Analysis and Comparison of Generalized Peukert's Equations Parameters for Nickel-Cadmium Batteries with Pocket and Sintered Electrodes

Nataliya N. Yazvinskaya, Nikolay E. Galushkin^{*}, Dmitriy N. Galushkin, Boris Yu. Kalmykov

Don State Technical University, Laboratory of electrochemical and hydrogen energy, 1 Gagarin Square, Town of Rostov-on-Don, Russia, 344000. *E-mail: <u>galushkinne@mail.ru</u>

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In this paper, it is proved by experiments that the generalized Peukert's equations $C=C_m/(1+(i/i_0)^n)$ and $C=C_m erfc((i/i_k-1)/n)/erfc(-1/n)$, are applicable for the released capacity evaluation by nickel-cadmium batteries at any discharge currents and with electrodes of any types (pocket, sintered, plastic bonded). It was shown that in limits of experimental error, the parameter *n* coincides for batteries of various capacities but with the same type of electrodes and with the same discharge mode. But the parameter *n* depends on the batteries' discharge mode (medium, high) and electrodes type (pocket, sintered, plastic bonded). The explanation is proposed for the parameters changing of the generalized Peukert's equations for batteries of different discharge modes and having the different types of electrodes. The explanation is based on the statistical interpretation of the generalized Peukert's equations and the electrochemical process distribution along the porous electrode depth.

Keywords: Peukert's equation, modeling, battery, nickel-cadmium, discharge current

1. INTRODUCTION

At the present time, the electrochemical batteries are increasingly used in various devices and appliances of both domestic and special intention [1].

Despite certain limitations of the nickel-cadmium batteries use in Europe, their production is not reduced; instead, it even grows. At present, the alkaline batteries are used in the railway transport, the aviation, the electric vehicles, etc. [2–4].

However, in order for optimal designing and control of batteries belonging to various technical systems, necessary are quite reliable battery models. The performance of the systems containing the batteries is largely determined by their state of charge (SoC). That is why the modern battery models must allow the reliable evaluation of the batteries' SoC.

Now, the various battery models and SoC evaluation methods exist. Historically, one of the first methods was the method of the SoC evaluation based on the open circuit voltage. [5] However in some cases, this method causes errors up to 20% and more. [6] The use of the Kalman Filter and the models based on it is the other method of the SoC evaluation. [7–9] At the present time most often for the SoC evaluation, the combination of the two following methods is used: the ampere-hours calculation and the voltage profiles use.

From the theoreticaloй point of view, the most accurate SoC evaluations can be obtained only based on the models taking into account all the electrochemical processes running inside of the batteries [9-11]. However in practice for the batteries' SoC evaluation, such models are used rarely if ever. The reason is that those models require great computing power, which is unacceptable for onboard computers of airplanes, electric vehicles and other mobile systems containing batteries [12].

Often, an alternative to the above specified methods are the analytical models based on the generalized Peukert's equations [3,12-14] or on the non-linear structural models. [15] It should be noted that the analytical models can be used also in frame of the fundamental electrochemical models, where it is necessary to take some poorly studied phenomena or processes into account. For example, such are the thermal runaway [16-18], the hydrogen accumulation in battery electrodes [19,20] and the gas generation during batteries cycling [21,22].

This paper is aimed at the analysis of the parameters changing of the generalized Peukert's equations for the nickel-cadmium batteries with the sintered electrodes and at comparison of those parameters values with ones obtained by us earlier for the nickel-cadmium batteries with the pocket electrodes [23,24].

2. PEUKERT'S EQUATION GENERALIZATION

The classical Peukert's equation [25]

$$C = \frac{A}{i^n} \quad , \tag{1}$$

is inapplicable at small discharge currents as - according to this equation - in a case of reduction of the discharge current *i*, the batteries' released capacity *C* tends to infinity, which doesn't make a physical sense.

