



(51) International Patent Classification:

H01M 4/04 (2006.01) *H01M 10/0525* (2010.01)
H01M 4/13 (2010.01) *H01M 10/0567* (2010.01)
H01M 4/139 (2010.01) *H01M 10/44* (2006.01)

(21) International Application Number:

PCT/EP2016/054111

(22) International Filing Date:

26 February 2016 (26.02.2016)

(25) Filing Language:

English

(26) Publication Language:

English

(71) Applicant: TOYOTA MOTOR EUROPE [BE/BE]; Avenue du Bourget 60, 1140 Brussels (BE).

(72) Inventor: SAKURAI, Takahiro; Boulevard du Souverain 292, 1160 Auderghem (BE).

(74) Agents: INTÈS, Didier et al.; Cabinet Beau De Lomenie, 158 Rue de l'Université, 75340 Paris Cedex 07 (FR).

(81) Designated States (unless otherwise indicated, for every kind of national protection available): AE, AG, AL, AM,

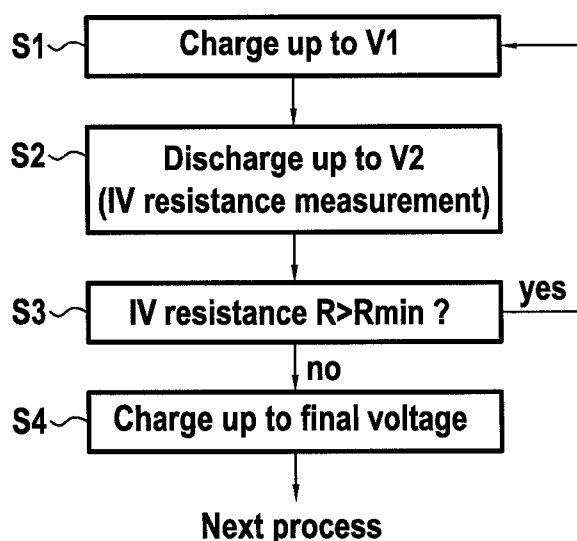
AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CZ, DE, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IR, IS, JP, KE, KG, KN, KP, KR, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, ME, MG, MK, MN, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, SM, ST, SV, SY, TH, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, ZA, ZM, ZW.

(84) Designated States (unless otherwise indicated, for every kind of regional protection available): ARIPO (BW, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SD, SL, ST, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, 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, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

Published:

— with international search report (Art. 21(3))

(54) Title: LITHIUM-ION BATTERY FORMATION PROCESS

**FIG.2**

(57) Abstract: A method of performing a formation process for a lithium-ion cell (10) comprising an anode (12), a cathode (16), an electrolyte (22) and a separator (20), the formation process including: - performing a charge-discharge cycle, wherein the cell is charged up to a first predetermined voltage level (V_1) and discharged until a second predetermined voltage level (V_2) being lower than the first predetermined voltage level, - determining a resistance R of the cell during discharge, wherein $R = (V_1 - V_2)/I$, I being the discharge current, and - repeating the charge-discharge cycle until the determined resistance of the cell reaches a predetermined lower resistance limit (R_{min}).

LITHIUM-ION BATTERY FORMATION PROCESS

FIELD OF THE DISCLOSURE

[0001] The present disclosure is related to rechargeable cells, in particular
5 to lithium ion batteries or cells, and more particularly to an improved method
for initially charging such batteries (formation process).

BACKGROUND OF THE DISCLOSURE

[0002] Lithium-ion batteries are part of a family of rechargeable battery
10 types in which lithium ions move from the negative electrode to the positive
electrode during discharge and from the positive electrode to the negative
electrode when charging.

[0003] There are various types of lithium-ion battery. The anode comprises
generally carbon and the cathode comprises a lithium compound. The anode
15 and the cathode are separated by a separator made from a porous polymer,
such as a micro-perforated plastic sheet, which allows ions to pass through.
The anode, cathode and separator are immersed in an electrolyte.

[0004] Lithium-ion batteries are classified according to the cathode material.

