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Analysis of the Performance of the Electrodes in a Natural Gas Assisted Steam Electrolysis Cell

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Abstract

The performance of solid oxide electrolysis (SOE) cells while operating in the natural gas assisted steam electrolysis (NGASE) mode was evaluated. The SOE cells used yttria-stabilized-zirconia (YSZ) as the oxygen ion conducting electrolyte, Co-CeO₂-YSZ as the H₂-H₂O electrode Pd-doped, CeO₂-YSZ CH₄-oxidation electrode. The cell electrochemical performance was evaluated as a function of the H₂O/H₂ ratio and the extent of conversion of CH₄. The results of this study provide insight into the factors that control electrode performance and further demonstrate the viability of an NGASE cell for the production of H₂.

Keywords: catalysis, electrolysis, energy, fuel, hydrogen production, steam

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Introduction

Solid oxide electrolyzers (SOE) have the potential to be efficient devices for the production of high purity hydrogen from water. The ability to produce hydrogen free from impurities other than water is particularly attractive for polymer exchange membrane (PEM) fuel cells where the electrodes are highly susceptible to poisoning by impurities such as CO and sulfur. In a SOE water or a mixture of water and hydrogen is fed to the cathode where the water is dissociated to produce H₂ and oxygen anions that are transported through an oxygen ion conducting ceramic electrolyte (typically yttria-stabilized zirconia, YSZ) to the anode. In conventional designs molecular oxygen is evolved into an air stream on the anode and, thus, the oxygen partial pressure on the anode is significantly higher than that on the cathode producing a large chemical potential barrier for the transport of oxygen through the electrolyte. In order to overcome this barrier an external power supply must be used to “pump” the oxygen anions through the electrolyte. The chemical potential difference between the cathode and the anode and the external electrical power requirement can be reduced significantly by exposing the anode to a reducing gas such as methane that can react with the evolved oxygen. This approach is called natural gas assisted steam electrolysis (NGASE) [Martinez-Frias et al., 2003]. A schematic diagram of an NGASE cell is displayed in Figure 1. Note that the overall reaction being performed in an NGASE cell is equivalent to the steam reforming of methane followed by water gas shift. This is accomplished, however, in a single membrane reactor that separates the CH₄/CO₂ stream from the H₂/H₂O stream, thereby eliminating the need for difficult gas phase separations in order to produce pure H₂.

Another advantage of NGASE is that, at least for some operating conditions, the Nernst potential is positive and the electrolysis reaction will proceed without application of an external potential. Efficient operation in this mode requires, however, electrodes that have high activity for methane oxidation and are stable when exposed to hydrocarbons under reducing conditions. This latter constraint rules out Ni/YSZ cermet electrodes that are commonly used in solid oxide fuel cells (SOFC) since they catalyze the formation of carbon deposits from methane which rapidly degrades performance. Our group has developed alternative electrode designs that circumvent this problem by either replacing Ni with Cu which is not prone to coking [Gorte et al., 2000; Kim et al., 2001; Lu et al., 2003] or using all ceramic electrode formulations [Gross et al., 2007; Gross et al., 2007]. For example, we have shown that electrodes based on thin porous YSZ layers that are impregnated with high loadings of CeO₂ (40 wt %) and dopant levels of a catalytically active metal such as Pd exhibit excellent performance on both H₂ and CH₄ in SOFCs [Gross et al., 2007; Gross et al., 2007]. One of the objectives of the work presented here was to evaluate the performance of electrodes of this design in an NGASE cell. Since there have been very few studies of electrolysis cells operating in the NGASE mode a second objective was to evaluate how the performance of the anode and cathode in an NGASE cell vary as a function of the fuel utilization (i.e. anode and cathode gas composition). This insight is needed to further evaluate the potential of the NGASE approach and to predict the overall performance and efficiency of this type of hydrogen production device.

