

Real-time artificial intelligence for solid-state lithium metal batteries

Corresponding Author: Professor Xin Li

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

In this manuscript, the authors employ real-time AI-driven state perception and reinforcement learning approaches to analyze the critical role of intelligent charging strategies in suppressing dendrite growth and delaying the "knee point" phenomenon of solid-state lithium metal batteries. Furthermore, the effectiveness of the strategy and the evolution path of battery states are validated through SEM/EDS characterization.

1. Line 24: Suggest changing "to decarbonization" to "for decarbonization."
2. Line 34: Suggest changing "In Li-ion battery it can..." to "In Li-ion battery, it can..."
3. Line 86: The different solid-state batteries in the dataset should be explained. Different electrode and electrolyte materials often exhibit distinct electrochemical degradation pathways. How can the universality of the network trained on different battery systems be demonstrated?
4. Line 128: The cycling results of the 7 benchmark batteries mentioned, specifically the cycling condition of 18C-4.25V, should be evaluated for representativeness. It is generally assumed that lower rates lead to longer lifespan. When the benchmark group is cycled at a lower rate, does the intelligent cycling strategy still maintain its advantage? Suggest supplementing experiments.
5. Please explain the supporting role of the analysis in Fig. 4b, c, e, and f for the main contributions of this article. The PCA analysis demonstrates differences between intelligent cycling and benchmark testing in state space, but the analysis remains superficial and does not explain the specific physical meaning of the principal components or their relationship to battery performance. Suggest providing a detailed explanation.
6. In Fig. 4 h, it is evident that the capacity retention of the 6 cut-off group is higher than that of intelligent cycling. Please provide an explanation.
7. Line 172: The evaluation of the impact of adding charging actions to manually programmed battery test procedures should compare the degradation of the two groups under the same rate and cut-off voltage frequency as in intelligent cycling. Suggest supplementing this comparison.
8. Line 448: "dacays" should be corrected to "decays".

(Remarks on code availability)

Reviewer #2

(Remarks to the Author)

This work presents a closed-loop battery testing platform incorporating AI for real-time adaptive cycling. The authors propose a system in which a deep learning-based state perception module and a reinforcement learning (RL) agent interact with a commercial battery tester to optimize cycling conditions (battery C-rate and voltage cutoff) based on recent cell behavior. The approach is demonstrated on solid-state lithium metal batteries (SS-LMBs), and the results show significant improvement in battery longevity and accumulated energy throughput compared to fixed-strategy testing benchmarks.

The work's main conceptual innovation integrating AI models into the experimental loop of battery cycling—is timely and highly relevant. It aligns well with emerging trends in intelligent BMSs, where ML could extend cycle life and reduce safety

risks, and potentially uncover hidden, incubating aging mechanisms to prevent failure or avoid knee point. This conceptual framework is a promising direction and represents a necessary evolution beyond static, model-based or heuristic cycling strategies.

Still, in reviewers view, the work as presented does not fully attain the proposed goals and objectives, and several critical issues limit its impact, reproducibility, and generalizability. While the paper introduces interesting elements, it falls short in several areas, which shall be commented on below:

1. System architecture, battery testing infrastructure and reproducibility

A key concern relates to the implementation of the AI–hardware integration, which is insufficiently described given that the manuscript aims to present a hardware–software control platform.

The authors state that adaptive cycling was conducted using a Solartron XM MTS system, while fixed benchmark testing was performed on an Arbin BT2000. The Solartron, however, is a commercial battery testing machine, but not a standard platform for high-throughput or standard programmable BMS-level testing. It remains unclear:

- A comprehensive description of the protocol(s) or software layer(s) used to enable real-time communication between the Python-based AI control and the Solartron system and Windows Operating System that controls the Solartron
- Whether the feedback control was synchronous or batch-based, and how latency and reliability were evaluated and handled
- Is the AI module running on a separate workstation and received the Solartron data and then sends updates the control parameters? How?
- How the system could be generalized to commercially relevant test stations (e.g., BioLogic, Maccor, Neware) or implemented in edge computing

In addition, without a detailed system diagram (Fig. 1 is too general) or communication/control workflow, the platform's portability and reproducibility cannot be assessed. If the authors intend to present a novel AI-powered battery control system, this aspect must be central in the manuscript. Currently, the AI agent appears more like an external script that interfaces with a test station, rather than an integrated, deployable BMS architecture.

2. Data sufficiency and model generalizability

The developed AI models rely on a dataset of just only 28 solid-state cells, collected from historical lab experiments, to train the ResNet50-based state classifier. While the authors attempt to validate the classifier on independent cells, this dataset is extremely limited in terms of size, diversity, and representativeness—especially for training deep learning models and RL agents.

It is also unclear:

- How the dataset was split (train/validation/test)
- Whether cells from similar configurations were grouped across splits, risking data leakage
- If any data augmentation or uncertainty quantification was applied to improve robustness

Furthermore, all data appears to originate from lab made LMBs, which to this stage suffer from high variability and low reproducibility (as shown in Ext. Fig. 1). The use of such limited and variable data raises substantial questions about model generalizability, especially across different chemistries or battery formats.

What remains strange to the reviewer is that, had the authors used commercial, off-the-shelf lithium-ion batteries—which are more reproducible and come in diverse chemistries—they could have better demonstrated the robustness and wide applicability of the AI platform. And saving time wouldn't be an issue, either (i.e., cycling at high temperatures, high rates, plating, etc.). This makes the case that solid-state LMBs were not necessary for this demonstration, and in fact, their use may have constrained both the model's performance and its relevance and add too much information not needed to present the platform.

3. Title and scope

The manuscript title emphasizes “real-time AI-powered solid-state lithium metal batteries”, which is misleading. The innovation lies not in the materials science of solid-state batteries, but in the AI-based control methodology. Also, if the AI logic isn't embedded but is a software agent controlling a lab instrument externally, then calling this “real-time AI-powered” control requires qualification. It's more of a scripted closed-loop AI-supervised platform.

Similarly, the manuscript devotes extensive space to microstructural evolution, SEM/EDS imaging, and even modeling of interfacial degradation using a 2D grid simulation. While informative, these analyses appear secondary to the main contribution. The platform, which should be central, is lightly described, lacking architectural detail, block-level diagrams, or algorithm–hardware flowcharts. This mixing of material characterization with high-level systems and AI objectives creates conceptual fragmentation, detracting from the manuscript's clarity and weakening the scientific message. If the core novelty is the intelligent cycling framework, then the focus must shift more decisively towards the architecture description, the experimental integration of control and validation under diverse scenarios and the comparison with other heuristic and conventional adaptive methods.

In total, this manuscript aims to introduce a novel methodology that combines data-driven state classification with reinforcement learning for adaptive cycling, which is a novel and attracting idea. However, the work as presented lacks critical validation, experimental detail, and architectural clarity to support the strength of its claims.

The main issues are:

- The integration of AI into battery control is more conceptual than demonstrable at this point
- The limited dataset and battery format restricts the generalizability of the conclusions
- The overemphasis on solid-state lithium metal batteries is misaligned with the core innovation, which is platform-level
- The system engineering aspects, particularly the AI–hardware interface, are under-described and difficult to reproduce

which in reviewers' opinion weakens the case for publication in Nature Communications on its current form.

(Remarks on code availability)

The computer code generated and used during this study would be available from the corresponding author on reasonable request.

Reviewer #3

(Remarks to the Author)

This work presents a novel approach leveraging AI and reinforcement learning to both estimate the state of solid-state lithium metal batteries and to adjust their cycling protocols to delay accelerated degradation and extend energy throughput. This work appears to represent a considerable contribution to the space of battery diagnostics and operation, however, presentation issues mean that the work should not be published in its current state. Specifically, the language and figures must be improved prior to publication.

The following comments are presented in no particular order:

The readability of the article is considerably degraded by the clarity of the language and grammar. There are many tools and resources available to refine the language. I encourage the authors to leverage them.

For the title, I would suggest instead, "Real-time AI for solid-state lithium metal batteries." The current title seems to indicate that the batteries are AI powered in real time, which doesn't make a lot of sense.

