FUEL CELL THERMAL MANAGEMENT SYSTEM BASED ON MICROBIAL FUEL CELL 3-D MULTI-PHASE FLOW NUMERICAL MODEL

by

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The paper tests the changes in the pH value of the anolyte and catholyte. The 3-D multi-phase 3-D multi-current conductivity values analyze the electricity generation process and energy utilization of the microbial fuel cell (AMFC) and provide a theory for improving the AMFC following the performance. The test results show that with the operation of AMFC, the pH value of the anolyte and the 3-D multi-flow conductivity show a downward trend, the pH value of the catholyte and the 3-D multi-flow conductivity show an upward trend, and the ratio of the pH value of the catholyte the pH value of the anolyte is about 0.30-0.50 higher, and the average 3-D multi-current conductivity of the anolyte and catholyte does not change much. When AMFC operates stably, the internal ohmic resistance is 29.69 Ω, the limiting current is 2.69 mA, the maximum output power is about 0.8 mW, and the corresponding internal resistance is about 95.72 Ω. The mass transmission of potassium ferricyanide is the limiting factor of limiting current. Numerical analysis of 3-D multi-phase flow found that other microorganisms consume 91.1% of the glucose in AMFC anolyte, and only 8.9% of the glucose is used for power generation. The 88.5% of the energy of the glucose used for power generation is converted into other forms of energy, only 11.5% of the energy is converted into electricity.

Key words: *3-D multi-current conductivity, electricity generation process, energy utilization, mass transfer, pH value*

Introduction

Solving the increasingly serious environmental pollution problems and exploring new energy sources are the fundamentals for achieving sustainable development and circular economy. The AMFC is one of the new technologies that organically combines environmental pollution and the production of new energy. It is a device that uses microorganisms as a biocatalyst to convert organic biomass that pollutes the environment into electricity. It can use various sewage as raw materials and can generate clean energy [1]. It has diversified fuel sources, pollution-free, high energy utilization efficiency, and mild operating conditions. Strong biocompatibility, safety, high efficiency, continuous and other advantages. Its distinctive features and multifunctional characteristics have attracted widespread attention, and it has the prospect of being widely used in microbial sensors, bioremediation, sewage treatment*, etc.*

The AMFC usually consist of an anode chamber, a cathode chamber, and a proton exchange membrane. The substrate in the anode compartment is oxidized under the catalysis of microorganisms, and the generated electrons are transferred to the anode through the elec-

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tron carrier (such as cytochrome) located in the outer membrane of the cell, and then reach the cathode through the external circuit. The protons reach the cathode through the proton exchange membrane. The electron acceptor reacts with protons and electrons on the surface of the cathode electrode to produce water. The process of proton transmission is related to the pH of the solution. Some scholars have conducted experimental research on the pH value of the anolyte and found that the current is the largest when the pH is 7.00-8.00, which is related to microorganisms' activity at different pH values. But generally speaking, the pH value of the anode compartment solution has little effect on the battery current, and the difference is within 10%. At the same time, the study found that if the buffer solution is added, the anode solution's pH value and the cathode solution changes little, and *vice versa*. Some scholars have conducted experimental studies on the catholyte's pH value and found that when the pH value of the cathode compartment solution increases from 2.40-9.40, the cathode potential and electrical energy output are significantly reduced. The reason is that the oxygen reduction reaction rate constant is affected by the pH value. Some scholars have found that the internal resistance of AMFC mainly depends on the resistance of the electrolyte between the electrodes and the resistance of the proton exchange membrane [2]. Reducing the internal resistance and optimizing AMFC performance can be achieved by shortening the distance between the electrodes and increasing the ion concentration.

