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Fully automatic test facilities for the characterisation of DMFC and PEFC MEAs

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Abstract

Membrane electrode assemblies (MEAs) for polymer electrolyte fuel cells with hydrogen fuel (H_2-PEFC) and direct methanol fuel cells (DMFC) are under development at DLR. For their characterisation fully automatic test units have been designed and realised to guarantee reproducible test results.

The identical oxidant supply at the cathode side of the H_2-PEFC and DMFC as well as similar test cells and test conditions offer the possibility to realise both modifications in one test unit. The pipework system and all fittings of the cathode supply can be used simultaneously. Different conditions have to be realised particularly in the anode supply. At the anode of the DMFC liquids (methanol/water) and in the H_2-PEFC gas (hydrogen) are supplied.

By integration of an electronic software-supported control unit operating modes can be changed in the test unit depending upon requirement. In order to show the reproducibility of fuel cell operations it is necessary that parameters will be kept within very low deviation limits. An automatic regulation permits impact onto all controllable parameters e.g. pressures, temperatures and mass flow rates. When achieving stationary operating conditions current–voltage-curves can be recorded by automatic change of the electronic cell load. Measured values for current, voltage and all operating parameters are recorded by the software and stored for later interpretation. During data acquisition the parameters are visualised on a graphic interface. It is possible to influence the control at any time.

To permit an unguarded long-term experimental operation a sophisticated safety system is necessary. The pre-defined safety parameters are monitored by computer software as well as by an industrial type Programmable Logic Controller (PLC).

Keywords: MEAs; H_2-PEFC; DMFC

1. Introduction

The development of polymer electrolyte fuel cells (PEFC) requires intensive experimental work for investigating the characteristics of this technology. It is important to make available an experimentation platform that permits a wide range of parameter variations.

In order to understand the behaviour of PEFCs, numerous experiments with a multiplicity of parameter variations are necessary.

Operating parameters which have influence on the behaviour and performance of a fuel cell and which are given by the test facility are:

- cell temperature;
- pressure;
- composition of the supplied media (methanol concentration, oxygen partial pressure);
- mass flows;
- humidity condition in the cell.

A laboratory test set-up must have the potential to examine reliably, to visualise and to document as well these parameters.

A test set-up that meets these requirements was developed and built at DLR.

A completely integrated Programmable Logic Controller (PLC) makes it possible to set given parameters automatically, to visualise currently measured values and to store these values for a later evaluation. Furthermore it should be possible to set previously defined operating points successively in a fully automatic way. Current–voltage characteristics have to be recorded under quasi-stable conditions. This allows to make statements about the performance of fuel cells [1].

A comprehensive security concept serves to conduct unguarded experiments. This makes continuous long-term tests possible.

Of this standard 10 test set-ups for PEFC, three for DMFC and three for both PEFC and DMFC MEAs are

