

Article

A Study on Capacity and State of Charge Estimation of VRFB Systems Using Cumulated Charge and Electrolyte Volume under Rebalancing Conditions

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Abstract: Extensive research has been conducted on energy storage systems (ESSs) for efficient power use to mitigate the problems of environmental pollution and resource depletion. Various batteries such as lead-acid batteries, lithium batteries, and vanadium redox flow batteries (VRFBs), which have longer life spans and better fire safety, have been actively researched. However, VRFBs undergo capacity reduction due to electrolyte crossover. Additionally, research on the capacity and state of charge (SOC) estimation for efficient energy management, safety, and life span management of VRFBs has been performed; however, the results of short-term experimental conditions with little change in capacity are presented without considering the rebalancing process of the electrolyte. Therefore, herein we propose a method for estimating the capacity of a VRFB using the cumulative charge and electrolyte volume amount under long-term cycle conditions, including rebalancing. The main point of the estimation method is to design a capacity estimation equation in the form of a power function with the measured cumulative charge of the battery as a variable and to update the initial capacity value applied to the estimation equation with the amount of electrolyte measured at the time of rebalancing. Additionally, the performance verification results of the SOC estimation algorithm using the capacity estimation model were presented using the long-term charge/discharge cycle test data of a 10 W-class single cell.



Citation: Jung, H.; Lee, S. A Study on Capacity and State of Charge Estimation of VRFB Systems Using Cumulated Charge and Electrolyte Volume under Rebalancing Conditions. *Energies* **2023**, *16*, 2478. <https://doi.org/10.3390/en16052478>

Academic Editor: Balakumar Balasingam

Received: 1 February 2023

Revised: 26 February 2023

Accepted: 3 March 2023

Published: 5 March 2023



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Keywords: vanadium redox flow battery (VRFB); energy storage system (ESS); crossover; capacity estimation; state of charge (SOC); rebalancing

1. Introduction

Environmentally friendly new and renewable energy sources, such as solar energy and wind power, are attracting attention due to global warming, environmental pollution, and resource depletion caused by the continuous use of fossil fuels. Owing to intermittent power generation characteristics due to environmental conditions, the utilization of energy storage system (ESSs) for stable and efficient energy use such as frequency regulation and peak load shaving is also steadily growing [1,2]. Currently, lithium-ion batteries with high energy densities are employed in ESSs to maximize the efficiency of energy use [2,3].

However, current battery packs and systems that use lithium batteries have several issues. In fire accidents involving large-capacity batteries in electric vehicles, energy storage devices composed of lithium-ion batteries using flammable nonaqueous organic solvents are vulnerable to fire. In addition, when the battery ignites due to thermal runaway caused by internal and external short circuits, the fire is difficult to extinguish [4–6]. In particular, ESSs manufactured by combining a number of lithium battery cells in series and parallel are vulnerable to fire when the maximum and minimum voltages of the applied battery cells are exceeded and failed cells are detected and managed. Therefore, significant efforts must be made to prevent fires when lithium-ion batteries are used in large-scale ESS applications.

In addition, lithium batteries with poor life spans pollute the environment by emitting large amounts of waste [7]. To solve these problems, additional studies, such as the diagnosis of deterioration and reuse of lithium-ion batteries, have been conducted, but this increases the operational complexity and cost of the system [8,9].

On the other hand, in ESS applications, research on alternative battery systems that can replace lithium-ion batteries, such as the vanadium redox flow battery (VRFB) using an aqueous solvent, has been carried out [10–18]. VRFBs, which use nonflammable aqueous solvents, have a low risk of fire and easy thermal management because the circulation of electrolytes removes heat from the battery cell and stack [19]. In addition, compared to lithium-ion batteries, which have a life span of approximately 10 years with 2500 cycles, VRFBs have a life span of approximately 15 to 20 years and are capable of charging and discharging for more than 10,000 cycles [3,10,11,19]. In the case of the electrolyte, the risk of crossover contamination is low, and it can be used semipermanently because the same electrolyte is used for the anode/cathode; thus, VRFBs are suitable for a long-cycle energy storage system. As shown in Figure 1, the VRFB system consists of two vanadium electrolyte tanks that store positive and negative electrolytes, a fluid pump that circulates the vanadium electrolyte, and a battery cell. The output of the VRFB was determined by the area of the electrode, and the voltage of the system was determined by the number of cells connected in series. Compared to lithium-ion batteries, which increase capacity through parallel connections, the VRFB capacity is determined by the amount of electrolyte. Therefore, VRFBs have the advantage of easily increasing capacity and not having to manage many batteries [11]. However, VRFBs have lower energy density and efficiency than lithium batteries; therefore, VRFB systems require more installation area and initial investment cost to have the same energy capacity as ESSs using lithium batteries. Nevertheless, research on the dimension, morphology, composition, and architecture of electrode materials to overcome the low ionic/electronic conductivity of VRFBs is being conducted, and efficiency is expected to improve [18]. Also, VRFBs are suggested to be more economical as large-capacity long-cycle energy storage devices, because ESSs using batteries require fire prevention facilities and more effort to manage individual cells [12–19].

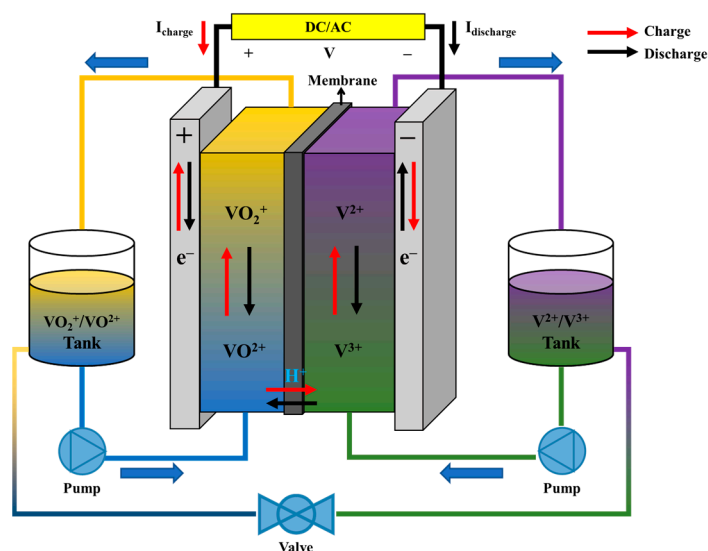
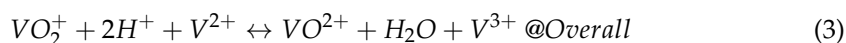


Figure 1. Diagram of a VRFB energy storage system reproduced from [10], Alotto et al., 2014.

