Formulation for the Performance Analysis of a Lithium-Carbon Monofluoride Cell

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ARTICLE INFO	ABSTRACT
Published Online: 04 April 2023	The formulation presented in this paper is for the performance analysis of a primary lithium-carbon monofluoride cell. The formulation can also be used to compute the theoretical cell open-circuit voltage; the fractional conversion of the limiting reactant, carbon monofluoride and hence, the cell fractional state-of-charge; total amount of charge required to be involved for the complete utilization of the cell cathode active material, carbon monofluoride as well as the transient specific amount of charge involved; the ratio of the thermal energy production to the electric energy production; and the
Corresponding Author: **Sarwan S. Sandhu**	rate of heat exchange between the cell exterior surface and its surroundings required for the determination of the cell average temperature.
KEYWORDS: theoretical open-circuit voltage, carbon monofluoride, lithium, primary, fractional conversion; heat generation	

I. INTRODUCTION

The work reported in the paper is in continuation of our recent effort to understand the performance behavior of the [lithium/electrolyte/$CF_{(x,(s),(x=1))}$] cells expressed in the form of the cell voltage versus time data during their discharge period [1, 2]. Based on the understanding of such experimented data, the formulation sets are developed to explain and analyze the cell performance data quantitatively; with the overall objective of identifying and determining the cell optimum design parameters and operational conditions leading to higher cell power and energy densities during the cell discharge periods.

A typical lithium-ion cell is sketched in Figure 1 below. Notation: L^A, L^S, and L^C = thickness of a lithium foil anode – electrode, porous separator, and particle packed – bed cathode electrode, respectively. The porous separator contains the electrolytic solution of LiFSI (lithium bis(fluorosulfonyl) imide); or a solvent mixture of, e.g., DME (dimethoxyethane) and EC (ethylene carbonate). The cell cathode contains the electrolytic solution in the pores between the solid composite particles containing the solid reactant material, $CF_{(s)}$ (carbon monofluoride), $L^A_{Cu\,(s)}$, $L^C_{Al\,(s)}$ = thickness of the cell anode and cathode current collector, respectively.

In the porous, packed bed cathode of a cell shown in Figure 1, the solid active material, $CF_{(s)}$ particles are wetted by the electrolyte solution of LiFSI in a solvent or solvent mixture. Lithium ions move in the micro-channels (e.g., size range: 1.5 – 5.0 Å), present in the active material, toward the particle centroids. During the cell discharge period, the electrochemical reaction: $Li_{(s)} \rightarrow Li^+ + e^-$, produces lithium ions (Li_s^+) and electrons (e_s^-). Lithium ions enter the electrolyte solution which makes an interface with the lithium foil anode. The reaction generated electrons enter the anode current collector for their supply to an external electrical load circuit. Lithium ions migrate through the electrolyte solution present in the cell porous separator made of Celgard 3500 and packed – bed cathode to arrive at the cell cathode solid active material particle – electrolyte solution interfaces. There they interact with the electrons transported via an electronic conductor, such as: graphene or carbon nanotubes (CNT's), from the cathode current collector, $Al_{(s)}$ sheet, which receives electrons from an external, electrical load circuit.

The basic formulation, to be presented in Section 2, links the amount of charge involved during the discharge period of the above – mentioned cell with the fractional conversion of the limiting reactant, $CF_{(s)}$, present in the cathode packed – bed of the solid particles composed of $CF_{(s)}$; electron – conducting material, such as graphene or carbon nanotubes; and a binding material, e.g., PVDF (polyvinylidene fluoride). The overall cell discharge process – activation energy barrier is here assumed to be equal to the sum of cell voltage losses associated with the electrochemical reactions as well as charge transfers at the cell electrode – electrolyte solution interfaces and the transport of ions and electrons under the effect of the electric and species concentration gradient fields in the various cell components.

