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Article

A closed-loop degradation-aware self-healing battery framework for ultra-long-duration energy storage

Binggui Lu[1,*], Manisha Sagar Pawar[2], Budigi Prabhakar[3], Simranjeet Nanda[4], Tusha[5], Kasturi Pohini[6], Yagna B. Adhyaru[7]

[1] Faculty of Education, Shinawatra University, Pathum Thani 12160, Thailand
[2] Department of Engineering Sciences and Humanities, Vishwakarma Institute of Technology, Pune 411037, India; manisha.pawar@vit.edu
[3] Department of Physics, Noida International University, Greater Noida 203201, India; budigi.prabhakar@niu.edu.in
[4] Center for Research Impact and Outcome, Chitkara University Institute of Engineering and Technology, Chitkara University, Rajpura 140401, India; simranjeet.nanda.orp@chitkara.edu.in
[5] Quantum University Research Center, Quantum University, Roorkee 247667, India; tusha@quantumeducation.in
[6] Centre for Multidisciplinary Research, Anurag University, Hyderabad 500088, India; KasturiRohini2024@protonmail.com
[7] Faculty of Engineering, Gokul Global University, Siddhpur 384151, India; ybadhyaru.ce.hcet@gokuluniversity.ac.in
*** Corresponding author:** Binggui Lu, lubinggui1110@163.com

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Abstract: Degradation in lithium-ion batteries employed for ultra-long duration energy storage (LDES) greatly restricts the performance, dependability, and remaining useful life (RUL) predictability of such devices. Current methods tend to emphasize either material degradation or data-driven prognostic approaches and seldom incorporate the notions of self-healing and intelligent prognosis. A novel paradigm of degradation-aware self-healing electrodes is introduced by combining a composite core-shell electrode, a reversible self-healing matrix, embedded multi-modal sensing, and a physics-informed hybrid predictive model. It enables monitoring of mechanical stress, impedance increase, temperature, and capacity degradation, leading to a closed loop of degradation detection, self-healing, and adaptive RUL prediction. The approach was analyzed via simulations under representative long-duration battery operation and contrasted with the traditional lithium-ion electrode. The suggested framework demonstrated substantial improvements in electrochemical durability and prediction performance. The cycle life of the battery has been extended to 2,100 charge-discharge cycles from 1,200 charge-discharge cycles, and capacity retention after 1,000 cycles rose from 68% to 86%. The impedance increase was significantly decreased by approximately 40%, and the normalized stress increase decreased from 1.00 to 0.62. The capacity fade rate dropped from 1.8% to 0.9% per 100 cycles. Also, the hybrid prediction framework lowered the error rate of RUL prediction from 18.4% to 6.7%, outperforming traditional predictive frameworks. The statistical analysis conducted across 10 simulation runs proved the significance of the observed changes (two-tailed paired t-test, $p < 0.001$). The suggested degradation-aware self-healing framework proves the potential of combining autonomous recovery and hybrid prediction in order to improve both the battery's durability and its prognostic performance at once. This closed-loop system is able to prevent the negative effects caused by degradation on battery performance and improve reliability.

Keywords: degradation-aware electrodes; self-healing batteries; long-duration energy storage; remaining useful life prediction; hybrid predictive modeling; battery degradation; intelligent energy storage

1. Introduction

1.1. The criticality of advanced energy storage solutions

The global transition away from carbon-intensive energy systems toward a modern and sustainable low-carbon infrastructure has driven increased demand for high-tech energy storage technologies [1]. Due to the rapidly expanding integration of volatile and intermittent renewable energy sources like wind and solar on the grid, the need for long-duration energy storage (LDES) for ultra-long-duration applications to guarantee a reliable and continuous energy supply has become immediate [2]. Among energy storage options, batteries have been identified as the optimal solution owing to the inherent modularity, scalability, and speed required to maintain equilibrium between energy supply and demand within today's energy grids [3].

1.2. Challenges of degradation in long-duration systems

Despite their potential, the long-term usefulness of existing battery systems remains limited, despite their advantages, due to complicated electrochemical degradations that gradually result in capacity fading and a rise in internal resistance [4]. Several types of such structural degradations are known to limit device longevity, such as growth and aggregation of solid electrolyte interphase (SEI) layers, consumption of active material, deposition of Li plating, and fractures of electrodes, all exacerbated through operation over many charge/discharge cycles over extended time scales, causing continuous mechanical and chemical loadings [5]. To tackle the problem in a practical manner, one typically employs material durability approaches, predictive approaches, or their combined versions: material durability approaches, which can provide structural stability for device components; however, they usually lack in situ monitoring for actual system states. Purely predictive models, on the other hand, typically make assumptions that electrode material properties are static during their service time [6].

1.3. Degradation-aware self-healing electrodes

To counter such fundamental limitations, the current work develops a smart electrode architecture that transcends passive protection against degradation in favor of an active, adaptive solution. Introducing self-healing constituents, specifically dynamic polymers, which can readily remake broken bonds in the electrode network structure, enables the self-healing capability that proactively counteracts degradation caused by microcracks and structural fatigue [7]. When this self-healing functionality is synergized with a degradation-aware sensing layer, the battery acquires the capability of sensing degradation markers like strain accumulation, impedance increase, and capacity loss simultaneously [8]. An intelligent predictive framework integrating both physics-based modeling and deep learning-based predictions, long short-term memory (LSTM)/Gated recurrent unit (GRU) networks, allows this hybrid scheme to operate within a closed-loop configuration, not only repairing structural damage but also iteratively improving its future projections of RUL for improved condition monitoring of future grid-scale batteries [9].

1.4. Research objectives

The key focus of the current research work is on developing an intelligent electrochemical degradation-aware electrode structure that can help mitigate electrochemical degradation and also improve the prediction of battery lifetime in the application of ultra-long duration energy storage. In particular, this research work proposes the combination of self-healing materials, degradation detection, physics-based degradation modeling, and LSTM/GRU-based lifetime prediction analytics in a single closed-loop system.

