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Simple model for the overcharge reaction in valve regulated lead/acid batteries under fully stationary conditions

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Abstract

A model for the float charge of valve regulated lead/acid batteries is presented. The model relies on a description of the intrinsic kinetics of hydrogen and oxygen evolution, and corrosion. The mass transport is characterized by a single parameter. An important result is that polarizations are highly dependent on both the hydrogen and oxygen overvoltages. The model enables the calculation of the minimum float charge voltage.

Keywords: Valve regulated lead/acid batteries; Lead/acid batteries; Overcharge reactions

1. Introduction

Stationary valve regulated lead/acid batteries are continuously charged at a voltage slightly above the open-circuit voltage (OCV). In spite of their widespread use and great commercial importance, the fundamental principles of their operation are still poorly understood. This paper presents a simple model that can explain some of the features observed in modern valve regulated lead/acid batteries.

Stationary lead/acid batteries are used as the emergency source of energy in telecommunication's power plants. The batteries are connected in parallel with the load and the rectifier. The voltage level for charging has to be carefully chosen both with respect to the load and the battery. Low voltage may lead to poor charging of the battery while high voltage will lead to higher corrosion and water loss.

In valve regulated batteries, the electrolyte has been immobilized either by turning it into a gel by the addition of thermal silica or by the absorption in a non-woven glass fibre mat [1]. The most important features of valve regulated batteries are position-independent operations, no need to refill with water, and greatly reduced gas emissions. The greatly decreased gas emissions and water-loss rate are results of a process, recombination, by which the overcharge current can pass without electrolysis of water. Recombination occurs when oxygen is transferred from the positive to the negative electrode where it is rapidly reduced electrochemically. Recombination occurs both in batteries with fully liquid electrolyte and those with immobilized electrolyte. In batteries where the electrolyte is immobilized, recombination can be increased greatly by creating passages of gas through the electrolyte. In general, these gas passages are created by limiting the total volume of electrolyte between the plates.

Modern valve regulated batteries have good recombination, only a small fraction of the float charge current, 1–10%, is spent in the electrolysis of water. Likewise, the float charge current is very low, 10–50 mA/100 Ah, which leads to a very low rate of water loss. Diffusion of water through the polymeric battery box material is the dominant cause of water loss [2].

Different ideas have been put forward to explain the efficient operation of valve regulated batteries. Early ideas involve the view that the negative electrodes should have a larger capacity than the positive ones to prevent the negative electrodes from becoming fully charged and thereby preventing hydrogen evolution. Other ideas involve lead sulfate as a necessary intermediate in the oxygen recombination process. A high internal pressure as a prerequisite for recombination to occur is also an idea that has been put forward. One of the original designs for a sealed lead/acid battery was based on a high working pressure, 4 bar approximately. Modern designs use a low venting pressure, around 0.1 bar, but still recombination is efficient. Recent studies show that the negative electrodes do not need to be oversized for recombination to be efficient, nor are there are any indications pointing to lead sulfate as an intermediate product [3]. Today, there is a consensus that the recombination is set up primarily as the result of the creation of oxygen passages between the plates and that no overpressure is needed.

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In a valve regulated lead/acid battery the float charge voltage is slightly higher than the OCV. The OCV is very close to the equilibrium voltage because of the very low self-discharge rate in the lead/acid system. The float charge voltage needs to be higher than the OCV in order to have a reasonably fast charge and to fully compensate self-discharge. Studies indicate also the existence of an optimal potential for the positive plate [4,5], where the corrosion is at a minimum. A theoretical study of the lead/acid battery at typical float voltages was first made by Milner [6]. This study dealt exclusively with flooded lead/acid batteries. Miller introduced some ideas that have proved to be useful:

- Tafel behaviour of the overcharge reactions;
- apparent overvoltages of overcharge reactions substituted for true overvoltages;
- constant corrosion rate for the positive plate;
- currents related to capacity, and
- limiting current for oxygen reduction on the negative electrodes.