II. FORMULATION

The overall reaction between lithium $\left(Li_{(s)}\right)$ and carbon monofluoride $\left(CF_{(s)}\right)$ – the limiting reactant of the cell cathode is:

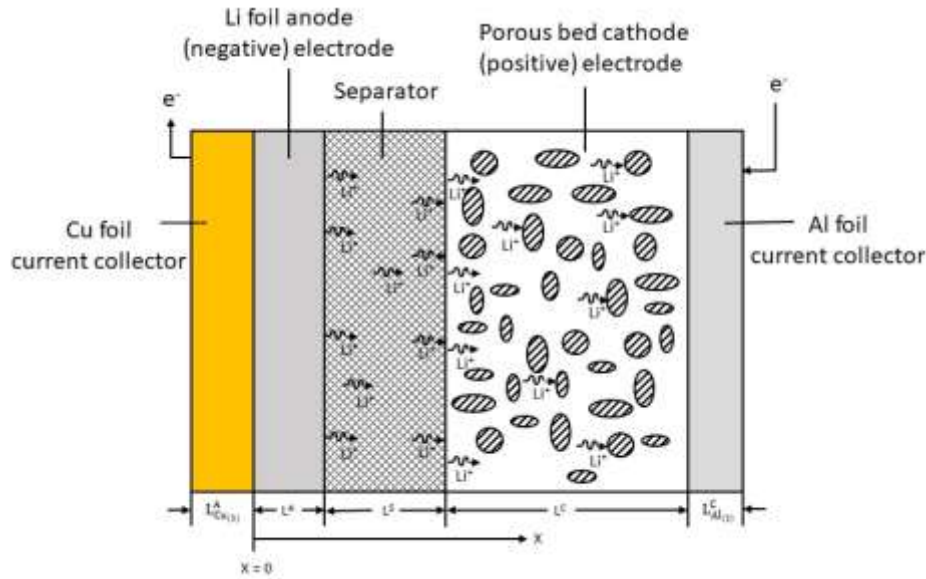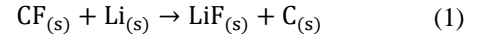$$CF_{(s)} + Li_{(s)} \rightarrow LiF_{(s)} + C_{(s)} \qquad (1)$$

Figure 1: Sketch [2] of a lithium – based galvanic cell during its discharge period.

Theoretical open – circuit cell voltage (OCV) at T [K] is given by:

$$E^{\circ}_{OCV} = \left(\frac{-\Delta G^{\circ}_{T}}{F}\right), [V] \qquad (2)$$

where

$$\Delta G^{\circ}_{T} = \left[\left(G^{\circ}_{LiF_{(s)}, T} + G^{\circ}_{C_{(s)},T}\right) - \left(G^{\circ}_{CF_{(s)},T} + G^{\circ}_{Li_{(s)},T}\right)\right], [J\ mol^{-1}] \qquad (3)$$

If ΔG°_{T} is not available, it can be determined from the following celebrated equation provided in the books of thermodynamics.

$$\frac{\Delta G^{\circ}_{T}}{RT} = \left(\frac{\Delta G^{\circ}_{T_0} - \Delta H^{\circ}_{T_0}}{RT_0}\right) + \frac{\Delta H^{\circ}_{T_0}}{RT}$$
$$+ \frac{1}{T}\int_{T_0}^{T}\left(\frac{\Delta C^{\circ}_p}{R}\right)dT$$
$$- \int_{T_0}^{T}\left(\frac{\Delta C^{\circ}_p}{RT}\right)dT \qquad (4)$$

where

$$\Delta H^{\circ}_{T_0} = \left(H^{\circ}_{LiF_{(s)},T_0} + H^{\circ}_{C_{(s)},T_0}\right)$$
$$- \left(H^{\circ}_{CF_{(s)},T_0} + H^{\circ}_{Li_{(s)},T_0}\right)$$
$$= \left(\Delta H^{\circ}_{f,LiF_{(s)},T_0} + \Delta H^{\circ}_{f,C_{(s)},T_0}\right)$$
$$- \left(\Delta H^{\circ}_{f,CF_{(s)},T_0} + \Delta H^{\circ}_{f,Li_{(s)},T_0}\right) \qquad (5)$$
$$= \left(\Delta H^{\circ}_{f,LiF_{(s)},T_0} - \Delta H^{\circ}_{f,CF_{(s)},T_0}\right), [J\ mol^{-1}]$$