In the VRFB system, the positive and negative electrolytes are stored in two separate electrolyte tanks, and the electrolyte is circulated and supplied to cells or stacks through a fluid pump. In the transferred electrolyte, oxidation/reduction reactions occur at the electrode, ions move based on the ion-exchange membrane made of Nafion, and energy is stored in the electrolyte [10–16]. As shown in Figure 1, the oxidation/reduction reaction of the VRFB can be expressed as an electrochemical reaction at the anode and cathode, as

expressed in Equations (1) and (2), and the overall electrochemical reaction is given by Equation (3). When the vanadium electrolyte is charged, V^{3+} ions on the negative electrode are reduced to V^{2+} ions, and V^{4+} ions on the positive electrode are oxidized to V^{5+} ions. During the discharging process, V^{5+} ions are oxidized to V^{4+} ions at the anode, and V^{2+} ions are oxidized to V^{3+} ions at the cathode [10–19].



In the case of a VRFB system, the concentrations of the electrolytes of the positive and negative electrodes change during charging and discharging, and an osmotic pressure phenomenon occurs because of the concentration difference. Therefore, a phenomenon occurs in which water moves through the ion exchange membrane, leading to a crossover phenomenon in which the initially stored equal amounts of positive and negative electrolytes become unequal [13,14,16–21]. In addition, an electrolyte imbalance may occur due to oxidation of the electrolyte when exposed to air or a reaction in which hydrogen is generated. The crossover phenomenon causes a capacity reduction in the VRFB system [12,14,16–21]. Therefore, it is necessary to apply a method capable of recovering the capacity reduced by the crossover phenomenon to maximize the usable capacity of a VRFB system.

Research is being conducted to solve the imbalance of electrolytes caused by crossover. Corcuera and Skyllas-Kazacos presented research results on the causes of crossover [17]. Cunha et al. [19] and Haisch et al. [21] suggested a remixing method in which the anode and cathode electrolytes were mixed to match the initial amount of electrolyte, because VRFBs use the same electrolyte as the anode and cathode. Similar to the previous method, in this study a VRFB cell experiment was set that can rebalance the electrolyte through valve control installed in the flow path between the anode and cathode electrolyte tanks, and then a battery charge/discharge cycling experiment, including rebalancing, was conducted. By applying a bubbler, the inside of the electrolyte tank was purged with nitrogen to minimize contact with air. Figure 2 shows the volume change of the cathode electrolyte at the time of full charge measured during long-term charge/discharge cycling (99 cycles) of the VRFB single cell under the rebalancing condition tested in this study. At this time, the electrolyte volume was measured by image processing of data captured using a camera at 5-min intervals. As shown in the figure, the amount of cathode electrolyte, which was at the level of 38 mL, decreases as cycling proceeds. When the cathode electrolyte was less than a certain amount (29 mL or less in the case of this system) due to crossover, rebalancing was performed by manually controlling the valve connected between the electrolyte tanks, and rebalancing was performed six times. When rebalancing was performed, the amount of electrolyte in the negative electrode increased again, and the battery capacity was restored after the ion stabilization process as charging and discharging proceeded [17,19,21].

Because the state of charge (SOC), which is an indicator of the remaining charge of the battery, cannot be directly measured, it is estimated through various methods. If the SOC of the battery is not accurately estimated, the energy cannot be efficiently managed, and the aging of the battery can be accelerated as the battery is overcharged and discharged. Therefore, as mentioned above, because the capacity of the VRFB system varies depending on the amount of electrolyte, the capacity and SOC estimations are required simultaneously. However, there are few research results on estimating the capacity and SOC of VRFBs considering rebalancing.

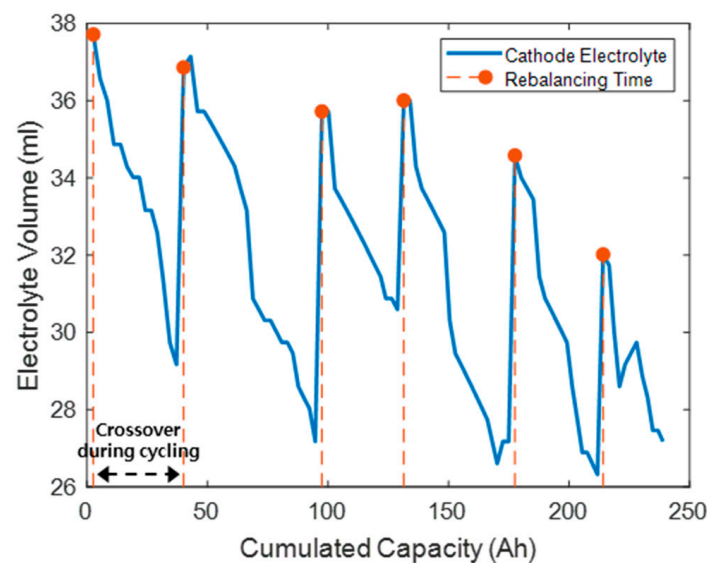


Figure 2. Volume change of VRFB cathode electrolyte.

Studies related to the capacity and SOC estimations for VRFB battery systems are as follows: after modeling the VRFB cell or stack as an electrical circuit model, estimation methods have been developed using an extended Kalman filter (EKF) or neural network [22,23]. However, previous studies have only presented estimated results for short-term experiments without a significant reduction in capacity. In addition, a method for estimating the energy SOC using the ampere-hour counting method after expressing the capacity fade as a function of charge and discharge cycles was proposed [24]. However, this study did not address electrolyte crossover and rebalancing. Although the cycle count was defined as an independent variable for capacity reduction, it was difficult to determine the application criteria for the cycling count. In another study, a method for estimating SOC by measuring the potential of the anode and cathode using a reference cell or half-cell was proposed [17,21]. Corcuera and Skyllas-Kazacos installed half-cells on the anode and cathode, respectively, and then monitored them, analyzed the SOC of the half-cell electrolyte, and conducted research on methods to detect electrolyte imbalance and restore capacity [17]. Haisch et al. conducted a study to estimate capacity using the Nernst equation; open circuit potential (OCP) was measured after installing half-cells in positive and negative electrolytes, and the rebalancing timing according to the electrolyte imbalance was determined [21]. As reported in a previous study, the capacity estimation method using a half-cell or reference cell requires an additional cell to measure the potential of the electrolyte solution for SOC and capacity estimations. In addition, there is a problem implementing it in real time because the additional equipment/sensors and chemical analysis for analyzing the ion concentration of the electrolyte solution are required.

