A parameter estimation method for a zinc-nickel-single-flow battery

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ABSTRACT

Battery modeling is important for the battery management systems of zinc-nickel-single-flow batteries in which energy storage systems are applied to enhance the stability of power systems for new energy applications. To create a precise model, parameter identification and the model structure are studied for the case of a dynamic working current. First, a second-order equivalent circuit model is used to evaluate the influence of different fitting data on parameter identification, and then, a systematic comparison study of the models is conducted in which the parameters are identified based on different fitting data. Second, to further validate the accuracy of the proposed model, a dynamic stress test is performed. The simulated voltage and experimental voltage results demonstrate that the established battery model, in which the parameters are identified by the terminal voltage of the constant-current discharge curve, is suitable for simulating the dynamic characteristics of a zinc-nickel-single-flow battery under dynamic current loads.

I. INTRODUCTION

Along with the progress of mankind and global economic development, the demand for energy is continually increasing. Many countries are thus beginning to turn their attention to renewable energy resources. However, renewable energy resources have many disadvantages, such as randomness and volatility, which limit their large-scale application to power grids. Since an energy storage system could compensate for power fluctuations and improve the grid-connection ability of intermittent power by absorbing or releasing power, it is recognized as a key technology for the development of renewable energy resources. The zinc-nickel-single-flow battery is considered to be a particularly promising system for grid-scale energy storage because of its greenness, environmental friendliness, low-cost, and simple structure (without membranes).

Substantial research has been carried out since the zinc-nickel-single-flow battery was first proposed. The majority of these works have focused on battery performance, such as the influence of electrolyte additives, zinc ions in the electrolytes, and zinc morphology. However, a variety of issues remain to be resolved before the zinc-nickel-single-flow battery can be utilized in industrial applications. Battery modeling is important for the development of battery technology. The state of charge (SOC) can only be assessed by applying advanced battery modeling and estimation techniques, and accurate estimation of the SOC is one of the core functions of battery management systems (BMSs).

In recent years, many modeling studies on vanadium redox flow batteries and lithium batteries have been performed using both electrochemical models and equivalent circuit models (ECMs), but modeling studies on the zinc-nickel battery are relatively lacking. Arise et al. established a two-dimensional electrochemical model for a zinc-nickel battery and studied the variations in the zinc surface morphology by simulation; Liu et al. proposed a mathematical model for a zinc-nickel-single-flow battery that was used to simulate the performance of nickel electrodes; and Yao et al. established a two-dimensional transient model and a three-dimensional stationary model. The first model was built according to the conservation principles of mass, momentum, and charge and was used to study the influence of the circulating flow rate and current density on the voltage change. The second was built on a kinetic model for the chemical reaction between hydroxide and zinc ions and was applied to describe the performance of a zinc-nickel-single-flow battery.
battery cell stack. The type of model established by the above research was an electrochemical model, which is based on the mechanisms of the electrochemical reactions of batteries, making it possible to simulate the inner chemical and physical processes. However, significant computational power is required to solve the partial differential equations (PDEs); thus, the ECM is more suitable for battery design. The ECM does not consider the reaction processes inside the battery but instead uses electronic components, such as resistors and capacitors, to describe the dynamic and static characteristics. Compared to the electrochemical model, the lower computational cost of this model makes it more suitable for simulation and computation in embedded microcontrollers. Although second-order Thevenin ECMs have already been established for zinc-nickel-single-flow batteries,\textsuperscript{16–18} the established models were validated only under constant-current loads. Thus, the ECM of zinc-nickel-single-flow batteries requires further investigation.

In this paper, the influence of the experimental design and parameter identification methods on the accuracy of the model is analyzed and discussed. First, considering the balance among the generalization performance of the model, the computational requirements of the hardware, and the stability of the algorithm, the second-order ECM is selected. Second, to evaluate the influence of different fitting data on parameter identification, an experiment is designed and different experimental datasets from the experiment are used to fit the parameters. A systematic comparison study of the simulation results of models based on different parameters is performed. Finally, the accuracy of the different models under dynamic current loads is validated by experiments and simulations. The most appropriate fitting dataset is provided and used to identify the model parameters.

II. EXPERIMENTAL DETAILS AND BATTERY DATASET GENERATION

A. Experimental setup

The dimensions of the positive and negative electrodes are 240 mm \( \times \) 150 mm. The negative electrode is composed of eight pieces of an inert current collector, the positive electrode is nickel oxide, and the electrolyte is 7 mol/l KOH and 0.5 mol/l K\(_2\)Zn(OH)\(_4\). The electrolyte inlet is at the bottom of the battery to ensure the upward flow of the electrolyte, and the flow is maintained by a circulating pump. The nominal capacity is 56 Ah, the charge cut-off voltage is 2.1 V, and the discharge cut-off voltage is 1.2 V. The test platform is a BTS-3000 programmable battery test system produced by Neware, all experimental data (including the current, voltage, and charge and discharge capacities) are collected at a 1 Hz sampling frequency, and all of the tests are run at room temperature.

