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Search for an optimized cyclic charging algorithm for valve-regulated lead-acid batteries

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Abstract

Valve-regulated lead-acid (VRLA) batteries are characterized by relatively poor performance in cyclic applications of the order of two hundred to three hundred 100% depth-of-discharge (DoD) cycles. Failure is due to sulfation of the negative plate and softening of the positive active-material. It is felt that this failure mode arises from abnormally high levels of oxygen recombination that arise due to decreases in separator saturation levels as VRLA batteries age. Charging algorithms have been developed to address this changing condition throughout life. The key step is the finish of charge where, traditionally, low currents and low overcharge limits have been employed with poor results. It has been found that using high finishing currents in an alternating charge–rest algorithm results in proper recharge of the negative plate without creating unacceptable temperature increases. This has resulted in deep-discharge lifetimes of 800 to 1000 cycles, particularly when using a charging algorithm employing only partial recharges (97–100% return) interspersed with full conditioning recharges every 10th cycle. With such minimal average overcharge levels, deep-cycle lifetimes approaching 1000 cycles have been achieved without experiencing failure due to massive grid corrosion. © 2000 Elsevier Science S.A. All rights reserved.

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1. Introduction

Valve-regulated lead-acid (VRLA) batteries generally have relatively poor cycle lives in deep-cycling applications, particularly in thin-plate designs. Typical cycle lives are of the order of 200–300 cycles and autopsies of failed batteries indicate relatively little grid corrosion, a traditional mode of failure for lead-acid products in deep-cycling applications. Rather, it appears that the batteries fail due to a combination of negative-plate sulfation and positive-paste softening. This is usually more severe at the bottoms of the plates and is apparently due to excessive oxygen recombination. The discharge capacity is generally acceptable up to 150–200 cycles, but then exhibits a relatively rapid decline.

Typical algorithms for VRLA batteries employ constant-voltage (CV) charging with low finishing currents, and with limited overcharge (between 5% and 20%). Early in life, when the separator saturation level is high and the oxygen cycle is relatively inefficient, this algorithm is effective in recharging VRLA batteries, which are behaving much like flooded analogues. As VRLA batteries age, however, electrolyte and water are removed from the separator by a number of mechanisms, namely water– vapour transport, grid corrosion and redistribution from the separator into the plates. The net effect is a significant increase in the void space of the separator. This results in a progressive increase in the oxygen-reduction efficiency (ORE), which consumes a growing percentage of the overcharge current and ampere-hour input supplied to the battery. When this amount exceeds the allowed overcharge percentage, the battery is undercharged and the discharge capacity immediately begins to decline. This process is portrayed conceptually in Fig. 1 [1].

The decline in discharge capacity can be avoided temporarily by removing the overcharge limit (dashed/dotted line in Fig. 1), but eventually the amount of overcharge becomes unwieldy. In addition, the increasing amount of overcharge promotes the creation of more void space, which makes recharge more difficult. This cyclic spiraldown results in an improvement in cycle-life, but not by a

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Fig. 1. Conceptual model of the effect of the oxygen cycle on VRLA capacity-cycle performance.

great amount. In order to realize a quantum leap in battery life from the current norm of 200–300 to 800–1000 cycles, an entirely different strategy is clearly required.

The need for a new charging strategy appears to be a fundamental issue in the cyclic charging of VRLA batteries. It was first pointed out in quantitative terms by Atlung and Zachau-Christiansen [2], who developed a model for the process and applied it to the charging of both flooded and VRLA batteries. It was shown that as a VRLA battery ages (in contrast to a flooded analogue), the increased ORE takes up more and more of the overcharge applied to the battery, and eventually results in an undercharging of the negative plate. It was further demonstrated that at a given level of unsaturation where the oxygen cycle is highly efficient, any amount of overcharge will not succeed in adequately recharging the negative plate. When a VRLA battery reaches this state, the controlling factor is not the amount of overcharge applied, but rather its rate of application (a factor not addressed by Atlung and Zachau-Christiansen [2]). It is not surprising that conventional CV charging does not succeed in prolonging life in these situations, as finishing currents are intentionally quite low. Thus, a project was developed under the auspices of the Advanced Lead-Acid Battery Consortium (ALABC) to develop a new approach to the cyclic charging of VRLA batteries. It is based upon the concept that such batteries age significantly as they are cycled and that their physical configurations (primarily glass-mat separator void volumes) change accordingly throughout life. Thus, an adaptive charging algorithm is required to achieve high cycle numbers where grid corrosion, rather than negative-plate sulfation and/or positive-plate softening, is/are the ultimate failure mode(s). Some preliminary results from this

research (ALABC Project B-007.1) are reported here, and are intended to improve significantly the cycling performance of VRLA batteries in electric vehicle (EV) and other deep-cycling applications.