For the actual values of the parameters Miller [6] relied on work performed by Willhinganz that was unpublished when Milner's paper was published. Willhinganz [7] published later a paper in which the methodology of extracting the critical prarameters was explained. Milner's paper concentrated on the question if the normal float voltage would be sufficient for healthy operation of a battery taking into account factors such as spread in exchange current densities, limiting current for oxygen reduction, and temperature excursion for the whole battery as well as temperature variations within the battery. A factor of 2*X* was assigned as span for the exchange current densities. It was established by this model that a cell with an extreme behaviour in a string of 11 normal cells would not be properly charged at the standard float voltage. For a cell with a temperature of 35 °C with other cells at 25 °C the analysis showed that this cell would not float properly. Subsequent field trials proved Milner's model to be quite accurate [8].

When the valve regulated lead/acid batteries were introduced various concepts to explain their behaviour were put forward such as the ideas mentioned earlier in this text. A common idea of these explanations was that the negative electrodes should be at their open-circuit potential (OCP) and remain essentially unpolarized for the recombination to be effective [9–11]. Opinions that a completely unpolarized negative would mean elimination of water losses through electrolysis were opposed by Berndt et al. [12]. Their analysis showed that the water loss can never be lower than the water loss experienced at OCP of the negative electrodes. Subsequent descriptions of the operation of valve regulated batteries have clung to the opinion that the negative electrodes should remain at open circuit for recombination to be efficient. A constant limiting current density for oxygen reduction is assumed. This reasoning leads to a dilemma between the older view that a negative electrode should have a small polarization to stay charged and the view for valve regulated batteries where the negative electrodes should remain at open circuit. The first study to address this dilemma was published in 1994 by Teutsch [13]. His paper presented a model of the float charge behaviour of the valve regulated lead/acid battery. In this model, a high oxygen recombination rate was introduced by defining a limiting current for oxygen recombination, I_{lim}. It was assumed that below this current, the oxygen recombination would stay close to 100%. A parametrical analysis showed that the negative electrodes would go to OCP at a normal operating voltage of 2.27 V per cell when the apparent exchange current density for hydrogen evolution, I_{H0}, was approaching that of the oxygen evolution. A weakness in the model is the assumption of a constant oxygen recombination. It can be argued that this assumption cannot hold for any lead/acid battery if a steady state should prevail. As pointed out by Berndt et al. [12] and also by Teutsch [13] there must be venting as the result of the corrosion of the positive plates. The corrosion reaction will create an imbalance which leads to a surplus of hydrogen evolution. In the gases, within the battery, there must be a finite concentration of oxygen and hence some electrolysis gases will be vented. The model also fails to explain the steep Tafel slopes that many valve regulated cells have. Cell Tafel slope of 190–220 mV/decade have been observed when at the same time recombination has been over 96% [2]. Data from Calasanzio and McClelland [9] also give a good illustration to this phenomenon. They observed a valve regulated battery at elevated temperature of 50 °C. This battery type has been charged at different voltages. Fig. 1 shows the recombination for the three different charging voltages. The recombination increases during the test but is approximately the same for the three charging voltages at each period. As can be seen from Fig. 2, the Tafel slope for the cell is very high. Recombination may in fact be higher than in Fig. 1, because some of the water losses is probably the result of direct dry-out. The behaviour of this battery seems to be highly desirable. The recombination is high inspite of the extreme operating conditions. The Tafel slope is high which should indicate that the battery should be quite resistant to 'thermal runaway'. Because of the high Tafel slope for the cell overcharge the total overpotential should be divided

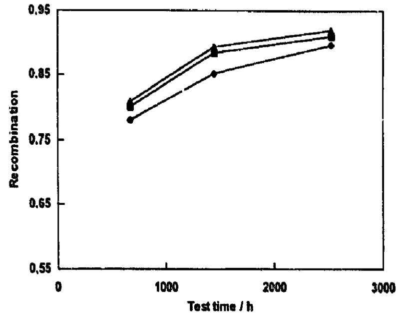

Fig. 1. Recombination for a valve regulated battery at different voltages at 50 °C: (▲) 2.46 V; (◆) 2.37 V, and (■) 2.27 V (from Calasanzio and McClelland, Ref. [9]).