In the first line of the abstract, "battery cycling lifetime" should be replaced by "battery degradation" or "battery health evolution"

Figure 1 is difficult to interpret. Especially the battery images on the top left of the figure that seem to indicate the battery state of health dramatically improving and then degrading in time, a characteristic not seen in real cells.

Figure 2 indicates that the normalized capacity in the transitional state ranges between 0.9 and 0.5 SOH. However, in industrial settings, 0.8 is typically selected as the threshold for end of life. Why is this transitional period defined to cross such a dramatic change in SOH? Intuitively, once the battery exhibits roll-over, you are no longer in a transitional period, you are deep into decay.

The interpretation of Fig 4a is unclear. For example, upwards of cycle 2000, it appears that the state perception is 1 across a large number of action indices simultaneously. I suggest that the figure be explained in greater detail, or another approach is used to represent the concept.

In all figures and in the text, Crate should be changed to C-rate

Figure 4 plots the "Euclidean distance" for the intelligent and benchmark case, but it is unclear what this distance is measured to and from.

It is unsurprising that the AI input features are scattered in input space for the intelligence case versus the benchmark due to the constantly changing charging protocols. This does not necessarily indicate anything about the state of health as these changes in cycling protocol are not deconvolved. I am not convinced of the value of this PCA analysis.

The analogy between the battery and neural networks is suspect. I suggest removing it as it does not help explain the phenomena nor is it well supported by evidence.

Should the data and code not be made publicly and easily accessible, the authors should make a reasonable justification. Not making these accessible significantly decreases the impact on the community and the chance of reproducing these results.

(Remarks on code availability)

code was not available

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The authors have made necessary revisions.

(Remarks on code availability)

Reviewer #2

(Remarks to the Author)

Dear authors,

I have carefully reviewed the revised version of the manuscript and I would like to thank the authors for the substantial improvements made in response to the reviewers' comments.

The revised manuscript addresses the concerns raised during the review process in a thorough and satisfactory manner.

The quality of the presentation, clarity of the arguments, and scientific rigor have all been significantly enhanced.

Thank you,

(Remarks on code availability)

Reviewer #3

(Remarks to the Author)

I thank the authors for properly addressing most of my presentation and technical concerns.

I would encourage the authors to improve the grammar throughout the article. For example, the first line of the introduction should be "The Li-ion battery is one of the most powerful technologies in our era for decarbonization and combating climate change" instead of "Li-ion battery is one of the most powerful technologies in our era for decarbonization and combat climate change." Please either leverage a native English speaker at one of your institutions, or even ask AI to review the document and provide suggestions on grammatical changes with the reasoning for these changes.

(Remarks on code availability)

I did not run and install the code, as this would require the requisite dataset and hardware. The necessary code appears to be present with comments. The codebase would be of much greater value to the research community if it came with a detailed demonstration notebook with associated datasets and instrument/battery simulators.

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II. RESPONSE TO REVIEWERS.

Reviewer #1:

In this manuscript, the authors employs real-time AI-driven state perception and reinforcement learning approaches to analyze the critical role of intelligent charging strategies in suppressing dendrite growth and delaying the "knee point" phenomenon of solid-state lithium metal batteries. Furthermore, the effectiveness of the strategy and the evolution path of battery states are validated through SEM/EDS characterization.

1. Line 24: Suggest changing "to decarbonization" to "for decarbonization."

2. Line 34: Suggest changing "In Li-ion battery it can..." to "In Li-ion battery, it can..."

Response: Thank you for your suggestion. We have revised the manuscript accordingly and corrected the relevant descriptions in Lines 24 and 34 as recommended.

3. Line 86: The different solid-state batteries in the dataset should be explained. Different electrode and electrolyte materials often exhibit distinct electrochemical degradation pathways. How can the universality of the network trained on different battery systems be demonstrated?

Response: Thank you for your insightful comment. We would like to clarify that although the dataset was collected from different group members, all the solid-state cells used in this study were based on a consistent battery chemistry system—namely, NMC811 cathodes paired with lithium metal anodes. The primary variation among the cells lies in the electrolyte formulations, which were intentionally preserved to capture realistic diversity in degradation behaviors within the same electrochemical system.

Importantly, we did not tailor the model to any specific failure mechanism or degradation mode, thereby preserving its ability to generalize. The proposed data processing and network training were originally developed in our previous machine learning studies, where it was successfully applied to commercial Li-ion battery datasets [36], and Na-ion battery datasets [Ref. R1]. This consistency in methodology further supports the model's universality across different battery systems.

We acknowledge that different cathode/anode materials would require retraining or transfer learning with data from the respective systems. Actually, the current results demonstrate promising robustness and adaptability of the model within the lithium-metal battery family. We have added a clarifying note in the revised manuscript to prevent any confusion regarding the battery systems used.

[36] Chen, Xi, Jeeseon Choi, and Xin Li. "Machine Learning a Million Cycles as 2D Images from Practical Batteries for Electric Vehicle Applications." *ACS Energy Letters* 7.12 (2022): 4362-4367.

[R1] Xi Chen, Luhan Ye, Yichao Wang, Xin Li†, "Beyond expert-level performance prediction for rechargeable batteries by unsupervised machine learning" *Advanced Intelligent Systems*, 1, 1900102 (2019)

“This consistent cathode/anode configuration ensures a focused evaluation of the model's ability to generalize across different degradation patterns induced by electrolyte variations.”

“Despite the diversity in data sources, all solid-state cells share the same electrochemical configuration that NMC811 cathodes paired with lithium metal anodes. These electrolyte differences among the cells introduce measurable variations in degradation behavior, which can evaluate the neural network's generalization performance across different degradation pathways. Importantly, the model was not trained to target any specific failure mechanism, allowing it to capture broader degradation dynamics without overfitting to a particular pathway.”

4. Line 128: The cycling results of the 7 benchmark batteries mentioned, specifically the cycling condition of 18C-4.25V, should be evaluated for representativeness. It is generally assumed that lower rates lead to longer lifespan. When the benchmark group is cycled at a lower rate, does the intelligent cycling strategy still maintain its advantage? Suggest supplementing experiments.

Response: Thank you for this important question. While it is true that lower C-rates can extend battery lifespan in many liquid electrolyte batteries, recent studies have shown that such assumptions do not universally apply to solid-state lithium-metal batteries. For instance, as demonstrated in our previous work [5], low C-rates such as 1.5 C can result in inferior cycling stability compared to higher C-rates like 15 C and 20 C, and 15 C shows worse cycling stability than 20 C.

In this work, the benchmark cycling rate of 18C was intentionally selected as a representative midpoint within the high C-rate regime from 15 C to 20 C. This choice allows us to meaningfully quantify and compare the performance of intelligent cycling, which dynamically explore a range of rates both above and below 18C. Meanwhile, among the 7 benchmark batteries, three were cycled at 20C, and their stability was found to be comparable to those at 18C (see Figure3c and Supplementary Figure S4). The action of more frequent attempts toward 15 C itself after 500 cycles in our intelligent cycling thus will not lead to better cycling stability, because it should have moved toward the opposite C-rate direction of 20 C instead for better cycling stability, as 15 C shows worse cycling stability than 20 C. Therefore, the complete avoid of the decay state (state #2) by our intelligent cycling has to be the result of the combination of low and high C-rates in consecutive cycles. This also further supports the representativeness and robustness of our benchmark selection across a range of high C-rates.

We therefore believe that the current experimental setup sufficiently supports the proof-of-concept nature of this study, especially given the unique behavior of solid-state lithium-metal batteries under high-rate conditions.

[5] Ye, Luhan, and Xin Li. "A dynamic stability design strategy for lithium metal solid state batteries." *Nature* 593.7858 (2021): 218-222.

“The benchmark conditions were fixed from within the regime of charging actions, which 18C-4.25V was selected as a representative midpoint. This allows a meaningful and quantifiable comparison with the intelligent cycling, which dynamically explores C-rates and voltage cutoff both above and below this benchmark. Also, three of the benchmark cells were cycled at 20C, and their performance was found to be comparable to those cycled at 18C. This reinforces the representativeness and robustness of the chosen 18C condition, and highlights that higher C-rates do not necessarily lead to inferior cycling performance in solid-state lithium-metal batteries, which is consistent with our earlier findings [5].”

