

TOPICAL COLLECTION: ELECTRONIC MATERIALS FOR RENEWABLE ENERGY APPLICATIONS

Lithium Battery Transient Response as a Diagnostic Tool

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Lithium batteries are currently used as the main energy storage for electronic devices. Progress in the field of portable electronic devices is significantly determined by the improvement of their weight/dimensional characteristics and specific capacity. In addition to the high reliability required of lithium batteries, in some critical applications proper diagnostics are required. Corresponding techniques allow prediction and prevention of operation interruption and avoidance of expensive battery replacement, and also provide additional benefits. Many effective diagnostic methods have been suggested; however, most of them require expensive experimental equipment, as well as interruption or strong perturbation of the operating mode. In the framework of this investigation, a simple diagnostic method based on analysis of transient processes is proposed. The transient response is considered as a reaction to an applied load variation that typically corresponds to normal operating conditions for most real applications. The transient response contains the same information as the impedance characteristic for the system operating in linear mode. Taking into account the large number of publications describing the impedance response associated with diagnostic methods, it can be assumed that the transient response contains a sufficient amount of information for creation of effective diagnostic systems. The proposed experimental installation is based on a controlled load, providing current variation, measuring equipment, and data processing electronics. It is proposed to use the second exponent parameters U_2 and β to estimate the state of charge for secondary lithium batteries. The proposed method improves the accuracy and reliability of a set of quantitative parameters associated with electrochemical energy sources.

Key words: Lithium battery, diagnostics, time response, linear mode, time analysis

INTRODUCTION

Lithium batteries (LBs) provide high-density power and weight/dimensional characteristics as well as long service life in comparison with traditional types of batteries,¹ and these characteristics explain the wide use of LBs in portable electronics, electric vehicles, large-scale energy storage,² and other applications. The rapid development of the mentioned fields has been determined significantly by progress in LB technology. In addition to the high reliability of LBs, in some critical applications, diagnostics require detection and prediction of malfunction, prevention of operation interruption, and avoidance of expensive battery replacement procedures. These needs pose the urgent problem of developing new methods for LB diagnostics. This is urgently needed to provide reliable estimation of technical conditions during normal LB operation.

Many methods exist for technical diagnosis of electrochemical systems. They have different capabilities and fields of applications. Mainly, these methods are based on analysis of polarization

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curves, because this allows evaluation of the quality of electrochemical reactions, value of charge-transfer resistance, and losses associated with limited mass transfer processes.^{3,4} The disadvantage of this approach is the impossibility of diagnostics administration during battery operation.

One of the most convenient methods for battery state-of-health (SOH) assessment is based on charge capacity evaluation during discharge in nominal mode.⁵ Accordingly, the process of battery degradation is expressed in terms of the decreasing capacity value. The key disadvantage of this approach is the relatively long measurement time, which does not allow detection of temporally localized failures, e.g., due to local overheating.

A promising method for electrochemical systems diagnosis is the method of noise spectroscopy.⁶⁻¹⁰ This approach does not imply direct interference in the operation of the battery and allows a researcher to carry out diagnostics during its operation; however, expensive measuring equipment and considerable time costs are required to accumulate the necessary statistics. In addition, a general approach for building a noise diagnostic system for all types of batteries is lacking.

The most promising strategy for implementation of battery diagnostic systems during their operation involves methods that use weak disturbances of the operating mode. These methods include impedance spectroscopy,^{11–13} the current interrupt method,¹⁴ and fast electrochemical impedance spectroscopy.¹⁵ Impedance spectroscopy is one of the most effective and informative methods; however, it suffers from a number of significant drawbacks: (1) it requires expensive, technically complex equipment; and (2)the measurement of the impedance frequency response takes a long time, making it difficult to provide the necessary stability for most electrochemical systems. The current interrupt method permits acquisition of information about the battery internal resistance and provides an effective tool for technical state monitoring during operation. The basic drawback of these methods is associated with the fact that only one parameter is determined. In other words, these methods provide reliable but not a sufficient amount of information to build a complete diagnostic system. The main advantages of the current interruption method are that the test procedures are localized in time and the measurement equipment used is simple. In addition, the high information impact of impedance spectroscopy is combined with the possibilities of fast electrochemical impedance spectroscopy. In this paper, a corresponding approach is designed to build the desired diagnostic tool.

