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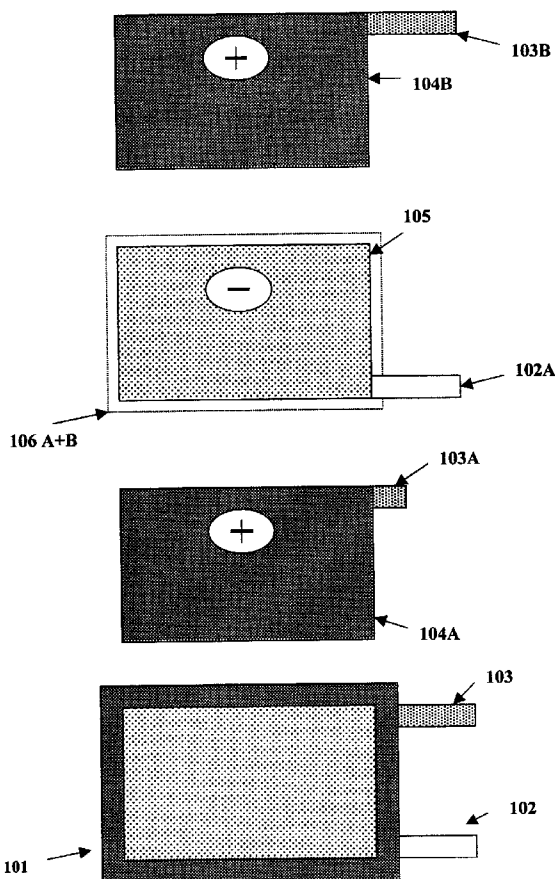
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(54) Title: LITHIUM-ION ELECTROCHEMICAL CELL AND BATTERY



(57) Abstract: The present invention provides new and useful concepts in a lithium-ion electrochemical cell and battery, including the use of carbon-carbon composite as active material for the anode, to improve (i) cycle life and self discharge characteristics, (ii) overcharge and overdischarge acceptance ability, and (iii) cathode utilization.



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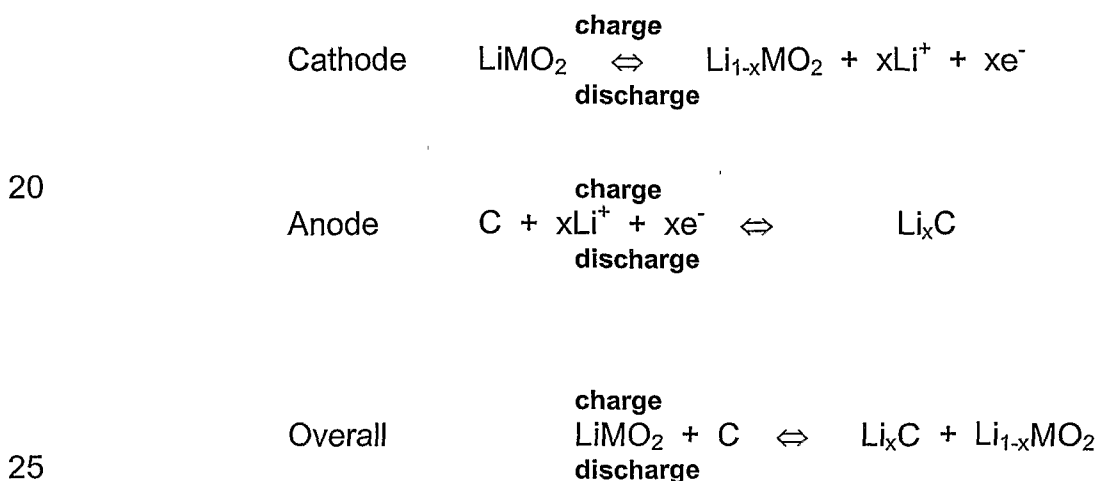
LITHIUM-ION ELECTROCHEMICAL CELL AND BATTERY

TECHNICAL FIELD

The present invention relates generally to improvements in a lithium-ion electrochemical cell and battery, including the use of carbon-carbon composite as active material for the negative electrode (anode) to
 5 improve (i) cycle life and self discharge characteristics, (ii) overcharge and overdischarge acceptance ability, and (iii) cathode utilization characteristics of such cell and battery.

BACKGROUND

Since its introduction and commercialization in 1991, rechargeable (or
 10 secondary) lithium-ion battery systems have received considerable interest not only to the battery community but also to the electronic and automobile industries. In lithium-ion batteries, carbon or graphite is used as an anode, a lithiated transition metal intercalation compound is used as a cathode and LiPF_6 is used as an electrolyte in carbonate-based organic solvents. For
 15 example, the reactions at the electrodes and overall cell reaction of an oxide-containing lithium intercalation compound are as follows:



where LiMO_2 represents the lithiated metal oxide intercalation compound.

Current Anodes

At present, hard carbon or graphite is used as active anode material in commercial lithium-ion batteries. Polyvinylidene fluoride (PVDF) is used as a binder to improve the mechanical integrity of the electrode. Copper is
5 universally used as the substrate for anode. Hard carbon or graphite material is mixed with PVDF in an organic solvent and the mixture is coated on the copper substrate to produce the anode.

Cycle Life and Self Discharge Issues

During charge-discharge process, due to intercalation and de-
10 intercalation of lithium-ions, a significant expansion and contraction of anodes occurs that can loosen the mechanical integrity and thereby increase the impedance of the electrodes. This increase in impedance of the anode causes capacity fade of lithium-ion batteries during cycling. The present state-of-the-art lithium-ion battery delivers approximately 500 cycles at 100%
15 depth of discharge with 80% capacity retention. There are many applications (e. g., aerospace and transportation) that demand higher cycle life.

Another disadvantage of the state-of-the-art lithium-ion battery is its relatively high self-discharge. The present lithium-ion battery loses 7% to 12% capacity per month at ambient temperature. The loss is even higher at
20 higher temperatures.

Overcharge/Overdischarge Issues

The present state-of-the-art lithium-ion cells require overcharge/overdischarge protection circuits and/or devices so that the cells can charge and discharge within certain voltage regimes. Overdischarge
25 causes dissolution of copper that is used as the substrate for carbon anode and degrades cell performance.

During overcharge, more lithium-ions are transported to the carbon anode and since the anode does not have enough room to accommodate them, overcharge may cause metallic lithium deposition on the anode, heat
30 build-up, and ultimately thermal run away of the cell.

Overcharge/overdischarge protection circuits and/or devices also increase(s) the weight and cost of the lithium-ion cell. Reliable and inexpensive overcharge/overdischarge protection for multi-cell lithium-ion battery is a major obstacle to commercialization of the systems in electric vehicle and other high voltage applications.

Cathode Utilization Issues

Because of their high electrochemical potential, LiCoO_2 , LiNiCoO_2 , and LiMn_2O_4 are attractive as cathode materials for lithium-ion battery systems. LiCoO_2 and LiNiCoO_2 have a layered structure, where lithium and transition metal cations occupy alternate layers of octahedral sites in a distorted cubic close-packed oxygen-ion lattice. The layered metal oxide framework provides a two-dimensional interstitial space, which allows for easy removal of the lithium ions. The LiMn_2O_4 spinel framework possesses a three-dimensional space via face sharing octahedral and tetrahedral structures, which provide conducting pathways for the insertion and extraction of lithium ions.

The removal and insertion of the lithium ions for the lithiated transition metal oxides are



In the present lithium-ion cells based on graphite or carbon anode with copper substrate, the value of x for LiCoO_2 is less than or equal to 0.5 (~140 mAh/g), for LiNiCoO_2 , x is less than or equal to 0.6 (~165 mAh/g) and for LiMn_2O_4 , x is less than or equal to 0.85 (~125 mAh/g). The value of x of these cathode materials can be increased at if the cell is charged at higher voltage. If an anode can accept higher cathode capacity at higher voltage without plating metallic lithium and deteriorating mechanical integrity (cycle life), a lithium-ion battery system with higher specific energy and energy density may result.

The present state-of-the-art lithium-ion battery systems, which use anodes of relatively poor mechanical integrity and low active materials limit their charge voltages to relatively low values and thereby limit the cathode capacity utilization. If the present anode with copper substrate was loaded
5 with more active anode material, the resulting anode will be thicker and heavier and will result in lower cycle life without significant improvement in specific energy (Wh/kg) and energy density (Wh/l).

SUMMARY OF THE PRESENT INVENTION

The present invention provides new and useful concepts in a
10 lithium-ion electrochemical cell and battery, including the use of carbon-carbon composite as active material for the anode, to improve (i) cycle life and self discharge characteristics, (ii) overcharge and overdischarge acceptance ability, and (iii) cathode utilization.

Cycle Life and Self Discharge

15 It is a primary objective of the present invention to improve the cycle life of lithium-ion electrochemical cells and battery systems using such cells.

Another objective of the present invention is to improve the self-discharge characteristics of lithium-ion electrochemical cells and battery systems using such cells.

20 In this aspect of the present invention the anode comprises carbon-carbon composite material, having high electronic conductivity and high lithium-ion intercalation capacity. The composite can accept repeated expansion and contraction as a result of intercalation and de-intercalation of lithium-ions during charge-discharge process with a little or no change in
25 mechanical integrity. The impedance of the anode, therefore, remains almost the same. The cycling behavior of the lithium-ion cells made with the carbon-carbon composite shows significant improvement. Moreover, a carbon-carbon composite electrode consists of a single phase, does not contain any binder and there is no metal carbon interface. The self-discharge behavior of

an electrochemical cell made with the carbon-carbon composite material as anode is, therefore, improved.

Overcharge/Overdischarge

It is also a primary objective of the present invention to improve the
5 overcharge/overdischarge acceptance ability of lithium-ion cells and battery systems, thereby eliminating the need for or simplifying overcharge/overdischarge protection circuits and/or devices, and reducing the cost and weight of lithium-ion cells and battery systems.

In accordance with this aspect of the invention, carbon film of high
10 electronic conductivity provides a carbon based substrate, and on which carbon material having high lithium-ion intercalation capacity is coated to produce the anode. Carbon-carbon composite of high electrical and thermal conductivity can also be used to form the anode. The substrate of the carbon-carbon composite anode is carbon, and the lithium-ion cell can accept
15 repeated overdischarge without performance degradation.

In each of the foregoing embodiments, the anode substrate and anode itself comprises carbon material, and each can accept lithium-ions during charge from cathode of lithiated intercalation compounds. The substrate can, therefore, act as a sink of lithium-ions. During overcharge, additional lithium-
20 ions from the cathode can, therefore, be stored to the anode substrate without causing metallic lithium deposition.

Thus, the anode made according to this aspect of the present invention allows the acceptance ability for overcharge and overdischarge of a lithium-ion cell.

25 Improved Cathode Utilization

It is yet another primary objective of the present invention to improve the content of anode active material and thereby the cathode capacity utilization of lithium-ion electrochemical cells and battery systems using such cells without the sacrifice of cycle life. The present invention improves the

specific energy and energy density of lithium-ion electrochemical cells and battery systems using such cells.

In this aspect of the present invention, a lithium-ion cell structure is provided having an anode comprising a carbon-carbon composite with a deliverable capacity at least 65% of the theoretical capacity of the cathode of the cell. The cell is charged at a potential configured to optimize the energy density of the cell, to improve cathode capacity utilization and thereby improve the specific energy and energy density of the cells and batteries.

BRIEF DESCRIPTION OF THE DRAWINGS

10 In the accompanying drawings:

Figure 1 is a schematic representation of a lithium-ion cell embodying a carbon substrate based anode in accordance with the present invention.