$\Delta H^{\circ}_{f, i_{(s)},T_0}$ = enthalpy of formation of the chemical species $i_{(s)}$ in its defined standard state° from its elements at the reference state temperature of $T_0 = 298.15K$.

The relation between $\Delta G^{\circ}_{T_0}$ and $\Delta H^{\circ}_{T_0}$ is:

$$\Delta G^{\circ}_{T_0} = \Delta H^{\circ}_{T_0} - T_0 \Delta S^{\circ}_{T_0} \qquad (6)$$

where,

$$\Delta G^{\circ}_{T_0} = \left(\Delta G^{\circ}_{f,LiF_{(s)},T_0} + \Delta G^{\circ}_{f,C_{(s)},T_0}\right)$$
$$- \left(\Delta G^{\circ}_{f,CF_{(s)},T_0} + \Delta G^{\circ}_{f,Li_{(s)},T_0}\right) \qquad (7)$$
$$= \Delta G^{\circ}_{f,LiF_{(s)},T_0} - \Delta G^{\circ}_{f,CF_{(s)},T_0}, [J\ mol^{-1}]$$

where, $\Delta G^{\circ}_{f,C_{(s)},T_0} = \Delta G^{\circ}_{f,Li_{(s)},T_0} = 0.0\ [J\ mol^{-1}]$, and

$$\Delta S^{\circ}_{T_0} = \left(S^{\circ}_{LiF_{(s)},T_0} + S^{\circ}_{C_{(s)},T_0}\right)$$
$$- \left(S^{\circ}_{CF_{(s)},T_0} + S^{\circ}_{Li_{(s)},T_0}\right), [J\ mol^{-1}\ K^{-1}] \qquad (8)$$

is equal to the change in the entropy associated with the reaction, Eq. (1), with the species involved (taken in their defined standard states at the reference state temperature of $T_0 = 298.15K$).

The reaction, Eq (1), related heat capacity change, ΔC°_p , is given as:

$$\Delta C^{\circ}_p = \left[\left(C^{\circ}_{p,LiF_{(s)}} + C^{\circ}_{p,C_{(s)}}\right) - \left(C^{\circ}_{p,CF_{(s)}} + C^{\circ}_{p,Li_{(s)}}\right)\right], \qquad (9)$$
$$[J\ mol^{-1}\ K^{-1}]$$

where

$C_{p,i}^\circ$ = heat capacity of a species i in its defined standard state at the reaction temperature, T [K], [J mol^{-1} K^{-1}] (10)

and, R = the universal gas constant = 8.314 [J mol^{-1} K^{-1}].

For the initial amount of the limiting reactant, $CF_{(s)}$, in the cell cathode = $m_{CF_{(s)},0}$ [g]; the $CF_{(s)}$ moles are given by

$$N_{CF_{(s)},0} = \frac{m_{CF_{(s)},0}}{M_{CF}}, [mol] \quad (11)$$

where, M_{CF} = molecular weight of CF = 31.0096 [g mol^{-1}].