LBs are usually used in combination with a battery management system (BMS), which has protective functions by simultaneously monitoring temperature, electrical current, and voltage. The main functions of the BMS are related to accurate measurement of cell voltages, evaluation of battery status, carrying out battery balancing, diagnosing battery failures, monitoring state-of-charge (SOC),³ etc. The main aim of the present work is to provide an effective diagnostic tool that can be effectively introduced into a conventional BMS without significantly increasing its cost.

In the framework of this paper, we propose an approach that includes the organization of the battery diagnostic system. It is based on a combination of the high information capabilities of impedance spectroscopy and measurement procedures related to the localization of the current interruption method. The basic ideas of this paper were presented and discussed at the European Conference on Renewable Energy Systems (ECRES-2017) and 2018 Systems of Signal Synchronization, Generating and Processing in Telecommunications (SYNCHROINFO).¹⁶

EXPERIMENTAL PROCEDURES

Lithium Battery Transient Response

The transient response is traditionally used to describe time relaxation processes in linear electric circuits (see, for example, Ref. 17). In the case of a small perturbation, an electrochemical system remains linear, making it possible to use transient response apparatus for analysis of such systems.

The transient response of an LB can be defined as its response to a step variation in voltage or current. In the framework of this paper, we consider the voltage response $h_u(t)$ to a stepwise current variation $1_i(t)$. In this case, the measured signal is a transient voltage process caused by a step change in the electrical current of the lithium battery (Fig. 1). However, it should be mentioned that all conclusions are also valid for the current response due to a step voltage variation. Two contradictory restrictions are imposed on the amplitude of the considered current variation: (a) maintaining the linear operating mode of the lithium battery while (b) obtaining reliable (relatively stable) experimental data.

Figure 2 shows a typical transient voltage response of an LB to a step current variation. Note that the transient response exhibits complex behavior that can be considered as a combination of two parts (Fig. 2): an instantaneous voltage step (1) and a relatively slow relaxation process (2). The instantaneous voltage step is related to ohmic polarization, whereas the relaxation process corresponds to charge-transfer processes within the LB.



Fig. 1. Lithium battery transient response generation.



Fig. 2. Typical transient voltage response of an LB to a step current variation.



Fig. 3. Verification of the simplified model (1) according to criterion (2).

Taking into account the relaxation processes, it can be assumed that the transient response of an LB can be described by an exponential equation,

$$U(t) = U_{\rm L} - U_1 e^{-\alpha t},\tag{1}$$

where $U_{\rm L}$ is the total voltage change after termination of the relaxation process, U_1 is the exponential amplitude, and α is the inverse time constant. The accuracy of this assumption can be verified if one considers the product of the first derivative of the relaxation process and an exponent

$$h'_{II}(t) \cdot e^{\gamma t}.$$
 (2)

If model (1) is valid, this product will be monotonic for any real factor γ . Figure 3 shows that the product (2) is not monotonic but rather presents a bell-shaped form for some positive γ values. This indicates that the transient response cannot be described by the simple Eq. 1.

The transient response of a battery can be described in terms of an equivalent circuit, for example, the equivalent circuit most frequently used for lithium batteries, presented in Fig. 4,¹⁸ i.e., an *RC* circuit of second order. Here, R_1 and C_1 are, respectively, the charge-transfer resistance and interfacial capacitance (which includes the



Fig. 4. Equivalent electrical circuit for an LB.

capacitance of the double electric layer and associated capacitive components caused by adsorption processes, formation of passive films, etc.) for the first electrode; R_2 is an ohmic resistance, which is a function of the resistance of the electrolyte, electrodes, and interelement junctions; and R_3 and C_2 are, respectively, the charge-transfer resistance and the interfacial capacity for the second electrode.^{16,19}

