Review Article

Prediction of the Remaining Useful Life of Supercapacitors

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As a new type of energy-storage device, supercapacitors are widely used in various energy storage fields because of their advantages such as fast charging and discharging, high power density, wide operating temperature range, and long cycle life. However, the degradation and failure of supercapacitors in large-scale applications will adversely affect the operation of the whole system. To maximize the efficiency of supercapacitors without damaging the equipment and to ensure timely replacement before reaching the end of their useful life, it is critical to accurately predict the remaining useful life of supercapacitors. This paper presents a comprehensive review of model-based and data-driven approaches to predict the remaining useful life of supercapacitors, introduces the characteristics of the various methods, and foresees future trends, with the expectation of providing a reference for further research in this field.

1. Introduction

With the rapid development of the global economy, the living standard of human beings has been improving, and the consumption of resources has been further increased. For example, the massive burning of coal will also produce numerous solid wastes [1], the generation and discharge of industrial wastewater and cause serious water pollution [2], and the problems of energy crisis and environmental pollution are becoming more and more serious [3, 4]. In this context, the concept of green environmental protection, energy-saving, and low carbon has gradually become popular. Tens of thousands of plastics produced by humans have caused immeasurable harm to the environment, while energy consumption is becoming more and more serious, and a study has proposed a triboelectric nanogenerator (TENG) based entirely on waste plastic bags [5]. In addition, a study has formed high-efficiency photovoltaic cells based on PdSe2 [6]. Some studies combine energy storage elements with material directions

in an attempt to develop efficient and low-cost energy-consuming devices [7–12].

In addition, new high-performance, low carbon, and green energy storage power systems are a key approach to improve this situation [13]. Similarly, energy-storage power systems have a wide range of applications in biomedical and medical devices and motion sensors [14, 15]. Energy storage systems also have unique advantages in industrial, military, transportation, and power fields [16–18]. Lithium batteries [19, 20] and capacitors play an important role in new energy-storage power systems, where capacitors are mainly classified as dielectric energy storage capacitors and widely studied supercapacitors [21]. To solve the problem that electrical conductivity and cycle life cannot meet the requirements of applications, various types of metal-organic frameworks (MOFs) materials [22] have been tried to be used in supercapacitors and metal cells to explore their electrochemical energy storage mechanisms, stability of electrode materials, charge transfer pathways, mass transfer, and electrochemical reactions. Dielectric energy storage

capacitors are mainly lead-free energy storage ceramics [23], which have a certain potential for application due to their higher effective energy storage density compared to conventional linear dielectrics. Lead-free energy storage ceramics are both environmentally friendly and less dense than lead-based materials because they are lead-free materials, which facilitates lightweight in applications [24]. Supercapacitors (SCs), as a new type of energy storage device, have the advantages of fast charging and discharging, high power density, wide operating temperature range, long cycle life, and high reliability [25].

At this stage, ultracapacitors are widely used in standby or emergency power, alone or mixed with batteries as peak power, as well as in control systems, communication fields, hybrid electric vehicles, and smart grids. Meanwhile, a study has proposed an optimized power distribution method for hybrid energy-storage systems for electric vehicles (EVs) [26]. The hybrid energy storage system (HESS) uses two isolated soft-switched symmetrical half-bridge bidirectional converters connected to a battery and a supercapacitor (SC) as a protection structure for the composite structure. It helps to improve energy utilization and reduce the battery aging effect.

However, since an energy storage system consisting of ultracapacitors is a complex nonlinear system, degradation and failure of individual ultracapacitors in an application will adversely affect the operation of the entire system. Therefore, accurate prediction of the remaining useful life (RUL) of ultracapacitors is crucial to improve the reliability of energy storage systems and effectively reduce the occurrence of failures.

The mainstream methods are mainly model-based and data-driven approaches. The model-based approaches consider the battery loading conditions, material properties, and degradation mechanisms, mainly including equivalent circuit models, electrochemical models, and empirical degradation models [27, 28]. However, the principles of model-based approaches are complex and vulnerable to external factors, making it difficult to build stable models. Notably, data-driven methods do not require complex modeling and internal mechanism analysis, are highly flexible and scalable, and have been widely used in recent years [29].

This paper reviews model-based and data-driven approaches to predict the remaining useful life of supercapacitors and analyzes the characteristics and problems of each approach as well as future research trends.