5. Please explain the supporting role of the analysis in Fig. 4b, c, e, and f for the main contributions of this article. The PCA analysis demonstrates differences between intelligent cycling and benchmark testing in state space, but the analysis remains superficial and does not explain the specific physical meaning of the principal components or their relationship to battery performance. Suggest providing a detailed explanation.

Response: Thank you for this thoughtful comment. We appreciate the opportunity to clarify the physical and analytical roles of the PCA-based analysis.

First, the shape-based features used in this work were derived from cycling profiles (voltage and capacity over time), and their relevance has been demonstrated in our prior works [36, R1], where shape images were shown to encode information-rich electrochemical characteristics.

In our current study, PCA was applied to these shape images to distill dominant degradation patterns. Extended Data Fig. 4 illustrates the reconstructed contributions of PC1, PC2, and PC3, revealing that:

- PC1 captures global capacity fading trends by horizontal compression of voltage plateaus; PC2 and PC3 are linked to localized fluctuations and distortions in voltage curves, which may be related to internal resistance changes or interfacial instability.

Based on these interpretations, we analyze how intelligent cycling and benchmark traverse the PCA-defined state space:

- Fig. 4b shows that intelligent cycling induces broader and more divergent motion in the PC1-PC2 space under charging actions, implying greater dynamical exploration of battery states through jumping between states to access the PC space that otherwise will not be accessible at all in a normal cycling, which significantly slows down the evolution (and thus degradation) speed of the state trajectory in the PCA space for the intelligent cycling. Intelligent cycling leverages the dynamics to effectively enable recovery from transitional states back into stable states by dynamically varying rates and voltage ranges, though jumping out of the routine battery evolution pathway in the PCA space of a normal cycling with fixed rate and voltage range. Also, Fig. 4c quantifies this effect by showing larger Euclidean distances that signal jumping and reversible state evolution pathways.

In contrast, Fig. 4e and 4f show that benchmark cycling follows a narrow and unidirectional trajectory in PCA space of strong path dependence and minimal Euclidean movement, consistent with irreversible degradation and the early emergence of capacity knees (Fig. 4d)

We have revised the main text and Methods section accordingly to enhance these explanations.

[36] Chen, Xi, Jeesoon Choi, and Xin Li. "Machine Learning a Million Cycles as 2D Images from Practical Batteries for Electric Vehicle Applications." *ACS Energy Letters* 7.12 (2022): 4362-4367.

[R1] Xi Chen, Luhan Ye, Yichao Wang, Xin Li†, "Beyond expert-level performance prediction for rechargeable batteries by unsupervised machine learning" *Advanced Intelligent Systems*, 1, 1900102 (2019)

“Specifically, PC1 reflects the global capacity fading trend (noted by horizontal shrinking of voltage plateaus), while PC2 and PC3 are linked to local fluctuations and distortions, likely representing internal resistance changes or interfacial instability.”

“This suggests the intelligent cycling of dynamic strategy enables a broader and more flexible exploration of electrochemical states, allowing the battery to jump between states of little path dependence and access the principal component space that otherwise will not be accessible in a normal cycling of fixed strategy and unidirectional trajectory evolution in the principal component space of strong path dependence. Euclidean distances calculated between adjacent cycles in the PCA space quantify the state transitions induced by each action step (Fig. 4c). For intelligent cycling, these distances reveal more frequent and larger shifts between consecutive steps, reflecting rapid and reversible transitions between stable and transitional states, which effectively elongates the trajectory of the state evolution in the PCA space and slows down the degradation. In comparison, benchmark cycling exhibits narrow, unidirectional state trajectory (Fig. 4e, Supplementary Fig. 6 and animations) and minimal Euclidean displacement (Fig. 4f), corresponding to irreversible fast degradation and early emergence of knee points. The limited state variability under fixed charging actions restricts the ability to recover once degradation accelerates.”

6. In Fig. 4 h, it is evident that the capacity retention of the 6 cut-off group is higher than that of intelligent cycling. Please provide an explanation.

Response: Thank you for pointing this out. While it is true that the 6-cutoff group in Fig. 4h exhibits slightly higher capacity retention than the intelligent cycling group at 1500 cycles, we would like to clarify it.

This phenomenon aligns with our main conclusion that increasing the number of available charging actions yields diminishing returns in improving battery longevity beyond a certain level. As shown in Fig. 4h and Extended Data Fig. 6, the correlation between action number and performance is non-linear, and beyond a certain threshold, additional actions may not further improve, even slightly reduce performance. The 6-cutoff group may fall into such a marginally optimal point. Meanwhile, insights from our 2D grid simulations (Extended Data Fig. 5) support this interpretation. In the model, we define a fluctuation ratio of interaction

field to represent the extent of the perturbation introduced by varying charging actions. This nonlinear trend provides a conceptual explanation for why a group with limited but well-distributed actions like 6 cutoff group might perform well within a specific range.

On the other hand, it is important to emphasize that knee points exhibit abrupt capacity change, which is not determined by the current SOH. The intelligent cycling framework aims to proactively detect and avoid these transitions through real-time state perception. Therefore, a slightly higher capacity retention of 6 cut-off group at 1500 cycles does not guarantee longer lifetime.

We have added additional clarification in the revised manuscript to better reflect these insights.

“The 6 cutoff group shows slightly higher capacity retention at 1500 cycles compared to intelligent cycling. This outcome likely reflects the diminishing returns effect when increasing the number of available charging actions beyond a certain level. As also illustrated in the 2D grid model (Extended Data Fig. 5), a moderate level of fluctuation may strike a balance between the extent and distribution of local degradations, leading to temporarily favorable outcomes. The 6-cutoff group may happen to lie near such a marginal optimum. However, given the abrupt nature of capacity knee points, short-term advantages in capacity retention do not necessarily imply superior long-term performance. Intelligent cycling is designed to dynamically adapt to evolving battery states, aiming to delay the onset of irreversible degradation.”

7. Line 172: The evaluation of the impact of adding charging actions to manually programmed battery test procedures should compare the degradation of the two groups under the same rate and cut-off voltage frequency as in intelligent cycling. Suggest supplementing this comparison.

Response: We appreciate the reviewer’s valuable comment and suggestion. Regarding the frequency of rate and cut-off voltage, in our study, both the intelligent cycling and random testing (ED Fig. 6h) groups were operated under the same frequency of switching the charging actions every three cycles, where intelligent cycling shows obviously better capacity retention than the random cycling using the same ranges of C-rates and voltage cutoffs as the intelligent cycling (Fig. 4h).

We fully agree that ideally it will be great if we can run long-cycle tests with random or other programmed changing actions every 3 cycles for the conditions in ED Fig. 6a-e as well. However, due to practical constraints including the long duration required for these cycling tests, limited channels of the testing instrument, and high instrument occupancy from other long-term battery studies, these tests could not be performed.

That said, we hope that our explanation and analysis of the existing data could lift the concerns from the reviewer. ED Fig. 6a-e are at the same frequency of changing actions every 50 cycles, while they show very different capacity retention, suggesting that frequency is not the most dominant factor here. Instead, there is a general trend that the group of 6 actions (6 cutoffs or 6 rates) shows better retention than 4 actions (2 cutoffs and 2 rates), which is better than the group of 2 actions (2 cutoffs or 2 rates), suggesting that increasing the number of different actions is more important to better retention when the number of actions is smaller than 6. In contrast, further increasing the action number to 18 with the random switch every 3 cycles reduced the capacity retention, suggesting the importance of intelligent cycling when more actions are included.

We would like to emphasize that Figure 3ab presents statistically meaningful results across multiple batteries, confirming the effectiveness of intelligent cycling, without the need to manually test and program the number and frequency of actions to include. The action number dependence in Fig. 4h also helps probe finer aspects of the charging strategies, and the observed diminishing returns support a deeper mechanistic understanding that is consistent with our modeling work (ED Fig. 5).

We have clarified these points in the revised manuscript to avoid any potential confusion and highlight the experimental rationale.