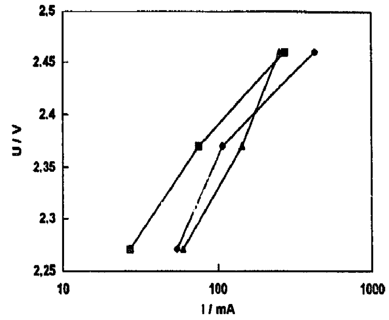

Fig. 2. Overcharge characteristics of the valve regulated battery in Fig. 1, after (■) 28 days; (♦) 60 days, and (▲) 105 days.

between both the positive and the negative plate. This behaviour cannot be explained with the assumption of a limiting current for oxygen reduction.

Therefore, we suggest the following simple model for the overcharge reaction of the valve regulated lead/acid battery.

2. Model

2.1. Assumptions

The overcharge reaction's hydrogen and oxygen evolution follow Tafel equations and their kinetic relations are expressed relative to the OCVs of the lead and lead dioxide electrodes, respectively. The battery is assumed to be tight except for the valves which may open and let out gas. Hydrogen diffusive leakages are encountered in valve regulated batteries but as they are quite small are not taken into consideration [2]. Hydrogen oxidation on the positive electrodes is assumed to be negligible. Earlier research information [14–16] indicates that some oxidation may take place but the rate is, in general, very low and does not influence the potential.

An even current distribution is assumed, likewise an even gas compositon. Corrosion will be characterized by two different relations.

Instead of a limiting current density for oxygen reduction (recombination) we assume a constant mass transfer rate, characterized by the parameter k.

2.2. Model equations

We set up mass balance for the removal and production and venting of oxygen within the battery:

(i) current balance:

$$i_H + i_R = i_O + i_K \tag{1}$$

where the subscripts denote hydrogen evolution, recombination of oxygen, oxygen evolution and corrosion, respectively.

(ii) mass transfer:

$$i_R = ky \tag{2}$$

where k is a mass transport parameter and y the mole fraction of oxygen.

(iii) mass balance for oxygen:

$$(i_O - i_R)(V/4F) = i_H(y/(1-y))(V/2F) \tag{3}$$

The left side of the equation denotes the net production rate of oxygen. The right side denotes the amount of oxygen vented. V is the molar volume.

Eqs. (1)–(3) can be used to solve for y when the values of i_K and i_O are known. A quadratic equation for y with i_K and i_O, and k is the result. With

$$B = (3i_O + 2i_K + k)/6k \tag{4}$$

the solution of the equation can be expressed:

$$y = B - (B^2 - i_O/3k)^{0.5} \tag{5}$$

The limiting solutions are: high k values and low currents (high recombination efficiency!):

$$y = i_O/k \tag{6}$$

and high current and low k value (no recombination!):

$$y = 1/3 \tag{7}$$

When all partial currents are known, all of the polarizations can be solved. The basis for our calculations are the Tafel equations assumed for the kinetics of the hydrogen and oxygen evolution and the modified Tafel expression for the corrosion.

(iv) oxygen evolution:

$$i_O = i_{O0}\exp(\eta_+/b_O) \tag{8}$$

where i_{O0} is the apparent exchange current density for oxygen evolution, and η_+ is the apparent overvoltage for oxygen evolution which is related to the equilibrium potential of the positive electrode instead of the standard potential of the oxygen electrode; b_O is the Tafel slope for the oxygen evolution reaction.

(v) hydrogen evolution:

$$i_H = i_{H0}\exp(\eta_-/b_H) \tag{9}$$

We use the apparent overvoltage for this electrode and apparent exchange current density for convenience. η_- denotes the overvoltage related to the equilibrium voltage of the negative electrode.

(vi) corrosion reaction:

$$i_K = i_{K0}\exp(\eta_+/b_K) \tag{10}$$

We use either a Tafel reaction with a high Tafel slope of 180 mV/decade, corrosion type-I or a curve derived from experimental data, corrosion type II. This is illustrated in Fig. 3. In this case i_{k0} means corrosion at $\eta_+ = 0$.