Figure 2 is a graph representing the cycling behavior of a lithium-ion cell made with carbon-carbon composite anode in accordance with the present invention;

Figure 3 represents the cycle life of a lithium-ion cell made in accordance with the prior art;

Figure 4 represents a comparison of the cycling behavior of lithium-ion cells made in accordance with the prior art (1) and in accordance with the present invention (2);

Figure 5 represents a comparison of the voltage decay of lithium-ion cells made in accordance with the prior art (1) and in accordance with the present invention (2);

Figure 6 represents the discharge behavior (before-1 and after storage-2) of a lithium-ion cell made in accordance with the prior art;

Figure 7 represents the discharge behavior (before-1 and after storage-2) of a lithium-ion cell made in accordance with the present invention; and

Figure 8 is a graph representing the overdischarge characteristics of a lithium-ion cell made in accordance with the present invention;

5 Figure 9 represents voltage and temperature responses during overcharge of a lithium-ion cell made in accordance with the present invention;

Figure 10 represents repeated overcharge/overdischarge characteristics of a lithium-ion cell made in accordance with the present
10 invention; and

Figure 11 is a graph representing the cathode capacity utilization of a lithium-ion cell made with carbon-carbon composite anode and lithium nickel cobalt dioxide cathode at different charge voltages in accordance with the present invention;

15 Figure 12 represents the deliverable capacity of a lithium-ion cell with respect to the total weight of the negative and positive electrodes made in accordance to the present invention;

Figure 13 represents the cycling behavior at higher cathode capacity utilization of a lithium-ion cell made in accordance with the present invention;

20 Figure 14 represents the cathode capacity utilization of a lithium-ion cell made in accordance with Example 7;

Figure 15 represents the deliverable capacity of a lithium-ion cell with respect to the total weight of the negative and positive electrodes made in accordance with Example 7; and

25 Figure 16 represents a comparison of the cycling behavior of lithium-ion cells made in accordance with Example 7 (1) and in accordance with the present invention (2);

DETAILED DESCRIPTION

In a preferred form of the present invention, a lithium-ion cell is comprised of a negative electrode of carbon-carbon composite material heat treated in the range of 1000°C to 3000°C and having high electronic and thermal conductivity, and a positive electrodes containing LiCoO₂, LiNiO₂,
5 LiNiCoO₂, LiMn₂O₄, LiMnO₂, LiV₂O₅, LiV₆O₁₃, LiTiS₂, Li₃FeN₂, Li₇VN₄, Li₇MoN₄, Li₂ZrN₂ or combinations of these materials. The carbon-carbon composite material is also used as the substrate for the negative electrode.

The electrolyte used in a lithium-ion cell and battery of the present
10 invention is a non-aqueous aprotic organic electrolyte and preferably a non-aqueous solution consisting of a solute, such as LiPF₆, LiBF₄, LiAsF₆, LiCF₃SO₃, LiN(CF₃SO₂)₂ or LiClO₄, dissolved in a solvent such as propylene carbonate, ethylene carbonate, diethyl carbonate, dimethyl carbonate, ethyl methyl carbonate and as well as mixtures thereof.

15 There are a number of known approaches suitable for producing carbon-carbon composite material, which are described e.g. in the following review Books: Essentials of Carbon-Carbon Composites, Edited by C. R. Thomas, The Royal Society of Chemistry, Cambridge, 1993 and Carbon-Carbon Composites, by G. Savage, Chapman & Hall, New York, 1993. The
20 disclosures of such review books are incorporated herein by reference. The carbon-carbon composite is made for this invention by heat treating at the temperature range of 1000-3000°C in inert atmosphere and can have density, specific resistance, and thermal conductivity in the range of 1.3-2.0 g/cc, 50-1,000 μohm-cm, and 50-600 Wm⁻¹K⁻¹, respectively. The carbon fiber used
25 to make the carbon-carbon composite can be pitch-, PAN-, and/or rayon-based fiber. For purpose of present invention, pitch and PAN-based fibers are preferable. Other than the foregoing general parameters, the present invention is not limited to any specific approach to produce carbon-carbon composite.

A preferred form of lithium-ion cell embodying a carbon-carbon composite anode is shown in Figure 1. Wherein the assembled cell 101 is shown with the anode, cathode, and electrolyte enclosed in a sealed sandwich structure with the carbon-carbon composite anode electrically accessible by means of protruding nickel conductive tab 102 and the lithiated intercalation compound cathode electrically accessible by means of a protruding conductive aluminum tab 103. The anode (not shown) and cathode (not shown) of the assembled cell 101 are separated by a porous separator (not shown) that is permeated with an aprotic non-aqueous electrolyte (not shown) that is in effective contact with both the anode and cathode.

In Figure 1, above the assembled cell 101, there is also shown the components of the cell 101, comprised of a pair of one-sided cathodes 104A and 104B and a carbon-carbon composite anode 105, suitable to be assembled as a sandwich (cell 101) with the anode 105 positioned between the respective cathodes 104A and 104B being separated from the anode 105 by respective porous separators 106A and 106B that are permeated with an aprotic, non-aqueous electrolyte that is in effective contact with both the cathode and the facing anodes.

Conductive aluminum tabs 103A and 103B are provided for the respective cathodes 104A and 104B and a nickel conductive tab 102A is provided for the anode 105, whereby the respective electrodes of the cell 101 are electrically accessible when assembled as a sandwich and enclosed within a sealed enclosure.

The materials of the respective anodes and cathodes of the cell 101 may be formed of materials, as described herein in further detail. For example, as described above, the anode comprises carbon-carbon composite. The cathode may be formed of LiCoO_2 , LiNiCoO_2 , LiNiO_2 , LiMn_2O_4 , LiMnO_2 , LiV_2O_5 , $\text{LiV}_6\text{O}_{13}$, LiTiS_2 , Li_3FeN_2 , Li_7VN_4 , Li_7MoN_4 , Li_2ZrN_2 or a combination thereof, supported by an aluminum foil substrate. The respective anode and cathode electrodes are maintained spaced from one

another by a respective electrically non-conductive separator that is permeable, whereby the aprotic, non-aqueous electrolyte is carried by the spacers and maintained in effective electrochemical contact with both the cathode and facing anode. The permeable separator may be formed of a micro-porous poly-olefin film.

It is to be understood that a plurality of electrochemical cells as described above can be used to assemble a battery of such cells by connecting the respective electrodes of the assembly of cells in an electrical circuit and in a known fashion to produce a battery with the voltage or current characteristics as determined by the number of cells connected in series or parallel circuit relationship.

The following specific examples are given to illustrate the practice of the invention, but are not to be considered as limiting in any way.

EXAMPLES

15 Cycle Life and Self Discharge Aspect of Invention

Example 1

Carbon-carbon composite heat-treated to 2600°C in an inert atmosphere was used as an anode of a lithium-ion cell to evaluate the concept of the present invention. The carbon-carbon composite was used as a thin film of thickness 300 µm. The lithium-ion cell included a negative electrode made from the carbon-carbon composite, a lithiated nickel dioxide positive electrode and 1M LiPF₆ electrolyte in a mixture (1:1 v/v) of ethylene carbonate/dimethyl carbonate (EC/DMC) solvents. A micro-porous poly-olefin (Celgard 2400) separator was used in between the positive and negative electrodes to isolate them electronically. The positive electrode was made from a mixture of 85% LiNi_{0.8}Co_{0.2}O₂, 6% carbon black, and 9% PVDF in DMF by coating on to an aluminum foil.

The aprotic, non-aqueous 1M LiPF₆ electrolyte mixture permeated the micro-porous poly-olefin separator, whereby the electrolyte was in effective contact with both the positive and negative electrodes, which were nevertheless maintained space and electrically isolated from one another.

5 The developed cell was charged at a constant current of 0.5 mA/cm² to 4.0 V and then at a constant voltage (4.0 V) for 3 hours or until the current dropped to 0.02 mA/cm². The cell was then discharged at a constant current of 0.5 mA/cm² to a cut-off voltage of 2.75 V. The charge discharge process was repeated in order to evaluate the cycle life. Figure 2 shows the cycling
10 characteristics of the developed cell according to the present invention. The cell delivered 580 cycles with 91.8% capacity retention.

A lithium-ion cell was made with the same components as described above except the negative electrode was made from a mixture of 90% MCMB 2528 carbon and 10% PVDF in DMF by coating on to a copper foil. It is
15 noteworthy to mention that MCMB 2528 carbon is used as an active material of anode for commercial lithium-ion cell. The cell was charged and discharged under the same conditions as the previous cell. Figure 3 shows the cycling behavior of this prior art cell. The cell lost 20% capacity after delivering only 557 cycles. Figure 4 shows a comparison to the cycling
20 behavior of the cell made in accordance to the present invention (shown at 2 in Figure 4 and that made according to the prior art (shown at 1 in Figure 4).

Example 2

A lithium-ion cell was made as in Example 1 with the carbon-carbon composite heat treated to 2800°C as anode, lithiated cobalt oxide as active
25 cathode material and an electrolyte comprising 1M LiPF₆ in a mixture of ethylene carbonate and diethyl carbonate (1:1 v/v). The cell was first charged at a constant current of 0.5 mA/cm² to 4.1 V and then at a constant voltage (4.1 V) for a period of three hours or until the current dropped to 0.02 mA/cm². The cell was then discharged at a constant current of 0.5 mA/cm² to a cut-off
30 voltage of 3.0 V. The charge-discharge process was repeated at least two

times in order to obtain a cycling efficiency greater than 99%. The cell was then fully charged and left at open-circuit voltage (OCV) at ambient temperatures to evaluate the self-discharge behavior. The cell was left at OCV for a period of a month (720 hours). After storage, the cell was
5 discharged at a constant current of 0.5 mA/cm² to a cut-off voltage of 3.0 V.

A lithium ion cell was made with the same components as described above except the negative electrode was made from a mixture of 90% MCMB 2528 carbon and 10% PVDF in DMF by coating on to a copper foil. The above experimental steps were repeated with the cell.

10 Figure 5 shows a comparison of voltage decay with time for the lithium-ion cells made in accordance with the present invention (shown at 2 in Figure 5) and in accordance with the prior art (shown at 1 in Figure 5). A voltage decay of only 10 mV was observed with the cell made in accordance to the present invention. For the prior art cell, a voltage decay of 60 mV was
15 observed.

The discharge behavior (before and after storage; the "before" is shown at 1 and "after" is shown at 2 in Figure 6) of the lithium-ion cell made in accordance with the prior art is shown in Figure 6. The cell lost a capacity of 11% due to storage for a month. Figure 7 shows the discharge behavior
20 (before and after storage; the "before" is shown at 1 and "after" is shown at 2 in Figure 7) of the lithium-ion cell made in accordance with the present invention. This cell lost only 2% capacity after storage for a month.

Overcharge/Overdischarge Aspect of Invention

Example 3

25 Commercially available carbon conductive film (COER-X from REXAM) was used as a substrate for anode of a lithium-ion cell to evaluate the overcharge/overdischarge aspect of the present invention. The anode was made from a mixture of 90% MCMB 2528 graphite and 10% PVDF in DMF by coating on to the carbon film. The lithium-ion cell included the negative
30 electrode, a lithiated cobalt dioxide positive electrode and 1M LiPF₆ electrolyte

in a mixture (1:1 v/v) of ethylene carbonate/dimethyl carbonate (EC/DMC) solvents. A micro-porous poly-olefin (Celgard 2400) separator was used in between the positive and negative electrodes to isolate them electronically. The positive electrode was made from a mixture of 85% LiCoO₂, 6% carbon black and 9% PVDF in DMF by coating on to an aluminum foil.

The aprotic, non-aqueous 1M LiPF₆ electrolyte mixture permeated the micro-porous poly-olefin separator, whereby the electrolyte was in effective contact with both the positive and negative electrodes, which were nevertheless maintained space and electrically isolated from one another.

The developed cell was charged at a constant current of 30 mA to 4.1 V and then at a constant voltage (4.1 V) for 3 hours or until the current dropped to 2 mA. The cell was then discharged at a constant current of 30 mA to a cut-off voltage of 3.0 V. The charge discharge process was repeated at least three times in order to obtain a stable charge discharge capacity. The cell was then exposed to repeated overdischarges. Figure 8 represents a plot of repeated overdischarge. The cell was discharged at a constant current of 30 mA to -1.0 V. For the first two cycles, the cell was charged to 4.1 V and for the last two cycles, the charge voltage was 4.2 V. The capacities delivered during overdischarges are 180 mAh, 179 mAh, 193 mAh, and 193 mAh for the 1 to 4 cycles, respectively. The cell did not show any performance degradation.

