Title: METHOD FOR DETERMINING EXTENT AND TYPE OF CAPACITY FADE

Abstract: A method of determining extent and type of capacity fade of an electrochemical cell in one embodiment includes identifying a first volume fraction of an active material in an electrode, identifying a first capacity of the first electrode at another time, identifying a second volume fraction of the first active material based upon the first capacity, identifying a first amount of the first active material lost from the first time to the second time based upon the first volume fraction and the second volume fraction, identifying a third volume fraction of an active material in another electrode, identifying a second capacity of the second electrode at a later time, identifying a fourth volume fraction of the second active material based upon the second capacity, and identifying a second amount of the second active material lost based upon the third volume fraction and the fourth volume fraction.

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ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, MK, MT, NL, NO, PL, PT, RO, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).
METHOD FOR DETERMINING EXTENT AND TYPE OF CAPACITY FADE

Field of the Invention

[0001] This invention relates to battery systems and more particularly to battery systems incorporating cells which exhibit capacity fade over the life of the system.

Background

[0002] A number of new battery chemistries are entering the market to provide capabilities required in specialized applications. At one time, the lithium-ion battery market was driven by the use of such batteries in portable electronics which require high energy but only limited life and power. More recently, other industries have focused on the use of batteries. By way of example, batteries are commonly incorporated into power tools and certain types of hybrid-electric vehicles. Each new industry requires different performance characteristics. Certain applications such as automotive applications require battery stability both in terms of battery safety for large packs and long life (at least 10 to 15 years).

[0003] Over the life of electrochemical cells, capacity can be lost due to side reactions and/or active material loss. Cell capacity is defined herein as the capacity of the cell between two particular cell voltages (e.g., 2.8 V and 4.1 V). The change in capacity usually results in changes in the relationship between the open cell potential (OCP) and capacity of the cell. The change in the relationship becomes problematic in that the OCP of a cell is a convenient measure of the state of charge (SOC) of the cell so long as the relationship between the OCP and the capacity of the cell is known.
A full discharge of a cell can be used to periodically establish the relationship between the OCP of a cell and the SOC of the cell. The opportunity to fully discharge a cell, however, is not readily available for various cell usages. By way of example, the entire available capacity range of the battery is not accessed during normal operation of a hybrid-electric vehicle. Moreover, it is inconvenient to interrupt operation of a vehicle merely to fully discharge a battery in order to diagnose the remaining capacity of the battery cells.

An article, Bloom, et al, "Differential voltage analyses of high-power, lithium-ion cells: 1. Technique and application," Journal of Power Sources, 139 (2005) 295, discusses a technique wherein low-rate measurements of cell voltage as a function of capacity could be used to determine the dominant contributors to capacity fade in a battery. The technique involves differentiation of the discharge voltage-vs.-capacity curves of a cell in order to obtain recognizable signatures that indicate the relative states of charge of the positive and negative electrodes. This technique, however, is limited in precision.

What is needed therefore is a method of determining the extent and type of capacity fade in a dual intercalation battery (e.g., a lithium-ion battery). A further need exists for a method which allows determination of the capacity of a battery when only a limited capacity or state-of-charge range is sampled. Additionally, the ability to more precisely identify features contributed by individual electrodes would be beneficial.
Summary

[0007] In accordance with one embodiment, a method of determining extent and type of capacity fade of an electrochemical cell in one embodiment includes identifying a first volume fraction of a first active material in a first electrode at a first time, identifying a first capacity of the first electrode at a second time, identifying a second volume fraction of the first active material based upon the first capacity, identifying a first amount of the first active material lost from the first electrode from the first time to the second time based upon the first volume fraction and the second volume fraction, identifying a third volume fraction of a second active material in a second electrode at about the first time, identifying a second capacity of the second electrode at about the second time, identifying a fourth volume fraction of the second active material based upon the second capacity, and identifying a second amount of the second active material lost from the second electrode from about the first time to about the second time based upon the third volume fraction and the fourth volume fraction.