“Comparing those tests with fewer actions of 2 or 4(ED. Fig. 6a-c) and with more actions of 6 (ED. Fig. 6d-e) with the action switch of every 50 cycles, it shows increasing the number of actions helps the retention before the effect of action number converges at 6 actions. All the 18 charging actions were executed in the random action test (ED Fig. 6h) and were updated every 3 cycles, matching the interval and actions used in intelligent cycling. The lower capacity retention in the random action test than the intelligent one and the 6-actions one suggests the importance of intelligent cycling when more actions (e.g. 18) are included, without the need to test and program the optimum number of actions and frequency to switch. Action number dependence (Fig. 4h) also offers insight into the underlying mechanisms of increasing charging complexity (ED. Fig. 5).”

8. Line 448: "dacays" should be corrected to "decays".

Response: Thank you for the careful reviewing. We have corrected this typo.

Reviewer #2:

This work presents a closed-loop battery testing platform incorporating AI for real-time adaptive cycling. The authors propose a system in which a deep learning – based state perception module and a reinforcement learning (RL) agent interact with a commercial battery tester to optimize cycling conditions (battery C-rate and voltage cutoff) based on recent cell behavior. The approach is demonstrated on solid-state lithium metal batteries (SS-LMBs), and the results show significant improvement in battery longevity and accumulated energy throughput compared to fixed-strategy testing benchmarks.

The work's main conceptual innovation integrating AI models into the experimental loop of battery cycling—is timely and highly relevant. It aligns well with emerging trends in intelligent BMSs, where ML could extend cycle life and reduce safety risks, and potentially uncover hidden, incubating aging mechanisms to prevent failure or avoid knee point. This conceptual framework is a promising direction and represents a necessary evolution beyond static, model-based or heuristic cycling strategies.

Still, in reviewers view, the work as presented does not fully attain the proposed goals and objectives, and several critical issues limit its impact, reproducibility, and generalizability. While the paper introduces interesting elements, it falls short in several areas, which shall be commented on below:

Response: We sincerely thank the reviewer for the thorough and constructive feedback. We are delighted that the reviewer recognizes the value and potential of this research direction. The insightful comments have been instrumental in improving the clarity and quality of our manuscript to better align with *Nature Communications* standards. We have carefully addressed each concern and revised the manuscript accordingly.

1. System architecture, battery testing infrastructure and reproducibility

A key concern relates to the implementation of the AI – hardware integration, which is insufficiently described given that the manuscript aims to present a hardware-software control platform.

The authors state that adaptive cycling was conducted using a Solartron XM MTS system, while fixed benchmark testing was performed on an Arbin BT2000. The Solartron, however, is a commercial battery testing machine, but not a standard platform for high-throughput or standard programmable BMS-level testing. It remains unclear:

Response: We thank the reviewer for pointing out this concern. In fact, no proprietary software modifications to the battery testing hardware were required. The integration of AI with the battery tester was realized by us entirely from the workstation side by leveraging standard communication protocols and application programming interfaces (APIs).

The Solartron XM MTS system used in this study is an electrochemical battery test station available in our lab, and it served the purpose of validating real-time communication and control with the AI agent. While it is not a high-throughput testing system, it served well for our proof-of-concept demonstration. Importantly, we have also consulted with commercial vendors such as Arbin and confirmed that their modern programmable BMS-level testing platforms also support integration with external control agents via open APIs. Unfortunately, the Arbin BT200 system in our lab is too old to support this functionality.

In light of the reviewer's comment, we have revised the manuscript to better clarify this point and emphasize that the proposed platform is readily transferable and scalable across other compatible battery testing systems.

“The platform integrates a Python-based control on a computer workstation with the battery testing hardware (Solartron XM MTS) for real-time data exchange and parameter updates. Notably, this implementation does not rely on proprietary software modifications to the hardware.”

- A comprehensive description of the protocol(s) or software layer(s) used to enable real-time communication between the Python-based AI control and the Solartron system and Windows Operating System that controls the Solartron

Response: We appreciate the reviewer's request for a more comprehensive description of the real-time communication architecture.

For software layer, real-time control was implemented using Component Object Model (COM) automation APIs officially provided by Solartron. COM is Windows-based technology for inter-process communication and dynamic object creation, enabling different software components to interact through well-defined interfaces. Here we deployed a Python environment on Windows 10 Operating System and accessed COM objects exposed by the Solartron APIs. These interfaces allow us to manage instrument connections and status events, execute experiments, and retrieve real-time measurement data directly.

Physically, communication between the workstation and the Solartron XM MTS is achieved via an RJ-45 Ethernet interface operating over TCP/IP. The instrument is visible on the local network and can be accessed through its assigned IP address.

This architecture enables real-time communication: the AI agent monitors the cycling data streams, makes decisions on the next charging action, and updates control commands accordingly. The streamline does not require proprietary software modification.

We have revised the Methods section to describe this architecture in more detail and to highlight the generalizability of this approach to other programmable battery testing systems with open API support.

“The control system was implemented using Component Object Model (COM) automation APIs, which expose programmable interfaces for accessing and managing hardware operations, including experiment execution, instrument status monitoring, and data retrieval. An established Python environment running was used to interact with these COM interfaces, enabling direct control from the workstation. Physically, the workstation is connected to the Solartron system through a standard RJ-45 Ethernet interface over TCP/IP. The instrument is visible on the local network and can be accessed through its assigned IP address.”

- Whether the feedback control was synchronous or batch-based, and how latency and reliability were evaluated and handled

Response: We thank the reviewer for this important question. Real-time experimental data are continuously streamed to the AI agent, which makes decisions based on the most recent 3-cycle data. The update frequency of the executed charging action is therefore every 3 cycles, forming the intelligent cycling framework.

Data acquisition is conducted via the same communication interface used by Solartron's commercial software, without introducing any additional layers or delay. The stability and availability of the interface are evidenced by its use in conventional cycling tests. On the other hand, each set of 3 cycles typically takes on the order of 10 minutes, while the AI agent's inference and command dispatch takes less than one second. This large difference in timescales ensures that the latency introduced by the control system is negligible.

The uninterrupted operation of the system and its stable performance demonstrate that latency and reliability are not limiting factors. We have added the relevant description to make this point clearer.

“The latency introduced by the closed loop is negligible relative to the cycling timescale. Reliability is also ensured by leveraging the same standard interface used in routine experiments, with no observable disruption or delay during long-term operation.”

- Is the AI module running on a separate workstation and received the Solartron data and then sends updates the control parameters? How?

Response: We thank the reviewer for this question. As previously detailed, the AI module is deployed on the Windows-based workstation that does not rely on the Solartron commercial control software. Our control system bypasses the vendor software entirely by using a Python-based environment that communicates directly with the hardware via COM automation APIs over a standard RJ-45 Ethernet connection using TCP/IP.

This design enables real-time bidirectional communication: experimental data is streamed directly from the instrument to the AI module, which then updates control parameters based on its decisions without the involvement of any proprietary vendor software. It ensures seamless integration with minimal latency, and offers a path toward future deployment in edge-intelligent battery management systems.

- How the system could be generalized to commercially relevant test stations (e.g., BioLogic, Maccor, Neware) or implemented in edge computing

Response: We appreciate the reviewer's insightful question. Traditionally, users rely on vendor-provided control software when operating commercial battery test stations, which often imposes constraints on control logic and automation.

We have actively communicated with several major vendors, including Arbin, BioLogic, and Neware, and confirmed that their modern systems support open communication protocols and API access. In fact, vendors have indicated that enhanced API accessibility is becoming a key competitive feature in the development of their instruments. Therefore, as long as the battery tester offers open communication interfaces, our framework can be readily deployed on either conventional computer workstations or embedded edge-computing devices.

We have added a statement in the revised manuscript to highlight this point.

“For broader applicability, commercial vendors generally confirmed that modern programmable battery testers support external integration via open interfaces. Therefore, the proposed platform is broadly transferable across different hardware systems.”

In addition, without a detailed system diagram (Fig. 1 is too general) or communication/control workflow, the platform's portability and reproducibility cannot be assessed. If the authors intend to present a novel AI-powered battery control system, this aspect must be central in the manuscript. Currently, the AI agent appears more like an external script that interfaces with a test station, rather than an integrated, deployable BMS architecture.

