



Short communication

Enhancement of glucose electro-oxidation by an external electromagnetic field in direct-mode fuel cells

J.-P. Spets^{a,*}, M.A. Kuosa^a, Y. Kiros^b, T. Anttila^c, J. Rantanen^c, M.J. Lampinen^a, K. Saari^a

^a Helsinki University of Technology, Department of Energy Technology, Applied Thermodynamics, PO Box 4400, FI-02150 Espoo, TKK, Finland

^b Royal Institute of Technology-KTH, Department of Chemical Engineering and Technology, S100-44 Stockholm, Sweden

^c Oy Hydrocell Ltd., Minkkikatu 1-3, FI-04430 Järvenpää, Finland

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ABSTRACT

In this study a direct-mode fuel cell in which the fuel and electrolyte are mixed with each other is tested. An alkaline electrolyte is used. The direct-mode fuel cell is exposed to an externally generated electromagnetic field between electrodes to cause both the splitting of the fuel molecule into smaller units (i.e. electrochemical reforming) and an increase in the activity of catalyst materials on the fuel before electrochemical oxidation. The target is to create a fuel cell with a capacity range of a few mW cm^{-2} with glucose as a fuel. In the selected fuel cell type with glucose as the fuel, a maximum current density of 13 mA cm^{-2} was obtained. On the basis of the tests it seems to be possible to use the glucose-fuelled cell in small-scale applications, e.g. in small electronic devices.

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1. Introduction

The ideal fuel for fuel cells in power generation systems is the direct use of a fuel like glucose, which is produced from cellulose by hydrolysis, in the form of a liquid phase [1–4]. This allows easier use with negligible energy input for the enrichment of the fuel. Fuels such as glucose or other carbohydrates can easily be utilised by mixing them directly in the electrolyte without the use of any auxiliary reforming equipment, membrane barriers, or additional microbial cultures.

Many earlier studies have been conducted on low-temperature direct or direct-mode fuel cells, in which metallic catalysts or enzymes were used in electrolytes with different pH values [1–17]. As far as glucose is concerned, the oxidation reactions are believed to take place to only a limited degree with very low Coulombic efficiency (CE) values of theoretical maximum value, and the total yield of electrons from the molecule is only two electrons from 24 available electrons [4,9,11,12,15].

In direct-mode fuel cells (DMFCs) in which a fuel and an aqueous electrolyte are mixed with each other, the current densities have been very low. The current densities have ranged from 5 to 8 mA cm^{-2} [2,3], in comparison to current densities of several hundred mA cm^{-2} in direct fuel cells, which are supplied with either hydrogen or alcohols [18,19]. However, the use of direct-mode fuel

cells is simple as they are equipped only with a metallic catalyst, avoiding the use of anion- or cation-selective membranes. Concerning the application of the direct-mode fuel cell, the membranes are regarded as being expensive and inducing resistance, and their effective operating times are limited. In the studies regarding direct bioorganic oxidations, the anodic catalyst materials have been composed of different Pt alloys or combinations with different materials [1–16].

2. Bioorganic materials as a fuel source

In this study, a DMFC with a function generator (FG) connection is used. The idea is to use the FG to produce an electromagnetic field between the electrodes. As a result of this the glucose molecule could start to vibrate and then split into smaller units in the electromagnetic field (i.e. electrochemical reforming). The operating capacities of the anode and cathode catalysts could also increase as a result of the increased activity towards the fuel molecules. Earlier, the FG was used in fuel cells for the determination of internal resistances [20,21]. The use of a monosaccharide glucose ($\text{C}_6\text{H}_{12}\text{O}_6$) fuel is of great importance, because it can be produced easily by catalytic hydrolysis at elevated temperatures between 150 and 250°C from both starch and cellulose (polysaccharides) [22]. The target in this work is to produce a direct-mode bioorganic fuel cell with an alkaline electrolyte and with a power density production of a few mW cm^{-2} through the use of an effective and simple cell arrangement. In the development of the selected fuel cell, it is important both to increase the oxidation reactions in the liquid phase and

* Corresponding author.

E-mail address: jukka-pekkaspets@tkk.fi (J.-P. Spets).