Therefore, in this study we present the results of capacity estimation based on the amount of cathode electrolyte and the cumulative charge used in the VRFB, considering electrolyte rebalancing. The proposed capacity estimation model is superior in terms of cost and ease of implementation, because it sets the initial capacity at the time of rebalancing by using the amount of electrolyte measured using image processing. Further, the method uses the accumulated charge amount of the measured current as an input variable. The proposed capacity estimation model was verified from the cumulative charge test results of 532.7 Ah, which is the result of 99 cycling experiments of a 10 W-class VRFB single cell and has an error performance of within 5% on average. In addition, the SOC estimation algorithm based on the ampere-hour counting method, to which the capacity estimation method proposed in this study was applied, could estimate the energy SOC within $\pm 5\%$ error during the charge/discharge test cycle.

2. VRFB Electrical Characteristics Analysis

2.1. VRFB Experimental Setup

The configuration of the VRFB single-cell charge/discharge experiment in this study is shown in Figure 3. The ambient temperature was maintained at 25 ± 3 °C. The VRFB system used in the experiment consisted of a 10 W-class VRFB cell, anode and cathode electrolyte tanks, a diaphragm fluid pump to circulate the electrolyte in each electrolyte tank, a manual valve connected between tanks for electrolyte rebalancing, and a bubbler for nitrogen purge. In addition, a camera was used to measure the amount of electrolyte, and a data logger was employed to measure the temperature, current, and voltage of the VRFB cell. Table 1 lists the specifications of the cell and the major components used in this experiment.

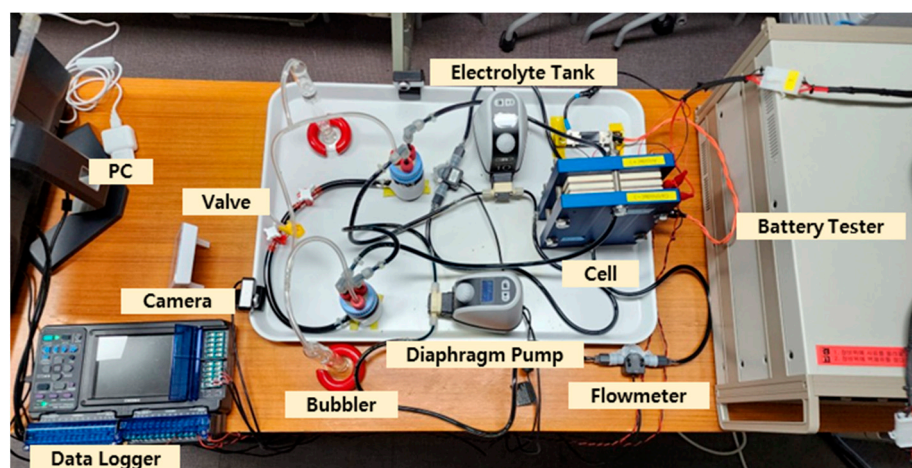


Figure 3. VRFB single-cell system configuration.

Table 1. Specification for VRFB single cell.

10 W VRFB Single-Cell System		
VRFB cell	Membrane	Nafion-212
	Electrolyte	1.6 M V3.5+ / 2 M H ₂ SO ₄
	Electrode Area	100 cm ² (10 × 10 cm)
	Electrode	Carbon Felt
	Electric Collector	Brass
Diaphragm pump		SIMDOS 10
Electrolyte volume		45 mL in anode and cathode tanks
Battery simulator		Maccor 4300 K

Nafion-212 was applied to the ion-exchange membrane of the cell, and the electrode area was 100 cm² (10 × 10 cm). In addition, 1.6 M V3.5+ / 2 M H₂SO₄ was used as the vanadium electrolyte, and 45 mL was equally distributed in each electrolyte tank of the anode and cathode during the experimental setting. A diaphragm fluid pump was used to transfer the positive/cathode electrolyte to the electrodes and was operated at a flow rate of 50 mL/min. The VRFB cells were charged and discharged using a Maccor 4300 K battery cycler with a maximum voltage/current specification of 5 V/15 A.

2.2. VRFB Charge and Discharge Profile

A VRFB charge/discharge cycle experiment was conducted, as shown in Figure 4, to satisfy the charge/discharge conditions provided by the cell manufacturer. The battery was fully charged under constant current (CC)–constant voltage (CV) conditions of 1.6 V/3 A (cutoff current 0.15 A). The battery was discharged for 4 min at a current of 3 A and then rested for 5 min, which was repeated until a cutoff voltage of 0.8 V was reached, to measure the battery's electrical parameters and open-circuit voltage (OCV) by the SOC. The afore-

mentioned full charge and full discharge profiles were continuously performed 10 times per experiment. Figure 5a shows the voltage and current waveforms of 10 charge/discharge cycling experiments, and Figure 5b illustrates the expanded waveforms of the first cycling results. Figure 5c shows the charge and discharge capacity results measured during the 10-cycle experiment. In the figure, if the VRFB undergoes a long charge/discharge cycle, the capacity continuously decreases, and thus, a rebalancing process is required to recover the capacity.

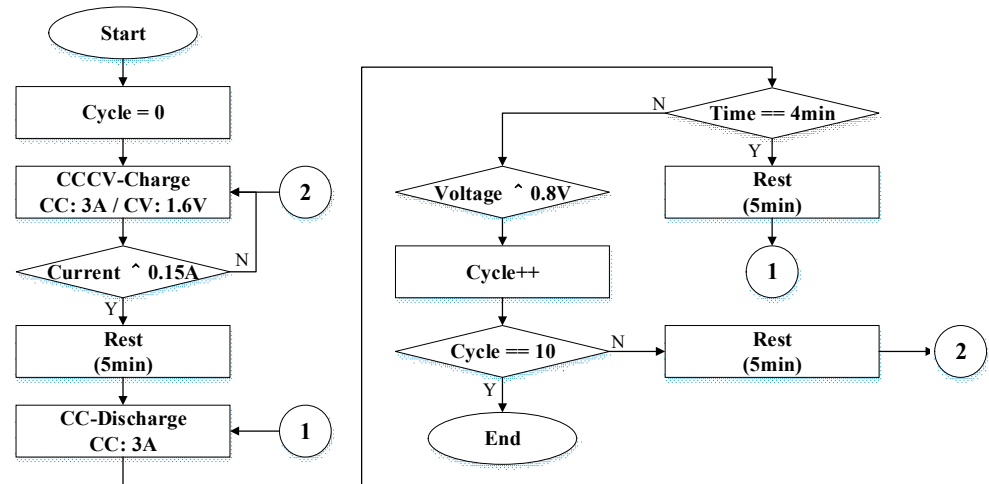


Figure 4. VRFB charging and discharging cycling procedure.

