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МОДЕЛЮВАННЯ РОЗРЯДКИ ЛІТІЄВОЇ БАТАРЕЇ З Bi_2Se_3
ПОРОШКОПОДІБНИМ ЕЛЕКТРОДОМ

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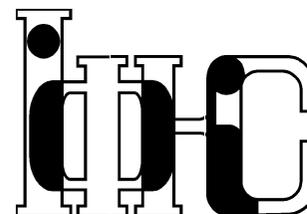
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Dmitry Portnyagin

MODELLING OF DISCHARGE OF LITHIUM BATTERY WITH
 Bi_2Se_3 POWDERED ELECTRODE

ЛЬВІВ

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Моделювання розрядки літієвої батареї з Bi_2Se_3 порошкоподібним електродом

Д.В. Портнягін

Анотація. Чисельно моделювалася циклічна зарядка-розрядка та розрядка літієвої батареї із Bi_2Se_3 порошкоподібним електродом в режимі заданої напруги та заданого струму. В дещо спрощеній моделі було отримано циклічні вольтамперні характеристики та розрядні криві, типові для двофазної системи.

Modelling of discharge of lithium battery with Bi_2Se_3 powdered electrode

D.V. Portnyagin

Abstract. Cycling under potentiodynamic control and galvanostatic discharge of lithium cell with Bi_2Se_3 powdered electrode has been modelled. Cyclic voltammograms and discharge curves typical to the two-phase system in a simplified model have been obtained.

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1. Introduction.

In recent years the simulation of the intercalation of lithium into the structure of porous electrode has been attracting the attention of several authors [1, 3, 5]. Mathematical simulation of charge/discharge processes allows to optimize the battery in order to obtain a higher performance. This can also help to analyze these processes to gain a deeper insight into the nature and courses of phenomena that occur during the cycling of these devices.

In these batteries lithium often serves as a cathode material because of its low electronegativity. Porous materials are used for anode due to their large surface area associated with high energy storage. Bi_2Se_3 can be viewed as an alternative material for anode in a high energy density portable power sources to be used in mobile phones, laptop computers, other portable devices and electric vehicles, the demand for which has raised in the market in recent years. The open-circuit potential curve of Bi_2Se_3 battery is characterized by plateau due to phase change.

It is widely held that the main driving force at the operation of the battery is diffusion and that the transport of ions across the electrode is governed by Fick's second law. In the present paper we have taken into account electrostatic interaction between ions and with the distribution of charge in the bulk of porous electrode.

We have made a comparison of the predictions of the model at different scan rates and different discharge currents, and obtained cyclic voltammograms and discharge curves, typical to the two-phase system.

2. Basic considerations.

We study the cycling under potentiodynamic control and galvanostatic discharge of lithium battery in a simplified model. The battery consists of lithium foil of thickness $L_{Li} = 0,55mm$, porous separator of thickness $L_s = 100\mu m$, Bi_2Se_3 powder electrode of thickness $L = 0,55mm$ made of spherical particles of radius $R_s = 50\mu m$, and current collector, and has the form of a cylinder with high $H = L_{Li} + L_s + L_1$ and radius of the base $R_c = 10mm$. The battery is immersed in 1M solution of $LiClO_4$ in propylene carbonate. Our assumptions of the model are (i) a uniform concentration of the electrolyte over time and throughout the cell, (ii) a uniform concentration of the intercalated lithium ions throughout anode, (iii) neglecting the electrodes' expansion and contraction.

During the discharge of the battery, lithium is dissolved into lithium ions from the negative electrode, migrates through the separator and

finally intercalates into the anode. During the charge the reverse process takes place. We assume that the intercalation of lithium ions into anode continues until the concentration of intercalated ions in anode reaches the value of that of solid lithium, which is associated with the formation of film of pure lithium.