Analysis of this equivalent circuit indicates that the following model, comprising two exponents, should be used to describe the transient response:

$$U(t) = U_{\rm L} - U_1 e^{-\alpha t} - U_2 e^{-\beta t}, \tag{3}$$

where $U_{\rm L}$ is the total voltage change after termination of the relaxation process; U_1 and U_2 are the exponential amplitudes; and α and β are the corresponding inverse time constants. The procedure of computer fitting allows determination of the parameters of model (3), which can be preliminarily considered as the diagnostic features for estimation of the technical state of the LB. It can be shown that the parameters of the electrical equivalent circuit of a lithium battery with known parameters $U_{\rm L}$, U_1 , U_2 , α , and β can be determined according to the following system of equations:

$$\begin{cases} R_{1} = -\frac{U_{1}}{M_{0}}; \\ C_{1} = \frac{\Delta I_{0}}{-\alpha U_{1}}; \\ R_{2} = \frac{U_{L}}{M_{0}} + \frac{U_{1}}{M_{0}} + \frac{U_{2}}{M_{0}}; \\ C_{2} = \frac{\Delta I_{0}}{-\beta U_{2}}; \\ R_{3} = -\frac{U_{2}}{M_{0}}, \end{cases}$$
(4)

where ΔI_0 determines the amplitude of the applied current step 1(t).

Smoothing Procedure

To smooth the original experimental data, it is proposed to use the procedure of optimal linear smoothing (POLS).^{20,21} This method permits determination of the optimal and smoothed trend (defined as the pseudofitting function) and its separation from high-frequency fluctuations. The smoothed trend \tilde{y} for the discretized signal is found as

$$\tilde{y}_j(w) \equiv \frac{\sum_{j'}^N K(\frac{x_j - x_{j'}}{w}) y_{j'}}{\sum_{j'}^N K(\frac{x_j - x_{j'}}{w})},$$
(5)

where $\{x\}$ is the set belonging to the input variable, *y* is an initial function, *w* is a smoothing window, and

$$K(t) = \exp\left(-\frac{t^2}{2}\right) \tag{6}$$

determines the Gaussian kernel. The Gaussian kernel is used here for two important reasons: (1)the smoothed function is determined by the equation obtained as a result of linear conversion that does not lead to uncontrolled errors, and (2) the smoothing width w (determined also as a smoothing window) itself represents a fitting parameter and could take any value. However, it is necessary to remark here on two limiting values of this parameter: (a) for $w \gg 1$, function (5) becomes the usual arithmetic mean, while (b) for $w \rightarrow 0$, the kernel (6) tends to a delta function, and therefore, in this case, the initial function coincides with the smoothed one. The function \tilde{y} coincides with the so-called pseudofitting since it is not directly related to a specific model used for description of the process under study. The amplitudes of the initial fluctuations are reduced by means of an iterative procedure, decreasing the value of the smoothing window. The optimal value w_{opt} is calculated from the relative error RelErr minimization procedure:

$$\operatorname{RelErr}(w) \equiv \left[\frac{\operatorname{StDev}(\Delta y(w))}{\operatorname{StDev}(\tilde{y}(w))}\right],\tag{7}$$

where

$$\Delta y_j(w) \equiv y_j - \vec{y}_j(w), \tag{8}$$



Fig. 5. Experimental installation structure: TS is a temperature sensor, R_{load} is a controllable electrical load, R_{CS} is a current sensor, ADC is an analog-to-digital converter, PXI is a PXI-platform, DAC is a digital-to-analog controller, and LPF is a low-pass filter.

$$\operatorname{StDev}(\Delta y(w)) \equiv \sqrt{\frac{1}{N-1} \sum_{j=1}^{N} \Delta y_j^2(w)}. \tag{9}$$

This method allows the optimal and smoothed trend (the desired pseudofitting function) to be determined and its separation from high-frequency fluctuations without *a priori* information associated with the initial signal. The computer curve-fitting procedure achieves rather high accuracy, and its results can be used for further analysis.

According to the Kramers-Kronig relation, all actions applied to the real part of a function describing an electrochemical system response are also valid for its imaginary part, since there is a single-valued integral connection between them and the system obeys the causality principle. This allows us to consider the proposed approach for diagnosis of lithium batteries as a fully fledged alternative to traditional impedance spectroscopy.

Experimental Installation

To test and verify the method proposed herein, an experimental installation was developed to allow measurement of the transient response of an LB together with simultaneous recording of the temperature during the experiment. The structure of the installation is presented in Fig. 5.

