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Progress towards an advanced lead–acid battery for use in electric vehicles

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Abstract

The attributes which are essential for a battery to be successful as the energy store for an electric vehicle are reviewed. These are then matched against the substantial advances in the technology of valve-regulated lead–acid (VRLA) batteries that have been posted during the course of the technical programme of the Advanced Lead–Acid Battery Consortium (ALABC). A project which was designed to draw together several desirable features, identified during the early years of the ALABC programme, into a test battery has provided much useful information. The design target for specific energy (36 W h kg^{-1}) has been achieved successfully. Cycle-life is short, but it appears likely that an inappropriate charging regime with an unrestricted charge factor was largely responsible. Benchmark tests with a commercial product also yield very short life with this regime, but provide good performance when the charge factor is kept in check. Attention to the deployment of suitable charging regimes continues to be a fruitful area in extending the life of VRLA batteries, and the ALABC's programme to enhance both specific energy and life, while shortening recharge time, is making good progress. © 1999 Elsevier Science S.A. All rights reserved.

Keywords: Cycle-life; Electric vehicle; Lead–acid batteries; Rapid recharge; Specific energy; Valve-regulated

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1. Essential characteristics for electric vehicles

Ever since the Air Resources Board in California proposed [1], at the beginning of the 1990s, to mandate the sale of large numbers of electric vehicles by the major automobile manufacturers, there has been a vigorous debate over what are the essential features that such vehicles should offer in order to be acceptable to the majority of the purchasing public. Initial preoccupation with the sole issue of range per charge of the battery, and hence specific energy, has given way to a recognition that cost is a major issue and that range per charge is much less of a problem provided that it is possible to recharge the vehicle battery quickly. Indeed, it is clear that if it is not possible to recharge the vehicle battery quickly, then specific energies of even two or three times greater than that of lead–acid may not render the prospect of an electric vehicle sufficiently attractive to a potential purchaser. A recent EPRI survey [2] expressed the view that there will be a market for vehicles with a range of between 160 and 190 km that should be of the order of 1.5 to 2% of total vehicle sales in the USA (in the next several years).

The current status of the performance of vehicles available with lead–acid batteries has been evaluated by EV America. Their report shows [3] that the most up-to-date offerings of the major automobile manufacturers (the General Motors EV1 and the Ford Ranger) offer a range of around 110 km on a prescribed driving cycle and significantly more than this at a constant speed of 70 km h^{-1}. Lead–acid batteries currently used in these vehicles are characterized by a specific energy of some 35 W h kg^{-1}, so it is clear that in order to achieve a range of over 160 km, a specific energy of around 50 W h kg^{-1} should be the target. A recent survey [3] of daily driving range of drivers in North America shows that a range of 130 km would satisfy the needs of 90% of drivers and that there is a long tail for the remaining 10% which extends into well over 240 km, probably to 480 or 640 km. The message here is that a reasonable range per charge (of around 160 km), coupled with the ability to recharge quickly, will be far more useful than a range per charge of 240 km followed by a period of hours when the vehicle is out of commission.

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Cycle-life of course is always important, and so ongoing research programmes for batteries for electric vehicles tend to emphasize these three parameters: specific energy, rapid recharge, and cycle-life.

2. Advances in valve-regulated lead–acid (VRLA) battery technology

Uniquely, among the battery systems quoted as candidates for powering electric vehicles, the lead–acid battery is produced by well-established manufacturing organizations around the world. Uniquely too, this system is being developed for electric vehicles through a global consortium of all interested companies who have set aside their competitive instincts in favour of a cooperative drive towards a product that should address all of the needs of the emerging electric vehicle industry. This is the Advanced Lead–Acid Battery Consortium (ALABC).

The lead–acid battery is often presented as an ancient technology with limited scope for improvement. Although the traditional flooded lead–acid battery does indeed have a long history, it was clear to all concerned at the beginning of the present drive for electric vehicles that the need was for a sealed product. Therefore, the VRLA battery has been adopted for modern electric vehicles and this has a history scarcely longer than those of the newer battery chemistries. At the beginning of the 1990s, the VRLA battery available for consideration in electric vehicles offered promising cost and specific-power characteristics, but it had a very poor cycle-life coupled with a modest specific energy, and required a long time for recharge (see Fig. 1).