[0008] In accordance with another embodiment, a method of determining extent and type of capacity fade of an electrochemical cell includes quantifying a first change in the volume fraction of active material in a negative electrode, quantifying a second change in the volume fraction of active material in a positive electrode, comparing the first change and the second change, and determining a side reaction loss based upon the comparison.
Brief Description of the Drawings

[0009] FIG. 1 depicts a graph of the OCPo/SOCo relationship for an exemplary cell along with the OCP_A/SOC_A relationship for the exemplary cell after the exemplary cell has been operated for a number of cycles;

[0010] FIG. 2 depicts a flow chart of a procedure which may be used to determine the extent and type of capacity fade in a battery system in accordance with principles of the present invention;

[0011] FIG. 3 depicts a graph of the relaxation voltage of an exemplary cell obtained in accordance with the procedure of FIG. 2 showing various curves used to obtain a best-fit of the data; and

[0012] FIG. 4 depicts a graph of the derived OCP_A/SOC_A relationship for the cell using data obtained through the best-fit analysis of FIG. 3 for an exemplary cell along with the modeled OCP_A/SOC_A relationship.

Description

[0013] For the purposes of promoting an understanding of the principles of the invention, reference will now be made to the embodiments illustrated in the drawings and described in the following written specification. It is understood that no limitation to the scope of the invention is thereby intended. It is further understood that the present invention includes any alterations and modifications to the illustrated embodiments and includes further applications of the principles of the invention as would normally occur to one skilled in the art to which this invention pertains.
FIG. 1 shows a graph of the relationship between the open cell potential (OCP) of an electrochemical cell and the state of charge (SOC) of the cell. The line 102 identifies the relationship between the OCP and the SOC of the cell at an earlier time of life while the line 104 identifies the relationship between the OCP and the SOC of the cell at a later time of life. The relationship between the OCP and the SOC for the cell represented by the lines 102 and 104 is the cumulative result of the contribution of each of the active materials within a cell to the SOC of the cell. A cell will have at least two active materials, with one active material in each electrode. Electrodes may further be blended with two or more active materials to provide desired operational characteristics.

As evidenced by the graph, the relationship between OCP and SOC for the cell changes over time. Thus, while an OCP of 3.8 volts is associated with a SOC of about 39% at the time of life represented by the line 102, the same voltage is associated with a SOC of about 58% at the time of life represented by the line 104. The 19 per cent difference indicates that the exemplary cell has experienced capacity fade. Moreover, the shape of the line 102 is different from the shape of the line 104. For example, at 3.8 volts, the slope of the line 102 is greater than the slope of the line 104. The differences in the shape of the lines 102 and 104 reflect changing contributions to the SOC of the various active materials over the life of the cell.

The data available in the graph may be used to further understand the nature of the capacity fade if the capacity of the cell is known. In particular, the capacity of the cell (C_{cell}) at any given time is given by:

\[ C_{cell} = \hat{C}_{cell} L_{cell} e_{cell} P_{cell} (SOC_{mix} - SOC_{mm}) , \]

where \( \hat{C}_{cell} \) is the cell specific capacity.
\( \varepsilon_{c,e} \) is the volume fraction of active material in the cell, pceii is the density of the active material in the cell, and Lc,ei is the thickness of the electrodes in the cell.

[0017] Of the foregoing, \( \dot{C} \), L, and p can be assumed to be invariant. Accordingly, if the original capacity of the cell is known, and the capacity of the cell at a later time along with the OCP/SOC relationship of the cell is known, the active material loss (AML) can be determined by resolving the foregoing equation such that:

\[
AML = 1 - \frac{\varepsilon_f}{\varepsilon_0}
\]

where \( \varepsilon_0 \) and \( \varepsilon_A \) are the earlier and later volume fraction of active material in the cell, respectively.

[0018] By further identifying the capacities and the OCP/SOC relationships of each of the electrodes within a cell, even greater insight into the internal processes of the exemplary cell may be obtained using the foregoing equations. For example, the capacity (Cpos) of the positive electrode of the cell at any given time is given by:

\[
C_{\text{pos}} = \dot{C}_{\text{pos}} L_{\text{pos}} \varepsilon_{\text{pos}} P_{\text{pos}} (y_{\text{max}} - y_{\text{min}}),
\]

where \( \dot{C}_{\text{pos}} \) is the positive-electrode specific capacity,
\( \varepsilon_{\text{pos}} \) is the volume fraction of active material in the positive electrode,
\( P_{\text{pos}} \) is the density of the positive-electrode active material,
\( L_{\text{pos}} \) is the thickness of the positive electrode,
\( y_{\text{max}} \) is the maximum SOC of the positive electrode, and
\( y_{\text{min}} \) is the minimum SOC of the positive electrode.
Similarly, the capacity ($C_{\text{neg}}$) of the negative electrode of the cell at any given time is given by:

$$C_{\text{neg}} = \hat{C}_{\text{neg}} L_{\text{neg}} e_{\text{neg}} \rho_{\text{neg}} (x_{\text{max}} - x_{\text{min}}).$$