2.3. Basis for calculations

We used capacity as the basis for our calculations. We express current density as μA/Ah which gives reasonably

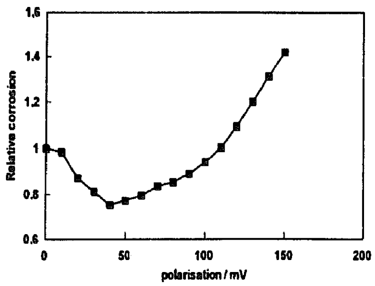

Fig. 3. Relative corrosion rates from Brecht et al. [5].

large numbers for partial currents and constants. In a spreadsheet program, i_K and i_O are calculated for positive polarizations from 0 to about 120 mV; from the equation system above, the oxygen mole fraction and hydrogen current are solved. Derived quantities such as recombination, negative plate polarization and total polarization are also calculated.

2.4. *Estimate of constants*

The activity of both electrodes varies much, see Table 1. The Willhinganz figures were measured on American stationary flooded batteries while the later-cited figures refer to valve regulated cells. It is known that the oxide used may influence the hydrogen overvoltage to a large extent [17]. A great influence is also exerted by the expander [13]. It should also be assumed that the oxygen overvoltage may be affected by the oxide used and by curing and formation methods. In the above-mentioned figures it should be stressed that differences in acid density exist. The typical cell of Willhinganz used an acid with a sp. gr. of 1.21 while in the other studies sp. gr of 1.28 or higher were used. As the acid density influences the charge acceptance of the positive electrode, it may, as well, influence the oxygen overvoltage. We used somewhat higher values for evaluation purposes, both for the exchange current of the hydrogen and for oxygen. The data of Calasanzio and McClelland [9] indicate that the apparent exchange current density for the oxygen electrode can be as high as 80 μA/Ah.

One parameter in the model, the mass transfer parameter *k*, has not been evaluated elsewhere. It may be estimated from various figures of 'limiting currents'. If we take the limiting current to be the maximum recombination current we may assume that this corresponds to a stoichiometric hydrogen oxygen mixture, where the mole fraction of oxygen is 1/3. The recombination 'limiting current' can be as high as a few A/100 Ah, while flooded batteries have recombination limits of a few mA/100 Ah. This will lead to *k* values of 100 000–50 μA/(Ah, mole fraction).

We found various corrosion data in the literature. In addition to the general corrosion level there is also a debate concerning the relation between potential and corrosion. According to Fiorino [18] a Tafel slope of 180 mV/decade can be applied. Most of the studies, however, indicate that the corrosion is quite constant for low overpotentials. A corrosion minimum occurs at a polarization of 40–60 mV. Both Willhinganz [7] and Brecht et al. [5] have published data on the plate growth as a function of potential. These data are based on long-term studies and should be reliable. It should be remembered that the Willhinganz and Brecht data concern positive plates which are almost pure-lead–calcium alloys. Another factor to be taken into consideration is the difference in acid concentration. The Willhinganz and Brecht data concern acids with a sp. gr of 1.21 which is considerably lower than that of modern valve regulated batteries. The open circuit of the PbO_2 electrode is higher in the more concentrated acid and this may move the position of the corrosion minimum closer to the OCP. More recent-developed alloys include tin, which may influence strongly the corrosion around the OCP. It may also be debated if the plate growth is directly proportional to the corrosion current. At low potentials, in vicinity of the equilibrium potential of the positive plate, the corrosion products contain lead sulfate in addition to lead dioxide, which means that less charge is needed per volume of corrosion products. For sake of simplicity, however, we have taken the Brecht data to be directly related to the corrosion current.

3. Results

3.1. *Standard case: good mass transfer*

The behaviour of a cell with assumed standard parameters has been calculated in Figs. 4 and 5. The recombination remains high in the whole current interval for which calculations have been made. There is a gradual decrease in recombination as the current increases. Concerning the question of the Tafel slopes, the assumed corrosion kinetics type plays a decisive influence. For corrosion type II we will have a Tafel

Table 1
Activity of electrodes: currents and Tafel slopes

Refs.	i_{HO} (μA/Ah)	i_{OO} μA/Ah)	i_K (μA/Ah)	b_H (mV/decade)	b_O (mV/decade)
Willhinganz [7]	1–5	1–8	5	110	70
Berndt et al. [12]	34	10	NA	NA	90
Teutsch [13]	8	40	13	120	90
Berndt [10]	1–3	10–30	20	120	80