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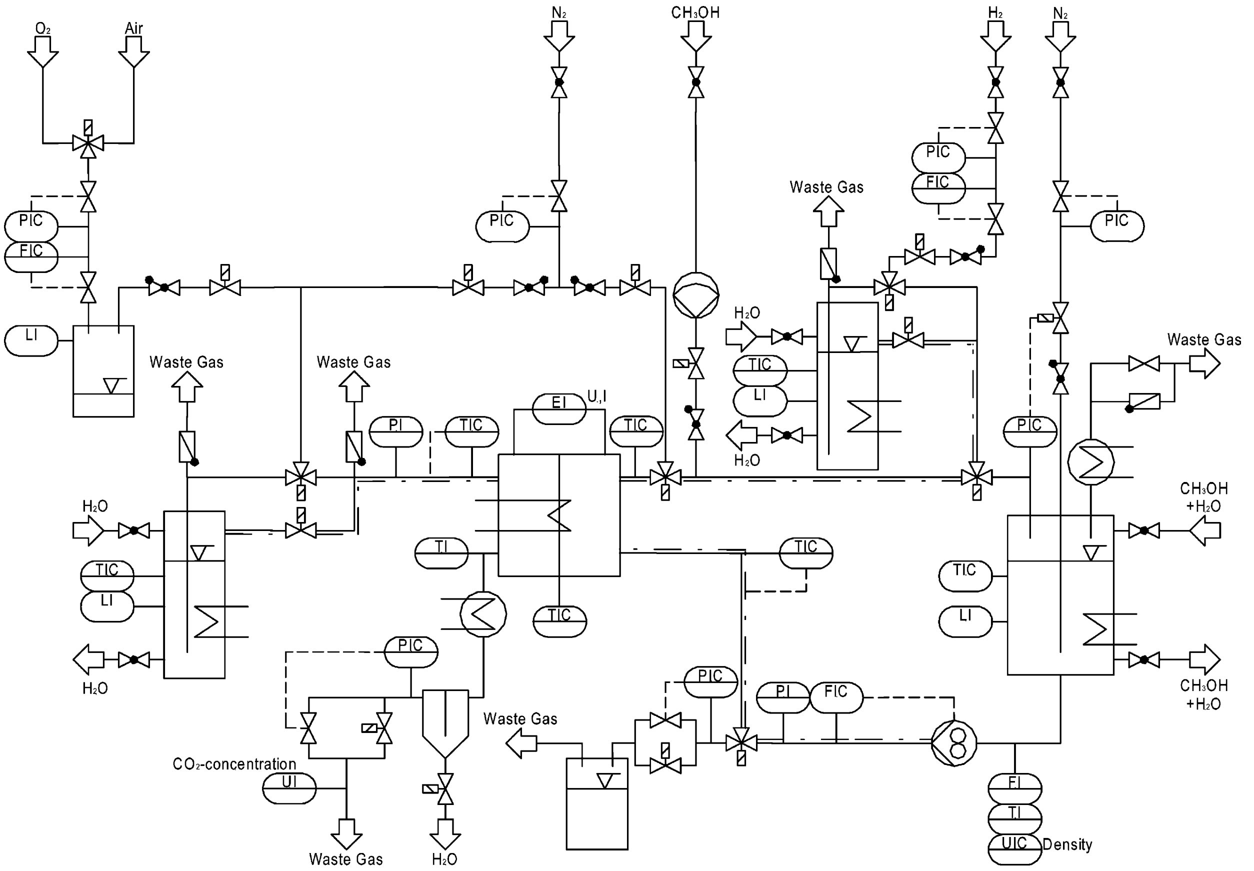

Fig. 1. Flow sheet of a fully automatic test facility.

now available at DLR. A typical test facility is described in the following and displayed in Fig. 1.

2. Functionality of the test facilities

2.1. Requirements of the test facilities

In order to ensure sufficiently large reproducibility it is important to be able to accomplish analyses under always the same parameters and conditions. To make this possible, standard procedures should be automated as far as possible. Furthermore it is necessary that given parameters will be kept within very low deviation limits.

For a later evaluation and to compare different results, it is necessary to acquire and store data. To ensure this independently of the user, measured values in a given time interval are to be saved.

To be able to adapt the test facilities later to new experimental requirements a sufficient continuity in planning and realisation of equipment must be ensured. That means for example that the compatibility between old and new construction units has to be guaranteed.

In order to ensure stand alone experiments and to exclude faulty operations all parameters have to be supervised by the control unit (Table 1).

2.2. Cathode supply

The gas supply of the cathode can be performed alternatively via oxygen or air. The switching of the supply gas is done automatically. A mass flow controller provides a