![Fig. 1](image1.png)

**FIG. 1.** Sampling curves of current and voltage for one cycle: (a) Overall view and (b) close-up view of the first 100 s.

![Fig. 2](image2.png)

**FIG. 2.** Sampling curves of current and voltage for one cycle: (a) Current and (b) voltage.
B. Parameter identification dataset

A pulse discharge test is used to study and analyze the characteristics of the zinc-nickel-single-flow battery. To determine the influence of the different fitting parameters, a pulse discharge test is designed, with steps as follows:

1. Charge the battery at 0.5 C (28 A) until the maximum capacity limit is reached, and then, let it rest for 1 h.
2. Discharge the battery at 0.5 C (28 A) for 10 s, and then, let the battery rest for 40 s.
3. Apply a constant-current discharge of 0.5 C for 5 min and 50 s, and then, let the battery rest for 1 h.
4. Repeat steps 2 and 3 until the lower cut-off voltage of the zinc-nickel-single-flow battery is reached.

It can be seen that the zinc-nickel-single-flow battery discharges 5% of the SOC in a cycle. The sampling curves of the current and voltage for one cycle are shown in Fig. 1.

C. Validation datasets

A model based on MATLAB/Simulink should be used to evaluate the validity of the model. In this work, the validation is performed by a dynamic stress test (DST). The periodic current and voltage profiles are shown in Fig. 2. The data obtained from these tests will be used to compare the accuracy of the different models.

III. PARAMETER ESTIMATION

A. Parameter estimation algorithm

The structure of the second-order ECM is shown in Fig. 3. $V_{OCV}$ is the open-circuit voltage, which is equal to the difference between the positive and negative potentials, and $R_0$ is the internal resistance of the battery, which is used to describe the resistance characteristics of the battery. Two RC networks are used to describe the polarization characteristics of the battery during charging and discharging to simulate the fast and slow dynamic diffusion processes. The values of the electronic device are related to the SOC.

According to Kirchhoff’s voltage law and Kirchhoff’s current law, the electrical characteristics of the ECM are expressed as the following state-space equations:

$$\begin{bmatrix}
\frac{dV_{\text{short}}}{dt} \\
\frac{dV_{\text{long}}}{dt}
\end{bmatrix} =
\begin{bmatrix}
-1/C_{\text{short}} R_{\text{short}} & 0 \\
0 & -1/C_{\text{long}} R_{\text{long}}
\end{bmatrix}
\begin{bmatrix}
V_{\text{short}} \\
V_{\text{long}}
\end{bmatrix} +
\begin{bmatrix}
1/C_{\text{short}} \\
1/C_{\text{long}}
\end{bmatrix} I,
$$

(1)

$$V_{\text{cell}}(t) = V_{OCV}(\text{SOC}) + V_{\text{long}} + V_{\text{short}} + R_0 I,$$

(2)

where $V_{\text{long}}$ and $V_{\text{short}}$ are the polarization voltages that correspond to the two RC networks. Equation (1) is the equation of state, and Eq. (2) is the output equation in which the open-circuit voltage of the battery is the external expression of the electromotive force in the steady state of the battery. The polarization effect can be considered to be completely eliminated when the battery is stationary for a sufficient amount of time. Therefore, the voltage measured after each stationary hour can be regarded as the open-circuit voltage corresponding to the current state. The values under different conditions are shown in Fig. 4(a). The SOC can be calculated by Eq. (3), where $C_p$ represents the battery capacity in Ah. The values of $R_0$ are obtained directly from each pulse cycle by Eq. (4). The three corresponding variables $[V(t_1), V(t_2), I]$ can be obtained from Fig. 1. The values of $R_0$ under different SOCs are shown in Fig. 4(b).