1.5. Key contributions

The proposed work makes the following contributions:

- Formulates a self-healing electrode design for long-term energy storage that is aware of its degradation.
- Incorporates embedded sensing of multiple modes to monitor degradation in real time.
- Proposes a hybrid physics-informed LSTM/GRU framework to predict RUL adaptively.
- Constructs a feedback-based closed-loop approach that associates the process of healing with prediction.

The rest of the paper is structured as follows. Related works on self-healing electrodes, degradation models, and prediction methods are presented in Section 2. The proposed model of the degradation-aware self-healing electrode is introduced in Section 3, with an introduction to the methodologies employed. Simulation results and a comprehensive performance evaluation are provided in Section 4. The broader implications of these findings, along with a detailed analysis, are discussed in Section 5. Finally, the paper is concluded in Section 6.

2. Literature review

2.1. Review of current research on self-healing materials in energy storage applications

The quest for ultra-long-duration energy storage (LDES) has spurred extensive interest in merging material science advances with sophisticated prognostic algorithms. Contemporary reviews point out that modern battery reliability is compromised by intrinsic electrochemical failure modes like solid electrolyte interphase (SEI) formation, lithium plating, and mechanical failure mechanisms [10]. In response to this issue, current research directions emphasize the incorporation of self-healing designs as an approach to bolster structural robustness [11]. Self-healing polymer binders and nanostructure-based electrodes have recently demonstrated effective mitigation of mechanical fracture via auto-repair of microcracks created during electrochemical cycling [12]. Lifecycle awareness of battery architectures has similarly noted the importance of incorporating recyclability with self-healing for achieving sustainable, high-density renewable energy storage systems [13].

The recent advances in the field of energy storage are mostly concentrated on combining the self-healing components into these systems to compensate for the demerits associated with the conventional batteries. Recent research reported that the use of the self-healing properties of dynamic polymer binders and the use of nanomaterials on electrodes have shown promising results that enable them to heal on their own from minor crack formation and structural damage generated while performing charge-discharge operations [14]. This is indeed necessary for retaining the mechanical stability and expanding the lifespan of Li-based batteries and supercapacitors [15]. In addition to this, some reports have also indicated that the synergy of such material developments with the multisensory data-based AI is needed for building smart energy storage systems [16].

2.2. Discussion of the mechanisms of degradation in energy storage systems

Degradation in energy storage devices is the result of an intricate combination of electrochemical interactions. Among the most detrimental, and often co-dependent mechanisms, are SEI layer growth, lithium plating, and the consumption of active materials, all of which degrade the capacity and increase the internal resistance of the cell over time [17]. These processes are amplified by mechanical interactions in electrode components, which may be exacerbated by structural integrity issues (such as cracking of active particles or delamination) caused by the volumetric changes inherent in battery cycling [18]. Without any form of mitigation or control of these degradation pathways, the capacity degradation curve is nonlinear, and premature failure will result in an overestimation of a battery's life [19].

In parallel with these material innovations, the field of battery prognostics is also seeing significant developments, with ML becoming the primary technology used for predicting remaining useful life (RUL) and state-of-health (SOH). Increasingly sophisticated modeling methods, specifically with recurrent neural networks such as long short-term memory (LSTM) and gated recurrent unit (GRU) networks, have been reported to capture the complex and nonlinear nature of degradation, along with the dynamic properties of battery usage [20]. More recent literature has also focused on the combination of physics-based models that reflect underlying mechanistic aging phenomena like SEI growth with ML learning methods, reporting higher prediction accuracies than standalone methods by including dynamic phenomena such as capacity regeneration and polarization recovery [21]. Moreover, uncertainty estimation using methods like quantile Transformers has been proposed for making better predictions of battery degradation in real and complicated conditions [22].

2.3. Overview of existing strategies for mitigating degradation in electrodes

To counteract these degradation mechanisms, existing methods typically work in two largely isolated approaches: material-level strategies and predictive maintenance modeling. The first enhances battery electrode longevity by employing some mechanism of structural recuperation and/or through the introduction of nanomaterials to defer performance decay [23]. The latter employs machine learning models, including LSTM and GRU neural networks, for predicting the state-of-health (SOH) and remaining useful life (RUL) [24]. Both have proven effective but were applied separately, and predictive models generally assumed a fixed state of the electrodes, not accommodating adaptive, self-healing material attributes. This work offers a unifying closed-loop methodology by blending the fields of real-time sensing, autonomous self-healing, and hybrid predictive modeling in order to proactively control battery degradation [25].

Despite these strides, a distinct gap remains in the literature: the absence of a comprehensive closed-loop framework integrating real-time sensing of degradation coupled with active self-healing and prognostic predictions. Previous works have individually identified and demonstrated several key techniques—namely mechanical stress assessment, autonomous diagnostics, and prognosis—although thus far operated as independent non-interacting solutions. Increasingly, evidence indicates that a new era of sapiential or smart battery technology needs to transcend passive acceptance of degradation in favor of active, health-aware adaptive capabilities. The fusion of multi-modal sensing, which allows for detection of both mechanical stress accumulation and impedance growth, is gradually illustrating that battery cells can be designed to possess a sense of self, and then to activate healing exactly at the point structural limits are exceeded.

This research directly attempts to fill the aforementioned knowledge gap by putting forward a comprehensive and degradation-aware self-healing electrode concept. Adopting the current state-of-the-art approaches of hybrid modeling and material design, this work presents a systematic procedure by taking system performance feedback to manipulate prediction in order to reduce error drift and prolong the lifetime of the device. By combining material science using a dynamic polymer network with the methodology of an advanced predictive maintenance approach for LDES, a practical solution for deploying the device in the field of large-scale renewable applications may be realized. By integrating these two approaches, this proposal may contribute to a resilient operational state that helps to ensure a stable supply of renewable power.

2.4. Recent Advances in self-healing polymer binders and electrolytes

Recently, considerable progress has been made towards the design of self-healing materials for lithium-ion batteries, where the focus of study has shifted from electrode structures to self-healing polymer binders and electrolytes. Reversible covalent or supramolecular dynamic binders have been shown to heal cracks created during repeated cycling, thereby retaining the integrity of the electrodes and contributing to improved cycling performance [26]. Such binders are capable of accommodating electrode volume change and preventing electrode particle isolation and, thus, are mechanically robust.