[0005] Once the lithium-ion battery is assembled, before the battery is
20 suitable to be used, the lithium-ion battery may be put through at least one
precisely controlled charge/discharge cycle to activate the working material.
This step is called the formation process. This formation process provides the
initial full charge of the battery.

[0006] During the formation process, a solid electrolyte interface (SEI) is
25 formed on the anode. The SEI formation is important for the lifetime of the
lithium-ion battery or cell.

[0007] Methods for initial charging, i.e., for the formation process, of a
lithium-ion battery have been proposed.

[0008] Typically, the battery is charged at a constant charge rate. The
30 charge rate is also expressed as a C-rate, which represents a charge or a
discharge rate equal to the capacity of a battery in one hour. It has been found
that the SEI is best formed at small C-rate, which means that the initial
charging is performed over an extended period of time. Indeed, fully charging a
battery at a C-rate equal to C/5 would take approximately five hours. The
35 battery is charged at a small C-rate up to the fully charged voltage of the

battery in order for the SEI to form on the carbon anode during the first charge and then the battery is held constant at the fully charged voltage until the current drops below a threshold. The battery is then left to rest for two hours and is discharged at a small C-rate to a pre-set voltage, i.e., the discharge cut-off voltage. This formation process may be cycled at least once.

[0009] In order to reduce the manufacturing time of lithium-ion batteries, so-called dynamic forming processes have been proposed. In such processes, the battery is charged at a small C-rate up to the end of SEI layer formation on the anode, corresponding to a threshold voltage value, and then, a large C-rate is used to charge the battery up to the fully charged voltage. For example US 2015/060290 discloses such a formation protocol which still involves at least charging the battery up to the fully charged voltage at least twice, and resting the cell for two hours between each charge/discharge of the cell, the total duration of the dynamic formation process being greater than forty hours. However, in US 2015/060290, the end of SEI layer formation voltage value on the anode being determined by a method using differences of temperature, the determination is an approximate.

[0010] Additives have also been added to the electrolyte to improve the formation of the SEI and therefore enhancing the anode stability.

[0011] A further charging method of a secondary battery is known from JP 2011-222358 (A). In the method a lithium ion secondary battery is initially charged during high temperature ageing. The SEI film formation is determined by an impedance variation and the charging method is changed when the SEI film formation is not completed. However, impedance measurements require further equipment like sensors, etc., and may lead therefore to higher costs.

SUMMARY OF THE DISCLOSURE

[0012] Currently, it remains desirable to reliably and efficiently perform the formation process while having a battery that will exhibit good properties over a large number of charge/discharge cycles.

[0013] Therefore, according to embodiments of the present disclosure, a method of performing a formation process for a rechargeable cell, in particular a lithium-ion cell having an anode, a cathode, an electrolyte and a separator is provided. The method including:

- performing a charge-discharge cycle, wherein the cell is charged up to a first predetermined voltage level V_1 and discharged until a second predetermined voltage level V_2 being lower than the first predetermined voltage level;

5 - determining a resistance R of the cell, wherein $R = (V_1 - V_2)/I$, I being the discharge current, and

- repeating the charge-discharge cycle until the determined resistance of the cell reaches a predetermined lower resistance limit.

[0014] By providing such a method, repeating the charge-discharge cycle
10 may improve the capacity retention, i.e. the lifetime of the cell can be increased.

[0015] The resistance is desirably determined during discharging, e.g. based on one or several measured values of the current during discharging. The current may namely be based on the constant current discharge. The current is
15 usually most stable during discharging. However, the resistance may also be calculated at the end of or after ending discharging.

[0016] The resistance may also be determined based on an averaged value of the current during discharging. This way of determination regularly provides the most exact resistance calculation. For example, the average may be based
20 on the values between 1s after starting discharging and 10s (end). The start point of discharging (0s) may be avoided because discharge current is usually unstable at this time.