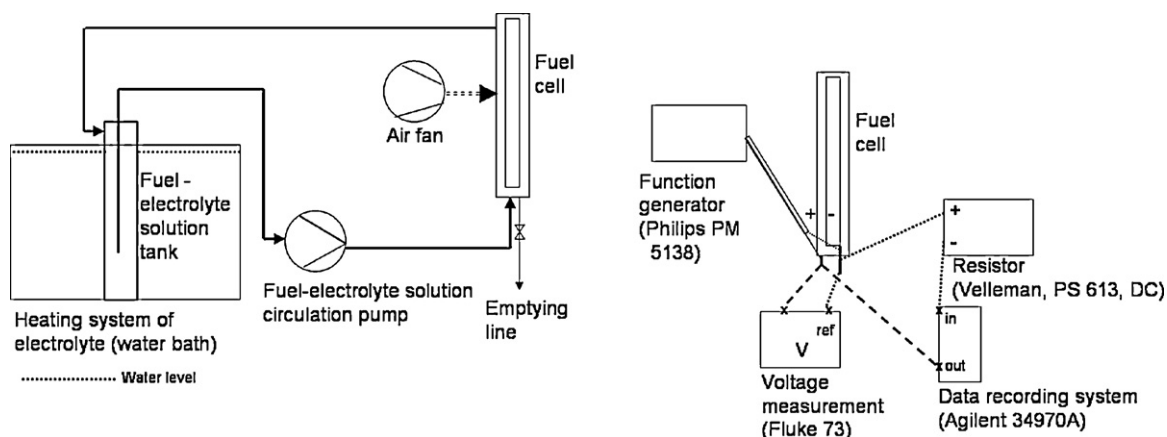


Fig. 1. The test fuel cell equipment (on the left) and the electrical connections (on the right).

to increase the change in the oxidation number. This would make possible a high electronic yield and electric power production from one molecule of bioorganic material. In this study, the electromagnetic signals are fed to the direct-mode fuel cell before the onset of electrochemical reactions in order to improve the electrochemical oxidation of the glucose. An anode catalyst of platinum and palladium (Pt–Pd) and a combined cathode catalyst of cobalt porphyrin complex (CoTPP) and of spinel (MnCo_2O_4) are tested in the test fuel cell with an alkaline electrolyte.

3. Experiments

3.1. The test equipment

The test fuel cell (FC) equipment included a direct-mode fuel cell (developed from the commercial fuel cell model HC-100 of Oy Hydrocell Ltd.), a recirculation pump (capacity 100 ml/min), an air fan (i.e. a PC cooling fan) and a recirculation tank (a glass bottle) for the fuel–electrolyte solution. The system also included a heating system (water bath) for the fuel–electrolyte solution. The test fuel cell equipment and the electrical connections are shown in Fig. 1 [3].

The direct-mode fuel cell in Fig. 1 consisted of a two-cylinder structure. The inner cylinder was an anode electrode and the outer one was a cathode electrode. The anode catalyst structure was wetted on both sides of the catalyst surfaces. The distance between the anode and cathode was 1 mm. The catalyst loadings of the anode electrode were 2.46 mg cm^{-2} for both the Pt and Pd in the bimetallic combination (in a weight ratio of 1 to 1) with concentrations of 10 wt.% on carbon. The catalyst loadings of the combined cathode electrode contained a catalyst loading of 3.15 mg cm^{-2} of CoTPP with a concentration of 18 wt.% on carbon and of 17.5 mg cm^{-2} of MnCo_2O_4 . The anode electrode consisted of a single-layer structure without diffusion material. The cathode electrode was of the double-layer type containing both the active (catalyst) and the diffusion layers. The electrodes were sintered at a temperature of 350°C . The geometric areas of both electrodes were 200 cm^2 . The Pt–Pd catalyst was a commercial grade purchased from Johnson–Matthey. The CoTPP and the MnCo_2O_4 catalysts were commercial products obtained from Oy Hydrocell Ltd.

The electrical equipment in Fig. 1 includes the electrodes, current/voltage measurements, resistor, and the function generator. To operate the fuel cell, an auxiliary device (Velleman, PS 613, and DC) was used as a resistor. The voltage values were measured by Fluke 73 measurement. The current and voltage values were recorded with an Agilent 34970A data logger. The current was determined

indirectly from the voltage loss over shunt with known voltage loss and current values (60 mV and 5 A).