Experimental

Two identical solid oxide electrolysis cells were used in this investigation and were constructed using a three-layer YSZ (8 mole % yttria) disc which consisted of a dense YSZ electrolyte layer sandwiched between two porous electrode layers. The multi-layer disc was fabricated by laminating three YSZ tapes. The center electrolyte tape contained only YSZ and organic binders, while sacrificial pore formers (graphite and/or polyethylene) were included in the tapes used for the anode and cathode. The tapes were laminated under a pressure of ~ 6 MPa at 340 K and then sintered to 1773 K. During firing the pore formers combusted resulting in a dense, 50 μm thick YSZ electrolyte disc, 1.25 cm in diameter, sandwiched between 15 μm and 300 μm thick porous YSZ layers.

The active components of both electrodes were added using wet impregnation as has been described in detail previously [Huang et al., 2004, 2004, 2005; Lee et al., 2004, 2005; McIntosh et al., 2003; Costa-Nunes et al., 2005]. The 300- μm thick porous electrodes were loaded with 15 wt % CeO_2 and 30 wt % Co by impregnation of aqueous solutions of $\text{Co}(\text{NO}_3)_2$ and $\text{Ce}(\text{NO}_3)_3$. The 15- μm thick porous electrodes were loaded with 40 wt% CeO_2 and 1 wt % Pd by impregnation of aqueous solutions of $\text{Ce}(\text{NO}_3)_3$ and $(\text{NH}_3)_4\text{Pd}(\text{NO}_3)_2$. The 15- μm electrode had an active area of 0.35 cm^2 . The electrical conductivity in the thinner electrode was supplied by the ceria and the Pd dopant was used to enhance the catalytic activity for the oxidation of CH_4 . We have previously shown that SOFC anodes of this design exhibit high performance for the direct oxidation of hydrocarbon-based fuels [Gross et al., 2007].

Electrical contacts were made using Ag wire and Ag paste on both electrodes. The cells were sealed onto 1.0 cm diameter alumina tubes using a ceramic adhesive (Aremco,

Ultra-Temp 516) with the anode on the inside of the tube. This tube was then placed into a second larger tube that was sealed at one end. This two-tube set-up allowed the composition and flow rates of the gasses exposed to both electrodes to be controlled. During electrolysis cell testing the Co-ceria-YSZ electrode was exposed to streams containing mixtures of H₂O and H₂. In some experiments N₂ was included as a diluent. While the flow rate of the carrier gas was maintained at 130 ml/min, the flow rate of the gas sent to the Co-ceria-YSZ electrode varied from 130 to 360 ml/min depending on steam content. The feed gas to the Pd-ceria-YSZ electrode consisted of mixtures of CH₄, H₂O, and CO₂. Since it was not possible to operate the button cells used in this study at high conversions, CO₂ was added to the anode feed gas in order to simulate the gas composition at high conversions. For example, a mixture of CH₄:H₂O:CO₂=10:40:80 was used to simulate 80% conversion. Because the flow of oxygen ions was negligible compared to the gas flow rates, conversion in each compartment was truly differential, so that analysis of exit-gas compositions was not useful.

In addition to being an excellent hydrocarbon oxidation catalyst, Pd/CeO₂ has high activity for steam reforming of methane [Wang and Gorte, 2001]. Thus, some reforming of the methane will occur on the anode. Since the residence time for the gas in the button cell is short, the extent of reforming in the anode is likely to be relatively low. This would not be the case, however, in larger cells running at high conversions where the amount of steam reforming will be much higher. Therefore, in order to more accurately simulate the gas compositions that would be present under realistic operating conditions, the anode gas was passed through a small pre-reactor containing a Pd/CeO₂ catalyst prior to being sent to the electrolysis cell in order to allow the gas-phase

composition to come to equilibrium. The pre-reactor was maintained at the same temperature as the electrolysis cell. To calculate values of the OCV for each operating condition we assumed that the following reforming reactions came to equilibrium within the pre-reactor (only reaction 1 was used when the Pd-ceria-YSZ electrode was exposed to only CH₄ and H₂O):