During the course of the world-wide programme of research and development carried out by the ALABC through the 1990s, the performance of the VRLA battery for electric vehicles has improved dramatically. The present phase of the ALABC program (Fig. 2) is implementing advances at the component level, in battery design, in building improved batteries, in testing, and in vehicle programmes.

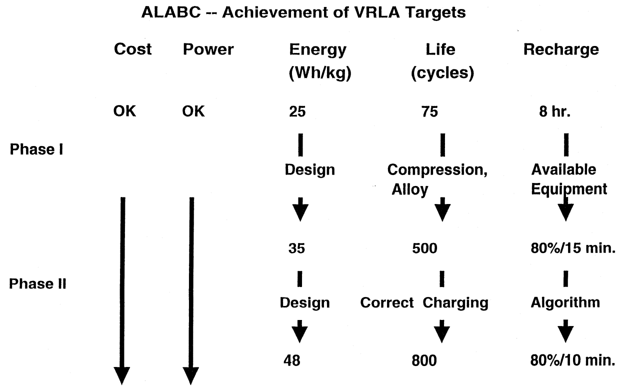

Fig. 1. Evolution of performance parameters for VRLA batteries from 1990 through Phases I and II of the ALABC programme.

2.1. Improving cycle-life

The source of early limitations on life has been thoroughly studied and addressed directly. It has been shown [4,5] that the plate active materials in VRLA batteries need to be properly compressed, and attention to this requirement is rewarded by substantial improvements in life.

Projects have been initiated in Japan, Europe and Australia to develop improved separator systems that will maintain the positive active-material under the ideal degree of constraint while allowing good acid accommodation, good short-circuit resistance, and the avoidance of acid stratification. The research at the Japan Storage Battery (JSB) seeks [6] to develop improved cycle-life performance by exploring alternative materials in VRLA batteries of the absorptive glass-mat (AGM) design and by an improved approach to the construction of granular silica batteries. A problem with conventional AGM separators is that they tend to relax the force they apply to the active mass both when the material is wetted with sulfuric acid and when the batteries are cycled. The glass-free materials tested for AGM batteries in the JSB project performed less well than conventional separators when they were dry but performed better when they were wet. The granular silica product does not appear to relax at all.

The Australian research project at CSIRO [7] is also investigating two materials—a mixed glass-organic substance for AGM cells and a novel microporous separator for a high-compression gel cell. Early results look promising with high utilization of active material. In Europe, too, novel separator materials are being sought for flat-plate designs and also for improved gauntlets for tubular plates.

A number of ALABC projects have shown [8,9] that it is absolutely essential to charge the VRLA battery correctly in order to achieve significant life. There appear to be major benefits for cycle-life to be gained if the battery is recharged rapidly and if the degree of overcharge is restricted carefully.

A fundamental study at the University of Chicago is examining the consequences of fast charging in terms of the crystal structure and the microstructure of the active material. Progressive changes in the Pb_xO_2 stoichiometry, the lattice parameter ratio and the positional parameter of the oxygen atom have been observed. There is also an interesting progressive change in the shape of the lead atomic displacement ellipsoid. None of these changes, however, correlates closely with the end of life of the battery from which the materials were extracted. Nevertheless, there does appear to be a correlation with the change from a fine, needle-like crystal form at the start of life to a large grain size at the end of life [10]. The fine crystal form is sustained for more cycles in the case of fast charging than in the case of conventional charging. It is