Another class of self-healing materials used in the field of lithium-ion batteries includes self-healing polymer electrolytes, which could provide better safety and electrochemical performance of batteries. Self-healing polymer electrolytes allow self-repair of damaged parts without compromising ionic conductivity of the system [27], hence preventing electrolyte failure. In comparison to conventional electrolytes, self-healing electrolytes possess greater crack resistance, sealing properties, and interface stability.

While the aforementioned literature deals mainly with individual components at a material level, this paper presents a self-healing polymer matrix coupled with degradation sensing and feedback mechanisms as part of a single system. Thus, it not only allows the restoration of damaged structures but also the management of the battery state of health.

2.5. Research gap and novelty

While recent research focuses on the self-healing of the electrodes, modeling of the degradation process, and the application of machine learning to predict the remaining useful life separately, there is still an absence of an integrated degradation-aware energy storage framework. In fact, while the material-based methods focus only on the improvement of structural stability, they fail to continuously monitor the degradation process and update the models upon restoration of the structure.

In order to fill these gaps, the proposed degradation-aware energy storage framework incorporates a degradation-aware design of the electrode, the layer of sensors, the reversible polymer network, the modeling of degradation informed by physical principles, and the prediction model based on LSTMs/GRUs. The difference between this approach and those of the existing studies lies in the continuous updating of the degradation predictions based on feedback from the restored electrode condition.

The major novelties of this work are summarized as follows:

- Invention of a degradation-aware self-healing electrode design that incorporates sensing, material recycling, and prediction capabilities.
- Self-healing loop interaction with battery state-of-health prediction.
- Physics-informed and deep learning-based framework for dynamic RUL estimation.
- Degradation monitoring through built-in multi-modal sensing.
- Integrated material-computation framework for ultra-long duration energy storage use cases.

3. Methodology

3.1. Data collection and simulation setup

The proposed approach was validated through a simulation-based procedure for evaluating the behavior of the lithium-ion battery in ultra-long-duration energy storage scenarios. Degradation datasets, including capacity fading, impedance increase, temperature, and mechanical stress, were produced via a hybrid electrochemical battery degradation model, which was calibrated according to the degradation properties of lithium-ion batteries found in contemporary literature. The simulation involved a constant-current charge-discharge cycle to mimic long-term energy storage in the grid. The resulting degradation data were used to train the RUL prediction model.

3.2. System overview

The proposed system features a multi-level electrode structure designed to ensure high operating reliability over time. The electrode structure consists of three main cooperative elements integrated together to achieve energy storage, proactive degradation mitigation, and continual health assessment at the same time: (1) The electroactive substance acting as the material used for energy storage. (2) Self-healing material comprising dynamic polymers that act as the electrode binder, designed to react to mechanical stress-induced damage. (3) A dedicated degradation sensor layer providing high-fidelity information about electrochemical and mechanical health parameters.

Combining the above, a flexible structure was designed that is able to self-adjust its local degradation state to locally occurring phenomena at any cycle. Thus, the architecture has self-repair features, allowing it to avoid failure modes common in traditional long-lifetime energy storage systems.

3.3. Data analysis and predictive modeling

The degradation factors were normalized and then passed to the hybrid model for prediction purposes. A physics-based degradation model was used to calculate the loss of capacity due to growth of SEI and mechanical degradation, while an LSTM/GRU-based neural network was trained on historical battery states to predict non-linear degradation behavior. The prediction model continuously updated the predicted remaining useful life after every healing period through feedback provided by the degradation sensing layer. Evaluation was carried out using Root Mean Square Error (RMSE), RUL prediction error, capacity retention, impedance growth, Coulombic efficiency, and cycle life.

3.4. Self-healing–prediction framework

To maximize the system's longevity, this framework makes use of a closed-loop adaptive feedback process that integrates online sensing and prognostic knowledge. This prognostic model can consistently examine for subtle departures from nominal performance trends in observed degradation. If such patterns go above prescribed safety limits, pointing to the probability of imminent essential failure, the system may proactively activate the self-healing mechanism throughout the electrode matrix; this may result in the physical remodeling of reversible bonds that help close microcracks and resume electrochemical paths. Crucially, this entire approach works as an intelligent loop feedback system; after the healing operation has been carried out, the revised well-being of the electrode is assessed once more via the sensors, and reinserted into the prediction model. The recurrent neural networks, like LSTM or GRU-based algorithms, then use this data to adjust the subsequent forecasts to make sure that their predictions of the RUL stay accurate and dependable even as dealing with the non-stationary character of battery aging.

Figure 1 shows an integrated closed-loop energy storage scheme, incorporating a degradation-sensitive electrode design, live measurement of critical parameters such as stress, capacity loss, impedance, and temperature, and a predictive model based on physics (SEI evolution prediction) and deep learning (LSTM/GRU model). Self-healing is triggered when the system detects degradation, and it is also continuously improving its predictions using a feedback loop.