From the two foregoing relationships, the volume fraction of active material may be determined if the capacities of the respective electrodes and the respective state of charge ranges are known and with the assumption that $\hat{C}$, $L$ and $p$ are invariant. Given the foregoing, the positive-electrode active-material loss (AML$_{\text{pos}}$) and negative-electrode active-material loss (AML$_{\text{neg}}$) at a given time are:

$$\text{AML}_{\text{pos}} = 1 - \frac{e_{\text{pos}}}{\epsilon_{\text{pos}0}} , \text{ and}$$

$$\text{AML}_{\text{neg}} = 1 - \frac{e_{\text{neg}}}{\epsilon_{\text{neg}0}},$$

where $\epsilon_{\text{pos}0}$ and $\epsilon_{\text{neg}0}$ are the earlier active-material volume fractions of the positive and negative electrodes, respectively.

The definition of capacity loss due to side reactions or other changes in capacity such as active material that becomes isolated from the electrodes depends upon whether more active material has been lost from the negative or positive electrode. If AML$_{\text{pos}}$ exceeds AML$_{\text{neg}}$, then the side-reaction loss (SRL) is

$$\text{SRL} = 1 - \frac{y_{\text{max}} - y_{\text{mm}}}{y'_{\text{max},0} - y'_{\text{min},0}},$$

where $y'_{\text{max},0}$ and $y'_{\text{min},0}$ are the initial SOC bounds for the positive electrode.

Similarly, if AML$_{\text{neg}}$ exceeds AML$_{\text{pos}}$, then

$$\text{SRL} = 1 - \frac{x_{\text{max}} - x_{\text{mm}}}{x'_{\text{max},0} - x'_{\text{min},0}},$$
where $x_{\text{max},o}$ and $x_{\text{min},o}$ are the initial SOC bounds for the negative electrode.

Accordingly, the overall capacity ratio (present capacity $C_A$) divided by initial capacity ($C_0$) is given by

$$\frac{C_A}{C_0} = (1 - \text{SRL})[1 - \max(\text{ARL}_{\text{pos}}, \text{ARL}_{\text{neg}})].$$

Thus, once the change in capacity of each of the electrodes in a cell is identified, the OCP/SOC relationship of an aged cell compared to the OCP/SOC relationship of the cell before aging can provide useful insight into the extent and type of capacity fade in a dual intercalation battery (e.g., a lithium-ion battery). The initial OCP/SOC relationship of an intercalation battery is easily obtained. If desired, actual values for the OCP at full charge and full discharge may be measured. Alternatively, the intrinsic thermodynamic properties of the cell materials can be used to provide the initial OCP/SOC relationship if the initial design parameters of $\hat{C}, \varepsilon, L$, and $p$ are known. Alternatively, the current discharged during a complete discharge of the cell can be measured to obtain the capacity of the cell.

Establishing the capacity and OCP/SOC relationship of an aged cell can be accomplished in a similar manner. In an operational cell, however, establishing the proper conditions to obtain the values can be problematic as discussed above. Accordingly, an aged OCP/SOC relationship in one embodiment is established using the procedure of FIG. 2.

FIG. 2 depicts a procedure 200 for determining, for an intercalation cell, the extent of capacity fade, and the type of capacity fade, wherein the type of capacity fade is categorized as positive-electrode active-material loss, negative-electrode active-material...
loss, and side-reaction loss. The procedure 200 begins at block 202 which may be prior to initial use of a cell. Initially, the relationship between the OCP (OCPo) and the SOC (SOCo) is determined for each material used in both electrodes of the cell (block 204).

[0027] The intrinsic thermodynamic relationships between the OCPo and SOCo for each electrode are dependent upon material properties which do not typically change. Exceptions can occur in materials whose crystal structure can change during cell use. Furthermore, the OCP-SOC relationship for a blended electrode, i.e., an electrode with more than one active material, can change over time (e.g., if active material of one type is destroyed to a greater extent than the other type); hence OCP-SOC data must be obtained for each active material used in a blended electrode. For the purposes of this example, the exemplary cell discussed in reference to the procedure 200 incorporates a crystal structure that does not change and neither the negative nor the positive electrode is a blended electrode.