The concentration of lithium inside the particle is a function only of radial distance, governed by the equation

$$\frac{\partial y}{\partial t} = \frac{1}{R^2} \frac{\partial}{\partial R} \left(R^2 \frac{D_s}{R_s^2} \frac{\partial y}{\partial R} \right) \quad (2.1)$$

$$y = y_0, \quad \text{at } \tau = 0, \forall R; \quad (2.2)$$

$$\frac{\partial y}{\partial R} = 0 \quad \text{at } R = 0, \forall \tau; \quad (2.3)$$

$$\frac{\partial y}{\partial R} = -\frac{j_n^+}{D_s} \frac{R_s}{C_{s,max}} \quad \text{at } R = 1, \forall \tau; \quad (2.4)$$

where t is time, $y = C_s/C_{s,max}$, $R = r/R_s$; are dimensionless variables. D_s is the diffusion coefficient in the solid phase, R_s is the radius of the particle, C_s is the concentration of lithium ions inside the particle, $C_{s,max}$ is the maximum concentration of lithium ions inside the particle, j_n^+ is the flux of lithium ions at the surface of the particle. The initial value of y is equal to 0.01. The flux of lithium ions at the surface of the particle is equal to the electrochemical reaction rate per unit of surface area of the particle as given by a Butler-Volmer reaction rate expression

$$j_n^+ = K (C(1 - y|_{R=1}))^{\beta-1} (y|_{R=1})^\beta \left\{ \exp \left[\frac{(1 - \beta)F}{\Re T} (U_{app} - U) \right] - \exp \left[\frac{-\beta F}{\Re T} (U_{app} - U) \right] \right\},$$

where C is the concentration of the electrolyte, which is taken constant over time, because the particles are assumed to be immersed in an excess of electrolyte, K is the reaction rate constant ($K = k_c^{1-\beta} k_a^\beta$), F is the Faraday constant, \Re is universal gas constant, T is temperature, U_{app} is the applied potential between solid phase and electrolyte, and U represents the open-circuit cell potential with respect to a metallic lithium electrode which is evaluated at the surface of the particle where the electrochemical reaction takes place and which is given by

$$U = \sum_{m=0}^5 U_m (y|_{R=1})^m \quad \text{for } 0 < y|_{R=1} < 1.$$

We take the diffusion coefficient

$$D_s = \sum_{m=0}^5 D_m y^m \quad (2.5)$$

The formulas for the open-circuit potential and the diffusion coefficient have been obtained by means of least square regression of experimental graphs.

We add the current caused by electric field to the righthand side of equation (2.1).

$$\frac{\partial y}{\partial t} = \frac{1}{R^2} \frac{\partial}{\partial R} \left(R^2 \frac{D_s}{R_s^2} \frac{\partial y}{\partial R} \right) - \frac{1}{FC_{max,s}} \text{div}(\sigma E), \quad (2.6)$$

where E is the electric field, σ the ionic conductivity given by Einstein relation

$$\sigma = y C_{max,s} N_a \overline{D_s} e^2 / kT,$$

k is Boltzman constant, e is elementary charge, N_a is Avogadro number, $\overline{D_s}$ is the average diffusion coefficient.

In the first approximation we assume that the current of positive ions through the surface of the particle is entirely due to the uniform distribution within the particle of negative charge, which Bi_2Se_3 , being more electronegative, draws from lithium, and the distribution of charge caused by imposed external electric field. However, X-ray photoelectron spectroscopy (XPS) proved [4] that after insertion the lithium retains only a fraction of the positive charge $+\delta$, while the anode material takes a negative charge $-\delta$. Therefore to the distribution of charge in the bulk of the particle we add the term associated with the nonuniform distribution of lithium ions. This results in

$$\text{div}(E) = \frac{3}{R_s \sigma_{eff,sol}} j_n^+ - \delta \frac{FC_{max,s}}{\varepsilon_0} (y_{avr} - y),$$

where $\sigma_{eff,sol}$ is the effective conductivity of electrolyte, ε_0 is the dielectric constant, $y_{avr} = \int y dV / V = 3 \int_0^1 y R^2 dR$ is the average concentration of ions in the particle, δ is the delocalization factor which equals 1 when we have naked lithium ions and negative charge, drawn from lithium, uniformly spread over Bi_2Se_3 sites, and equals 0 when negative charge is maximally localized on lithium ions.

The ionic current across the anode, i is equal to the external current through the battery at the contact with separator, and is zero at current

collector. Between these two values the current is assumed to be linearly distributed:

$$\frac{\partial i}{\partial x} = aFj_n^+,$$

where a is the interfacial area of particles per unit volume of powder electrode, calculated by

$$a = 0.02 \cdot 3(1 - \epsilon)/Rs;$$

ϵ - porosity of anode. It appears quite obvious that after we have pressed and baked the Bi_2Se_3 powder, only a fraction of the particle's surface will be exposed to electrolyte, so we have introduced a suitable factor in the formula for the interfacial area. Thus the density of total current through the cell is

$$i_{total} = aFj_n^+L.$$

All the parameters of the cell are evaluated at $T=298K$ for the reasons explained in [2]. The values of the coefficients in the expression for open-circuit potential, the diffusion coefficient, and the kinetic parameters are given in Table I.