Table 1
Adjustable parameters of the test facility

Parameter	Unit	Value	Remark
Media temperatures/humidifier	°C	<160	
Cell temperature	°C	<160	
Pressure	bar_a	<6	bar_a means absolute pressure
Flow rate (methanol/water-mixture)	ml/min	10–500	Higher flows by exchanging pump head possible
Deviation limit of the methanol-concentration	mol/l	<0.1	
Flow rate, oxygen/hydrogen/air/nitrogen	sccm/min	0–5000	Higher flows by exchanging flow controller possible

constant gas flow. To avoid pressure fluctuations a buffer tank is integrated in the supply line. A further option, offered by the test facility is the possibility to humidify the cathode gas. The gas can be passed trough a heated container filled with water. Humidity control is done by controlling the temperature in the humidifier. To prevent a condensation of water at the cell inlet this part is heated. At the cell exit a condenser with filter separator is arranged downstream to dehumidify the gas flow. At the cathode exit an electronic pressure control valve generates defined pressure ratios in the cell. Pressure transmitters at cathode entrance and exit enable an accurate control and inform about the pressure loss in the cell.

For the determination of the methanol cross over on the cathode exit the CO_2-concentration is measured.

2.3. Anode supply

In order to test cells alternatively in hydrogen operation mode or in direct methanol operation mode, the possibility exists to change the anode fuel supply between both variants.

2.3.1. Hydrogen operation mode

If the supply of the anode is done with hydrogen, this happens according to the same principle as the cathode supply. A constant gas flow which can be humidified is adjusted by a mass flow controller. The heated gas is supplied to the cell and a pressure control valve is arranged behind the cell to adjust the gas pressure.

2.3.2. Methanol operation mode

A second possibility to feed the anode represents methanol. A tank serves to store a mixture of methanol and water. A speed adjustable gear pump sucks the mixture and pumps it into the cycle. With the help of a Coriolis-meter temperature and density of the mixture are determined. Based on the assumption that the mixture is incompressible the concentration ($c = f(\rho, T)$) can be determined by a fit curve calibrated before. If the methanol concentration decreases pure methanol can be dosed via a diaphragm pump.

The mixture can be preheated by a tubing heating to ensure constant conditions in the cell. At ambient pressure methanol has a boiling point of 64.7 °C. To ensure the supply in the liquid phase it is necessary to put the feed under pressure. The pressure in the tank is measured and regulated by pressurising with nitrogen gas. To adjust pressure fluctuations caused for example by developing carbon dioxide the tank is connected with the environment by a pressure control valve. If nevertheless critical increase of pressure occurs, the pressure control valve is bypassed by a security valve.

2.4. Electrical components

For adjusting the electrical load of the cell and for the measurement of current–voltage characteristics, an electronic load is integrated into the test facility. This load can be operated potentiostatically or galvanostatically. The cell current is measured by a shunt. A shunt is a precisely defined resistance with which the voltage drop, proportional to the current, can be measured.

2.5. Automatic adjustment of parameters

It is necessary to treat each cell similarly to ensure a sufficient reproducibility. This is reached by defined values for all given parameters. By the PLC preset operating points are successively adjusted and held at quasi-stable condition. Current–voltage characteristics are registered in the preset operating points. After analysis the test facility is shut down in a defined way.

2.6. Visualisation/data acquisition

At the test facility a visualisation is needed to observe the process and to display momentary measured values (Fig. 2). The histogram makes it possible to interpret the trends. The following values are displayed:

- cell voltage;
- cell current;
- pressures (media supply, humidifier, tank etc.);
- temperatures (cell, media supply, humidifier, tank etc.);
- concentration of the methanol/water mixture.

For the later evaluation of the data these values are permanently acquired and stored in adjustable intervals from 1 to 999 s. A computer generated text file can be evaluated afterwards by means of a spread-sheet program or a similar software.

This complete recording of measured values can also give information about transient processes.

2.7. Analytical equipment

The recording of current-density–voltage characteristics is most important and a common characterisation method for electrochemical systems. The usual measuring range for direct methanol fuel cells extends from 50 mV to the open circuit voltage, which is usually about 700–800 mV. The incrementation amount is 50 mV. The deviation of the value after the stage change and thus leaving the stationary operating point, is called transient.

Because the performance of a PE-fuel cell is strongly dependent on the water content in the membrane, it is desirable to have a measure of humidification in the membrane. This is realised by continuous measurement of the cell impedance at a high frequency (10 kHz). At this frequency the impedance is dominated by the ohmic resistance in the cell and thus by the membrane conductivity and the contact resistances [2].