The fractional conversion of $CF_{(s)}$ by its reaction with lithium is defined as:

$$\xi = \frac{\begin{pmatrix} \text{moles of } CF_{(s)} \text{ converted or} \\ \text{consumed; for example,} \\ \text{during the cell discharge} \\ \text{period, in the cell cathode} \\ \text{at any time} \end{pmatrix}}{\begin{pmatrix} \text{initial moles of } CF_{(s)} \\ \text{in the cell cathode} \end{pmatrix}} \quad (12a)$$

$$\xi = \frac{N_{CF_{(s)},0} - N_{CF_{(s)}}}{N_{CF_{(s)},0}} = 1 - \frac{N_{CF_{(s)}}}{N_{CF_{(s)},0}} \quad (12b)$$

where $N_{CF_{(s)}}$ = moles of $CF_{(s)}$ still available for the reaction, Eq. (1), with lithium at a time during the cell discharge period. The relation between ξ and the cell fractional state of charge, f_{SOC}, is:

$$f_{SOC} = 1 - \xi \quad (13)$$

The fractional state – of – charge, f_{SOC}, is a measure of the unreacted, limited solid reactant, such as $CF_{(s)}$ in the cell cathode still available for its utilization at any time during the cell discharge period.

Note that according to Eq. (13), at $\xi = 0$, $f_{SOC} = 1$ and at $\xi = 1$, $f_{SOC} = 0$ as it should be. At $\xi = 1$, all of the solid, $CF_{(s)}$, reactant is consumed. Then, during the cell discharge period, the total amount of charge, in coulombs, involved in the reaction, Eq. (1), is as follows:

$$Q = \begin{pmatrix} \dfrac{F}{1 \text{ mole of}} \\ CF_{(s)} \text{ consumed} \end{pmatrix} \begin{pmatrix} N_{CF_{(s)},0}, \\ \text{moles of} \\ CF_{(s)} \\ \text{consumed} \end{pmatrix} \quad (14)$$

$$= \left(N_{CF_{(s)},0} F \right)$$

Where F = Faraday's constant = 96487 [coulombs (g-equivalent)$^{-1}$].

The fractional conversion of $CF_{(s)}$ during the cell discharge period is related to the cell discharge current, I_{disch}^{cell}, as:

$$\xi = \frac{N_{CF_{(s)},0} - N_{CF_{(s)}}}{N_{CF_{(s)},0}} = \frac{\int_{t=0}^{t=t} I_{disch}^{cell} dt/F}{N_{CF_{(s)},0}}$$

$$= \frac{\int_0^t I_{disch}^{cell} dt}{FN_{CF_{(s)},0}} \quad (15)$$

If I_{disch}^{cell} is kept at a constant level, Eq (15) reduces to:

$$\xi = \frac{I_{disch}^{cell} t}{FN_{CF_{(s)},0}} \quad (16)$$

If some of the limiting reactant, $CF_{(s)}$, in the cell cathode is in the product form initially; then, the initial fractional conversion of $CF_{(s)}$ is given by:

$$\xi_{ini} = \frac{\left(-\Delta N_{CF_{(s)},ini} \right)}{N_{CF_{(s)},0}} \quad (17)$$

where $\left(-\Delta N_{CF_{(s)},ini} \right)$ = moles of $CF_{(s)}$ present in the reacted form in the cell cathode before the start of cell discharge at time, t = 0.0 sec. For this situation, the fractional conversion of $CF_{(s)}$ during the cell discharge period is given by:

$$\xi = \xi_{ini} + \frac{\int_0^t I_{disch}^{cell} dt}{FN_{CF_{(s)},0}} \quad (18)$$

Total amount of charge involved, in coulombs, corresponding to the fractional conversion of $CF_{(s)}$, ξ, at time, t seconds, during the cell discharge period is given by:

$$Q_{t, \text{ amount of charge involved}} = (1 - f_{soc})Q = \xi Q \quad (19)$$

The amount of charge still required to be involved for the conversion of the remaining cathode reactant, $CF_{(s)}$,

$$Q_{t, \text{ remaining amount of charge still required to be involved}} = f_{soc}Q = (1 - \xi)Q \quad (20)$$