The installation operates according to the following principles: The measuring signals from the current sensor (R_{cs}) and the LB voltage signal are passed to the inputs of an analog-to-digital converter (ADC 1) that is implemented on the basis of a PXI-5922 module (National Instruments Inc.). ADC 1 has 24-bit resolution with intrinsic noise level of 3.4 μ V (r.m.s.) within the range of -10 V to 10 V for sampling frequencies up to 50 kHz. The temperature sensor (TS) is connected to ADC 2, with sampling frequency of 1 Hz. The total error of the temperature-measuring channel does not exceed 1°C. The PXI controller (PXI) controls the electrical





Fig. 7. Variation of parameter $\Delta U'$ for primary lithium cell during discharging process.

load (R_{load}) to provide the required transient processes, processes the measured data, performs all the arithmetic and logical calculations, and provides diagnostic information about the LB under test.

The controllable load is implemented on the basis of a IRFZ44N high-power field-effect transistor to define the electrical mode of the battery. A currentshunting resistance made from a Kantal GS SY with extremely low temperature coefficient of resistance performs the function of the current sensor (R_{cs}) . A digital-to-analog converter (DAC, NI USB-6211) is used to generate the probing signal by changing the transistor operating point. The signal generated by the DAC is passed to the controllable electrical load through a low-pass filter (LPF), which suppresses quantization noise and improves the signal form. A further probing signal goes to the gate of the transistor, which changes the load in accordance with the variation in the control signal. Furthermore, in the experimental installation, the controllable load is allowed to run the battery under test in potentiostatic or galvanostatic mode. This function is implemented on the basis of a standard proportional-integral-derivative (PID) controller methodology and is not considered in detail here. More detailed information about these controllers can be found, for example, in Refs. 22 and 23.

RESULTS AND DISCUSSION

Primary Lithium Cell

The first series of experimental studies was carried out for a CR2032 primary lithium cell produced by GP Batteries with nominal capacity of 210 mA h and nominal voltage of 3 V. The discharge (Fig. 6) was carried out with current of 3 mA almost to the zero potential of the element. The value of the discharging current was selected to avoid the self-heating effect. During the experiment, temperature variations were no larger than 2° C. All cells were operated within the temperature range of 20° C to 22° C. The same temperature conditions were applied for the secondary battery tests.

Three regions can be distinguished in the discharge curve: small fluctuations (at the initial discharge), developed fluctuations (in the voltage range of 2 V to 0.75 V), and the postfluctuation region. In the charge process, the current produced by the lithium element was modulated using rectangular pulses generated by the controllable electrical load. The amplitude of the modulating impulses was selected as 0.3 mA to keep the element in linear operating mode.

Parameter identification for model (3) gives the following results. It was found that use of the amplitudes $U'_1 = U_1/U_L$ and $U'_2 = U_2/U_L$, reduced by the total voltage variation U_L , was preferable to describe the behavior of the primary LB. The parameters α and β remained almost constant during the discharging process of the cell, confirming correct model selection. The reduced amplitudes U'_1 and U'_2 decreased during the discharge process. Note that these parameters begin to fluctuate when the developed fluctuations appear. It was found that, for evaluation of the charging degree of an element, the value of $\Delta U' = 1 + U'_1 + U'_2$, which is proportional to the ohmic resistance R_2 , can be used.

Analysis of the variation of the parameter $\Delta U'$ for a lithium element during the discharge process (Fig. 7) shows monotonic growth, suggesting the possibility of considering it as an indicator of the discharge level of the element. Investigations conducted on lithium cells with different residual charge showed that, even in the case of restoration of the open-circuit potential, the parameter $\Delta U'$ indicated the remaining cell power with high confidence.

Several batteries demonstrated the same behavior, confirming the reliability of the proposed approach. The obtained results indicated that the transient response can be considered as a diagnostic feature reflecting the current technical state of the LB. The possibility of evaluating the remaining cell power was also established. It was found that the proposed evaluation method is faster and more

 Table I. Parameters of Li-Po battery (LP 502540)

Parameter	Value
Nominal voltage (V)	3.7
Capacity (mA h)	450
Operating temperature (°C)	0 - 45
Standard discharge current (mA)	90
Maximum discharge current (mA)	900
Cutoff voltage (V)	2.75



Fig. 8. Variation of fitting parameters U_2 (a) and β (b) of the used LB for different SOC: 100%, 50%, and 0%.

reliable in comparison with the convenient method based on open-circuit potential measurement. The obtained information can be used for more accurate detection of the current battery state in situations when the battery should be replaced.