[0028] After the cell has been in use (block 206), the relationship between the aged OCP (OCP_A) of the full cell and the aged capacity (C_A) of the cell is mapped (block 208). Various procedures may be used to obtain the relationship between the OCP_A and the C_A of the cell. In one approach, the cell is slowly discharged a fraction of its full capacity while measuring current output. After allowing the cell to relax, which can take on the order of one day, the OCP_A value for the remaining cell capacity may be obtained. The foregoing steps are repeated until the OCP_A for the capacity range of interest has been measured. For cells that exhibit open-circuit hysteresis, the procedure is repeated for the charge curve. This mapping procedure, while one of the more accurate procedures, is very time consuming.
[0029] An alternative approach to mapping the relationship between the OCP_A of the cell and the C_A of the cell is to slowly discharge and recharge the cell (e.g., at C/50, depending upon the impedance of the battery). The values obtained over the discharge/charge cycle are then averaged to obtain an estimate of the OCP_A/C_A relationship. This approach does not account for hysteresis. Thus, for cells which exhibit different charge/discharge relationships, such as two-phase materials, like LiFePO_4 which exhibits different diffusivities on charge and discharge, some inaccuracies are realized.

[0030] A further alternative for mapping the OCP_A/C_A relationship is to charge and discharge the cell at several different rates, generating a curve for both charge and discharge. The obtained curves are then extrapolated to zero current. This approach accounts for asymmetric battery impedance, but may not account for hysteresis. Some inaccuracies may be realized, however, since the relationship between current and the difference between the OCP_A and the OCPo may not be precisely linear, even at very low current flow. For cells without open-circuit hysteresis, the average of the two extrapolated curves may be used to obtain the OCP_A/C_A relationship.

[0031] In one of the quickest approaches, the cell is discharged a fraction of its capacity, and allowed to relax for a short time (e.g., 90 seconds). The shape of the voltage decay curve (the change in the measured OCP versus time) is then used to estimate the OCP_A. The fit that is used may be exponential or a series of exponential forms. Other functional forms may be used to fit the data to provide more accuracy in the OCPA /C_A estimate.

[0032] The time segment that is fit to the exponential curve in this fast approach depends upon cell chemistry and state of charge. Typically the first several seconds of
the relaxation is not used in the fitting procedure. The foregoing steps are then repeated to cover the capacity range of interest or the capacity range to which the data is limited by operational constraints. While the voltage decay curve obtained through this approach does not strictly follow a given functional form, the result may be close enough to derive a sufficiently good approximation of the \( \text{OCP}_A / C_A \) relationship. In systems where hysteresis is negligible, the average of the charge and discharge curves may be used. Alternatively, if the hysteresis as a function of the SOCs of individual half cells has previously been measured, the previously measured value may be used.

Finally, a physical model of the electrochemical cell can be used to estimate the \( \text{OCP}_A / C_A \) relationship from a history of measurements of the cell voltage and current.

Once the \( \text{OCP}_A / C_A \) data has been identified, the data may be filtered to reduce noise and/or charge/discharge curves may be averaged as desired. Next, various SOC parameters for each material in the aged cell are derived from the data (block 210) using the thermodynamic \( \text{OCP}_{0i} / \text{SOC}_{0i} \) relationships determined at block 204 for the materials in the two electrodes. Once the aged SOC (\( \text{SOC}_A \)) parameters are derived, the \( \text{SOC}_A \) parameters may be used to define the \( \text{OCP}_A / \text{SOC}_A \) relationship for the cell.

Derivation of the \( \text{SOC}_A \) parameters used to define the \( \text{OCP}_A / \text{SOC}_A \) relationship begins by defining the OCP for each of the electrodes as:

\[
\text{OCP}_{\text{neg}} = \text{OCP}_{n(i)x}, \quad \text{and}
\]

\[
\text{OCP}_{\text{pos}} = \text{OCP}_{p(0)y},
\]

where \( x \) is the SOC of the negative electrode and \( y \) is the SOC of the positive electrode. Accordingly, the OCP of the aged cell across both electrodes (\( \text{OCP}_A \)) is:

\[
\text{OCP}_A = \text{OCP}_{A\text{pos}} (y) - \text{OCP}_{A\text{neg}} (x)
\]
Next, the OCP_A as a function of C_A is defined to be V(C_A). Additionally, the remaining capacity of the cell at the OCP at which the OCP data measurement was initiated (block 208) is defined as C_1, and the remaining capacity of the cell at the OCP at which the OCP data measurement was terminated (block 208) is defined as C_2. The value of C_1 should be greater than the value of C_2. Accordingly, if a discharge segment was followed by a charge segment in the data, then C_1 defined as the capacity at the end of the charge segment, and C_2 is defined as the capacity at the end of discharge segment.