[0017] The determination of the predetermined lower resistance limit may be performed prior to applying a formation process to a cell. For example, test
25 cells may be charged by applying different numbers of charge-discharge cycles to the test cells, respectively. The resulting resistance of the test cells may be measured subsequently. The SEI, which is formed by the application of the charge-discharge cycles, decreases the resistance. The resistance saturates with an increasing number of charge-discharge cycles indicating the completion
30 of the SEI formation process, e.g. after the third cycle. Hence, the lower resistance limit may be determined to correspond to this saturated value of the resistance or to a value at which the resistance starts to saturate. The latter value may be determined for example by adding a tolerance range, e.g. 1, 2 or 3%, to the actual saturated value.

[0018] Thus, the lower resistance limit may be used to initially charge cells having the same configuration and the same components as the cells used to determine the lower resistance limit. Due to a suitable choice of the lower resistance limit, the determination of the required number of charge-discharge cycles until completion of formation is accurate, as the resistance can be reliably determined, without the requiring expensive measurement equipment.

[0019] Accordingly, the charge-discharge cycles can be limited to a minimum number which is necessary to complete the formation process. This limitation allows reducing the formation process duration.

[0020] When the determined resistance of the cell reaches the predetermined lower resistance limit, the cell may be charged up to a predetermined final voltage level, which is higher than the first predetermined voltage level.

[0021] The cell may reach at the predetermined final voltage level a fully charged capacity.

[0022] Accordingly, once, the SEI formation is completed, the cell may be charged up to a fully charged capacity.

[0023] In the charge-discharge cycle the cell may be charged at a first predetermined charge rate and discharged at a second charge rate, the second charge rate being greater than the first charge rate.

[0024] Charging the cell up to the first predetermined voltage level, in particular with the first predetermined charge rate, allows for the formation of the SEI on the anode. This first predetermined rate allows for the formation of a SEI with good electrochemical properties while not extending too much the duration of the full formation process.

[0025] The second predetermined charge rate allows reducing the formation process duration, as the second predetermined rate is greater than the first predetermined rate.

[0026] Each of the charge/discharge rates may be any value, preferably below 5 C.

[0027] The first charge rate may be smaller than 2 C, preferably smaller than or equal to 1 C.

[0028] The second charge rate may be equal to or greater than 2 C, in particular equal to or greater than 3 C.

[0029] In addition, also the step of charging the cell up to the predetermined final voltage level may be done with a third predetermined charge rate being greater than the first predetermined charge rate. The third predetermined charge rate may be equal to or greater than 2 C, in particular
5 equal to or greater than 3 C.

[0030] The first predetermined voltage level may be below the voltage of a fully charged state of the cell, e.g. 4 V. In particular the first predetermined voltage level may be between 2 V and 3.5 V, more particularly 3.2 V.

[0031] The second predetermined voltage level may be greater than 0 V
10 and smaller than 3 V, in particular 2 V.

[0032] Such a value is preferable because SEI can be formed in the beginning of formation. The SEI piles up on the surface of the anode. Electro chemistry of charge-discharge occurs on the anode surface. Moreover additives make SEI on the surface and improve the electro chemistry like catalyst.
15 Additives make SEI below 3V mostly. From these facts, SEI to improve performance can be formed in the beginning of formation. SEI cannot grow forever. If it reaches a saturated thickness, the growth stops.

[0033] The predetermined final voltage level may be 4 V.

[0034] The second predetermined voltage level may be determined by,
20 when the cell has the first predetermined voltage level, discharging the cell in the charge-discharge cycle for a predetermined time interval, in particular between 5s and 30s, more particularly 10s.

[0035] In this way the second predetermined voltage level may be determined based on the discharge time. Such an embodiment only requires a
25 simple control method. This is possible because the second predetermined voltage level may have any level lower than that one of the first predetermined voltage level.

[0036] The predetermined lower resistance limit may be determined as being a value, at which the determined resistance becomes constant when
30 repeating the charge-discharge cycle.

[0037] In other words, the predetermined lower resistance limit may correspond to a saturation value of the determined resistance, when the number of charge-discharge cycles is increased.

[0038] Indeed, when the determined resistance does not decrease any more with increasing number of charge-discharge cycles, it may be concluded that the SEI formation on the anode is completed.

5 [0039] Before performing the charge-discharge cycle, an additive may be added to the electrolyte for improving a solid electrolyte interface build-up on the anode, in particular the additive being selected from an oxalate salt, the additive being more particularly a Lithium bis(oxalato)borate.

