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Life prediction of batteries for selecting the technically most suitable and cost effective battery

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Abstract

It is necessary to be able to predict the lifetime of a battery in target applications in order to make sound technical and commercial decisions at the system design stage. In general, accurate lifetime prediction requires more than knowledge of ageing processes and the availability of battery models. A concise procedure linking user requirements, operating regimes and operating conditions of batteries to ageing processes and loss of performance has to be used. Quantified end-of-life criteria have to be defined with the details of the application requirements in mind. To verify lifetime prediction models it is necessary to have data of the battery when new and immediately before replacement, results of post mortem analysis and detailed data of the operation. This paper describes a procedure that can be used for lifetime prediction, outlines some of the requirements for a prediction and discusses the principles of battery models and their potential use for lifetime prediction.

Keywords: Battery life prediction; Modelling; Simulation; End-of-life criteria; Electrochemical model; Equivalent circuit model

1. Introduction

Being able to predict the lifetime of a battery is of immense technical and commercial importance when planning systems, selecting the most suitable battery, determining the operating conditions and planning replacement intervals for batteries. A successful prediction requires knowledge of (a) the ageing processes within a battery which lead to a loss of performance, (b) the stress factors which induce ageing and influence the rate of ageing, and (c) an understanding of the relationships between the stress factors and the ageing processes. Detailed models which describe the performance of a battery are necessary to predict the lifetime, but not sufficient—even if the models are capable of simulating the performance of aged batteries.

Some of the most important differences between the investigation of ageing processes and the development of performance models on the one hand and lifetime prediction on the other are:

A. Lifetime prediction has to take into account the operating conditions of the application which often lead to a

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complex combination of stress factors and do not usually induce one dominant ageing process. As a result, all ageing processes that may occur in a battery will have to be considered simultaneously, including different ageing rates, and the effect of every ageing mechanism on one another has to be considered. Abusive operating conditions have to be taken into account if they are the result of proper planning for the most cost-effective system.

B. Lifetime prediction has to combine all effects into one figure of merit: (EOL), regardless of the severity of the individual ageing processes. Clear and unambiguous quantitative definitions for end-of-life have to be established. End-of-life depends on the nature of the application. Design parameters of batteries can change EOL significantly even if the ageing processes are quantitatively identical.

C. Lifetime prediction deals with the entire battery system, not cells, and has to include battery-related effects which are difficult to take into account when focusing on ageing processes on an electrode or cell level.

D. Lifetime prediction has to be done from the application and user point of view and needs to be decision oriented. Two frequently-asked questions are:
   - How long will a battery of a specific type and manufacturer last under certain operating conditions and how will the lifetime change if the operating conditions would be changed?
   - How long will a new design of battery or a battery with new material compositions last in a well-known type of operation?

   Lifetime prediction will only be a useful tool if these questions can be answered.

E. Manufacturers cannot test their batteries for the full range of applications in which the batteries are used. This is partly a question of time and expense, but also a question of lack of detailed knowledge concerning the use of the batteries in various applications. For renewable energy systems, for instance, there are very few complete data sets which describe the use of the battery fully from commissioning to final replacement. The real challenge of lifetime prediction, therefore, is predicting the lifetime of a battery under operating conditions for which there are no life time tests and for which results of existing lifetime tests cannot easily be transferred because the conditions during the test are not sufficiently similar to those in the application.

This paper will discuss these points, describe the requirements for making lifetime predictions for batteries and provide an outlook on some promising approaches.

2. Terminology

In this paper the following terminology will be used to make a clear description of the concept of lifetime prediction possible.

2.1. Performance, function and lifetime

The performance of a battery as regards a certain requirement (e.g. capacity, high rate discharge power at various SOC and temperatures, charge acceptance and self-discharge) can be measured and assessed using a scalar value such as Ah or time until a certain voltage limit has been reached.

A function, the ability to achieve a technically or commercially[1] desirable effect, can either be fulfilled or not, and the term state-of-function which is often used [1] should be considered to have either the value 1 or 0 when making lifetime predictions during the planning stage. If an internal combustion engine can start with the high-rate discharge power delivered by the battery in its application-typical state-of-charge and temperature, then the state-of-function is 1, if the engine cannot be started, the state-of-function is 0. State-of-function therefore includes the properties of the system as a whole and is not only focussed on the performance of the battery.

The battery has reached its EOL, if the state-of-function is 0.[2] This definition is true even in cases where the battery will be able to fulfil the required function again once its application-typical condition is improved, e.g. after recharging, a regeneration regime or modification of the operating conditions (higher temperature and SOC in the example of starting an internal combustion engine (ICE), cycling to remove acid stratification, etc.). It is presumed that end-of-life criteria for planning purposes can always be linked to a single performance value and that it is not necessary to know the exact change of voltage or current over time when using the battery.

2.2. Short-term performance prediction and long-term lifetime prediction for planning purposes

For safety reasons and for customer satisfaction it is of interest to predict the performance of a specific battery in the immediate future. A warning that the battery may no longer fulfil a certain function must come so early that the required action can still be taken, but for cost reasons it is unacceptable to issue a warning signal much too early. Also, early warning of imminent failure which turns out to be incorrect will lead to ignoring the warning signal in consumer products such as portable electronic equipment or in automobiles. Products which provide performance prediction, for instance for the

[1] An example of a commercially desirable effect is the ability of a battery to prevent the frequent start up of a diesel generator in a renewable energy system. The more frequent the diesel has to start up because the battery has lost its capacity or its charging characteristics have degraded, the higher will be the cost of electricity delivered. The economic function which could be used to determine EOL is: cost of energy before and after replacement of the battery. Technically the battery can be continued to be operated in the renewable energy system after its state-of-function as defined here has become 0.