In our previous papers [23,24,26], it was proved experimentally that in the case of the nickelcadmium batteries with the pocket electrodes, the following generalized Peukert's equations best correspond to the experimental data:

$$C = \frac{A}{1 + Bi^{n}} , \qquad (2)$$
$$C = \frac{A}{2} \cdot erfc \left(\frac{i - i_{k}}{\sqrt{2\sigma}}\right). \qquad (3)$$

where *i* is discharge current, *C* is batteries' released capacity, i_k is the mean value of the statistical variable *i*, σ is the standard deviation and *A*, *B*, *n* are empiric constants. That is why only these equations will be studied by us in this paper.

Often, the equation (3) describes the phase transition in various processes [27]. But a battery discharge represents nothing but the phase transition of electrodes' active mass: the transition is going on from the charged state phase to the discharged state phase. That is why no wonder, the batteries' discharge process is well described by the equation (3).

For convenience of the empiric equations (2,3) comparison with the experimental data, let's rewrite them in the following form:

$$C = \frac{C_m}{1 + \left(\frac{i}{i_0}\right)^n} , \qquad (4)$$

$$C = \frac{C_m}{erfc\left(\frac{-1}{n}\right)} \cdot erfc\left(\frac{i/i_k - 1}{n}\right) . \qquad (5)$$

In this case, $C(0) = C_m$, i.e. C_m is top capacity of a battery. Besides for the equation (4), $C(i_0) = C_m/2$; i.e. i_0 is the discharge current value, at which the battery's released capacity is half of its top capacity. For the equation (5), $C(i_k) = C_m/erfc(-1/n)$. Hence, i_k is the discharge current value, at which the battery's releases capacity being erfc(-1/n) times less than its top capacity. In the paper [26], it was shown that at $n \le 1$, $1.85 \le erfc(-1/n) \le 2$.

3. EXPERIMENTAL

For our experimental studies, there were used the following models of the SAFT-produced batteries with the sintered positive electrodes and the plastic bonded negative electrodes: SRM 62, SRM 105 and SRM 200 (with the capacities 62 Ah, 105 Ah and 200Ah respectively, medium discharge mode) and SRX 800, SRX 1000 and SRX 1900 (with the capacities 80 Ah, 100 Ah and 190 Ah respectively, high discharge mode). The batteries' charging was performed in line with their operation manuals: $0.2C_n$ current for 7.5 hours (where C_n was the battery nominal capacity).

Also for the results comparison, there were studied the SAFT-produced batteries with the pocket electrodes: SBM 65, SBM 112 and SBM 231 (with the capacities 65 Ah, 112 Ah and 231Ah respectively, medium discharge mode) and SBH 69, SBH 118 and SBH 196 (with the capacities 69 Ah, 118 Ah and 196Ah respectively, high discharge mode). The batteries' charging was performed in line with their operation manuals: $0.2C_n$ current for 10 hours.

Their discharge was done with direct currents down to the voltage value 1V. The used discharge currents were in the range from $0.1C_n$ up to the currents, at which the battery's released capacity was close to zero.

In order to exclude various residual effects (in particular, the memory effect, etc.), prior to each change of the discharge current, the training cycles were fulfilled. The training cycles were performed in line with the operation manuals of the batteries under investigation (charge: the standard modes described above, discharge: direct current 0.2Cn to a voltage value of 1 V). The number of the training cycles performed was not less than five. However, if in three last cycles, the released capacity differed by more than 5%, the training cycles were continued.

At each discharge current, as the average value, the capacity value was taken obtained in three charge-discharge cycles in line. This enabled obtaining more reliable value of the battery's released capacity. However, if the released capacity values in these cycles differed more than by 5%, the experiment was repeated from the very beginning.

Besides it is for sure that even in the same batch of the identical batteries, there exists a statistical dispersion of their parameters connected with the statistical dispersion of their production parameters, times and modes of their operation, etc. This dispersion, for example, for batteries' top capacities, can reach 4-5% and sometimes even higher. In order to exclude this statistical dispersion as much as possible, we standardized the obtained experimentally batteries' capacities on their top capacity (being found experimentally for each battery).