[0040] The additive may present a decomposition potential at a smaller voltage than the electrolyte.

10 [0041] These additives may improve the formation of the SEI on the anode and provide a SEI having better in-use characteristics than SEI formed from the electrolyte only.

[0042] It is intended that combinations of the above-described elements and those within the specification may be made, except where otherwise
15 contradictory.

[0043] It is to be understood that both the foregoing general description and the following detailed description are exemplary and explanatory only and are not restrictive of the disclosure, as claimed.

20 [0044] The accompanying drawings, which are incorporated in and constitute a part of this specification, illustrate embodiments of the disclosure and together with the description, and serve to explain the principles thereof.

BRIEF DESCRIPTION OF THE DRAWINGS

[0045] Fig. 1 shows a lithium ion cell;

25 [0046] Fig. 2 shows a flow chart illustrating an exemplary method according to embodiments of the present disclosure;

[0047] Fig. 3 shows an exemplary and schematic voltage – time diagram of the charge-discharge cycles applied to a cell.

DESCRIPTION OF THE EMBODIMENTS

30

[0048] Reference will now be made in detail to exemplary embodiments of the disclosure, examples of which are illustrated in the accompanying drawings. Wherever possible, the same reference numbers will be used throughout the drawings to refer to the same or like parts.

[0049] Fig. 1 shows a schematic representation of an exemplary lithium ion cell 10. The lithium ion cell 10 includes an anode 12 fixed on an anode current collector 14 and a cathode 16 fixed on a cathode current collector 18. The anode 12 and the cathode 16 are separated by a separator 20, the anode 12, the cathode 16 and the separator 20 being immersed in an electrolyte 22.

[0050] Typically, the anode 12 is made of a carbonaceous material and/or graphite. The anode current collector 14 may be made of copper. The cathode 16 may be made of an intercalated lithium compound, e.g. $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$. The cathode current collector 18 may be made of aluminum. The separator 20 may be made of a film comprising polyethylene.

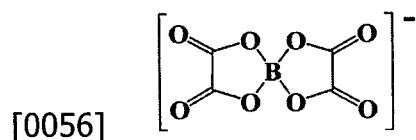
[0051] The electrolyte 22 may be a mixture of ethylene carbonate, dimethyl carbonate and ethyl methyl carbonate present in equal volume ratio. The electrolyte may also comprise LiPF_6 at 1 mol/L (mole/litre).

[0052] On the anode 12, a solid electrolyte interface (SEI) 24 is formed. The SEI 24 is formed during the formation process of the cell, i.e., during the initial charging of the cell.

[0053] Additive may be added to the electrolyte 22 to improve the formation of the SEI.

[0054] According to some embodiments, the additive provided in the electrolyte may be selected from an oxalate salt.

[0055] Examples of oxalate salts may include lithium salts of bis(oxalato)borate:



[0057] In particular, the additive may be Lithium bis(oxalato)borate, more in particular added at 5 wt% (weight percent) to the electrolyte 22.

[0058] Lithium ions present in the electrolyte 22 move from the anode 12 to the cathode 16 during discharge of the cell 10 and from the cathode 16 to the anode 12 when charging the cell 10.

[0059] Fig. 2 shows a flow chart illustrating an exemplary method according to embodiments of the present disclosure.

[0060] In step S1, the cell 10 is charged at a first predetermined rate C_1 up to a first predetermined voltage level V_1 , e.g. to 3.2 V.

[0061] For example, the first predetermined rate C_1 may be equal to 1 C. This first predetermined rate C_1 allows for the formation of a SEI 24 with good electrochemical properties while not extending too much the duration of the full formation process.

5 [0062] Then, in step S2 the cell 10 is discharged at a second predetermined rate C_2 until a second predetermined voltage level V_2 is reached, e.g. 2 V.

[0063] During or after the cell 10 is discharged in step S2 the resistance R of the cell is determined. For this purpose the formula $R = (V_1 - V_2)/I$ is used. I is the discharge current and may be measured during the discharging in step S2.