In the DMFC mode a measure for the amount of methanol that permeates through the membrane is requested. This is realised by measuring the CO_2 content in the cathode outlet

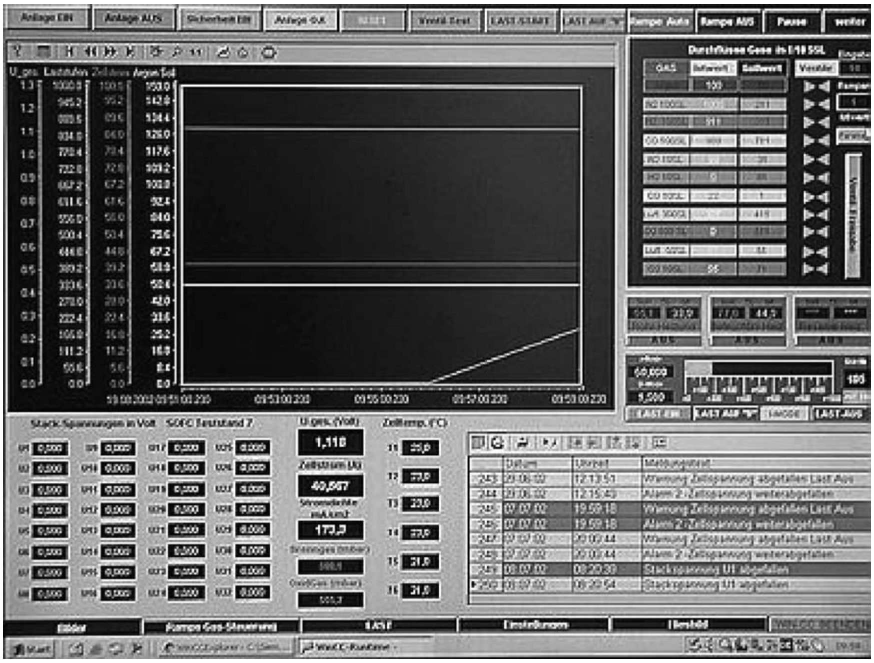

Fig. 2. Visualisation of present and recently measured values.

gas with an infrared measuring device. The amount of methanol that has been oxidised to CO_2 at the cathode is registered in the measuring device. Knowing the cathodic gas flow rate the amount of methanol can be calculated. However this CO_2 content does not give the absolute amount of methanol that permeates through the membrane. The methanol that reaches the cathode might not be completely be oxidised to CO_2. The methanol permeation value calculated from the CO_2 cathode outlet content would then be too low. Also CO_2 from the anode can permeate through the membrane and increase the CO_2 value at the cathode outlet [3]. In this case the calculated methanol permeation value would be too high. However when different membranes are considered the relative methanol permeation calculated from the CO_2 content in the cathode outlet using the same electrodes and the same operating conditions can be used for comparison.

It is also possible to measure the current-density distribution over a 25 cm^2 fuel cell without interfering with the cell operation. For this purpose a cell was designed and built at DLR that can be used as anode or cathode flow field and MEA holder [4]. The whole cell is divided into 16 segments where each segment is insulated against each other. The current values of each region of the cell are measured, registered and on-line displayed on a computer. Current-density distributions can be registered at time intervals as low as 2 s in the presently used equipment.

2.8. Safety system

An important issue working with the test facility is security which is also true for continuous unmanned test. The Programmable Logic Controller has the task to supervise the test facility and the parameters:

- pressures of the media and in the tank;
- temperatures of the media and the components;
- cell voltage;
- cell current.

The methanol tank and the humidifier are protected with a pressure relief valve against a too high pressure value. If the maximum pressure value is exceeded, the valve opens and a pressure balance with the ambience occurs.