4. RESULTS AND DISCUSSION

The released capacity of the nickel-cadmium batteries depends on two factors: the discharge current and the battery temperature. [12, 28-30] For example at reduction of the battery temperature, the released capacity falls because of slowing down of the chemical reactions inside of the battery. [12] However, the temperature influence is not taken into account by the classical Peukert's equation [1], which fact is able to cause considerable errors at evaluation of a battery capacity [31].

4.1. Batteries' released capacity dependence on temperature

In our experiments, there were used the batteries of the company SAFT production, namely the brands SRM 105 and SRX 1000. These batteries are equipped with the sintered positive electrodes and the plastic bonded negative electrodes. Also for the results comparison, the SBM 112 and SBH 118 batteries were studied with the pocket electrodes.

The experiments were implemented in the climatic chamber Binder MK240 (BINDER GmbH, Germany) in the temperatures range from -30°C to 55°C. The obtained experimental results are represented in the Figure 1.

It should be noted that the released capacities in the standardized coordinates (T, C/C_{mref}) of the batteries of both types (SRM 105 / SRX 1000 and SBM 112 / SBH 118) coincide in limits of experimental error. That is why in the Figure 1, the experimental data are given only for the batteries of the models SRM 105 and SBM 112.

In the paper [30], it was experimentally proved that the dependence between the released capacity of nickel-cadmium batteries and their temperatures is described by the following equation:

$$C = C_{mref} K \frac{\left(\frac{T - T_L}{T_{ref} - T_L}\right)^{\nu}}{(K - 1) + \left(\frac{T - T_L}{T_{ref} - T_L}\right)^{\beta}}, \qquad (6)$$

where T_{ref} is a battery reference temperature; C_{mref} is the top released capacity of the battery at the temperature T_{ref} ; T_L is a temperature, at which C=0, i.e. it is the temperature, at which running of all the electrochemical processes stops; and the parameter K shows how many times (theoretically) the capacity C in the equation (6) can be increased with the temperature growth from T_{ref} to infinity [30].



Figure 1. Released capacity dependence on temperature of studied batteries at discharge current value 0.2 C_n (C_n is battery nominal capacity). C_{mref} is top capacity released at the temperature $T_{ref}=25^{\circ}$ C. a) Batteries with sintered positive electrodes and plastic bonded negative ones; b) Batteries with pocket electrodes.

Using the experimental data represented in the Fig. 1, the optimal parameters for the equation (6) were found with aid of the least square method and the Levenberg–Marquardt optimization procedure. The obtained optimal parameters are shown in the Table 1.

Table 1. Optimal values of parameters for equation (6)

Model	Tref(°C)	$C_{mref}(Ah)$	β	$T_L(^{\circ}C)$	K	δ ^a (%)
SRM 105	25	105	2.987	-61.144	1.031	1.915
SRX 1000	25	100	3.054	-62.029	1.039	1.807
SBM 112	25	112	3.261	-64.345	1.042	1.585
SBH 118	25	118	3.273	-63.089	1.039	1.573

^a δ is relative error of experimental data approximation by the equation (6) in Fig. 1.

According to the experimental data (Fig. 1) in the temperature range from 25 to 55°C, the capacity relative deviation from the average value makes less than 1%. Hence, in the temperature range from 25 to 55°C, it is possible to neglect the temperature influence on the batteries' released capacity. This statement is fair for all the studied by us batteries. (Tables 2,3).

4.2. Batteries' released capacity dependence on discharge current

In the experiments, all investigated batteries were used.



Figure 2. Comparison of released capacity dependence on discharge current between batteries with pocket electrodes and ones with sintered electrodes of various discharge modes. a) Batteries of medium discharge mode; b) Batteries of high discharge mode. The parameters C_m and i_0 were taken from the Tables 2 and 3.

The batteries were cycled at the temperature $T=25^{\circ}C$ inside of the climatic chamber Binder MK240 (BINDER GmbH, Germany). Besides, to the batteries, the heat sinks were fastened with aid of a specially made fixing element and heat-conducting paste; so those precautions increased

considerably both the thermal flow from the batteries and their cooling. At any discharge currents, these measures allowed to keep the temperature of nickel-cadmium batteries much lower than 55°C.