10 [0064] In step S3 it is determined, whether the resistance R is greater than a predetermined lower resistance limit $R_{\min}$. If this is the case, the method moves back to step S1, i.e. the cell is charged up again to the first predetermined voltage level V_1 . Accordingly, the charge-discharge cycle is repeated.

15 [0065] However, in case the resistance R is not greater but reaches the predetermined lower resistance limit $R_{\min}$ (i.e. is equal to or smaller than $R_{\min}$), the cell may be charged up to a predetermined final voltage level V_{fin} in step S4. V_{fin} may be higher than the first predetermined voltage level, e.g. 4 V. This voltage level may correspond to a fully charged state of the cell. Hence, when
20 this voltage level is reached, the cell is fully charged for the first time.

[0066] With each charging procedure in step S1 the formation of the SEI is further advanced. The formation of the SEI decreases the resistance until the SEI formation is completed. Then a saturation level of the resistance is reached. Therefore the resistance can be used as a threshold to determine, at what time
25 the formation of the SEI is completed. Based on the saturation level the predetermined lower resistance limit $R_{\min}$ (being this threshold) may be determined.

[0067] The predetermined lower resistance limit $R_{\min}$ may be determined as follows.

30 [0068] A given number of test cells 10 (i.e. samples) having the same components were charged with a different number of applied charge-discharge cycles (i.e. repeat number of charge-discharge cycles), respectively. Table 1 summarizes the resulting resistance of the cells.

[0069]

	Charge rate	V1	Discharge rate	V2	Repeat number	First IV Resistance mΩ	Second IV Resistance mΩ
Sample 1	1C	3.2V	3C	2V	0	301	312
Sample 2					1	297	309
Sample 3					2	293	297
Sample 4					3	293	297
Sample 5					4	292	297

Table1: formation condition

5 [0070] As it can be seen in this example, after the second repetition, i.e. when the charge-discharge cycle has been applied three times, the resistance starts to saturate, in this example at 293 mΩ. Accordingly, for this type of cell, a suitable value for the predetermined lower resistance limit $R_{\min}$ may be 293 mΩ.

10 [0071] Fig. 3 shows an exemplary and schematic voltage – time diagram of the charge-discharge cycles applied to a cell.

[0072] At $t=0$ the battery is charged, starting from 0 V. This charging procedure corresponds to step S1 in fig. 2. When the voltage reaches the first predetermined voltage level V_1 , e.g. 3.2 V, the cell is discharged again until the
 15 second predetermined voltage level V_2 is reached, e.g. to 2 V. This discharging procedure corresponds to step S2 in fig. 2. At the time of discharging or when the second predetermined voltage level V_2 is reached, the resistance may be determined.

[0073] Such a charge-discharge cycle may now be repeated several times,
 20 until the determined resistance is equal to or lower than the predetermined lower resistance limit.

[0074] Thus, it is demonstrated that accurate determination of the required number of charge-discharge cycles based on the predetermined lower resistance limit may produce cells with good capacity retention and relatively
 25 short duration of formation process.

[0075] Throughout the description, including the claims, the term "comprising a" should be understood as being synonymous with "comprising at least one" unless otherwise stated. In addition, any range set forth in the description, including the claims should be understood as including its end
 30 value(s) unless otherwise stated. Specific values for described elements should be understood to be within accepted manufacturing or industry tolerances

known to one of skill in the art, and any use of the terms "substantially" and/or "approximately" and/or "generally" should be understood to mean falling within such accepted tolerances.

5 [0076] Although the present disclosure herein has been described with reference to particular embodiments, it is to be understood that these embodiments are merely illustrative of the principles and applications of the present disclosure.

10 [0077] It is intended that the specification and examples be considered as exemplary only, with a true scope of the disclosure being indicated by the following claims.

[0078] The method is described in terms of a single cell. However, it may be easily adapted for batteries having multiple cells. Moreover it may also refer to other cell types than lithium-ion cells.