2. Operating Principle and Aging Mechanism

2.1. Operating Principle.
According to the energy-storage mechanism, supercapacitors are divided into double-layer capacitors and Faraday capacitors. The double-layer capacitor uses carbon material as an electrode to store electrical energy by electrostatic effect, and the physical reaction occurs and the process is reversible. The energy storage is achieved by the potential difference between the two solid electrodes due to the adsorption of positive and negative ions on the surface between the solid electrode and the electrolyte, respectively. During charging, the anions and cations in the electrolyte gather on the surfaces of the two solid electrodes under the effect of charge gravity on the solid electrodes; during discharging, the anions and cations leave the surfaces of the solid electrodes and return to the electrolyte body, while the stored charge is released through an external circuit. The changes in the supercapacitor before and after charging are shown in Figure 1.

When a Faraday capacitor is charged, the ions in the electrolyte diffuse into the solution under the action of the applied electric field to the electrode/solution interface and then enter the electrode surface-active oxide through the electrochemical reaction at the interface; if the electrode material is an oxide with a large specific surface area, a considerable number of such electrochemical reactions take place and a large amount of charge is stored in the electrode. When discharging, these ions that enter the oxide are returned to the electrolyte, while the stored charge is released through the external circuit. Thus, it is able to supply power to the load.

Therefore, electrode materials and electrolytes can have a huge impact on the electrochemistry of supercapacitors and are also important factors in the aging of supercapacitors.

2.2. Aging Mechanism.
The study of the aging mechanism of supercapacitors is important for the accurate prediction of the remaining service life of supercapacitors.

Supercapacitors consist of electrodes, electrolyte, diaphragm, and fluid collector, so the aging characteristics of supercapacitors usually refer to case damage, electrolyte decomposition, and electrode degradation [30]. In practical applications, their service life is also influenced by external stresses. For example, voltage, current, and temperature are the main factors affecting the aging of supercapacitors. Water decomposition of supercapacitors generates a certain amount of air pressure inside the case, which may damage the case with long-term use or in extreme cases. In the temperature range, high temperatures will promote the chemical activity of activated carbon electrodes and accelerate their aging. The capacitance value of supercapacitors is directly proportional to the specific surface area of the electrode material, so changes in the electrode material often cause a decrease in the specific surface area and thus the capacity of supercapacitors [31]. The by-products of aging of supercapacitors and polymers will result in a smaller pore structure on the electrode surface, and impurities from electrolyte decomposition reduce the ability of ions to reach the cavities, leading to an increase in the equivalent series resistance (ESR). This affects the normal embedding and de-embedding of ions and makes the performance of the supercapacitor degraded.

In addition, during the preparation of electrode materials, a small number of impurities as well as oxygen-containing functional groups, remain on the electrode surface, which can lead to a faster decrease in capacitance value of supercapacitors in the early stages of aging [32].

Under the influence of these aging factors, the RUL of supercapacitors gradually degrades along a certain nonlinear

Before charging

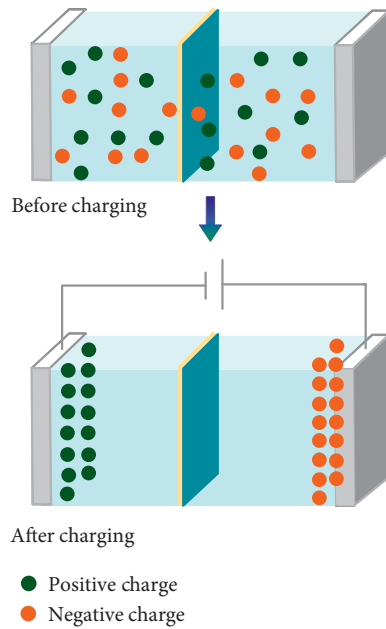After charging

● Positive charge
● Negative charge

FIGURE 1: Principle of operation of supercapacitors.

curve until it reaches the critical allowable use range [33]. Therefore, it is important to accurately predict the RUL of supercapacitors to ensure the safety and reliability of their operation.

3. Model-Based Prediction Methods

The model-based prediction method is an effective way to evaluate the lifetime of supercapacitor devices by building an equivalent circuit model based on the electrical performance or energy storage principle of the supercapacitor. The equivalent circuit model uses a parametric RC (capacitor-resistor) network to model the electrical behavior of the supercapacitor, and usually uses ordinary differential equations with simplicity and ease of implementation in the model formulation.

