

TRADITIONAL FLOAT CHARGES: ARE THEY SUITED TO STATIONARY ANTIMONY-FREE LEAD-ACID BATTERIES?

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ABSTRACT

The self-discharge phenomenon caused by side reactions such as corrosion, water decomposition and recombination is unavoidable in lead acid batteries. Self-discharge rates depend on several factors such as the state of charge, the temperature, grid alloys, etc. These rates are not equal between negative and positive electrodes and are independent of each other. In lead-acid batteries with antimony, the self-discharge rate of the negative electrode is several times stronger than the self-discharge rate of the positive electrode, depending mainly on the battery age and the antimony content in the alloy of the positive grid. In order to compensate for the self-discharge of lead-acid batteries, a current whose value exceeds the self-discharge of the negative electrode has to be supplied. This is traditionally achieved by applying a constant voltage whose value is slightly higher than the battery open circuit voltage. This is called “float charge”. It has to be kept in mind that the float polarization increases gas evolution rates at both electrodes. This float polarization voltage is adjusted to ensure that the battery is not under-charged. At typical float polarization voltages of the order of 120 mV/cell, batteries are permanently overcharged. The float current is then about 10 stronger than the self-discharge current at open circuit.

In antimony-free lead-acid batteries, e.g. VRLA batteries, the self-discharge rate of the negative electrode is largely reduced and is about a half of the self-discharge rate of the positive electrode. The question is: are traditional float charges really suited to antimony-free lead-acid batteries? Experimental results say no. In this paper, a new method of maintaining the charge of antimony-free lead-acid batteries using low currents and periodic charges is presented. This is called “Low-current” method. Low currents are about 5 to 10 times smaller than traditional float currents. This method reduces overcharge, thus decreasing water loss and keeping the battery voltage in a minimum corrosion zone.

Laboratory experiments are focused on studying rates of self-discharge at open circuit and state of charge evolutions of antimony free lead-acid batteries in three different cases: float charge, low-current charge and open circuit. The new Low-current method, associated to an improved storage architecture system is now used in several UPS systems at Electricite de France (EDF). This association is intended to provide long life span and high reliability to VRLA batteries.

Keywords: *lead-acid batteries, VRLA batteries, self-discharge, float charge, intermittent charge, corrosion.*

INTRODUCTION

Valve regulated lead acid batteries - VRLA batteries - have been developed and used for about 30 years in standby applications. They have shown several advantages compared to flooded lead acid batteries: spill-proof, reduced weight, free from excessive gas evolution or acid spillage, reduced maintenance and reduced cost. However, limitations have been also observed concerning system reliability and battery service life.

Several reliability prediction methods have been used such as complete discharge test [1], open-circuit voltage measurement [2], conductance testing [3], internal resistance and impedance measurements [1, 4-8]. Among these, the complete discharge test is well known as the most reliable one but it requires service interruption. This has driven EDF R&D to develop the “Stationary Multibat” system, which consists of a new design of the electrochemical storage and an adapted electronic battery management system [9]. This system, combining redundancy and automated periodical capacity measurements, increases reliability and allows a real time monitoring of the battery state of health. Redundancy not only ensures the continuity of service in the case of a cell failure, but also enables complete discharges to be periodically performed without service interruption.

Standby VRLA batteries maintained under a constant float voltage to compensate self-discharge encounter the problem of short service life, e.g. about 3-4 years compared to the so-called 20-year design and to the 20-year lifetime of conventional lead acid batteries in similar conditions [10]. Indeed, VRLA batteries under float charges are permanently overcharged and different failure modes have been observed, such as corrosion of positive grid alloys, electrolyte dry-out and thermal runaway [11-17].