With the foregoing convention, the total available capacity of the aged cell (C_A), C_1, and C_2 may be used to define a dimensionless factor (θ) as:

$$\theta = \frac{C_2 - C_1}{C_1 - C_2}$$

Accordingly, because the relationship between i) the SOC_A, ii) the SOC at the initiation of the OCP data measurement (SOC_1), and iii) the SOC at the termination of the OCP data measurement (SOC_2) follows the relationship of C_A, C_1, and C_2, the factor θ may be used to define the SOC_A for each of the electrodes as:

$$x = x_2 + \theta(x_1 - x_2), \text{ and}$$

$$y = y_2 + \theta(y_1 - y_2),$$

where X_1 is the negative-electrode SOC corresponding to cell capacity C_1, 

x_2 is the SOC corresponding to C_2, 

y_1 is the positive-electrode SOC corresponding to cell capacity C_1, and 

y_2 is the SOC corresponding to C_2.

The parameters X_1, x_2, y_1, and y_2 are thus determined by fitting the estimated or measured OCP(C_A) and V(C_A) relationship downward from values dependent upon material properties. Hence, a relationship between X_A, Y_A, and C_A is uniquely determined
for the particular point in the life of the cell. Furthermore, \( \text{SOC}_A \) can be determined as a function of \( C_A \) (or \( x_A \) or \( y_A \)) even outside the range of measurement, according to the above definition of \( \text{OCP}_A \).

[0039] By tracking the above defined relationships as the cell ages, the data needed to identify the extent of side reaction and active material loss can be determined. First, the lower cutoff voltage is defined to be \( \text{OCP}_{\text{min}} \) and the upper cutoff voltage is defined to be \( \text{OCP}_{\text{max}} \). The SOC of each electrode at these voltages can be determined using the inverse of the definition of \( \text{OCP}_A \) discussed above, resulting in the following equation:

\[
X = x_2 + \frac{y - y_2}{y_1 - y_2} (x_1 - x_2).
\]

[0040] The \( x \) or \( y \) term in the foregoing equation may be eliminated by using the definition of the SOC for the other of the electrodes discussed above.

[0041] Next, the maximum and minimum SOC values for each of the electrodes is defined as follows:

\[
y_{\text{min}} = y(\theta \text{CP}_{\text{min}}),
\]

\[
y_{\text{max}} = y(\text{OCP}_{\text{min}}),
\]

\[
x_{\text{min}} = x(\text{OCP}_{\text{min}}), \text{ and}
\]

\[
x_{\text{max}} = x(\text{OCP}_{\text{max}}).
\]

[0042] Once the SOC data for each of the materials in the cell is identified, the extent of side reaction loss and active material loss can be derived from the SOC of the individual materials and the cell capacity (block 212). Data needed to identify the extent of side reaction and active material loss can be derived using the equations:

\[
C_{\text{pos}} = \hat{C}_{\text{pos}} L_{\text{pos}} \varepsilon_{\text{pos}} \rho_{\text{pos}} (y_{\text{max}} - y_{\text{min}}), \text{ and}
\]
Once the AML and SRL data is available, battery management strategies are modified based upon the updated data (block 214). Additionally, the updated relationship between the SOC of the individual materials in the cell and the capacity of the full cell may be used to update battery management system models (block 216). The process may then be terminated at block 218.

The process of FIG. 2 was verified by initially obtaining voltage relaxation data from a cell in which current was applied or discharged for a short time and the cell was then allowed to relax toward its OCP. After the relaxation, the current was again applied or discharged. In this way, the voltage relaxation data was collected at several different SOCs, on both charge and discharge cycles. The voltage relaxation data obtained is depicted in FIG. 3. The graph 220 depicts the change in cell voltage over time from the beginning of relaxation. The collected data are identified by circles and best fit curves are shown for the various data sequences.

By way of example, curve 222 is a best fit curve for data points 224. The data points 224 were obtained after a discharge cycle, with data point 224 obtained about 0.5 second after termination of discharge and data point 224 obtained about 1 second after termination of discharge. The data points 224 show that initially after termination of a discharge, the cell OCP rises fairly rapidly. By about 90 seconds, however, the change between successive data points 224 is relatively small, and the zero current value of the cell may be accurately determined using the curve 222.