Example 4

Two lithium-ion cells were made as in Example 3 with the carbon-carbon composite heat treated to 2800°C as anode, lithiated cobalt oxide as active cathode material and an electrolyte comprising 1M LiPF₆ in a mixture of ethylene carbonate and diethyl carbonate (1:1 v/v). The cells were first charged at a constant current of 0.5 mA/cm² to 4.1 V and then at a constant voltage (4.1 V) for a period of three hours or until the current dropped to 0.02 mA/cm². The cells were then discharged at a constant current of 0.5 mA/cm² to a cut-off voltage of 3.0 V. The charge-discharge process was repeated at

least two times in order to obtain a cycling efficiency greater than 99%. The cells were then used to carry out the overcharge experiment.

The cells were overcharged at 1C and C/3 rates. The voltage and temperature responses of the fully charged cells during overcharge at two different rates are shown in Figure 9. The temperatures recorded were the outside body temperature of the cells. At 1C charge rate, the cell voltage raised to 4.7 V maximum with insignificant increase in temperature.

Example 5

A lithium-ion cell was made as in Example 3 with the carbon-carbon composite heat treated to 2600°C as anode, lithiated cobalt oxide as active cathode material and an electrolyte comprising 1M LiPF₆ in a mixture of ethylene carbonate and diethyl carbonate (1:1 v/v). The cell was first charged at a constant current of 0.5 mA/cm² to 4.1 V and then at a constant voltage (4.1 V) for a period of three hours or until the current dropped to 0.02 mA/cm². The cell was then discharged at a constant current of 0.5 mA/cm² to a cut-off voltage of 3.0 V. The charge-discharge process was repeated at least two times in order to obtain a cycling efficiency greater than 99%. The cell was then used to carry out repeated overcharge/overdischarge experiment.

Figure 10 shows the repeated overcharge-overdischarge characteristics at 50 mA of the lithium-ion cell made in accordance with the present invention. There was no safety hazard or thermal run away of the cell. The cell can accept repeated overcharge and overdischarge and still deliver excellent capacity.

Cathode Utilization Aspect of Invention

Example 6

Carbon-carbon composite heat-treated to 2400°C in an inert atmosphere was used as an anode of a lithium-ion cell to evaluate the concept of the present invention. The carbon-carbon composite was used as a thin film of 210 μm thickness. The lithium-ion cell included a negative electrode made from the carbon-carbon composite, a lithiated nickel cobalt

dioxide positive electrode and 1M LiPF₆ electrolyte in a mixture (1:1 v/v) of ethylene carbonate/dimethyl carbonate (EC/DMC) solvents. The total weight of the negative electrode was 0.76 g. A micro-porous poly-olefin (Celgard 2400) separator was used in between the positive and negative electrodes to isolate them electronically. The positive electrode was made from a mixture of 85% LiNi_{0.8}Co_{0.2}O₂, 6% carbon black and 9% PVDF in DMF by coating on to an aluminum foil. The weight of the cathode excluding the weight of aluminum foil was 1.1 g.

The aprotic, non-aqueous 1M LiPF₆ electrolyte mixture permeated the micro-porous poly-olefin separator, whereby the electrolyte was in effective contact with both the positive and negative electrodes, which were nevertheless maintained space and electrically isolated from one another.

The developed cell was charged at a constant current of 0.5 mA/cm² to 4.0 V and then at a constant voltage (4.0 V) for 3 hours or until the current dropped to 0.02 mA/cm². The cell was then discharged at a constant current of 0.5 mA/cm² to a cut-off voltage of 2.50 V. The cell was then charged at 4.2 V and 4.3 V and discharged to 2.5 V in order to obtain the deliverable discharge capacities under these conditions. Figure 11 shows the cathode capacity utilization of the cell. Figure 12 represents the deliverable capacity of the cell with respect to the total weight of negative and positive electrodes. The charge discharge process was repeated at a charge voltage of 4.2 V in order to evaluate the cycle life. Figure 13 shows the cycling characteristics of the developed cell according to the present invention.

Example 7

A lithium-ion cell was made with the same components as described in Example 6 except the negative electrode was made from a mixture of 90% MCMB 2528 carbon and 10% PVDF in DMF by coating on to a copper foil. It is noteworthy to mention that MCMB 2528 carbon is used as an active material of anode for commercial lithium-ion cell. The total weight of the negative electrode was 1.3 g of which 0.76 g was for MCMB, 0.08 g was for PVDF, and 0.46 g was for copper foil. The anode was 240 μm thick. The

objective of the cell of Example 7 was to use components similar to those currently used to create lithium-ion cells, but to compensate the theoretical capacity of the cathode, by using higher amount of anode material.

5 The cell was charged and discharged under the same conditions as mentioned for the previous cell. Figure 14 shows the cathode capacity utilization of the cell of Example 7 at different charge voltage. Figure 15 represents the deliverable capacities of the cell of Example 1 with respect to the total weight of negative and positive electrodes at different charge voltages. The deliverable capacities of this cell are significantly lower (and
10 hence energy density) than that obtained from the cell made in accordance to the present invention (Example 6) particularly at higher cathode utilization voltage. Figure 16 shows a comparison to the cycling behavior of the cells made in accordance to the present invention (shown at 2 in Figure 16) and that made according to Example 7 (shown at 1 in Figure 16). The capacity
15 fade on cycling of the cell of Example 7 is very high at higher cathode utilization voltage.

CLAIMS

What is claimed is:

1. A rechargeable electrochemical cell comprising a body of aprotic, non-aqueous electrolyte, first and second electrodes in effective
5 contact with said electrolyte, the first electrode comprising a lithiated intercalation compound, and the second electrode comprising carbon-carbon composite.
2. An electrochemical cell as defined in claim 1, wherein the carbon-carbon composite is made by heating in the temperature range of
10 1000°C to 3000°C.
3. An electrochemical cell as defined in claim 1, wherein the carbon-carbon composite has a density in the range of 1.3 g/cc to 2.0 g/cc.
4. An electrochemical cell as defined in claim 1, wherein the carbon-carbon composite possesses an electrical specific resistance in the
15 range of 50-1,000 $\mu\text{ohm-cm}$.
5. An electrochemical cell as defined in claim 1, wherein the carbon-carbon composite has a thermal conductivity in the range of 50- 600 $\text{Wm}^{-1}\text{K}^{-1}$.
6. An electrochemical cell as defined in claim 1, wherein the
20 carbon-carbon composite is made from pitch-based carbon fiber.
7. An electrochemical cell as defined in claim 1, wherein the carbon-carbon composite is made from PAN-based carbon fiber.
8. An electrochemical cell as defined in claim 1, wherein the carbon-carbon composite is made from rayon-based fiber.
- 25 9. An electrochemical cell as defined in claim 1, wherein the lithiated transition metal intercalation compound of the first electrode comprises a compound taken from a group comprising LiCoO_2 , LiNiCoO_2 ,

LiNiO₂, LiMn₂O₄, LiMnO₂, LiV₂O₅, LiV₆O₁₃, LiTiS₂, Li₃FeN₂, Li₇VN₄, Li₇MoN₄, and Li₂ZrN₂, and combinations of the foregoing.

10. An electrochemical cell as defined in claim 1, wherein the electrolyte is a non-aqueous organic electrolyte solution comprising a lithium
5 compound solute dissolved in a non-aqueous solvent.

11. An electrochemical cell as defined in claim 10, wherein the electrolyte comprises a solute selected from a group comprising LiPF₆, LiBF₄, LiAsF₆, LiCF₃SO₃, LiN(CF₃SO₂)₂, and LiClO₄.

12. An electrochemical cell as defined in claim 10, wherein the
10 electrolyte comprises a non-aqueous solvent selected from a group comprising propylene carbonate, ethylene carbonate, diethyl carbonate, dimethyl carbonate and ethyl methyl carbonate, and combinations of the foregoing.

13. An electrochemical cell as defined in claim 1, wherein the first
15 electrode is a cathode comprising a metal substrate having the lithiated intercalation compound affixed to a surface thereof, wherein the second electrode is an anode comprising a carbon-carbon composite and wherein said respective surfaces of the cathode and anode are separated from one another by a micro-porous electrically non-conductive separator that is
20 permeated by said aprotic, non-aqueous electrolyte which is in effective contact with said respective surfaces of the anode and cathode.

14. An electrochemical cell as defined in claim 13, wherein the separator comprises a micro-porous poly-olefin film.

15. An electrochemical cell as defined in claim 13, wherein the
25 cathode and anode form a sandwich that is contained within a sealed enclosure and wherein each of said cathode and anode has a connector extending out of the sealed enclosure for connecting said cell to an external electric circuit.

16. An electrochemical cell as defined in claim 13, wherein the cathode including its substrate, anode, and the electrolyte permeated separator are all contained within a sealed enclosure and wherein each of said cathode and anode has a connector extending out of the sealed enclosure for connecting said cell to an external electric circuit.

17. A battery comprising a plurality of electrochemical cells as defined in claim 1, having their respective electrodes connected in an electric circuit defining a battery of said cells.

18. A battery comprising a plurality of electrochemical cells as defined in claim 13, having their respective electrodes connected in an electric circuit defining a battery of said cells.

19. A battery comprising a plurality of electrochemical cells as defined in claim 16, having their respective electrodes connected in an electric circuit defining a battery of said cells.

20. A rechargeable electrochemical cell comprising a body of aprotic, non-aqueous electrolyte, first and second electrodes in effective contact with said electrolyte, the first electrode comprising a lithiated intercalation compound, and the second electrode having a substrate comprising carbon-based material.

21. A rechargeable electrochemical cell as defined in claim 20, wherein said second electrode has a substrate comprising carbon-carbon composite material.

22. A rechargeable electrochemical cell as defined in claim 20, wherein said electrode comprises carbon material disposed on a substrate comprising carbon-based material.

23. A rechargeable electrochemical cell as defined in claim 22, wherein said substrate comprises a carbon-carbon composite material.

24. A rechargeable electrochemical cell comprising a body of aprotic, non-aqueous electrolyte, first and second electrodes in effective contact with said electrolyte, the first electrode comprising a lithiated intercalation compound, said cell having a predetermined discharge level, and
5 the second electrode having a substrate configured to provide overdischarge acceptance for said cell.

25. An electrochemical cell as defined in claim 24, wherein said cell has a predetermined charge level, and said substrate is configured to provide both overcharge and overdischarge acceptance for said cell.

10 26. An electrochemical cell as defined in claim 25, wherein said substrate comprises carbon-carbon composite material.

27. A battery comprising a plurality of electrochemical cells, each cell comprising a non-aqueous secondary cell, each cell having a predetermined discharge level, and each cell having an anode with carbon
15 based material providing the cell with overdischarge acceptance.

28. A battery as defined in claim 27, wherein each cell comprises an anode with a substrate comprising said carbon-based material.

29. A battery as defined in claim 28, wherein each cell comprises an anode comprising carbon material disposed on a substrate comprising said
20 carbon based material.

30. A battery as defined in claim 29, wherein the substrate for each anode comprises a carbon-carbon composite material.

31. A method of forming a lithium-ion electrochemical cell comprising the steps of:

25 a. providing a lithium-ion cell structure having an anode comprising a carbon-carbon composite configured to optimize the content of anode active material in the cell, and

b. charging the lithium ion-cell at a potential configured to optimize the energy density of the cell.

32. A method as set forth in claim 31, wherein said step of charging the lithium-ion cell comprises charging the cell at a potential greater than that
5 used in the present state-of-the-art-lithium-ion cells and batteries.

33. A method as set forth in claim 31, wherein said anode comprising carbon-carbon composite has a deliverable capacity at least 65% of the theoretical capacity of the cathode of the cell.