In order to avoid an overheating of the system, all temperatures in the system are supervised. The test facility is shut down automatically at a temperature overrun. If a temperature sensor is damaged, it simulates a too high temperature value and the test facility is switched off likewise.

In the case of a short-circuit or in galvanostatic operation mode the cell voltage can decrease to critical cell conditions. If a minimum cell voltage is reached the PLC shuts down the test facility.

The power supply of the data acquisition is not affected by the shut down of the test facility. So a later fault-tracing is possible.

2.9. Operation of the cells

2.9.1. Operation of a H_2-PEFC

At the beginning of the test the humidifiers for hydrogen and air are heated up to the operating temperature of 70 °C. Then the inlet valves of the gases were switched on and the cell is operated in open circuit for 10 min. Afterwards the electronic load is regulated to 500 mV, after reaching a

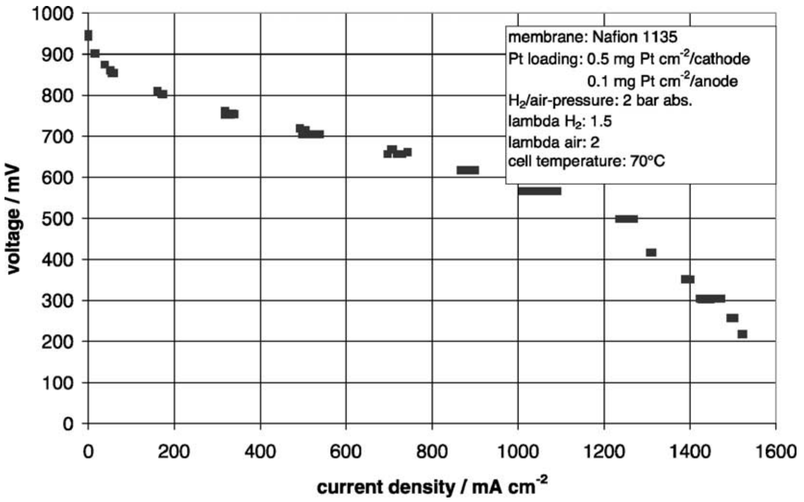

Fig. 3. Current-density–voltage characteristic of a H_2-PEFC-MEA made with DLR dry spraying-rolling technique.

constant current the cell temperature is set to 70 °C. For the next 20 h the cell is operated in galvanostatic mode at $1\ A\ cm^{-2}$ with lambda 2 for the air and lambda 1.5 for the hydrogen supply at 2 bar. A current–voltage-characteristic is measured after the 20 h of operation with the gas-flow rate for $1\ A\ cm^{-2}$. The cell potentials are then kept constant for 3 min, beginning with the open circuit voltage going down to 200 mV in 50 mV steps and back (Fig. 3).

2.9.2. Operation of a DMFC

After the installation of the MEA first the cathode supply is turned on. With reaching stable conditions at the cathode (constant pressure) the anode is supplied with methanol/water. After reaching stationary conditions (constant cell voltage, constant impedance at 10 kHz) the cell is heated up under maximum load (at 35 mV) to the first operating point. At this point a current–voltage characteristic is measured. Afterwards the next operating point is set (Table 2). Next step after finishing the measurement is purging the cell with nitrogen and switching off.

Table 2
Usual operating points

	Cell temperature (°C)	Cathode supply (N ml min^{-1} cm^{-2})
1	90	24 air
2	110	24 air
3	110	24 oxygen
4	120	24 air
5	110	120 air

In Fig. 4 different current–voltage-characteristics of an experimental MEA with backings of low hydrophobicity are illustrated. The performance of the cell is strongly influenced by the cathodic flow rate and the cell temperature. With a higher cathodic flow rate the power output of the MEA increases. Reason for this increasing is the better oxygen supply caused by the better water removal from the cell. Thus, flooding of the cathode is prevented and the transport rates of the reactants and products to and from the catalyst layer rise. A similar effect can be achieved by decreasing the cathodic