In Fig. 1, the working conditions of the battery can be divided into two categories. For steps t3 to t4 and t7 to t8, no current excitation occurs and they can be regarded as the zero-input voltage response of the two RC networks. Therefore, according to formulas (1) and (2), the terminal voltage response of the battery can be...
calculated by Eq. (5) as follows:

$$\text{SOC} = \text{SOC}(0) + \frac{1}{3600C_p} \int_0^t f(x) dx, \quad (3)$$

$$R_0 = \frac{V(t_1) - V(t_2)}{I}, \quad (4)$$

$$V_{\text{cell}} = V_{\text{OCV}} + V_{\text{short}}(0)e^{-t/\tau_{\text{short}}} + V_{\text{long}}(0)e^{-t/\tau_{\text{long}}}, \quad (5)$$

where $V_{\text{short}}(0)$ and $V_{\text{long}}(0)$ are the polarization voltages of the battery at static initiation and $\tau_{\text{short}} = C_{\text{short}}R_{\text{short}}$ and $\tau_{\text{long}} = C_{\text{long}}R_{\text{long}}$ represent the short-term and long-term time constants, respectively.

For steps $t_3$ to $t_4$ as the measured voltage. The parameters $20\text{ min},$ fitting data $B.$ RC network parameter estimation with different $\text{RC}$ networks is zero and the terminal voltage response of the battery can be expressed as follows [Eq. (6)]:

$$V_{\text{cell}} = V_{\text{OCV}} + IR_{\text{short}}(1 - e^{-t/\tau_{\text{short}}}) + IR_{\text{long}}(1 - e^{-t/\tau_{\text{long}}}) + IR_0. \quad (6)$$

The nonlinear least squares method is widely used to determine system parameters in various fields. This method can also be used to determine the parameters of the estimated charging state model. The cost function $J$ of curve fitting is the sum of the least square errors between the estimated results and the measured data and is limited by the following conditions:

$$J = \min_{R} \sum_{k=1}^{n} \left[ V_{\text{me}}(t_k) - V_{\text{e}}(R, \tau, t_k) \right]^2$$

$$s.t. R_{\text{short}}, R_{\text{long}}, \tau_{\text{short}}, \tau_{\text{long}} > 0, \quad (7)$$

where $t_k$ is the input time series; $n$ is the length of the fitting experimental dataset, which is determined by the selected fitting dataset; and $V_{\text{me}}$ and $V_{\text{e}}$ are the measured and estimated voltages under the current time series, respectively. Because the values of $V_{\text{OCV}}$ and $R_0$ are known, the parameters of the RC network can be obtained by fitting the experimental data and using the equations.

**B. RC network parameter estimation with different fitting data**

Current and voltage curves, as shown in Fig. 1, are used to study the influence of different fitting data. The terminal voltage at a long rest time is often used to identify model parameters, and the rest time used for the fitting is different in different studies, such as 6 min, 20 min, and 2 h. In this paper, the voltage data from time point $t_3$ to $t_4$ are used as the measured voltage. The parameters $V_{\text{short}}(0),$ $V_{\text{long}}(0),$ $\tau_{\text{short}},$ and $\tau_{\text{long}}$ are calculated by curve fitting with formula (5). Assuming that the current through the polarization resistance equals the load current at $t_2$ in the previous discharge process, the polarization voltage reaches its maximum at this time; therefore, the values of $R_{\text{short}}$ and $R_{\text{long}}$ can be obtained by formula (8). The SOC at the 1 h rest time can be taken as constant according to formula (4); thus, the obtained parameters correspond to the SOC(t4) as follows:

$$\begin{cases} R_{\text{short}} = V_{\text{short}}(0)/I \\ R_{\text{long}} = V_{\text{long}}(0)/I. \end{cases} \quad (8)$$

Short-time pulse discharge is also widely used to identify the parameters of a battery, and the segments from $t_5$ to $t_8$ can be used to identify the parameters of the ECM. The current is suspended for 40 s from time point $t_7$ to $t_8$, which can be regarded as the zero-input voltage response of the two RC networks, and the values of $V_{\text{short}}(0),$ $V_{\text{long}}(0),$ $\tau_{\text{short}},$ and $\tau_{\text{long}}$ can be obtained by formula (5) and the nonlinear least squares fitting method. Then, the discharge voltages from time point $t_5$ to $t_6$ are used as the measured data. $R_{\text{short}}$ and $R_{\text{long}}$ can be obtained by substituting $\tau_{\text{short}}$ and $\tau_{\text{long}}$ derived from the zero-input response, into formula (6). Because the discharge time is so short, the SOC during the entire pulse period can be considered unchanged; thus, the obtained parameters correspond to the SOC(t5).