Although a BMS is not usually used for primary cells, for primary batteries in critical applications, nondestructive testing is required to prevent malfunction and estimate remaining battery life. Therefore, application of such an inexpensive diagnostic tool could be reasonable for this type of battery.

Secondary Lithium Cell

An experimental study of a lithium-polymer (Li-Po) battery (LP 502540, produced by Robiton Inc.) with capacity of 450 mA h and nominal voltage of 3.7 V was also carried out. The parameters of the used battery are presented in Table I. The charging



and discharging process was carried out in galvanostatic mode with charging current of 100 mA and discharging current of 25 mA. The amplitude of the current step was 2.5 mA.

It was established that the transient response could also be described with high accuracy using model (3). Figure 8 shows that the parameters of the second exponent U_2 and β varied slightly at high charge levels, and started to change greatly when approaching the fully discharged battery state. When the battery was discharged, the parameter U_2 (Fig. 8a) increased by a factor of more than 2, indicating an increase in losses due to the influence of charge-transfer processes in the near-electrode region. The almost twofold decrease of the parameter β (Fig. 9b) is a sign of the slowing down of the charge-transfer process. These results are in good agreement with results authors.^{19,24} obtained by other

However, analysis of Fig. 8 revealed strong fluctuations in the behavior of the fitting parameters that impede their use for identification of the current technical state of the battery. It is believed that the origin of these strong fluctuations is the complexity and instability of the double-exponential fitting procedure realized in accordance with model (3). For practical applications, this technique should therefore be improved.

Model (3) shows that the step transient response comprises three components: an instantaneous voltage step and two exponential relaxation processes. The results of the standard fitting procedures suggest the conclusion that the exponential factors of the two relaxation processes differ from each other by more than 30-fold; For example, $\alpha \approx 30$ and $\beta \approx 0.8$ for the full-charge mode. This difference increases during battery discharge. This fact allows one to apply two independent fitting procedures, one for each exponential process. Time division can be applied in this situation due to the fact that the first exponent with factor α is located at the initial



Fig. 10. Dependences of fitting parameters U_1 (a), U_2 (b), α (c), and β (d) on battery open-circuit potential V_{bat} for two battery examples (battery 1 and battery 2).

section of the transient response curve, whereas the second exponent with factor β predominates when the first one is negligible. Analysis of the derivative of the experimental transient response (Fig. 9) allows the conclusion that the first exponent is localized within the time interval of less than 100 ms. This observation is confirmed by the fact that the obtained value of the exponential factor α for the value of 100 approximately corresponds to the value of $3\alpha^{-1}$. In other words, it is possible to carry out two independent fitting procedures in the different time regimes before and after $t = 3\alpha^{-1}$. This assumption implies that one of the relaxation processes becomes negligible and the secondary relaxation process remains relatively accurate if the exponential factors satisfy the criterion $\alpha \gg \beta$. The root-mean-square error of the fitting procedures did not exceed 10 μ V for all analyzed measurements, confirming the high accuracy of the proposed method.

The dependences of the fitting parameters for the case of the battery open-circuit potential were obtained in accordance with the approach described above (Fig. 10). These dependences confirm the previous results that the second (slower) exponential response is more sensitive to the discharge

processes. The amplitude (Fig. 10a) and exponential factor (Fig. 10b) of the first exponent do not show any significant variation during the discharge process. The amplitude of the second exponent increases more than twofold, while the preexponential factor decreases twofold when the open-circuit potential is decreased from 4.1 V to 3.1 V (Fig. 10a and b). The data for two batteries are presented in Fig. 10. Analysis of these data shows that their behavior is similar to each other. The small differences can be explained by the nonideality of the battery manufacturing process. It can be proved that the effect described above is not connected with the chosen battery. The obtained outcomes coincide with the results of other authors; For example, Rahmoun and Biechl²⁵ showed that the internal resistance increases during discharging. In Fig. 10d, one can see this as an increase of the second exponent amplitude. The same results can be found in Ref. 26, where the impedance curves and the results of their analysis allowed it to be proved that decreased SOC leads to a change in the lowfrequency part of the impedance.