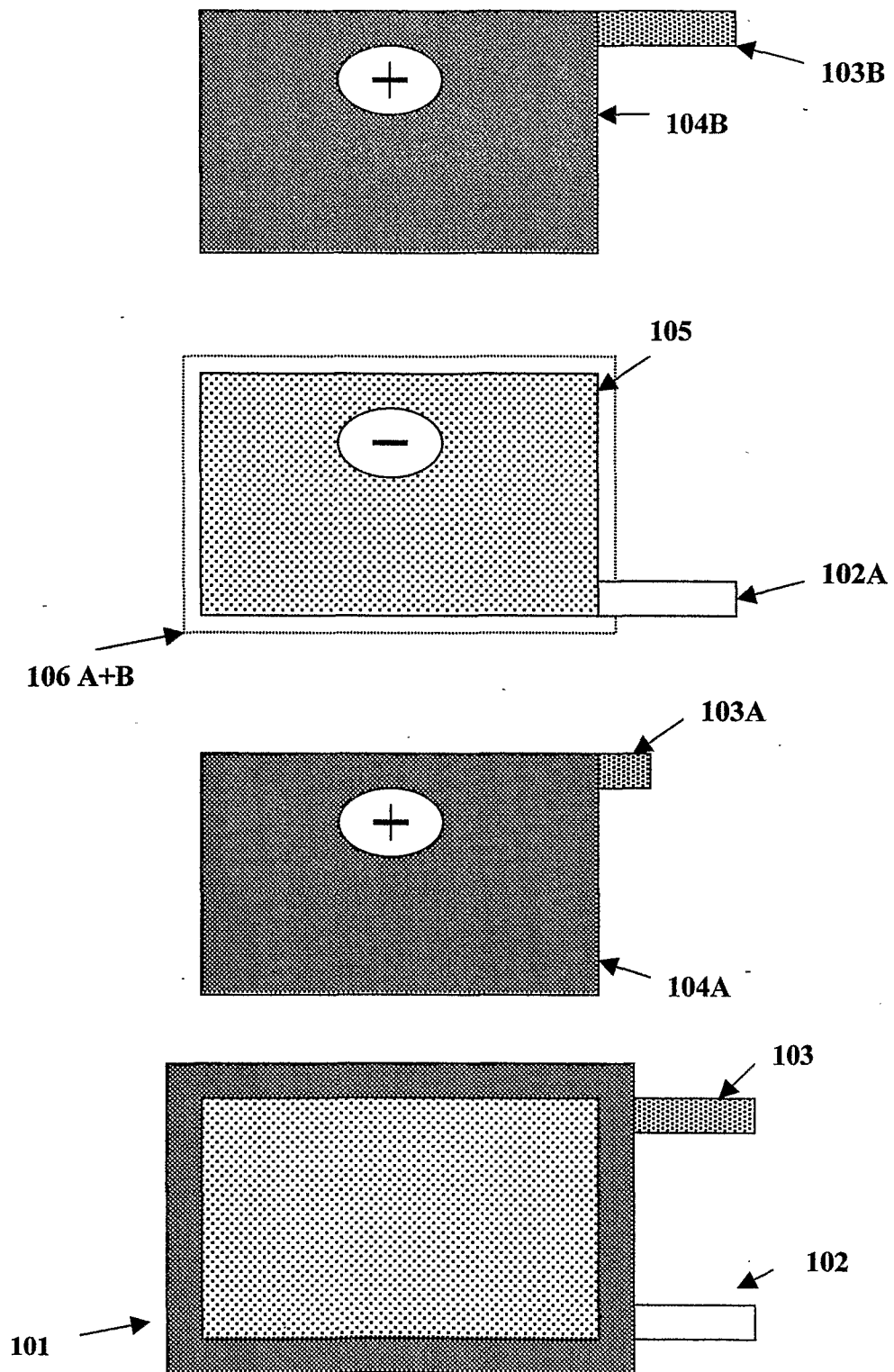


Figure 1

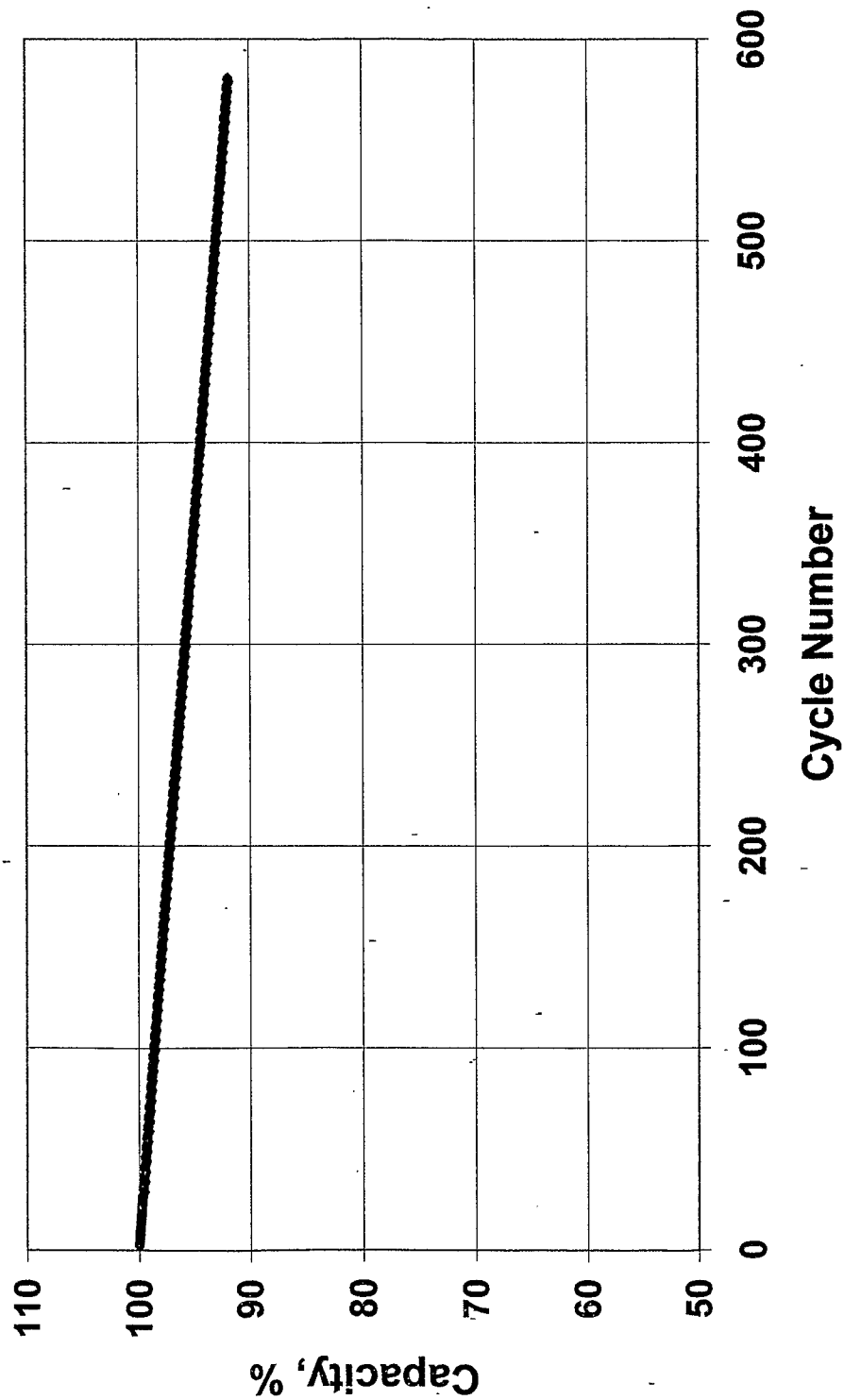


Figure 2

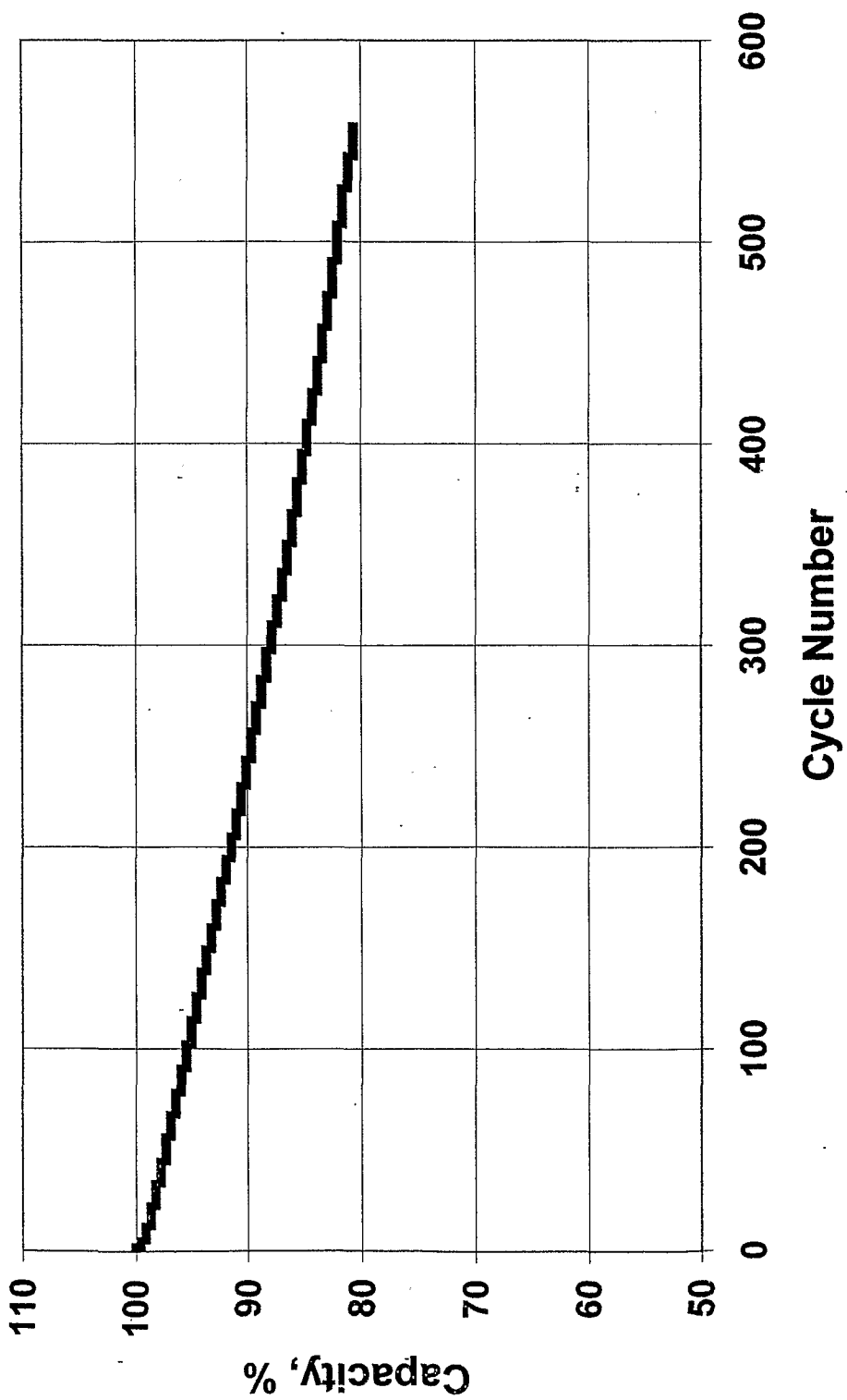


Figure 3

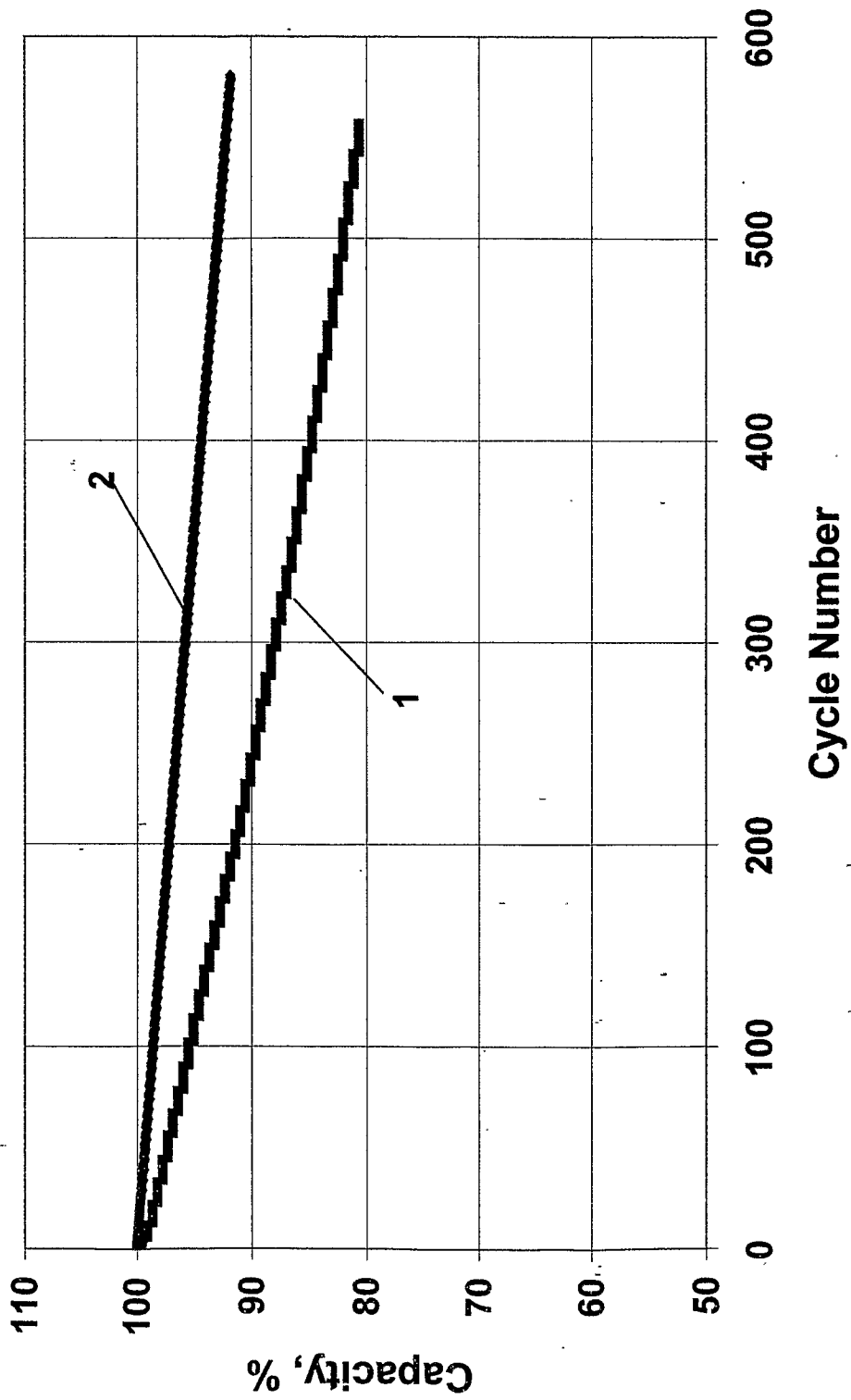


Figure 4