System (2.6), (2.2)-(2.4) has been solved numerically.

3. Cyclic Voltammograms.

The applied potential is changed linearly with time and is given by

$$U_{app} = U_{ini} + \omega t$$

where U_{ini} is the initial applied potential, ω is the sweep rate, t is time. At a sweep rates 1, 0.5 and 0.2 mV/s the battery was discharged from its initial state to 1.2V, then it was charged to 2.5 V, after that the battery was again discharged to 1.2V. These steps were repeated twice to reach a periodic state. The periodic state is the state, at which the results are uniform and sustained during consecutive cycles, when cycled under the same conditions.

Let us clear up how the speed of changing of j_n^+ during the cycling depends on the speed of changing of the applied voltage U_{app} . We have

$$\begin{aligned} \frac{d}{dt}j_n^+ &= KC^{\beta-1} [(\beta - 1)(1 - y|_{R=1}) - \beta(y|_{R=1})] \times \\ &\times ((1 - y|_{R=1}))^{\beta-2} (y|_{R=1})^{\beta-1} \left\{ \exp \left[\frac{(1 - \beta)F}{\Re T} (U_{app} - U) \right] - \right. \end{aligned}$$

$$\begin{aligned} &- \exp \left[\frac{-\beta F}{\Re T} (U_{app} - U) \right] \left. \right\} \frac{dy}{dt} + \\ &+ KC^{\beta-1} \frac{F}{\Re T} ((1 - y|_{R=1}))^{\beta-1} (y|_{R=1})^\beta \{ (1 - \beta) \times \\ &\times \exp \left[\frac{(1 - \beta)F}{\Re T} (U_{app} - U) \right] + \end{aligned}$$

Table I. Model parameters for the lithium cell and physical constants.

Parameter	Value
U_{ini}	1.9387 V
U_0	1.9387 V
U_1	-4.2547 V
U_2	27.1704 V
U_3	-75.0395 V
U_4	93.1909 V
U_5	-43.0055 V
D_0	0.1323×10^{-12} m ² /s
D_1	0.1765×10^{-11} m ² /s
D_2	0.1400×10^{-10} m ² /s
D_3	0.3633×10^{-10} m ² /s
D_4	0.3950×10^{-10} m ² /s
D_5	0.1533×10^{-10} m ² /s
$C_{s,max}$	76,945 mol/m ³
β	0.5
K	10^{-7} mol ^{1/2} /m ^{1/2} s
C	1000 mol/m ³
T	298 K
$y_{initial}$	0.01
$\sigma_{eff,sol}$	6.0×10^{-1} S/m
R_s	50×10^{-6} m
L	0.55×10^{-3} m
k	1.381×10^{-23} J/K
N_a	6.022×10^{23} mol ⁻¹
\Re	8.314 J/(mol · K)
F	96,487 C/mol
ε_0	8.854×10^{-12} C ² /(N · m ²)
e	1.9×10^{-19} C
δ	10^{-9}
ϵ	0.7

$$+ \beta \exp \left[\frac{-\beta F}{\Re T} (U_{app} - U) \right] \left\{ \left[\frac{U_{app}}{dt} - U' \frac{dy}{dt} \right] \right\}.$$

Substituting for $\frac{dy}{dt}$ its expression from the diffusion equation in which we neglect electrostatic interaction, and substituting 1 for y and its spacial derivatives, since they are dimensionless magnitudes, hence we get the following estimate for the order of magnitude:

$$\frac{d}{dt} j_n^+ \simeq j_n^+ \left(\sum_{m=0}^5 |D_m| \right) / R_s^2 + (\pm 1) j_n^+ \frac{F}{\Re T} \left\{ \beta \frac{U_{app}}{dt} - \beta \left(\sum_{m=0}^5 |U_m| \right) \left(\sum_{m=0}^5 |D_m| \right) / R_s^2 \right\}, \quad (3.1)$$

where we take "+" when j_n^+ is positive, "-" when it is negative. Hence we can conclude that the hysteresis is the more significant, the steeper is the relation graph of j_n^+ vs. applied voltage, and the stronger, in turn, is the following inequality:

$$\frac{U_{app}}{dt} \gg \frac{\Re T}{F} \left(\sum_{m=0}^5 |U_m| \right) \left(\sum_{m=0}^5 |D_m| \right) / R_s^2. \quad (3.2)$$