The obtained experimental data are represented in the Figure 2. In the Fig.2, the experimental data were approximated by the equation (4) as the equation (4) had the least relative error of approximation (Tables 2,3) in all our experiments.

Using the experimental data given in the Fig. 2, the optimal parameters for the equations (4,5) were found with aid of the least square method and the Levenberg–Marquardt optimization procedure. The obtained optimal parameters are shown in the Tables 2 and 3.

C_n (Ah) ^a	$C(\Lambda h)$	· (A) · (A)		hs (a)				
	$C_m(AII)$	$lo(\mathbf{A})$ or $lk(\mathbf{A})$	n	°∂ (%)				
Equation (4)								
62	61.219	159.129	2.527	1.337				
105	104.042	239.337	2.525	1.304				
200	198.953	405.483	2.536	1.961				
80	80.386	316.761	2.799	1.956				
100	102.425	390.323	2.805	2.212				
190	193.238	746.383	2.922	1.751				
Equation (5)								
62	64.534	140.699	1.324	3.492				
105	110.033	209.098	1.345	3.101				
200	211.555	345.609	1.390	3.105				
80	83.747	302.361	1.016	3.79				
100	106.747	372.634	1.019	4.25				
190	201.185	721.969	0.953	4.028				
	62 105 200 80 100 190 62 105 200 80 100 190	Equation 62 61.219 105 104.042 200 198.953 80 80.386 100 102.425 190 193.238 Equation 62 62 64.534 105 110.033 200 211.555 80 83.747 100 106.747 190 201.185	$\begin{array}{c c} Equation (4) \\ \hline Equation (4) \\ \hline 62 & 61.219 & 159.129 \\ 105 & 104.042 & 239.337 \\ 200 & 198.953 & 405.483 \\ 80 & 80.386 & 316.761 \\ 100 & 102.425 & 390.323 \\ \hline 100 & 193.238 & 746.383 \\ \hline Equation (5) \\ \hline 62 & 64.534 & 140.699 \\ 105 & 110.033 & 209.098 \\ 200 & 211.555 & 345.609 \\ 80 & 83.747 & 302.361 \\ 100 & 106.747 & 372.634 \\ 190 & 201.185 & 721.969 \\ \end{array}$	$\begin{array}{c c c c c c c c c c c c c c c c c c c $				

Table 2. Optimal parameters of generalized Peukert's equations (4,5) for nickel-cadmium batteries with sintered positive electrodes and plastic bonded negative ones

^aNominal capacity of a cell, ^bRelative error of experimental data approximation by the Equations (4,5) in Figure 2

Let's analyze the obtained experimental results.

Firstly, the relative error of experimental data approximation by the equations (4,5) is less than 4.5%. Such error is quite acceptable at practical evaluations of batteries' SoC. [12] Hence, the generalized Peukert's equations (4,5) can be used for evaluation of released capacities of batteries with any types of electrodes (pocket, sintered, plastic bonded).

Secondly, in the Figure 2, the experimental data coincide (in the coordinates C/C_m , *i/io* (или *i/ik*)) for batteries of various capacities, but with the same type of electrodes and of the same mode of discharge.

In the standardized coordinates, the equations (4,5) depend only on the single parameter n. Hence, also the parameter n must coincide for these batteries. Indeed, in limits of the experimental error according to the data given in the Tables 2 and 3, the parameter n coincides for batteries of various capacities but with the same type of electrodes and of the same mode of discharge.