In order to reduce overcharge, intermittent charges (open-circuit and periodical charges) have been used instead of float charges and have given positive results in reducing thermal runaway and increasing the battery life span [18-21]. Nevertheless, corrosion of the positive grid is not at minimum during open-circuit periods of intermittent charges. Indeed, the literature shows that there is a potential zone where corrosion is minimum [11, 14, 22-26]. This minimum would be at positive polarizations (ρ^+) between zero (at open-circuit) and polarizations at float charges ($\rho^+ \approx 100$ mV). According to Brecht et al. [26] this minimum zone is situated in the range of [30;70] mV. Taking into account this minimum corrosion zone, a new method for maintaining the charge of standby lead acid batteries, called “Low-current” method, has been developed at EDF [27-29]. This method is derived from the intermittent charge: open-circuit periods are replaced by constant low current periods. These constant currents, whose values are smaller than traditional float currents, are intended to compensate, at least partially, self-discharge currents, and preferably place the battery in the minimum corrosion zone. Expected results are: (i) a slow down of the corrosion rate of positive grids, (ii) a reduction of the need of periodical charges.

In this paper, a management system for standby VRLA batteries improving both reliability and service life is presented, associating the Stationary Multibat system with the Low-current method.

EXPERIMENTAL

Laboratory experiments aim to evaluate the state of charge of batteries subjected (1) to open circuit, (2) to float charges and (3) to polarizations whose values are intermediate between zero and float polarizations. For the third case with intermediate charge conditions, the choice of piloting the current instead of the voltage relates to the fact that maintaining a voltage with a value very close to the battery open voltage is difficult and can accidentally discharge the battery [18]. Moreover, the scattering phenomenon when a voltage is applied to the battery consisting of cells in series configuration is unavoidable. Monitoring the voltage share for each cell is difficult and expensive while applying a current ensures that every cell receives a given current value. Several current values below float currents have been tested. These currents are called low currents.

The methods used to evaluate the self-discharge rate and the state of charge of lead-acid batteries were:

- lead sulfate content measurement in active materials by a chemical titration
- capacity measurement by the discharge test.

Batteries used for lead sulfate measurements were antimony-free vented SLI batteries, Exide L01033C24A, 40 Ah-12V. Four batteries were subjected first to open circuit during 231 days, then to low currents of 25, 50, 100 and 200 μ A/Ah during 356 days at room temperatures ($23 \pm 4^\circ\text{C}$) and were finally opened to analyze their lead sulfate content. The plates were pulled out of the batteries and rinsed with distilled water for 1 week before picking up active material samples. Lead sulfate contents on the later samples were measured chemically: the successive formation of complexes is achieved using nitric acid, acetic acid, xylenol orange and titriplex III (EDTA). The last complex is reached using a titrated solution of EDTA. The final stage of this reaction induces a color change: violet to yellow. Sulfate content was calculated from the sample mass and the quantity of EDTA used.

Low currents were applied using METRIX power supplies AX 502 2.5A – 30 V. As their accuracy does not allow a fine control of currents as low as a few mA, these power supplies were used as voltage regulators and the low currents were set using resistors in series.

Positive polarizations were evaluated using a lead acid battery reference electrode, which was first charged and then stabilized at open circuit. A current path between the tested cell and the reference electrode was provided by a tube filled with immobilized electrolyte.

Batteries used for capacity measurements were AGM VRLA batteries, PowerSafe V, 400 Ah/2V. These batteries were taken from the field and replaced by new ones after 6-year service, which corresponds to their mid-life (they are designed to last 12 years or more). These batteries were first submitted to 15 discharge-charge cycles in order to stabilize their capacities. The last capacity obtained is the reference capacity. They were then subjected to tests at room temperatures ($18 \pm 3^\circ\text{C}$): open circuit, low-currents of 29 and 105 $\mu\text{A}/\text{Ah}$ and float charge at 2.27V (current response is 414 $\mu\text{A}/\text{Ah}$). After 6 months, these batteries were discharged with the same rate (C/10) to determine their final capacities.

The discharge-charge cycles were performed with BITRODE Battery Charging and Test System, Module Type LCN 100A – 5V.

Voltages, currents, positive polarizations and temperatures were recorded with Personal Daq 56™ USB Acquisition Systems.

