이완 시간 분포의 파라미터를 이용한 리튬 이온 배터리의 SOH 추정

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SOH Estimation of the Lithium-ion Batteries using the Parameters of Distribution of Relaxation Times

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ABSTRACT

Lithium-ion batteries are increasingly deployed across a range of applications, due to their favorable characteristics. As the demand for these batteries increases, addressing future challenges related to secondary life of batteries becomes increasingly important. Electrochemical Impedance Spectroscopy (EIS) is a widely used technique for evaluating battery life span, valued for its simplicity and cost-effectiveness. Nevertheless, EIS frequently comes across limitations in distinctly isolating and analyzing internal processes. Recently, Distribution of Relaxation Times (DRT) has emerged as a valuable analytical method for deconvoluting overlapping electrochemical processes, such as ohmic resistance, solid electrolyte interface (SEI) resistance, charge transfer, and diffusion, from EIS data. In this paper, we propose the use of DRT peak parameters of the most prominent charge transfer process across the various State of Charge (SOCs) as inputs to an AI-based model for accurate State of Health (SOH) estimation. Experimental data were gathered from aged lithium-ion batteries, and the LSTM-based model was developed and evaluated using these features. Accuracy of the results demonstrates the novelty of the approach, in addressing the need of reliable battery state estimation methods which is critical of lithium-ion batteries across various applications.

1. Introduction

Lithium-ion batteries (LIBs) are fundamental to a wide range of modern technologies, powering devices from smartphones to electric vehicles due to their high energy density, long cycle life, and operational efficiency. As these batteries approach the end of their first-life usage, there is increasing interest in repurposing them for second-life applications, such as energy storage systems. However, one of the key challenges in extending the useful life of LIBs is the accurate estimation of their state of health (SOH), which is crucial for determining their viability in second-life applications. Developing reliable methods for battery state estimation is therefore essential for optimizing the performance and safety of these batteries [1].

Electrochemical Impedance Spectroscopy (EIS) has long been utilized as a critical technique for evaluating the health of LIBs. EIS provides insight into the electrochemical properties of the battery by measuring its impedance response over a range of frequencies, revealing information on internal resistive and capacitive elements. This technique has proven useful in assessing battery degradation and aging, offering a macroscopic view of the processes occurring within the cell. However, EIS alone has limitations in capturing the detailed chemical processes at play, leaving gaps in understanding the internal mechanisms responsible for battery aging and degradation.

Distribution of Relaxation Times (DRT) has gained attention as an advanced method for refining impedance analysis to address these limitations. DRT deconvolutes impedance spectra, offering enhanced resolution of individual electrochemical processes and their associated timescales. This enables a more nuanced understanding of the internal dynamics governing battery performance. By combining EIS with DRT, it becomes possible to extract more detailed information regarding the internal chemical behavior of the battery, providing a comprehensive framework for evaluating battery health. This study applies both techniques to enhance the accuracy of SOH estimation in lithium-ion batteries, with a focus on second-life applications [1].

In this paper, the DRT method is utilized to analyze EIS data with the aim of extracting key parameters that represent critical internal processes within the lithium-ion battery. Specifically, the charge transfer peak, identified as the most prominent feature within the DRT spectrum, is employed to train a Long Short-Term Memory (LSTM) model for the estimation of the SOH of the Galaxy S9+ battery cell. By leveraging the model-free nature of DRT, this approach allows for a more detailed and process-specific understanding of battery aging. The integration of DRT-derived features into the AI-based model offers an advanced framework for improving the accuracy of SOH estimation, thereby contributing to the development of data-driven methods for monitoring the long-term performance of lithium-ion batteries.

2. Methodology

2.1 Battery Aging Data

In this section, the aging test of the Galaxy S9+ smartphone lithium-ion battery is described [2]. The specifications of the battery used in the test are presented in Table 1. The aging of the battery cell was carried out through charge and discharge cycles, with capacity and impedance spectra measurements taken every 20 cycles. The aging test was conducted with lithium battery of the same specifications over a duration of 1000 cycles. The cycle aging and EIS tests for the lithium battery were conducted using the HYSCLAB chamber, maintaining a constant temperature of 25°C. Battery charge and discharge, as well as EIS tests, were conducted using WonATech's WBCS3000 M2 and ZIVE MP2A equipment, respectively.