CLAIMS

1. A method of performing a formation process for a rechargeable cell (10) comprising an anode (12), a cathode (16), an electrolyte (22) and a separator (20), the formation process comprising:
- performing a charge-discharge cycle, wherein the cell is charged up to a first predetermined voltage level (V_1) and discharged until a second predetermined voltage level (V_2) lower than the first predetermined voltage level,
 - determining a resistance R of the cell during discharge, wherein $R = (V_1 - V_2)/I$, I being the discharge current, and
 - repeating the charge-discharge cycle until the determined resistance of the cell reaches a predetermined lower resistance limit ($R_{\min}$).
2. The method according to claim 1, further comprising the step of: when the determined resistance of the cell reaches the predetermined lower resistance limit, charging the cell up to a predetermined final voltage level (V_{fin}), which is higher than the first predetermined voltage level.
3. The method according to claim 1 or 2, wherein the cell reaches at the predetermined final voltage level (V_{fin}) a fully charged capacity.
4. The method according to any of claims 1 to 3, wherein in the charge-discharge cycle the cell is charged at a first predetermined charge rate (C_1) and discharged at a second charge rate (C_2), the second charge rate (C_2) being greater than the first charge rate (C_1).
5. The method according to any of claims 1 to 4, wherein the first charge rate (C_1) is smaller than 2 C, preferably smaller than or equal to 1 C.
6. The method according to any of claims 1 to 5, wherein the second charge rate (C_2) is equal to or greater than 2 C, in particular equal to or greater than 3 C.
7. The method according to any of claims 1 to 6, wherein

the first predetermined voltage level (V_1) is below 4 V, in particular between 2 V and 3.5 V, more particularly 3.2 V, and/or

the second predetermined voltage level (V_2) is greater than 0 V and smaller than 3 V, in particular 2 V, and/or

5 the predetermined final voltage level (V_{fin}) is 4 V.

8. The method according to any of claims 1 to 7, wherein the second predetermined voltage level (V_2) is determined by, when the cell has the first predetermined voltage level (V_1), discharging the cell in the charge-discharge
10 cycle for a predetermined time interval, in particular between 5s and 30s, more particularly 10s.

9. The method according to any of claims 1 to 8, wherein the predetermined lower resistance limit (R_{min}) is determined as a value, at which
15 the determined resistance becomes constant when repeating the charge-discharge cycle.

10. The method according to any of claims 1 to 9, wherein before performing the charge-discharge cycle, an additive is added to the electrolyte
20 (22) for improving a solid electrolyte interface (24) build-up on the anode (12), in particular the additive being selected from an oxalate salt, the additive being more particularly a Lithium bis(oxalato)borate.