V-i curves were measured over a potential range from 0.4 to -0.5 V. AC impedance data were recorded in the galvanostatic mode using a Gamry Instruments impedance spectrometer, with a frequency range from 0.01 Hz to 100 kHz and perturbation amplitude of 5 mV for various DC polarizations. Throughout this study, the H₂O/H₂ ratio was maintained below 13. This set the upper limits for P_{O₂} at the Co-ceria-YSZ electrode to be 3×10⁻¹⁹, 6×10⁻¹⁸ and 9×10⁻¹⁷ atm at 973, 1023 and 1073 K, respectively. These are one to two orders of magnitude lower than the equilibrium P_{O₂} required for cobalt oxidation [Gaskell, 2003] insuring that cobalt remained metallic in each experiment. N₂ was used as a diluent for H₂ in order to vary the H₂O/H₂ ratio and H₂O partial pressure over a wide range.

Because reference-electrode measurements tend to be unreliable [Adler, 2000], electrode performance was analyzed the V-i polarization curves and 2-probe impedance spectra. Electrolyte losses are certainly proportional to the current density and can be calculated from the tabulated resistivities of YSZ [Gross, et al, 2007]. It has also been previously shown that the Co-ceria-YSZ electrode exhibits an impedance that is independent of current density in H₂-H₂O mixtures under both SOE and SOFC conditions

[Wang, et al, 2006]. Therefore, the losses within each electrode be calculated from the data provided.

While we did not perform detailed studies of electrode stability in this work, the performance of the cells remained stable for a period of at least several days at 973 K. At 1073 K, there was a gradual loss in the performance of the Pd-ceria-YSZ electrode due to a loss of conductivity associated with sintering of the ceria, as discussed elsewhere in more detail [Gross, et al, 2000].

Results and Discussions

The performance of the electrolysis cell was initially studied as a function of the H₂O/H₂ ratio and H₂O partial pressure in the feed gas to the Co-ceria-YSZ electrode with the composition of the feed gas to the Pd-ceria-YSZ electrode held fixed at 97% CH₄ and 3% H₂O. As noted in the experimental section, the CH₄-H₂O mixture was first contacted with a Pd/CeO₂ catalyst in order to more accurately simulate the conditions expected in a larger area cell. Figure 2 shows V-i curves obtained at 973 K for several H₂O/H₂ ratios. For each curve, from the OCV to 0 potential, the current flow and H₂ generation is driven by the chemical potential difference between the two electrodes and no external power was supplied to the cell. At negative cell potentials, an external power source was used to supply the additional potential required to maintain current flow. For H₂O/H₂ =13 and P_{H₂O} = 0.57 atm, the measured OCV is 0.21 V and the short circuit current is 0.14 A/cm². The measured OCV for this cell is 0.36 V less than the calculated value of 0.57 V. Due to the low reactivity methane, it is likely that for high CH₄ concentrations equilibrium is not established in Pd-ceria-YSZ electrode and this may be the cause of the lower than expected OCV.

As shown in Figure 2, decreasing the H₂O/H₂ ratio to 0.9 while maintaining the P_{H₂O} relatively constant at 0.53 atm resulted in an overall decrease in the cell performance. The OCV for these operating conditions was 0.13 V which is 0.33 V less than the calculated value of 0.46 V. This can again be attributed to the low reactivity of methane and equilibrium not being established on the Pd-ceria-YSZ electrode. The short circuit current for this cell condition was 0.09 A/cm². Note that the primary effect of decreasing the H₂O/H₂ ratio from 13 to 0.9 while holding P_{H₂O} relatively constant was to decrease the OCV, which is expected based on the change in the P_{O₂} with the change in the H₂:H₂O ratio. Except at very low current densities, the slope of the V-i curve remained constant. This indicates that the kinetics of the water dissociation reaction are essentially zero order with respect to the hydrogen partial pressure, since the equal slope implies that the impedance is unchanged.

