



## Triode operation of CO poisoned PEM fuel cells: Fixed and cyclic potential triode operation

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### ABSTRACT

An alternative approach for enhancing the Polymer Electrolyte Membrane Fuel Cell (PEMFC) performance under CO poisoning conditions was investigated by the use of the recently developed triode fuel cell design and operation. In this mode of operation a third, auxiliary, electrode is introduced in addition to the anode and cathode electrodes. It is shown that the application of electrolytic potential between the cathode and an auxiliary electrode can affect the performance of the fuel cell circuit in a controllable and reversible manner. The applied electrolytic current or potential can be fixed (steady-state) or can be cycled between chosen limits. In the present study, both triode operation modes were investigated and it was found that the time-averaged power output of the fuel cell can be increased up to three times.

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### 1. Introduction

Triode operation is a recently developed method for enhancing the power output and thermodynamic efficiency of batteries and fuel cells [1,2]. A triode fuel cell consists of three electrodes, i.e. apart from anode and cathode, a third electrode is also introduced. Hence, in addition to the conventional fuel cell circuit, a second auxiliary circuit is formed which is run in the electrolytic mode and permits operation of the fuel cell circuit under previously inaccessible anode–cathode potential differences [1]. Thus during triode operation some power is sacrificed in the auxiliary circuit in order to enhance the overall production of electrical power.

The concept of the triode fuel cell operation was first developed and validated using a Solid Oxide Fuel Cell (SOFC) with polarizable Pt electrodes [1]. Afterwards, the triode fuel cell design was demonstrated in state-of-the-art Polymer Electrolyte Membrane Fuel Cells (PEMFCs) operated in absence of noble metals in the anode [3] and also in PEMFCs using Pt-based anodes operated under severe CO poisoning [4] and under mild CO poisoning conditions [5]. These studies have shown that the power output of the fuel cell can be very significantly increased via application of auxiliary potential or current. Furthermore, this increase,  $\Delta P_{fc}$ , was found to be comparable with or higher than the power,  $P_{aux}$ , sacrificed in the auxiliary circuit [1–5]. Recently, Cloutier and Wilkinson [6] have pioneered the use of the triode design of a PEMFC type reactor in order to improve the

electrolysis of water and liquid methanol in a non-flowing system at ambient conditions.

Fig. 1a shows schematically the triode PEMFC design. The three electrodes are all in electrolytic contact and form two electrical circuits (Fig. 1a):

- (i) the fuel cell circuit, which comprises the anode, the cathode, and a variable resistance,  $R_{ext}$ , for dissipating the electrical power,  $P_{fc}$ , produced, and
- (ii) the auxiliary circuit, which comprises the auxiliary electrode, the cathode of the fuel cell and a potentiostat–galvanostat.

When the applied auxiliary current,  $I_{aux}$ , is zero then the fuel cell operates in the conventional mode. It produces a power, denoted by  $P_{fc}^0$ , which is the product of the fuel cell potential,  $\Delta V_{fc}^0$ , and of the fuel cell current,  $I_{fc}^0$ . Both  $\Delta V_{fc}^0$  and  $I_{fc}^0$  vary as the external resistive load of the fuel cell circuit,  $R_{ext}$ , is varied.

The triode fuel cell operation consists of imposing an electrolytic current  $I_{aux} < 0$  or potential on the auxiliary system. In this way protons migrate from the fuel cell cathode to the auxiliary electrode (Fig. 1a).