Similarly, curve 230 is a best fit curve for data points 232. The data points 232 were obtained after a charge cycle, with data point 232 obtained about 0.5 second
after termination of discharge and data point 232 obtained about 1 second after termination of discharge. The data points 232 show that initially after termination of a discharge, the cell OCP drops fairly rapidly. By about 90 seconds, however, the downward drift of the data points 232 is relatively small, and the zero current value of the cell may be accurately determined using the curve 230.

[0047] A model was then used to simulate voltage relaxation data for a lithium-ion cell with typical parameters. Noise was added to the data in order to simulate actual data. Results of the simulation are depicted in the graph 240 of FIG. 4. Graph 240 depicts data points 242 derived from the graph 220. The data points 242 were used to derive the curve 244, defining a relationship between OCP and SOC. The curve 246 depicts the actual relationship between the modeled OCP and SOC with the values of \( x_{\text{min}}, x_{\text{max}}, y_{\text{min}}, \) and \( y_{\text{max}} \) set to 0.21, 0.361, 0.6459, and 0.7368, respectively.

[0048] The slight difference between the curve 244 and the curve 246 indicates that the estimated OCP/SOC relationship using the procedure of FIG. 2 was very accurate even though data was only collected between about 65% SOC and 82% SOC. The best-fit values determined by the algorithm for \( x_{\text{min}}, x_{\text{max}}, y_{\text{min}}, \) and \( y_{\text{max}} \) were 0.2099, 0.3638, 0.6502, and 0.7407, respectively, giving an error of less than 1%.

[0049] While the example discussed with reference to FIGs. 3 and 4 was directed to a cell with a single active insertion material, the procedure of FIG. 2 may be applied to blended electrodes as well. For blended electrodes, the ratio of active material volume fractions is incorporated as an additional fitting parameter.

[0050] While the invention has been illustrated and described in detail in the drawings and foregoing description, the same should be considered as illustrative and not
restrictive in character. It is understood that only the preferred embodiments have been
presented and that all changes, modifications and further applications that come within
the spirit of the invention are desired to be protected.
Claims

Claim 1. A method of determining extent and type of capacity fade of an electrochemical cell comprising:

identifying a first volume fraction of a first active material in a first electrode at a first time;
identifying a first capacity of the first electrode at a second time;
identifying a second volume fraction of the first active material based upon the first capacity;
identifying a first amount of the first active material lost from the first electrode from the first time to the second time based upon the first volume fraction and the second volume fraction;
identifying a third volume fraction of a second active material in a second electrode at about the first time;
identifying a second capacity of the second electrode at about the second time;
identifying a fourth volume fraction of the second active material based upon the second capacity; and
identifying a second amount of the second active material lost from the second electrode from about the first time to about the second time based upon the third volume fraction and the fourth volume fraction.

Claim 2. The method of claim 1, further comprising:
identifying a fifth volume fraction of a third active material in the first electrode at
about the first time;
identifying a third capacity of the first electrode at about the second time;
identifying a sixth volume fraction of the third active material based upon the
third capacity; and
identifying a third amount of the third active material lost from the first electrode
from about the first time to about the second time based upon the fifth volume fraction
and the sixth volume fraction.

Claim 3. The method of claim 1, wherein identifying a first volume fraction
comprises identifying a relationship between an open cell potential and a state of charge
of the first electrode.

Claim 4. The method of claim 3, wherein identifying a third volume fraction
comprises identifying a relationship between an open cell potential and a state of charge
of the second electrode.

Claim 5. The method of claim 4, wherein identifying a third volume fraction
comprises:
identifying an intrinsic thermodynamic relationship between the open cell
potential and the state of charge of the second electrode based upon a material property of
the second active material.
Claim 6. The method of claim 1, further comprising:

identifying a side reaction loss based upon the identified first amount and the identified second amount.

Claim 7. The method of claim 1, wherein the identification of the second volume fraction comprises:

determining a first dimensionless factor ($\theta$) based upon the identified first capacity; and wherein the identification of the fourth volume fraction comprises:

determining a second dimensionless factor ($\theta$) based upon the identified second capacity.