And vice versa, the hysteresis is the less significant, the stronger is the reverse inequality:

$$\frac{U_{app}}{dt} \ll \frac{\Re T}{F} \left(\sum_{m=0}^5 |U_m| \right) \left(\sum_{m=0}^5 |D_m| \right) / R_s^2 \quad (3.3)$$

(in this case $\frac{d}{dt} j_n^+$ dose not become large, because the whole righthand side in (3.1) is multiplied by the derivative of y with respect to radial distance, which, the concentration profile being sloping, is small). The above said is verified by Figure 4.

The cyclic voltammograms of our battery are characterized by two peaks typical to the two-phase system, as Figures 1-3 indicate. One peak corresponds to discharge, the other - to charge. They are accounted for by the formation of double layer at the solid phase-solution interface and avalanche-like increase in the number of charge carriers. From the viewpoint of mathematical simulation they can be explained by the fact that when we move on a plateau the difference between the open-circuit

potential of the battery and the applied potential, as well as the current across the surface of the particle, increases rapidly, which corresponds to the peak. At the same time, when we aren't on a plateau anymore and the open-circuit potential begins to decrease, the aforementioned difference diminishes, which corresponds to the drop in absolute value of the current. The fact that these two peaks aren't identical testifies to that the charge /discharge processes are not completely reversible.

Figure 5 show the profile of dimensional concentration y vs. dimensional radial distance R at 1.6V of the applied potential corresponding to the discharge.

4. Discharge curves.

The battery is discharged from its initial state with $U_{imi} = 1.9387V$ to 0.01V cutoff voltage at current densities 120.46A/m² and 12.05A/m².

The local surface overpotential, i. e. the difference of potentials between solid and liquid phases is given by solving the equation

$$j_n^+(\eta, y) = \frac{-i_{total}}{aFL_1}$$

with respect to η . The total voltage of the cell is calculated by

$$V_{total} = \eta + (\phi_1 - \phi_2)_{kin},$$

where $(\phi_1 - \phi_2)_{kin}$ is given by kinetic expression

$$-i_{total} = FK_{Li}C^{0.5}(\exp((0.5F/(\Re T))(\phi_1 - \phi_2)) - \exp(-(0.5F/(\Re T))(\phi_1 - \phi_2)))$$

with K_{Li} - the reaction rate constant at the lithium electrode.

The utilization of the cell u is defined by

$$u = (3/L_1) \int_{x=L_s}^{x=L_s+L_1} dx \int_{R=0}^{R=1} yR^2 dR.$$

The discharge curves of our battery are characterized by plateaus typical to the two-phase system, as Figures 6-7 indicate. The plateau on the open-circuit potential curve and on the discharge curves corresponds to the formation of a new phase, when the potential remains constant. The graph of a larger discharge current lies below that of a smaller one and below the open-circuit potential curve due, firstly, to

the contribution of a kinetic term to the expression of a total voltage of the cell. Secondly, the graph of high discharge current takes on zero value earlier, because at larger discharge currents the change of concentration caused by diffusion process, which is associated with the establishment of equilibrium inside the particle, doesn't manage to overtake the change of concentration due to the influx of lithium ions through the surface of the particle, which in this case is large. Thus the given difference between local surface overpotential and the open-circuit potential is attained at smaller average concentrations, i. e. smaller utilizations. That is why the discharge curves, as Figure 10 shows, look contracted in horizontal direction, as compared to the open-circuit potential curve, and the more, the larger is the discharge current.