Some scholars found that when 0.3 mol/L sodium chloride was added to the anode solution, the voltage increased by 20 mV, and the coulombic efficiency increased by 1.2 times. The 3-D multi-current conductivity is an important parameter related to the ionic strength. Many documents mention the 3-D multi-current conductivity, but there is no change in the 3-D multi-current conductivity of the anolyte and catholyte during stable operation of the AMFC. Detailed study. Some scholars divide AMFC electricity production process into three-stages, but they have not analyzed it in detail. This paper mainly analyzes the changes in the pH value of the anolyte and the catholyte and the conductivity of the 3-D multi-stream during the operation of the AMFC through experimental research, besides, the electricity generation process and energy utilization of the AMFC are also analyzed and calculated. Improve the performance of AMFC to provide a theoretical basis.

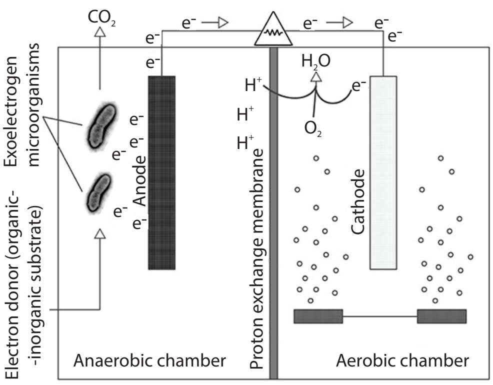

Figure 1. Schematic diagram of the AMFC system

Materials and methods

Configuration of AMFC

The AMFC is shown in fig. 1. This rectangular AMFC is a two-chamber system. The cathode chamber and the anode chamber use the same rectangular structure, and the effective volume is 1300 mL, separated by a proton exchange membrane, and the size is 50 mm × 50 mm. The anode chamber cap has three small holes with a diameter of 6 mm, used for sampling, nitrogen aeration, and fixed electrodes. The cathode chamber cap also has three small holes used for sampling stomata and fixed electrodes [3]. Both the cathode electrode and the anode electrode use graphite felt with an effective area of 100 cm^2. The external circuit load is the output voltage of the ZX36 resistance box, which is recorded by the UT70D digital multimeter and automatically stored in the personal computer.

Inoculation and operation of AMFC

The experimental inoculation sludge came from a sewage treatment plant, using pre-filmed anode electrodes, but did not use anolyte for domestication. Both the anode solution and the cathode solution use sterilized deionized water as the solvent. The anode solution's composition is glucose 0.91, KH_2PO_4 6.81, NaOH 1.17 g/L, and other trace elements 20 mL. The cathode solution's composition is KH_2PO_4 6.81 g/L and NaOH 1.17g/L. Use 1000 mg/L potassium ferricyanide $K_3Fe(CN)_6$ as the electron acceptor. Before the operation of the AMFC, nitrogen is introduced into the anode solution remove the oxygen in the solution. The sealing of the anode chamber is strengthened during operation maintain the anaerobic environment of the anode chamber. The AMFC is placed in a constant temperature water tank with a constant temperature of 35 °C. The ambient temperature varies between 13-18 °C, and the relative humidity varies between 10-30%. During operation, the load's external resistance remains unchanged at 100 Ω, and the external resistance of the load changes only when the test voltage changes with the current [4]. The experiment adopts an intermittent feeding method and runs continuously for three weeks to observe the AMFC operating parameters.

Data measurement and calculation

The experiment uses a UT70D digital multimeter to measure the output voltage. Uses a RotronicK15 temperature and humidity sensor to measure the ambient temperature. Uses a Multi340 multi-parameter water quality tester to measure the 3-D multi-flow conductivity and pH value of the solution. Uses a 5B-3C COD tachometer to measure the anode liquid COD. Use eq. (1) to calculate current, use eq. (2) to calculate output power, use eq. (3) to calculate internal resistance, use eq. (4) to calculate Coulomb efficiency, and use eq. (5) to calculate COD removal rate:

$$I = \frac{U}{R_0} \tag{1}$$

$$P = UI \tag{2}$$

$$R_i = \frac{E}{U-1} R_0 \tag{3}$$

$$\varepsilon_{Cb} = \frac{M \int_0^{tb} I \mathrm{d}t}{Fb\Delta m} \tag{4}$$

$$\varepsilon_{CoD} = \frac{\Delta COD}{COD} \tag{5}$$

where I [mA] is the current, U [mW] – the output voltage, P [mW] – the output power, R_i [Ω] – the internal resistance, R_0 [Ω] – the external resistance, M [180 gmol^{-1}] – the molar mass of glucose, F [96485 Cmol^{-1}] – is Faraday constant, b [24 molmol^{-1}] – the number of electrons gained and lost per mole of glucose, Δm [g] – the change in glucose in time, and tb [s] – the time period.

Results and analysis

Changes in pH

After AMFC inoculation runs, test the pH value of anolyte and catholyte once a day until AMFC stops running. The test found that the anolyte's pH value quickly dropped from

6.89-6.43 and then rose to about 6.60. When AMFC is running, the anolyte's pH is maintained at about 6.50, fig. 2. The following two reasons may cause the decrease in the pH value of the anode solution:
- The product CO_2 dissolves in the anode solution generate carbonic acid, which causes the pH value of the anolyte to drop, and the solubility of CO_2 is limited, so the degree of subsequent change is not big.
- Biogas production by anaerobic microorganisms is divided into acid production stage and methane production stage [5]. It may also be caused by microbial acid production. The microorganism consumes the substrate to produce many H^+, and AMFC consumes H^+ during the electricity generation process. Finally, the two reach a balance, so that the pH value of the anolyte is maintained at about 6.50, the pH value of the catholyte does not change much, showing a slight increase the trend has risen from 6.92 at runtime to 7.05. The reason may be that the H^+ that migrates from the proton exchange membrane to the catholyte is the same as the H^+ consumed by AMFC in the process of generating electricity. Simultaneously, the catholyte is reduced at a high temperature, which increases the pH value, fig. 2. During the operation of AMFC, the pH of the catholyte is about 0.30-0.50 higher than the pH of the anolyte.

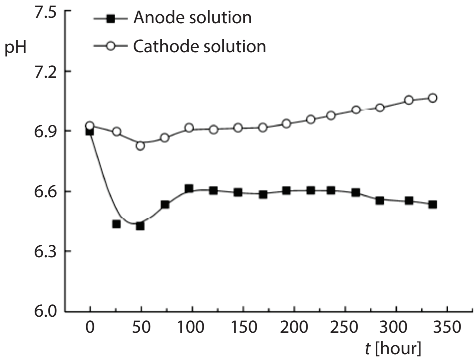

Figure 2. Changes in pH over time

Changes in 3-D multi-current conductivity

After AMFC inoculation runs, the 3-D multi-current conductivity of the anolyte and catholyte is tested once a day until the AMFC stops running. The experiment found that with the operation of AMFC, the 3-D multi-flow conductivity of the anolyte showed a downward trend, from 7.51 ms/cm during operation 6.97 ms/cm, while the 3-D multi-flow conductivity of the catholyte showed an increase, the trend, from 7.37 ms/cm during operation 10.10 ms/cm. The average value of the 3-D multi-current conductivity of catholyte and anolyte does not change much, from 7.44 ms/cm during operation 8.53 ms/cm, fig. 3. The reason may be that the ions in the anolyte enter the catholyte through the proton exchange membrane, so that the 3-D multi-stream conductivity of the anolyte decreases, and the 3-D multi-stream conductivity of the catholyte increases, at the same time, the catholyte decreases at high temperatures. For one part, the average 3-D multi-current conductivity of the anolyte and catholyte shows a slight upward trend [6]. The average 3-D polynomial current conductivity of AMFC is 7.77 ms/cm in stable operation. Ohmic resistance:

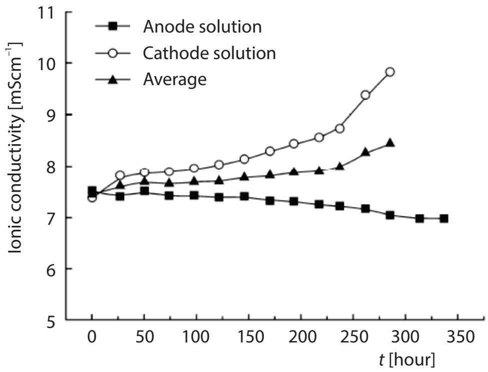

Figure 3. The 3-D polynomial current conductivity changes with time

$$R_{\text{ohmic}} = \frac{L}{A\sigma} \tag{6}$$

where A [cm^2] is the cross-sectional area, σ [Scm^{-1}] – the 3-D polynomial conductivity of the solution, and L [cm] – the transmission distance. When the cross-sectional area is 25 cm^2, and the transmission distance is 7 cm, the internal ohmic resistance can be obtained as 36.04 Ω according to the aforementioned formula.

Analysis of electricity production process

Analysis of the power generation process after the AMFC inoculation is running, the external resistance of 100 Ω is connected, and the output voltage is recorded with a multimeter. The COD of the anolyte is tested every two days, and the curve shown in fig. 4 is obtained. It can be seen that the AMFC electricity production process is divided into three-stages: the first stage is the initial stage. Because the inoculated microorganisms are not domesticated and cultivated, the microorganisms are not adapted to the anolyte environment, and the electricity production is small, so the output voltage is very low. With the operation of AMFC, microorganisms have gradually adapted to the anolyte after domestication and culture. Simultaneously, the number of microorganisms has also increased greatly, and the output voltage has increased rapidly. The second stage is the intermediate period when the output voltage increases to a certain extent and then begins to drop. When 1 g potassium ferricyanide is added to the catholyte again, the output voltage rises sharply, showing that the electron acceptor's mass transfer limits the output voltage [7].

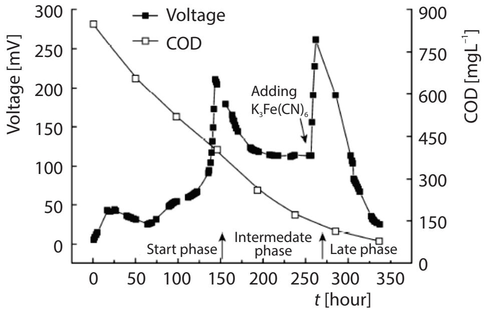

Figure 4. Changes in output voltage and COD over time

Theoretical analysis can also get the same results, tab. 1: a current of 1 A is generated, the mass-flow of glucose is $7.77 \cdot 10^{-5}$ g/s, and the mass-flow of potassium ferricyanide is $3.41 \cdot 10^{-3}$ g/s. The consumption of potassium ferricyanide is 43.9 times that of glucose. So, the consumption of potassium ferricyanide is extremely fast. Simultaneously, the diffusion coefficient of potassium ferricyanide in water is 20.1 times that of glucose, so the mass transmission of potassium ferricyanide is not necessarily the limiting factor of output voltage when AMFC is running. The third stage is the later stage. The output voltage gradually decreases. When 1 g potassium ferricyanide is added to the cathode again, the output voltage continues to decrease (not shown in the figure). It can be seen from fig. 4 that the COD of the anolyte has dropped below 150 mg/L, which shows that the quality of the substrate has become a limiting factor for the output voltage.