To study the dependence of P_{H₂O} on the kinetics of the H₂O-H₂ reaction, the cell was also operated with a P_{H₂O} of 0.1 atm and an H₂O/H₂ ratio of 1. The OCV for this gas composition was 0.14 V and the short circuit current was 0.055 A/cm². Note that decreasing the P_{H₂O} by a factor of five from 0.53 to 0.1 atm while holding the H₂O/H₂ ratio nearly constant caused the cell current at each voltage to decrease by roughly 50%. While data at additional P_{H₂O} values are needed to accurately determine the kinetic expression for the H₂O-H₂ reaction, this result suggests that the order of the reaction with respect to P_{H₂O} is less than one. This is an important result since it indicates that it may be possible to run solid oxide water electrolysis cells at relatively high water utilizations and still obtain reasonable performance.

Figure 3 displays data on the effect of the gas composition for the CH₄ side on the cell performance at an operating temperature of 973 K. The goal of this series of experiments was to evaluate how the performance of the electrode varies as a function of the CH₄ conversion. As described in the experimental section, the gas composition for high conversions was simulated by adding H₂O and CO₂ to the CH₄ feed. For example, a mixture of CH₄:CO₂:H₂O = 1:1:2 was used to simulate 50% conversion. This gas stream was passed over a Pd/CeO₂ catalyst at the cell operating temperature in order to allow it to equilibrate and to obtain a composition that would be more representative of that present in a large area cell. The gas composition at the opposite electrode was held fixed with an H₂O/H₂ ratio of 13 and P_{H₂O} = 0.57 atm. For comparison purposes, data is also included for gases consisting of humidified CH₄ (3% H₂O) and humidified H₂ (3% H₂O) at the Pd-ceria-YSZ electrode. These gases were also passed over the Pd/CeO₂ catalyst bed.

For simulated CH₄ conversions of 10, 50, and 80%, the measured OCVs were 0.22, 0.12, and 0.07 V respectively. These values are all within 0.1 V of those calculated using the method described above with the overall difference between the experimental and calculated values decreasing with increasing conversion. The short circuit currents for CH₄ conversions of 10, 50, and 80% were 0.33, 0.17, and 0.09 A/cm² respectively. It is noteworthy that the OCV is still positive even with CH₄ conversions as high as 80%. This indicates that the benefits of using methane as a reducing gas on the anode are maintained for relatively high CH₄ conversions and further demonstrates the viability of the NGASE approach.

Note that the slopes of the V-i curves for the 10, 50 and 80% CH₄ conversion data are nearly the same. This demonstrates that while increasing the CH₄ conversion lowers the OCV, it has little effect on the kinetics of the electrode reaction. This is further confirmed by the impedance data presented in Figure 4 which shows that the total area-specific resistance (ASR) of the electrodes is ~0.35 Ω cm² for the 10, 50, and 80% CH₄ conversions. In contrast to these results, the V-i curve for humidified CH₄ has a much higher slope corresponding to an ASR of 1.1 Ω cm². The origin of this difference in the electrode ASR when using humidified CH₄ compared to CH₄ streams with simulated conversion of 10% or greater becomes apparent when one examines the results obtained when exposing the Pd-ceria-YSZ electrode to humidified H₂ with 3% H₂O. Note that the V-i curves for humidified H₂ and 10% CH₄ conversion in Figure 3 are almost the same. The impedance data in Figure 4 also show that the total electrode ASR for both gasses is ~0.35 Ω cm². The most likely explanation for this similarity in the electrode performance for these two different gas compositions is that steam reforming over the Pd/CeO₂ catalyst results in a high concentration of H₂ in the 10% CH₄ conversion gas stream. The kinetics of the electrode reaction and the performance of the Pd-ceria-YSZ electrode are thus more representative of the operation on H₂ rather than that for the direct oxidation of CH₄. Since the electrode ASR was the same for the 50 and 80% CH₄ conversions, the electrode performance for these conditions also appears to be dominated by the H₂ oxidation reaction.