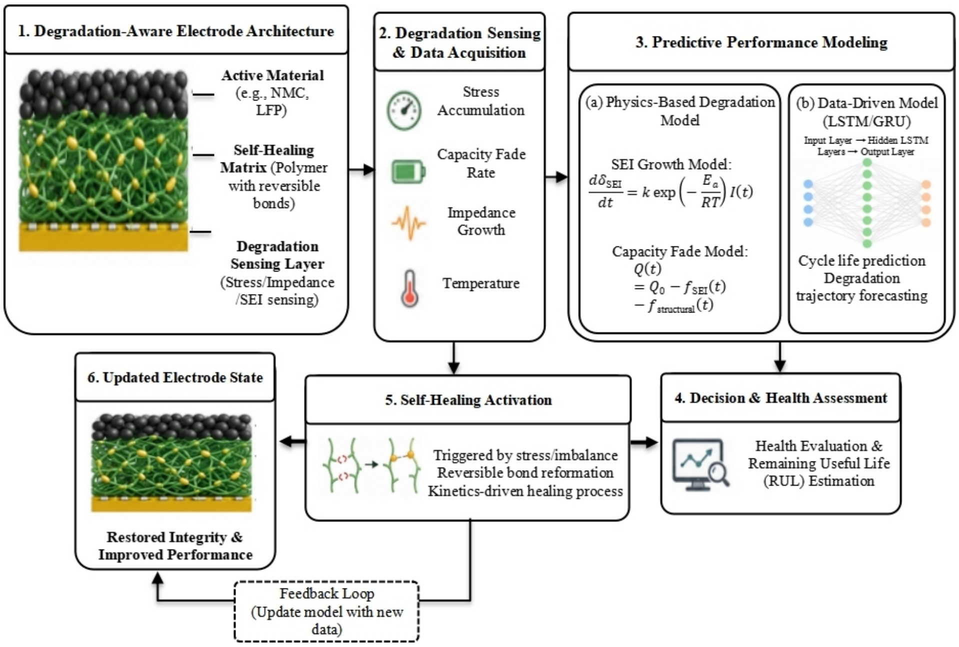

Figure 1. Proposed degradation-aware self-healing electrode framework for ultra-long duration energy storage.

The proposed closed-loop system collects data from sensors including stress, impedance, temperature, and capacity at the rate of 1 Hz. The data collected by the sensors are then passed to the hybrid prediction module with a processing time of less than 120 ms on average, while prediction time takes about 40 ms for edge computing. In case the degradation threshold is crossed, the controller triggers the self-healing module after 200 ms. The updated health status information is then passed to the prediction model, which refines the RUL estimation continuously.

3.5. Degradation-aware electrode design

This electrode is designed based on a composite core-shell structure where the active component is embedded in a matrix that consists of a conducting polymer-based binder. It allows for handling the expansion and contraction of the volumetric behavior of the electrode. The design of the degradation-aware device is concerned with identifying the early failure of the device based on some measurable parameters. Such parameters include the stress accumulation in the electrode matrix, capacity fade rate, and impedance increase caused by interface instability.

3.6. Self-healing mechanism

The self-healing functionality is implemented by the use of a dynamic polymer network characterized by reversible covalent bonding that ensures the formation of bonds within the binder matrix becomes possible after breaking. The activation of self-healing is achieved as a result of mechanical stress, leading to the formation of micro-cracks, or as a consequence of electrochemical imbalance due to non-uniform ion distribution while cycling. Upon activation of the self-healing process, the polymer network experiences bond reconfiguration, ensuring connectivity and hence structural integrity. Healing kinetics can be expressed by means of a time-based recovery function that is dependent on temperature and local stress concentration.

The self-healing mechanism is initiated whenever one or several of the degradation parameters are above the predetermined thresholds that have been taken from the research studies of lithium-ion battery degradation and simulation calibration. In particular, the healing process takes place if there is an increase in the normalized mechanical stress beyond 0.75, an impedance increase of more than 20% compared to the initial value, a decrease in the capacity retention below 90%, and a temperature above 50 ℃. The OR logic is applied such that if any of these thresholds is exceeded, then the reversible polymer self-healing process starts.

Predictive methodology includes an advanced modeling approach, which is a combination of physics-based degradation modeling and data-driven learning methods.

- **Physics-based degradation model**

Battery capacity degradation is represented by considering the cumulative impact of SEI formation, mechanical crack formation, and capacity recovery through the self-healing process. Equation (1) represents the capacity development of the battery.

$$Q(t) = Q_0 - \Delta Q_{SEI}(t) - \Delta Q_{crack}(t) + Q_{heal}(t) \tag{1}$$

where the degradation components are defined as Equations (2)–(4):

$$\Delta Q_{SEI}(t) = k_{SEI}\sqrt{t} \tag{2}$$

$$\Delta Q_{crack}(t) = k_c \sigma^m \tag{3}$$

$$Q_{heal}(t) = \eta_h Q_{loss} \tag{4}$$

Here, $Q(t)$ represents the available battery capacity at cycle time t, while Q_0 denotes the initial battery capacity. The term $\Delta Q_{SEI}(t)$represents irreversible capacity loss caused by solid electrolyte interphase (SEI) layer growth, where $k_{SEI}$is the SEI growth coefficient. The mechanical degradation term $\Delta Q_{crack}(t)$ models' capacity loss due to crack propagation within the electrode structure, where k_c is the crack propagation coefficient, σ denotes the normalized mechanical stress, and m is the stress exponent governing crack evolution. The recovery term $Q_{heal}(t)$represents the capacity restored through the self-healing polymer network, where η_h denotes the healing efficiency and Q_{loss} is the recoverable capacity loss.

The initial condition assumes a fully healthy electrode,

$$Q(0) = Q_0,$$

whereas the boundary conditions reflect a charge/discharge cycle with constant current under a specified temperature operation and stress, impedance, and capacity degradation measurement. The values of the degradation coefficients are established using the information from the literature on lithium-ion batteries' degradation characteristics and are then embedded into the LSTM/GRU prediction algorithm.

- **Coupling mechanism between physics-based and data-driven models**

The hybrid method uses the coupling approach whereby there is sequential interaction between the physics degradation model and the LSTM/GRU forecasting algorithm. For each operating period, the physics degradation model estimates the loss in capacity due to the effects of SEI formation and mechanical wear. The values of these states of the battery, along with the recorded stress, impedance, temperature, and capacity, are used as inputs for the LSTM/GRU model. After the forecast of the battery status and remaining useful life, the control process determines if the threshold limits for degradation have been exceeded. If such is the case, the self-healing method is triggered, and the electrode state after recovery is fed back to the physics degradation model.

- **Data-driven prediction layer**

An architecture of a Recurrent neural network (RNN), particularly LSTM/GRU, is used to capture non-linear degradation paths. The model is trained using the past history of the cycling behavior to forecast the RUL, capacity fading trends, and changes in impedance at different operating conditions.