2. Experimental details

Cycling experiments were carried out on two VRLA products, viz., an Optima 12-V, 16-A h hybrid electric vehicle (HEV) developmental battery and a 12-V, 50-A h Optima 'Yellow Top' (YT) deep-cycling commercial battery used in a number of EV programmes. All batteries were first conditioned via 10 to 20 deep cycles with constant-current (CC) recharge which employed overcharge levels of ~ 30%. All batteries also had to pass screening tests with acceptable open-circuit voltage (OCV), impedance, weight and high-rate discharge values. These are spiral-wound products with thin plates of 1 mm or less and all-glass separators with compression levels of ~ 30% to 40%. The grids are cast-punched sheet made from a binary lead-tin (~ 0.65% Sn) alloy.

Cycle testing at Optima was carried out by means of Bitrode LCN testers, as well as two 'home-built' units. Temperature monitoring was done on the outer case in such a way that the difference between interior and exterior temperatures was of the order of 2 to 3° C. A temperature limit of 60° C was used, at which point the battery was subjected to a 45-min rest before cycling was recommenced. Some tests were carried out with fan cooling. All testing was continuous, with minimal rest periods of the order of a few minutes or less between charge–discharge steps. Cycle testing at the National Renewable Energy Laboratory (NREL) was undertaken with an AeroVironment ABC-150 cycle tester in conjunction with temperature monitoring and fan cooling. Each discharge was taken to 10.5 V, i.e., 100% depth-of-discharge (DoD) by BCI Standards, at the $C_1/1$ (12-V, 16-A h units) or $C_2/2$ (12-V, 50-A h units) rates, unless otherwise stated. Batteries were cycled to failure at 50% of the initial or rated capacity, with the 80% level also indicated.

Reference-electrode measurements were made with 'home-built' mercury/mercurous sulfate electrodes that were inserted into the top of the separator/plate stacks between the plates. Pressure measurements were taken with Iomega Model 154 (0–15 psig) sensors; each probe was positioned in the head space of the battery in order to minimize contact with electrolyte. The sensors were sealed in the battery cases with silicone-Teflon gasket seals.

3. Results and discussion

As for all thin-plate VRLA batteries, the Optima products exhibit cycle-lives in the range of 200 to 300 deep cycles to failure with standard CV charging. Work at Optima has determined that addition of a 2-A, 1-h CC finishing charge for the 12-V, 50-A h YT battery is beneficial, but cycling performance is still not acceptable. This is an algorithm where the battery is CV charged at 14.5 V with a 25-A current limit. When the CV current taper reaches 1 A, the charge is completed with a 2-A, 1-h CC step; the total charge return is limited to 120%. Loss of capacity is gradual right from the beginning, with an increase at ~ 300 cycles, at which point the battery has reached the 50% failure level (Fig. 2). What is remarkable - and typical of VRLA battery behaviour - is a sharp increase in the current taper at the end of the CV stage, as shown in Fig. 3. By the time the battery was taken off test

at cycle 400, the discharge capacity had dropped to $\sim 33\%$ of initial and the charge current at the end of the CV stage was on the verge of going into thermal runaway. Roughly accompanying the sharp rise in current is a drop in the end-of-charge voltage, also indicative of a system which is going into thermal runaway, as seen in Fig. 3. All of this behaviour is representative of a situation where the ORE is extremely high and drawing large currents at the end of charge. If the finishing charge current level is below what the battery will draw for the oxygen cycle, the battery will not be fully recharged, regardless of the percent overcharge, i.e., it doesn't matter how long the battery is charged at this current level because virtually all the current is going into oxygen reduction at the negative plate rather than completing the conversion of lead sulfate to sponge lead. The CC finish at 2 A, while being somewhat effective in extending life, is not adequate to maintain the battery at anything close to full discharge capacity beyond ~ 150 cycles. With this as a backdrop, testing was initiated at Optima and NREL to develop charging algorithms which would either overcome this situation or delay its onset to such a degree that significantly higher cyclelives would be obtained for the Optima batteries in deepcycling tests.