5. Temperature of the cell.

To determine the temperature of the cell we make the assumptions that the distribution of temperature throughout the cell is uniform at a given instant in time and the enthalpy of mixing and phase-change terms are neglected. With these assumptions the temperature is calculated according to the equation

$$\varrho C_p \frac{\partial T}{\partial t} = a_1 h (T_{amb} - T) + i_{total} \left(U - V_{total} - T \frac{d\bar{U}}{dT} \right),$$

where ϱ is the density of the cell, C_p is the heat capacity the cell, calculated as the average of the components of the cell, a_1 is the ratio of external cell surface area to geometric electrode surface area, $a_1 = (2(R_c^2 + RcH))/(R_c^2)$, T_{amb} is the ambient temperature, and h is the heat transfer coefficient. The entropy term $\frac{d\bar{U}}{dT}$ is

$$\frac{d\bar{U}}{dT} = \frac{\Re}{F} \ln \left(\frac{1-y|_{R=1}}{y|_{R=1}} \right) \text{ for.}$$

The mean cell heat capacity is calculated by formula:

$$C_p = C_{p,PC} \varrho_{PC} (L_1 \epsilon + L_s \epsilon) + (1 - \epsilon) C_{p,1} \varrho_1 L_1 + C_{p,Li} \varrho_{Li} L_{Li},$$

where ϱ_{Li} , ϱ_{PC} , and ϱ_1 are, respectively, the densities of lithium, propylene carbonate and Bi_2Se_3 electrode; $C_{p,PC}$, $C_{p,1}$, and $C_{p,Li}$ are the heat capacities of propylene carbonate, Bi_2Se_3 electrode, and lithium. Heat parameters of the cell are given in Table II.

Table II. Heat parameters of the cell.

Parameter	Value
T_{amb}	298 K
h	5 W/(m ² · K)
ϱ_1	7500 kg/m ³
ϱ_{Li}	534 kg/m ³
ϱ_{PC}	1200 kg/m ³
$C_{p,PC}$	720 J/(kg · K)
$C_{p,1}$	190.2798 J/(kg · K)
$C_{p,Li}$	3.72T + 2423 J/(kg · K)

As Figures 8,9 indicate, at smaller discharge currents the cell runs for a longer period of time, as Table III shows, generating more ohmic heat and, thus, heats to a higher temperature.

Table III. Discharge times.

Model and discharge rate	Time to discharge
$i_{total} = 12.05 A/m^2$	1797 s
$i_{total} = 120.46 A/m^2$	130 s

6. Conclusions.

We have made a simulation of the cycling of lithium cell with Bi_2Se_3 powder electrode under potentiodynamic control. The obtained voltammograms correspond to those of the two-phase systems with phase change, characterized by two peaks, the one of which corresponds to the discharge, the other - to the charge of the battery.

We have obtained discharge curves typical to the two-phase system with phase change. We have obtained the graphs of absolute temperature vs. utilization testifying to that at smaller discharge currents the battery heats to higher temperature.

Acknowledgment

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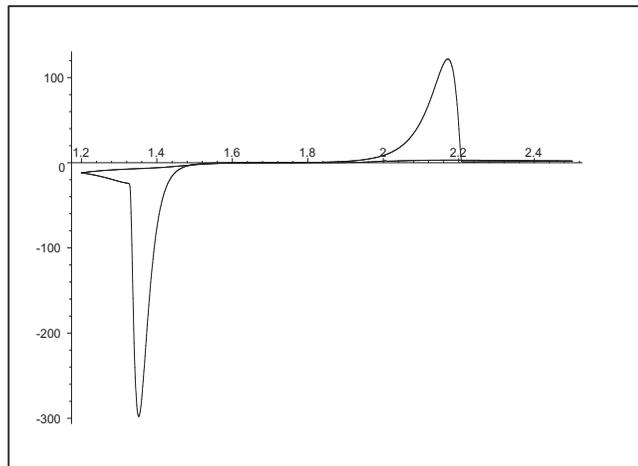


Figure 1. Current density (A/m^2) vs. applied voltage (V) at scan rate 1mV/s.

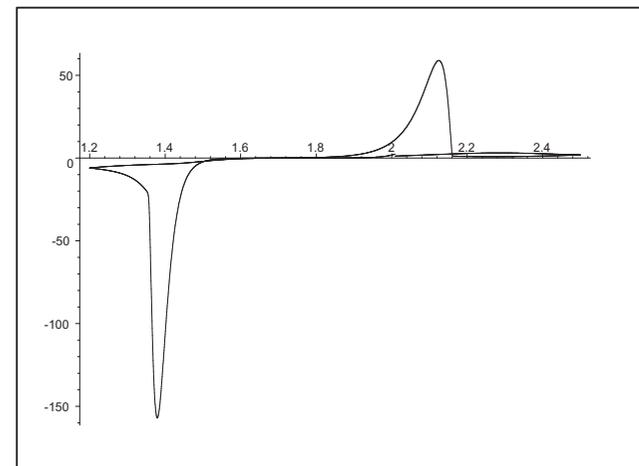


Figure 2. Current density (A/m^2) vs. applied voltage (V) at scan rate 0.5mV/s.

