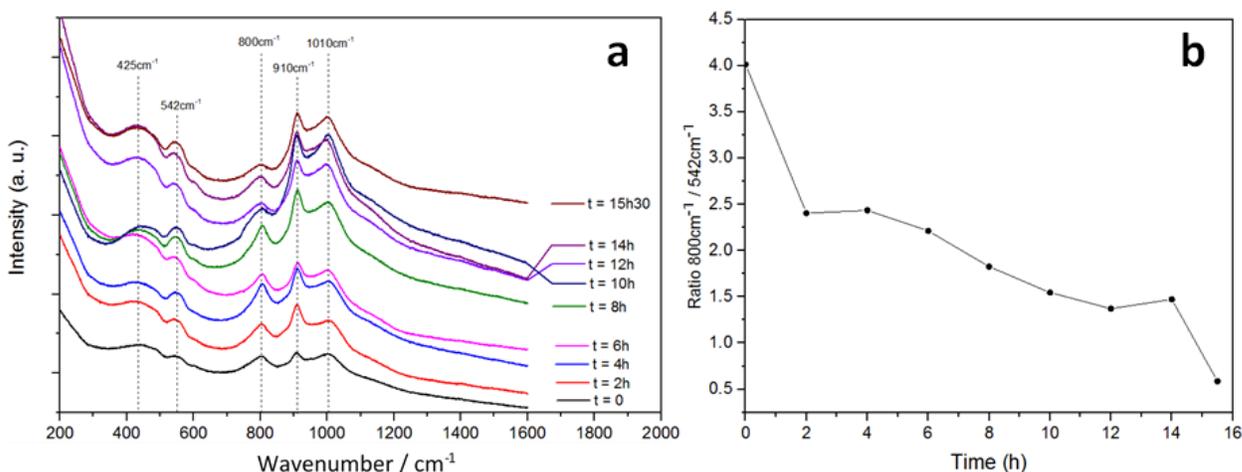


Figure 4. a) *in-situ* Raman spectra of a Ag/YSZ electrochemical catalyst. Pretreatment: 6 % H₂ in N₂, total flow 1.5 L h⁻¹, 1 hour, 300 °C. Spectra taken at 300 °C under O₂ 8 % (total flow 1.5 L h⁻¹, N₂ balance). b) Evolution of the ratio of the band areas at 800 cm⁻¹ and 542 cm⁻¹ with time.

3.3. Electrochemical and catalytic properties in ethylene epoxidation conditions

Fig 5a shows the impedance spectra under different reaction conditions: A first spectrum was measured under 1 % O₂ environment (He balance, 10 L h⁻¹, 300 °C), and a second one under an ethylene epoxidation reaction atmosphere (4 % C₂H₄, 1 % O₂, He balance, 300 °C). A full spectrum under ethylene epoxidation conditions is shown in the supporting information (Fig. S3). In addition, Fig. S4 shows a light-off experiment to assess the catalytic performance of the Ag/YSZ electrochemical catalyst.

Under ethylene epoxidation conditions, with a significant excess of ethylene in the feed (as typically used for this catalytic process), the electrode polarization resistance significantly increased (Fig. 5a, Fig S3). In parallel, the catalytic performances are poor, reaching a very low ethylene conversion of around 0.045 % at 300°C. We did not detect any ethylene oxide production, suggesting that the Ag electrode is only active for ethylene combustion. These results indicate that an important amount of ethylene is adsorbed on the Ag catalyst-electrode under these conditions, hindering in a significant manner the O₂ chemisorption and then the electrode reaction and the catalytic reaction. In addition, a new impedance spectrum was obtained under 1 % O₂ reaction atmosphere (Fig. 5a), right after stopping the flow of ethylene. In comparison with the spectrum under similar conditions before the ethylene epoxidation treatment, an important decrease in the polarization resistance (mostly concerning R4) is observed. This effect was similar but less pronounced to that observed

after a H₂ treatment (Fig. 3b). It could be attributed to the consumption of the silver oxide species with the ethylene present in the reaction atmosphere. In other words, ethylene seems to have an important reducing effect on the Ag catalyst-electrode. This effect was further verified by following the evolution of the impedance spectra under 1 % O₂ with time, right after ethylene epoxidation reaction conditions (Fig. 5b). A significant increase of the polarization resistance is observed with time, similar to that observed on Fig. 3a, which could be most likely attributed to the slow and gradual formation of silver oxide species.