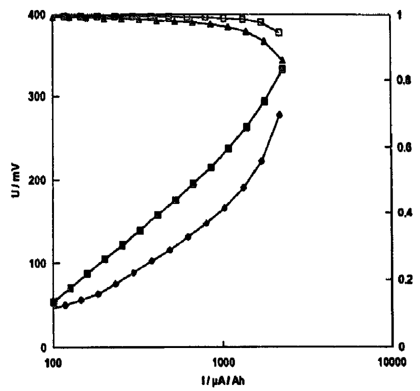

Fig. 4. Total cell overvoltage and recombination for standard parameters; $i_{O0}=60$, $i_{H0}=10$, $i_{K0}=20$ and $k=7000$. Two different corrosion kinetics are assumed. (□), (▲) recombination for corrosion type II and I, respectively; (■), (◆) overvoltage for corrosion type I and type II.

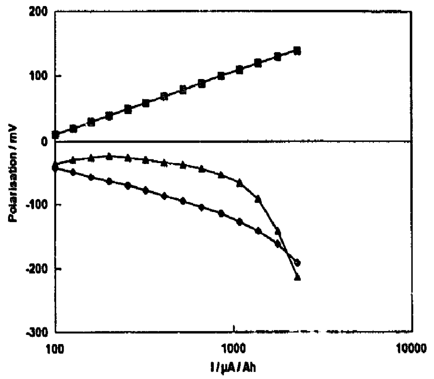

Fig. 5. Individual overvoltages and polarizations for standard case. Parameters are the same as in Fig. 4, only one positive curve as the curves nearly coincide. (■) positive electrode; (▲), (◆) negative electrode for corrosion type II and I, respectively.

slope close to 100 mV/decade at low overpotentials at which the corrosion is at or below the corrosion minimum. At somewhat higher overpotentials the Tafel slope gets much steeper approaching 200 mV/decade. At overpotentials above 0.1 V both corrosion kinetics will give high Tafel slopes and high recombinations. The model gives qualitative agreement with earlier observations of a high Tafel slope and a high recombination.

3.2. *Influence of electrode materials: hydrogen overvoltage*

The hydrogen overvoltage influences strongly the operation of a valve regulated battery. In Fig. 6 we have assumed a higher i_{H0}, three times higher than the standard value. The positive polarizations for the negative plate in this case indicate that the active material, metallic lead, will start to discharge. As this reaction is so fast in comparision to the hydrogen reaction, polarization will be much smaller. In comparision to the standard case there has to be a minimum charging current. If the battery is charged at the same total overpotential as in the standard case, the float current will be two to three times higher which is shown in Fig. 7. In this figure we have plotted only the currents which do not discharge the negative electrodes. Below this minimum current, we would get the polarization from the postitive electrodes only, resulting in a low Tafel slope of only 90 mV/decade.

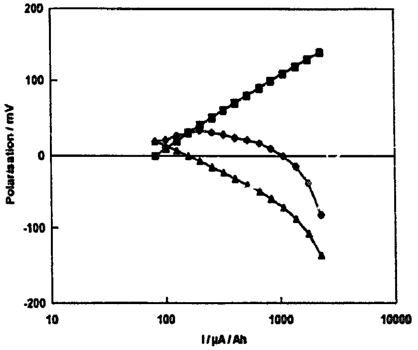

Fig. 6. Individual polarizations for the case with low hydrogen overvoltage material, $i_{H0}=30$, other parameters as in standard case. (■) positive electrode; (▲), (◆) negative electrode for corrosion type II and I, respectively.