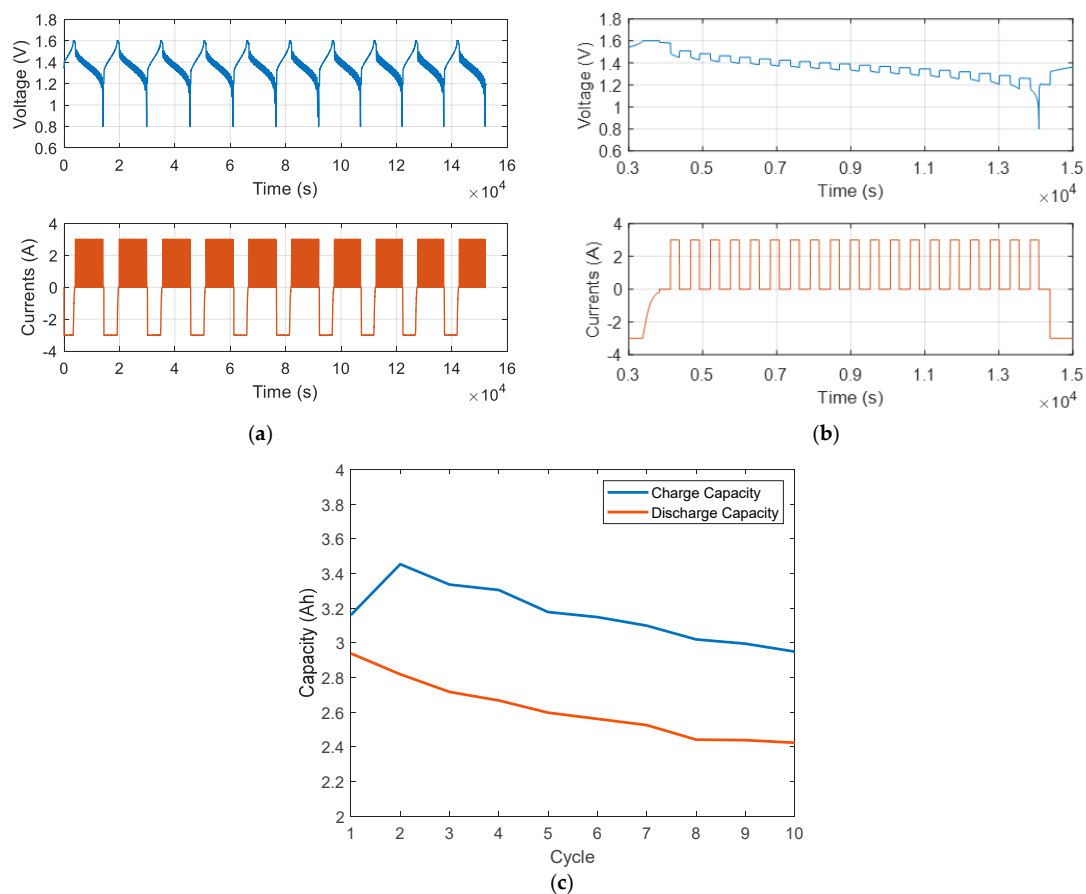


Figure 5. VRFB charge/discharge experimental results. (a) Voltage/current results during 10 cycles, (b) enlarged voltage and current waveforms of the first cycle, and (c) charge/discharge capacity during 10 cycles.

3. Capacity Estimation Methodology of VRFB System

3.1. Capacity Variation during Charge/Discharge Cycles Including Rebalancing

Figure 6 shows the charge and discharge capacities and Coulombic efficiency for the 99-cycle experiment of a VRFB single cell, including 6 rebalancing processes. Figure 6a shows the changes in charge and discharge capacities. The charge and discharge capacities initially decrease exponentially starting from a discharge capacity of 2.634 Ah (charge capacity of 4.184 Ah), but the capacity recovers upon electrolyte rebalancing, which is indicated by the yellow dots. Table 2 summarizes the sizes of the charge and discharge capacities at each rebalancing time. The Coulombic efficiency, which is the efficiency of the discharge capacity versus the charge capacity of the battery, shows an average efficiency of 81.5%, excluding the temporary decrease in efficiency at the time of rebalancing and the time of charging and discharging after long-term rest. The decrease in capacity during the 99 cycles is due to evaporation of the electrolyte; however, as previously mentioned, the capacity recovers based on the rebalancing time point and decreases as the cycling proceeds. Therefore, in this study a capacity estimation model was designed using the amount of electrolyte and accumulated charge based on the results of this experiment.

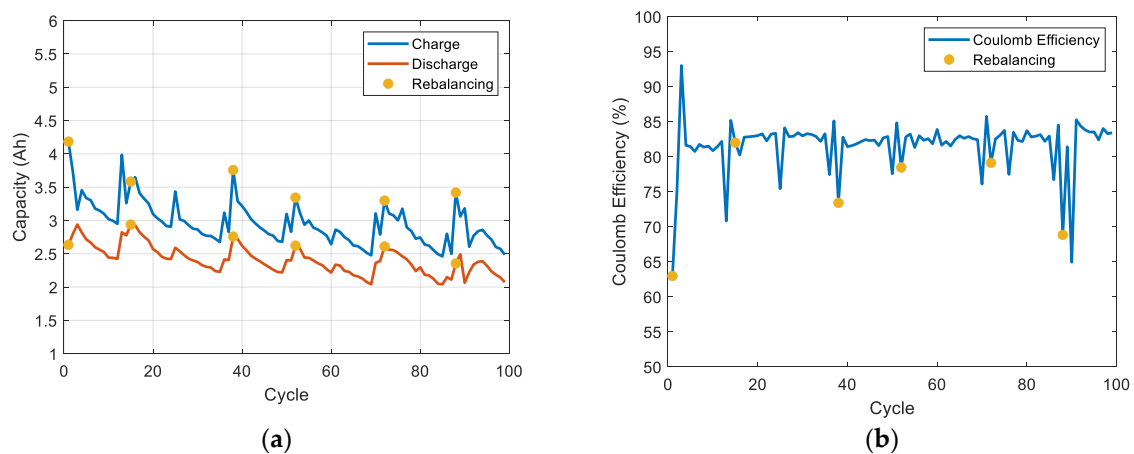


Figure 6. Capacity and Coulombic efficiency during 99-cycle charge/discharge experiment of VRFB cell. (a) Charge/discharge capacity result, and (b) Coulombic efficiency result.