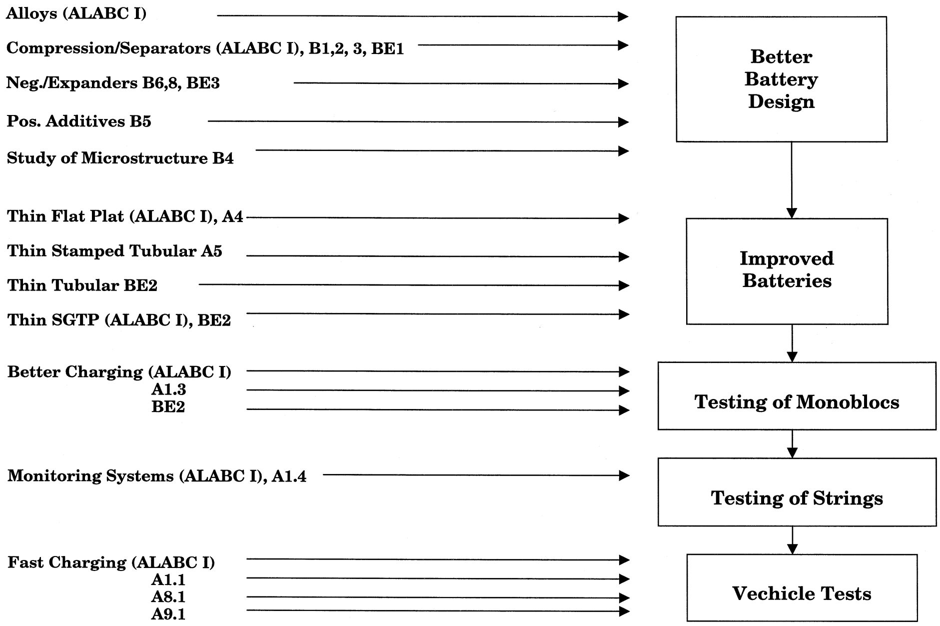

Fig. 2. Outline of main themes of the ALABC technical programme, 1997–1999. ALABC I indicates major advance made during ALABC programme 1993–1996. Other symbols refer to component projects within the present ALABC programme.

interesting to note that the electron energy loss spectrum of the fine needles (Fig. 3) is quite different from that of the coarser-grained material; this indicates a difference in electrical characteristics. During a later stage of this study, structural changes will be observed in situ by means of neutron diffraction from a lead–acid cell which is being charged and discharged within the neutron beam.

The importance of restricting overcharge was clearly demonstrated by a supplementary outcome from a project to develop a test VRLA battery in the European part of the ALABC programme. Although the battery met the design predictions for specific energy very closely, its cycle-life was extremely short and there was a correlation between the falling capacity and the increasing charge factor applied to the battery [9]. In order to assess the effect of the charge factor in the test employed, a commercially available VRLA battery was cycled under the same conditions (ECE15L discharge)—first with the charge factor unchecked and then with the charge factor pegged at 1.08. The results are shown in Fig. 4. These show a very much better performance for a string (14 monoblocs) cycled with a restricted charge factor. This result adds to a growing body of evidence that correct charging is far more important for VRLA batteries than for flooded counterparts. If sufficient attention is paid to this factor, then lives of many hundred cycles can be obtained (see Table 1 below).

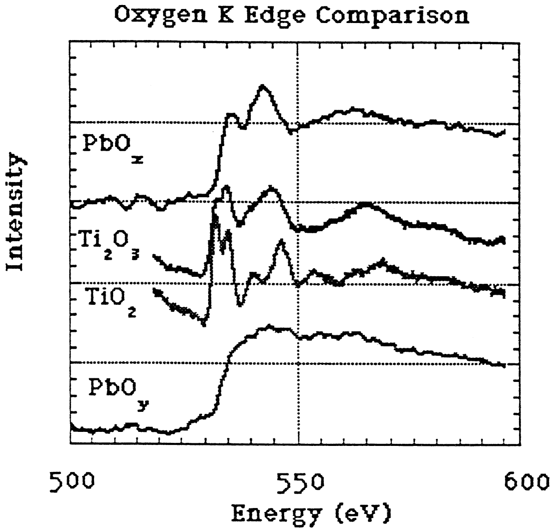

Fig. 3. Comparison of oxygen K edge from electron energy loss spectra of PbO_2 fine crystals (PbO_x) and large crystals (PbO_y). Spectra for TiO_2 and Ti_2O_3 are included as standards for reference.

As longer cycle-lives are achieved, particularly at high rates, it is increasingly being found that it is the negative plate, rather than the positive plate, that limits performance. Conventional (lignosulfonate) expander formulations are becoming a limiting factor. Accordingly, projects in Europe and in the USA have been placed to identify expander materials which will remain effective over longer periods of service. To date, some 34 materials have been evaluated for metal impurity content, acid stability, pH/solubility, and thermal stability. Eight materials, some natural and some synthetic, are being taken forward to more detailed testing.