Specific amount of charge involved in the conversion of $CF_{(s)}$ in the cathode corresponding to its conversion level of ξ is given as:

$$Q_{t, \text{ specific amount of charge involved}} = \frac{Q_{t, \text{ amount of charge involved}}}{m_{CF_{(s)},0}} \quad (21)$$

$$= \left(\frac{\xi Q}{m_{CF_{(s)},0}} \right) \quad (22)$$

$$= \left[\frac{\xi \left(N_{CF_{(s)},0} F \right)}{m_{CF_{(s)},0}} \right] = \left(\frac{\xi F}{M_{CF}} \right), [\text{coulomb g}^{-1}] \quad (23)$$

$$= (2.7778 \times 10^{-4}) \left(\frac{\xi F}{M_{CF}} \right), [\text{Ah g}^{-1}] \quad (24)$$

$$= (0.2778) \left(\frac{\xi F}{M_{CF}} \right), [\text{mAh g}^{-1}] \quad (25)$$

It is here assumed that the total cell voltage loss, E_{loss}, at any time during the cell discharge period, is a measure of the total resistance to the occurrence of the overall cell discharge process composed of sub – processes of the electrochemical reactions and charge transfers at the cell electrode – electrolyte solution interfaces, and the transport phenomena involved in the movement of ions and electrons under the effects of the electric and species concentration gradient fields in the various

material components of the cell. Also, the overall energy barrier, $\Delta G_{ov,barrier}$, in [J mol^{-1}], to the occurrence of the overall cell discharge process is given by:

$$\Delta G_{ov,barrier} = E_{loss}F = (E_{OCV} - E_{actual})F \quad (26)$$

where E_{actual} = actual cell voltage, [V], at any time during the cell discharge period.

The cell electric power production rate [W] at any time is given by:

$$\dot{P}_{electric} = E_{actual}I_{disch}^{cell} \quad (27)$$

Thermal energy production rate, $\dot{P}_{thermal}$ [W], at any time during the cell discharge period due to the irreversible loss of some of the Gibbs free energy change of the cell reaction, Eq (1), is given by:

$$\dot{P}_{thermal} = E_{loss}I_{disch}^{cell}$$
$$= (E_{OCV} - E_{actual})I_{disch}^{cell} \quad (28)$$

It is noted here that $\dot{P}_{thermal}$ does not include the "reversible heat", $(T\Delta S)$, of the reaction, Eq. (1).

Total electric energy [J] production in time, t seconds, is given by:

$$E_{electric\ energy}^{cell} = \int_{t=0}^{t=t} \dot{P}_{electric}\ dt$$
$$= \int_{0}^{t} \left(E_{actual}I_{disch}^{cell}\right) dt \quad (29)$$

Total thermal energy [J] production in time, t seconds, is

$$E_{thermal\ energy}^{cell} = \int_{0}^{t} \dot{P}_{thermal}\ dt$$
$$= \int_{0}^{t} \left(E_{loss}I_{disch}^{cell}\right) dt \quad (30)$$

Assuming T_{skin} [K] is the surface average temperature of the cell at any time during the cell discharge period; then, the heat exchange [W] between the cell and its surroundings can be determined using the following expression:

$$\dot{q}_{heat-exchange} = h_{ov}\left(T_{skin} - T_{surroundings}\right)A_s \quad (31)$$

where $T_{surroundings}$ = temperature of the cell surroundings [K], A_s = outer surface area of the cell case, $[cm_{surf}^2]$, and h_{ov} = the overall heat – transfer coefficient for the heat exchange between the cell and its surroundings via both mechanisms of heat transfer of natural convection and thermal radiation processes, $[J\ cm_{surf}^{-2}\ sec^{-1}\ K^{-1}]$ or $[W\ cm_{surf}^{-2}\ K^{-1}]$.