[2] This definition presumes that the battery, when new, can always fulfil the required function.

back-up time in uninterruptible power systems or for starting of ICEs, are commercially available or are under development [2,3]. They usually rely on the measurement of the battery performance in the recent past and extrapolate to the likely performance and state-of-function under future conditions.

In this paper, such short-term prediction of performance for a specific battery for which detailed measurements under operating conditions are always required will not be discussed. Instead, the focus is the long-term prediction of lifetime as required for planning purposes, addressing issues of financing and likely replacement periods during the life of the system. To do this, the average properties of the battery (model, type and manufacturer) at the beginning of its lifetime have to be used and prior knowledge of their degradation under various conditions of use has to be available. Lifetime prediction as defined here is carried out at the planning stage for developing products and optimizing the operating regime.

2.3. User requirements, operating conditions and operating regime

The user requirements are power and energy demand, the necessity to store energy and restrictions on the installation conditions, such as ambient temperature. The user requirements determine the operating conditions current $I(t)$, voltage $V(t)$, temperature $T(t)$ and state-of-charge SOC(t) which is a calculated value. The operating conditions depend on the systems design, the selection of the size and type of the battery, the energy and time availability for recharging and the operating regime. Temperature is not only affected by heat generation within the cells but also by heat transfer to or from the cells and therefore the installation conditions. The other operating conditions are also affected by the installation conditions via the effect of temperature on the battery voltage. The operating conditions of a battery are also a function of the operating regime which includes the selection of the charging characteristics, voltage settings during charging and discharging, and special settings such as frequency of equalization charge or full charge.

Operating conditions can be abusive under certain conditions, but still reflect good planning principles and lead to the overall technically and economically best solutions. An example of abusive operating conditions within the context of good planning principles is the long time between full recharges from which most batteries in renewable energy systems suffer. Even if an optimal charge controller is used, the time between full recharges can be months and in certain installations a full charge with full conversion of all discharged material and creation of homogeneous acid concentration and state-of-charge of the electrode may never occur throughout the whole lifetime. For lifetime prediction, therefore, abusive conditions must be taken into account and it is not possible to ignore them on the grounds of poor planning.

2.4. Stress factors and ageing processes

Ageing processes are changes in the structure of the components of a battery or in the materials used in the battery. The changes lead to changes in the properties of the battery and are, under the normal constraints of operation, irreversible, e.g. corrosion and sulphation once it has accumulated over a long period of time. In flooded lead acid batteries, acid stratification can usually be reversed at any point in time and is therefore not considered an ageing process whereas acid stratification in batteries with immobilized electrolyte is irreversible and therefore an ageing process.

There are two different types of ageing processes: those which lead to slow degradation and a gradual loss of performance, and those which have no or virtually no impact on the performance until they suddenly lead to a major problem (sudden death). In cells, this is for instance the occurrence of a short circuit or dendrite formation, and in batteries, corrosion of the interconnection between cells.

Stress factors are statistical parameters or weighted scalar variables which are calculated from the time series of the operating conditions voltage, current, temperature and SOC and link the operating conditions to the lifetime of batteries observed in an application. Stress factors are, for instance, cycling under partial-state-of-charge, operation at high temperature and high charging voltage or long time intervals between reaching full charge.[3] These terms are normally used to describe empirically well-known damaging operating conditions but do not in themselves describe any irreversible changes of the battery components or materials. Stress factors only create conditions under which ageing processes occur and their rate (overall or at certain locations) increases.

Stress factors are therefore the result of user requirements, ambient conditions, operating regimes and battery design, and are a convenient way of describing damaging conditions which induce aging and/or increase the ageing rate. It should be realized that, under identical user requirements, ambient conditions and operating regimes, a VRLA battery, a flooded lead–acid battery, and a flooded lead–acid battery with an electrolyte circulation system will have a different temperature at the end of charging by virtue of their designs. Also, even if the same charging characteristics would be used, the voltage and current time series would differ and the resulting stress factors for these three types of batteries would be different.

Operating conditions, stress factors and ageing processes can be linked in a simplifying manner by using a matrix (see Table 1, [4]) which describes the effect of the operating conditions and stress factors on each ageing mechanism.

[3] For the development of models it is not necessary to use and define stress factors as all damaging conditions are completely described by the operating conditions. However, for discussion of damaging conditions it is convenient to use stress factors rather than the raw time series of voltage, current, temperature and SOC because they link the experience of what characterizes damaging operating conditions to the operating conditions.