An FG of the Philips PM 5138 type was used to create a change in the electromagnetic field over the electrolyte space in the fuel cell. In the tests two types of electromagnetic signals (sine and square) were used. In the tests a sine signal with a direct current (DC) or an alternating current (AC) of $0.6 \text{ V} (\pm 10\%)$ with 20 kHz was used. The square signal had AC and DC currents of $0.6 \text{ V} (\pm 20\%)$ with 5 MHz. The current outputs were measured to be 20 mA. The reason for using the low-voltage values of the signals was to prevent the electrochemical decomposition of the water in the test fuel cell under 1 V. The voltage and frequency values from the function generator were dependent on each other, so at 0.6 V the maximum possible frequency was 20 kHz for the sine signal and 5 MHz for the square signal, respectively.

For the determination of the transferred electrons the chemical oxygen consumption values (COD) were analysed with the help of a heating device and an analyser (Merck Spectroquant TR 320 and Merck Spectroquant Nova 60). The test cuvettes used for the analysis were of the Merck No. 14541 type. Pre-programmed heating at 148°C for 20 min was used, because this COD analysis method requires elevated temperatures in order to work.

3.2. Test routine

The test routine in the entire test was as follows: an aqueous alkaline 2 M potassium hydroxide (KOH, Sigma–Aldrich) solution electrolyte was prepared. The fuel, glucose (anhydrous, Oriola), was added to the electrolyte in a concentration of 1 M. The electrolyte–fuel solution was added to the fuel cell. The air fan was started and the open-circuit voltage (OCV) of the fuel cell was measured at room temperature (20°C) or at an electrolyte temperature of 28°C until a steady state was attained. The tank with the circulated solution was heated in a water bath, the temperature of which was 60°C in order to produce a temperature of 28°C for the solution in feed to the FC. The OCV value was recorded until the generation rate decreased to a minimum value of $2\text{--}4 \text{ mV min}^{-1}$. After the generation of OCV for a short period (i.e. 30 s) current was taken from the test fuel to secure that the catalysts were activated. The open-circuit generation was started again. The FG was connected to the test fuel cell with a coaxial cable: an internal wire was connected to a cathode electrode and the external insulator netting was connected to the anode electrode of the fuel cell. The function generator was set to the values as a DC or AC of $0.6 \text{ V} (\pm 10\%, 20 \text{ kHz, sine})$ or $(\pm 20\%, 5 \text{ MHz, square})$ and it was used for a period of 90 min. It was assumed that a long enough reforming time was capable of causing the phenomena of both the electro-

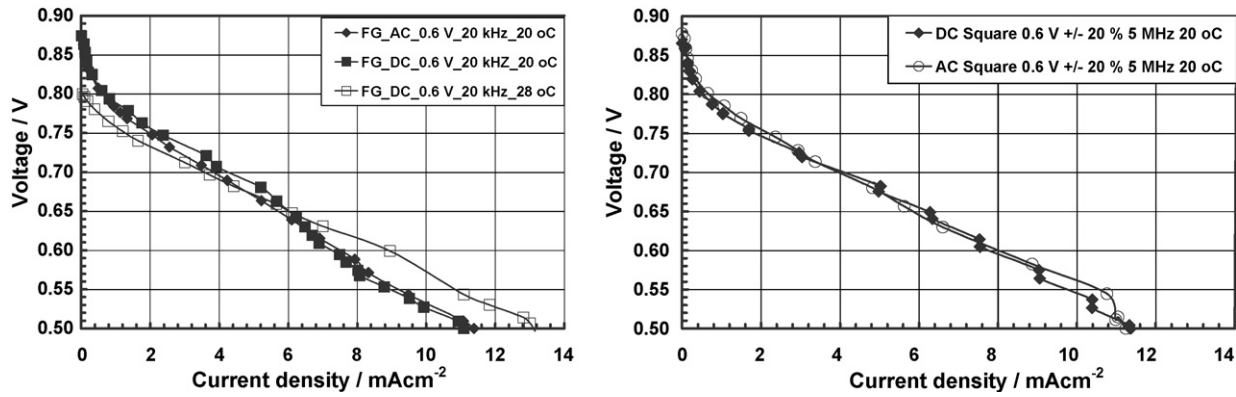


Fig. 2. Polarisation curves of direct-mode alkaline FC after the use of function generator with DC and AC sine (on the left) and square signals (on the right).

chemical reforming and the splitting of the fuel molecule in the electrolyte. After the selected time period, the function generator was switched off to avoid possible malfunctioning of the function generator when taking current from the test fuel cell. The polarisation curves and the current-versus-voltage values were recorded in each case. The test was stopped when the voltage of the direct-mode fuel cell decreased to a value of 0.5 V [2–4] to avoid alkaline penetration into the cathode electrode at lower voltage values.