The terminal voltage of the constant-current discharge curve is rarely used for parameter identification, and one of the major reasons is that constant-current discharge over a long period leads to great changes in the SOC. Because the battery parameters vary with the SOC during discharge, the battery parameters change during a constant-current discharge process. A hypothesis for a zinc-nickel–single-flow battery is proposed so that the influence of the parameter changes is less. According to the hypothesis, the voltage and current are acquired from the constant-current discharge period and can be used for parameter identification. As the zinc-nickel–single-flow battery discharges 5% of the SOC in a test cycle, the model parameters $\tau_{\text{short}}, \tau_{\text{long}}, R_{\text{short}},$ and $R_{\text{long}}$ can be assumed as constant to simplify the calculation and the variations in $V_{\text{OCV}}$ and $R_0$ during the discharge period can be calculated by linear interpolation. The terminal voltages from $t_1$ to $t_2$ are used as the measured data, and the curves are fit with formula (6).

Although all the three fitting methods adopt the same model structure, the parameters of the model obtained by fitting are different due to the differences in the fitting datasets; thus, the models obtained with these methods can be regarded as three different models and are denoted as models I–III, where model I is identified by fitting the voltage curve during the rest time, model II is identified by fitting the short-time pulse curve, and model III is identified by a constant-current discharge process.

**IV. PARAMETER VALUES AND ANALYSIS OF THE SIMULATION RESULTS**

**A. ECM parameter extraction results**

The parameter identification results of the three types of battery models are shown in Fig. 5. The SOC that corresponds to the parameters of model III is taken as the value when the battery discharges half of the electricity in one cycle. The parameters for all of the models vary, but all generally increase with decreasing SOC due to mass transport and diffusion limitations, while the parameters fitted by the different methods are quite different. Because the same structure of the battery model is used in the three fitting methods, the differences in the parameter estimates lead to different parameter values of the battery voltage under a dynamic current. Clearly, the curves of the parameters of model III are smoother and less volatile than the others. The $R_{\text{short}}$ in model II is so small that it is almost negligible compared with $R_{\text{long}}.$ The values of $\tau_{\text{short}}$ and $\tau_{\text{long}}$ in model I are larger than those in the other models; thus, the polarization voltage estimation of model I is the last to reach a stable state under constant-current.
discharge conditions, and the terminal voltage is the smallest. The terminal voltage is larger and takes a shorter time to reach a stable state in model II than in model I.

B. Analysis of the simulation results under the one cycle test

To obtain a clearer understanding of the differences among the three models, first, the curves of the current and voltage shown in Fig. 1 are used to evaluate the validity of the three models. The polarization voltage at the beginning of each cycle is already known, so regarding the current and voltage of one cycle as a set of test results can reduce the effect of error accumulation on the simulation accuracy, which simplifies the discussion of the reasons for the error in the three models. A comparison of the three model voltages with the experimental battery voltages is shown in Figs. 6 and 7. Figure 6 shows the voltage estimations and errors of the three models for one cycle, and the corresponding root mean square error (RMSE) and mean absolute error (MAE) are given in Table I. As the rest time and constant-current charge time are longer than the short-time pulse discharge time, it is hard to analyze the characteristics of the established models under the dynamic working condition with Fig. 6 and Table I. Thus, a separate statistical analysis of the simulation of the first 100 s must be performed. A close-up view of the first 100 s is shown in Fig. 7, and the corresponding RMSE and MAE values are presented in Table II.

Although model II uses a short pulse period to fit the model parameters, the MAE of the simulation is the largest because the polarization voltage coincidence between the start time of the plateau and the end time of the discharge is not accounted for in the fitting. The $V_{\text{short}}(t_7)$ and $V_{\text{long}}(t_7)$ obtained by fitting the curves with the rest time are 4.9 mV and 11 mV, respectively, but the $V_{\text{short}}(t_6)$ and $V_{\text{long}}(t_6)$ obtained by fitting the curves with the discharge time are 8.6 mV and 2.1 mV. The reason for the discrepancy may be overfitting; the voltage response of the zinc-nickel-single-flow battery is irregular for several seconds after the current state changes, as shown in Fig. 7. Outliers account for a larger proportion of the fitting data points under a short-time pulse discharge because the fitting data are very few. The amount of fitting data is insufficient to obtain the global characteristics of the model, and the fitting
parameters are more affected by local features. The model over-
remembers the noise characteristics but neglects the real relationship
between the current and voltage.

Model I is established on the premise that the parameters of
the mode remain unchanged when the battery has a long rest time,
and this assumption might be incorrect for a zinc-nickel-single-flow
battery. To further analyze the influence of the length of the rest
time, the parameters are fitted under different rest-time durations.