2.2. Improved specific energy

The limitations of specific energy of the battery have also been tackled during the course of the ALABC's technical programme. Strong projects have been put in place to develop high specific energy by novel approaches

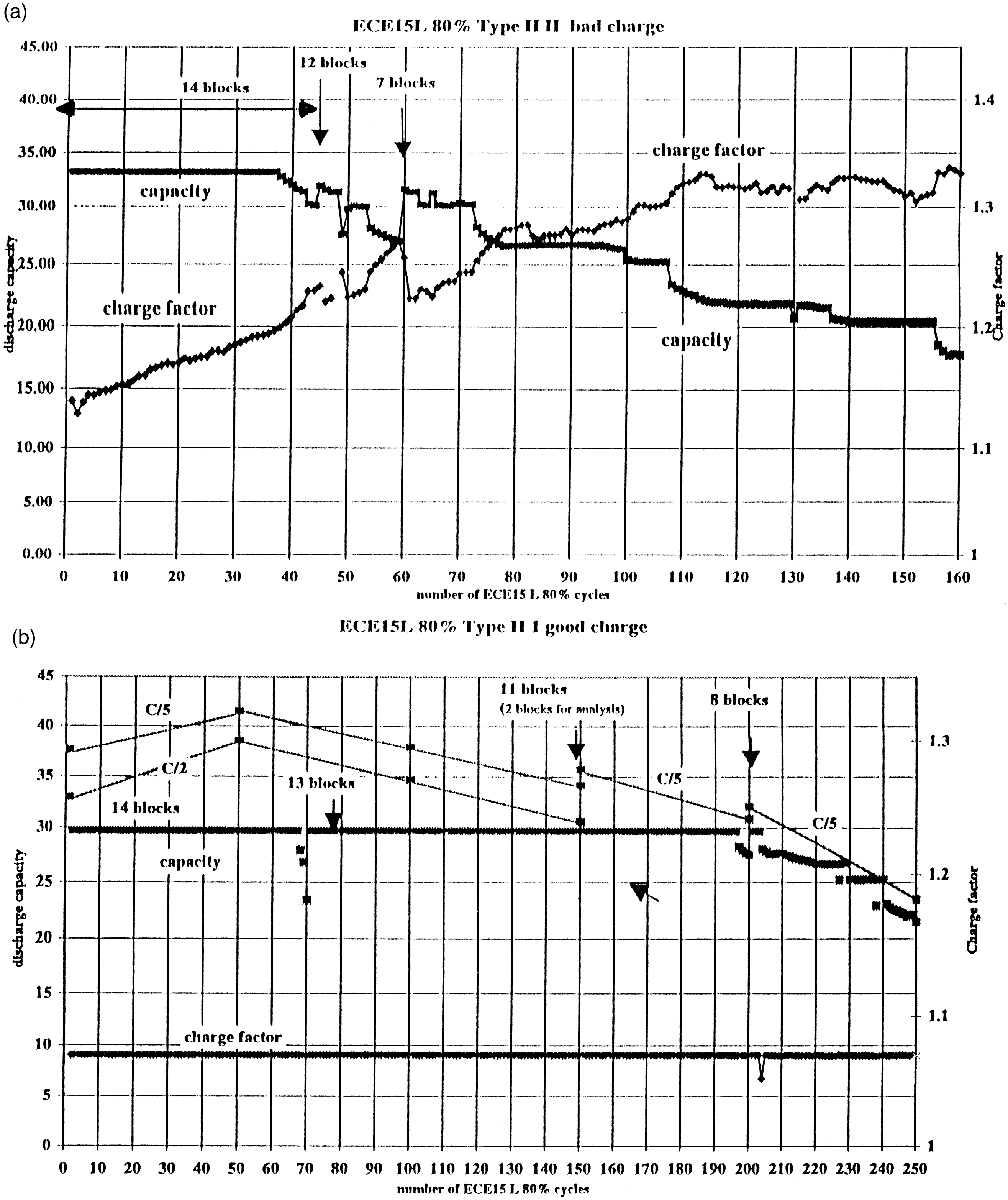

Fig. 4. Discharge capacity vs. cycle-life of strings of commercial batteries discharged under the ECE15L regime. In case A, the string (14 blocs) is charged without controlling the charge factor. In case B, the charge factor is constrained to 1.08.