Table 2. Charge and discharge capacity at the time of rebalancing.

Rebalancing Points	Charge Capacity (Ah)	Discharge Capacity (Ah)
#1	4.184	2.634
#2	3.584	2.938
#3	3.757	2.758
#4	3.343	2.623
#5	3.298	2.609
#6	3.420	2.354

3.2. Capacity Estimation of VRFB Using Cumulative Charge and Electrolyte Volume

In this study, we proposed a capacity estimation model in which capacity degradation occurs based on the accumulated charge amount in the form of a power series, as shown in Equation (4), for capacity estimation. In Equation (4), parameters A and B are variables for modeling the rate of decrease in the initial capacity as charge accumulates and are determined by the VRFB battery characteristics. Parameter C is an initial capacity value and is a parameter determined by the amount of electrolyte stored in the electrolyte tank. The *cumulated capacity* represents the cumulative charge amount due to the current used.

$$\text{Capacity} = A \times \text{Cumulated Capacity}^B + C \quad (4)$$

The proposed capacity estimation model was designed using experimental data from the first to fourth rebalancing cycles. The parameters of the capacity estimation model for the first 3 cycling sections from the time of rebalancing to the time of the next rebalancing were estimated using the least-squares method, and the capacity estimation results are shown in Figure 7. Parameters A and B were set to the average values of the parameters extracted for each section, and constant C was initialized by scaling the discharge capacity for the electrolyte measured at the time of rebalancing, as shown in Figure 8. The scaling value was determined through an analysis of the experimental results, and in this study it was 1.06. In addition, the cumulative capacity was initialized to zero when rebalancing was performed. Table 3 lists the parameter values of the capacity estimation equation mentioned above.

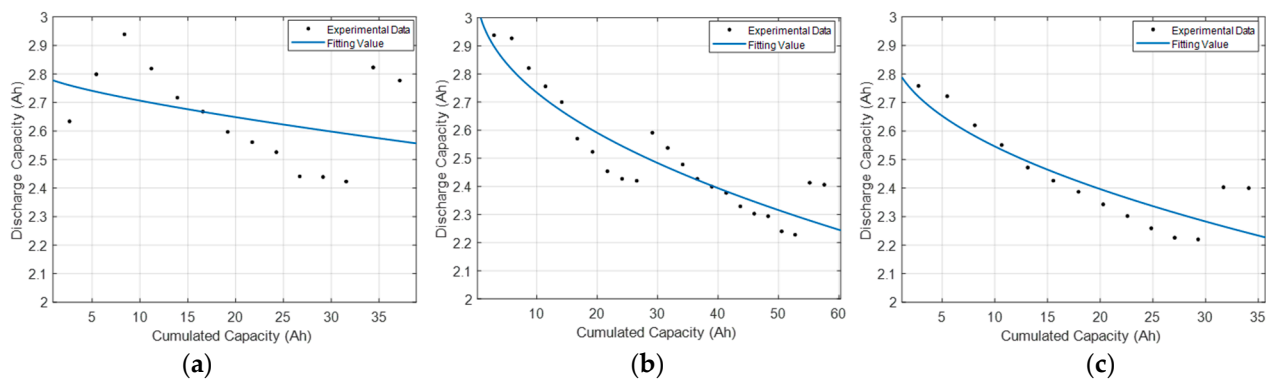


Figure 7. Estimated capacity by discharge interval after rebalancing. (a) First rebalancing period, (b) second rebalancing period, and (c) third rebalancing period.

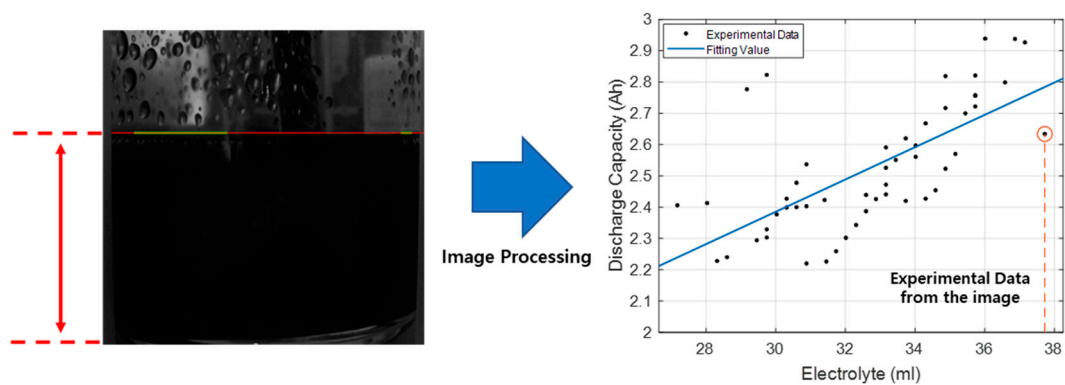


Figure 8. Discharge capacity according to the volume of the cathode electrolyte.

Table 3. Parameters of VRFB capacity estimation model.

$Capacity = A \times Cumulated\ Capacity^B + C$	
A	-0.0905
B	0.5626
C	$(0.0517 \times Electrolyte\ Volume + 0.8349) \times 1.06$

Figure 9 shows the capacity estimation results that reflect the estimated parameters. Figure 9a shows the measured actual discharge capacity and estimated capacity. The capacity estimation model can simulate the capacity decrease as the cycling proceeds, and the capacity is estimated by updating the value of the constant C of the capacity estimation model even at the time of rebalancing, which is indicated by a yellow dot. The capacity estimation result shows a large error of 18.9% at the point where the accumulated charge is 34.38 Ah, and the actual capacity is estimated within an error of 4.94% on average.