3.3. *Influence of mass transfer: water losses*

When mass transfer (for oxygen) is enhanced, float current and recombination increase. In Fig. 8, float current, recombination and hydrogen evolution have been plotted versus mass transfer parameter. The two different corrosion kinetics give different hydrogen evolution rates because the hydrogen evolution is determined to a large extent by corrosion. The hydrogen evolution rate relates directly to the rate of water loss. The hydrogen that is lost is taken from the water in the electrolyte. The oxygen part of the water will either be vented or bound to PbO_2: the maximum allowable water loss rate may be estimated from the amount of water in a battery and

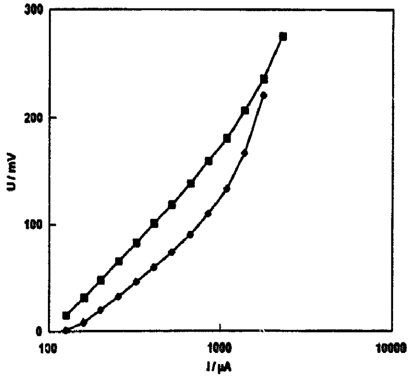

Fig. 7. Total cell overvoltage for the case in Fig. 7. (■), (◆) corrosion type I and II, respectively.

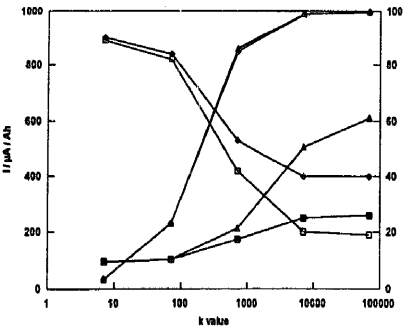

Fig. 8. Influence of mass transport parameter on total current, hydrogen evolution and recombination. Parameters as in standard case and at 100 mV total polarization. (◇), (△) recombination for corrosion type I and II, respectively; (◆), (□) hydrogen evolution current x10 for corrosion type I and II, respectively; (■), (▲) total current for corrosion type I and II, respectively.

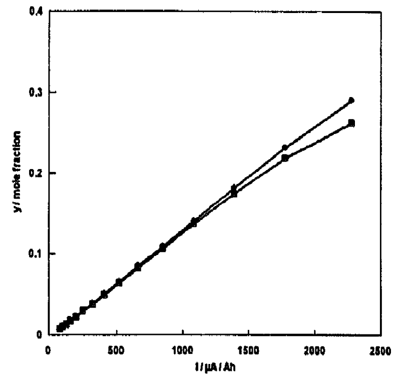

Fig. 9. Oxygen concentration vs. applied current density. Standard parameters as in in Fig. 4. (■), (◆) corrosion type I and II, respectively.

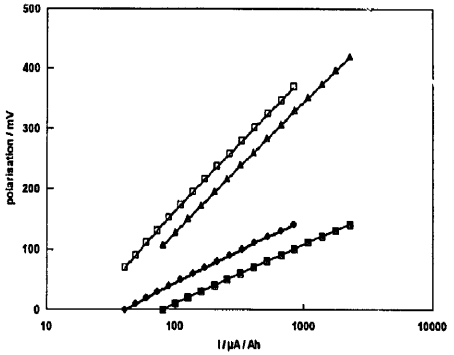

Fig. 10. Polarization for total cell and positive electrode at poor mass transfer conditions, $k=7$; corrosion type I assumed. (▲), (□) cell polarization, $i=30$ and 60, respectively; (■), (◆) positive electrode, $i_{00}=60$ and 30, respectively.

the anticipated life. A water loss of approximately 10% may seriously affect performance. In 10 years, 40 µA/Ah of hydrogen evolution current will lead to a loss of 10% in a battery with 1 kg of electrolyte.

3.4. Oxygen concentration

The oxygen concentration inside the battery increases almost lineary with current, see Fig. 9 for standard conditions. The concentration difference between the different corrosion kinetics at similar currents is small. However, overvoltages are different. At normal overvoltage, the oxygen concentration is below the flammability limit.

3.5. Positive electrode at poor mass transfer conditions

When the mass transfer conditions are poor, at low *k* values, the postitive elecrode may go to OCP. If a part of a valve regulated cell has a too high acid saturation, the mass transport conditions become even worse than in a flooded cell because there is no free convection of electrolyte. In Fig. 10, the overvoltages of the cell and that of the positive electrode have been calculated for a case of very poor mass transfer. When the oxygen kinetics is fast, the positive electrode will go to open circuit, if the overvoltage is 100 mV (for $i_{00}=60$). However, when the active materials have a high intrinsic oxygen overvoltage (low $i_{00}=30$), the cell can be operated at a lower voltage without the risk self-discharge of the positive electrodes.

