

Activation of Electrochemical Power Devices Using Microwaves

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The efficiency of fuel cells and batteries is currently gaining more importance, particularly in light of the increasing demand for alternative power. The efficiency is highest in the temperature range where the electrolyte has maximum ionic conductivity. Because the kinetics of electrochemical reactions is dependent upon temperature, reliable operation of fuel cells requires stable temperature conditions. Therefore, independent temperature management is very important for stable and efficient operation of fuel cells. The structural parts made from electrically conductive materials can be also utilized to transmit microwave radiation and can be enabled to use microwave radiation as a heat source. This offers an interesting concept for temperature equilibration of fuel cells. In addition, micro-reactors and micro-fluidic devices can be efficiently heated by microwaves when the flow field structures are adapted to serve also as microwave patch antennas. In a design study, the applicability of different flow field structures to serve simultaneously as microwave antennas has been investigated by simulation with *QuickWave*®.

Introduction

In order to ensure highest electrical efficiency of galvanic devices operated in large temperature ranges, particularly also at temperatures below 0°C, pre-heating and thermal conditioning of such systems is required. For example, modern polymer electrolyte membrane fuel cell units (PEMFC) consist of single cells with an active cell area between 5-500 cm² and are operated between room temperature and 90°C. PEMFC are supplied by metal hydride-, pressure- and cryogenic liquid storage systems with pure hydrogen or with reformat gas, which is obtained from natural gas. In contrast to hydrogen storages, liquid natural gas storages are easy to build, inexpensive, and have low weight and high energy density. A disadvantage of the technique is the reaction of harmful products (CO₂) and complex additional equipment.

As an alternative to a hydrogen fuel cell, the direct fuel cell, which uses the alcohols such as methanol and ethanol, dimethyl ether, etc, could be provided. Due to lower power density and poorer efficiency compared to the hydrogen operation, the direct oxidation of organic fuels in the medium to high power (100 W – 100 kW) range is currently regarded as uninteresting. However, when the disadvantages could be eliminated, this approach shows significant advantages compared to the H₂ technology.

An unsolved problem is the rapid start-up of the vehicle at very low temperatures. The ionic conductivity of the electrolyte is too low to provide enough energy for the start. Because the ionic conductivity of all electrolytes depends strongly on temperature, it is required to provide additional energy. In the low power (0.1 – 100 W) range, the complexity of the system and the volume play a larger role, so that the direct methanol fuel cell is advantageous. Compared to rechargeable secondary cells, a longer operating time by a factor of 2-3 is achieved with the same volume. Regeneration by replacing the methanol tank can be quick and network-independent. Disadvantages of the systems are low efficiency and high cost of the noble metal catalysts (platinum, ruthenium). In the power range of 10 W the peripheral components such as

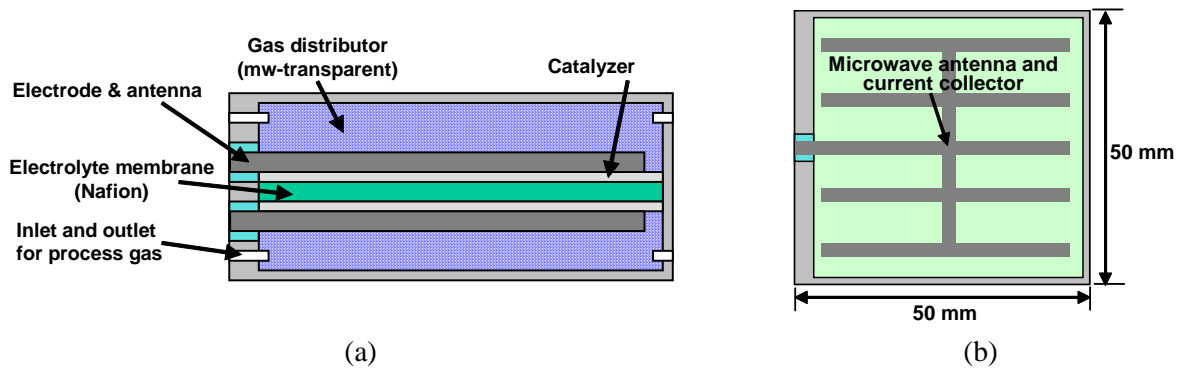


Fig. 1: Cross-section through the fuel cell and top view of the membrane assembly of an electrochemical cell with a current collector antenna (CCA).

pumps represent a major barrier for further volume and cost reduction. However, the improved activation of the catalyst could bring significant benefits.