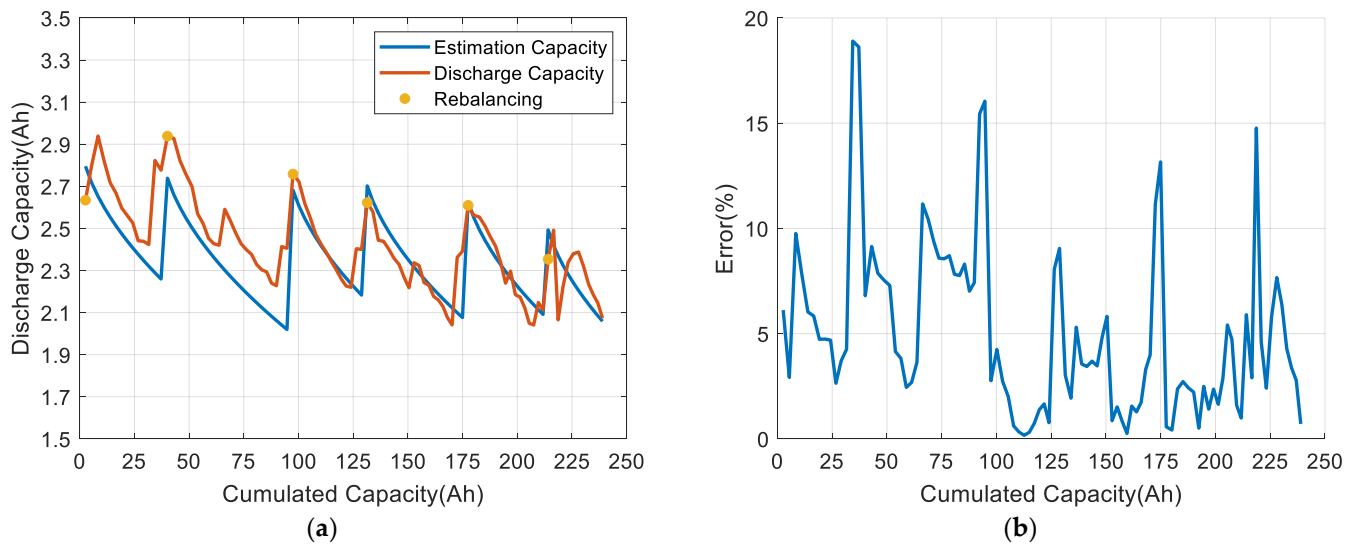


Figure 9. Proposed capacity estimation results. (a) Measured/estimated capacity, and (b) percentage error.

4. State of Charge Estimation Algorithm

The SOC of the battery is estimated using the ampere-hour counting method in terms of ease of implementation and reliability, and a method of resetting the SOC using an OCV is applied to overcome the disadvantages of the ampere-hour counting method [25,26]. In this section, we present the SOC estimation algorithm to which the proposed capacity estimation method was applied. The performance of the proposed method was verified through simulations using the experimental data of the measured voltage and current.

4.1. SOC Estimation Algorithm Based on the Ampere-Hour Counting Method

Figure 10 shows a flowchart of the SOC estimation algorithm based on the VRFB ampere-hour counting method proposed in this study. When the algorithm begins, the accumulated capacitance and SOC are initialized to the previous values, and the voltage, current, electrolyte amount, and rebalancing flag of the VRFB cell are sensed. Then, the capacity of the battery is updated using the proposed capacity estimation model in Equation (4) after calculating the cumulative capacity (Ah) from the measured current. When rebalancing is performed, the constant C value of the capacity estimation model is updated using the measured amount of the electrolyte. The SOC of the battery was calculated using the ampere-hour counting method, considering the charging efficiency (denoted by η). The charging efficiency in this study was set to 81%, as previously mentioned.

4.2. Simulation Result

The performance of the proposed capacity and SOC estimation algorithm was verified from the experimental results of 99 charging and discharging cycles. In this section, we present the estimation results for 15–24 cycles with large capacity estimation errors and 76–85 cycles with small errors.

Figure 11 shows the capacity and SOC estimation results for the 15–24 cycles. Figure 11a shows the measured voltage and current waveforms from the experiment, and Figure 11b shows the measured real capacity (blue solid line) and estimated capacity (red solid line) from the experiment. The capacity was estimated to be within the maximum error of 0.2675 Ah compared to the actual capacity measured in the experiment. Figure 11c,d illustrate the actual SOC, which is denoted as real and estimated SOC and estimation error, respectively. Based on the figure, the maximum error is 9%, and the root mean squared error (RMSE) error performance is 3.97%.