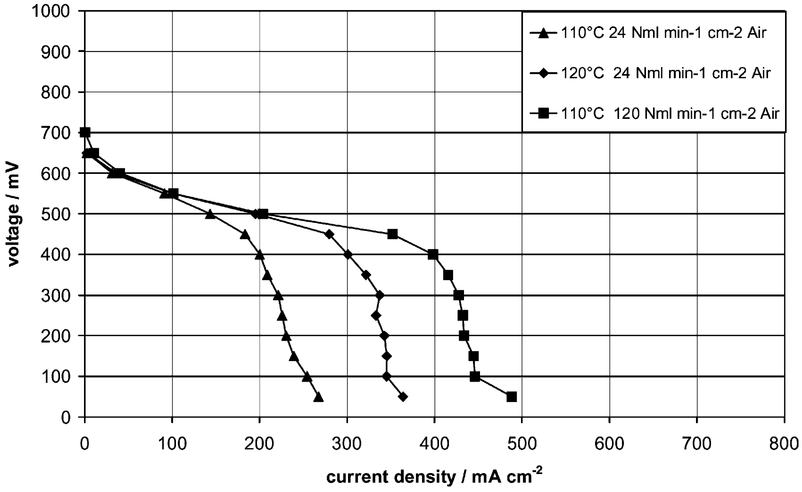

Fig. 4. Variation of cell temperature and cathodic flow rate of an experimental MEA with backings of low hydrophobicity.

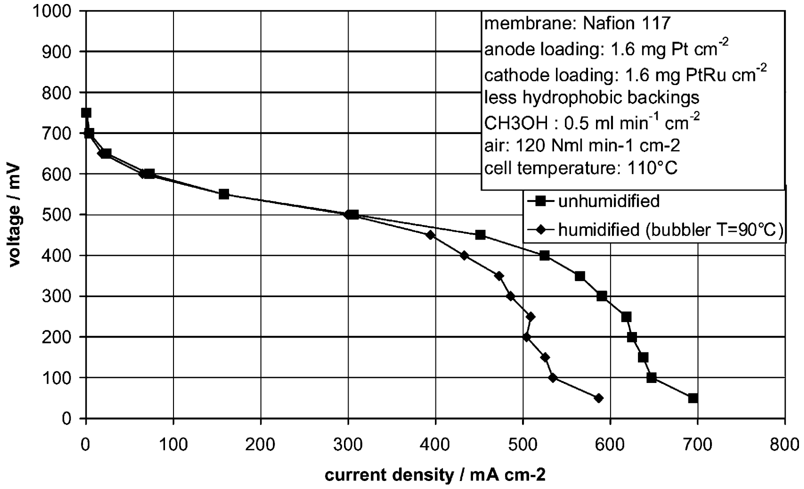

Fig. 5. Current-density–voltage characteristics of a DMFC-MEA made with DLR dry spraying-rolling technique.

pressure. It would be expected that with a higher pressure drop from the anode to the cathode the methanol crossover and also the flooding of the cathode increases [4,5]. But measurements have shown that the methanol crossover does not change and with a lower cathodic pressure the water removal will be better.

Also the increase of the cell temperature has a crucial influence on the performance of the cell. The kinetics improves and the water absorption capacity of air rises.

However therefore it is very important to keep the operating parameters (temperatures, pressures and material flow rates) within very low deviation limits.

In Fig. 5 the comparison between humidified and unhumidified operation mode is shown. When using a MEA with less hydrophobic backings additional transport inhibition arises within the range of high current-densities. The cathode is flooded and the power output decreases.

3. Summary

By the presented test facility the DLR has available an experimental set-up for a quick, reliable and efficient testing which favours an efficient development of PEFC MEAs. Interfaces exist for the collection of gases and liquids for further analysis (e.g. cathodic reaction water and anodic reaction gas).

The data from a multiplicity of measuring points can be recorded and processed automatically.

The high degree of automation of the equipment is the basis for reproducible results and makes stand alone long-term experiments possible.

A sophisticated safety system decreases the risk of faulty operations and allows riskless experiments.

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