to weight reduction. These are being carried out in the factories of major battery manufacturing companies. At East Penn, the use of very thin, flat plates, around 20% of the thickness of the conventional technology, offers substantial weight savings [11]. In another approach, at Yuasa [12] very thin, flattened tubular designs are being explored

Table 1
Effects of fast-charging on charge efficiency and cycle-life (50-A h battery)

	Slow	Fast
Charge scheme	5-h rate	12-min rate
Discharge scheme	at 2-h rate to 11.6 V (80% DoD)	at 2-h rate to 11.6 V (80% DoD)
After every 50 cycles	discharged to 10.5 V and fully charged for three cycles	discharged to 10.5 V and fully charged for three cycles
Charge efficiency	87%	97%
Cycles	250	900 +
Lifetime discharge (Ah)	10 000	36 000 +
	failed	still healthy

with plates (Fig. 5) prepared by stamping from thin foil which is rendered rigid and creep-resistant by a rolling process. In both instances, the technologies are being developed in a range of different variants in order to optimize the design. The first stages of the optimization process in the two projects will yield a product in 1999 and design calculations show an expected specific energy well in excess of what is currently available. Ultimately, it is likely that these initiatives will lead to specific energies approaching double what they were in 1990.

In support of the novel design projects, there is an extensive investigation of positive plate additives at the Trojan Battery [13]. This involves an evaluation of the most promising candidate materials available to date coupled with a theoretical study at the University of Idaho. The utilization of positive active-material in most of the cells containing additives is reported to be increased by at least 25% as compared with the controls. Cycle tests show capacities sustained well through 200 cycles without significant degradation.

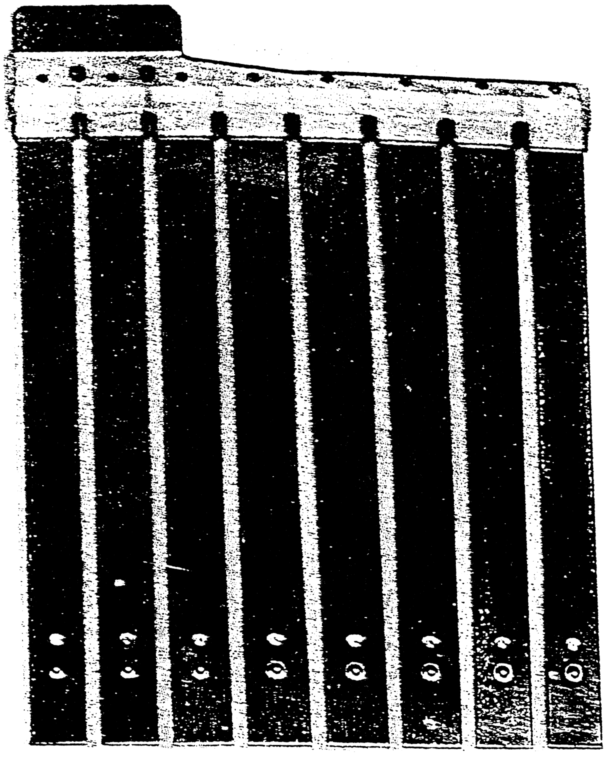

Fig. 5. Stamped, positive spines prepared for ‘flattened’ tubular plate design.

2.3. Recharge time

The capability to recharge rapidly impinges directly on the public attitude to the electric vehicle. It is widely accepted that most journeys for most people on most days of the year run for far less than 160 km. Any of the candidate battery systems should ultimately be able to satisfy this requirement. The major concern over range relates to those few occasions in the year when the driver wishes to journey further—250 to 500 km, for example. This requirement would only be satisfied by a system of rapid recharging. In a thorough study of all types of VRLA battery, it has been demonstrated [14] that 50% of charge can be returned in no more than 5 min. In fact, it has been shown that in some circumstances, the lead–acid battery actually benefits from the rapid recharging process. Table 1 shows an example of a comparative cycle-life test for a commercially available product in which conventional charging gives a life of 250 cycles, while fast charging leads to a life of over 900 cycles.