Response: We thank the reviewer for this important comment. We fully agree that the communication/control workflow is essential to assess the platform's portability and reproducibility. The original Fig. 1 was intended to serve as a high-level conceptual overview for general readability.

To address this concern, we have added a new Fig. 1b that explicitly illustrates the communication, data exchange, and control workflow between the AI agent and the battery testing hardware. Fig. 1b provide implementation-level clarity to complements the original schematic.

We sincerely appreciate this feedback and believe that the improved illustration strengthens the manuscript's contribution in terms of platform's portability and reproducibility.

Besides, our current work primarily serves as a proof-of-concept to demonstrate the feasibility and effectiveness of real-time AI integration into battery testing/controlling platforms. While it is not a full-featured deployable BMS at this stage, the proposed framework validates a scalable control mechanism that we envision being embedded into future edge-computing-based BMS hardware.

We have revised the manuscript text accordingly to clarify this positioning and avoid potential misinterpretation as a commercial-grade BMS product.

“As shown in the detailed workflow (Fig. 1b), the AI workstation directly communicates with the tester hardware via COM APIs over TCP/IP connection. Every 3-cycle data is employed to evaluate the battery state and adaptively control the charging strategy. This implementation supports real-time closed-loop control without relying on any proprietary vendor software.”

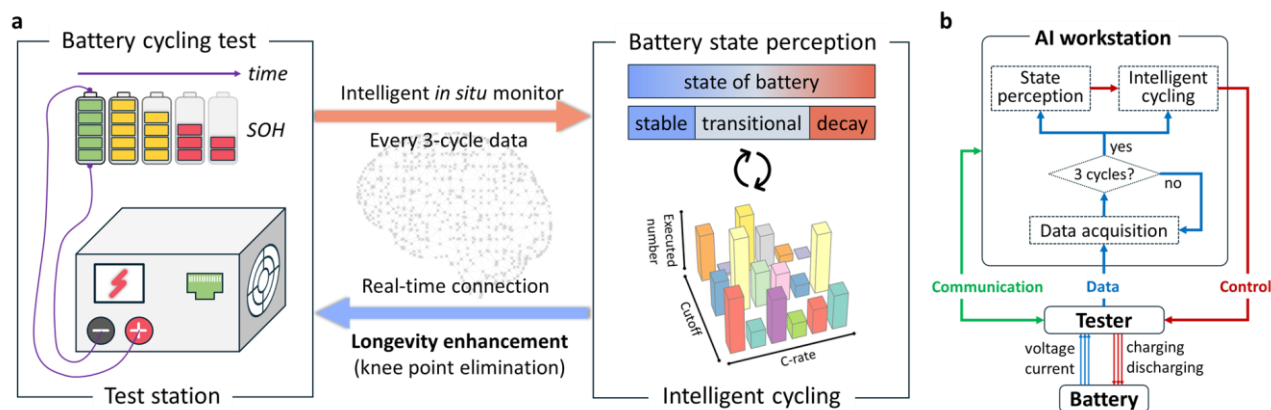


Fig. 1 | Real-time AI for intelligent cycling system of batteries. a, Schematic of the integrated platform. A trained state perception module is implemented based on the real-time cycling data from every 3 cycles, serving as an intelligent in-situ monitor to identify the state of the operating battery, including stable, transitional, and decay states. Reinforcement learning (RL) further enables intelligent cycling by adaptively optimizing fast charging strategies for solid-state batteries across cycles based on the battery state. An electrochemical test station is used to cycle actual batteries controlled by the above AI workstation. **b,** Engineering-oriented workflow showing the real-time communication, data and control loop. The AI workstation enables bidirectional communication with the tester hardware. Every 3-cycle data is continuously collected to evaluate the battery state, which is then used to adaptively control the charging strategy for the next segment. The goal of the current work is to demonstrate the effective longevity enhancement by intelligent cycling.

2. Data sufficiency and model generalizability

The developed AI models rely on a dataset of just only 28 solid-state cells, collected from historical lab experiments, to train the ResNet50-based state classifier. While the authors attempt to validate the classifier on independent cells, this dataset is extremely limited in terms of size, diversity, and representativeness—especially for training deep learning models and RL agents.

Response: We appreciate the reviewer’s comment on dataset size and generalizability. While the dataset is built from 28 solid-state cells, we would like to clarify that in our state perception module, the unit of training data is not the battery itself but the 3-cycle segmental data from each cell that can often run from $\sim 1,000$ to $\sim 10,000$ cycles. Each such segment produces a structured shape image input, and thus a total of 83,422 training samples were used. This dataset size is sufficient for the training and convergence of the current AI model, as supported by the validation and test performance.

Moreover, the long lifespan and fast-charging capability of our solid-state batteries enable efficient generation of large volumes of available data. This is a key motivation behind choosing this battery chemistry system for proof-of-concept demonstration. The 28 cells varied in electrolytes and cycling parameters, which intentionally capture realistic diversity in degradation behaviors within a controlled system.

These steps will be meaningful for moving from a working prototype toward a general-purpose intelligent battery management system. In future work, it can be planned to expand the dataset to include more battery systems to improve generalizability and robustness.

The relevant revision has been made in the updated manuscript to clarify the data structure used for model training.

“To construct the training samples, the battery itself is not treated as the data unit; instead, the model ingests abundant 3-cycle segmental data from each cell.”

“A total of 83,422 such samples were used for training.”

It is also unclear:

- How the dataset was split (train/validation/test)

Response: We thank the reviewer for raising this point regarding data splitting. In our study, we adopted a 16:6:6 split ratio for training, validation, and test sets, corresponding roughly to 60%, 20%, and 20% of the full 28 batteries. Importantly, the split was performed at the cell level, data from each battery only appears in one subset, thereby preventing data leakage and ensuring a fair evaluation to previously unseen cells. This splitting strategy adheres to commonly accepted practice.

The corresponding clarification has been added in the revised manuscript.

“The overall dataset was split into training, validation, and test sets at a 16:6:6 ratio, corresponding approximately to 60%, 20%, and 20% of the full 28 batteries.”

“Each battery's data was included exclusively in one subset to prevent data leakage and ensure generalization to unseen batteries.”

- Whether cells from similar configurations were grouped across splits, risking data leakage

Response: We appreciate the reviewer's concern regarding potential data leakage. To avoid such risks, our group deliberately masked the detailed configuration information of each cell during dataset preparation. The researchers conducting this study were thus blinded to the historical configuration specifics.

A corresponding clarification has been added to the revised manuscript.

“The detailed configurations of the batteries were masked during dataset preparation.”

- If any data augmentation or uncertainty quantification was applied to improve robustness

Response: We appreciate the reviewer's insightful question. In this study, we did not apply explicit data augmentation techniques. However, the data originated from real-world battery cycling experiments, which inherently include substantial variability such as measurement noise, voltage fluctuations, and differences in degradation rates. This natural diversity contributes to the robustness of the model. As noted in the manuscript, “with the augmentation of battery dataset in the future, more complex large models can also be employed to achieve generalization effects.”

The current ResNet50-based state classifier generates deterministic predictions. Although uncertainty quantification was not incorporated in this version, we acknowledge it is worth trying in the future for battery state estimation. In principle, methods such as Bayesian neural networks or ensemble learning are compatible with our method and can be employed to enable reliable uncertainty estimation and improve model interpretability.

Furthermore, all data appears to originate from lab made LMBs, which to this stage suffer from high variability and low reproducibility (as shown in Ext. Fig. 1). The use of such limited and variable data raises substantial questions about model generalizability, especially across different chemistries or battery formats.

Response: We appreciate the reviewer's professional comment. We choose solid-state batteries as we think its property is very suitable to a fast and reliable proof-of-concept demonstration of real-time AI research on batteries.

While Extended Data Fig. 1 shows certain degrees of variability, such variability serves as a valuable source of perturbations for enhancing model robustness. Our primary motivation for selecting solid-state LMBs lies in their long-term application potential and their ability to support high-rate charging, which significantly shortens the time required to generate large-scale cycling data. It's an important advantage for developing and evaluating AI-driven methods. Furthermore, all these batteries show similar three states of stable, transitional, and decay, as shown in Fig. 2.