SULFATE CONTENT MEASUREMENTS

Results

Table 1 gives the results of PbSO_4 contents at initial state (after 231 days on open-circuit), and PbSO_4 final contents (after 356 more days at open-circuit and at constant low currents varying from 25 to 200 $\mu\text{A}/\text{Ah}$). Sulfate content variations during 356 days are the difference between final and initial sulfate contents. The Effective Resulting Currents associated to these sulfate content variations are calculated using the first Faraday's law. Current Balances are the calculated sums of self-discharge currents at open-circuit and applied low currents.

Table 1: After 231 days at open circuit, four batteries were subjected to low-currents during 356 days at room temperatures $23 \pm 4^\circ\text{C}$. Contents of lead sulfate were measured at the negative active material (NAM) and the positive active material (PAM). Active material samples were taken from the middle of the negative and positive plates.

Executed service during 356 days	Applied Current ($\mu\text{A}/\text{Ah}$)	PbSO ₄ content (% mass)		PbSO ₄ content (% mol)		PbSO ₄ content variation during service (% mol)		Effective Resulting Current ($\mu\text{A}/\text{Ah}$)		Calculated Current Balance ($\mu\text{A}/\text{Ah}$)	
		NAM	PAM	NAM	PAM	NAM	PAM	NAM	PAM	NAM	PAM
Initial state (231 days at open circuit)	-	16	30	11.5	24.4	-	-	-	-	-	-
Self-discharge (Open-circuit)	0	21	45	15.4	37.3	+3.9	12.9	-9	-31	-9	-31
Low current	25	16.8	24.0	12.1	19.4	+0.6	-5	-1.4	12	16	-6
idem	50	11.5	19.1	8.1	15.4	-3.4	-9	8	22	41	9
idem	100	1.9	8.6	1.3	6.8	-10.2	-17.6	23	42.6	91	69
idem	200	1.4	4.8	1	3.8	-10.5	-20.6	24	50	191	169

In Figure 1, effective resulting currents are compared to calculated current balances for negative and positive plates.

Discussion

Open-circuit

At open-circuit, variations of sulfate content in mole percent are +3.9% for the NAM and +12.9% for the PAM [27]. One can calculate from these sulfate content variations the self-discharge currents during 356 days of the experiment: -9 $\mu\text{A}/\text{Ah}$ and -31 $\mu\text{A}/\text{Ah}$ for the negative and positive electrodes respectively. The self-discharge rate of the positive electrode is more than 3 times faster than the self-discharge rate of the negative electrode.