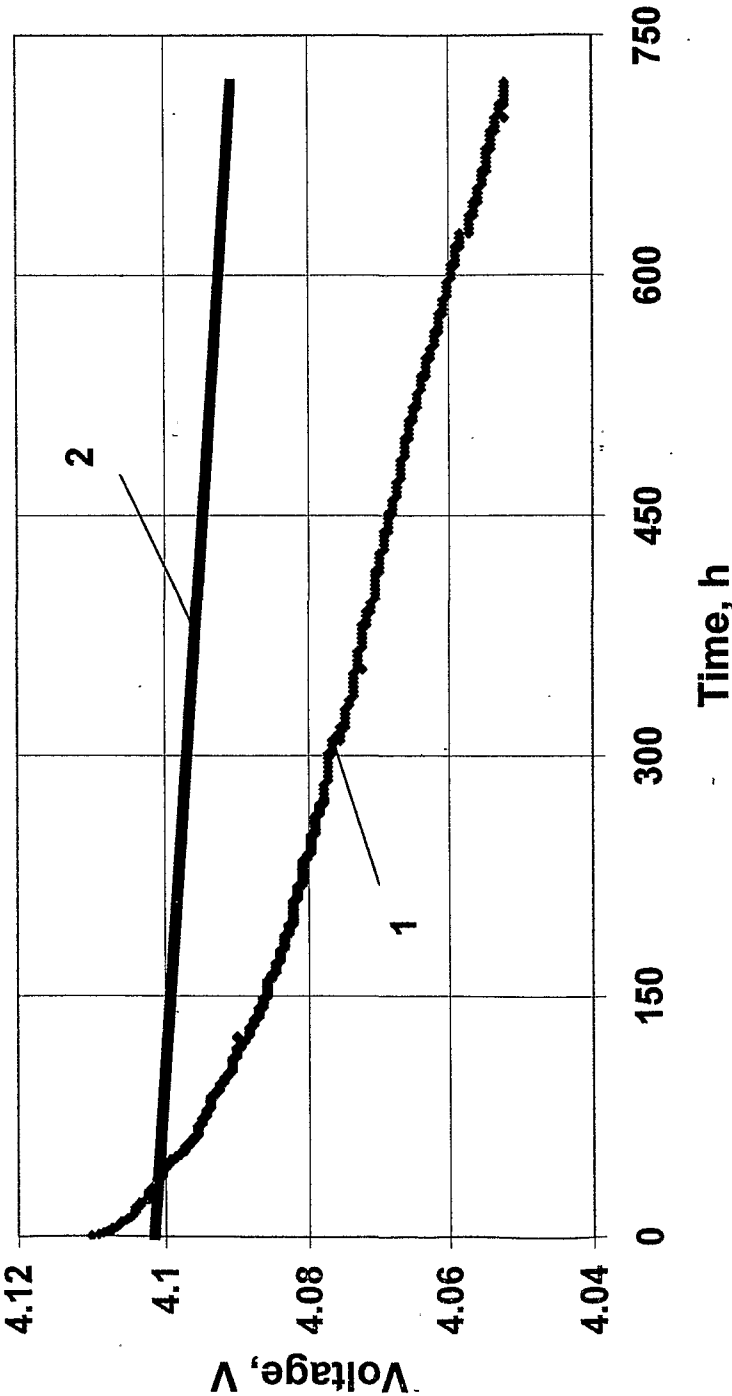


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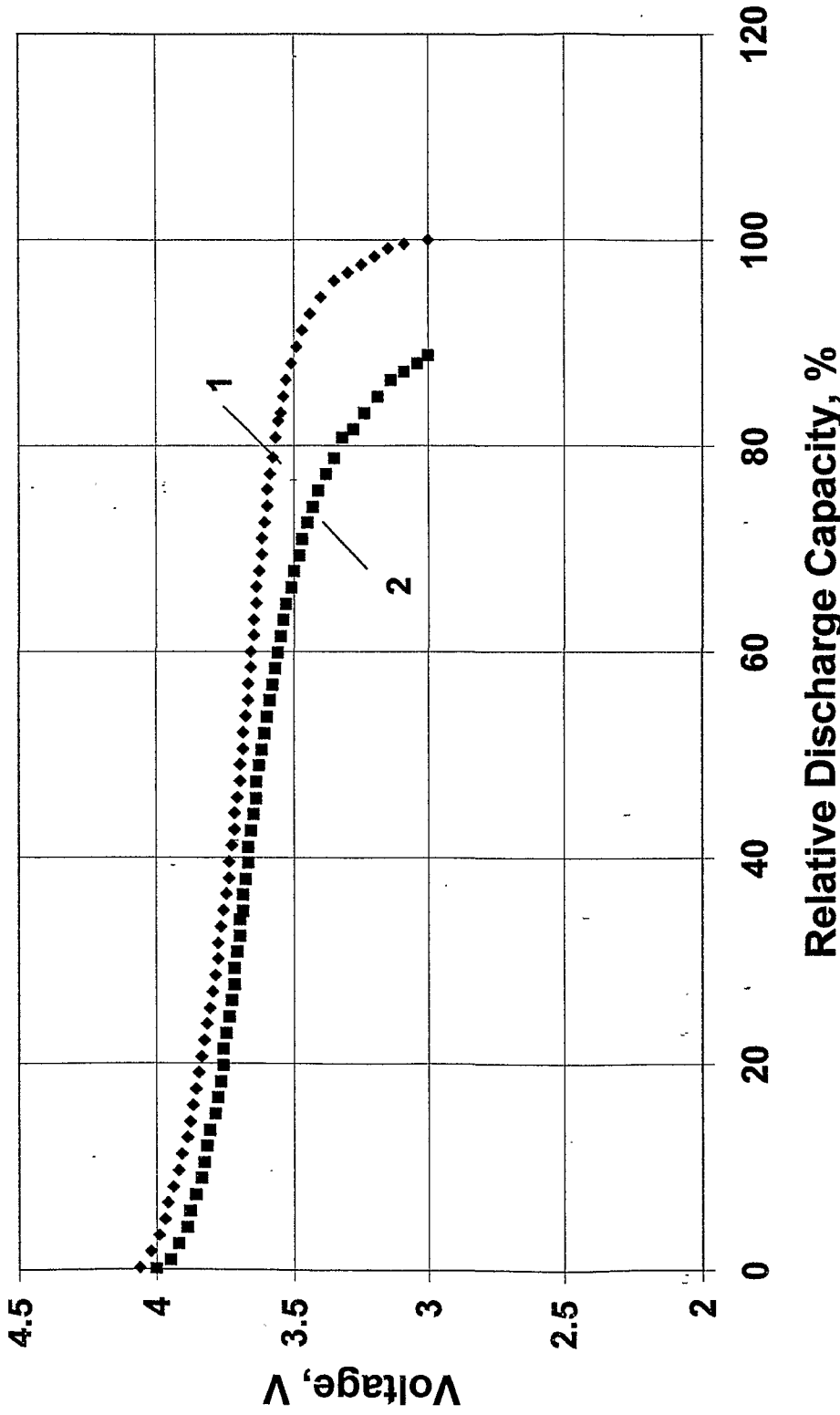


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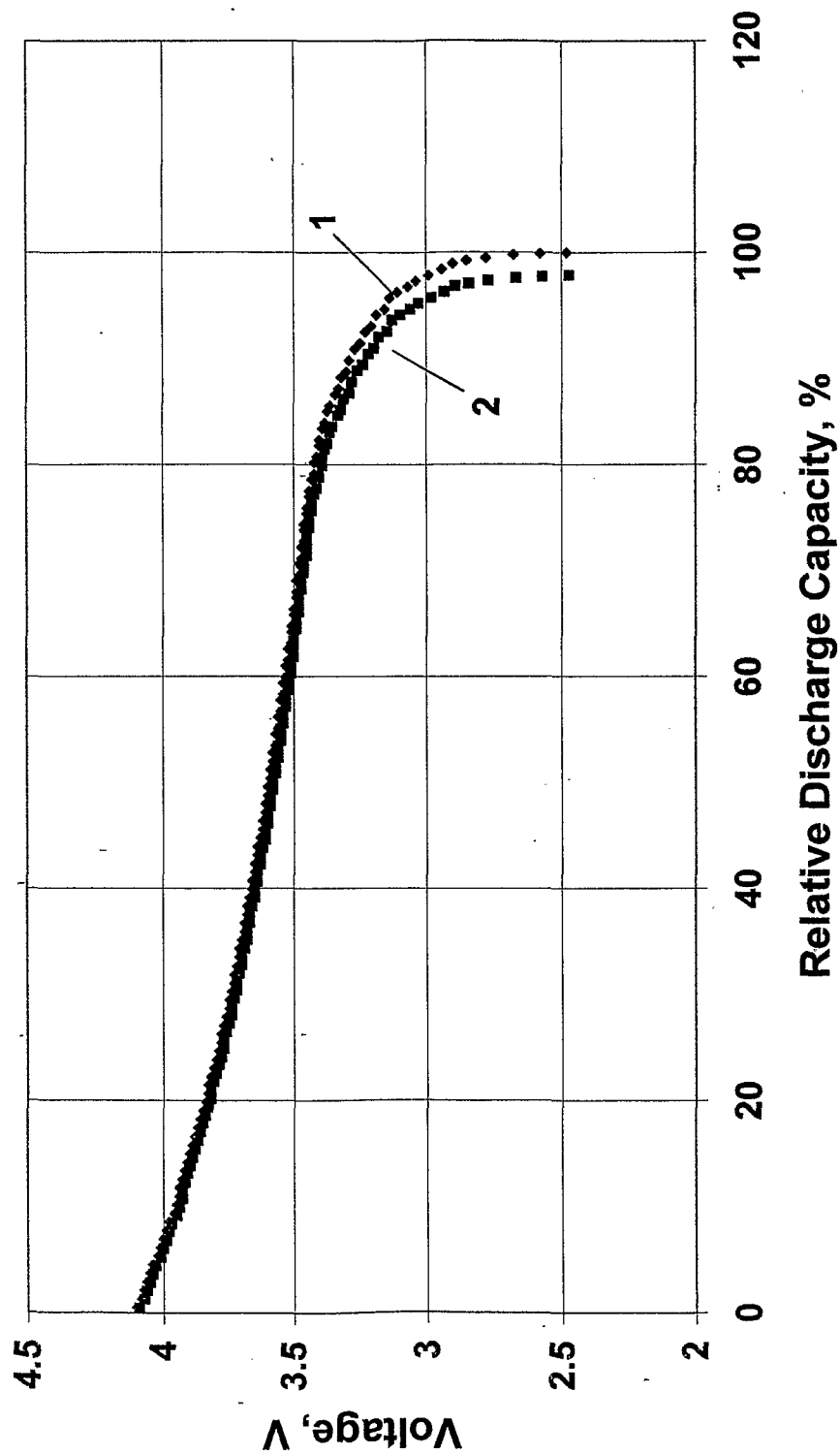