Table 1
Qualitative description of the relationships between some stress factors and ageing processes [4]

Ageing processes (stress factors)	Corrosion of the positive grid	Hard/irreversible sulphation	Shedding (loss of material)	Water loss/drying out	AM degradation (reduction of surface)	Electrolyte stratification
Long time at low states of charge	Indirect through low acid concentration and low potentials	A strong positive correlation: longer time at a low SOC accelerates hard/irreversible sulphation	No direct impact	None	None	Indirect effect. Longer time leads to higher sulphation and thus influences the stratification
Ah-throughput	No impact	No direct impact	Impact through mechanical stress	No direct impact	Loss of active material surface, larger crystals	A strong positive correlation: higher Ah-throughput leads to higher stratification
Charge factor	A strong indirect impact because a high charge factor and an extensive charge is associated with a high charging voltage (high polarisation of electrode)	Negative correlation, impact through regimes with high charge factors which reduce the risk of sulphation	Strong impact through gassing	Strong impact	No direct impact	A strong positive correlation: higher charge factor leads to lower stratification
Time between full charge	Strong negative correlation: shorter time increases corrosion	Strong positive correlation: frequent full recharge decreases hard/irreversible sulphation	Negative correlation, increasing with decreasing time	Negative correlation	No direct impact	A strong positive correlation: higher Ah-throughput leads to higher stratification
Temperature	Strong impact, positive correlation	High temperature helps to more fully recharge the battery (more sulphate can be recharged). On the other hand, high temperature leads to more hard sulphate at a low SOC	No direct impact	Increasing with increasing temperature	Low impact high temperature degrades negative electrode expanders	No direct impact

For instance, a high temperature will accelerate the rate of corrosion, but will reduce the rate of formation of hard, irreversible sulphation products. Such a matrix does not take into account that ageing processes may have an effect on the magnitude of a stress factor—e.g. corrosion leads to lower mass utilization and therefore a higher Ah-throughput of the available active mass—and that the rate of an ageing process might be affected by the degradation caused by other ageing processes. Mathematically, therefore, a set of differential equations rather than a set of linear independent equations have to be used to describe the relationships correctly.

Table 1 shows the qualitative links between stress factors and ageing processes. Although only a few stress factors and their impact on the ageing processes are shown it is clear that stress factors can simultaneously reduce the rate of progress of one ageing process and increase the rate of progress of another. Quantifying these effects requires many simplifications and will only be possible if minor effects are ignored.

3. Description of lifetime prediction concept

The concept for lifetime prediction proposed in this paper is shown in Fig. 1.

The user requirements (load profile and energy availability over time), ambient temperature and installation conditions, operating regimes, battery design and size will lead to the operating conditions of the battery, the time series of voltage, current, temperature and SOC.

From the operating conditions, stress factors are calculated which describe those aspects of the operating conditions which are well-known to be linked to a reduced lifetime. In a recent paper [5], a number of stress factors have been defined for classifying batteries into categories of similar use, and the mathematical formula presented which were used to calculate the stress factors from the voltage, current, temperature and SOC time series of the battery:

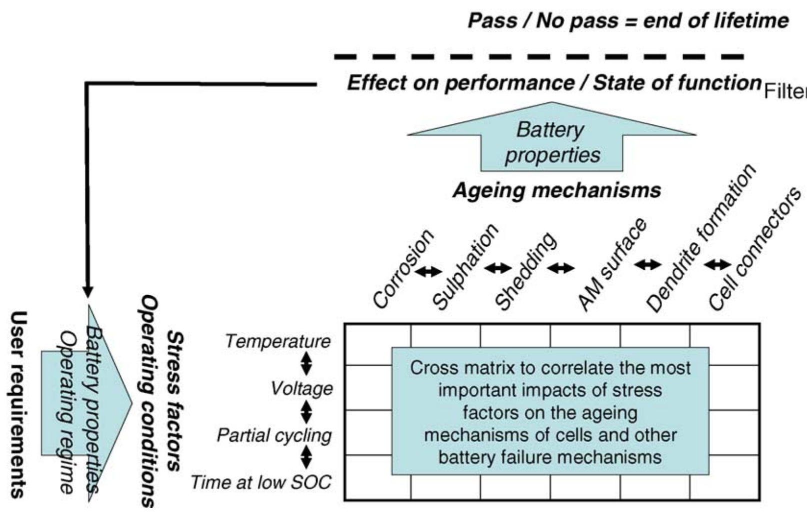

Fig. 1. Schematic diagram of the concept of lifetime prediction: The operating conditions of the battery and the stress factors which induce ageing processes and modify the rate of ageing are the result of the user requirements, operating regime and battery properties. Ageing processes affect the performance of the battery depending on its properties. Changes of the performance can be used to modify the operating regime and determine the end-of-life if a suitable quantitative criterion has been defined.

- temperature acceleration factor and low temperature factor;
- charge factor;
- cycling at partial-state-of-charge;
- Ah-throughput;
- average time between full charge;
- time at low SOC;
- partial cycling.

In addition, voltage is of course a stress factor, but calculating a single scalar value from the voltage time series of a battery which is used in cyclic operation with long time intervals between full charge can lead to results which are difficult to interpret. High voltage with the associated risk of corrosion is also contained in the stress factors charge factor and time between full charge. A publication containing a full discussion of stress factors used for a categorization process is currently under preparation [6].

When using battery models which use $V(t)$, $I(t)$ and $T(t)$ as inputs, it is not necessary to define stress factors because all the information is directly available from the data.

The interaction of operating conditions, stress factors and ageing processes is only schematically represented in Fig. 1 but shown in more detail in Table 1. It is obvious that a quantitative relationship between operating conditions, stress factors and ageing processes has to be limited to those with the greatest impact and even with this limitation is exceedingly difficult. The interdependence of ageing processes and the impact of ageing processes on stress factors is taken into account by the feedback loop linking performance changes to the operating conditions.