4. Discussion

The model presented in this paper may be used to explain and get some insight into phenomena which may be difficult to observe directly.

For instance, float charging voltages recommended today are approximately 100 mV above the corresponding open circuits. For standard parameters this seems motivated. There is always a risk that there are zones in a battery with poor recombination as a result of the locally higher acid saturation. The higher saturation could for instance be the result of locally higher compression. Another explanation is that the acid is unevenly administred to the plate groups and that some separators will have a higher saturation than the others. The zones with a poor recombination may remain unnoticed. The overall recombination will still be high as very little current will pass through them. These wet zones will be in danger if the total overvoltage is not high enough to keep the positive electrodes charged. A gelled battery can be treated as an extreme example of unevenly distributed recombination. It is generally assumed that recombination occurs in the cracks that exist in a gelled cell. The cracks are widely spaced in a typical gelled cell, from one centimetre to a few centimetres. This is more than the distance between the plates and the likely diffusion length of oxygen. An unequal current distribution must be suspected in that case. Then the relations valid for the wet parts of the plates apply. In order to be on the safe side concerning the full state of charge, the recommended

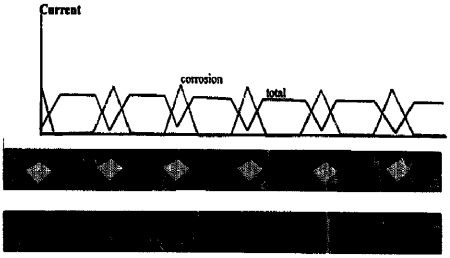

Fig. 11. Scheme of uneven current distribution around grid members.

float voltage should be such as to permit operation in a flooded mode.

Although the model does not take into account uneven current distribution, some qualitative reasoning may be applied to cases of uneven current distribution. The reasoning earlier treated the case of uneven distribution of electrolyte between the plates. Another reason that should be considered is uneven current generation within the plates. Electrodes contain metallic grid members embedded in active materials. Where there are no grid members, more current will be generated than in the zones with the grid parts; a schematic illustration is given in Fig. 11. The corrosion reaction is more unevenly distributed than the gas evolution; it is confined to the metallic parts. This observation is significant for the negative plate. The negative plate has to be polarized, otherwise it will self-discharge. If mass transfer for oxygen is good and there is no corrosion the negative will self-discharge. The earlier case of low hydrogen overvoltage illustrates this. Where k is high, the corrosion has to be higher than i_{H0}, the negative electrode self-discharge rate, in order for the negative electrode not to self-discharge. Such conditions are prevalent in the centre of grid openings, where the distance to the corroding grid members is the largest.

For further modelling work of the valve regulated battery it would be interesting to include thermal effects and transient behaviour.

It will also be interesting to take into account the uneven current distribution caused by, e.g.: cracks in the gel; discontinuity around grid members, and thermal gradients.

5. List of symbols

b	Tafel slope (mV/decade or natural decade) [a]
b_H	Tafel slope, hydrogen reaction
b_K	Tafel slope, corrosion reaction
b_O	Tafel slope, oxygen evolution
F	Faraday constant (96 500 A s/mol)
i	current density (μA/Ah)
i_H	hydrogen evolution
i_{H0}	apparent exchange current density for hydrogen evolution
i_K	corrosion current density
i_{K0}	apparent exchange current density for corrosion
i_O	oxygen evolution current density
i_{O0}	apparent exchange current density for oxygen evolution
i_R	recombination current
k	mass transfer coefficient (μA/(Ah, molefraction))
V	molar volume (dm^3/mol)

Greek letters

η	overvoltage (mV)
η_+	overvoltage of positive electrode in relation to its equilibrium voltage
η_-	overvoltage of negative electrode in relation to its equilibrium voltage

[a] Note: when the natural exponential expression is used the corresponding b-values for natural logarithms are used.

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