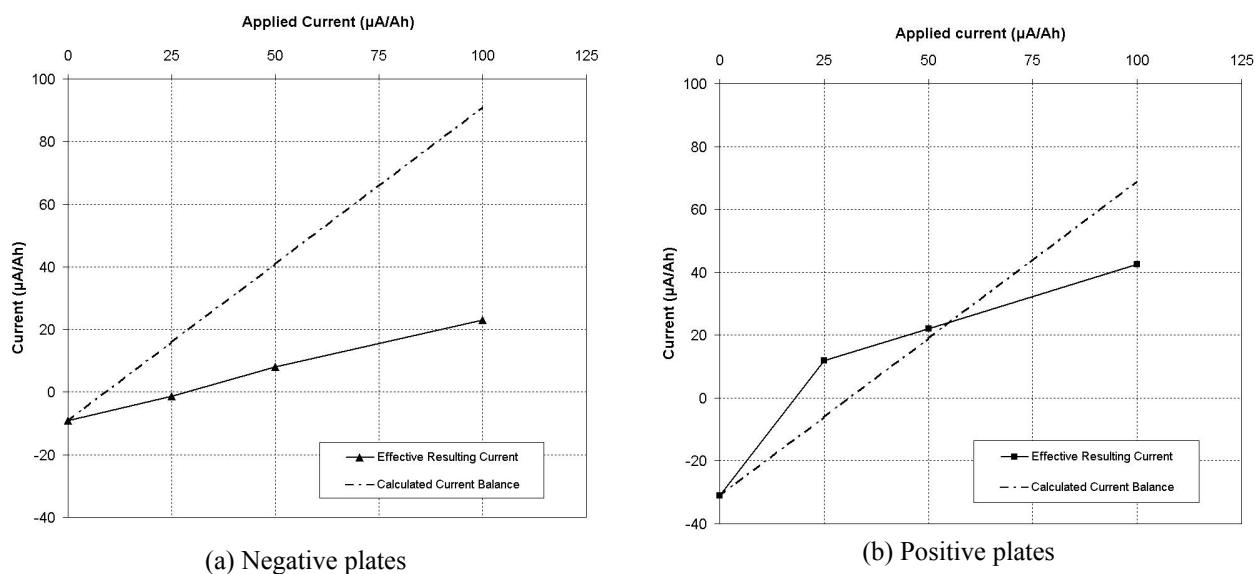


Figure 1: Balances of currents vs. applied currents: (a) negative plates, (b) positive plates. Dashed lines show the calculated current balances by the sum of the self-discharge currents and of the applied low currents. Full lines show the effective resulting currents corresponding to sulfate content variations.

Low currents

The lowest tested current, 25 μA/Ah, induces a drastic change in the sulfate content evolution inside the active materials. The sulfate content in the NAM remains almost constant (0.6 % increase) while the sulfate content in the PAM decreases by 5 %. So, the positive plate behaves worse than the negative plate at open-circuit but it has a better behavior as soon as the battery receives the lowest applied current.

a. Negative plates

Fig. (1, a) shows the calculated current balances and the effective resulting currents for the negative plates.

According to the lead sulfate content variation at 25 μA/Ah, the effective resulting current for NAM is -1.4 μA/Ah. This negative value means that the low current compensates only partially the self-discharge. The sum of the self-discharge current at open-circuit and the applied low current (called calculated current balance in table 1) is: $-9 + 25 = 16$ μA/Ah.

The difference between the current balance and the effective resulting current can be associated to losses of the applied current. Another explanation could be an increase of the secondary reactions involved in the self-discharge process under the polarization resulting from the applied current. These two hypotheses are in fact the same as secondary reactions involved in the self-discharge process and loss reactions during charging are identical: mainly hydrogen evolution and some oxygen recombination.

b. Positive plates

Fig. (1, b) shows the calculated current balances and the effective resulting currents for the positive plates.

A surprising result is observed for the 25 μA/Ah applied current. The effective resulting current, indeed, is higher than the calculated current balance. Of course, one cannot have an efficiency that is higher than 100%. The low current effect is then not only a compensation of the self-discharge process, but a modification and/or a rate reduction of the secondary reactions involved in this process.

Secondary reactions at the positive electrode are the oxygen evolution and the positive grid corrosion.

Oxygen evolution takes place at open-circuit, charge and discharge potentials. This reaction is known to increase its rate with positive polarization, so it cannot be the reason of the reduction of the self-discharge process observed in this case.

On the contrary, corrosion can slow down under positive polarization. This phenomenon is well known in the field of corrosion of metals as anodic protection, or more generally as corrosion passivation.

In the case of lead acid batteries, it is also well known that corrosion reactions are different at open-circuit and during charge. More exactly, corrosion reactions of the positive grid consist of 2 steps:



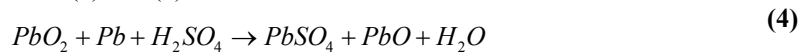
In which, O^{2-} ions are brought by migration from the active material across oxidation layers.

Only the first step (reaction 1) operates at open-circuit while these both steps occur during charge.

At open-circuit, divalent lead (Pb^{2+}) is the stable state for lead. Only the first step of the corrosion reaction occurs but not the second step (reaction (2), in which Pb^{2+} would oxidize into Pb^{4+}), because Pb^{4+} is not the thermodynamically stable state for lead. On the contrary, as at open-circuit the two electrons of the reaction (1) cannot be evacuated by the external circuit, they are used by reduction of Pb^{4+} into Pb^{2+} in the discharge reaction of the positive active material:



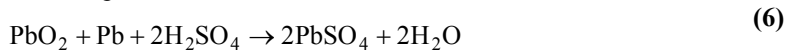
The combination of reactions (1) and (3) is then:



When PbO is not protected with a dense PbO_2 layer, it reacts chemically with sulfuric acid to form PbSO_4 as follows:



The overall corrosion reaction at open-circuit is then:



When this reaction occurs, usually in prolonged open-circuit conditions, positive grids are no more protected. Indeed, the formation of lead sulfate in the corrosion layers leads to mechanical stress, causing the formation of cracks and the destruction of the protective layer.