1/1

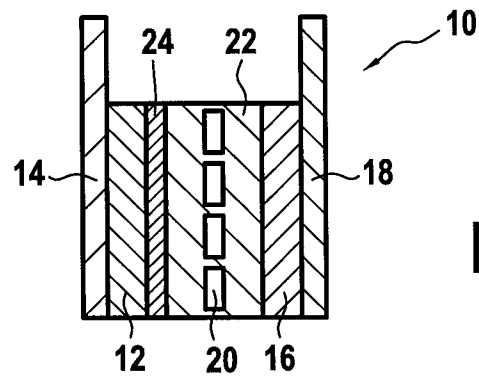


FIG.1

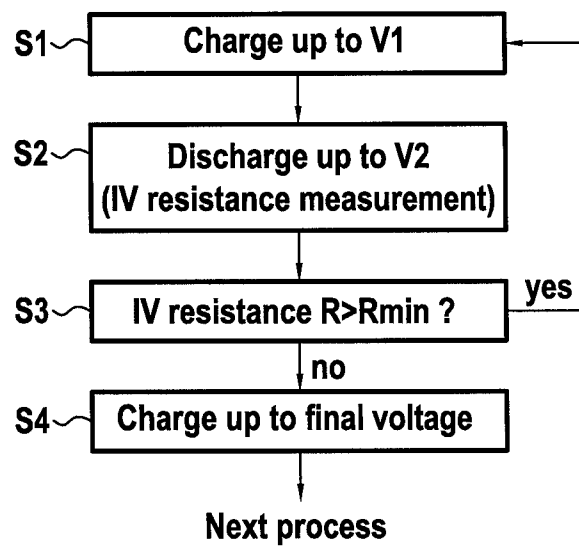


FIG.2

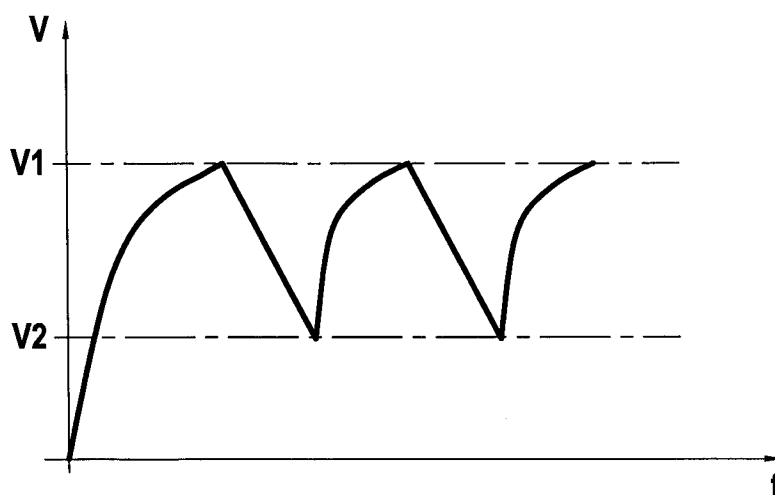


FIG.3

INTERNATIONAL SEARCH REPORT

International application No
PCT/EP2016/054111

A. CLASSIFICATION OF SUBJECT MATTER INV. H01M4/04 H01M4/13 H01M4/139 H01M10/0525 H01M10/0567 ADD. H01M10/44		
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		
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 practicable, search terms used) EPO-Internal, WPI Data		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 336 381 A2 (HABRA ELEKTRONIK [DE]) 11 October 1989 (1989-10-11) column 9, line 10 - column 10, line 10	1,7-9
Y	-----	2,3,5,10
A	US 2011/236751 A1 (AMIRUDDIN SHABAB [US] ET AL) 29 September 2011 (2011-09-29) paragraphs [0049], [0099], [0112]; figure 8	4,6
Y	-----	2,3,5,10
A	US 2014/329141 A1 (ONIZUKA HIROSHI [JP] ET AL) 6 November 2014 (2014-11-06) paragraphs [0040] - [0043]	1,4,6-9
A	-----	1-10
☐ Further documents are listed in the continuation of Box C. ☒ See patent family annex.		
* Special categories of cited documents :		
"A" document defining the general state of the art which is not considered to be of particular relevance "E" earlier application or patent but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed	"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "&" document member of the same patent family	
Date of the actual completion of the international search 31 May 2016	Date of mailing of the international search report 06/06/2016	
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Authorized officer Hofer, Astrid	

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2016/054111

Patent document cited in search report		Publication date		Patent family member(s)		Publication date
EP 0336381	A2	11-10-1989	BR	8901579 A		21-11-1989
			CN	1036865 A		01-11-1989
			DD	283695 A5		17-10-1990
			DE	3811371 A1		19-10-1989
			EP	0336381 A2		11-10-1989
			JP	H0237674 A		07-02-1990
			TR	24057 A		21-02-1991
			US	4947124 A		07-08-1990
			ZA	8902442 A		27-12-1989

US 2011236751	A1	29-09-2011	CN	102859778 A		02-01-2013
			EP	2553753 A2		06-02-2013
			JP	2013524413 A		17-06-2013
			KR	20130060180 A		07-06-2013
			TW	201145651 A		16-12-2011
			US	2011236751 A1		29-09-2011
			WO	2011119386 A2		29-09-2011

US 2014329141	A1	06-11-2014	CN	103959545 A		30-07-2014
			DE	112012004863 T5		28-08-2014
			JP	5754358 B2		29-07-2015
			JP	2013110022 A		06-06-2013
			KR	20140098150 A		07-08-2014
			KR	20160035100 A		30-03-2016
			US	2014329141 A1		06-11-2014
			WO	2013076893 A1		30-05-2013