To develop a useful charging algorithm, several principles were employed, namely:

- use of high-inrush charging currents, of the order of $C_2/2$ or higher;
- use of stepped constant-current stages to charge the battery rapidly without inducing excessive heating;
- provision for a moderate level of finishing current (actually, a high level compared with conventional CV charging) to minimize the time on charge and to have charging currents available in excess of the amount required for the oxygen cycle;



Fig. 2. Typical plot of capacity vs. cycle number for an Optima 12-V, 50-A h deep-cycle YT battery. Discharge: 25 A to 10.5 V. Charge: 14.5 V CV charge with a 25 A current limit and maximum charge return of 120%; 2-A, 1-h CC finishing charge is applied when current taper reaches 1 A or the 120% limit.

Fig. 3. Current at end of CV charge step (1 A or 120% charge return limit) and EOCV as a function of cycle number for battery shown in Fig. 2.

 selection of a charge-termination algorithm which provides for full recharge of both plates, but with a minimum of overcharge.

These were based upon the perception that the key to charging VRLA batteries is being able to deal with, and overcome, the increasing current demands of the oxygen cycle as the battery ages. The principles also assume that the life of VRLA batteries is limited by degradation of the negative plate, which causes a shift from positive-plate discharge capacity limitation to one involving the negative plate. In order to test this, several 12-V batteries were fitted with mercury/mercurous sulfate reference electrodes (one cell only) throughout cycle-life. A clear shift from positive- to negative-plate limitation was observed; moreover, the negative-plate capacity continues to degrade with cycling.

According to the work carried out by Atlung and Zachau-Christiansen [2] on cyclic applications and by Jones and Feder [3,4] on float charging, insufficient charge or recharge of the negative plate is due to the depolarizing effect of the oxygen cycle on the negative plate. Because the standard potential for oxygen reduction is more positive than the OCV for the negative half-cell, it is difficult or impossible to recharge fully the negative plate in the presence of a significant amount of oxygen reduction. Consequently, the development of effective charging algorithms focused on ensuring that polarization of the negative plate was substantial toward the end of the recharge process. A harbinger of the charge algorithm's inability to polarize sufficiently the negative plate is a drop in the end-of-charge voltage (EOCV). As this study utilized CC charging, this is easy to monitor. With the current levels used, EOCV values were initially in the range of 16.0-16.5 V. As cycling progressed, these voltages would typically hold steady for ~ 100 cycles and then begin to slowly drop (see Fig. 3). Unless the charge algorithm is modified,

this drop continues and is accompanied by a similar downward trend in discharge capacity. Paralleling this is an increase in the temperature at the end of the charge cycle; it approaches levels of 60 to 65°C when high CC charge currents ($C_5/5$ to $C_{10}/10$) are employed. When the EOCV values approach ~ 14 V, the battery capacity is likely to be at, or near, the failure level.

Initial cycling tests were performed with a simple stepped-CC algorithm with a fixed amount of overcharge, typically $\sim 20\%$. The drop in EOCV and rise in temperature quickly limited cycle life and it was found that late in life the CC finish could not be used due to overheating of the batteries. Moreover, it appeared that the high overcharge level was rapidly creating significant separator void space and the oxygen cycle was becoming dominant. This created a situation where the cycle-life levels attained were well short of what could be achieved with even conventional CV–CC charging.

In order to minimize overcharge and yet still recharge fully the battery, the termination algorithm was changed from a simple ampere-hour count to using the 'zero delta V' (ZDV) point on the voltage-time curve. The ZDV point is on the forward portion of the curve where the voltage flattens out in the gassing region. As the battery is only 97% to 98% recharged at this point, 5% to 10% additional charge was provided to the battery to complete recharge. Because the amount of overcharge was limited to a low value, this algorithm resulted in cycle-lives of ~ 250 cycles to 50% of rated capacity, i.e., roughly the same as with conventional CV–CC charging.