Fuel cells are composed of structural parts made from electronically conductive materials. Such structures can also be utilized to transmit microwave radiation that serves as an energy source for heating. The selective thermal activation of the catalytic electrode of fuel cells by microwaves provides a more efficient heating. The effective operating temperature of the cell can therefore be higher than the temperatures previously possible. This achieves an increase of the power density of the cell stacks and an increased CO tolerance. Since the reactants and products of the cell reaction, e.g. water and alcohols, have high dielectric losses in the microwave range, the microwave energy could be used to evaporate excess water from the electrode at very high current densities.

To increase the efficiency of fuel cells, some publications on the treatment of gas are available [1-3]. There are also investigations regarding the influence of microwaves on the kinetics of electrochemical reactions. Compton et al. [4] studied, for example, the influence of local overheating at microelectrodes in the microwave field. No further publications on this topic are known.

The scope of the work is to develop a low-cost and efficient microwave feeding system to electrochemical cells for selective activation of individual components, in particular the electrodes and catalyst. To implement the method of microwave heating of an electrochemical cell under the specified requirements in the technical task, two types of concepts for the supply of high-frequency radiation have been developed. The basic idea of the first one is to use the electrode of the electrochemical cell as antennas to radiate the radio frequency. This bi-functional electrode is referred to as a current collector antenna (CCA).

As the second concept, a design study has been investigated for the applicability of different flow field structures to serve as microwave antenna systems simultaneously.

Technique and Results

A. Simulation and construction of the current collector antenna

Fig. 1(a) shows the current collector antenna integrated with the gas diffusion layer and a catalyst supported on the surface. This design allows feeding in of microwave radiation through the thin components. The antenna shape shown as a top view in Fig. 1(b) is a planar antenna. It is only

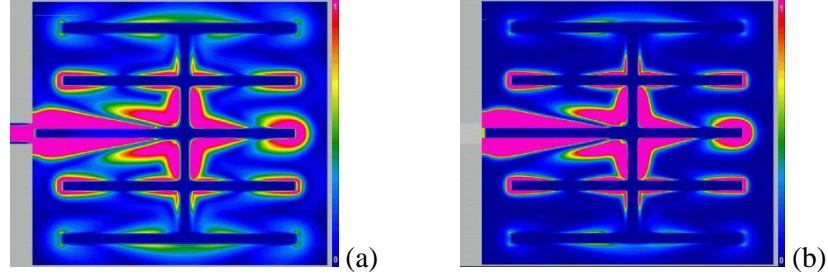


Fig. 2. Distribution of the electric field (a) and the power density (b) in the upper electrode catalyst.

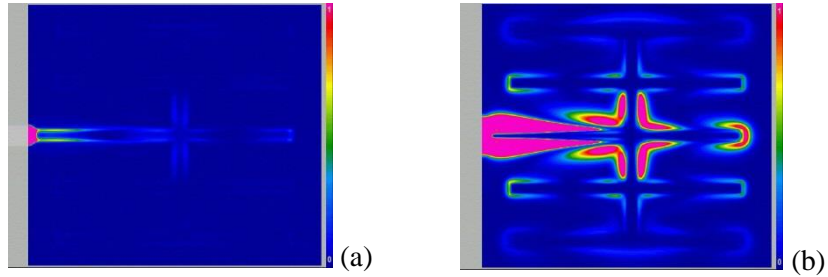


Fig. 3. Power density in the electrolyte membrane (a) and in the lower catalyst layer (b).