The time constants and polarization resistors obtained by fitting the
terminal voltage are shown in Fig. 8, indicating that the values of
the time constants and polarization resistors vary with the length
of the rest time. The two time constants present the trend of first
rapidly increasing and then slightly decreasing with the length of
the rest time, and afterwards, they tend to be steady. Thus, it can
be speculated that the time constants increase with the rest time.
Thus, if the time constants are recognized as fixed values and
the parameters of the model are obtained by all of the terminal volt-
ages of the rest time, the estimated time constants will be larger
than the actual values, which will cause the voltage response of
the model to severely lag behind the experimental data. This behaviour
is one of the reasons why the estimated voltage is always larger
than the measured voltage, as shown in Figs. 6 and 7. The two
short and long polarization resistors increase and decrease, respec-
tively, but they gradually converge with the increasing length of
the rest time. It is noteworthy that the decrease rate of \( R_{\text{long}} \) is greater
than the increase rate of \( R_{\text{short}} \); this difference makes the sum of
the polarization resistors in model I smaller than that of the others.
The long-term RC network under the discharge period is unsatu-
rated, which can explain the phenomenon for lithium-ion batter-
ies.\(^{26,27}\) A more plausible explanation is that untenable assumptions
lead to overfitting for the modeling of the zinc-nickel-single-flow
battery.
The MAE of the simulation of model III for the first 100 s is the smallest among the three models, so model III can track the terminal voltage variation of the battery when the current is changed by a short-time pulse. Although the voltage estimation of model III is more accurate under a pulse and a constant current, tracking the terminal voltage variation over long rest times is difficult, as shown in Fig. 6(b). The voltage estimation errors increase with time, and the simulated voltage reaches the equilibrium potential in less time than the real voltage, which means that the time constants of model III are so small that the simulated terminal voltage recovers faster than the real voltage. This result is consistent with the previous analysis.

FIG. 8. (a) Polarization resistance variation with different rest-time durations and (b) time constant variation with the rest-time durations.

FIG. 9. (a) Overall view of the voltage response in the DST and (b) overall view of the voltage error in the DST.

FIG. 10. (a) Close-up view of the voltage response in the DST from 3600 s to 3960 s and (b) close-up view of the voltage error in the DST from 3600 s to 3960 s.
C. Verification of terminal voltage predictions under the DST

Second, in order to further demonstrate the differences among the three models during the dynamic working condition, the DST is used to validate the accuracy of the models. The curves of the simulation results of the three models and the measured voltage are shown in Fig. 9(a), and an enlarged image of one segment from 3600 s to 3960 s is shown in Fig. 10(a). The RMSE and MAE for the entire DST trial period are shown in Table III. The error of model III is the smallest among the three models. Although the time constants increase with the length of the rest time, the influence of this characteristic on the simulation of the battery is small when the rest time span is short, so model III can be used to simulate the dynamic characteristics if the battery is operated only under high-frequency variable-current conditions. As shown in Fig. 9(a), the estimated voltage of model I is greater than the measured terminal voltage most of the time, and the results of model II are the opposite because one time constant is too large while the other is too small. In Fig. 10(b), the voltage estimation error changes abruptly after a change in current because of the characteristics of the zinc-nickel-single-flow battery proposed in Sec. IV B.

V. CONCLUSIONS

The characteristics of lithium batteries and zinc-nickel-single-flow batteries are different, and thus, the use of the same model structure and identification method for the two types of batteries is inappropriate. To build a more appropriate model for the zinc-nickel-single-flow battery, different datasets are used to fit the parameters of the second-order ECM. The difference in fitting parameters is systematically analyzed in this work, and the following three basic conclusions were drawn: (1) The short-time pulse current curve cannot be used to fit parameters due to the influence of outliers. (2) The time constants increase with the length of the rest time, which make the terminal voltage of a long rest time unsuitable for fitting model parameters, but this characteristic has little effect on the use of the two-order ECM for a zinc-nickel battery if the battery is operated only under high-frequency variable-current conditions. (3) Even though the SOC that corresponds to the fitting parameters cannot be accurately determined, the terminal voltage of the constant-current discharge curve can be used for parameter identification because the established model is more accurate and effective than the two other models.

Notably, the proposed model of the zinc-nickel-single-flow battery was built and tested only under high-frequency variable-current conditions. If the current conditions do not satisfy this assumption, such as when the working conditions have longer rest times, the proposed model will likely no longer be applicable; therefore, the subsequent work will focus on the study of the model structure, which will make the model more accurate under different working conditions.

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| TABLE III. Model errors in the DST. |
|---------------------------------|
| Method | Model I | Model II | Model III |
|-------|---------|----------|-----------|
| RMSE (mV) | 7.75 | 7.87 | 4.83 |
| MAE (mV) | 6.11 | 6.12 | 3.26 |