In order to overcome this limitation, the overcharge amount was raised successively as the battery aged, beginning with the first significant drop in discharge capacities at about cycle 220. As seen in Fig. 4, each increase in overcharge resulted in a temporary recovery of discharge capacity. As the battery aged, however, it required more and more overcharge to drag the capacity back up and

Fig. 4. Capacity as a function of cycle number for Optima 12-V, 16-A h batteries under a standard CV charge regime and stepped-CC charge with increasing overcharge.

there was still a downward trend overall. At 150% charge return (50% overcharge) and about 375 cycles, the test was terminated.

This experiment demonstrated an important point in the cyclic charging of VRLA batteries. As they age, the overcharge requirement of the battery increases due to the higher ORE; unless the amount of overcharge is increased (at a given rate of charge), it will become difficult and then impossible to recharge fully the battery. This is why charging algorithms involving fixed overcharge amounts (and, worse, in combination with low-level finishing currents) eventually fail, as demonstrated in Figs. 1 and 4.

3.1. Current-interrupt finishing charge

Most of the work on finishing charge described above was performed at the $C_{10}/10$ rate, which was the maximum level useable for the 12-V, 50-A h battery without incurring excessive heat build-up. As noted earlier, it was realized that the *rate* of charge during finish is more important than the *amount*, but with CC charging the upper current for finishing is limited, particularly for larger batteries. In order to provide higher currents during finish, which is necessary for polarization of the negative plate, a pulsed-charge approach was adopted. Because of the relatively long charge and rest times employed, this technique is termed 'current-interrupt', or CI.

The CI technique makes it possible to use high finishing currents (pulsed-current amplitude) and still adequately dissipate the heat generated by the oxygen cycle (during rest periods). An example of a CI curve is shown in Fig. 5. The first portion of the charge curve is a single-step CC stage, with termination at 85% charge return. The algorithm is then switched to CI at about 20 min and this is

Fig. 5. Example of current-interrupt finishing charge, cycle 326 for a 12-V, 16-A h HEV battery. CI is applied until the battery reaches 14.1 V at the end of the rest periods.

continued until the rest-period voltages are above ~ 14.1 V and are steady. For this experiment, the internal pressure was monitored and is shown as the lower curve. Interestingly, even though the battery is heavily polarized during the charge periods, there appears to be little or no build-up of hydrogen gas. Instead, the pressure readings track the course of the increase in ORE and it appears that most or all of the gas generated is oxygen. This would imply that there is a significant kinetic hindrance to hydrogen generation during the charge periods. If this is verified, it allows for the brief charge excursions required for CI with little or no danger of hydrogen accumulation and, possibly, venting. Clearly, more work is needed in this area to determine quantitatively the gassing behaviour during CI.

Initially, CI was used to attempt to recover batteries which had declined in capacity to the 50% cut-off level, or below. One example of this is shown in Fig. 6. Using the stepped-CC algorithm with limited overcharge, the performance was almost identical to that obtained for the HEV battery using the Optima standard CV-CC finish approach, viz., ~ 220 cycles to 80% capacity. Capacity "spikes" were obtained when the overcharge level was increased, but the capacity-cycle plot flattened out at about 6 A h and cycling in this mode was terminated at \sim cycle 370. At this point, CI was initiated and there was a capacity recovery to almost the 80% line. Changes to the CI algorithm produced rises and drops in capacity and the battery was terminated at 500 cycles at the 50% level. This is not a "one-off" result, as similar experiments were carried out on other batteries that went to 600-700 cycles. It is clear from these results that capacity loss is reversible to some extent, but it is also clear that restoration of the batteries to 100% capacity could not be achieved.

When CI finishing was applied from the beginning of cycling, better results were obtained, as shown in Fig. 7.