Table 1. Dielectric and Electric Properties of Used Material

| | ε' | ε'' | σ [S/m] |
|------------------------------------|----------------|-----------------|----------------|
| Dielectric of coaxial antenna | 2.1 | 0.0002 | |
| Gas distributor (SiO_2) | 2.9 | 0.0003 | |
| Catalyst layer | 20 | 7.3 | 4.0E4 |
| Electrolyte | 23 | 2.7 | 270 |

one of several geometries of possible current collector antennas, which was chosen as an example to show the technical feasibility of the process to validate the electromagnetic simulation. The fact that such a CCA achieves uniform heating over the entire electrode surface, while causing no additional volume and weight, will be shown in a simulation for a very simple geometry of the CCA. The CCA consists of a material of high electrical conductivity and low damping ratio of the spread of the radiation. Especially suitable materials are metallic conductors, such as gilded copper.

Just one microwave coupling on the upper catalyst electrode was simulated due to the symmetry. In real applications a fuel cell stack is contained in a metal case, which can serve as a microwave cavity. Power is supplied via a coaxial strip at a frequency of 2.45 GHz. The dielectric properties used for the simulation are shown in Table 1.

The electrolyte membrane and the catalyst layer absorb microwaves and thereby generate heat. The dielectric loss in the catalyst layer and thus the ability to heat in the microwave field is higher than in a moist membrane electrolyte. The metallic parts and the surrounding housing are made of an electrical conductor, and reflect the microwave radiation approximately losslessly. Figs. 2 and 3 show the field distributions and absorbed microwave power in different layers of the fuel cell.

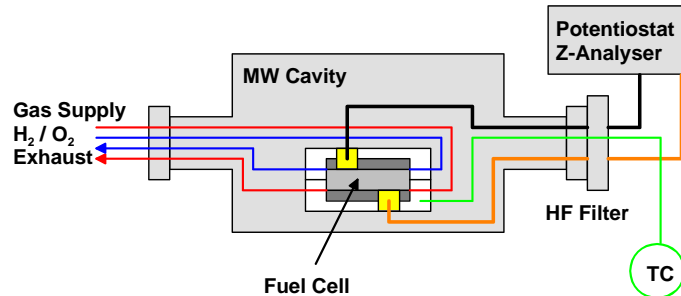


Fig. 4. Setup for the heating experiment.

The field distribution in the upper catalyst layer directly below the antenna shows that microwaves can be distributed over the entire electrode surface, but are mainly present at the location of the centre of the antenna coupling with strong field concentration. The power density (power dissipated as heat) shows a similar pattern because the absorbed power is proportional to the square of the electric field strength. In the electrolyte membrane, the absorption of microwaves is determined only in the centre area where the coaxial antenna is positioned. This is due to the relatively low dielectric loss of the electrolyte membrane. The result of simulation shows that a planar distribution of the microwave is possible in a very thin configuration. In order to achieve a more uniform field distribution, the antenna shape needs to be optimized.

B. Experimental validation in a membrane electrode assembly

A qualitative experimental verification has been performed by measuring the temperature increase inside a membrane electrode assembly (MEA) exposed to microwave radiation. The flow field was adjusted according to the results of the simulation.

As shown in Fig. 5, selective heating is observed upon exposure of the MEA to microwave radiation. The MEA is placed inside a multi-mode microwave cavity.

The electrodes of the electrochemical cell must be made of a composite material with a sufficiently high dielectric loss so that heating is possible. The heating behavior typically used in the PEMFC electrode, consisting of platinum catalysts on carbon black and solid electrolyte, is shown below. Such electrodes are efficient microwave absorbers and can be effectively heated. For separation of microwave radiation and DC on the CCA, the passive electronic RF filters are sufficient.

The microwave energy can be supplied as pulses to the electrodes to produce short-term temperature peaks. Such peaks are useful to desorb catalyst poisons like CO. It adsorbs strongly to the Pt catalysts of the PEMFC without raising the operating temperature, which sustained above the allowable maximum for efficient operation of fuel cells.