The study conducted by Liu et al. [34] can fit the parameters of the power function model better based on the decay trajectory of supercapacitor capacitance and extrapolate the failure life model which conforms to the Weibull distribution. The validation results of the life model show that the relative error decreases as the number of cycles increases, and the cycle life of supercapacitors can be better predicted by choosing the appropriate number of cycles. In the literature [35], a prediction model of supercapacitor capacity decay including temperature, current intensity, and cycle number factors was established based on the classical Arrhenius model through the performance decay law exhibited by supercapacitors under different charging and discharging currents and temperature conditions. The validation results show that the fitted results are in good agreement with the actual decay data, and most of the relative errors are within 3%. In the literature [36], an online estimation scheme based on particle filtering (PF) is proposed to estimate the state of health (SOH) of SC by combining the electrical equivalent circuit model (ECM)

and thermal model and to monitor the SOH of SC by estimating the equivalent series resistance (ESR) and device capacity in real-time.

Based on this, a supercapacitor RUL prediction model considering aging conditions such as temperature and voltage was proposed in the literature [37]. Experiments conducted under different aging conditions found that the method can predict the capacitance and resistance as well as the RUL under different initial conditions well in addition to considering voltage and temperature, an improved reliability model was proposed in the literature [38], which simulates the voltage, temperature, and humidity levels to which the supercapacitor is exposed during operation under actual operating conditions, further improving the prediction accuracy of the model.

Model-based prediction methods usually combine different models and filtering methods to achieve data tracking and prediction of the remaining lifetime of supercapacitors. However, model-based prediction methods are very complex and difficult to implement due to the complexity of supercapacitors.

4. Data-Driven Forecasting Methods

Compared with model-based methods, data-based methods do not require complex mathematical models to simulate the internal aging mechanism of supercapacitors. They are methods to predict the trends of device parameters during the aging process mainly based on historical data of supercapacitor aging process and state data, such as artificial neural networks and fuzzy logic.

A simple recurrent neural network (SIM RNN) was proposed in the literature [39] for supercapacitor lifetime prediction, but SIM RNN has the disadvantage of long-term dependent learning. If the information is stored for too long, the gradient will disappear and the SIM RNN cannot continue to learn. Zhao et al. [40] proposed a lifetime prediction method based on long short-term memory neural network (LSTM RNN), which used the Dropout algorithm to prevent overfitting and optimized the neural network using Adam's algorithm. The input of the neural network is the measurement data under different operating conditions that are divided into training and prediction sets, and the root mean square error (RMSE) of the prediction results is about 0.0261. The life prediction of offline data has a mean absolute error (MAE) of about 0.0338, which proves the applicability of the algorithm. The gated recurrent unit (GRU) structure is simpler than the LSTM. The advantage of the GRU is that it is a simpler simple model with fewer parameters, and GRU is more likely to converge. However, the LSTM RNN has better performance when the data set is larger [41]. Therefore, the long-short memory neural network is very suitable for the prediction of the remaining lifetime of supercapacitors.