The results obtained in this study allowed, for the very first time, to relate the catalytic properties of an electrochemical catalyst (Ag/YSZ) with electrochemical measurements (Electrochemical Impedance Spectroscopy) concerning the oxygen electrode reactions. The experiments were performed under reaction conditions compatible with the ethylene epoxidation reaction (low reaction temperature: 300 °C, and high ethylene excess). The presence of ethylene seems to hinder the oxygen adsorption on the Ag catalyst-electrode, increasing the overall polarization resistance attributed to the oxygen electrode reactions. A slow electrochemical process is a requirement for achieving significant electrochemical promotion. However, our results also emphasize a low oxygen coverage on Ag which will be detrimental for the lifetime of O^{δ-} ionic promoters. In addition, this low oxygen coverage also explains the poor catalytic performances of the catalyst. We decided to prepare this design of Ag/YSZ catalysts with a low porosity and large micrometric Ag agglomerates to mimic early studies on EPOC for ethylene epoxidation [9,16,17]. However, according to the findings of this study, this design has to be improved to enhance the oxygen coverage on Ag by preparing a catalytic layer containing Ag nanoparticles. Our results also emphasize that micrometric Ag particles interfaced on YSZ can stabilize Ag₂O on their surface at 300°C as shown by Raman spectroscopy. It has been found that the dissolution of oxygen in the Ag bulk or near surface plays a key role [27] in the stabilization of electrophilic oxygen weakly adsorbed on Ag which are considered as the active sites for the production of EO [28]. Therefore, the combination of micrometric and nanometric Ag particles supported on YSZ could lead to a selective catalyst for the ethylene epoxidation.