During charge (under low currents), reaction (2) takes place. Under positive polarization, Pb^{4+} (in PbO_2) is indeed the stable species of lead. PbO_2 produced in the reaction (2) is formed in the outer part of the corrosion layer. As it is dense, issued from the dense inner layer, and stable in sulfuric acid solution, it takes the role of protecting the PbO inner layer and the positive grid from the electrolyte [11, 14, 30].

Under positive polarization, grid corrosion, which is part of the self-discharge process of the positive electrode, is then slowed down by the formation of a protective layer of dense PbO_2 . Therefore, for the positive electrode, this can be the reason why the effective resulting current is sensibly higher than the calculated current balance, as long as corrosion is important part of the positive self-discharge process. Benefit of 25 $\mu\text{A}/\text{Ah}$ low current is then triple:

(i) Compensation of the self-discharge:

For intermittent charge, in the prospect of replacing open-circuit periods by low current periods, it is obvious that periodical charges would no longer be necessary or at least would be required less frequently.

(ii) Slowing down of the corrosion rate:

As evoked previously, several authors indicate a minimum corrosion zone. This minimum is generally situated at positive polarizations in the [30, 80] mV range [11, 14, 22-26]. It must be noticed that our results, obtained at room temperatures, differ from these published results, generally obtained at accelerated conditions. Indeed, at 25 $\mu\text{A}/\text{Ah}$, the strong effect observed can be attributed to an important reduction of the corrosion rate. But in this case the positive polarization is in the order of 2 mV, much lower than the preceding published values.

(iii) Reduction of water consumption:

It concerns water involved in corrosion reactions. In these reactions, the oxygen is taken from the positive active material. In turn, the active material takes oxygen from water of the electrolyte. The oxygen, finally locked in the corrosion product layers, cannot be recombined to reform water. So, the decrease of water consumption due to corrosion is important for the battery life span.

CAPACITY MEASUREMENTS

Results and discussion

Table 2 gives the capacities of VRLA batteries before and after 6 months at open-circuit, at low currents of 29 and 105 $\mu\text{A}/\text{Ah}$, and at 2.27V float charge. These experiments were done at room temperatures ($18 \pm 3^\circ\text{C}$). The battery at open-circuit for 6 months lost $(264 - 327)/327 = -19.3\%$ of its capacity compared to its reference capacity; the capacity of the battery subjected to 105 $\mu\text{A}/\text{Ah}$ increased by $(329 - 313)/329 = +5.1\%$.

Table 2: Capacities of the VRLA batteries, which were subjected to open circuit, to 29 and 105 $\mu\text{A}/\text{Ah}$ low-currents and to 2.27 V float charge during 6 months at room temperature ($18 \pm 3^\circ\text{C}$) as well as their reference capacities before this service.

Reference Capacity C_{ref} (Ah)	Applied Current during 6 months at room temperatures $18 \pm 3^\circ\text{C}$ ($\mu\text{A}/\text{Ah}$)	Final Capacity C_{fin} (Ah)	Capacity Variation $(C_{\text{fin}} - C_{\text{ref}})/C_{\text{ref}}$ (%)
340.0	~ 414 (float charge at 2.27V)	379.0	11.8
313.0	105	329.0	5.1
318.0	29	308.0	-3.1
327.0	0	264.0	-19.3

Figure 2 shows the capacity variations given for each battery in percent of their reference capacities. These percentages are also the state of charge variations induced by the applied currents during the 6 months of experiments. The capacity variation at open-circuit, i.e. -19.3% , allows to calculate the average self-discharge current, which is $36 \mu\text{A}/\text{Ah}$.