Zhou et al. [42] proposed an algorithm combining a long short-term memory neural network and a hybrid genetic algorithm, whose structure is shown in Figure 2. The sequential quadratic programming is used as a kind of local
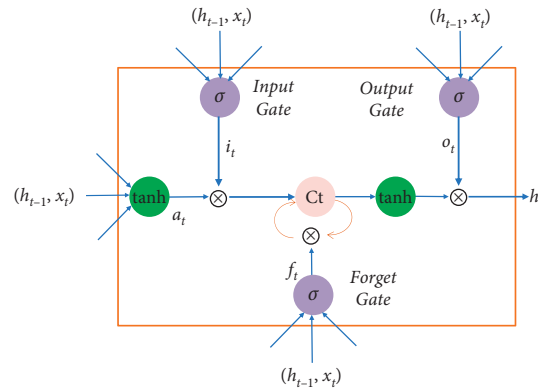
FIGURE 2: A structural diagram of the long short-term memory, a type of neural network. Here, a gate component is embedded to process the memorizing and forgetting and input as well as output inside such active memory [42]. Reproduced with permission. Copyright 2020, applied energy.
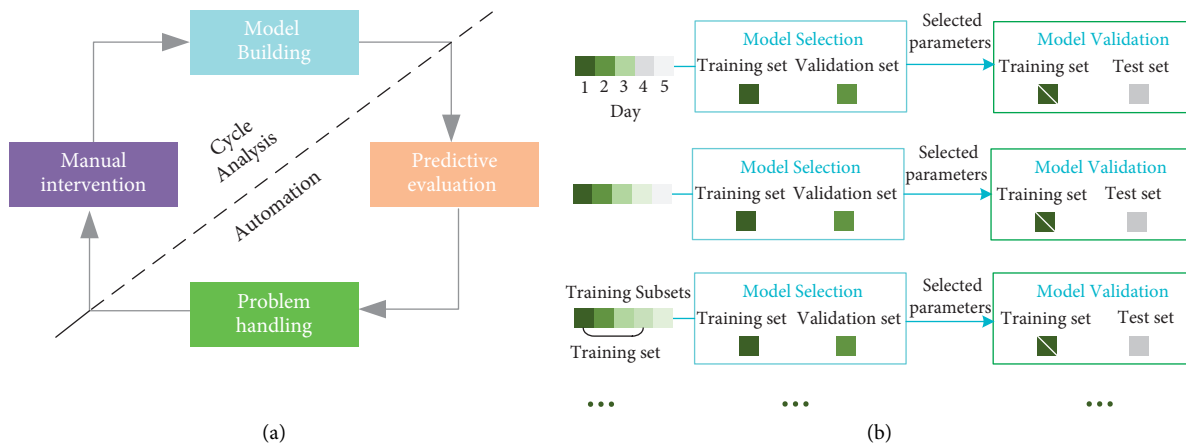

(a)

(b)

FIGURE 3: (a) Flowchart of the prophet model. (b) Schematic diagram of the forward chain method [44]. Reproduced with permission. Copyright 2021, Chinese Journal of electrical engineering.

search operator of the genetic algorithm, which enhances its global search capability and enables it to search the local optimal solution quickly by the exit probability and the number of hidden layer units. This prediction method can estimate the remaining lifetime of supercapacitors in steady-state charging mode well, and also works well for supercapacitors with dynamic operation cycles.

The study by Liu et al. [43] uses a stacked bidirectional long- and short-term memory recurrent neural network model, which adds a reverse recurrent layer with t-time and subsequent time values in the input sequence to the traditional long- and short-term memory recurrent neural network. Among them, the stacked network can ensure sufficient capacity space. When the number of hidden layers is 2, the predicted RMSE and MAE are 0.0275 and 0.0241, respectively, indicating that the network has better performance. A method for predicting the life of supercapacitor modules based on the monitoring data of buses under actual service conditions is proposed in the literature [44]. The qualified memory least squares method is used for parameter identification of service condition data to obtain the resistance and capacitance values with time-series and

seasonal characteristics, and then, the RUL is predicted based on this method using the Prophet algorithm. The forward chain method is also introduced to validate the results, which is better than the cross-validation method in machine learning that ignores the time-series characteristics of the time-series data. The running time of Prophet is only about 20% of the running time of LSTM RNN, and the prediction accuracy is higher and the time required is shorter for data with periodic and seasonal characteristics, provided that the model remains unchanged and the same prediction accuracy is obtained. The structure diagram of Prophet and the flow chart of the forward chain method are shown in Figure 3.