This battery was subjected to a 90% DoD regime. The battery was removed at 762 cycles at $\sim 50\%$ capacity. As can be seen, it received very heavy levels of overcharge, particularly late in life. In addition, end-of-charge temperatures rose steadily throughout life and for the last 150 cycles were at or above 60°C. For the last 100 cycles, the charge return curve is flat due to thermal kickouts. This battery failed because the charge return, even with CI, was not 100% efficient. Inspection of the CI curves shows that the current used (20 A) was not sufficient to polarize the negative plate and thus the rest voltages could not be raised above ~ 13.7 V. At the time this test was terminated, CI testing was not done at higher currents. In retrospect, this battery would probably have continued to higher cycle numbers with higher-current pulses. Duplicate testing with higher CI current amplitudes is now under way.

The experiments with CI finishing charge have validated an important point in the effective charging of VRLA batteries, particularly at high cycle numbers. As VRLA batteries age in cyclic duty, the current draw for the oxygen cycle increases at a significant rate. Physically, this means that the magnitude of the current draw for the oxygen cycle correspondingly increases. In order to recharge effectively the battery, the charge rate at finish must be increased throughout life, not just the amount of overcharge (commonly done at a low rate of charge).

3.2. Partial-state-of-recharge cycling

It is clear from the work presented above that forced polarization through the use of high-current pulses is a viable approach to extending the cycle-lives of VRLA

Fig. 6. Example of a failed 12-V, 16-A h battery which was revived with CI finishing charge after stepped-CC charging with a pure CC finish.

Fig. 7. Capacity as a function of cycle number for Optima 12-V, 50-A h deep-cycle YT battery. Discharge: 25 A to 11.0 V (90% DoD). Charge: 200 A to 60% charge return, 50 A to 80%, 15 A to 100%, CI overcharge (20 A for 15 s, 20 s rest, to 14.2 V).

batteries. This is something of a losing game, however, as heavier overcharge creates more void space that requires higher-current finishing, which then creates more void space, and so on. It is also clear that these products are very sensitive to overcharge, which must be kept to an absolute minimum in order to optimize cycle-life. This creates a 'Catch 22' situation, as it is known that overcharge is necessary for long-time performance but at the same time it is slowly killing the battery. As noted previously, overcharge at low current amplitudes is the worst possible approach because of its ineffectiveness in overcoming the oxygen cycle. In an effort to reduce overcharge without experiencing capacity walkdown, an approach called "partial-state-of-recharge", or PSoR, has been developed. This algorithm involves terminating recharge using a voltage at the base of the rapidly-rising portion of the voltage-time curve, a point where charge return is at $\sim 97\%$ to 99%. As there is some charge inefficiency, this will only return the battery to $\sim 95\%$ to 98% SoC, so the battery experiences capacity walkdown on discharge. In order to compensate for this, on every 10th cycle the battery is given a conditioning charge with 20% to 40% overcharge. Because there is little or no overcharge for the intervening cycles, the per-cycle overcharge percentage is only of the order of 2% to 4%. An example of this type of cycling over the first 100 cycles is shown in Fig. 8 and a full capacity plot is given in Fig. 9, both for 12-V, 50-A h YT modules. For the full cycling test, the Optima standard conditioning charge of 4 A for 16 h was used every 10th

Fig. 8. PSoR cycling of Optima 12-V, 50-A h YT battery, first 100 cycles. Discharge: 25 A to 10.5 V (100% DoD). Initial charge: 25 A to 80% charge return, 10 A to 15.5 V; full recharge every 10 cycles at 5.2 A for 10 h followed by 20 A/15 s CI with 20 s rests to 14.2 V rest voltages. Solid line is discharge capacities; dotted line is charge return.

Fig. 9. PSoR cycle testing of an Optima 12-V, 50-A h YT battery. Discharge: 25 A to 10.5 V (100% DoD). Charge: 25 A to 80% charge return, 10 A to 16.0 V; conditioning cycle every 10th cycle at 4 A for 16 h.

cycle; on intervening cycles, the 10-A finishing current step was terminated at, first, 16.0 V and later (at ~ 440 cycles when the battery was not reaching 16.0 V on charge) at 14.5 V. When the termination voltage was stepped down, the 10-cycle charge was also reduced to 60 A h. This is an encouraging result, but at high cycle numbers the 4-A conditioning current amplitude is not sufficient to recharge fully the negative plate. This test was repeated, but with the use of CI for finishing on the conditioning cycles. The results thus far are shown in Fig. 10, where it can be seen that the battery is still delivering $\sim 80\%$ of initial capacity at cycle 780 following the conditioning charge steps. This is a 100% DoD duty cycle $(C_2/2$ discharge rate to 10.5 V) and it is clear that the limitation of overcharge is exerting a beneficial effect on life.