For the determination of h_{ov}, the expressions are available in the references [1, 3, 4]. At any time, during the cell discharge period, the net heat exchange between the cell system and its surroundings is given as follows:

$$\dot{q}_{net,\ heat-exchange} = \left(\dot{P}_{thermal} - \dot{q}_{heat-exchange}\right) \quad (32)$$

If $\dot{P}_{thermal} > \dot{q}_{heat-exchange}$, thermal energy would accumulate in the cell and the cell temperature would increase as a function of time. If $\dot{P}_{thermal} < \dot{q}_{heat-exchange}$, the cell temperature would decrease. One may use the analytical equation provided in reference [1] to determine the average cell temperature as a function of time.

Specific electric energy $[Wh\ g_{CF_s}^{-1}]$ production as a function of ξ during the cell discharge period,

$$E_{sp-electric} = \left(\frac{\xi EF}{M_{CF}}\right) = 0.8643\xi E \quad (33)$$

Specific thermal energy $[Wh\ g_{CF_s}^{-1}]$ production as a function of ξ during the cell discharge period,

$$E_{sp-thermal} = \left(\frac{\xi FE_{loss}}{M_{CF}}\right) = 0.8643\xi E_{loss} \quad (34)$$

Ratio of the cell specific thermal energy production to its specific electric energy production,

$$\left(r_{thermal-to-electric\ production}\right) = \frac{E_{loss}}{E} \quad (35)$$

III. CELL EXPERIMENTAL DATA, APPLICATION OF THE FORMULATION, AND DISCUSSION

The data presented in Table 1 have been computed from Figure 1(f) of reference [5] using the technical information reported in the references [6 – 8]. The reported data of Ref. [5] is for the cell: $Li_{(s)}$/electrolyte in glass fiber/$CF_{x,(s),x=0.88}$; where the cell electrolyte is: 1M $LiPF_6$ in ethylene carbonate: dimethyl carbonate (1:1, v/v). The cell composite cathode is: $CF_{0.88\ (s)}$: acetylene black: PVDF (7:2:1, by weight). The overall cell reaction during the cell discharge period is:

$$CF_{0.88\ (s)} + 0.88\ Li_{(s)} \rightarrow 0.88\ LiF_{(s)} + C_{(s)} \quad (36)$$
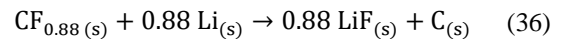
This reaction stoichiometric equation was utilized to calculate the data, shown in Table 1, from the experimental data of Figure 1(f) of Ref. [5].

Figure 2 shows that the cell operational voltage, E, decreases with an increase in the fractional conversion, ξ, of the limiting solid reactant, $CF_{x,(s),x=0.88}$ in the cathode electrode of the galvanic cell, while the cell's fractional state-of-charge, f_{SOC}, decreases during its discharge period. Ratio of thermal energy production (due to the loss of some of the Gibbs free energy change made available for useful work or electric energy production by the overall electrochemical reaction, Eq. (1)) to the electric energy production, $r_{thermal-to-electric}$, is presented as a function of ξ in Figure 3. It increases as ξ increases. At $\xi = 0.1872$, $r_{thermal-to-electric} = 0.6565$ is equivalent to 39.63% of (the cell-reaction Gibbs free energy change) conversion into thermal form and the remaining 60.37% of it appearing in the electrical form for its application in an external electrical load circuit. Similarly, at $\xi = 0.7313$, $r_{thermal-to-electric} =$