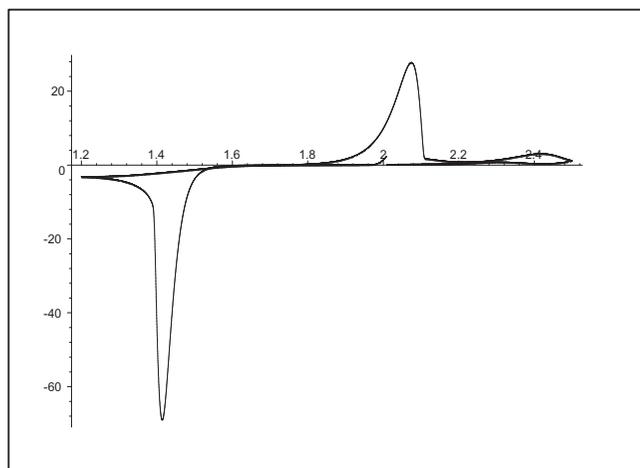


Figure 3. Current density (A/m^2) vs. applied voltage (V) at scan rate $0.2mV/s$.

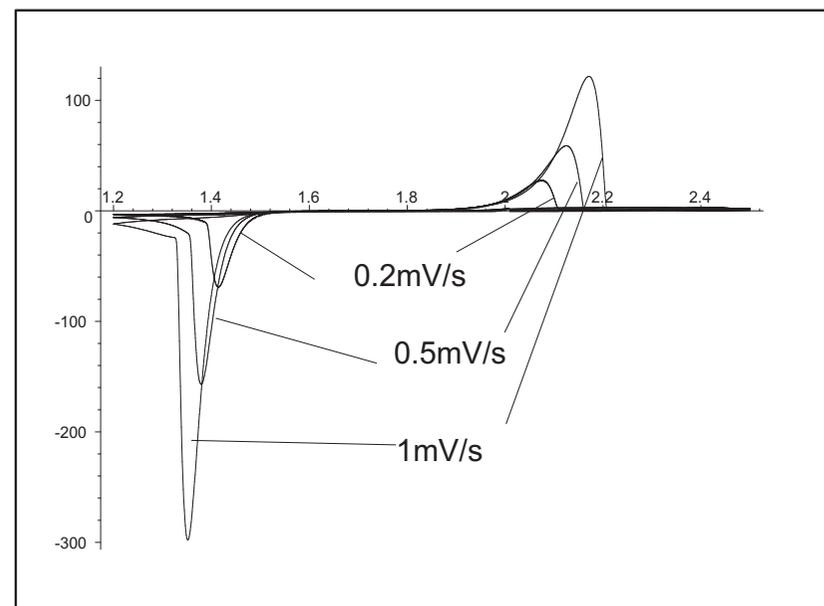


Figure 4. Comparison of graphs of current density (A/m^2) vs. applied voltage (V) at scan rates $1mV/s$, $0.5mV/s$, $0.2mV/s$.

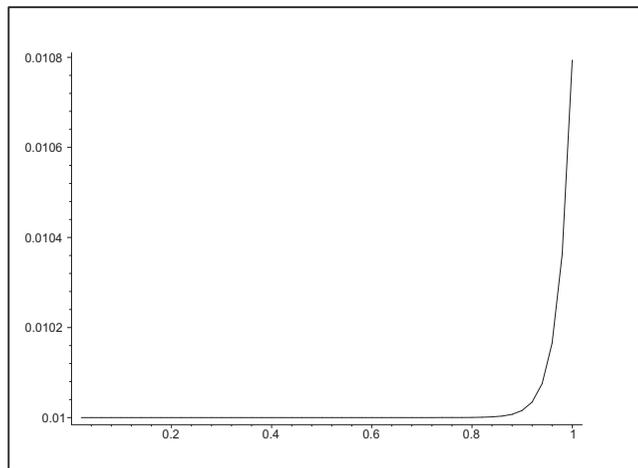


Figure 5. Profile of dimensionless concentration vs. dimensionless radial distance at applied voltage $U_{app}=1.6V$.