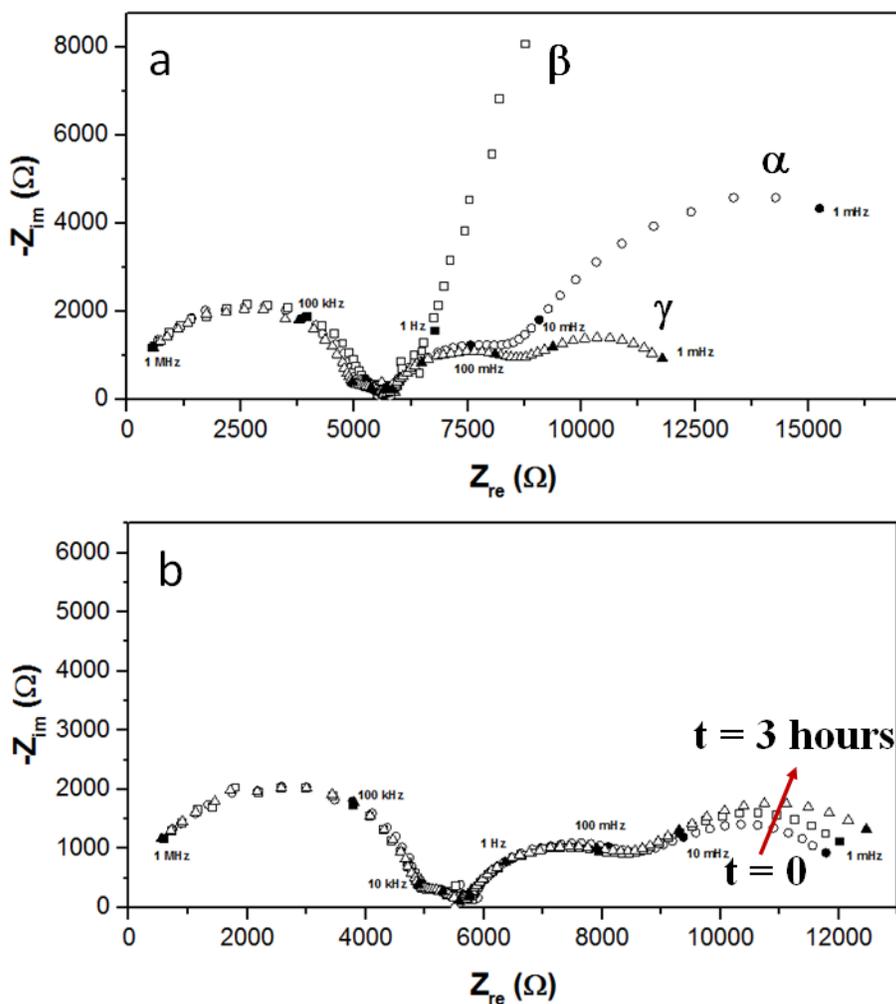


Figure 5. a) Impedance spectra of the Ag/YSZ/Ag symmetric electrochemical catalyst under different reaction atmospheres: α) 1 % O_2 in He (10 L h^{-1} , $300 \text{ }^\circ\text{C}$), β) 1 % O_2 + 4 % C_2H_4 in He (10 L h^{-1} , $300 \text{ }^\circ\text{C}$), γ) 1 % O_2 in He (10 L h^{-1} , $300 \text{ }^\circ\text{C}$) after β (ethylene epoxidation).

b) Impedance spectra of the Ag/YSZ/Ag symmetric electrochemical catalyst under 8% O_2 (10 L h^{-1} , Temperature = $300 \text{ }^\circ\text{C}$): Influence of time in stream after ethylene epoxidation conditions. The first spectrum started at $t = 0$, the second one at $t = 1.5$ hours and the third one at $t = 3$ hours.

4. Conclusions

EIS measurements were performed to relate the O_2 electrode reaction kinetics with the catalytic performances of Ag/YSZ electrochemical catalysts. Experiments were implemented in oxygen and in an oxygen/ethylene mixture representative of the ethylene epoxidation reaction (300°C , ethylene rich composition). Under O_2 , two different polarization resistances were observed, which were attributed to the electron transfer and the oxygen dissociative adsorption, the latter being the rate determining step. This polarization resistance was found

to increase with time, most likely due to the formation of silver oxide species on the surface or sub-surface, as confirmed by in-situ Raman characterizations. The presence of high ethylene excess in the feed hinders the O_2 electrode reaction. This was attributed to the preferential chemisorption of ethylene (in competition with O_2) on the Ag catalyst-electrode, in good agreement with poor measured catalytic performances. These results give new insights for the design of active and selective electro-promoted Ag/YSZ catalysts for ethylene epoxidation.

Acknowledgments

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Appendix A. Supplementary data.

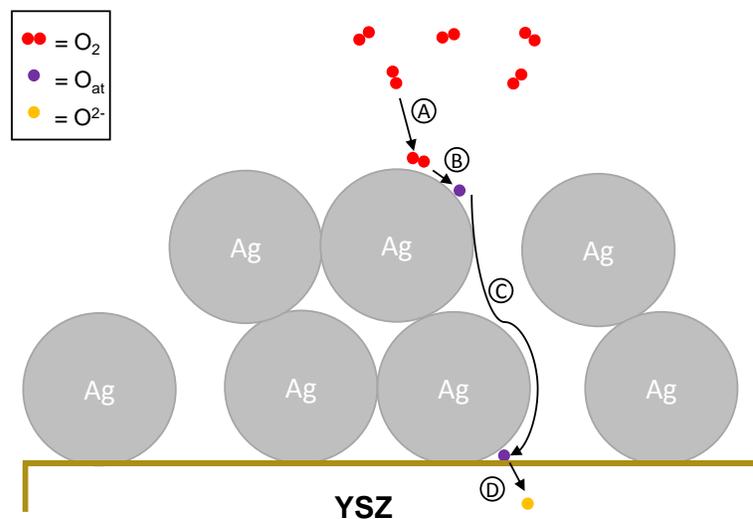


Figure S1. Proposed pathway for O_2 electrode reaction on Ag/YSZ electrochemical catalysts.