During the charging phase of each cycle, a constant current of 2A was applied until the battery reached its cut-off voltage of 4.4V, after which it transitioned to a constant voltage phase, continuing until the charging current decreased to 0.02C. For discharging, an accelerated test was conducted with a discharge current of 1.35C, with the discharge ending once the voltage dropped to 2.8V. A rest period of one hour followed each discharge before beginning the next cycle. After every 20 charge-discharge cycles, Electrochemical Impedance Spectroscopy (EIS) measurements were initiated. These measurements were conducted at 20% intervals of state of charge (SoC) from 0% to 100%. Once the EIS measurement at 100% SoC was completed, the battery was discharged at a rate of 0.2C until reaching the cutoff voltage of 2.8V, and the cycle process was repeated. The EIS tests used a 100mV perturbation and spanned a frequency range of 0.1 Hz to 4 kHz. While increasing the frequency of EIS measurements could provide more detailed impedance data, it also accelerates battery degradation and significantly extends testing time. Therefore, a frequency of every 20 cycles was selected to maintain the balance between accuracy, testing time, and minimizing additional battery wear.

Table 1. Specification of the Galaxy S9+ Battery used for the Aging Test.

Property	Value
Nominal Capacity	3,500mAh
Nominal Voltage	3.85V
Maximum Voltage	4.4V
Charge Current	2,000mAh

During the battery aging test, impedance data were measured at every 20-cycle interval up to 1000 cycles. The initial capacity and the remaining capacity of the battery relative to its nominal capacity was calculated. After the 1000 cycles battery capacity decreased from 97.71% to 90.76% [2].

3. Analysis of Electrochemical Impedance Spectroscopy and Distribution of Relaxation Time

EIS is widely used technique for characterizing the internal dynamics of the battery. It captures the battery impedance response, revealing insights into various electrochemical processes, these processes can be interpreted by Nyquist plots. In this study, EIS measurements are taken at every 20 cycles and at six different SOCs to track the battery internal electrochemical processes [2]. Fig. 1 represents Nyquist plots of impedance spectra for 0-1000 cycles at 100% SOC for highlighting the changes in the impedance over time. High frequency intercept of the Nyquist plot, representing ohmic resistance, remains stable initially but increases over time. SEI layer's semi-circle also acts similarly, second semicircular arc in the mid-frequency region, which is associated with the charge transfer resistance which dominates in aging process of this battery [3]. Due to the specific measurement range of the frequency, the diffusion process is present in the spectra but not fully represented.

In Fig. 1, the overlapping semi-circles complicate the clear distinction of electrochemical processes, which should ideally be represented separately. The height and width of these semi-circles increase with battery cycling, but their similar reaction time constants lead to superimposition in the impedance spectrum. This overlap obstructs the isolation of individual relaxation times and amplitudes, making it more challenging to interpret the impedance spectrum and understand the mechanisms of battery aging.

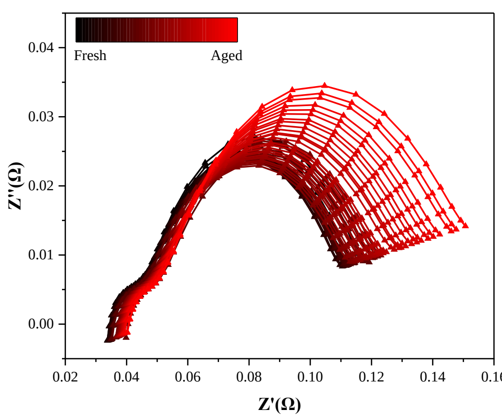

Fig.1. Nyquist plots of S9+ at 0-1000 cycles (SOC 100%)

However, by using DRT analysis, we can more effectively deconvolve and understand the individual electrochemical processes within the battery. DRT enhances this analysis by breaking down the impedance spectrum into distinct peaks, each corresponding to a different internal process. A wider range of frequencies provides more detailed information about what is happening inside the battery. The higher frequencies region reveals faster processes like chemical reactions, while lower frequencies reveal slower processes like ion diffusion within the battery [1].

DRT plots, as illustrated in Fig. 2, show a prominent peak associated with charge transfer resistance, which broadens and shifts to longer relaxation times with cycling. Additionally, smaller peaks related to other processes, such as ohmic resistance, double-layer capacitance, and diffusion also change with the number of cycles, providing deeper insights into the battery's internal condition. The ability of DRT to isolate these individual processes provides a clearer understanding of battery degradation, which is critical for improving SOH estimation models.