Ageing processes such as sulphation, shedding, loss of active material surface, corrosion and drying out of the electrolyte (for VRLA batteries) lead to irreversible changes in the structure and composition of materials of the components of the battery. Battery models capable of calculating the performance of the battery during the ageing processes can use exact end-of-life criteria as a filter which determines whether the required function can still be fulfilled. If not, the state-of-function is zero and EOL is reached. However, as discussed in Section 4.1, other battery models can also be used to translate the accumulated ageing processes into an end-of-life criterion.

The number of ageing processes contained in Fig. 1 can be extended to include “sudden death” ageing processes, such as an internal short circuiting, the result of poor maintenance, etc., by using stochastic models and probability functions which are dependent on all the other factors.

The speed with which the ageing processes proceed and performance values are reduced can be used to modify the operating regime of the battery for planning purposes or ultimately of course as a closed control loop for on-line process control. Although variations of the operating regime are limited, there are usually some parameters which can be altered in such a way that the ageing processes proceed more slowly.

The examples given so far have always been made with clear reference to lead–acid batteries. However, it should be pointed out that both the terminology and the concept of lifetime prediction are applicable to all types of battery chemistries.

4. Requirements for lifetime prediction

In this section the various requirements for making a successful lifetime prediction are discussed. Even if a different conceptual process of lifetime prediction is used, it is likely that the following considerations will have to be made, too.

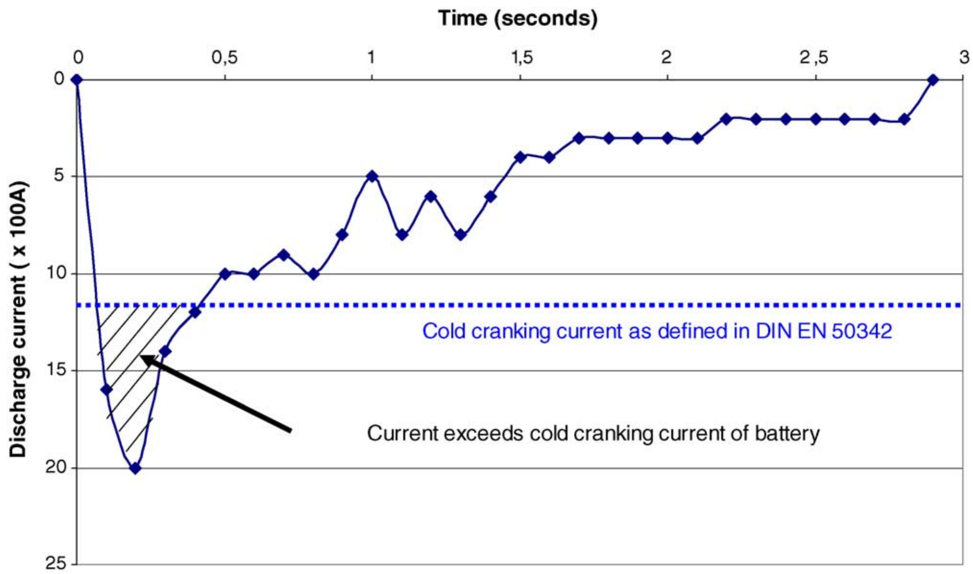

Fig. 2. Current of a battery (225 Ah at C_{20}) during starting of a railway traction engine. The cold cranking current of the battery as defined in DIN EN 50342 (current which the battery can supply at −18 °C for at least 10 s without the voltage dropping below 7.5 V; 1.25 V/cell) is also shown. The maximum current drawn exceeds the cold cranking current for approximately 0.2 s.

4.1. *Precise and quantified definition of end-of-life (EOL)*

Lifetime prediction is a projection of possibility or probability in a single figure of merit, end-of-life, which describes whether a particular function (e.g. starting an internal combustion engine or operation of a safety system for a certain duration) is possible or not at the desired moment. A quantified definition of when the state-of-function will become 0 and end-of-life is reached is therefore necessary. To do this, a threshold condition of the battery has to be defined in terms of state-of-charge, temperature, acid stratification, etc. and a performance value defined which can be linked to state-of-function. The process of starting an internal combustion engine provides a good example.

Fig. 2 shows the current and power of a battery during starting of a diesel engine of a railway traction vehicle under normal standby conditions. For a short-term performance predictions, the voltage is measured every time the engine is started and the voltage/time curve analyzed. By comparing this curve with parameters from engine start up in the past, a projection into the future at different temperatures and SOC can be made, see for instance [2,3].

For lifetime prediction for planning purposes, such measurements are not available and the performance value of the battery, which will enable starting of the engine, has to be defined differently. Fig. 2 also shows the cold cranking current which the battery used in this application has to be able to provide at −18 °C for 10 s without the battery voltage dropping below 1.25 V/cell (DIN EN 50342). Fig. 2 suggests that the engine will start if the battery can provide this current for only 3 s (disregarding the much higher current required for the first 0.2 s). A suitable end-of-life criterion for instance could be that the battery has to be able to provide its nominal cold cranking current for 3 s at the lowest temperature which has to be taken into account and at the lowest state-of-charge and the most unfavourable conditions that will exist during operation.