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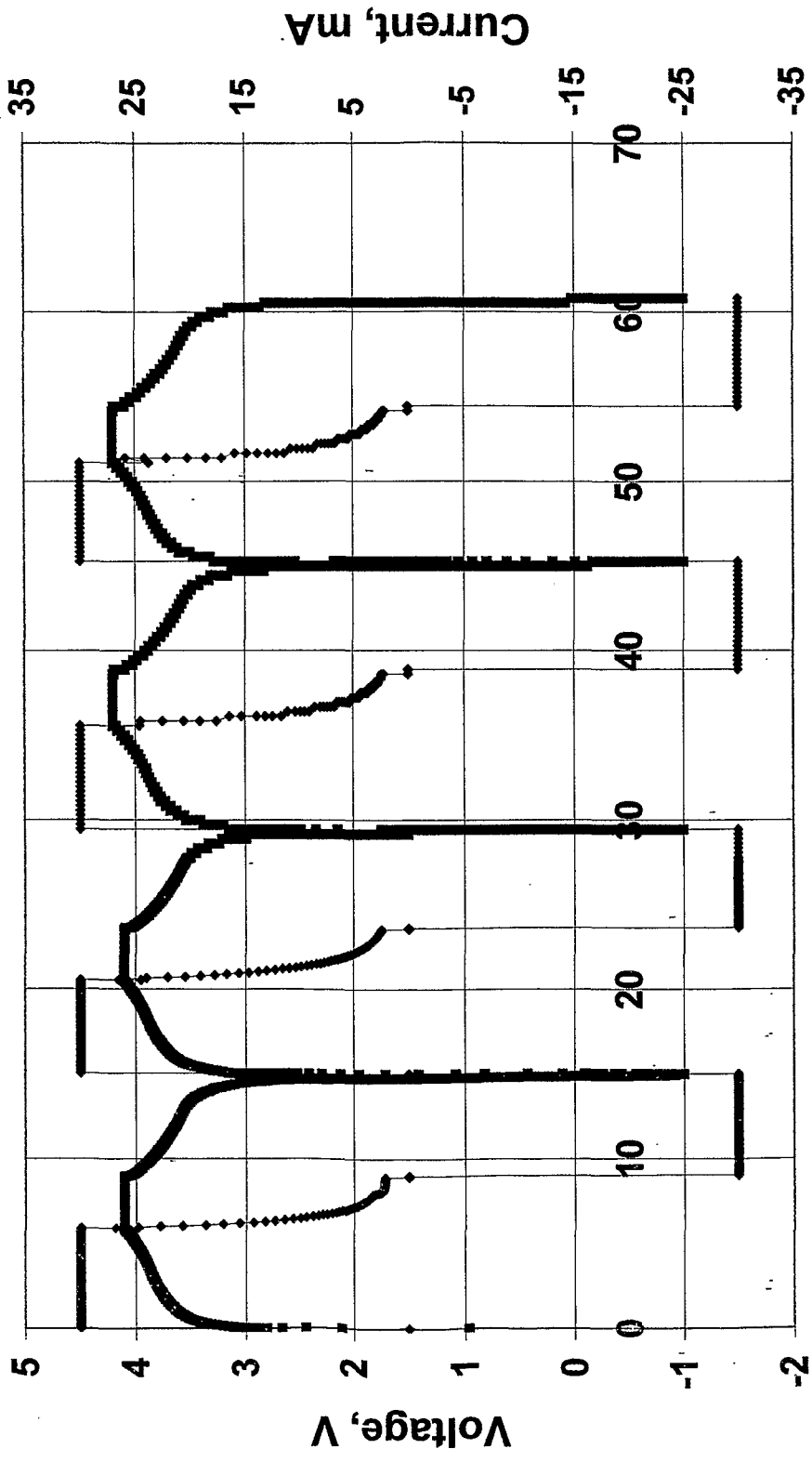


Figure 8

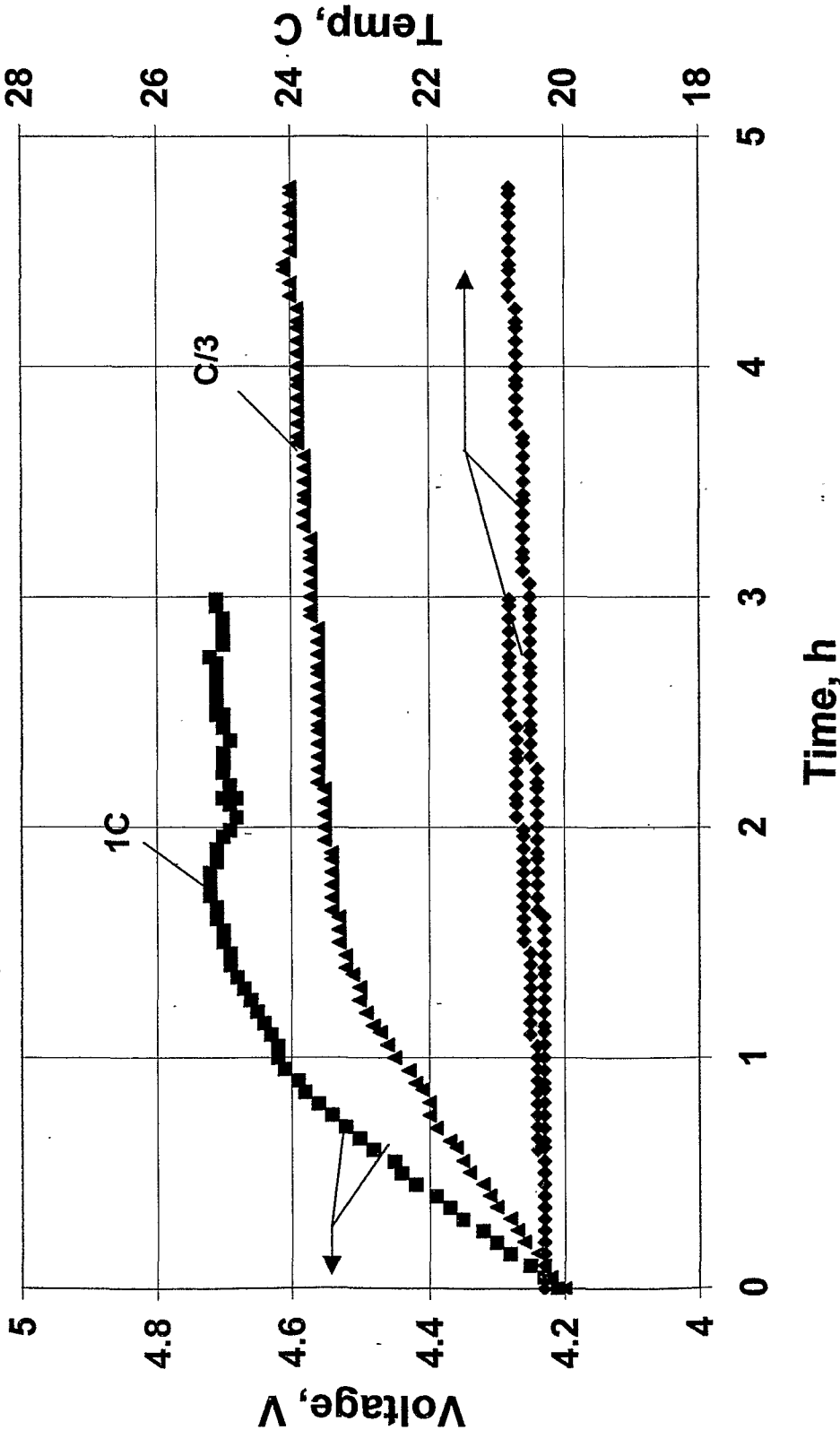
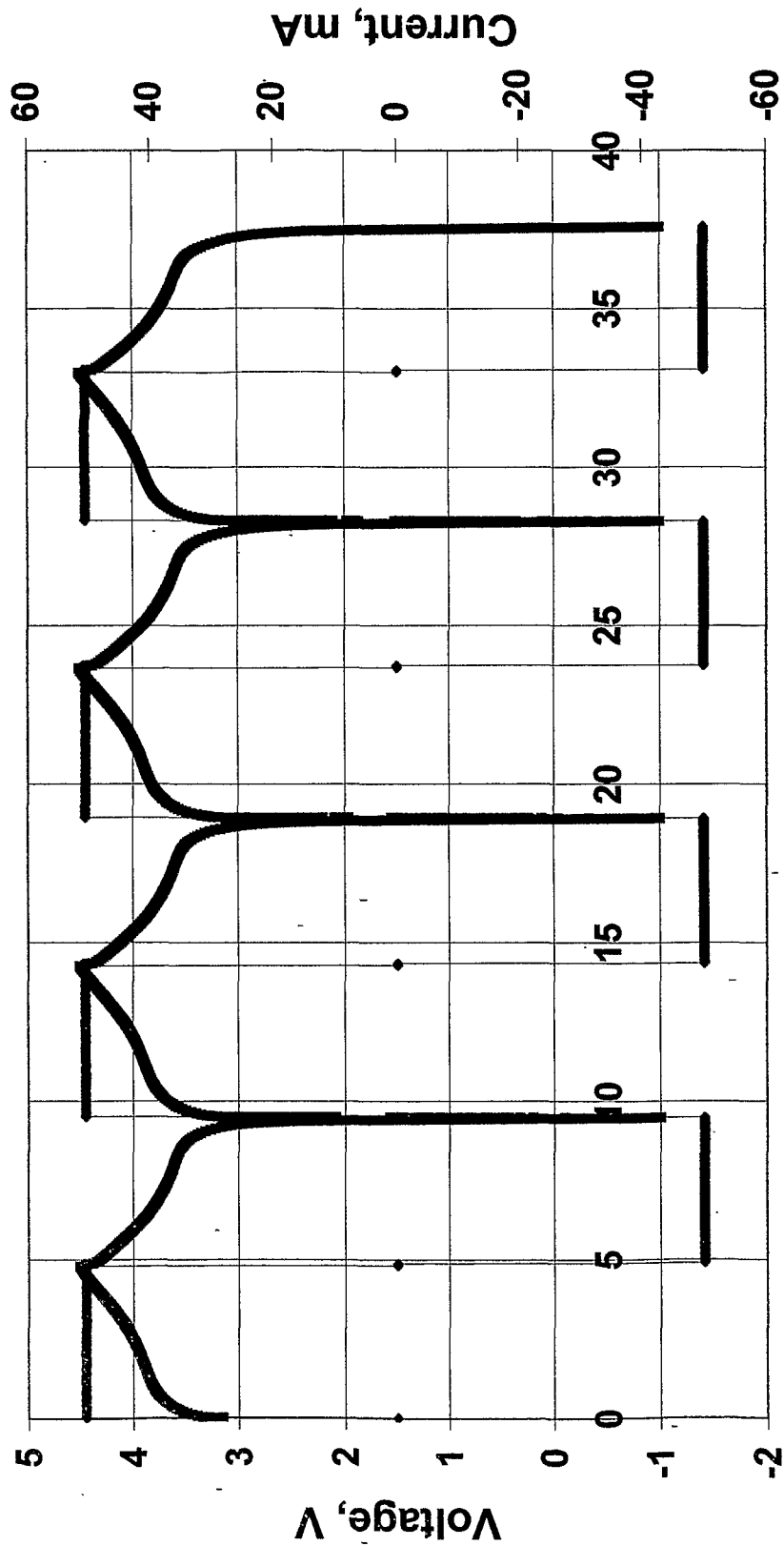