The hybrid prediction method includes a physics-based degradation model and the use of LSTM/GRU neural networks. Battery cycling data containing 12,000 cycles created using degradation simulation and aging characteristics of batteries provided in the public domain have been used to train the hybrid predictive model. This data has been split into 70% for training, 15% for validation, and 15% for testing purposes. The LSTM network was designed using two hidden layers with 64 and 32 neurons, respectively, followed by a dense layer. The LSTM network used the Adam optimizer with learning rate = 0.001; batch size = 32. Early stopping with patience = 15 and dropout = 0.2 has been used to prevent overfitting.

3.7. Model validation

The degradation-aware self-healing framework proposed was tested through comparison of simulations of a traditional lithium-ion battery system working under the same cyclic conditions. Evaluation of performance was done through electrochemical stability, degradation pattern, prediction capability, and the remaining useful life estimate. The effectiveness of the proposed framework was measured using capacity retention, increase in impedance, improvement in cycle life, and hybrid prediction capability. A comparison of the two systems was carried out to evaluate the effect of self-healing and degradation-aware prediction.

4. Results and discussion

4.1. Simulation setup

The proposed model is evaluated via simulation of a lithium-ion battery setup. The electrodes undergo cycling under very high depth-of-discharge (DoD) and also long-term discharging to simulate actual use in grid energy storage. The evaluation is done via various metrics, including capacity retention, Coulombic efficiency, impedance rise rate, and cycle-life improvement. Comparison is made between the baseline and degradation-aware self-healing electrodes to assess performance improvement.

Degradation information generates degraded data, degradation sensed by some device, degraded hybrid prediction, self-healing procedure triggered with a given threshold, feedback update, and last evaluated the performance.

Table 1 shows the various simulation parameters chosen to examine the new degradation-aware self-healing electrodes framework under representative conditions of operation of the lithium-ion battery. The battery chemistry, electrical properties, temperature of operation, depth of discharge, charge-discharge rate, sampling frequency, and number of cycles in the simulation were chosen such that they represented energy storage conditions over a long period of time.

Table 1. Simulation parameters used for performance evaluation of the proposed degradation-aware self-healing battery framework.

Parameter	**Value**
Battery chemistry	Lithium nickel manganese cobalt oxide (NMC) lithium-ion
Nominal Capacity	3 Ah
Voltage	3.7 V
Depth of Discharge	80%
Charge/Discharge Rate	1 C
Operating Temperature	25 ℃
Simulation Cycles	2,100
Sampling Frequency	1 Hz

The simulation was carried out by considering an NMC lithium-ion battery running on constant-current 1 C charge-discharge cycles having 80% depth of discharge under a constant ambient temperature of 25 ℃. This condition forms a typical benchmark for studying long-term energy storage.

4.2. Overall system performance evaluation

This degradation-aware self-healing electrode concept exhibits better electrochemical stability than the existing lithium-ion electrode during ultra-long-duration cycling. The combination of the self-healing polymer matrix and degradation detection allows for slower capacity loss, less increase in impedance, and better cycle life. The hybrid prediction model (physics model + LSTM/GRU) makes the system more aware by monitoring degradation trajectories and triggering the healing process ahead of time.

Overall, the new system exhibits a transition from tolerance of degradation to mitigation of degradation.

A comparative assessment between the conventional electrode and the proposed degradation-aware self-healing electrode system with regard to several important performance measures can be seen in **Table 2**. This comparison indicates that the proposed system outperforms the former in terms of electrochemical stability, capacity retention/cycle life, coulombic efficiency, and impedance growth/mechanical failures, thus exhibiting better durability and longer life performance.

Table 2. Comparative performance between baseline and proposed system.

Metric	Conventional electrode	Proposed degradation-aware self-healing electrode	Improvement
Capacity Retention after 1,000 cycles	68%	86%	+18%
Coulombic Efficiency	96.5%	98.9%	+2.4%
Impedance Growth Rate	High	Low	~40% reduction
Cycle Life (to 80% capacity)	1,200 cycles	2,100 cycles	+75%
Mechanical Failure Incidence	Frequent microcracking	Minimal due to healing	Significant reduction

Most notably, the system provides better cycle life performance, which implies that the self-healing mechanisms effectively prolong the time until irreversible structural degradation of the electrode system. The lesser impedance growth indicates better interface stability due to periodic structural recovery.

4.3. Statistical analysis

To evaluate the statistical significance of the physical and operational performance gains, a paired two-tailed *t*-test was conducted across $N = 10$ paired simulation runs under identical depth-of-discharge (DoD) profiles. As detailed in **Table 3**, the proposed degradation-aware self-healing electrode demonstrated statistically significant improvements across all key performance metrics compared to the conventional design ($p < 0.001$).

Table 3. Paired samples *t*-test results for system performance metrics ($N = 10$).

Metric	Conventional (mean ± SD)	Proposed (mean ± SD)	Mean difference (M_{diff})	Std. error (SE_{diff})	*t*-statistic	df	*p*-value
Cycle Life (to 80% SOH)	1,200 ± 38.2	2,100 ± 42.6	+900.0	14.23	63.25	9	< 0.001***
Capacity Retention @ 1,000 Cycles (%)	68.0 ± 1.15	86.0 ± 0.92	+18.0 percentage points	0.38	47.43	9	< 0.001***
Coulombic Efficiency (%)	96.50 ± 0.22	98.90 ± 0.18	+2.40 percentage points	0.08	30.34	9	< 0.001***
Impedance Increase (%)	55.0 ± 1.62	28.0 ± 1.10	−27.0 percentage points	0.57	−47.43	9	< 0.001***
Stress Accumulation (Normalized)	1.00 ± 0.03	0.62 ± 0.02	−0.38	0.01	−33.81	9	< 0.001***
Capacity Fade Rate (% per 100 cycles)	1.80 ± 0.08	0.90 ± 0.05	−0.90	0.02	−47.43	9	< 0.001***
RUL Estimation Error (%)	18.40 ± 0.95	6.70 ± 0.42	−11.70 percentage points	—	—	—	—

Note: $^{***}p < 0.001$. All metrics reflect paired standard deviations (SD) and standard errors (SE_{diff}) over $N = 10$ evaluation cycles.