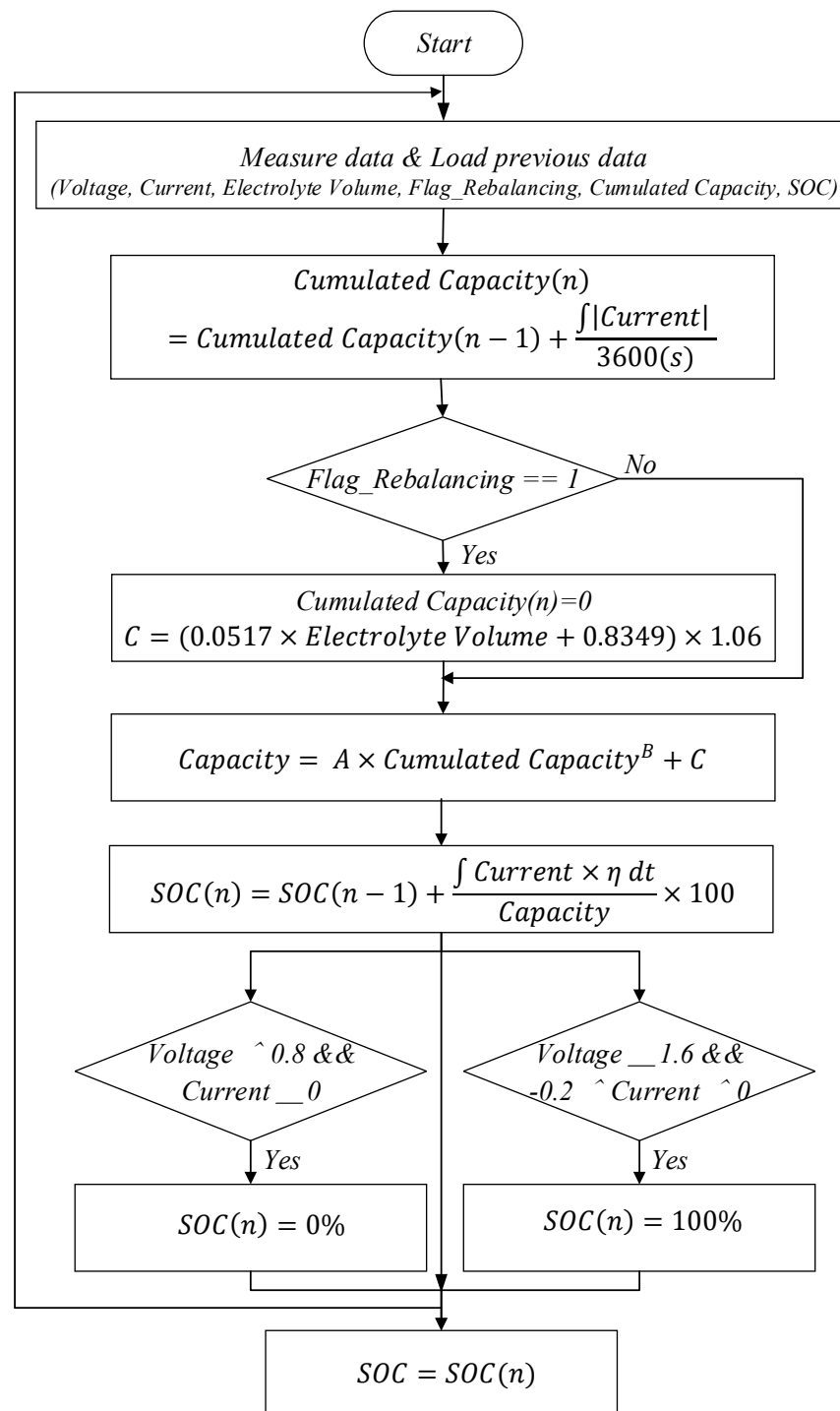


Figure 10. Flow chart of VRFB's SOC estimation algorithm based on the ampere-hour counting method.

Figure 12 shows the capacity and SOC estimation results for the 76–85 cycles. Figure 12a shows the measured voltage and current waveforms in the experiment, and Figure 12b shows the measured actual and estimated capacities. The capacity can be estimated within an error of 0.0596 Ah. Figure 12c,d show the actual and estimated SOC and error, respectively. From the figure, it can be confirmed that the maximum error is 5% and that the SOC is estimated within the RMSE error of 1.82%.

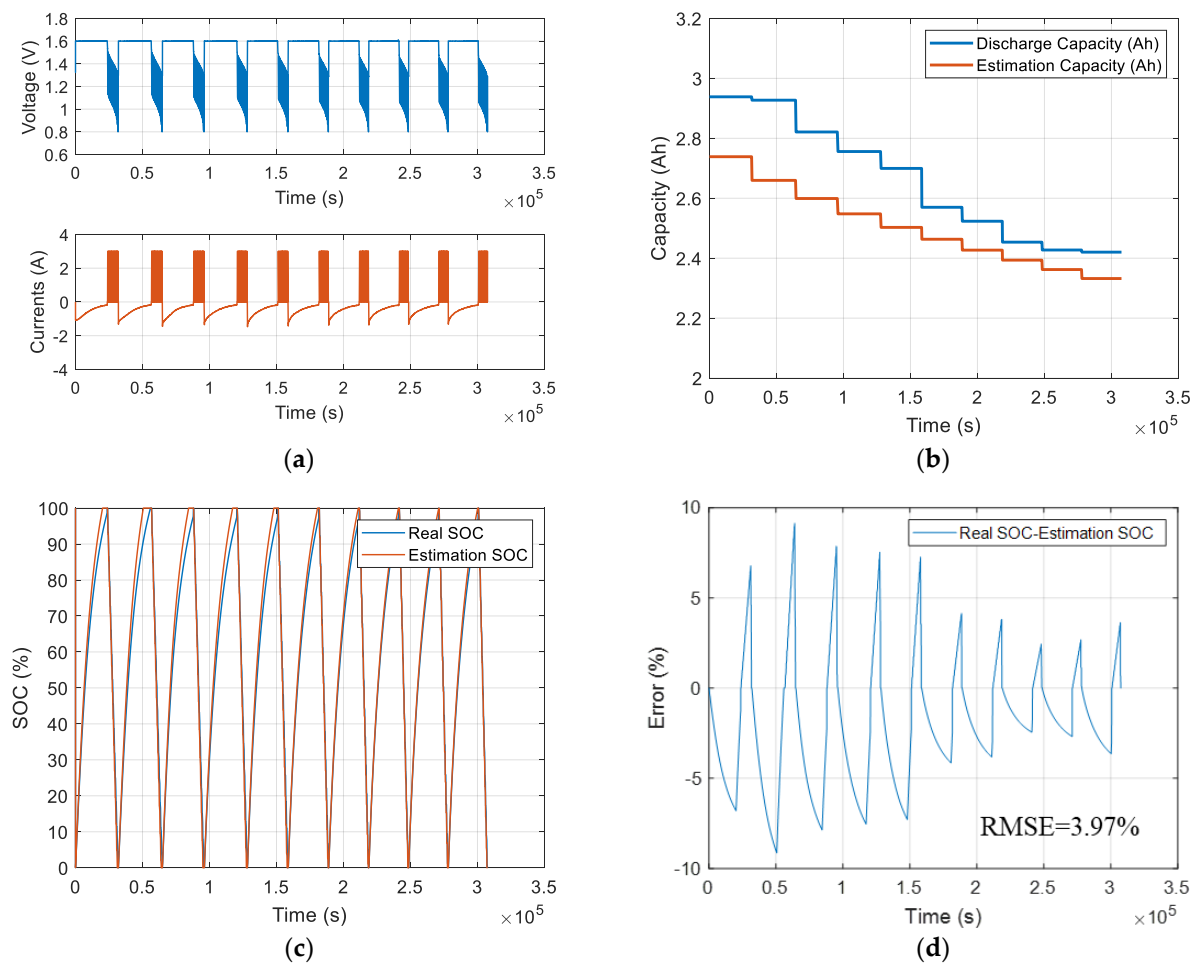


Figure 11. Capacity and SOC estimation results of the VRFB cell during 15–24 cycles (cumulated capacity 40–64 Ah). (a) voltage and current waveform, (b) capacity estimation result, (c) SOC estimation result, and (d) SOC error.

The estimation performance of the proposed method shows an average error performance of 4.94% for capacity and 3.97% for SOC. The results of this study have the following differences compared to the results of previous studies. Wei et al. presented a capacity estimation error of 3.06% and SOC error performance of 0.69% using the estimator of the extended Kalman filter [22]. Although it shows higher accuracy than this study, additional research is needed under long-term cycle conditions, because the study only presented estimation performance for short-term cycles in which capacity fade did not occur. Cao et al. conducted a test on a total of 20 short-time samples using 2 probabilistic neural network models and presented an average error performance of 5.4% for capacity and 2.3% for SOC [23]. Although Cao et al.’s result is similar to the performance of the results of the present study, it is necessary to additionally analyze the performance of the estimation algorithm for a long continuous time because the neural network was evaluated for each short period in this study.