C. Optimizing of the antenna shape

The penetration of microwave radiation into a fuel cell MEA structure inside a microwave cavity is simulated. The different flow field structures are shown in Fig. 7. The aim of the study is to evaluate the penetration depth and microwave power density in catalyst layers of a real anode for a PEM fuel cell.

The images in Fig. 8 are from the plane of the catalyst layer, which is the critical area of the fuel cell. Only a certain flow field channel arrangement enables transmission of microwave

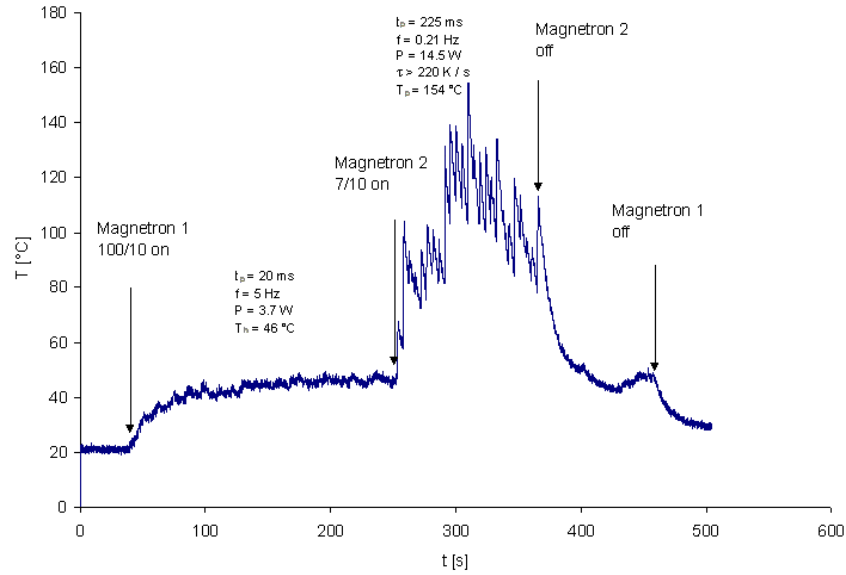


Fig. 5. Selective heating of the MEA compartment upon microwave exposure.

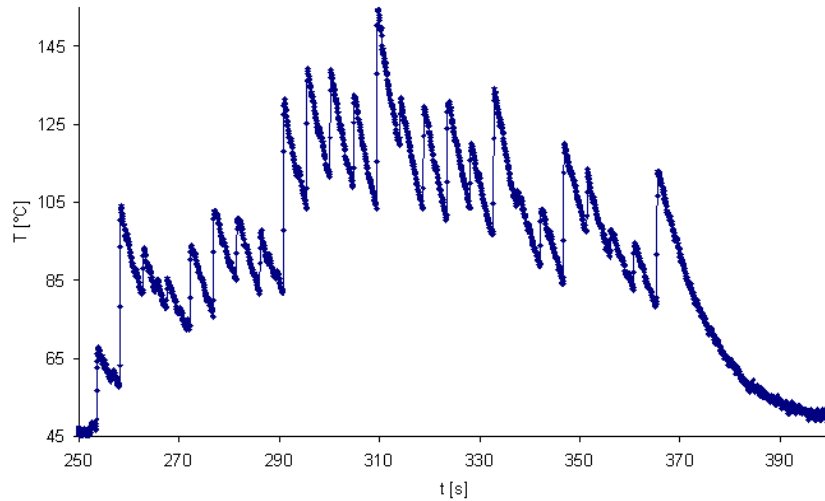


Fig. 6. Detail from Fig. 5, pulse heating of the catalytic electrode with 225 ms pulses at 0.21 Hz.

radiation into the volume between the electrodes. The line and square structure is blocking the microwave radiation, as shown in Fig. 8 (a), (b).

Opposite to this, the spiral structure supports transmission of the microwave radiation into the space between the electrodes. The catalyst and the electrolyte polymer can be efficiently heated by dissipation of microwave radiation (Fig. 8 (c), (d)).