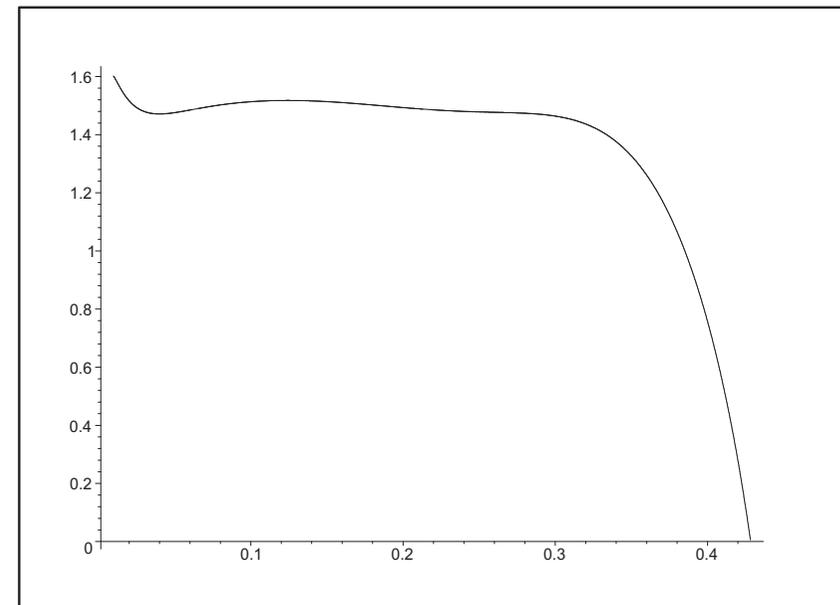


Figure 6. Cell voltage (V) vs. utilization at discharge rate 12.05 A/m^2 .

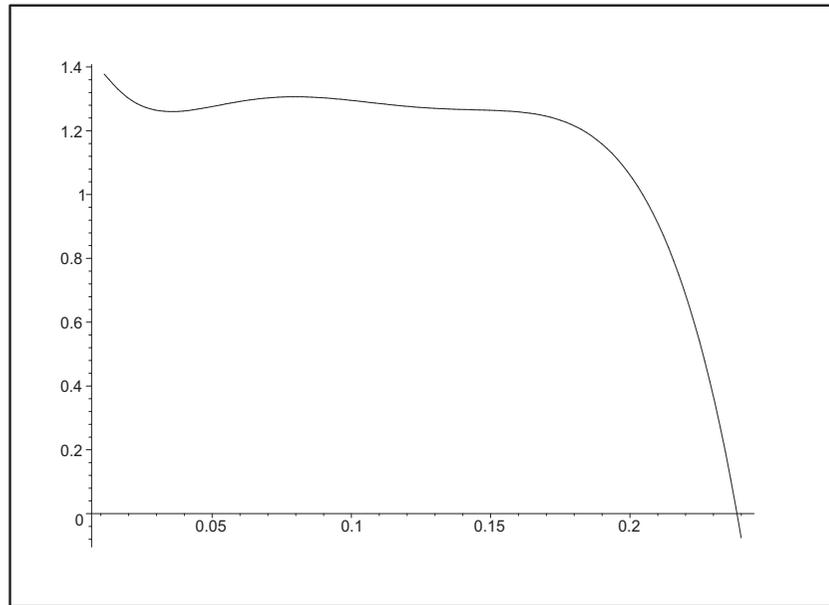


Figure 7. Cell voltage (V) vs. utilization at discharge rate 120.46 A/m^2 .