Table 1. Physical parameters of substrate and electron acceptor

Reactant	Molecular formula	Mol.wt. [gmol^{-1}]	Solubility [gm^{-3}]	Diffusion coefficient [m^{2}s^{-1}]	e [molmol^{-1}]
Glucose	$C_6H_{12}O_6$	180	$1.1 \cdot 10^6$	$0.69 \cdot 10^{-9}$	24
Sodium acetate	CH_3COONa	82	$3.3 \cdot 10^6$	$1.26 \cdot 10^{-9}$	8
Oxygen	O_2	32	8.4	$0.24 \cdot 10^{-8}$	4
Potassium ferricyanide	$K_3Fe\ (CN)_6$	329	$3.6 \cdot 10^5$	$1.39 \cdot 10^{-8}$	1
Potassium permanganate	$KMnO_4$	158	$6.38 \cdot 10^4$	NA	3

Voltammetry curve

After the AMFC runs stably, change the size of the external resistance (9999-10 Ω) to obtain the volt-ampere curve, fig. 5. The paper uses a simple 1-D model to fit it to get the relationship:

$$v = 591.45 - 29.55 - 9.12I - 29.69\ln I - 99.56\ln\frac{2.69}{2.69 - I} \tag{7}$$

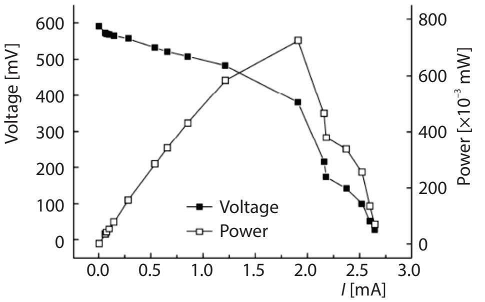

Figure 5. Output voltage and output power changes with current

It can be seen that the internal ohmic resistance is 29.69 Ω, the limiting current is 2.69 mA. The maximum output power is about 0.8 mW, and the corresponding internal resistance is about 95.72 Ω. These data indicate that for AMFC that rely solely on diffusion achieve the mass transfer of substrates and electron acceptors, the internal resistance caused by concentration loss increases greatly [8]. If you want to increase the output power, you must increase the limiting current, improving the mass transfer of the substrate and the electron acceptor. Several measures can be taken:

– using convection mass transfer instead of diffusion mass transfer may have side effects, and detailed studies have not been seen in the literature,
– the number of electrons produced per unit mass is large, Substrate and electron acceptor with large diffusion coefficient, and
– optimize the configuration of electrode and AMFC.

Energy analysis

During AMFC inoculation operation, the anolyte COD was 848.6 mg/L, the residual COD in the anolyte at the end of power generation was 76.58 mg/L, and the COD removal rate was about 91%. At the beginning of the reaction, the anolyte contained 1.18 g glucose, so the total amount of glucose consumed by AMFC was 1.07 g. According to the conservation of charge, fig. 6, the Coulombic efficiency can be calculated to be 8.9%, so the glucose used to generate electricity is 0.095 g. The energy that 1 g glucose can provide is 16.73 kJ (4 kcal), and the maximum power generation capacity of 0.095 g glucose is 1589.92 J. Integrating the power, fig. 6, can calculate that the electrical work done by AMFC is 182.28 J, and the energy utilization rate of AMFC is only 11.5%. From the previous calculation, it can be seen that other microorganisms consume 91.1% of the glucose in the AMFC anolyte, and only 8.9% of the glucose is used for power generation; 88.5% of the energy of the glucose used for power generation is converted into other forms of energy, only 11.5 % of energy is converted into electricity [9]. The measures to improve the energy utilization rate of glucose in AMFC include:

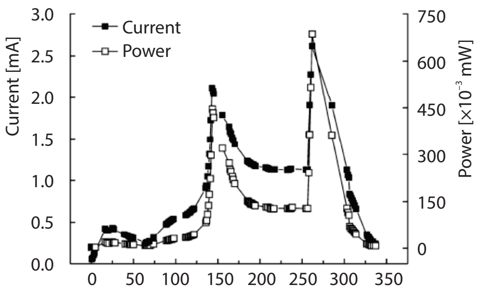

Figure 6. Current and output power changes over time

– speed up the filming process of the anode electrode, reduce the consumption of suspended microorganisms on the substrate, and increase the utilization rate of the substrate by the electricity-producing microorganisms,

– reduce the AMFC activation loss, ohmic loss, and concentration loss improve the energy utilization rate of AMFC electricity production, and
– increase the current when the AMFC is running so that the AMFC is in a state of high output power.