As mentioned above, this study presented a method for estimating capacity and SOC using variables of the amount of electrolyte in addition to voltage and current while rebalancing the electrolyte, and the algorithm is considered superior to previous studies because it was verified under long-term cycle conditions. To improve the performance of the proposed capacity estimation model, additional research on designing a deep learning model using experimental data of voltage, current, rebalancing time, and amount of electrolyte can be conducted and will be conducted in the future [27,28].

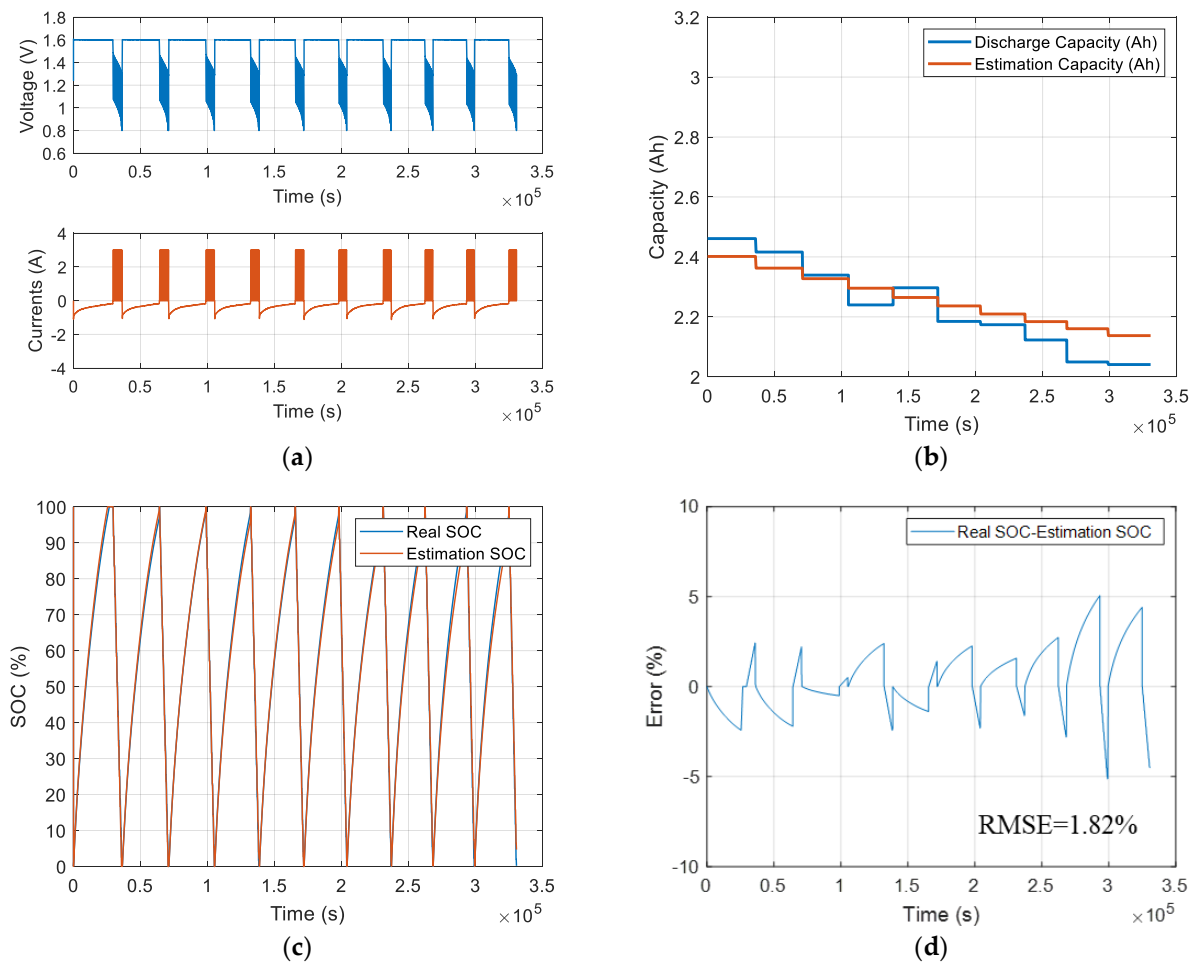


Figure 12. Capacity and SOC estimation results of the VRFB cell during 76–85 cycles (cumulated capacity of 188–208 Ah). (a) Voltage and current waveform, (b) capacity estimation result, (c) SOC estimation result, and (d) SOC error.

5. Conclusions

In this study, we proposed a method to set the initial capacity at the time of rebalancing by using the amount of electrolyte measured using image processing and to estimate the capacity as a variable of the accumulated charge amount of the measured current. In addition, an ampere-hour counting method that can estimate the SOC using the proposed capacity estimation method was presented. The performance of the proposed algorithm was verified from the results of 99 charge/discharge cycling experiments of a 10 W-class VRFB cell. The capacity estimation error performance was 4.94%, and the SOC showed a maximum RMSE error of 3.97%.

In this study, the capacity and SOC estimation that considers rebalancing, which was not covered in previous studies, was conducted. In addition, the proposed method was found to be superior to those presented in previous studies. In particular, it was easy to implement, as it involved applying only the amount of electrolyte, current, and voltage of the battery to the estimation algorithm.

However, the proposed estimation method has a limitation in that it has been verified only for a single cell. Since the VRFB stack system in which many cells are connected in series has a problem of parasitic loss of power and energy due to shunt current, it is necessary to analyze the effect of shunt current on the proposed method. Therefore, we plan to conduct additional algorithm research with a VRFB stack in which four cells are connected in series. In addition to verifying the performance of the proposed method in the VRFB stack system, the results of designing a deep learning model using measured

experimental data such as voltage, current, and amount of electrolyte will be presented in the future.

Author Contributions: Conceptualization, S.L.; Methodology, H.J. and S.L.; Software, H.J.; Validation, H.J.; Formal Analysis, H.J. and S.L.; Investigation, H.J.; Resources, S.L.; Data Curation, H.J.; Writing—Original Draft Preparation, H.J.; Writing—Review & Editing, S.L.; Visualization, H.J.; Supervision, S.L.; Project Administration, S.L.; Funding Acquisition, S.L. All authors have read and agreed to the published version of the manuscript.

Funding: This study was supported by the Korea Institute for Advancement of Technology (KIAT) grant funded by the Korean Government (MOTIE) (P0002092, The Competency Development Program for Industry Specialist), and the National Research Foundation of Korea (NRF) grant funded by the Korean Government (MSIT) (No. NRF-2021R1F1A1063150).

Conflicts of Interest: The authors declare no conflict of interest.

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