The use of such an application-oriented end-of-life criterion may appear unwieldy; however, it leads to decisions on the probable lifetime of the battery which closely reflects the technical and financial requirements. The usual criterion for EOL used in many tests, i.e. capacity under nominal conditions has dropped to a certain percentage, will lead to very similar results if it is assumed that the performance values of the battery will drop very quickly once the value used in the test has been reached. This, however, cannot be automatically assumed to be correct. In the Qualibat project [7], lifetime tests have been carried out where the battery capacity which was used as performance value for lifetime decreased more or less linearly for VRLA batteries (see Fig. 3) and an

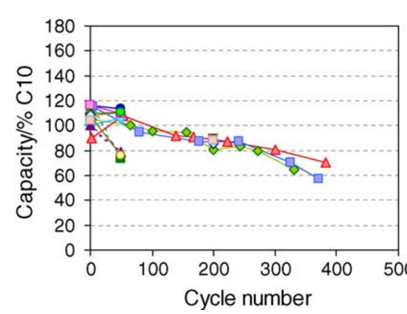

Fig. 3. Capacity loss of VRLA batteries as a result of cycling to a DOD of 66% at C_{10} [7]. A linear capacity loss is observed for most of the batteries which were tested (15 in all).

EOL criterion of 60% versus 80% doubles the lifetime of the battery.

Other frequently used end-of-life criteria such as total Ah-throughput, total number of cycles or years of operation do not take any application-specific requirements into account. They are based on standard lifetime tests and presume that the operating condition in the application can be linked to the conditions of the tests and that the same EOL values apply. This obviously is not a very accurate and application-specific criterion for EOL.

4.2. Lifetime prediction and lifetime tests

Lifetime prediction is relatively simple if there is either a very good empirical link between test results and lifetime achieved or there is a close similarity between the test procedure used and the operating conditions in the application. Close similarity is, however, difficult to establish when using accelerated tests. A good empirical basis exists, for instance, in the car industry for conventional SLI application. The way SLI batteries are tested and the performance values that have to be achieved during operation cannot easily be linked to the normal operating conditions in a car. However, experience has shown that batteries which perform well in tests also achieve a lifetime which customers find satisfactory. Lifetime prediction providing information on the lifetime of different batteries in different usage patterns of vehicles is not possible but also not required.

If the operating conditions of the battery in its application are very similar to life tests which have been carried out in the laboratory, then predicting the lifetime of a battery is also reasonably straightforward using simple extrapolation. However, such cases are rare and probably only float operation of batteries in uninterruptible power supplies and systems with a battery stabilized dc intermediary circuit allow accurate prediction of lifetime with the use of the results of lifetime tests. Another case that seems to allow a straightforward use of lifetime tests is cyclic operation of batteries with frequent full recharges, such as those found in forklift trucks. A closer look, however, reveals that the operating conditions of forklift truck batteries, such as depth-of-discharge, charging conditions as a function of temperature and depth of discharge, average discharge current, variations of discharge current and rest periods at low SOC vary greatly. It is by no means clear what the impact of these operating conditions is on the lifetime of the battery.

In applications such as renewable energy systems, normal test procedures carried out to characterise the battery provide only limited information on the suitability of the battery because the conditions of use in the application and the test procedure differ too much. Only dedicated test procedures have the potential to provide meaningful results. Work in the Qualibat project [7] which has devised test procedures which cause failure of batteries from predominantly one ageing process only is a first step to make lifetime predictions possible for batteries in applications where failure usually occurs from only one ageing process. However, as soon as there is a number of ageing processes which together lead to the failure of a battery, this approach has its limitations. Fig. 4 schematically shows the problem of linking test procedures, for which lifetime tests exist, to the range of operating conditions which are to be investigated and the uncertainties of extrapolating from the results.

Battery testing takes a long time, and the use of accelerated tests raises concerns about the lack of similarity between test procedures and operating conditions of the application, where the range of operating conditions is large. Although experience can additionally be used to enhance prediction methods, this requires knowledge of many well-documented and comparable installations. As a result, the need for lifetime prediction methods to interpolate between test results and extrapolate outside the tested range remains and cannot be substituted even by a large test program.

4.3. Verification of lifetime prediction

Verifying lifetime prediction is very difficult because a number of data sets from many different installations are nec-

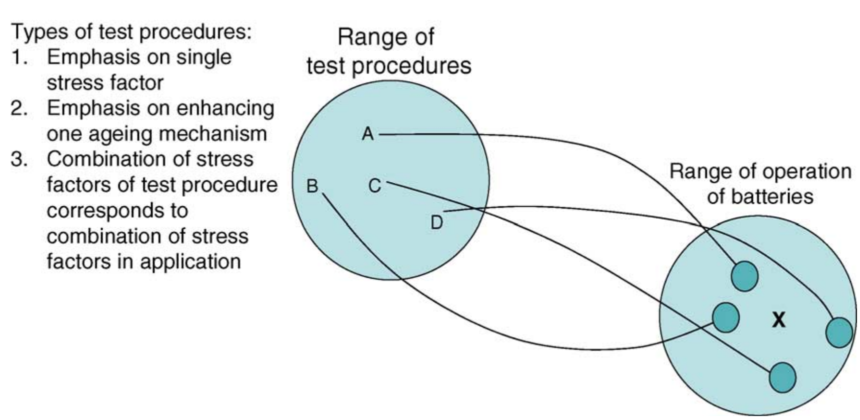

Fig. 4. Schematic diagram illustrating the link between test procedures and operating conditions. How to extrapolate between test procedures marked A, B, C and D to the operating conditions marked X is still problematic.

essary and they are difficult to obtain: battery properties (in particular capacity) at the beginning of installation, operating conditions during the application and a determination of the battery properties once the end-of-life has been reached. A track record of calculating from a range of different field data the correct lifetime of a number of batteries with their individual characteristics or, as a lower requirement, calculating the correct order of lifetimes that have been achieved by a number of different batteries will then allow predictions for planned systems:

- lifetime of a well-characterised and known battery for a planned system with sufficiently well-defined future operating conditions;
- effect of changes in the operating regime on the lifetime expectancy, e.g. changing the charging characteristics and voltage thresholds or the frequency with which a full charge will be carried out;
- lifetime of a newly developed battery for which there exists no track record.