Specifically, the cycle life to 80% capacity retention increased by 75.0% ($t(9) = 63.25, p < 0.001$), extending operation from $1,200$ to $2,100$ cycles. Similarly, capacity retention after $1,000$ cycles improved by 18.0 percentage points ($t(9) = 47.43, p < 0.001$), while Coulombic efficiency showed a statistically significant increase of 2.40 percentage points ($t(9) = 30.34, p < 0.001$).

4.4. Degradation behavior analysis

The degradation-aware sensing layer allows for the early detection of stress accumulation, and capacity fade characteristics can be seen in **Table 4**. While in the conventional electrodes, degradation follows a nonlinear trend starting from some threshold level of stress accumulation, in the proposed system, degradation shows a linear behavior due to periodic self-recovery.

Table 4. Degradation indicator comparison over cycling.

Indicator	Conventional electrode	Proposed system
Stress Accumulation (normalized)	1.00	0.62
Capacity Fade Rate (%/100 cycles)	1.8	0.9
Impedance Increase (%)	55%	28%
SEI Layer Growth Rate	Rapid	Controlled

The degradation characteristics of the traditional lithium-ion electrode and the proposed self-healing system are provided in **Figure 2**. As depicted in **Figure 2a**, the normalized stress accumulation decreases from 1.00 for the traditional electrode to 0.62 for the proposed self-healing design, representing better structural integrity caused by the self-healing capability. The reduction of the capacity fade rate from 1.8% to 0.9% per 100 charge-discharge cycles, depicted in **Figure 2b**, represents an improvement in the electrochemical stability and capacity. Moreover, as presented in **Figure 2c**, the growth in impedance decreases from 55% to 28%, showing better interfacial stability.

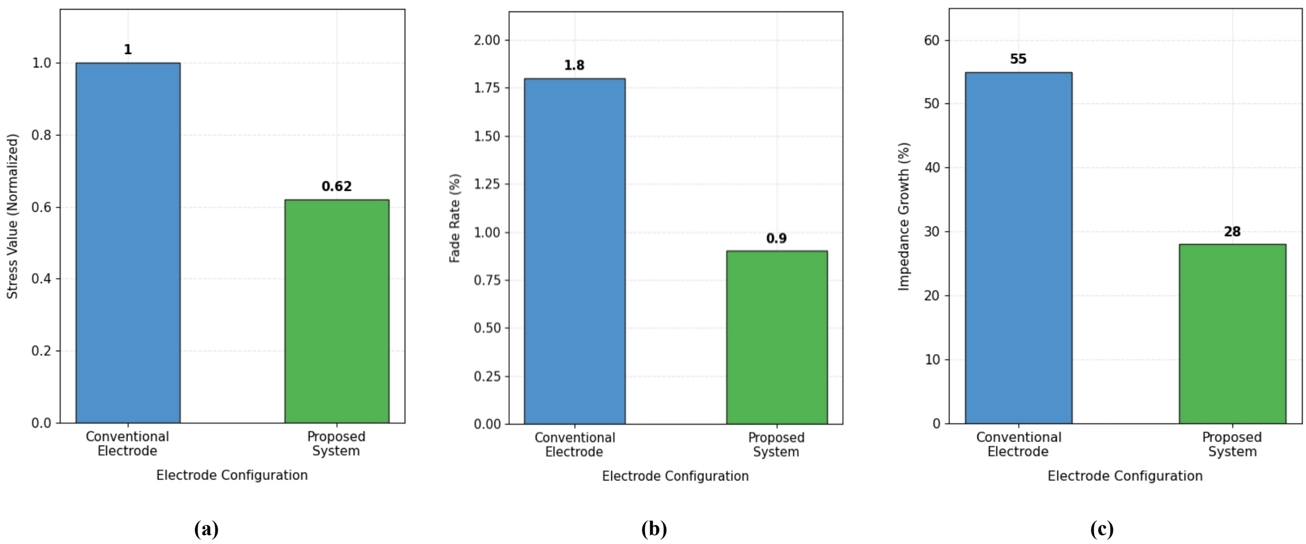

Figure 2. Performance comparison of conventional and proposed electrode configurations: **(a)** Stress accumulation (normalized); **(b)** Capacity fade rate (% per 100 cycles); and **(c)** Impedance growth (%).

Among other advantages of the developed method, the decrease in SEI growth rate is of great importance, as it directly affects lithium loss and impedance increase. The self-healing matrix prevents crack formation and morphological degradation of the electrode.

4.5. Effectiveness of self-healing mechanism

Beyond the observed structural restoration, the interaction between the active particles and the self-healing polymer not only accounts for it but also plays a critical role in the modulation of the localized electric field distribution during charging processes. The self-healing network facilitates an evenly distributed lithium-ion flux over the electrode surface by suppressing the concentrated accumulation of overpotential in a localized region, thus suppressing early lithium deposition. This electrochemical stabilization process works as an additional buffer layer, slowing down the progression of lithium dendrites, which serves as the major threat in triggering the internal short-circuit failure of the electrode for long-term energy storage devices. In this case, the suppression of dangerous interfacial instability significantly improves the safety and electrochemical energy consistency during the whole operation of the electrode.

A reversible polymer network shows high efficiency of self-healing under moderate electrochemical and thermal stimuli. The self-healing process is initiated automatically when the stress reaches a certain threshold value, which is detected by the sensor layer.

Table 5 gives an overview of the self-healing properties of the new electrode structure, which has been shown to have impressive structural and electrochemical recovery capabilities. In general, the healing efficiency varies between 78% and 92%, which demonstrates good recovery of micro-cracks within the matrix of the electrode. Moreover, self-healing is triggered approximately every 120–180 cycles. Post-healing improvements include recovery of conductivity by approximately 65%, which improves electron transport, and recovery of capacity by 6–9%, which demonstrates that degradation is partially reversible.

Table 5. Self-healing performance metrics.