To complete this study, we conducted multiple rounds of experiments with long-term cycling and statistical analysis, which required substantial time and experimental effort. Without solid-state LMBs, it would be difficult to manage the time cost associated with exploring real-time AI concept with the resource from a university lab.

Actually, we do not claim that the developed model is directly applicable to all battery chemistries or systems, although we believe the general framework and methodology are transferrable to other battery systems. Our study validates the feasibility of state perception and intelligent cycling for LMBs. Particularly, the knee point phenomenon is common across multiple battery systems, and their degradation pathways share certain universal patterns. Thus, we believe that the conceptual framework and AI workflow can be extended to broader battery systems in future work.

What remains strange to the reviewer is that, had the authors used commercial, off-the-shelf lithium-ion batteries—which are more reproducible and come in diverse chemistries—they could have better demonstrated the robustness and wide applicability of the AI platform. And saving time wouldn't be an issue, either (i.e., cycling at high temperatures, high rates, plating, etc.). This makes the case that solid-state LMBs were not necessary for this demonstration, and in fact, their use may have constrained both the model's performance and its relevance and add too much information not needed to present the platform.

Response: We thank the reviewer for this thoughtful comment. While commercial lithium-ion batteries offer high reproducibility and chemical diversity, their use in this specific study would not necessarily reduce the experimental time as suggested. In fact, our solid-state lithium-metal batteries support ultrafast cycling C-rates up to 20C [5], allowing a full lifetime test (from pristine state to below 80% SOH) within in a significantly shorter timeframe. Moreover, this approach preserves a large number of usable cycling data, making it particularly valuable for real-time AI control to extend battery lifespan

Due to safety constraints and performance limitations, commercial lithium-ion batteries typically cannot support such high-rate operation with sufficient cycles. Consequently, AI-driven experiments would require months or even years of continuous cycling, especially once the AI control begins to extend the cell's life. It is not practical for concept validation and model iteration.

Therefore, the choice of solid-state LMBs reflects a designed trade-off between experimental tractability and conceptual ambition. It not only facilitates timely and safe experimentation but also supports the feasibility demonstration of real-time AI control.

While we agree that commercial batteries represent an important direction, especially for broader deployment, such an extension is beyond the scope of this work and is planned for future studies. Specifically, the group has a long-term collaboration with LG Energy Solutions [36], and thus we understand that to work on commercial batteries, it would be a better idea to directly collaborate with battery industry, such as LG, to obtain big data from them for training, and implement the AI modulation approach to their BMS system. However, such work may be limited by certain regulations to publish.

A corresponding clarification has been added in the revised manuscript.

[5] Ye, Luhan, and Xin Li. "A dynamic stability design strategy for lithium metal solid state batteries." *Nature* 593.7858 (2021): 218-222.

[36] Chen, Xi, Jeesoon Choi, and Xin Li. "Machine Learning a Million Cycles as 2D Images from Practical Batteries for Electric Vehicle Applications." *ACS Energy Letters* 7.12 (2022): 4362-4367.

“As a proof-of-concept demonstration, we utilize the extremely fast charging capability of our solid-state batteries to 20 C-rate at low cathode loading [5], to greatly accelerate the data generation and reduce the feedback time in our development. This enabled rapid lifetime cycling and efficient model iteration. While commercial Li-ion batteries offer better reproducibility, their limited rate capability would substantially prolong the experimental timeline, especially under potential AI-extended lifetimes. However, implementing our approach to commercial Li-ion battery and BMS system will be an obvious next step through the collaboration with the battery industry.”

3. Title and scope

The manuscript title emphasizes “real-time AI-powered solid-state lithium metal batteries”, which is misleading. The innovation lies not in the materials science of solid-state batteries, but in the AI-based control methodology. Also, if the AI logic isn’t embedded but is a software agent controlling a lab instrument externally, then calling this “real-time AI-powered” control requires qualification. It’s more of a scripted closed-loop AI-supervised platform.

Similarly, the manuscript devotes extensive space to microstructural evolution, SEM/EDS imaging, and even modeling of interfacial degradation using a 2D grid simulation. While informative, these analyses appear secondary to the main contribution. The platform, which should be central, is lightly described, lacking architectural detail, block-level diagrams, or algorithm – hardware flowcharts. This mixing of material characterization with high-level systems and AI objectives creates conceptual fragmentation, detracting from the manuscript’s clarity and weakening the scientific message. If the core novelty is the intelligent cycling framework, then the focus must shift more decisively towards the architecture description, the experimental integration of control and validation under diverse scenarios and the comparison with other heuristic and conventional adaptive methods.

Response: We sincerely thank the reviewer for the thoughtful feedback. We regret that our original presentation may have conveyed a sense of conceptual ambiguity. We have revised the manuscript title and clarified related expressions throughout the text. The updated title is “Real-time AI for Solid-State Lithium Metal Batteries”

Regarding the phrase “real-time AI-powered”, we agree that clarification is warranted. Although our AI module is not embedded within the battery hardware, it operates in tightly synchronized communication with the experimental instruments with minimal latency. The real-time interaction is essential for the system to perceive and control during cycling. We have added a new workflow diagram (Fig. 1b) illustrating the communication, data, and control loops. Also, we have revised the main text to more prominently highlight the design and realization of the overall platform.

We acknowledge that materials characterization and interfacial modeling are described specifically, but they are not intended as parallel contributions. Other reviewers also expressed interest in and asked more questions about the materials characterization presented in our work, which aligns with our original intent. While intelligent cycling is the central contribution, we believe that the scientific validity of its outcomes must be grounded in physical interpretation. Otherwise, the real-time AI control would risk becoming a “black box optimization”.

When AI proves effective in scientific research, it is often because it captures or leverages underlying physical mechanisms that conventional strategies fail to address. As such, we believe it is our responsibility to investigate and discuss those mechanisms underpinning the AI actions. The materials analyses in our work are therefore not auxiliary, but essential for providing interpretability and theoretical grounding. We consider this coupling between AI-driven charging strategies and material-level insight to be a key scientific contribution of this study.

In total, this manuscript aims to introduce a novel methodology that combines data-driven state classification with reinforcement learning for adaptive cycling, which is a novel and attracting idea. However, the work as presented lacks critical validation, experimental detail, and architectural clarity to support the strength of its claims.

Response: We thank the reviewer for the overall assessment and constructive criticisms. We have carefully addressed each specific comment raised above. For the main issues summarized below, we would also like to provide the following point-by-point response.

The main issues are:

- The integration of AI into battery control is more conceptual than demonstrable at this point

Response: We are sorry for the misunderstanding caused by our previous version of the manuscript. In this proof-of-concept study, we choose solid-state LMBs as a validation platform to demonstrate the real-time AI potential over complex systems. The experiments are already conducted as a full-process, closed-loop control, rather than just simulations or isolated module tests. This represents an important step toward adaptive, AI-driven battery management. While further engineering efforts are certainly needed, this does not diminish the scientific innovation of the current work.

According to the reviewer's professional suggestions, we have revised the manuscript to more clearly present the positioning of this work with experimental grounding and forward-looking value.

- The limited dataset and battery format restricts the generalizability of the conclusions

Response: We appreciate the reviewer's comment on dataset size and generalizability. As clarified in a previous response, while the dataset originates from 28 batteries, the training unit is not full battery lifetimes but every 3-cycle data, each converted into structured shape images. This yielded 83,422 samples, which are sufficient for model training through validation and test performance.

Moreover, the long lifespan and ultrafast charging capability (up to 20C) of the selected solid-state batteries enabled efficient generation of cycling data. This was a key motivation for choosing this battery system as a testing ground for our proof-of-concept study.

We emphasize that the current work aims to demonstrate the feasibility of real-time AI-based control in complex battery systems, rather than claiming universal generalization across all chemistries or commercial formats. That said, the knee point behavior and degradation patterns observed are common across battery chemistries, and the overall framework can be extended in future studies to other battery formats, including the collaboration with battery industry for commercial batteries. We have revised the manuscript to clarify this issue.