The Coulombic efficiency (CE) values of each test were calculated using the recorded current and time values according to Eq. (1) [23]:

$$CE = \frac{(M_w \int Idt) 100\%}{Fn_e V_{an} \Delta C} \quad (1)$$

In Eq. (1) M_w is the molecular weight of glucose, n_e is the number of electrons transferred, V_{an} is the volume of liquid in the anode (0.3 dm^3), F is the Faraday constant ($96,500 \text{ As/mol}$), and ΔC is a change in the substrate mass concentration ($C_0 - C$). $\Delta C = C_0$ is often used. In Eq. (1) the term $(\int Idt)$ corresponds to the integration of the recorded Coulombs, and the denominator corresponds to the maximum available Coulombs from the initial glucose molar amount in the aqueous electrolyte.

The COD measurements of the fuel–electrolyte solution were obtained from the test that gave the highest current values. Samples were taken from the fresh and the used fuel–electrolyte solutions. The samples were diluted. 3-ml volumes of the diluted samples were injected into test cuvettes. Each cuvette was heated up to a temperature of 148°C for 20 min. After a 10-min cooling time the test cuvette was shaken and cooled down to the ambient temperature and the COD values were measured with the analyser.

4. Results and discussion

4.1. Measurements

Fig. 2 shows the measured polarisation curves with glucose as the fuel in the alkaline electrolyte. The tests were conducted at two different temperatures for the sine DC signals and at room temperature for all the AC and DC square signals. The measured current values were then compared to values that had been reported earlier [2–4] without the use of the function generator. The FG was in use for 90 min at 20°C and for 50 min at 28°C . The reason for the lower FG operation time at 28°C was the reduction of the OCV value to 0.8 V.

The measured polarisation curves at a temperature of 20°C in Fig. 2 do not show any major differences between the sine and square DC and AC signals at the frequencies of 20 kHz and 5 MHz. It is assumed that the frequency of the signals has to be increased

much more from the value of 20 kHz to cause a noticeable increase in the current density values. The measured OCV values in Fig. 2 were of an order of magnitude of 0.87 V at 20°C and 0.80 V at 28°C . The OCV value at 20°C was 0.07 mV higher than the values reported in the tests without the use of the FG (in Refs. [2,3]). However, at the temperature of 28°C the OCV value (0.8 V) was equal to the results [2,3] without the FG at the temperature of 51°C . It can be assumed that both the high temperature and the use of the FG accelerate the decomposition of the glucose in the electrolyte, and reduce the OCV from 0.87 V to 0.8 V. There was also a noticeable change in the colour of the used fuel–electrolyte solutions after the operation of the fuel cell. At 20°C the solution remained the colour of clear light urea, whereas at higher temperatures the colour of the spent electrolyte solution was black. The decomposition of the glucose probably started at the temperature of 60°C in the water bath. The selected capacity tests of the DMFC with the use of the FG with DC sine and with AC square signals are shown in Fig. 3. The voltage values of the fuel cell were recorded with changing current intervals. The capacity recordings were calculated from the linked runs as follows: first the polarisation curve (Fig. 2) was determined and then the capacity test (Fig. 3) was repeated until a decrease in the time period from the decrease of the voltage from the OCV until the value of 0.5 V was noticed (Figs. 2 and 3).

From the results shown in Figs. 2 and 3 it can be concluded that when the electromagnetic field is used before the operation of the fuel cell the current density values of the DMFC are increased, compared to earlier tests [2–4]. At 20°C the current density value at 0.5 V increases from 5 mA cm^{-2} (i.e. 1 A) to 11 mA cm^{-2} (i.e. 2.2 A) with the use of both DC and AC signals, i.e. an increase of 120% was reached. The raising of the electrolyte temperature improves the current density and the voltage values both without and with the usage of the FG. An increase from 8 mA cm^{-2} (1.6 A) to 13 mA cm^{-2} (2.6 A) with the use of the DC signals was reached, i.e. an increase of 62.5%. However, it is not desirable to use higher temperatures than 40°C in the water bath because sugars start to decompose at higher temperatures, resulting in an increased loss of the current density values.

The recorded Coulombs for the experimental tests are shown in Table 1 and the evaluations of the CE values for the fuel cell accord-

Table 1
The recorded Coulombs for the experimental tests.