Model	$C_n (Ah)^a$	$C_m(Ah)$	$i_0(A)$ or $i_k(A)$	n	$b\delta(\%)$			
Equation (4)								
SBM 65	65	65.569	71.043	3.073	2.536			
SBM 112	112	112.709	121.575	3.176	2.799			
SBM 231	231	232.258	249.982	3.169	2.629			
SBH 69	69	67.306	210.774	4.482	3.056			
SBH 118	118	115.131	360.489	4.473	3.148			
SBH 196	196	191.016	599.493	4.488	3.162			
Equation (5)								
SBM 65	65	68.701	68.469	0.897	4.491			
SBM 112	112	117.858	118.384	0.850	4.039			
SBM 231	231	242.961	243.132	0.857	4.086			
SBH 69	69	68.482	212.996	0.557	3.641			
SBH 118	118	117.194	364.265	0.561	3.645			
SBH 196	196	194.401	605.679	0.558	3.651			

Table 3. Optimal parameters of generalized Peukert's equations (4,5) for nickel-cadmium batteries with pocket electrodes

^aNominal capacity of a cell, ^bRelative error of experimental data approximation by the Equations (4,5) in Figure 2

This experimental result clearly follows from the fact that in all studied batteries (with the same type of electrodes and the same mode of discharge), the same electrodes are used. The difference in batteries capacities is connected only with a number of electrodes and their square area.

In the standardized coordinates, the study of any battery gets reduced to the study of a battery of single capacity, which – at discharge current $i/i_0 = 1$ – releases the capacity equal to C_m/2. Hence, in the standardized coordinates for batteries of any capacity, the experimental functions C(i) and the parameter *n* must coincide (in limits of the experimental error) if the batteries have their electrodes of the same type and were produced with use of the same production technology. This is exactly what is observed in the experiments (Tables 2 and 3).

Thirdly, the parameter n in the generalized Peukert's equations (4,5) depends on the mode of discharge (medium, high) and on the type of electrodes in the batteries (pocket, sintered, plastic bonded). Notably, for the equation (4), the parameter n grows at transition from batteries in the sequence (medium, high), while for the equation (5), it falls. This is fair for batteries with any type of electrodes (Tables 2 and 3).

Fourthly, at transition from batteries with pocket electrodes to batteries with sintered/plastic bonded ones, the parameter n grows in the equation (5) and falls in the equation (4) (for batteries of the same mode of discharge).

Of theoretical interest, the equation (5) is. It describes various phase transitions subjected to the normal distribution law [27]. However, the batteries discharge process itself is the phase transition: it is the transition from phases of active mass of electrodes corresponding to batteries' charged state to phases of the active mass corresponding to the batteries' discharged state.

Thus, based on the good correspondence (see Tables 2,3) of the equation (5) to the experimental data (Fig.2), one can conclude that at the level of ions, the process of discharge is a statistical process subjected to the normal distribution law.

Now, let's analyze the above established experimental facts on the basis of a statistical interpretation of the generalized Peukert's equation (5) and on distribution of the electrochemical process along the depth of a porous electrode.

If to suppose that the electrochemical process is distributed uniformly along the depth of the porous electrode, in this case, all the active mass of the electrodes will be discharged uniformly regardless of the discharge current. Hence, always the battery released capacity C will be maximal C_m regardless of the discharge current. However, in this case at a very high discharge current I_k , necessarily, a situation will appear, when already at the moment of switching on of a battery discharge mode, the voltage on the battery terminals will be equal to the lower cutoff voltage (because of the battery internal resistance). Hence, at the discharge current I_k , the battery released capacity will be equal to zero. Thus, in the case of the uniform distribution of the electrochemical process along the depth of the porous electrode, the function C(i) must have the rectangular form (Fig. 3 (3)).

However, as is known [15, 32], the electrochemical process decreases exponentially along the depth of the porous electrode.



Figure 3. Generalized Peukert's equation (5) in standardized coordinates for various values of the parameter n

In the case of the one-dimensional model of the porous electrode and the linear Butler-Volmer function ($i = \alpha \cdot \varphi$) at the boundary electrode/electrolyte, the discharge current distribution j(x) along the porous electrode depth will have the following form: [33,34]

$$j(x) = J_{\sqrt{\frac{\alpha\rho}{s}}} \frac{\cosh\left(\sqrt{\rho s \alpha} (l-x)\right)}{\sinh\left(\sqrt{\rho s \alpha} l\right)}$$
(7)

In this model, the beginning of the porous electrode is situated in the point x=0; J is outer discharge current density; ρ is specific resistance of electrolyte in porous electrode; s is square area of

pores surface in an electrode volume unit; *l* is half thickness of electrode at bilateral current supply; α is specific conductivity for activation process on electrode/electrolyte boundary; ϕ is polarization inside of porous electrode.