Discussion

Transmission analysis of electron acceptor and substrate

The AMFC usually uses sodium acetate and glucose as substrates. The test results found that when glucose is used as the substrate, AMFC performs better. From the point of view of mass-flow, the same current is produced. The mass-flow of sodium acetate is 1.37 times that of glucose, and the consumption of glucose is smaller, from the point of view of mass transmission, the diffusion coefficient of sodium acetate is 1.83 times that of glucose. Sodium transports faster, from the perspective of solubility, sodium acetate's solubility is three times that of glucose, and sodium acetate can form a larger concentration gradient. Existing documents are all based on the same mass concentration for comparison [10]. Under the same mass concentration, the COD value of glucose solution is significantly higher than that of the sodium acetate solution. Besides, glucose is the most easily degradable substrate and is easily consumed by microorganisms. However, different microorganisms have different preferences for substrates. Most electricity-producing microorganisms prefer acetic acid and lactic acid as electron donors. It is estimated that the carbon chain of glucose is long, and that of acetic acid and lactic acid is short, so most electricity-producing microorganisms prefer the latter. The degradation of glucose by microorganisms may first degrade glucose into acid and further degrade it into CO_2. Whether glucose is superior to sodium acetate as a substrate needs further verification.

The most commonly used electron acceptors include oxygen, potassium ferricyanide, and potassium permanganate. The experiment found that when potassium ferricyanide and potassium permanganate are used as electron acceptors, AMFC performs better. From the point of view of mass-flow, the same current is produced, the mass-flow of potassium ferricyanide is 41.13 times that of oxygen, and the consumption of oxygen is much smaller, from the perspective of mass transmission, the diffusion coefficient of potassium ferricyanide is 5.79 times that of oxygen, potassium ferricyanide transmits faster; from the perspective of solubility, the solubility of potassium ferricyanide is $4.28 \cdot 10^4$ times that of oxygen, and potassium ferricyanide can form a larger concentration gradient, tab. 1. Overall, using potassium ferricyanide as an electron acceptor is more conducive to mass transfer than oxygen [11]. However, potassium ferricyanide and potassium permanganate can only be used in the laboratory because they are not renewable, and the cost of generating electricity is too high. The direction of future research is still to use oxygen as the electron acceptor.

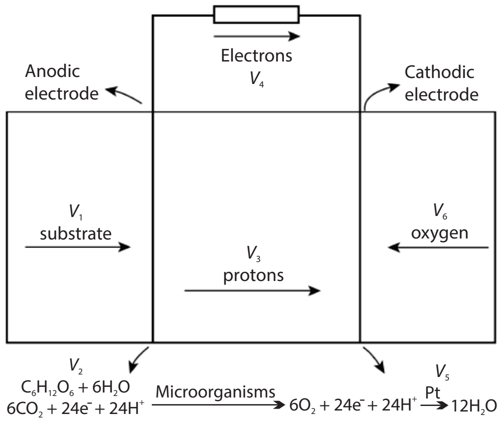

Figure 7. Schematic diagram of influencing factors of current

Current limiting factors

The electricity generation process of AMFC includes six steps, fig. 7:
– the mass transfer of the substrate in the anolyte,

- the decomposition of the substrate by the electricity-producing microorganisms on the surface of the anode electrode,
- the penetration of protons by protons; the mass transfer of the exchange membrane,
- the mass transfer of electrons through the external circuit,
- the reduction reaction of oxygen on the surface of the cathode electrode, and
- the mass transfer of the electron acceptor in the catholyte.