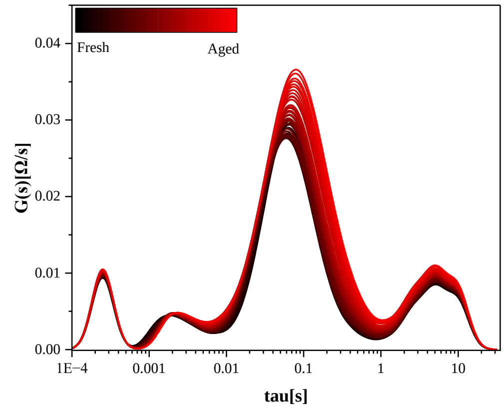

Fig.2. DRT plots of S9+ at 0-1000 cycles (SOC 100%)

4. Results and Analysis

4.1 DRT Parameters Evolution with Battery Aging

Fig. 3 presents the variation of the key parameters including the peak area, full width at half maximum (FWHM), center relaxation time and the peak height of charge transfer peak in the DRT plots over 1000 cycles across the six different SOCs (0%, 20%, 40%, 60%, 80%, and 100%). These parameters are obtained by applying the gaussian fitting on the charge transfer peak for each DRT plot [4]. The DRT parameters exhibit distinct trends that vary with both the number of cycles and the SOC. Notably, the FWHM generally increases with the number of cycles,

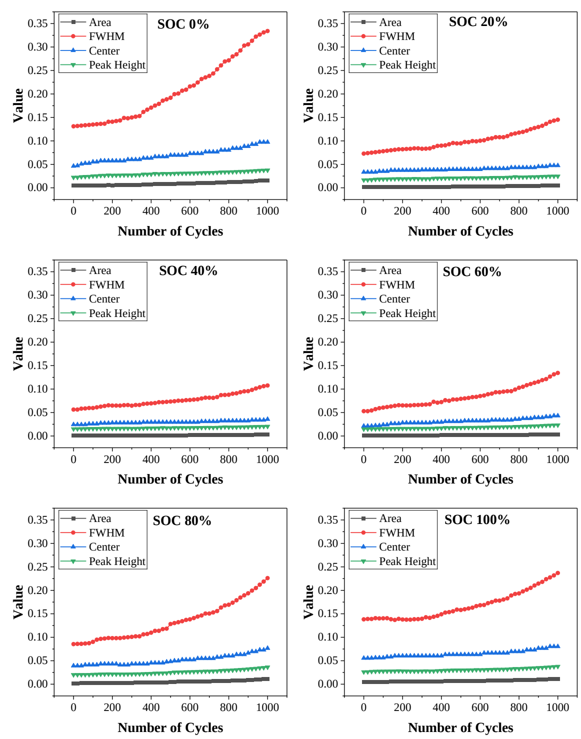

Fig.3. Variation in DRT Parameters of charge transfer peak at different SOCs over

1000 cycles

suggesting a distribution of relaxation times becomes more pronounced due to the increasing complexity of the electrochemical processes involved. Conversely, the peak area, center relaxation time and peak height also tends to broaden, indicating an enhancement in the charge transfer resistance as the battery ages [4].

The incremental trend of the parameters is observed with higher rate at 0%, 80% and 100% SOCs which means at both ends of SOC. This could be due to increased electrochemical activity and increased stress on battery materials. The analysis also reveals that the charge transfer resistance peak shifts towards longer relaxation times with increased cycling. Overall, these DRT parameters provide valuable insights into the aging mechanism of the battery which can help to build the reliable model for battery state estimation.

4.2 LSTM Model Performance

The Long Short-Term Memory (LSTM) model is a type of recurrent neural network that is selected particularly effective in learning from sequential data [5-6], making it ideal for modeling the time-dependent behavior of battery degradation, represented by Fig. 4. The input features for this model included four DRT parameters of charge transfer peak (Peak Area, FWHM, Center Relaxation Time and Peak Height) and SOC as an input and SOH as an output collected over 1000 cycles, which is then divided into training (80%) and testing (20%) sets, ensuring the balanced distribution of varying battery states.

These features are processed through two LSTM layers with 210 and 200 memory units, respectively, using the tanh activation function. A single output layer with a linear activation function predicted the SOH. To prevent overfitting, a 40% dropout rate was applied, and the Adam optimizer, with a learning rate of 0.001, minimized the loss. The model was trained for 175 epochs, with a batch size of 16, balancing training time and accuracy. This setup was chosen after tuning hyperparameters to ensure optimal prediction performance.

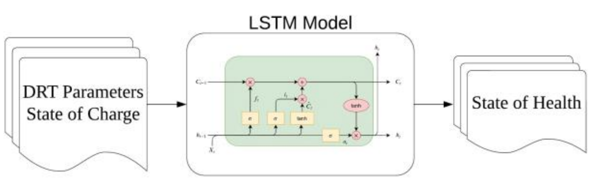

Fig.4. Architecture of LSTM for SOH estimation

The LSTM model was applied to predict the state of health (SOH) of the Galaxy S9+ battery across 1000 cycles. Fig. 5 presents the comparison between the actual SOH and the predicted SOH. As shown in the figure, the model effectively tracks the actual trend of SOH degradation over time. The predicted SOH values closely follow the actual SOH, with the majority of predictions falling within a reasonable margin of error. This demonstrates the model's capability to capture the battery's aging process.