Claim 8. The method of claim 7, wherein the identification of the second volume fraction comprises:

determining a state of charge of the first electrode using the first dimensionless factor ($\theta$); and wherein the identification of the fourth volume fraction comprises:

determining a state of charge of the second electrode using the second dimensionless factor ($\theta$).
Claim 9. A method of determining extent and type of capacity fade of an electrochemical cell comprising:

quantifying a first change in the volume fraction of active material in a negative electrode;

quantifying a second change in the volume fraction of active material in a positive electrode;

comparing the first change and the second change; and
determining a side reaction loss based upon the comparison.

Claim 10. The method of claim 9, wherein the quantification of the first change comprises:

identifying a first volume fraction of a first active material in the negative electrode;

identifying a first capacity of the negative electrode at time later than the first volume fraction identification; and

identifying a second volume fraction of the first active material based upon the first capacity.

Claim 11. The method of claim 10, wherein the quantification of the first change further comprises:

identifying a third volume fraction of a second active material in the negative electrode;
identifying a second capacity of the negative electrode at time later than the third volume fraction identification; and

identifying a fourth volume fraction of the second active material based upon the second capacity.

Claim 12. The method of claim 10, wherein the quantification of the second change comprises:

identifying a fifth volume fraction of a third active material in the positive electrode;

identifying a third capacity of the positive electrode at time later than the fifth volume fraction identification; and

identifying a sixth volume fraction of the third active material based upon the third capacity.

Claim 13. The method of claim 10, wherein identifying a first volume fraction comprises identifying a relationship between an open cell potential and a state of charge of the negative electrode.

Claim 14. The method of claim 13, wherein the quantification of the second change comprises:

identifying an initial relationship between an open cell potential and a state of charge of the positive electrode;
identifying a third capacity of the positive electrode at time later than the initial relationship identification; and

identifying a sixth volume fraction of the third active material based upon the third capacity.

Claim 15. The method of claim 14, wherein the identification the initial relationship between an open cell potential and a state of charge of the positive electrode comprises:

identifying an intrinsic thermodynamic relationship between the open cell potential and the state of charge of the positive electrode based upon a material property of the active material in the positive electrode.

Claim 16. The method of claim 9, wherein the quantification of the first change comprises:

determining a first dimensionless factor (θ) based upon an identified capacity of the negative electrode; and wherein the quantification of the second change comprises:

determining a second dimensionless factor (θ) based upon an identified capacity of the positive electrode.

Claim 17. The method of claim 16, wherein the quantification of the first change comprises:

determining a state of charge of the negative electrode using the first dimensionless factor (θ); and wherein the quantification of the second change comprises:
determining a state of charge of the positive electrode using the second dimensionless factor ($\theta$).
1. Identify baseline

2. Map OCP vs. SOC for each material in cell

3. Operate cell

4. Map OCP for full cell vs. CAP

5. Derive SOC of individual materials from OCP vs. CAP

6. Derive AML and SRL from SOC of the material vs. cell CAP

7. Update battery management strategies with AML and SRL data

8. Update battery management systems with material SOC vs. CAP

END

FIG. 2
## INTERNATIONAL SEARCH REPORT

### A. CLASSIFICATION OF SUBJECT MATTER

**INV.** H01M10/48  G01R31/36  H02J7/00
**ADD.** H01M10/0525

According to International Patent Classification (IPC) or to both national classification and IPC

### B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

H01M  G01R  H02J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal , INSPEC

### C. DOCUMENTS CONSIDERED TO BE RELEVANT

<table>
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<tr>
<th>Category</th>
<th>Citation of document, with indication where appropriate, of the relevant passages</th>
<th>Relevant to claim No</th>
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<tr>
<td>X</td>
<td>SIKHA G ET AL: &quot;Effect of the Porosity on the Capacity Fade of a Lithium-Ion Battery. Theory&quot;</td>
<td>1,2, 6-12,16, 17</td>
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<td>, pages A1104-A1114, XP002594495</td>
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<td>ZHANG ET AL: &quot;Calendar life study of Li-ion pouch cells&quot;</td>
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<td></td>
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Further documents are listed in the continuation of Box C

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Date of the actual completion of the international search

30 July 2010

Date of mailing of the international search report

12/08/2010

Name and mailing address of the ISA/European Patent Office

P B 5818 Patentlaan 2

NL - 2280 HV Rijswijk

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Riba Vianova, Marta
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<tr>
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<td>CHRISTENSEN J ET AL: &quot;Cyclable Lithium and Capacity Loss in Li-Ion Cells&quot;</td>
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