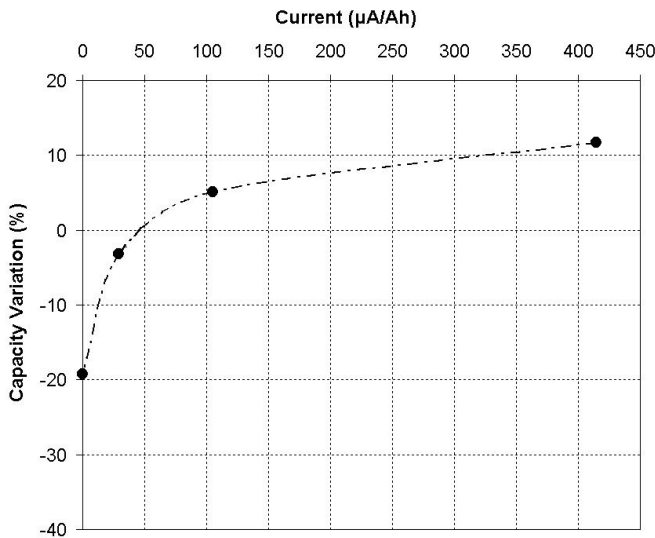


Figure 2: Relative capacity variations (state of charge variations) vs. applied currents. VRLA Batteries were tested during 6 months at room temperatures ($18 \pm 3^\circ\text{C}$).

The capacity variation has a strong increase for the lowest applied current ($29 \mu\text{A}/\text{Ah}$). It appears that this low current, whose value is 80 % of the average self-discharge current, compensates 84 % of the self-discharge. Such a high efficiency suggests that these VRLA batteries could be positive limited at this mid-life state. Further experiments confirmed that these batteries were indeed positive limited. Increases of the state of charge at $105 \mu\text{A}/\text{Ah}$ and at float charge ($414 \mu\text{A}/\text{Ah}$) indicate that the charge procedure we used does not lead to a real full charge. Charge is then completed slowly at currents beyond $50 \mu\text{A}/\text{Ah}$.

According to experiment results, we supposed that when combined with low currents in the order of 25 to $50 \mu\text{A}/\text{Ah}$, a refresh charge every 6 months or every single year is sufficient to maintain antimony-free batteries in a good state of charge. Higher currents, as traditional float currents (10 times higher), are not necessary and not suited, as they would increase corrosion and water loss.

We consider using a refresh charge only when the battery has been subjected to a service discharge. This is a common point with the Multibat Stationary system where batteries are periodically discharged and charged to evaluate the battery state of health.

NEW MANAGEMENT SYSTEM FOR STANDBY VRLA BATTERIES

A new management system for standby VRLA batteries has been developed at EDF using the Stationary Multibat system [9] to ensure system reliability and the Low-current method [27] to improve the battery life span.

Fig. (3, a) describes a simplified schematic diagram of the Stationary Multibat system. 3 battery-pack strings in parallel instead of 1 pack are used in order to improve the system reliability. In case of a cell failure such as an open circuit, the system loses only one battery-pack string, i.e. one-third of the total capacity. This configuration also allows each string to be discharged completely across the test resistance to evaluate its real state of health. Each battery string is periodically (e.g. every 6 months) discharged and charged.