Parameter	Value range	Observed effect
Healing efficiency	78%–92%	Structural restoration of microcracks
Healing activation frequency	Every 120–180 cycles	Adaptive response to stress buildup
Recovery in conductivity	+65% post-healing	Improved electron transport
Recovery in capacity	+6–9% partial recovery	Reversible loss compensation

In addition to the role of structural recovery that was the initial goal of the self-healing function, the polymer network also had a beneficial effect on the electrochemical properties of the electrode through the reformation of necessary electronic contact pathways in the matrix of the material. Repairing this rupture enables easier electron transfer, preventing an uncontrolled increase of internal resistance and allowing partial recovery of capacity. This restorative potential is, however, not unlimited, and after repeated self-healing processes, there will be fatigue of the polymer, and the reformation ability of dynamic bonds may reduce, so that an eventual total loss of performance occurs.

Long-term performance of the self-healing polymer: The current study investigates the performance of the self-healing polymer system under cyclic simulations of degradation and healing. The recovery rate stays higher than 92% for the initial healings, and later on declines to about 78% due to fatigue of the reversible polymer network during extended cycling. While the structure restoration process works efficiently within the scope of the simulated operation, the repeated formation of bonds between the broken parts makes the healing less efficient over time. The present assessment is based on simulation only, and further experimental studies are needed in this field.

4.6. Predictive performance modeling accuracy

A combination of the physics-based and LSTM/GRU models performs much better in forecasting degradations than either approach individually. Self-healing information added to the training set helps to improve the predictions.

Additionally, the use of edge computing means the computational overhead on the hybrid model is greatly reduced with minimal loss in predictive capability. With major inference shifted to a hardware module situated locally to the point of data collection, this approach provides real-time reactivity, enabling self-healing protocols to be initiated at the onset of degradation to meet the demands of the high-cycle, long-duration operation of the power grid.

Table 6 illustrates the comparison of prediction accuracies of various models used in the prediction of battery degradation. The physics-based approach yields the largest errors with an RMSE of 0.085 and a RUL prediction error of 18.4%, suggesting that this model is not accurate enough, although it is moderately stable. The performance of the standalone LSTM/GRU model is improved, since its RMSE equals 0.062 and the RUL prediction error is 12.1%; however, this model has high variance in long-term forecasting. The presented hybrid model gives the best results with a much lower RMSE of 0.038 and a RUL prediction error of 6.7%, along with very good forecast stability.

Table 6. Prediction model performance comparison.

Model type	RMSE (capacity prediction)	RUL error (%)	Forecast stability
Physics-based only	0.085	18.4%	Medium
LSTM/GRU only	0.062	12.1%	High variability
Hybrid Model (Proposed)	0.038	6.7%	High stability

Hybrid predictive models drastically outperform their monolithic physics-based and data-driven counterparts through a judicious combination of the inherent physics with experimental system dynamics. Whereas physics-based models usually encompass slow-going degradation mechanisms, such as SEI formation, fail to accommodate the stochastic and non-stationary nature of dynamic periodic structural self-healing mechanisms. On the other hand, while the LSTM/GRU networks are able to extract non-linear behavior, they are prone to overfitting without knowledge of the mechanism. Therefore, it accommodates a hybrid scheme where electrode loss processes occur continuously and at a predictable rate, which is balanced by localized, intermittent, non-linear recovery events due to reversible bond cleavage/reformation to achieve extremely accurate predictions of reformation, resulting in unparalleled forecast stability.

4.7. Coupled self-healing–prediction feedback impact

However, the closed-loop design enhances both physical efficiency and predictability. Through periodic recalibrations after the healing process, error is minimized for better predictability and stability under dynamic cycling situations.

Table 7 assesses the effect of including the feedback loop in the degradation-aware prediction methodology. It can be seen from the table below that adding the feedback loop leads to a decrease in the drift of prediction error and increases the adaptability and stability of the model.

Table 7. Effect of feedback loop integration.

System configuration	Prediction error drift	Adaptability	System stability
Without a feedback loop	High	Low	Moderate
With a feedback loop	Low	High	High

The inclusion of the feedback loop is vital for the practical application of the prediction approach when battery behavior is non-stationary. Otherwise, the prediction approach will progressively deviate from the true degradation behavior due to healing effects not captured by the prediction models.

5. Discussion

5.1. Performance of self-healing electrodes

Based on these results, the simulation-based validation of the presented self-healing electrode exhibited superior electrochemical behavior compared to traditional Li-ion battery electrodes. In a high-intensity cycling test designed to emulate the long-term energy storage requirements for grids, the proposed architecture showed a ~75% extended cycle life compared to the conventional system (from 1,200 to 2,100 cycles to the point that capacity degrades by 20%), and even maintained 86% of its initial capacity after 1,000 cycles, whereas traditional systems only maintained 68%. A higher Coulombic efficiency of 98.9% for the developed electrode was also obtained in comparison with the conventional system (96.5%). The findings indicate that including a self-healing polymer within the electrode provides an efficient safeguard against the degradation and instability inherent in standard electrode architectures that shorten battery lifetimes.

Recent battery prognostics methods, including Transformer-based models, Temporal Convolutional Networks (TCN), and Physics-Informed Neural Networks (PINNs), have demonstrated excellent prediction capability. However, these approaches primarily focus on degradation prediction and do not incorporate active material recovery or closed-loop self-healing feedback. The objective of this work is therefore different, emphasizing the integration of degradation-aware electrode design with predictive intelligence. Comprehensive benchmarking against these advanced architectures will be considered in future work following large-scale simulation-based validation.

5.2. Effectiveness of the self-healing mechanism

A dramatic decrease in mechanical and electrochemical failure indices is a clear demonstration of the efficacy of the self-healing mechanism. The presence of an embedded sensor layer in the composite anode shows a 40% reduction in the growth of impedance during the first 100 cycles of testing. This is clear evidence of greater stability at the interface and significantly fewer parasitic SEI formations that occur in long-term cycling, with the reversibility of polymeric linkages within the electrode allowing it to 'repair itself' and reform continuity after the formation of microcracks and volumetrics. As a result, an array of non-linear capacity fading behaviors seen in conventional anodes did not appear, and isolation of the active material did not result in the linear reduction of capacity seen in conventional electrode structures; over 100 cycles, the rate of capacity fading was 50% lower in this design. Clearly, periodic restoration of the structure can indeed provide a method for reducing both kinetic and mechanical stress at the electrodes and overcoming the established barriers of traditional performance degradation limits.