- The overemphasis on solid-state lithium metal batteries is misaligned with the core innovation, which is platform-level

Response: We thank the reviewer for this important observation. The solid-state lithium metal batteries were selected as a challenging but efficient platform due to its ultrafast cycling capability, which significantly reduces experimental time and enables rapid model iteration.

While we acknowledge that solid-state battery chemistry is not the focus of this study, material-level characterization was included to provide physical interpretation and avoid black-box optimization. These analyses support scientific soundness of the AI decisions. We have revised the manuscript to clarify this distinction.

- The system engineering aspects, particularly the AI-hardware interface, are under-described and difficult to reproduce

Response: We appreciate the reviewer's concern. As clarified in our previous responses, the AI-hardware interface is fully implemented using standard communication protocols available on modern programmable battery testers, enabling real-time data and control loops with minimal latency. Although the AI workstation is not embedded, it operates in a closed-loop manner with the instrument and can be reproduced using commercially accessible equipment and open APIs. A workflow diagram (Fig. 1b) and these implementation details have been added in the revised manuscript to improve reproducibility and transparency of the system design.

which in reviewers' opinion weakens the case for publication in Nature Communications on its current form.

Response: We acknowledge the reviewer's assessment and have carefully addressed each of the concerns in our revision. We hope that the revised manuscript is substantially strengthened and more aligned with the journal's expectations.

(Remarks on code availability):

The computer code generated and used during this study would be available from the corresponding author on reasonable request.

Response: We thank the reviewer for pointing this out. In fact, we had uploaded the source code in the public repository in our initial submission to the journal: <https://github.com/wmy1994/Real-time-AI-powered-solid-state-batteries>

Notably, we would like to clarify that the full closed-loop operation requires integration with specific battery testing instruments via their communication interfaces. Therefore, while the core code is available, executing the complete system workflow requires compatible hardware.

Reviewer #3:

This work presents a novel approach leveraging AI and reinforcement learning to both estimate the state of solid-state lithium metal batteries and to adjust their cycling protocols to delay accelerated degradation and extend energy throughput. This work appears to represent a considerable contribution to the space of battery diagnostics and operation, however, presentation issues mean that the work should not be published in its current state. Specifically, the language and figures must be improved prior to publication.

Response: We thank the reviewer for the positive assessment of the technical contribution. We fully acknowledge the concern regarding language clarity and figure quality, and we have undertaken a thorough revision of the manuscript to address these comments.

The following comments are presented in no particular order:

The readability of the article is considerably degraded by the clarity of the language and grammar. There are many tools and resources available to refine the language. I encourage the authors to leverage them.

Response: We thank the reviewer for the helpful suggestion. We have carefully revised the manuscript and improved the language throughout to enhance clarity and readability.

For the title, I would suggest instead, "Real-time AI for solid-state lithium metal batteries." The current title seems to indicate that the batteries are AI powered in real time, which doesn't make a lot of sense.

Response: Based on your suggestion, we have revised the title to: “Real-time AI for solid-state lithium metal batteries”.

In the first line of the abstract, "battery cycling lifetime" should be replaced by "battery degradation" or "battery health evolution"

Response: Thank you for the helpful suggestion. We have revised the corresponding description in the abstract to improve clarity.

Figure 1 is difficult to interpret. Especially the battery images on the top left of the figure that seem to indicate the battery state of health dramatically improving and then degrading in time, a characteristic not seen in real cells.

Response: We appreciate the reviewer’s helpful observation. In response, we have revised the top left of the figure to reflect a gradual SOH decline, and added textual SOH labels to clarify the misunderstanding.

Additionally, we have included a new figure (Fig. 1b) to illustrate the workflow of communication, data, and control loop of our AI platform, thereby improving the technical clarity of the system architecture.

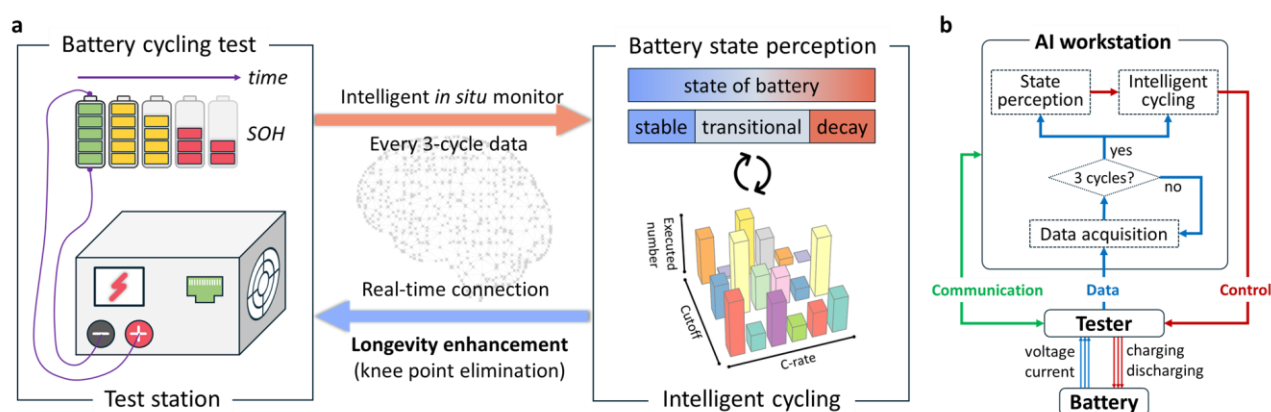


Fig. 1 | Real-time AI for intelligent cycling system of batteries. a, Schematic of the integrated platform. A trained state perception module is implemented based on the real-time cycling data from every 3 cycles, serving as an intelligent in-situ monitor to identify the state of the operating battery, including stable, transitional, and decay states. Reinforcement learning (RL) further enables intelligent cycling by adaptively optimizing fast charging strategies for solid-state batteries across cycles based on the battery state. An electrochemical test station is used to cycle actual batteries controlled by the above AI workstation. **b,** Engineering-oriented workflow showing the real-time communication, data and control loop. The AI workstation enables bidirectional communication with the tester hardware. Every 3-cycle data is continuously collected to evaluate the battery state, which is then used to adaptively control the charging strategy for the next segment. The goal of the current work is to demonstrate the effective longevity enhancement by intelligent cycling.

Figure 2 indicates that the normalized capacity in the transitional state ranges between 0.9 and 0.5 SOH. However, in industrial settings, 0.8 is typically selected as the threshold for end of life. Why is this transitional period defined to cross such a dramatic change in SOH? Intuitively, once the battery exhibits roll-over, you are no longer in a transitional period, you are deep into decay.

Response: We appreciate the reviewer’s insightful comment. From a conventional battery engineering standpoint, an SOH of 0.8 is commonly used in industry as a threshold to mark end-of-life. While in this study, the definition of the “state segmentation” is designed to capture and dissect the knee point and its surrounding evolution.

Following the onset of the knee point, the battery typically undergoes a rapid degradation. This sharp change in capacity marks a transitional behavior, which we aim to characterize in detail. We view this region as a temporary regime where internal failure and side reactions are actively progressing but have not yet stabilized. Only after these sharp changes subside does the battery enter the final “decay state”, characterized by slower degradation. Therefore, our segmentation is not based solely on SOH values, but rather on the first and second derivatives of capacity evolution, reflecting changes in the underlying physical mechanisms.

We have added clarifying text in the revised manuscript to avoid potential confusion.

“While 0.8 SOH is commonly used in industrial settings as the end-of-life (EOL) threshold, the definition of state segmentation is to isolate and examine the rapid degradation behavior around the knee point. The transitional state represents a temporary yet critical period where internal failures and side reactions accelerate sharply but have not yet stabilized. That means a battery in the transitional state can still go back to the stable state through intelligent cycling (e.g., Fig. 2d, 2e). Only after this sharp degradation phase does the battery enter the final decay state, characterized by a slower rate of capacity loss.”

The interpretation of Fig 4a is unclear. For example, upwards of cycle 2000, it appears that the state perception is 1 across a large number of action indices simultaneously. I suggest that the figure be explained in greater detail, or another approach is used to represent the concept.