Verification is of course only possible if the variation of battery properties and operating conditions have a certain range so that interpolation and extrapolation is possible. If all data for verification are only for one type and manufacturer of batteries, then extrapolation to another battery is possible, but risky and certainly not verified. The same applies for extrapolation from one cell to a battery because the increasing differences between cells during ageing are highly relevant for reaching end-of-life.

5. Lifetime models

Lifetime prediction is impossible without models which interpolate and extrapolate between the results of lifetime tests. There are two completely different approaches:

1. Models based on simulating the change of performance values of the battery while the various ageing processes take place. These models will be called performance-based lifetime models and are potentially very accurate for making technical and financial decisions.
2. Models which link the end-of-life of a battery to some parameters which can easily be determined such as Ah-throughput, number of cycles and time since manufacturing. Once a predetermined value of the parameter has been exceeded, the battery is considered to have reached its end-of-life. These models will be called cycle counting or weighted Ah-throughput models and are inherently limited in their accuracy. However, they are the only models which are now available and most planning tools which incorporate lifetime models use them.

This section will discuss these two different approaches in the light of their suitability for lifetime prediction. A publication for a detailed discussion of the models is planned.

5.1. *Performance-based lifetime models*

Most of the performance values used to define end-of-life can be modelled for a new battery. Usually, a few tests are sufficient to parameterise the models and then the voltage, current, power output or uptake, state-of-charge, etc., can be predicted at different current profiles and temperatures. The Shepherd model [8] is one of the first of such models to simulate the voltage during discharging and charging.

Limited accuracy of these models is less a problem than the difficulty of handling the performance of a battery once ageing processes have started to influence battery performance noticeably or once severe inhomogeneity has been induced by partial cycling. The so-called 17.5% test of Volkswagen AG [9], for instance, leads to a reduction of the amount of Ah recharged once stratification builds up. Although the capacity after a full recharge does not change in the first few cycles, it is obvious that performance values such as cold start cranking and residual capacity are very difficult to model during such operation.

When the battery is new, the values P_i describing the performance requirement i can be written as a function of the operating conditions: $P_i = f_i(T, U, I, \mathrm{SOC})$.

Once the operation of the battery over time has led to inhomogeneity, this general expression has to be modified to include the history of the battery since the time t_1, when it was last fully charged and electrolyte and electrodes were in a homogeneous state, and the time t_2 at which the performance value has to be calculated: $P_i = f_i(T(t), U(t), I(t), \mathrm{SOC}(t))$. Alternatively this relationship can be written as $P_i = f_i(T, U, I, \mathrm{SOC}, S_j\ (j = 1\text{–}J))$ with J being the number of stress factors to be considered and S_j some numerical value which describes the inhomogeneity induced, mainly acid stratification and inhomogeneous state-of-charge of the electrode. If inhomogeneity is removed quickly, the impact on ageing processes can be ignored. “State-of-inhomogeneity” is therefore independent of ageing.

An example of the importance of including inhomogeneity is shown in Fig. 5. The voltage, current and Ah-balance of a starter battery are shown for the first 12 h of a 17.5% test and, ca. 77 h later, for the last 12 h of the test. After an initial increase in the Ah balance achieved at the end of each charging phase, the Ah-balance begins to fall again although the time for charging is longer than the time for discharging. At the end of the first such test, the battery has not suffered any capacity loss. Post mortem analysis at the end-of-lifetime reveal, however, severely different Ah-throughput in the different regions of the electrode. When calculating the performance of a battery in real applications with severe cycling at partial-state-of-charge, the effects of such severe inhomogeneity have to be taken into account.

Once ageing proceeds and the structure of the battery components and its materials undergo irreversible changes, the relationship has to be written as $P_i = f_i(T(t), U(t), I(t), \mathrm{SOC}(t), A_k\ (k = 1\text{–}K)$ with K being the number of ageing processes that have to be considered, or $P_i = f_i(T, U, I,$