The specific behavior of the fitting parameters can be considered to provide diagnostic features that determine the state of charge (SOC) of a secondary lithium battery. These diagnostic features are more convenient in comparison with the open-circuit voltage (OCV), because the OCV is restored over a sufficiently long period after disconnection of a given load. In contrast, the parameters U_2 and β can be found during operation, without load disconnection or involving any time-consuming processes.

The additional advantage of the proposed method consists in the inexpensive equipment required to implement the proposed diagnostic method. The transient response can be generated by a small additional load commutated in parallel with the main load by means of a transistor operating in switching mode. The measurement equipment requires only an ADC with high input impedance to prevent its possible influence on the measured results. It is well known that the majority of commercially available secondary lithium batteries include electronics to prevent overcharge and strong discharge, so introducing the proposed electronics will not significantly increase the total cost of the power source. Furthermore, for large power sources comprising a large number of lithium elements, the same measurement equipment could be commutated between many elements to test them sequentially one by one.

As shown above, the transient response allows estimation of the SOC using the parameters of the second exponent of the model (3). This component corresponds to relatively slow (low-frequency) processes inside the battery, such as diffusion, and thus is expected to be sensitive to the SOC. The first exponent reflects faster processes of charge transfer corresponding to the higher frequency range. It is believed that the parameters of the first exponent could be sensitive to the specific implementation of the battery (electrode materials, battery geometry, etc.), as shown by the results of other authors; For example, some authors 2^{26-28} have shown that the parameters of the high-frequency part of the electrochemical impedance spectrum are sensitive to the electrode materials. Reference 29 allows the assumption that the proposed approach can be applied to estimate the aging processes within lithium batteries.

The reliability of the proposed method is also confirmed by the reliability of impedance spectroscopy, as demonstrated in numerous studies. The proposed strategy represents a new method for its measurement. Increased reliability for nonstationary systems could be achieved using the proposed method due to the better time localization of the measurement procedure.

This method would also enable diagnosis of a power source during operation, as it only requires a small short-time step-like variation of the load or current, which could be implemented using a simple electronic switch commutating an additional small load. We have demonstrated the possibility of using two simple fitting procedures instead of one complex one, allowing implementation of the method on simple, low-speed microprocessors. Therefore, the low computational cost and relative simplicity and inexpensiveness of the measurement equipment would allow integration of the proposed method in an existing BMS.

It should also be mentioned that all the results presented herein were obtained using a step-like test signal; however, this approach could be generalized to a probing signal of arbitrary form, representing a possible basis for future development of simple diagnostic systems based on the natural variation of a given load and carried out during the normal operation of the battery as a diagnostic feature.

CONCLUSIONS

An experimental study of the transient response of lithium batteries to a step disturbance of the applied current or voltage was carried out. The results showed that the transient response reflected physical and chemical processes occurring inside lithium batteries and could be used for diagnostics of their technical state.

It is proposed that such measurement of the transient response of a lithium battery could be used as an alternative to electrochemical impedance spectroscopy. Based on the Kramers–Kronig relationship, we suppose that, for a linear system, the transient response contains the *same* information as the impedance curve. In the presented method, the amplitude of the probing signal is limited, which should allow the system to remain in the linear mode. However, note that the same limitations should be taken into account for pure electrochemical impedance spectroscopy.

It was shown that the transient response can be described by a function comprising an instantaneous step and two exponential relaxation processes, which can be represented by an equivalent circuit in the form of a second-order RC circuit. The results of the experiments presented herein prove that the transient response is sensitive to the operating mode and technical state of the battery. It was found that a reduced instantaneous voltage step response may serve as an indicator of primary lithium battery discharging. The possibility of using the second exponential parameters U_2 and β to estimate the state of charge was also established for secondary lithium batteries. Application of the proposed approach to a primary battery is reasonable for a number of critical applications. In this case, the corresponding experimental equipment could be embedded into the object powered by the primary battery. The diagnostic features could potentially be enhanced by means of high-frequency components; this will be one of the most promising directions for future research. Analysis of the experimental data suggested that the transient response is a prospective diagnostic tool. The simplicity of the testing equipment indicates that it would be possible to embed the required circuitry into the battery without significantly increasing its cost.

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