PSoR cycling appears to be the closest approach to using a single, fixed charge termination algorithm, but limitations still remain. As VRLA batteries are taken to high cycle numbers (500 or more), the shape of the CC charge voltage–time curve becomes poorly defined and drops to lower voltages as the oxygen cycle becomes dominant. This requires adjustments to the cut-off voltage used for PSOR or the battery will be heavily overcharged due to the charge voltage not reaching the cut-off value. If a low-voltage cut-off (13.5 to 14.5 V) is used from the beginning of cycling, discharge capacities early in life will be substantially lowered relative to using a higher value of 15-16 V. Thus, some adjustments must be made throughout cycle-life to optimize discharge performance.

As an example, the testing shown in Fig. 10 charging was terminated at 15.5 V for about the first 350 cycles, at

Fig. 10. PSoR testing of an Optima 12-V, 50-A h YT battery to 780 cycles. See legend of Fig. 8 and text for charging algorithms throughout life.

which point it was dropped to 15.2 V because the battery was no longer reliably reaching 15.5 V on charge. At about cycle 570, the battery was again not reaching the set cut-off voltage (Fig. 10) and several cycles with high overcharge levels were experienced; subsequently, the voltage was lowered to 14.6 V (cycle 610), 14.4 V (cycle 739) and 14.2 V (cycle 812, not shown). At higher cycle numbers, it is anticipated that the battery will have to be put on a timed charge, as the shapes of the voltage–time curves have become very indistinct and have flattened out.

The decline in discharge capacities between conditioning charges is clearly not acceptable for some applications. For EV use with limited range and weekend full recharges, however, it may do very well. This approach has not been 'fine tuned', but it is apparent that it holds great promise for charging VRLA batteries. The experiments shown in Figs. 9 and 10 demonstrate a further point for VRLA charging algorithms. The amount of overcharge on a percycle basis must be absolutely minimized in order to achieve very high cycle numbers for VRLA batteries. Overcharge creates more void space in the glass-mat separator and this in turn increases the efficiency of the oxygen cycle. As the ORE increases, more current is drawn just for oxygen reduction — a parasitic process which does nothing for the completion of recharge of the negative active-material. Finishing currents in excess of this parasitic draw must be applied to the battery to finish recharge. Higher finishing currents, however, tend to increase the total amount of overcharge and thus create more void space. This cyclic process requires an escalation of current-interrupt finishing currents at high cycle numbers in order to recharge effectively the negative plate.

4. Conclusions

Thus far, this study has been successful in extending cycle-life values for Optima thin-plate VRLA batteries by some 200% to 300% in deep-cycle, constant-current discharge tests at the $C_1/1$ (HEV battery) and $C_2/2$ (YT EV battery) rates. The following requirements are necessary in order to achieve high cycle numbers for these products:

• use of an adjustable charge-termination algorithm to compensate for the increasing influence of the oxygen cycle;

- strictly limited amounts of overcharge so that the rate of separator void space increase is kept to a minimum;
- gradually increasing current amplitudes for finishing of the charge process to satisfy the current draw for the oxygen cycle and still have charge available for completing the recharge of the negative plate.

The traditional charging procedures with low finishing currents and strict overcharge percentage limits ensures that at ~ 200 to 300 deep cycles, most thin-plate VRLA batteries will fail to deliver adequate discharge capacities because of insufficient recharge of the negative plate. Due to the increasing strength of the oxygen cycle, more and more aggressive charging conditions must be applied to VRLA batteries as they approach high cycle numbers. Both the amount of overcharge and the rate of finishing charge must be gradually increased as the batteries age. It has been demonstrated that this approach is successful in a qualified sense, but more effective measures involving fundamental design and materials changes can yield similar or superior results. Also, the periodic addition of water during long cyclic service may also be effective in controlling the influence of the oxygen cycle. Work toward these ends is continuing in earnest.

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