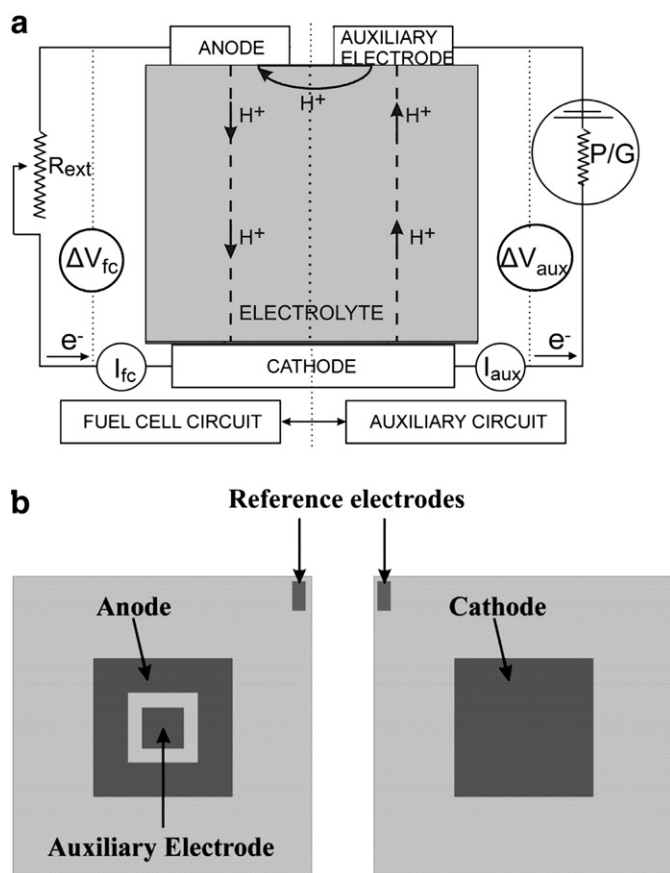
Denoting by  $I_{far}$  the net Faradaic fuel-consuming current, one notes that in view of Fig. 1 and Kirchoff's first law it is:

$$I_{far} = I_{fc} + I_{aux} \quad (1)$$

where  $I_{aux} < 0$  for triode operation, and the net Faradaic current  $I_{far}$  corresponds via Faraday's Law to the net consumption of fuel at the anode or the net consumption of  $O_2$  at the cathode.

It is well known, that one of the main problems associated with the practical utilization of PEMFC units is that of CO poisoning of

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**Fig. 1.** Schematic of the triode fuel cell concept, showing the fuel cell and auxiliary circuit in detail (a) and the geometry of the membrane electrode assembly (b). The fuel cell cathode acts simultaneously as an electrode of the auxiliary circuit. P/G: potentiostat-galvanostat.

the Pt-based anode [7–9]. The aim of the present study is to study the possibility of enhancing the performance of a CO poisoned PEMFC using the triode operation, both under steady-state (fixed) and under cyclic potential application in the auxiliary circuit.

## 2. Experimental

A state-of-the-art PEMFC [3] was used (NuVant) with state-of-the-art Pt and Pt-Ru electrodes deposited by NuVant on E-TEK carbon cloth and equipped with a third auxiliary electrode.

The exact geometry of the membrane electrode assembly (MEA) used in the present investigation is shown in Fig. 1b. The MEA also includes two reference electrodes which permit direct measurement of the anodic and cathodic overpotential.

The loading of the Pt electrode was  $0.5 \text{ mg Pt/cm}^2$  (unsupported Pt black) and the loading of the Pt (30%) Ru (15%), supported on Vulcan XC-72 carbon, was  $0.5 \text{ mg/cm}^2$ . The superficial surface area of the cathode (Pt) was  $5.29 \text{ cm}^2$ , of the anode (PtRu) was  $3.85 \text{ cm}^2$  and of the auxiliary electrode (PtRu) was  $0.49 \text{ cm}^2$  (Fig. 1b). The cathode was a square and the auxiliary electrode was a smaller square located in the center of the hollow square anode (Fig. 1b). The membrane was Nafion 117 with nominal thickness of  $185 \mu\text{m}$ . The membrane electrode assembly (MEA) was prepared by hot pressing in a model C Carver hot press at  $120 \text{ }^\circ\text{C}$  and under pressure of 1 metric ton for 3 min.