At the non-uniform distribution of the electrochemical process along the depth of the porous electrode, in the case of reaching the cutoff voltage on battery terminals, not all the active mass of the electrodes will be discharged; and hence, the battery released capacity will fall. This battery released capacity decrease will be the more noticeable, the more greater the non-uniformity will be of the electrochemical process distribution along the depth of the porous electrode.

The extent of the non-uniformity of the electrochemical process distribution along the depth of the porous electrode can be evaluated with aid of the equation

$$j(0)/j(l) = \cosh(\sqrt{\rho s \alpha l}) \quad . \tag{8}$$

Based on the equation (8), it is possible to explain the above established experimental fact, which is the decrease of the parameter n in the equation (5) for batteries with any type of electrodes in the modes sequence (medium, high). In this sequence, the electrodes thickness in the batteries is decreased. Hence, according to equation (8), with a decrease in the thickness of the electrodes, the statistical dispersion for the degree of discharge of the active substance along the depth of the porous electrode will decrease. In this case, the statistical dispersion of the entire process of battery discharge will fall. As it is seen from the equation (5), it is parameter n that determines the statistical dispersion for the batteries discharge process in the standardized coordinates. Hence, with the electrodes thickness decrease, the parameter n in the equation (5) is to fall, which indeed was found by us experimentally (Tables 2,3).

Thus, with the electrodes thickness decrease, the function C(i) will approach the rectangular form (Fig. 3 (1,2,3)) typical for the uniform distribution of the electrochemical process along the depth of the porous electrode. It should be noted that for the equation (4) the decrease of the statistical dispersion is connected with the growth of the parameter *n*.

Now let's compare batteries with electrodes of different types. According to experimental studies (Tables 2,3), at transition from the batteries with the pocket electrodes to the batteries with the sintered/plastic bonded electrodes, the parameter n in the equation (5) grow (for batteries of the same discharge mode). According to the conducted analysis this can only due growth to the non-uniformity of the electrochemical process distribution along the porous electrode depth . However, both the sintered & plastic bonded electrodes are thinner than the pocket electrodes for batteries of the same discharge mode. Hence, in this case, the non-uniformity growth of the electrochemical process distribution along the porous electrode depth is possible only due to the growth of the parameter $\rho s \alpha$ in equation (8). Indeed, our preliminary experimental studies have shown that the specific conductivity α for the activation process on the boundary electrode/electrolyte is much higher in the porous sintered electrode than in the pocket electrode. However, the more detailed measurement of the parameters ρ , s, α in various types of electrodes is a topic of our separate investigation.

5. CONCLUSIONS

The conducted experimental studies have shown that the generalized Peukert's equations (4,5) are applicable for the evaluation of the nickel-cadmium batteries' released capacity at any discharge currents and for batteries with any types of electrodes. It should be noted that the classical Peukert's equation (1) is not applicable at small discharge current values.

The parameter n in the equations (4,5) does not depend on a capacity of batteries under investigation; instead, it depends on a type of used electrodes and the discharge modes of the batteries. Hence, the parameter n depends on the constructive features of the electrodes used in the batteries (such as electrodes' type, thickness, etc.) and on their manufacture technology (their active mass composition, the presence of various additions, etc.).

The proposed statistical mechanism of the batteries discharge process enables explaining the equation (5) parameters changing in dependence on discharge modes of batteries and used electrode types.

As the Peukert's equation is widely used in various methods and models [6,12,13,31], the clarification of this equation and the finding of its underlying physical and electrochemical mechanisms is of a significant both practical and theoretical importance.

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