Finally, in addition to 973 K, we also investigated the performance of the NGASE cell at both 1023 and 1073 K. As expected, increasing the temperature resulted in slight increases in the OCV values and decreases in the ASR. It is useful to compare the

current obtained under short circuit conditions (0 V) as a function of temperature and CH₄ conversion, since this represents the transition point from where the cell runs spontaneously to where an external potential must be supplied to pump oxygen through the electrolyte. The short circuit current densities as a function of CH₄ conversion and temperature are listed in Table 1. These data were again obtained using a H₂O/H₂ ratio of 13 and P_{H₂O} = 0.57 atm at the opposite electrode. For all three conversions studied the short circuit current density roughly doubled upon heating from 973 to 1073 K. Also note that for a CH₄ conversion of 10% a short circuit current, which corresponds to one-half the H₂ production rate of 0.49 A/cm², was obtained at 1073 K and decreased to 0.09 A/cm² for 80% CH₄ conversion. In large area NGASE cells operated at high CH₄ utilization, the CH₄ conversion will vary across the cell and therefore the hydrogen production rate would be expected to be intermediate between these two values. The performance results obtained from the button cells used in this study suggest, however, that reasonable H₂ production rates could be obtained in a NGASE system without the need to provide additional electrical power to pump oxygen across the electrolyte.

Conclusions

In this study we have investigated the effect of the electrode gas compositions on the performance of an NGASE cell with a YSZ electrolyte, a Co/CeO₂/YSZ composite as the H₂-H₂O electrode and a Pd-doped CeO₂/YSZ composite as the CH₄ electrode. Studies of the cell performance as a function of the H₂O/H₂ ratio in the gas fed to the cathode indicate that the kinetics of the cathode reaction are nearly zero order in H₂ and less than 1st order in H₂O. Studies of the NGASE cell performance for simulated CH₄ conversions between 10 and 80 % showed that the kinetics of the oxidation reactions on

the Pd-doped CeO₂/YSZ composite anode were not a strong function of conversion. This result along with comparison to the performance when operating on humidified H₂ indicates that for these conditions the oxidation of H₂ produced by steam reforming of the CH₄ determines the performance of the anode. At low CH₄ conversions, however, direct oxidation of methane dominates and resulting in a higher ASR. Finally, the results of this study further demonstrate the viability of an NGASE cell for the production of H₂ and indicate that it may be possible to obtain high H₂ production rates without the need for applying an external potential to the cell.

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Figure captions

Figure 1. Schematic of a natural gas assisted steam electrolysis cell.

Figure 2. V-i curve as a function of composition of the cathode gas at 973 K. (○)

$\text{H}_2\text{O}/\text{H}_2=13$, $P_{\text{H}_2\text{O}} = 0.57$ atm, (◇) $\text{H}_2\text{O}/\text{H}_2=0.9$, $P_{\text{H}_2\text{O}} = 0.53$, and (Δ)

$\text{H}_2\text{O}/\text{H}_2=1$, $P_{\text{H}_2\text{O}} = 0.1$. The anode gas consisted of $\text{CH}_4+3\%\text{H}_2\text{O}$.

Figure 3. V-i curve as a function of anode gas composition at 973 K. Anode gas

composition: (+) $\text{H}_2+3\%\text{H}_2\text{O}$, (◇) CH_4 , 10% conversion, (○) $\text{CH}_4+3\%\text{H}_2\text{O}$, (Δ)

CH_4 , 50% conversion, and (X) CH_4 , 80% conversion 80%. The cathode gas

consisted had $\text{H}_2\text{O}/\text{H}_2=13$ and $P_{\text{H}_2\text{O}} = 0.57$ atm.

Figure 4. AC impedance spectra as a function CH_4 conversion at 973 K. Steam

electrode: $\text{H}_2\text{O}/\text{H}_2=13$, $p_{\text{H}_2\text{O}} = 0.57$ atm.