Temperature ($^\circ\text{C}$)/signal	Coulombs (As)
20/Sine DC	2115
28/Sine DC	3636
20/Sine AC	2778
20/Square DC	3182
20/Square AC	7321

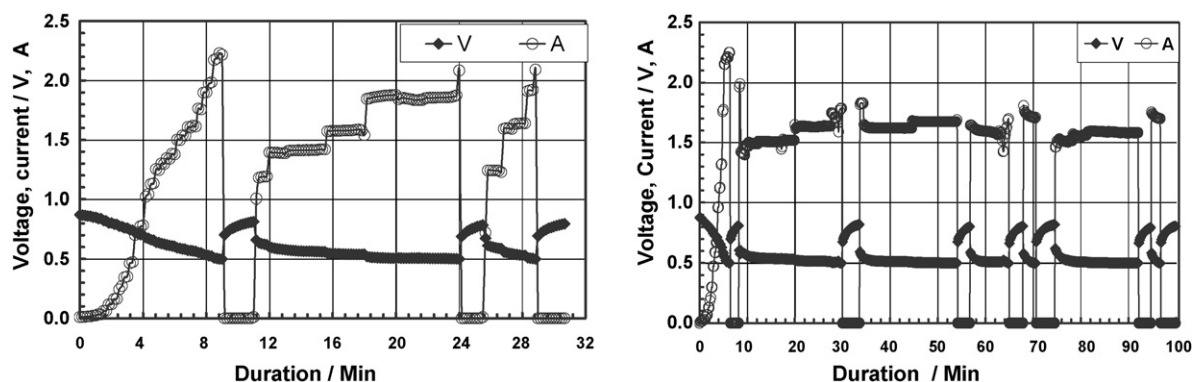


Fig. 3. Selected capacity tests of the direct-mode fuel cell at 20 °C. The results after the use of the function generator with DC sine are on the left and with AC square signals are on the right.

Table 2

The CE values as ratio to theoretical value with the release of two electrons per molecule.

Temperature (°C)/signal type	CE (%)
20/Sine DC	3.7
28/Sine DC	6.3
20/Sine AC	4.8
20/Square DC	5.5
20/Square AC	12.6

ing to Eq. (1) are shown in Table 2. The CE values are calculated as a ratio to theoretical value. Two electrons are supposed to be released from one molecule in the electrochemical oxidation.

The recorded Coulombs in Table 1 and Coulombic efficiency values (CE) in Table 2 were shown to be much higher than reported earlier in a direct-mode fuel cell without the use of the function generator at 20 °C (470 As and CE = 0.82%) [4]. In this study Coulombic values with a maximum of 3182 As (with a square DC signal) and 7321 As (with a square AC signal) at 20 °C and 3636 As at 28 °C (with a sine DC signal) were achieved. The improvements were 577% (with a DC signal) and 1458% (with an AC signal) at 20 °C. The use of the AC signal from the FG led to better Coulombs (As) from the DMFC than the use of the DC signal. The higher frequency of the signal made higher current values possible.

Table 3 shows the relative change in the chemical oxygen consumption (COD) values in the samples, which were taken from the fuel electrolyte solution at the beginning and, after the recording, the 7321 Coulombs (Tables 1 and 2).

Using the relative change in the COD value of the fuel–electrolyte solution in Table 3, the number of transferred electrons per glucose molecule can be approximated as two according to Eq. (1) when both the 23.3% change in the COD value (i.e. COD gives the moles of the consumed glucose Δn_{Glc} from the initial mole amount) and recorded Coulombs are taken into account [$n_e = (\int dt) (\Delta n_{\text{Glc}} \cdot F) = 7321 \text{ As} / (0.233 \times 0.3 \text{ dm}^3 \times 1 \text{ mol dm}^{-3} \times 96,500 \text{ As/mol}) = 1.1 \sim 2e^-$]. However, the increased CE values may indicate that the total activity of both the anode and cathode catalysts has increased as a result of the use of the FG, when compared to the earlier reported current

Table 3

Measured COD and pH values of fuel–electrolyte solutions before and after the recording of the highest amount of Coulombs (7321 Coulombs) in the direct-mode fuel cell.