Figure 9



Time, h

Figure 10

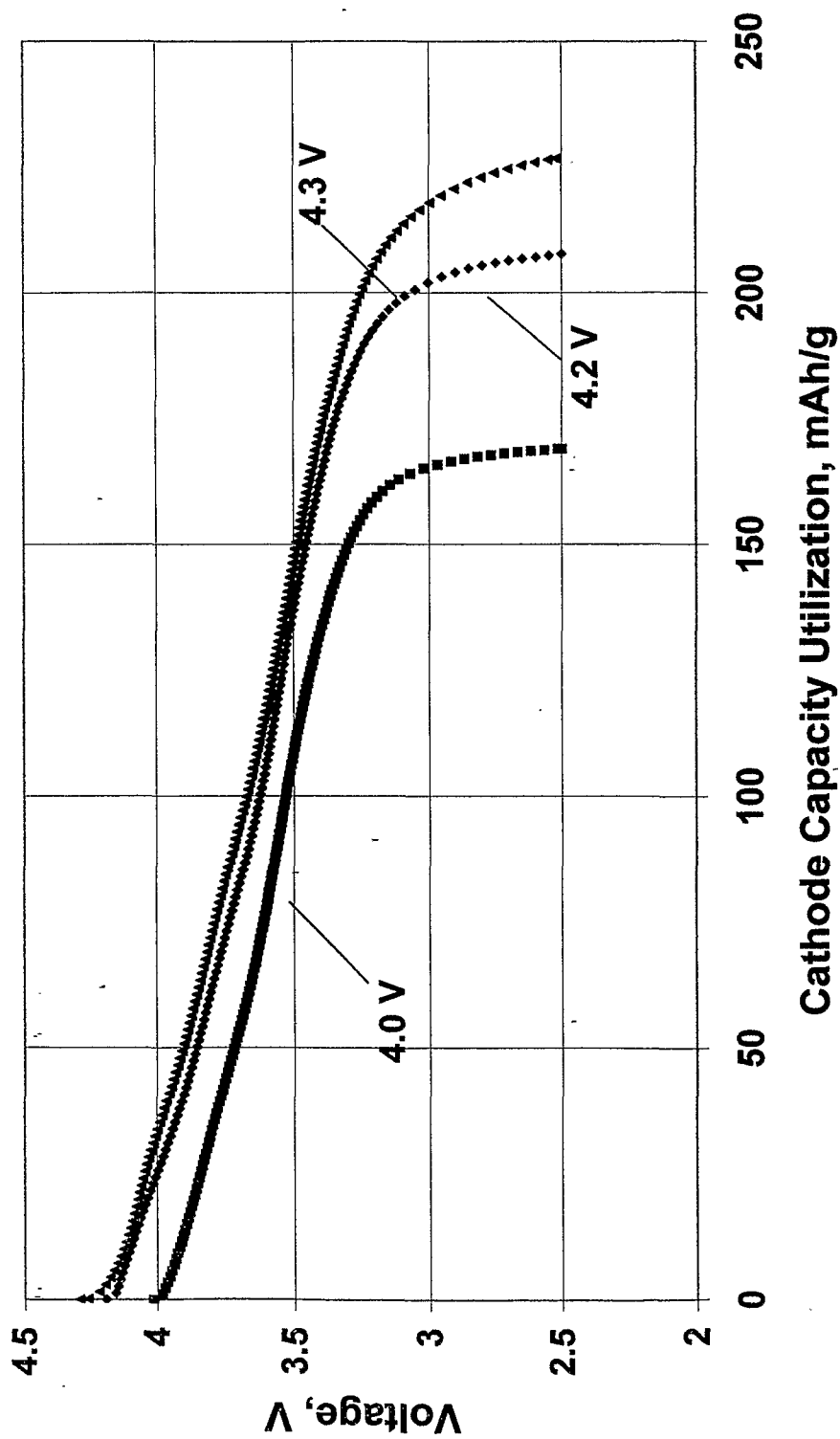


Figure 11

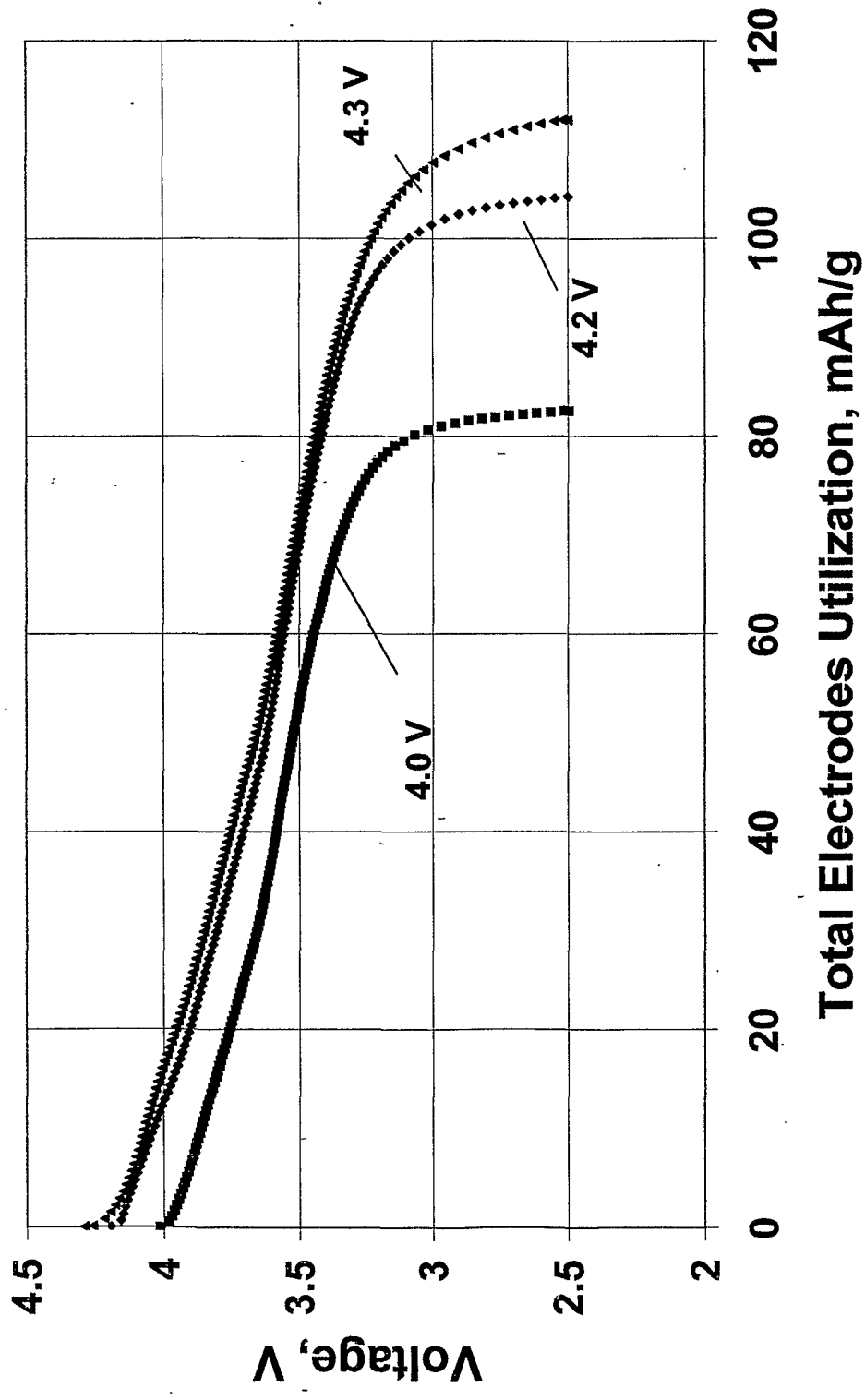
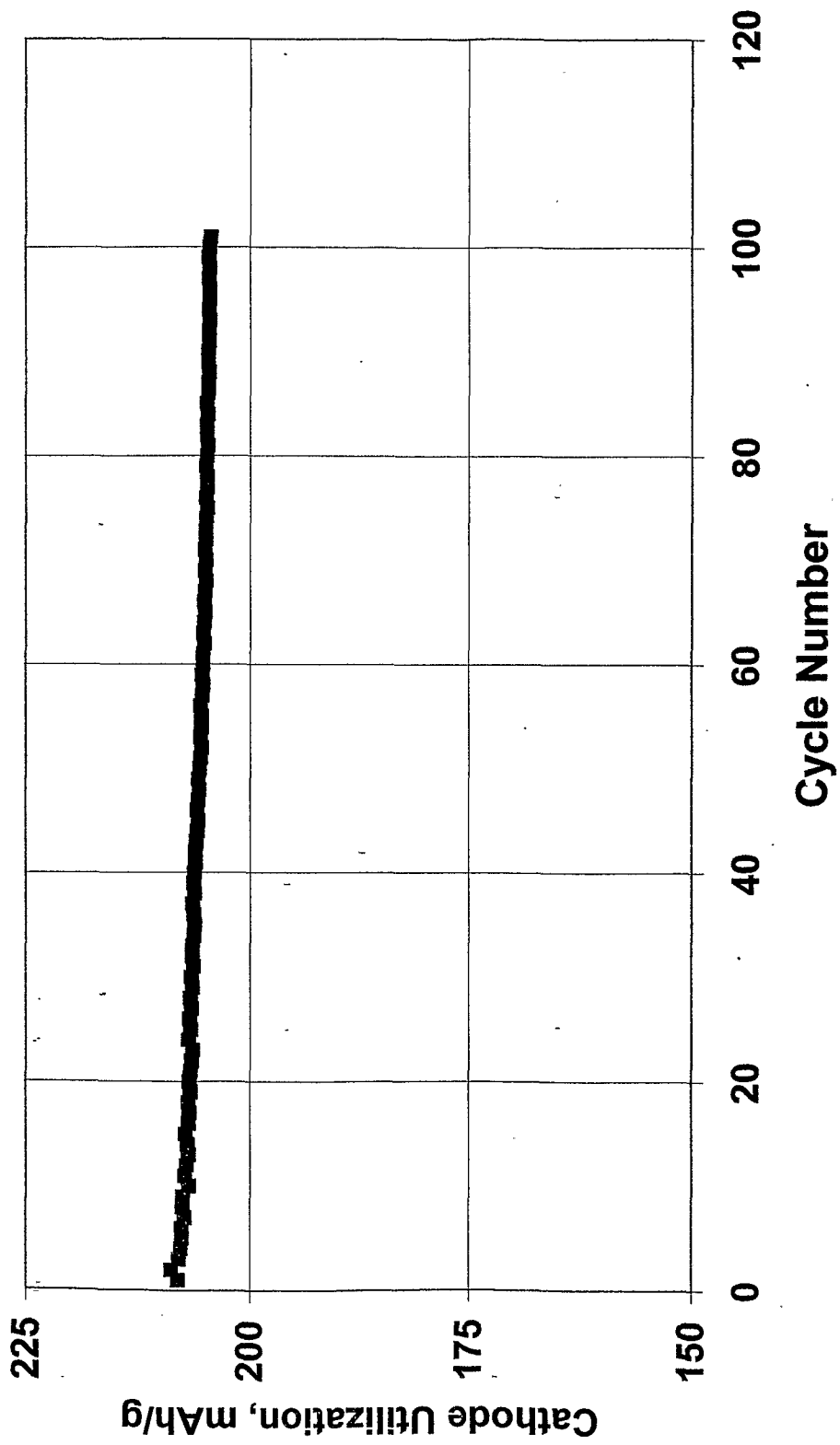