Conclusion

A type of CCA was constructed with the aid of the simulation. It is shown that the power density distribution of the antenna was homogeneous and the heating of the assembly was performed effectively.

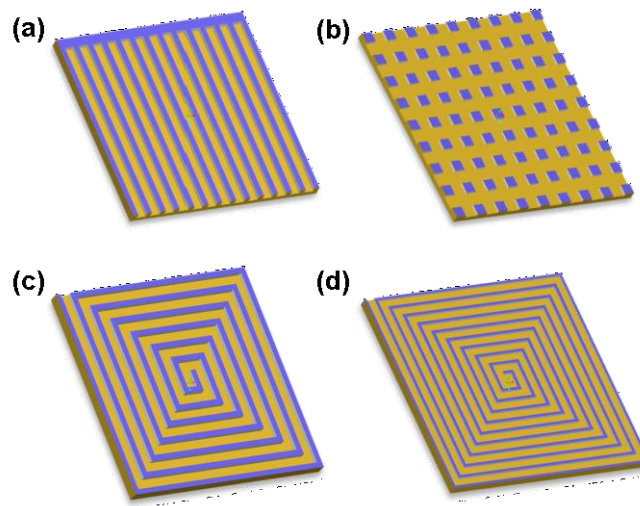


Fig. 7. Different flow field structures for MEA: the top row represents line (a) and square (b) flow fields; the bottom row represents continuous spiral structures with different thickness of the metallic channels (c) and (d).

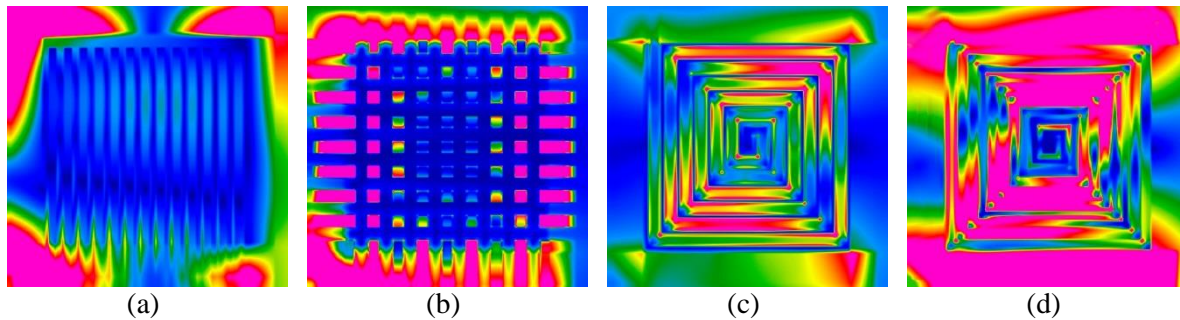


Fig. 8. Power density in the catalyst layer caused by different structures of the flow fields.

Comparing the different structure of flow fields, the fine spiral structure is particularly suitable for contacting. However, some aspects have been neglected in this simplified and purely qualitative analysis, e.g., the necessary components of the fuel cell and the connection points for the circuit. It is an optimizing problem that integrates many more parameters than the ones included in this analysis. Furthermore, the results should be examined by practical tests.

References

- [1] O. Katsuya, N. Toru, and H. Akira, Fuel cell power generation system and its starting method, Publication 2001 2103 49 A, Japanese Patent Office, 3 Aug 2001.
- [2] V. Sharivker, T. Honeycutt, and S. Sharivker, *Microwave Activation of Fuel Cell Gases*, Publ.# US 2003/0027021 A1, US Patent Application, Date of Publ. 6 Feb 2003.
- [3] V. Sharivker, T. Honeycutt, and S. Sharivker, *Microwave Activation of Fuel Cell Gases*, Publ.# WO 02/091505 A2, Intern. Patent Application, 14.Nov 2002.
- [4] R. G. Compton, B. A. Coles, and F. Marken, Microwave activation of electrochemical processes at microelectrodes, *Chem. Commun.*, pp. 2595-2596, 1998.