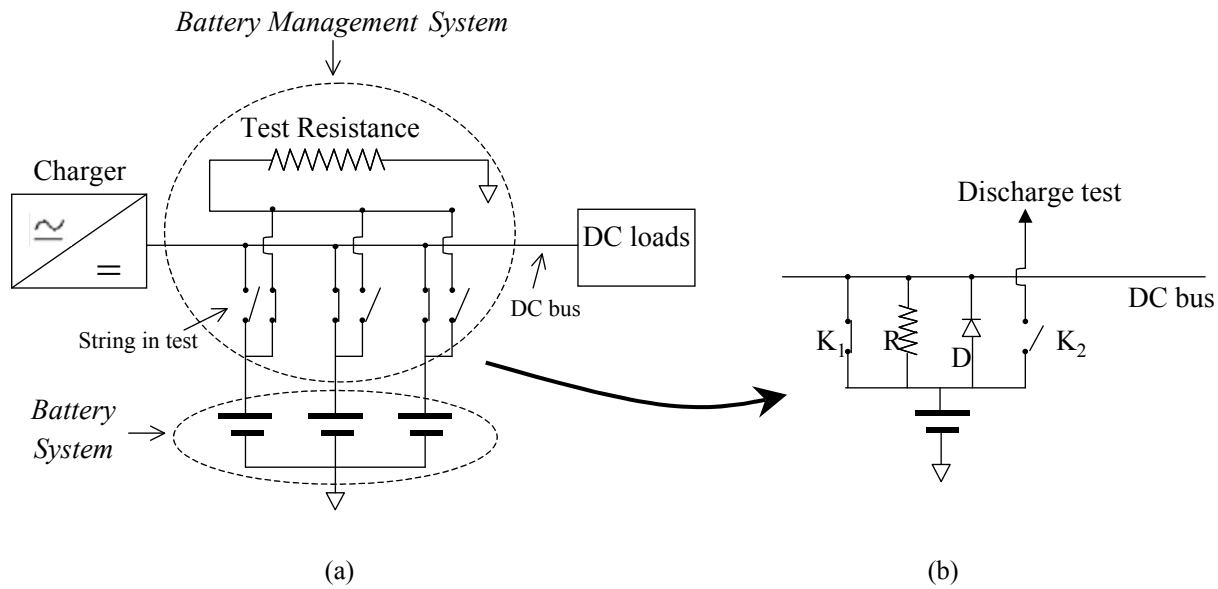


Figure 3: (a) Simplified schematic diagram of the Stationary Multibat system. (b) Zoom on the power electronic components of one of the three battery-pack strings using Low-current method.

Figure (3, b) describes how to integrate and to operate Low-current method of maintaining the charge on the Stationary Multibat system:

- A resistance branch is added to reduce the traditional float current with a factor of 5 to 10. In standby state, PLC K_1 and K_2 are open; the battery is maintained in charge with a low current via the resistance R .
- On backup demand, a voltage drop appears on the DC bus, the battery immediately provides electricity to the DC load via the diode D . Then PLC K_1 closes; DC load is directly supplied by the battery.
- During the periodical discharge test (e.g. every 6 months) to evaluate the battery state of health, PLC K_2 closes and K_1 stays open. The battery-pack string is discharged across the test resistance. The low current, still provided to the battery pack string, is a parasitic but negligible effect ($< 0.1\%$). Next, the charge is operated via K_1 . The charge current is controlled by a Pulse Wave Modulation (PWM). When the charge finishes, K_1 re-opens, the battery is re-maintained the charge with a low current via R .

All those operations are done sequentially and automatically so that no intervention is required from the maintenance.

This system is expected to fulfill 3 targets:

- Increasing the system reliability by a redundancy in the design of the battery system and by an assessment method to control battery states of health.
- Improving battery life spans by the Low-current method, leading to reduced corrosion and water loss.
- Decreasing maintenance costs.

CONCLUSION

Antimony-free lead-acid batteries were tested for several months at open-circuit, float charge and intermediate rates of charge – called “Low-current” in this paper. The following conclusions can be drawn from this study:

- In antimony-free lead acid batteries, the self-discharge rate of the positive electrode is higher than that of the negative electrode.
- Low-currents beyond 25 $\mu\text{A}/\text{Ah}$ appear to be able to maintain the state of charge for both positive and negative plates.
- The effect of 25 $\mu\text{A}/\text{Ah}$ low current on the positive active material is much higher than a simple compensation of the self-discharge. This can be explained by a passivation phenomenon of the positive grid corrosion. In other words, it constitutes an anodic protection of the positive grid.
- The use of low-current periods in place of open-circuit periods in the intermittent charging method should increase the life span of VRLA batteries, as it both lowers the water consumption and the corrosion rate of positive grids.

Combining the Low-current method, battery redundancy and automated periodical capacity measurements, VRLA batteries should provide long life spans and high reliability. Several management systems of this kind are being experimentally used at EDF.

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