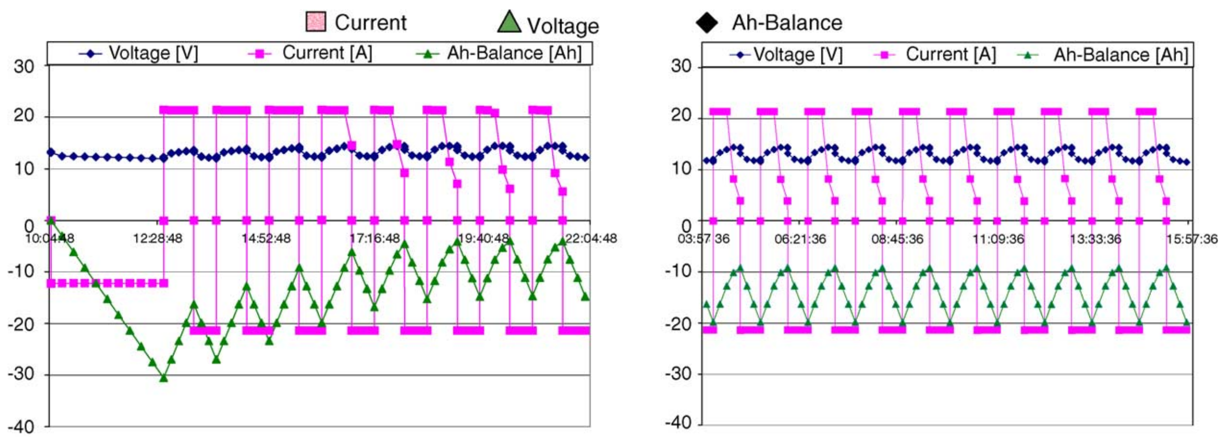

Fig. 5. Results of the so-called 17.5% test [9] of a 61 Ah, 12 V starter battery. First 12 h of the test at the left and, approximately 77 h later, last 12 h of the test at the right. Although the charging time (40 min) is one third longer than the discharging time (30 min) the voltage limit of 14.4 V during charging leads to a prolonged period of cycling at partial-state-of-charge. The Ah-balance reaches its maximum after seven charging events and then decreases slowly. Post mortem analyses of batteries cycled in this manner have revealed severe acid stratification and inhomogeneous distribution of state-of-charge on the electrode. Data courtesy of Akkumulatorenfabrik Moll GmbH + Co. KG, Bad Staffestelin, Germany.

SOC, S_j ($j = 1$–J), A_k ($k = 1$–K)) as inhomogeneity has to be considered in addition to ageing.

As ageing of batteries is usually accompanied by growing differences between cells which can lead to increased stress factors and accelerating ageing processes, it is important to take such effects into account as well.

Models which are capable of simulating performance and including inhomogeneity and ageing processes can be broadly distinguished into four groups.

5.1.1. First-principle electrochemical models

These models incorporate many factors including kinetic and mass transport properties, thermodynamic properties, mechanical, thermal and electric properties of materials and dimensions. Fig. 6 shows a schematic diagram of the interaction of such models. Inhomogeneity and ageing processes can be represented by the changes of the properties or dimensions. Mathematically, the relationships between operating conditions and loss of performance are expressed as a set of differential equations. Typical examples are, for instance, the models by Caselitz [10], Liaw et al. [11], Gu et al. [12] and Sauer [13,14]. Although the models are sometimes explained using an equivalent circuit diagram, they should be distinguished from models based explicitly on equivalent circuit diagrams.

5.1.2. Equivalent circuit diagrams

Every component of the battery which contributes to the transport of charge carriers or generation of a voltage drop is represented by a component of an equivalent circuit diagram: voltage and current sources, resistors, capacitors and inductances. Inhomogeneity and ageing processes are rep-

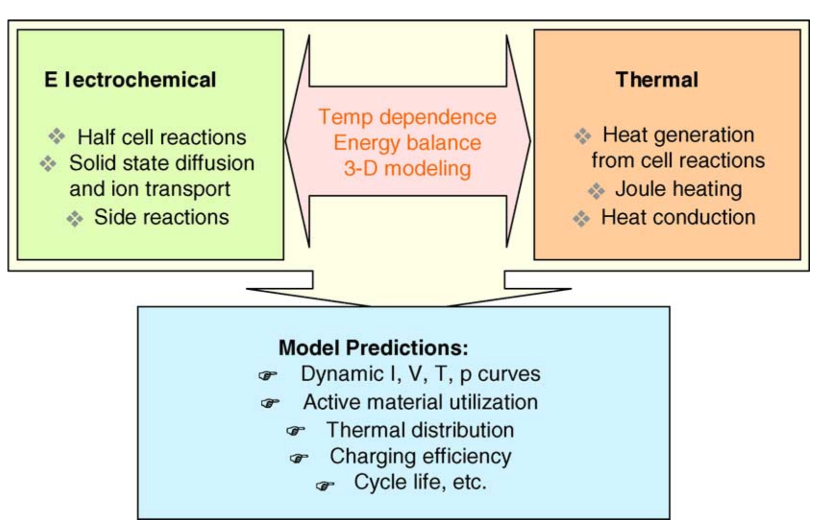

Fig. 6. A schematic diagram showing a “first-principles” electrochemical model that combines reactions and transport kinetics and coupled with thermal phenomena in an electrochemical cell to predict battery performance [12].

resented by the changes of the values of the components of the equivalent circuit diagram. Many one-dimensional models have been proposed, and very early Euler [14] has proposed a model to investigate the spatial distribution of the current in a new battery. More recently, an equivalent circuit diagram including spatial distribution modelling has been used by Sauer [15] to investigate electrolyte stratification [16].

5.1.3. Analytical models with empirical data fitting

Interpolation and extrapolation from test results and field data can be used for lifetime prediction by means of parameter fitting. The Shepherd model [8] belongs to this class of models. Where there is a wealth of data and the applications are reasonably uniform, this approach might be successful. Where there are only few data and the similarity between test results or field data and the application under investigation is unclear, this approach is not possible.

5.1.4. Artificial neural networks (ANN)

ANN (see for instance [17] have a tremendous potential to discover relationships between inputs (here operating conditions and stress factors) and outputs (here ageing processes and performance values) and do not rely on a detailed understanding of the mechanisms which link input and output. For lifetime prediction these models can only be used where there are sufficient data to train and verify the ANN. For models predicting the short-term performance, this problem can be overcome. For lifetime prediction, many more data sets than now available are required. Aspects of an artificial neural network can be used for any of the above models.