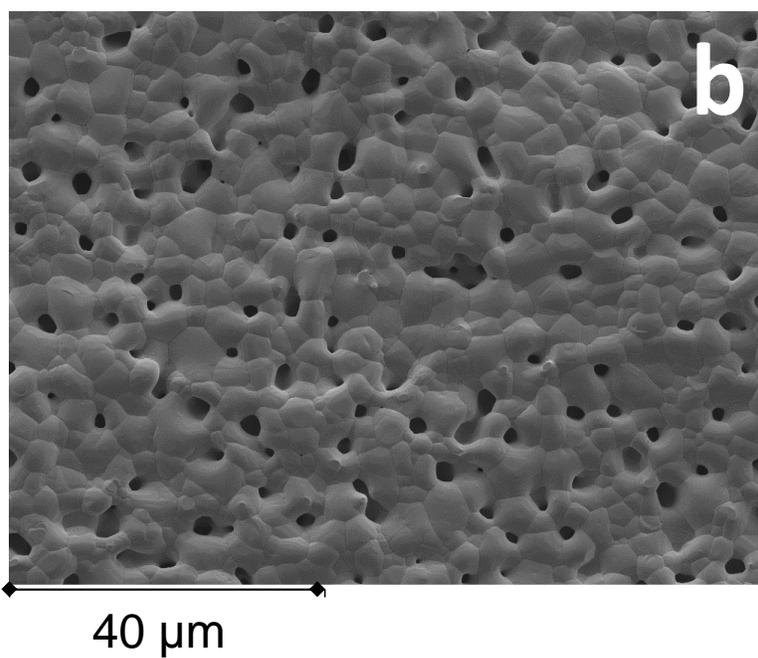
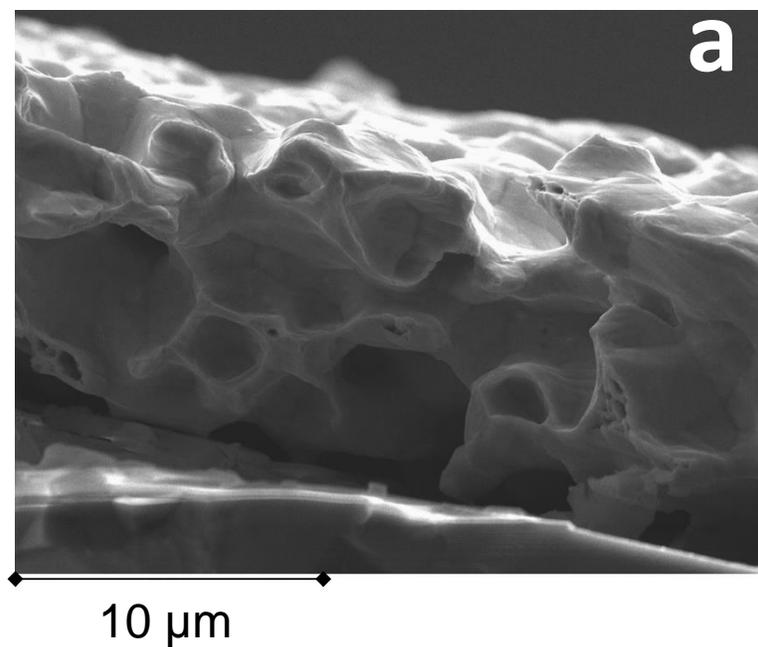


Fig. S2. SEM images of the Ag/YSZ electrochemical catalyst after calcination at 600 °C: a) cross-view, b) surface of the Ag catalyst-electrode.