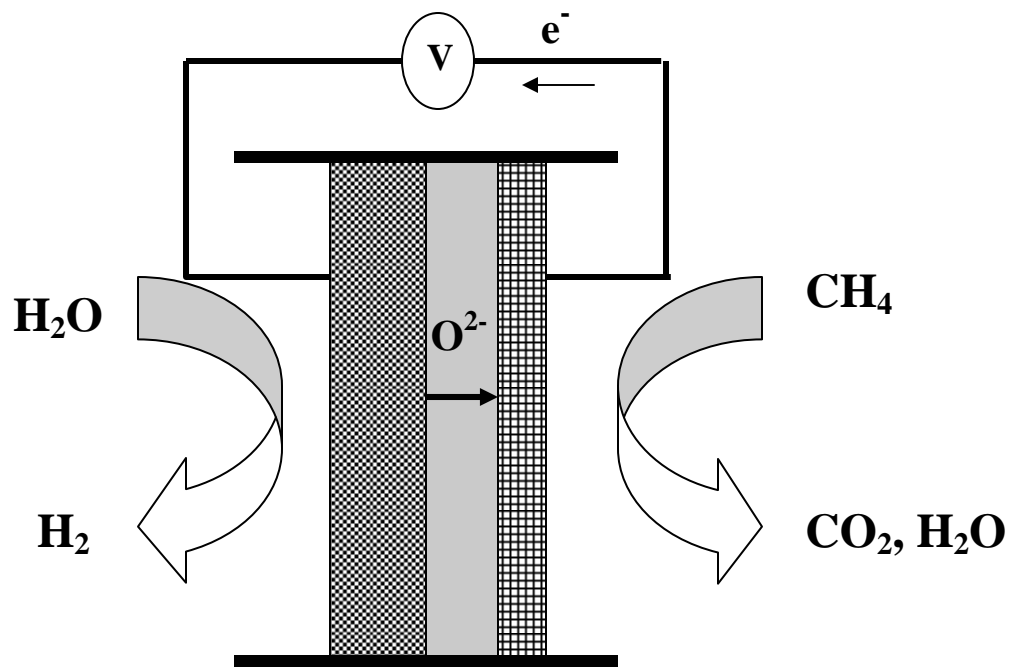


Figure 1. Schematic of a natural gas assisted steam electrolysis cell.