However, minor variations are observed, particularly in the early stages of the battery life cycle. The model tends to overestimate the SOH, which might be due to the insufficient historical data at the beginning of the cycle life, making it hard for the model to fully capture it. Conversely, during the later stages model show minor underestimations might be due to non-linearities in the battery aging process as it approaches the higher cycles.

The Mean Absolute Error (MAE) and Root Mean Squared Error (RMSE) below in equations may also be used to evaluate the prediction performance of the trained model [5].

$$MAE = \frac{1}{N}\sum_{I}^{N}|e_i| \tag{1}$$

$$RMSE = \sqrt{\frac{1}{N}\sum_{i}^{N}(e_i)^2} \tag{2}$$

The MAE defines how near estimates are to the corresponding results. The RMSE, which indicates the variation in errors, is more sensitive to large errors than the MAE. Despite these variations, model's performance can be evaluated by MAE and RMSE calculated as 0.38% and 0.541%, respectively. These performance metrices highlight the effectiveness of DRT parameters in handling the long-term predictions of SOH of the lithium-ion batteries. Future improvements could involve including additional parameters from electrochemical processes such as ohmic resistance, SEI dynamics, and diffusion, together with alternative models like GRU and XGBoost to enhance the accuracy and robustness of analysis.

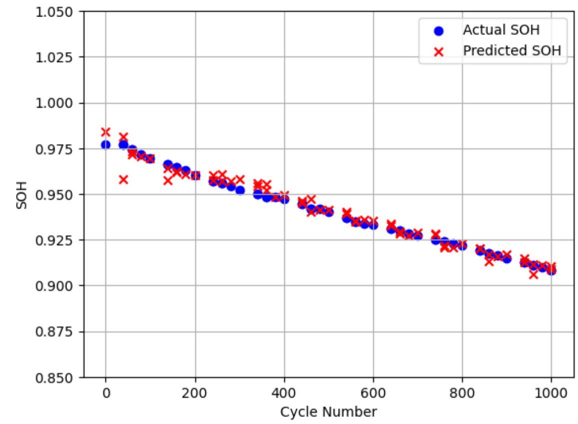

Fig.5. Actual SOH and Predicted SOH by the Model

5. Conclusion

This study highlights the effectiveness of integrating Distribution of Relaxation Times (DRT) with Electrochemical Impedance Spectroscopy (EIS) for enhanced battery state estimation. By analyzing DRT peak parameters over the cycle life of aged lithium-ion batteries at varying SOC levels, we successfully developed an LSTM-based model to estimate State of Health (SOH) with high accuracy. The model yielded significant predictive performance, with Mean Absolute Error (MAE) and Root Mean Square Error (RMSE) of 0.38% and 0.541% respectively, indicating its potential to substantially enhance the reliability of battery health assessments. This approach presents a sustainable solution for managing the lifecycle of lithium-ion batteries in diverse applications. Future work could refine these results by incorporating additional electrochemical parameters and exploring alternative models such as GRU and XGBoost to enhance predictive capabilities.

References

[1] Sohaib, M., & Choi, W. (2024). Investigation of the Aging Phenomena in Lithium-Ion Batteries Using Distribution of Relaxation Time Analysis. 전력전자학회 학술대회 논문집, 187-189.

[2] Kim, M., Akram, A., & Choi, W. (2024). A neuro fuzzy system to evaluate the remaining useful life of the lithium-ion battery using the impedance spectrum in the overall range of SOCs. Journal of Power Electronics.

[3] Wu, J., Meng, J., Lin, M., Wang, W., Wu, J., & Stroe, D. I. (2024). Lithium-ion battery state of health estimation using a hybrid model with electrochemical impedance spectroscopy. Reliability Engineering & System Safety, 252, 110450.

[4] He, R., He, Y., Xie, W., Guo, B., & Yang, S. (2023). Comparative analysis for commercial li-ion batteries degradation using the distribution of relaxation time method based on electrochemical impedance spectroscopy. Energy, 263, 125972.

[5] Zhang, Y., Xiong, R., He, H., & Liu, Z. (2017, July). A LSTM-RNN method for the lithuim-ion battery remaining useful life prediction. In 2017 prognostics and system health management conference (phm-harbin) (pp. 1-4). IEEE.

[6] Park, K., Choi, Y., Choi, W. J., Ryu, H. Y., & Kim, H. (2020). LSTM-based battery remaining useful life prediction with multi-channel charging profiles. Ieee Access, 8, 20786-20798.