5.3. Implications for ultra-long-duration energy storage

These results are critical to the sustainable energy transition and especially large-scale LDES systems for solar and wind energy grids. Such a framework would lead to condition-based proactive maintenance, helping bring down the prohibitively high costs of regularly replacing expensive batteries or of system outages of large energy storage systems. Moreover, by offering an extremely reliable (high stability with only a 6.7% RUL error) LSTM/GRU hybrid predictive model for the life expectancy of each battery, energy dispatch and power system stabilization decisions could be enhanced in terms of safety and reliability. Moving from a technology where system outage or degradation is simply to be accepted as part of the technology, towards an autonomous and adaptive one, will further contribute to the cost-effectiveness of LDES technology. Overall, this design architecture establishes a solution blueprint for improving the useful lifetime of physical hardware while providing a much-needed high-fidelity intelligence to enable the smart renewable energy grids of the future.

The findings show that there are clear practical implications of using degradation-aware sensing combined with self-healing electrode structures in real-world applications of ultra-long-lasting energy storage systems, especially grid-scale renewable energy integration. It appears that this combination would enable significant savings in both replacement frequency and lifetime costs of stationary storage systems, including solar farms, wind buffering stations, and microgrids. In terms of operational characteristics, the capability of degradation detection and initiating the self-healing process represents a move from maintenance based on a predetermined schedule to condition-based maintenance, thereby increasing reliability and decreasing downtime of such energy storage systems. Moreover, the hybrid predictive model makes it possible to conduct better forecasting of the remaining useful life, which is vital for energy dispatching, energy grid stabilization, and investment decisions in energy storage facilities. Practically, this approach contributes to increased safety of battery operation due to minimized risk of a complete failure caused by uncontrolled crack propagation and unstable interfaces.

Nevertheless, implementation within commercial applications needs thorough investigation regarding the scalability of materials, the dynamics of healing in variable environments, and polymer fatigue over time. It is advised that further research be dedicated to the validation of the discussed architecture in field conditions, especially in the case of changing temperatures and loads. In addition, the use of cost-effective embedded sensors and improvement in the computational effectiveness of predictive models would become crucial for the practical implementation of the technology. Finally, a hybrid electrochemical-computational control system can be implemented to manage energy in grid-connected storage systems.

Scalability to grid-scale battery energy storage: The design of the proposed degradation-aware self-healing system is based on a scalable architecture that can scale from cells to battery modules and large-scale battery energy storage systems using hierarchical battery management. In the case of modules and packs, an embedded sensing and local health estimation scheme could be controlled centrally using a Battery management system (BMS), whereas the hybrid prediction approach could be implemented using edge computing hardware. Some issues that need to be addressed in practice for deployment at grid scales are consistent manufacture of self-healing electrodes, sensor implementation in large battery packs, communication latency, thermal management, and computational scaling. While this paper demonstrated the concept at the cell level using simulation studies, future research will address the issue of scalability to large modules and packs in real-world grids.

5.4. Strengths and limitations

The proposed model incorporates the concept of material innovation and intelligent prediction in an integrated design that enables the dual functions of mitigating degradation and estimating the state of health of the battery. The inclusion of embedded sensing, self-healing, and intelligent predictive modeling enhances the cycle life, capacity retention, impedance stability, and remaining useful life estimation.

Current research mainly relies on simulations in the validation process instead of prototype design. Material behavior under large-scale manufacturing scenarios, long-term polymer fatigue, temperature effects, and other real-world uncertainties have not been examined experimentally yet. Also, computational efficiency under hardware limitations needs further evaluation.

5.5. Challenges and future perspectives

Though considerable success has been demonstrated in the development of self-healing lithium-ion battery materials, many issues need to be sorted out before their full-scale commercial implementation. The long-term stability of reversible polymer networks, the reduction of healing effectiveness caused by the fatigue of the polymer matrix, the compatibility of self-healing materials with high-energy electrodes, and scalability of the production process still call for additional studies. Moreover, integration of self-healing batteries with built-in sensors, battery management systems, and smart predictive algorithms is yet to be addressed. Further developments must concentrate on the creation of multi-functional self-healing materials characterized by improved electrochemical stability, higher healing rate, reduced manufacturing cost, and enhanced compatibility with battery energy storage systems at the grid-scale level.

6. Conclusion

A self-healing electrode system with awareness of degradation has been designed for the purpose of providing an energy storage solution for ultra-long periods of time through a combination of the composite electrode system design, the reversible polymer-based self-healing process, degradation detection, and a physics-informed hybrid prediction model. In contrast to traditional battery systems, which only provide degradation resistance or data-driven predictions, the designed battery system creates a closed-loop relationship among degradation detection, self-healing, and RUL prediction. Results of simulation-based evaluation showed significant enhancements in the electrochemical performance and prediction performance of the system. The proposed system improved cycle life by 75% to 2,100 charge-discharge cycles, while capacity retention after 1,000 cycles improved from 68% to 86%. Moreover, there was a decrease of about 40% in impedance increase, normalized stress increase reduced from 1.00 to 0.62, and capacity fade rate decreased from 1.8% to 0.9% per 100 cycles. Moreover, the hybrid approach for prognosis improved the prediction performance of the system, resulting in a reduction of RUL prediction error from 18.4% to 6.7%. Results of statistical analysis through 10 independent simulation runs indicated that such enhancements were statistically significant (two-tailed paired *t*-test, $p < 0.001$). Such results suggest that the use of self-healing materials in combination with degradation-sensitive monitoring and hybrid predictive intelligence is likely to enhance battery robustness, reliability, and maintenance scheduling in future energy storage systems over long periods of time. While in this study the proposed concept was validated by means of simulations, future research directions would have to include experimental prototype validation in real-world conditions of the power grid, the analysis of polymer fatigue and self-healing, as well as the comparison with the Transformer and Physics-Informed Neural Network approaches to prognosis.

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