Two cells were used in order to check reproducibility and both gave practically the same results. Preliminary investigation showed that the applied auxiliary potential,  $\Delta V_{aux}$ , should not exceed  $-1.9 \text{ V}$ , as this leads to  $\text{CO}_2$  formation at the cathode via oxidation of the

carbon support. The concentration of CO and  $\text{CO}_2$  in the anode and cathode feed and effluent was monitored using a Fuji Electric's Infrared Analyzer ZRJ-4UNOR 6 N IR CO/ $\text{CO}_2$  analyzer.

The gas feeds to the cathode and anode compartments (the latter includes also the auxiliary electrode, Fig. 1) were continuously humidified using thermostated gas saturators. The cell temperature was typically set at the same temperature ( $25 \text{ }^\circ\text{C}$ ) with the gas saturators. The anode compartment gas feed was Messer Griesheim certified gas mixtures of 500 ppm CO/ $\text{H}_2$ , which could be further diluted with Air Liquide (N4.5)  $\text{H}_2$ . The cathode feed was humidified by Air Liquide synthetic air.

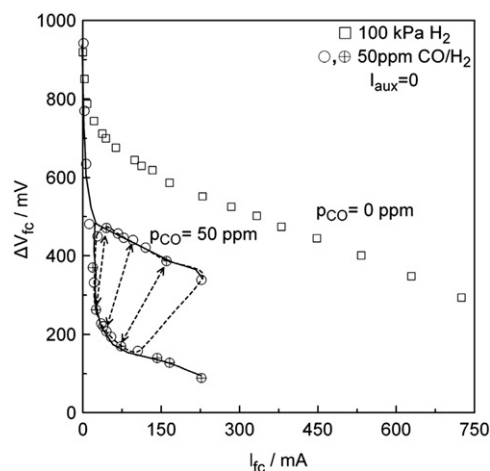
The fuel cell circuit included a decade resistance Box (Time Electronics Ltd 1051) in order to vary the external load. The current and the potential were measured by two digital multimeters (Metex ME 21). For the fixed triode operation, constant potentials or currents in the auxiliary circuit were applied using an AMEL 553 Potentiostat-Galvanostat. For the cyclic triode operation an AMEL 567 Function Generator was used together with the Potentiostat-Galvanostat.

It is evident from Fig. 1a that all three electrodes operate in a corrosion-type mode with part of their surface used for oxidation and part of their surface used for reduction. Despite this rather intense mode of operation no performance deterioration was observed during operation for several weeks.

## 3. Results and discussion

### 3.1. Conventional fuel cell operation

Fig. 2 shows the polarization curves obtained in the conventional fuel cell mode ( $I_{aux} = 0$ ) by varying the external resistance,  $R_{ext}$ , of the decade box from zero to  $200 \Omega$ . Data were collected both under 100 kPa hydrogen supply to the anode (squares) and under CO poisoning conditions a  $\text{H}_2$  stream containing 50 ppm CO, denoted hereafter 50 ppm CO/ $\text{H}_2$  mixture (cycles). As expected, one observes that the presence of CO causes a significant decrease in power output and creates three distinct regions [9]. In the high potential-low current region ( $R_{ex} > 19 \Omega$ ) and in the low potential-high current region ( $R_{ex} < 1 \Omega$ ) stable steady-state behavior is observed, while in the intermediate region ( $1 \Omega < R_{ex} < 19 \Omega$ ) self-sustained potential and current oscillations are obtained with a period of the order of 7 to 10 s. Similar oscillatory phenomena have been reported in the literature [3,10].



**Fig. 2.**  $I_{fc} - \Delta V_{fc}$  curves of the fuel cell circuit obtained with the conventional operation mode ( $I_{aux} = 0$ ) by varying the external resistive load,  $R_{ext}$ . Anode feed: 50 ppm CO/ $\text{H}_2$  (cycles).

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