Figure 12

**Figure 13**

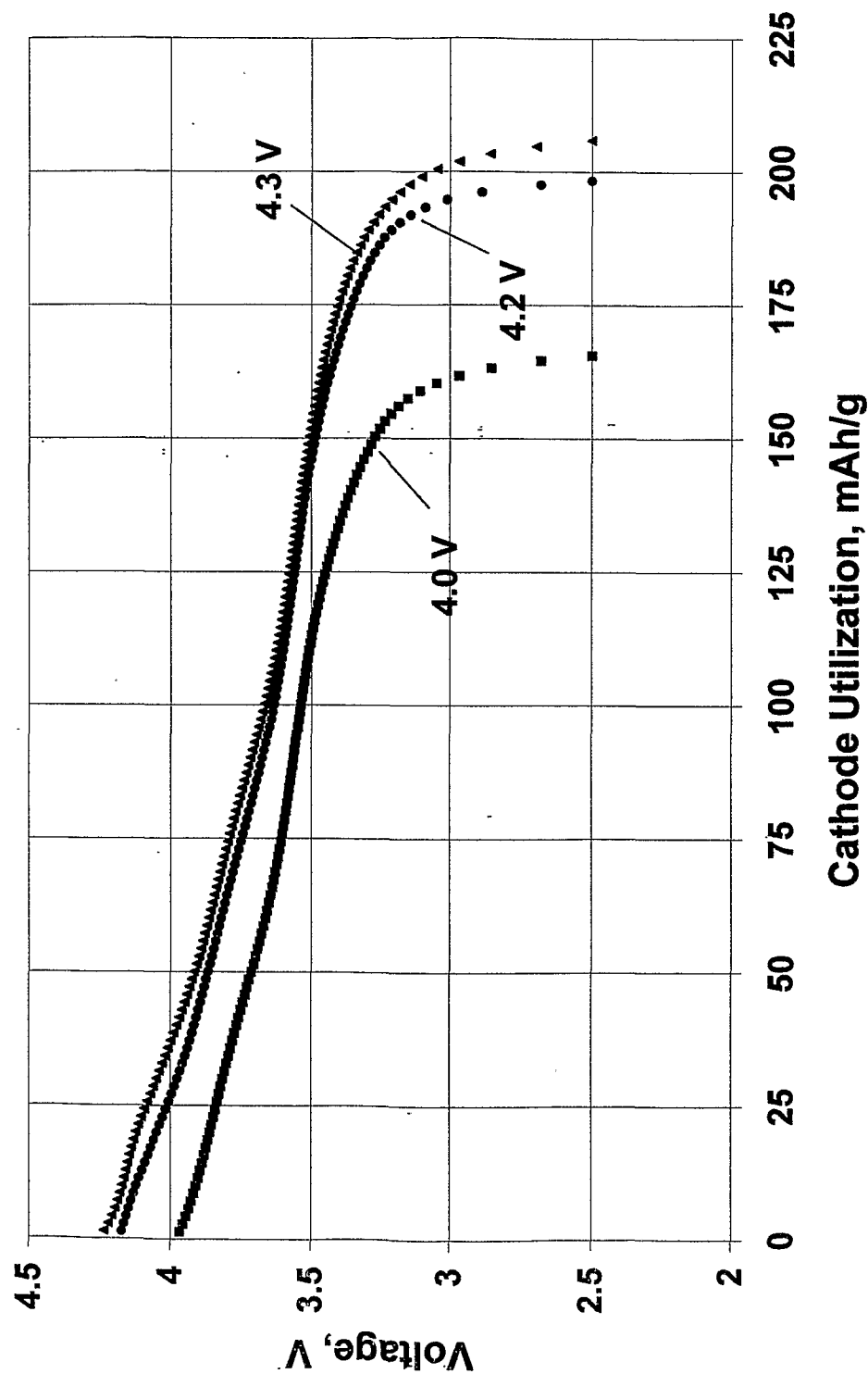


Figure 14

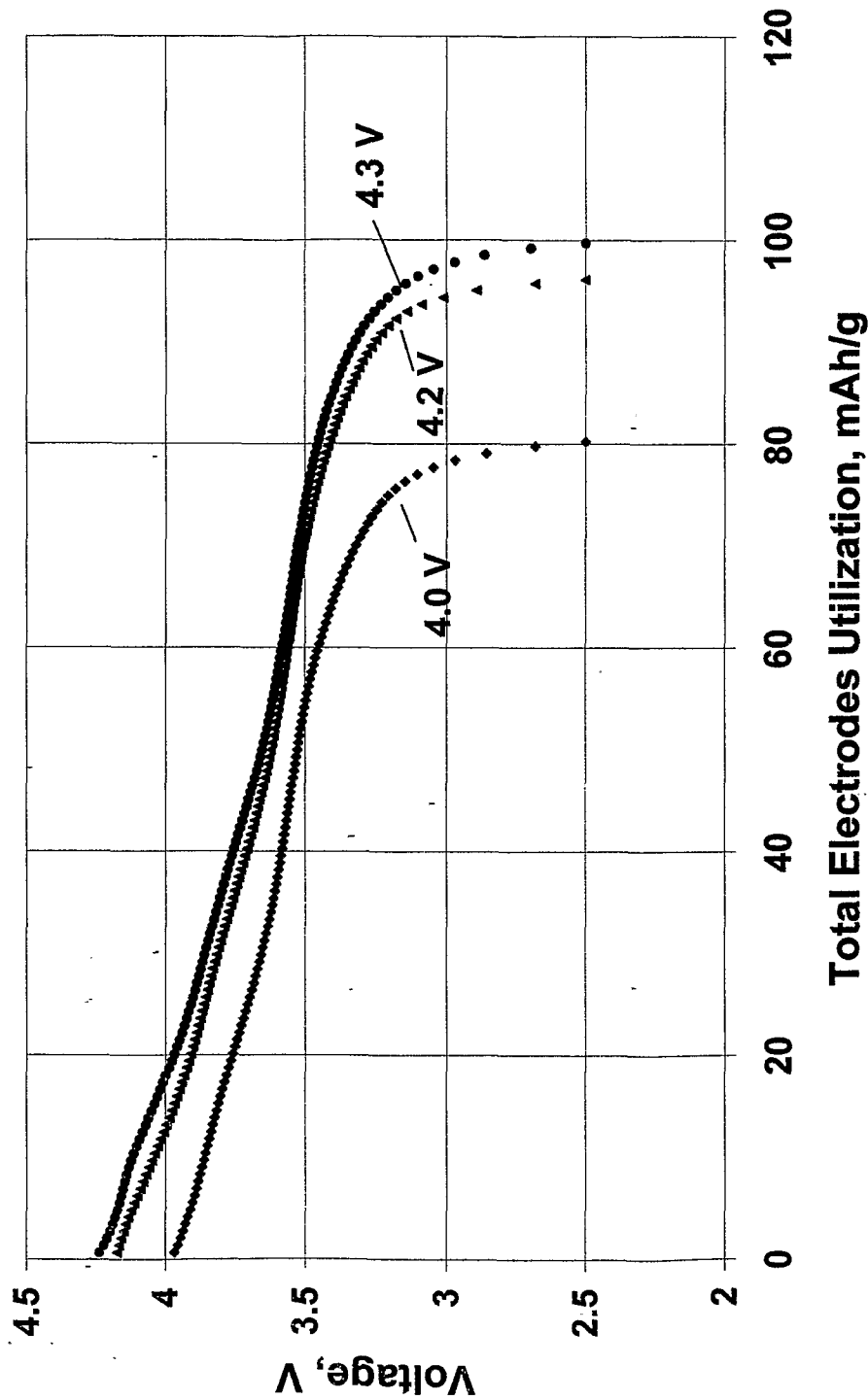


Figure 15

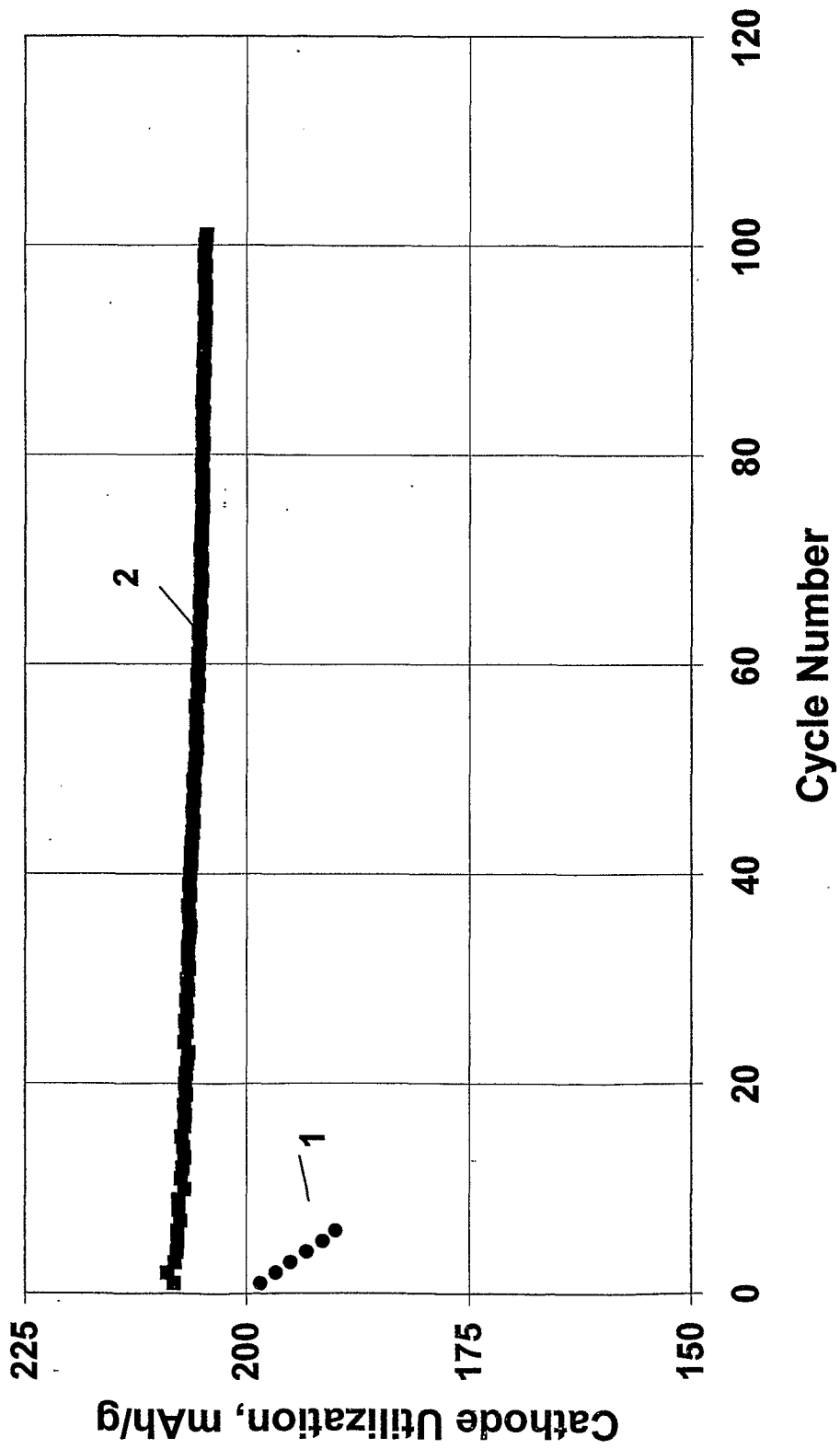


Figure 16