0.9622 is equivalent to 49.04% loss of the reaction Gibbs free energy change in the thermal energy form and the remainder 50.96% in the electric energy form. From this quantitative information, it is here concluded that at least the average interior temperature of a [lithium/electrolyte/CF$_{(s)}$ or CF$_{x,(s)}$] cell as well as that of a battery consisted of such cells connected-in-series must be controlled for the safe and efficient cell operation for electric energy production.
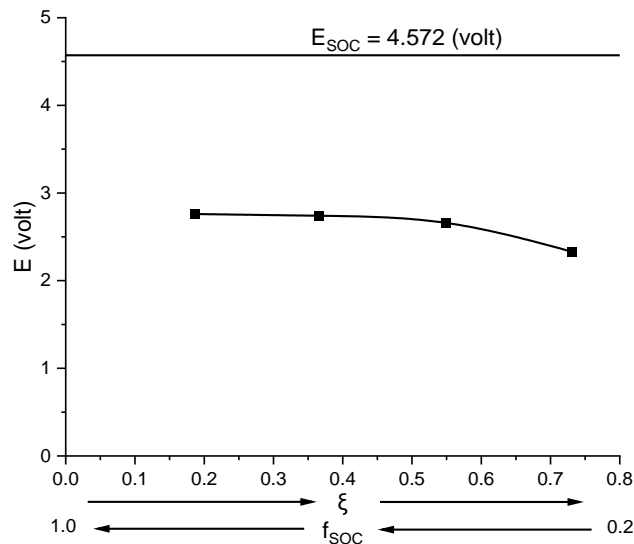
IV. CONCLUDING REMARKS

The formulation presented in this short paper was developed specifically for a [lithium/electrolyte/CF$_{(s)}$ or CF$_{x,(s)}$] galvanic cell. For the performance analysis of the experimental data obtained from such a cell during its discharge period, the provided formulation can be employed to compute the following:

(a) Theoretical cell open-circuit voltage.

(b) Fractional conversion of the limiting reactant, CF$_{x,(s),\ e.g.,\ x=1\ or\ 0.88}$ and hence, the cell fractional state of charge at any time during the cell discharge period at a fixed or transient cell current.

(c) Total amount of charge required to be involved for the complete utilization of the cell cathode active material, CF$_{x,(s)}$, as well as the specific amount of charge involved at any time.

(d) Ratio of the thermal to electric energy production.

(e) The cell electric power at any time and total transient electric energy production.

Table I: Computed Data from Figure 1 (f) of Reference [5]. E_{OCV} = 4.572 V, With the Lithium-ion Cell: $Li_{(s)}/CF_{x,(s),x=0.88}$; $I^{cell} = 0.02 \times (1C - rate) = \frac{1}{50}\left(\frac{821 mA}{g\ of\ CF_{(s)}}\right)$; Cell Discharge at Room Temperature = 25°C.

Fractional Conversion of CF$_{0.88\ (s)}$ $\xi = \dfrac{\left(-\Delta N_{0.88\ CF_{(s)}}\right)}{N_{CF_{(s)},0}}$	Cell fractional state of charge $f_{SOC} = 1 - \xi$	Operational Cell voltage E (V)	Cell voltage loss (V) $E_{loss} = (E_{OCV} - E)$ $= (4.572 - E)$	Ratio of thermal to electric energy production $r_{thermal-to-electric} = \dfrac{E_{loss}}{E}$
0.1872	0.8128	2.76	1.812	0.6565
0.3654	0.6346	2.74	1.832	0.6686
0.5482	0.4518	2.66	1.912	0.7188
0.7313	0.2687	2.33	2.242	0.9622

Figure 2: Cell voltage and voltage-loss versus ξ and f_{SOC} during the cell discharge period at room temperature for a Li$_{(s)}$/1M LiPF$_6$ in EC:DMC (1:1, v:v)/CF$_{0.88}$(s) cell.

(f) Rate of thermal energy production as well as the transient total thermal energy production due to the loss of some of the Gibbs free energy change of the overall cell reaction because of the irreversible occurrence of the species transport, charge transfer and electrode electrochemical reaction kinetics processes.

(g) Heat exchange between the exterior surface of the cell and its surroundings.

Finally, application of the presented formulation in conjunction with the experimental cell data from Ref. [5] shows an increase in the ratio of thermal-to-electric energy production with an increase in the fractional conversion of the cell cathode limiting reactant, $CF_{x,(s)}$.

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