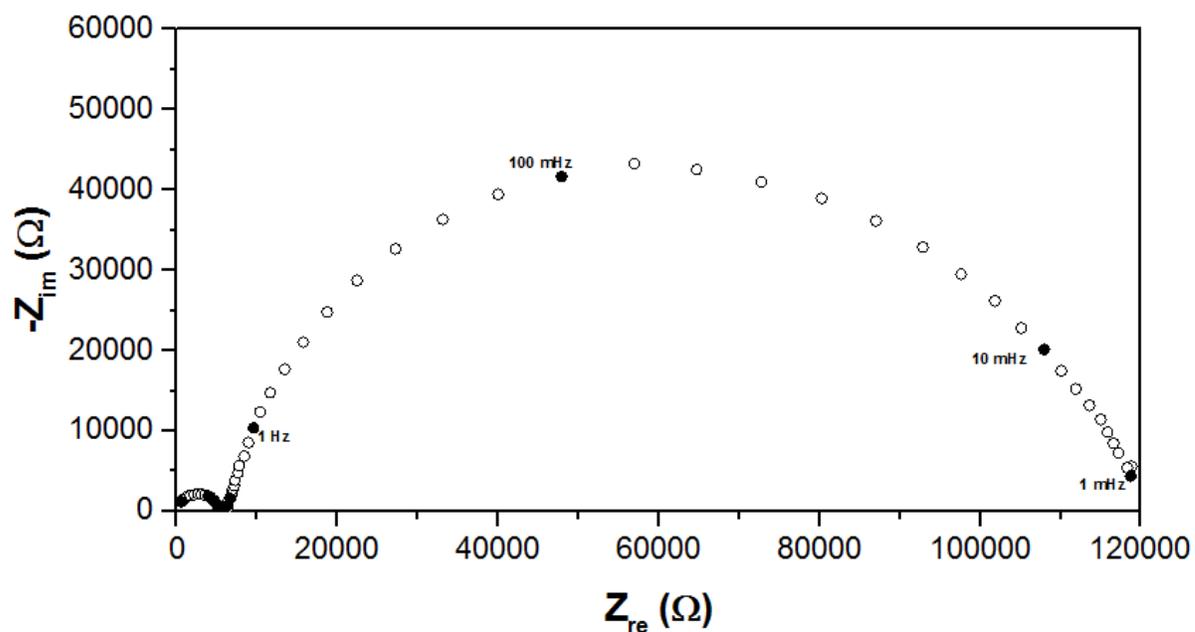


Figure S3. Impedance spectrum of the Ag/YSZ/Ag symmetric electrochemical catalyst under ethylene epoxidation reaction conditions: 1 % O₂ + 4 % C₂H₄ in He (10 L h⁻¹, 300 °C)

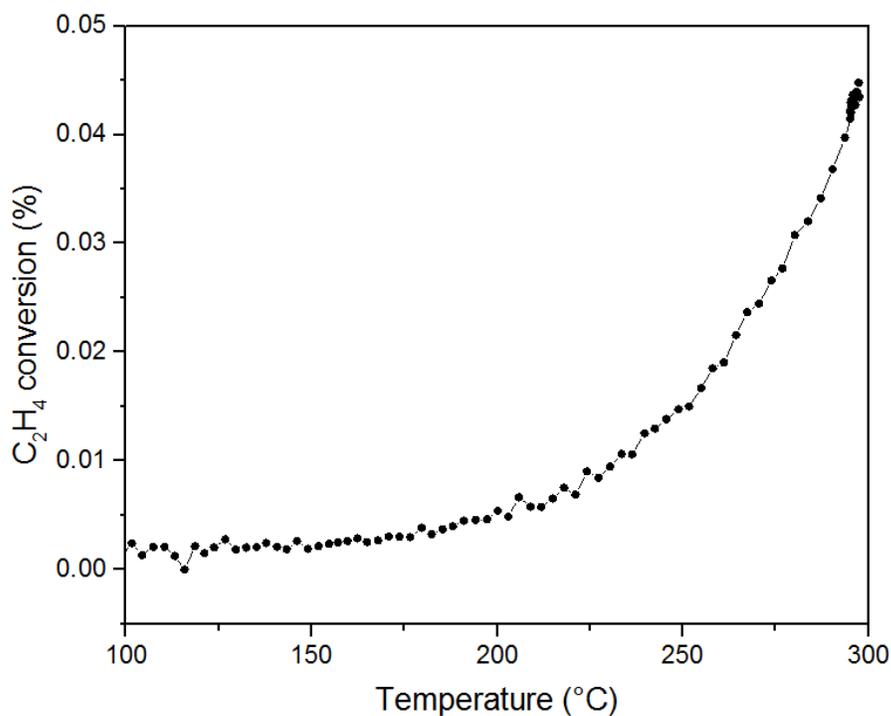


Figure S4. Variation of the ethylene conversion as a function of temperature over the Ag/YSZ/Ag symmetric electrochemical catalyst under ethylene epoxidation reaction conditions: 1 % O₂ + 4 % C₂H₄ in He (3 L h⁻¹, 300 °C).

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