The importance of having fast charging available when required cannot be over-emphasized. The ongoing ALABC programme takes full account of the need for a complete control over battery-charging regimes, with several projects working in detail on rapid recharge and on partial-state-of-charge (PSoC) operation. One such project, carried out in Phoenix, has as its goal an evaluation of the relative importance of fast charging and PSoC operation in determining battery life. The project involves the testing of battery packs both in the laboratory and in vehicles over a range of different PSoC windows and at different charging rates, as shown in Fig. 6. An initial test of Hawker Genesis 12-V, 38-A h modules in an S10 pick-up truck has provided very promising results. The vehicle is being charged using a 150 kW Norvik Minute$^{®}$ charger at a maximum current corresponding to the 5 C rate. The vehicle is operated three to four cycles per day from around 20–80% depth-of-discharge. During the first 20 000 km, the battery received over 500 cycles of which 476 were at the 5 C rate. In addition to a good cycle-life, the fast-charge

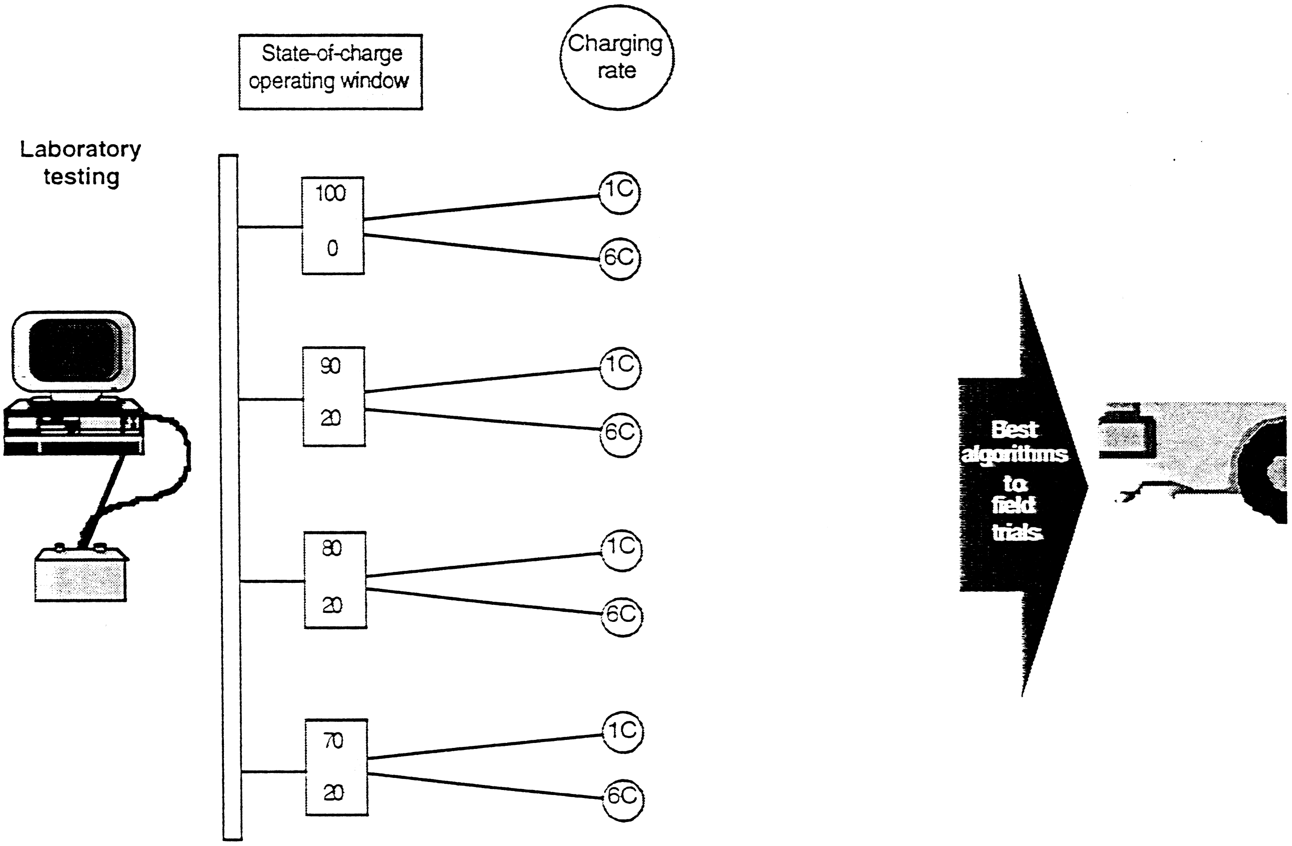

Fig. 6. Range of charge rate, PSoC range combinations to be tested in ALABC Project A-001.1.