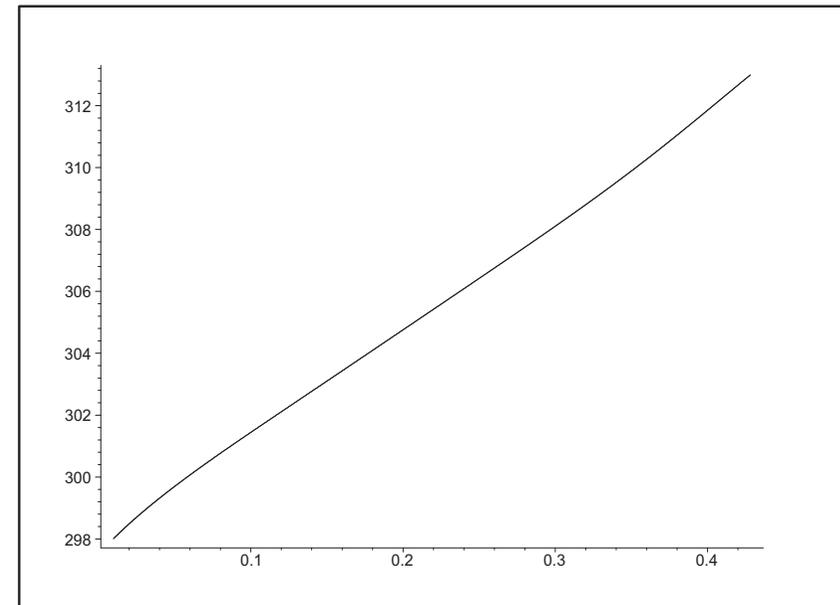


Figure 8. Temperature of the cell (K) vs. utilization at discharge rate 12.05 A/m^2 .

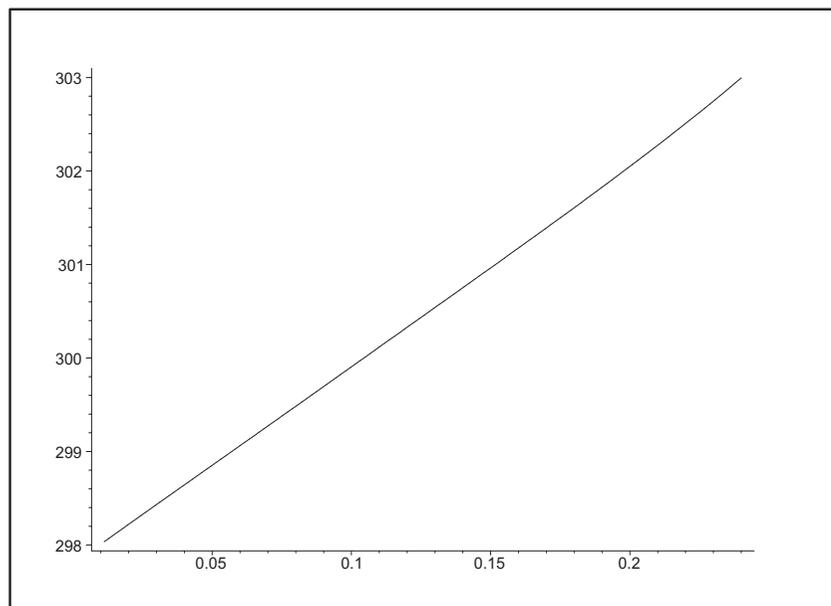


Figure 9. Temperature of the cell (K) vs. utilization at discharge rate 120.46 A/m^2 .

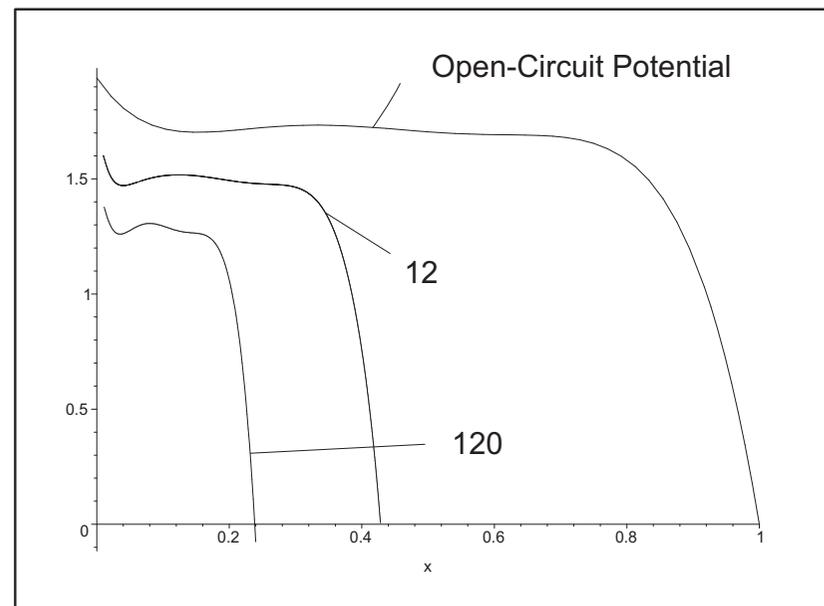


Figure 10. Comparison of cell voltage (V) curves at discharge rate, 12.05 A/m^2 and 120.46 A/m^2 , and of the open-circuit potential.

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