Therefore, six factors affect the AMFC current:

- the mass transfer rate of the substrate (V1),
- the anode electrochemical reaction rate (V2),
- the mass transfer rate of protons (V3),
- the electron mass transfer rate (V4),
- the cathode electrochemical reaction rate (V5), and
- the electron acceptor mass transfer rate (V6).

The mass transfer rate of electrons is much faster than the mass transfer rate of protons. The mass transfer of protons is driven by both the concentration driving force and the charge driving force, the substrate's mass transfer rate and the mass transfer rate of the electron acceptor are divided into diffusion transfer and convective transfer. It is currently mainly diffusion transfer, the type and number of microorganisms mainly limit the anode electrochemical reaction rate; the cathode electrochemical reaction rate is limited by the cathode area [12].

Analysis of ideal electricity production process

The ideal electricity generation process of AMFC should include three-stages, fig. 8. The first stage is the rising period. In this stage, the output voltage of AMFC gradually increases. The reason is that microorganisms need a period of domestication and culture for the anolyte, and the anode electrochemical reaction rate (V2) is the limiting factor of the output voltage. With the domestication and cultivation of microorganisms, the number of electricity-producing microorganisms gradually increased, V2 gradually increased, and finally, the output voltage reached the maximum. The second stage is a stable period. Due to the limitation of thermodynamics, the voltage of the AMFC cannot be increased indefinitely and due to the limitation of the number of microorganisms, the current of the AMFC cannot be increased indefinitely, so the output voltage of the AMFC will reach the maximum value. At this stage, V2, V3, or V5 are the limiting factors of the output voltage. The third stage is the down period. As the reaction progresses, the concentration of substrate and electron acceptor gradually decreases, and V1 or V6 becomes the limiting factor of the output voltage. The output voltage decreases as the concentration of substrate and electron acceptor decrease.

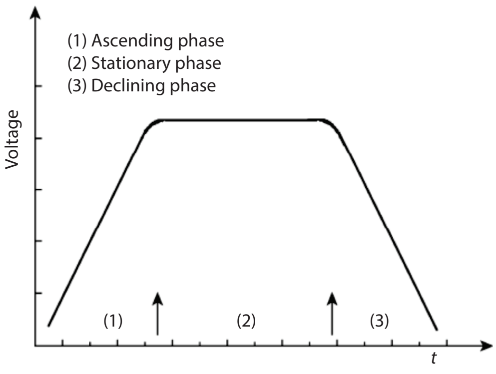

Figure 8. Ideal curve of output voltage *vs.* time

Conclusion

With the operation of AMFC, the pH value of the anolyte and the 3-D multi-flow conductivity show a downward trend, and the pH value of the catholyte and the 3-D multi-flow conductivity show an upward trend. The pH value of the catholyte is higher than the pH of the anolyte. The value is about 0.30-0.50 higher, and the average 3-D paleocurrent conductivity of the anolyte and catholyte does not change much. When AMFC operates stably, the internal ohmic resistance is 29.69 Ω, the limiting current is 2.69 mA, the maximum output power is about 0.8 mW, and the corresponding internal resistance is about 95.72 Ω. The mass transmission of potassium ferricyanide is the limiting factor of limiting current. Other microorganisms consume 91.1% of the glucose in the AMFC anolyte, and only 8.9% of the glucose is used for power generation. In comparison, 88.5% of the energy of the glucose used for power generation is converted into other forms of energy, and only 11.5% of the energy is converted into electrical energy. The ideal electricity generation process of AMFC includes three-stages: the limiting factor of the output voltage in the rising period is the anode electrochemical reaction rate, the limiting factor of the output voltage in the stable period is the anode electrochemical reaction rate, the cathode electrochemical reaction rate or the mass of protons transfer rate, and the limiting factor of the output voltage during the fall period is the mass transfer rate of the substrate or the mass transfer rate of the electron acceptor.

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