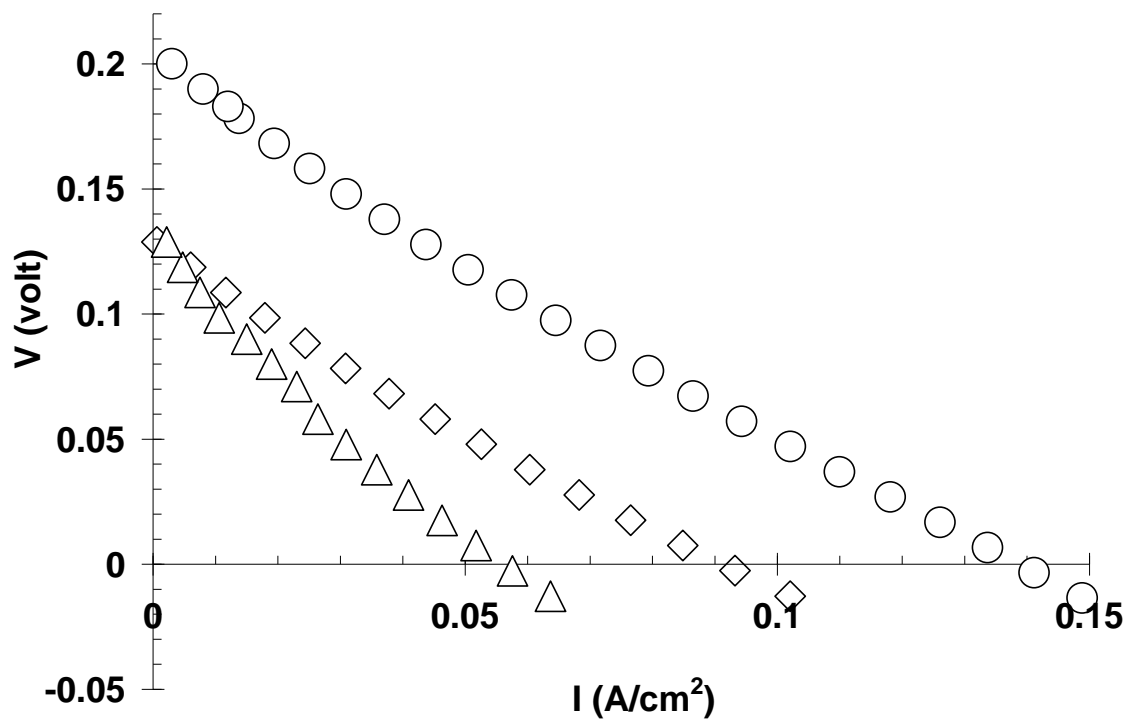


Figure 2. V-i curve as a function of composition of the cathode gas. (○) $H_2O/H_2=13$, $p_{H_2O} = 0.57$ atm, (◇) $H_2O/H_2=0.9$, $p_{H_2O} = 0.53$, and (△) $H_2O/H_2=1$, $p_{H_2O} = 0.1$. The anode gas consisted of $CH_4+3\%H_2O$.