Spatial differences and temporal changes can be modelled well using "first-principle" and "equivalent circuit models" if they have been devised in a suitable manner.

Details of these models, results, potential and limitations that may exist will be discussed in a paper which is under preparation.

5.2. Cycle counting or weighted Ah-throughput models

5.2.1. Cycle counting models

All of these models describe the lifetime of a battery in terms of a fixed magnitude (e.g. Ah-throughput or time) which is used up during use. A battery operated in float operation at a certain voltage and temperature for which lifetime tests predict a lifetime of 10 years, i.e. 3650 days, is considered to lose 10% of its lifetime each year. This is a generally used approach for lifetime prediction and can be formulated mathematically as "Proportion of lifetime which has been used up = $n \times 1/N$" with N being the number of events which can occur during the lifetime and n the number of events which have already occurred. For float operation, $N = 3650$ days for a battery with 10 year lifetime and n is the number of days that have already passed. For cycling operation, N is the total Ah-throughput of the battery achieved during cycling until the battery fails (e.g. number of full cycles multiplied by the nominal capacity C_N) and n denotes the Ah-throughput that has already occurred.

In mechanical engineering, this approach is used to determine the lifetime of components from bridges to aeroplanes and is extended to cope with different types of events E which occur sequentially and can all happen very often, i.e. N_E, the number of events E which can occur until the component fails, is very large (for a recent review see [18]).

A component will fail if $\sum n_E \times 1/N_E$ (sum over all events E) = 1 or the lifetime that has been used up is $L \times \sum n_E \times 1/N_E$ with L being the lifetime of the battery which can be expected under the combination of stress events E occurring n_E times each. As each stress event E is also associated with a certain duration t_E, the life expectancy under different combinations of stress events E can always be calculated.

Applying this approach to batteries requires the same assumptions as those made in mechanical engineering:

1. It is possible to define stress events which induce only a small amount of incremental loss of lifetime and thus can occur very frequently until the battery fails.
2. The loss of lifetime caused by a stress event does not depend on the previous stress event (for batteries this means that the end of a stress event has to be defined as full charge and homogeneous acid concentration and state-of-charge of the electrodes) and is independent of the accumulated loss of lifetime that the battery has suffered so far.
3. The total loss of lifetime caused is either independent of the sequence of the stress events or the stress events E which occur are distributed statistically throughout the lifetime of the battery, i.e. the battery is not operated first at float operation for half of its lifetime and then subsequently cycled for the other half of its lifetime, but float operation and cycling happen more or less alternatively.

Cycle counting methods—despite making some very crude and simplifying assumptions—are used in a number of planning tools which are used to simulate systems and to estimate the lifetime of the battery, such as Hybrid2 [19] or PVSyst [20]. A description of how this simple approach may be used without unreasonably simplistic assumptions will be described in a second paper.

5.2.2. Weighted Ah-models

Weighted Ah-throughput models are also based on the assumption that during use of the battery, the lifetime is used up proportionally to the total Ah-throughput. However, these models use a modified approach compared to the cycle counting model and take into account that certain operating conditions may lead to an increased rate of ageing whereas others may lead to a decreased rate of ageing. The equation $\sum n_E \times 1/N_E$ (sum over all events E) = 1 above is therefore modified and an effective Ah-throughput is calculated: $Ah_{eff} = \sum w_E \times n_E \times Ah_E$ (sum over all events E) with Ah_E being the Ah-throughput of an event E, n_E the number of events E and w_E the weight or severity associated with the event E. The battery is considered to fail once the effective Ah-throughput exceeds the total Ah-throughput of the battery. The effective Ah-throughput can be smaller or larger than the total Ah-throughput.

In a paper by Drouilhet and Johnson [21], this approach has been used to select the type of battery (NiCd or lead–acid) for a renewable energy system in Alaska where the battery was expected to be operated at reasonably high currents (I_1 range) for 30 min minimum time of autonomy. The weighting factors were based on depth-of-discharge and discharge rate and estimated from general principles and test results publicly available on the data sheet.

Puls et al. [22] have developed a model with PV systems in mind. The weighting factors used are temperature, time between full charge (to take growth of sulphate crystals and risk of sulphation into account) and a parameter based on the lowest SOC reached during cycling at partial-state-of-charge (to take the formation of acid stratification into account). The weighting factors were calculated using a parameter fit.

Another example for the use of this model is the lifetime prediction for forklift trucks used by the German battery industry association ZVEI [23]. The battery is considered to have a certain lifetime given in month which depends on the average discharge rate, the average daily Ah-throughput and the average temperature. The result is a prediction of lifetime in months.

It should be pointed out that none of these models has so far been validated.

The details of these models will also be discussed in the second paper.

6. Conclusions

Lifetime prediction is still at an early stage for applications where the variation of operating conditions is large and there is limited experience concerning the lifetime achieved in field experiments.

The availability of powerful software and computers has led to recent progress in model development where more and more effects leading to inhomogeneity and ageing are taken into account. However, the models can only be used for making technical and commercial planning decisions once they have proven their capability to predict the lifetime of a few different types of well-characterised batteries in well-documented applications or at least the order in which the batteries have failed. Specially developed test procedures are required in the beginning for verifying the models, but ultimately, the comparison with results of well-monitored field installations is necessary so that the models can be used for making sound technical and commercial decisions.

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