Haris et al. [45] proposed a new deep learning algorithm, deep belief network (DBN) combined with Bayesian Optimization and HyperBand (BOHB), with the structure shown in Figure 4, for predicting the RUL at the early stage of supercapacitor degradation. Compared with previous studies, the development time of the RUL prediction model was reduced by 54%, largely saving the time required to collect and measure supercapacitor cycle data, and the proposed model has good accuracy and robustness.
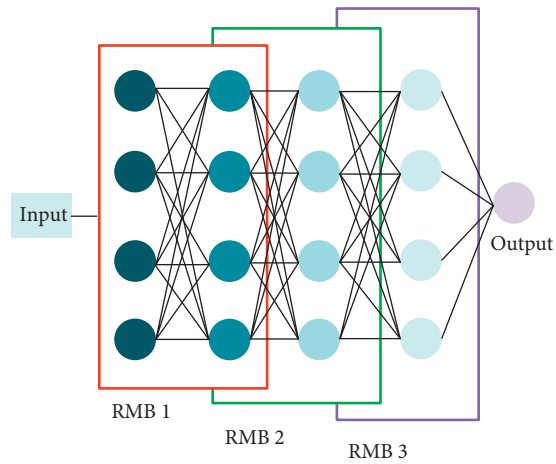
FIGURE 4: Deep belief network structure diagram [45]. Reproduced with permission. Copyright 2021, applied energy.
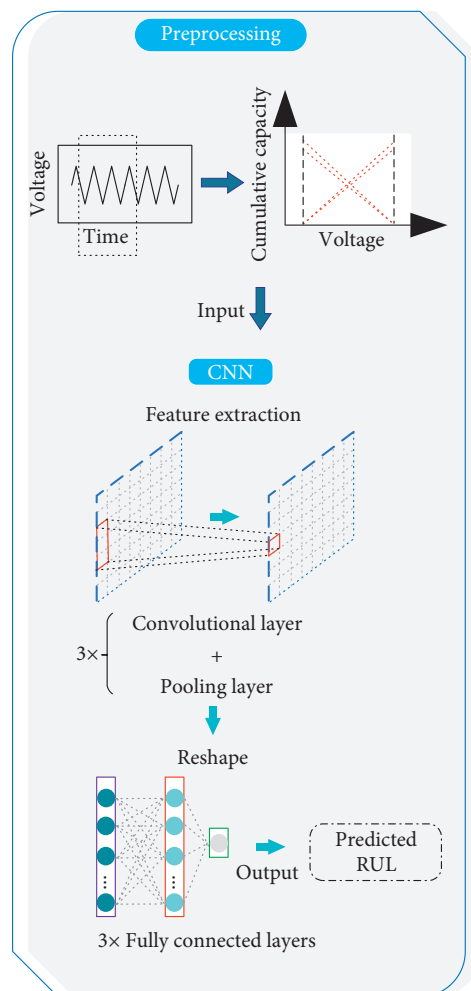

FIGURE 5: Framework of the proposed CNN [47]. Reproduced with permission. Copyright 2022, applied energy.

Ren et al. [46] presented a neural network-based RUL prediction method for supercapacitors that does not strongly depend on the data distribution and is less dependent on the correlation between variables and features, and the proposed model is suitable for data sets with a wide training distribution and has an accurate early diagnosis and prediction capability for the performance of complex supply chain systems. The testing error is less than 10.9%, and this error can be further adjusted by the dataset.

The above methods enable a significant reduction in the length of the input cycle, but they still require a large amount of data for their extraction. Since manually produced features inevitably lose aging information, the accuracy of prediction results is limited. In addition, manual feature extraction is cumbersome because it requires domain-specific knowledge and a long processing time. An end-to-end RUL prediction method based on convolutional neural networks (CNNs) is proposed in the literature [47]. The method has higher accuracy while requiring significantly fewer input data. Also, the method effectively reduces the need for data and improves prediction accuracy, which helps in the diagnosis and prediction of supercapacitors. The framework of the proposed CNN is shown in Figure 5.

Using a data-driven approach that does not require physical modeling of the component and is designed to simulate the relationship between measurement data and component degradation, it can effectively predict energy storage lifetime and discharge behavior without the need for detailed studies of internal chemical changes and side reaction disturbances.

5. Conclusion

Supercapacitors are widely used in many fields because of their advantages such as high power density, fast charging and discharging speed, and wide operating temperature range. Since the supercapacitors storage system is composed of a set of basic units, the inconsistent parameters of each unit, uneven charging voltage, and the difference in internal temperature in large-scale applications will lead to the degradation of its performance and the aging of the device. Therefore, accurate prediction of the remaining service life of supercapacitors can effectively reduce the occurrence of failures and accidents. Currently, there are mainly model-based and data-driven prediction methods. Due to the complexity of supercapacitors, model-based prediction methods are complicated to implement. The data-based approach does not require complex mathematical models to simulate the internal aging mechanism of supercapacitors, and combined with artificial neural networks can have better prediction accuracy and efficiency, which is the focus of future research. However, since the method relies only on relevant historical data to complete the prediction, the quality of the assessment depends heavily on the accuracy of the historical data. Because of the long lifetime of supercapacitors, it takes longer to collect cycle life data, which will increase the possibility of noise pollution and affect the prediction results. Also, the difficulty of obtaining high-precision historical data is a technical problem that needs to be broken in the process of achieving accurate evaluation.

Combining with transfer learning techniques to reduce the reliance on data will be a future research trend.

Conflicts of Interest

The authors declare no competing interests.

Acknowledgments

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