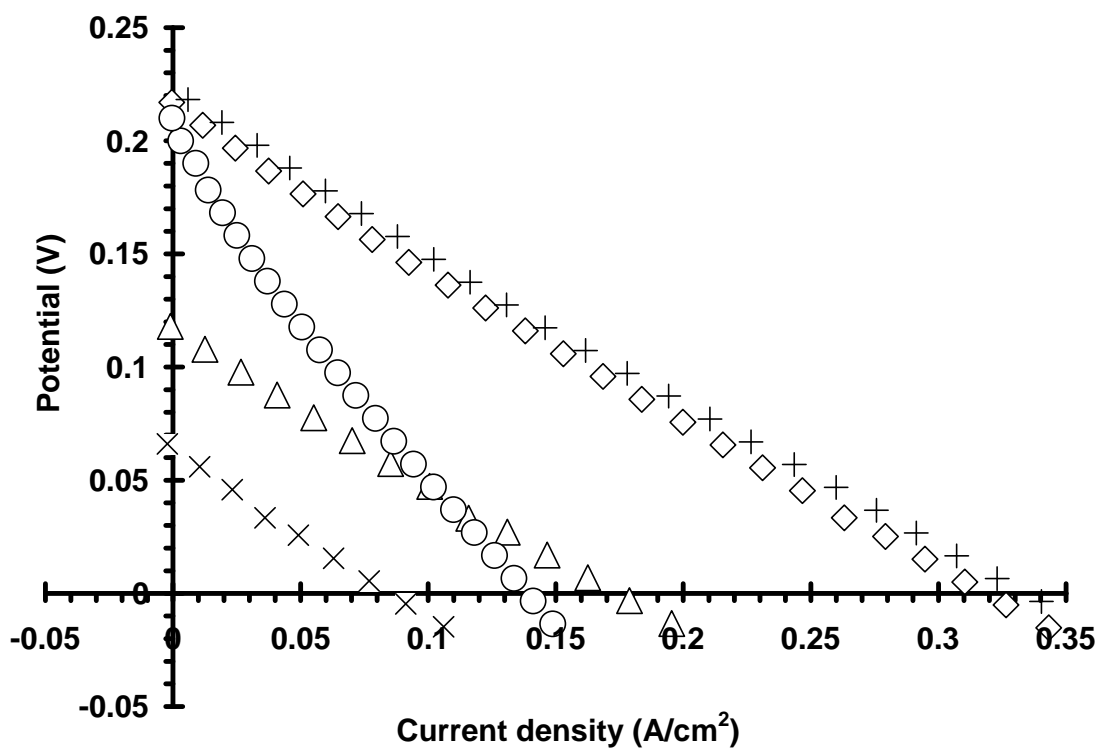


Figure 3. V-i curve as a function of anode gas composition at 973 K. Anode gas composition: (+) $\text{H}_2+3\%\text{H}_2\text{O}$, (\diamond) CH_4 , 10% conversion, (\circ) $\text{CH}_4+3\%\text{H}_2\text{O}$, (Δ) CH_4 , 50% conversion, and (X) CH_4 , 80% conversion. The cathode gas consisted had $\text{H}_2\text{O}/\text{H}_2=13$ and $p_{\text{H}_2\text{O}} = 0.57$ atm.

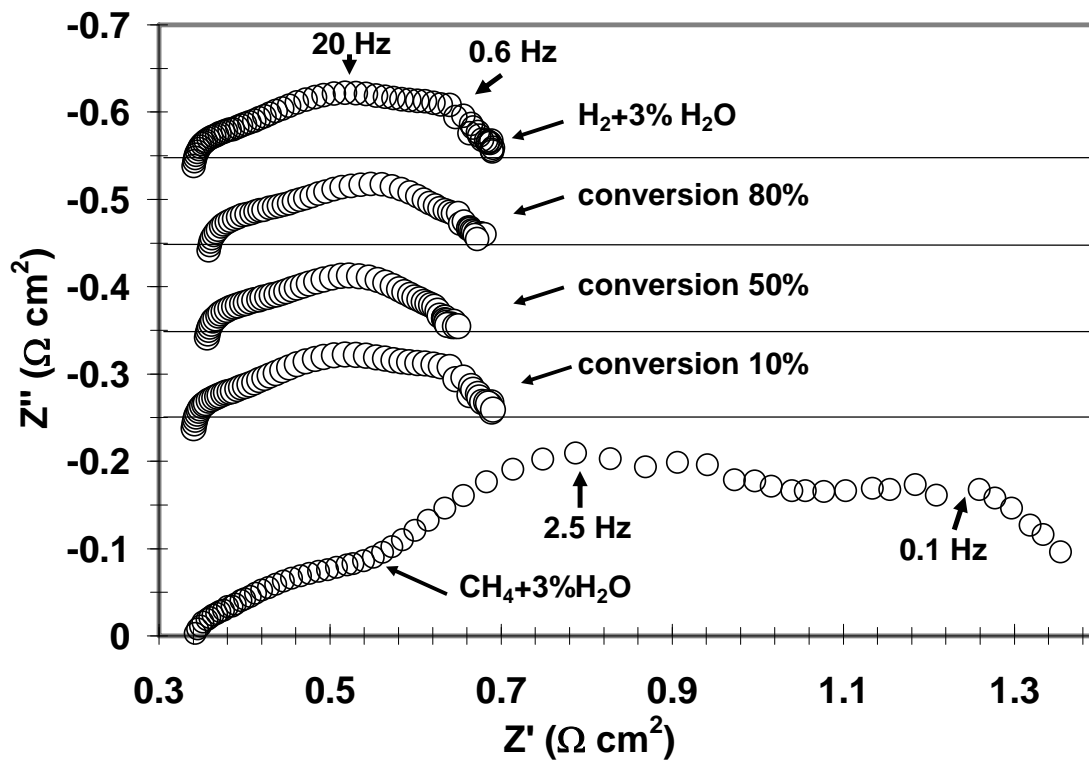


Figure 4. AC impedance spectra as a function CH_4 conversion at 973 K. Steam electrode: $\text{H}_2\text{O}/\text{H}_2=13$, $p_{\text{H}_2\text{O}} = 0.57 \text{ atm}$.

Table 1. The short circuit current densities as a function of CH₄ conversion and temperature

T (K)	Short circuit current density (A/cm ²)		
	10% CH ₄ conversion	50% CH ₄ conversion	80% CH ₄ conversion
973	0.25	0.1	0.05
1023	0.26	0.14	0.085
1073	0.49	0.19	0.09