Liquid sample	COD (pH value) g dm ⁻³
Before use in DMFC	131 (pH 13.70)
After use in DMFC	100 (pH 13.60)
Change in COD value	–23.3%

density values (without the use of the FG [1–4]). This needs to be confirmed by cyclic voltammetry measurements and rotating ring electrode analysis, together with durability tests in the future. The low change in the pH value of the fuel–electrolyte solution indicates that the carbonisation problem resulting from the release of CO₂ during glucose electrochemical oxidation can be considered to be negligible.

The current densities of the DMFC in Fig. 2 are lower when compared to those observed in direct fuel cells (hydrogen- or alcohol-fed) with membranes [18,19], but higher than the reported current density values for microbial and enzymatic biofuel cells [17]. However, when compared to direct fuel cells [18,19], the DMFC is simple to use and provides good application potentials for small portable applications. The glucose fuel, which is produced from cellulose materials, provides a good alternative to alcohols (methanol and ethanol) thanks to its renewable and sustainable nature and development. For example, because of the limitations of the direct use of ethanol in the fuel cell, it has to be reformed at high temperatures between 500 and 700 °C to provide high hydrogen conversion [24]. There are problems in the direct use of methanol and ethanol because of the carbonisation problems in alkaline electrolytes, when the applicable high current densities are to be extracted [12]. It is also assumed that the use of a direct-mode fuel cell with a different voltage and a different signal frequency will cause the release of more than two electrons per glucose molecule in electrochemical oxidation. If the electron yield from one molecule of glucose increases from 2 towards the 24 available electrons, glucose will be an even better fuel for low-temperature direct fuel cells than liquid alcohols.

4.2. Development needs in the future

It is assumed that the use of the function generator (FG) is mainly based on two principal assumptions, viz. the enhancement of the activity of the catalyst material on the fuel and the electrochemical reforming of the fuel in the electromagnetic field between the electrodes in the electrolyte space. The right forms of the AC or DC signals (sine, square etc.) from the FG, together with the voltage and the frequency values, in addition to the concentrations and temperature of the fuel–electrolyte solution are vitally important to provide higher current densities in the direct-mode fuel cell. The search for more active catalyst materials for both the anode and cathode electrodes could also improve the operating capacities of bioorganic fuel cells. Higher temperatures than room temperature have a positive influence on the current densities, as has been reported earlier and also in this research. When the operational parameters of the FG are optimised, the effects (i.e. the vibration of the fuel molecules in the electromagnetic field and the increased

activity of the catalyst materials) would strengthen the production of both the higher current densities and of the number of Coulombs in the bioorganic direct-mode fuel cell. Tests using changing electromagnetic fields have to be conducted with both anodic and cathodic half-cells in order to see the influence of the function generator on each catalyst material. It is important to know which reaction routes can be formed in the electrochemical oxidation of glucose when the function generator is used. Tests should be carried out in different near-neutral and alkaline electrolytes to indicate the improvements and differences in operating capacities with electrolytes of different pH values. It is already known that the electro-oxidation of glucose and other biofuels occurs with low current density values in fuel cells with near-neutral electrolytes with low conductivity values [1,25]. Further studies have to be carried out in order to find out if any poisoning of the catalyst materials, together with the possible carbonisation, affects the electro-oxidation of carbohydrate compounds in direct-mode fuel cells attached to a function generator. During the electro-oxidation of glucose, the Pt catalyst is reported to be poisoned by gluconic acid, especially in near-neutral-state electrolytes [26].

5. Conclusions

In this study glucose was tested as mixed with an alkaline electrolyte in the selected direct-mode fuel cell connected to a function generator. It was found that glucose in an aqueous alkaline electrolyte produced current densities in the range of 11–13 mA cm⁻² at a voltage of 0.5 V. These current density values were higher than reported earlier without the use of a changing electromagnetic field. The measured polarisation curves at the temperature of 20 °C did not exhibit any significant differences between the sine and square DC and AC signals with 20-kHz and 5-MHz frequencies. It is assumed that the frequency of the signals has to be increased much more from the values of 20 kHz and 5 MHz to cause a noticeable increase in the current density values. The use of the AC and the higher frequency signal produced a higher current capacity than with the use of the DC signal at a lower frequency value. The influence of the FG on the operation of the direct-mode fuel cell is assumed to be improved even more by the optimisation of the different parameters (the form, the voltage, and the frequency of the electromagnetic signals), together with the operating temperature. The concentrations of the glucose fuel and the electrolyte also have to be changed to find effective operating conditions for the anode and cathode catalyst materials. The extended yield of the

electrons from one glucose molecule would make it an important fuel for bioorganic cells.

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