regime has provided the ability to operate the vehicle continuously throughout a 24-h day. Throughout this period of testing, the phase composition and the BET surface-area of the active materials, as well as the rate of positive-grid corrosion, has been monitored. Fig. 7 shows the progressive evolution of BET surface-area for the

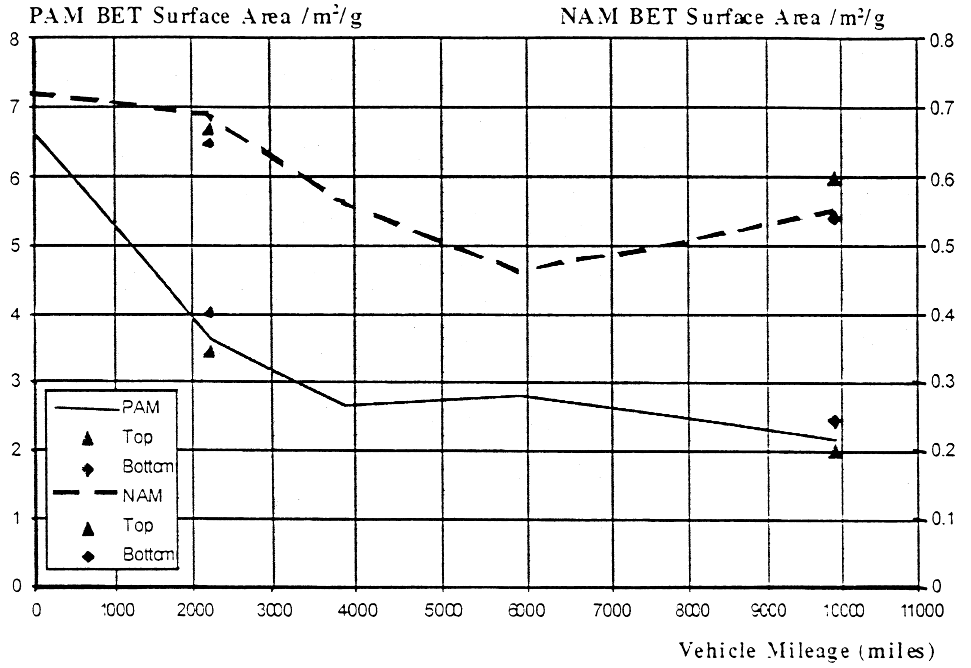

Fig. 7. Evolution of surface area of positive active-mass (PAM) and negative active-mass (NAM) with accumulated mileage in vehicle rapid-recharge test.

positive and negative materials through the first 16 000 km of the vehicle. The progressive decrease in surface area shown here is broadly in line with the results of the study carried out at the University of Chicago.

3. Conclusions

The improvements to cycle-life and specific energy involve substantial technical development in the way the battery is assembled, but are also intimately involved with the way the battery is charged. The fundamental mechanisms of the function of the valve-regulated variant of the lead–acid battery have been thoroughly studied, and their influence on the improved performance of the battery is beginning to be understood. One of the important factors is that high-rate charging produces high-surface-area active material. Another important point is that it is crucial to minimize the time during which the battery is in gassing mode rather than the amount of current that is passed during that time.

Improvements in the key parameters of the battery have been achieved through the course of the 1990s, as illustrated in Fig. 1. The initial values shown are a matter of historical record and the performance of the batteries for 1999 are the subject of ALABC projects, both in the laboratory and in vehicles.

In summary, it may be concluded that emerging VRLA batteries will be able to provide the electric vehicle with a range of 160 km per charge at a price which is likely to be well below those of other systems. The vehicle will be rechargeable in a few minutes so that on occasions when a range of more than 160 km is required, this will be accessible with minimum inconvenience. During the 1990s, the cycle-life of VRLA batteries has increased by a factor of 10 and the specific energy by a factor of around 2. Concomitantly, the charge time has been shortened by an order of magnitude.

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