Response: We appreciate the reviewer's valuable observation and suggestion. In Fig. 4a, the "action index" refers to different choices of charging actions. The plotted state perception output reflects the battery state under the specific action applied during cycling (0 = stable, 1 = transitional, 2 = decay).

Intelligent cycling significantly prolongs battery lifetime, extending the stable state well beyond what can be achievable under benchmark testing. However, the battery still eventually reaches a limit where degradation becomes unavoidable. Beyond approximately cycle 2000, the state perception output becomes state 1 across almost all action indices, indicating that the battery has thoroughly entered the transitional state regardless of which action is applied. In other words, many of the explored charging actions cannot maintain the battery in a stable state anymore, reflecting a transition into irreversible degradation. It marks the limit of intelligent cycling's ability to recover the battery to a stable state.

We have clarified this point in the main text to avoid any potential misunderstanding.

“As cycling progresses, the battery inevitably approaches a point where degradation becomes irreversible. Around cycle 2000, the state perception output shifts to 1 across most action indices, indicating that the available charging actions to preserve the battery in a stable state are limited. This simultaneous onset of transitional states across large number of actions signals the transition into irreversible degradation. This behavior highlights the ultimate constraint of the electrochemical system, reflecting the limit of intelligent cycling's ability to recover the battery to a stable state.”

In all figures and in the text, Crate should be changed to C-rate

Response: We thank the reviewer for the careful attention to detail. We have corrected all instances in the figures and text.

Figure 4 plots the "Euclidean distance" for the intelligent and benchmark case, but it is unclear what this distance is measured to and from.

Response: Thank you for raising this important point. Here, Euclidean distance represents the distance between two consecutive data points in the PCA space, where each point corresponds to a step composed of three consecutive cycles. This distance quantifies the magnitude of change in the battery's state from one step to the next.

As also suggested by Reviewer #1, we have revised the manuscript to explicitly clarify this definition.

“Euclidean distances calculated between adjacent cycles in the PCA space quantify the state transitions induced by each action step composed of three consecutive cycles.”

It is unsurprising that the AI input features are scattered in input space for the intelligence case versus the benchmark due to the constantly changing charging protocols. This does not necessarily indicate anything about the state of health as these changes in cycling protocol are not deconvolved. I am not convinced of the value of this PCA analysis.

Response: We thank the reviewer for raising this professional concern. We agree that the scatter observed in PCA space under the intelligent cycling scenario is partly influenced by the variability of charging protocols, and thus does not directly indicate battery state-of-health (SOH). However, that was not the main point we wanted to argue.

In our work, the purpose of PCA is to track the evolution of state-related features as influenced by the applied charging actions. PCA processing was performed on shape images derived from charging-discharging profiles. These shape features have been previously validated to contain rich information correlated with battery

degradation [36]. Therefore, while PCA does not explicitly deconvolve the effects of varying charging protocols, it provides a low-dimensional visualization of how the battery state presentations evolve under intelligent cycling, which is hidden in shape images.

The observation that intelligent cycling induces a more exploratory and reversible trajectory in the PCA space potentially reflects how the changing charging protocols modulates the battery's accessible state pathways. In contrast, the benchmark exhibits a narrow, monotonic progression, corresponding to irreversible degradation.

While the influence of varying protocols is a valid confounding factor, the trajectories in PCA space are not interpreted in isolation. The consistency of PCA-based observations with other independent characterizations strengthens the value of this PCA analysis. Namely, SEM characterizations and 2D grid simulations both reveal similar patterns of state divergence, which correlate with the PCA-based depiction. These cross-validated insights suggest that the PCA analysis offers indirect but meaningful support for understanding how intelligent cycling interacts with battery degradation dynamics.

In response to the reviewer's comment, we have revised the manuscript to explicitly acknowledge the potential confounding effects of charging protocol variations and to better frame this PCA analysis as an auxiliary, visualization-driven tool, rather than a standalone indicator of SOH.

[36] Chen, Xi, Jeesoon Choi, and Xin Li. "Machine Learning a Million Cycles as 2D Images from Practical Batteries for Electric Vehicle Applications." *ACS Energy Letters* 7.12 (2022): 4362-4367.

“The variability of charging actions in intelligent cycling inevitably introduces some confounding effects into the PCA results. Therefore, the PCA analysis should not be regarded as a direct indicator of state-of-health, but rather as a visualization tool to trace the evolution of battery state features.”

“This suggests the intelligent cycling of dynamic strategy enables a broader and more flexible exploration of electrochemical states, allowing the battery to jump between states of little path dependence and access the principal component space that otherwise will not be accessible in a normal cycling of fixed strategy and unidirectional trajectory evolution in the principal component space of strong path dependence. Euclidean distances calculated between adjacent cycles in the PCA space quantify the state transitions induced by each action step (Fig. 4c). For intelligent cycling, these distances reveal more frequent and larger shifts between consecutive steps, reflecting rapid and reversible transitions between stable and transitional states, which effectively elongates the trajectory of the state evolution in the PCA space and slows down the degradation. In comparison, benchmark cycling exhibits narrow, unidirectional state trajectory (Fig. 4e, Supplementary Fig. 6 and animations) and minimal Euclidean displacement (Fig. 4f), corresponding to irreversible fast degradation and early emergence of knee points. The limited state variability under fixed charging actions restricts the ability to recover once degradation accelerates.”

The analogy between the battery and neural networks is suspect. I suggest removing it as it does not help explain the phenomena nor is it well supported by evidence.

Response: We agree with the reviewer's suggestion. We have removed the analogy between the battery and neural networks from the manuscript. This revision does not affect the core methodology or conclusions of the work.

Should the data and code not be made publicly and easily accessible, the authors should make a reasonable justification. Not making these accessible significantly decreases the impact on the community and the chance of reproducing these results.

Response: Thank you for your suggestion. The source code has been made publicly available on GitHub in our initial submission.

Regarding the dataset, we regret that it cannot be released publicly at this stage due to institutional restrictions associated with ongoing research in solid-state batteries and the related commercialization activity. However, we welcome collaboration and data-sharing inquiries. Access can be granted to qualified researchers upon reasonable request to the corresponding author.

(Remarks on code availability):

code was not available

Response: We thank the reviewer for pointing this out. We had uploaded the source code in the public repository in our initial submission a few months ago in fact at: <https://github.com/wmy1994/Real-time-AI-powered-solid-state-batteries>

Notably, we would like to clarify that the full closed-loop operation requires integration with specific battery testing instruments via their communication interfaces. Therefore, while the core code is available, executing the complete system workflow requires compatible hardware.

RESPONSE TO REVIEWERS.

Reviewer #3:

(Remarks to the Author):

I thank the authors for properly addressing most of my presentation and technical concerns.

I would encourage the authors to improve the grammar throughout the article. For example, the first line of the introduction should be "The Li-ion battery is one of the most powerful technologies in our era for decarbonization and combating climate change" instead of "Li-ion battery is one of the most powerful technologies in our era for decarbonization and combat climate change." Please either leverage a native English speaker at one of your institutions, or even ask AI to review the document and provide suggestions on grammatical changes with the reasoning for these changes.

Response: We thank the reviewer for highlighting the need for improved grammar and for providing specific examples. In response, the text is revised for grammatical accuracy and clarity.

(Remarks on code availability):

I did not run and install the code, as this would require the requisite dataset and hardware. The necessary code appears to be present with comments. The codebase would be of much greater value to the research community if it came with a detailed demonstration notebook with associated datasets and instrument/battery simulators.

Response: We thank the reviewer for this valuable suggestion. Actually, the released codebase already includes a virtual instrument interface, which can be directly executed via `Exp_Pyctrl_solartron/Exp_ctrl.py` script to launch a simulated communication environment. We have updated the repository README to explicitly describe this functionality at <https://github.com/wmy1994/Real-time-AI-for-solid-state-batteries>. However, it will require the future readers to download our code, modify and connect to their own hardware, including their own running batteries and dataset, to run our AI system.

Regarding the dataset that we used for training, we regret that it cannot be released publicly due to various restrictions. However, we welcome collaboration and data-sharing inquiries, and access can be granted to qualified researchers upon reasonable request to the corresponding author.

These clarifications have been added to the “Code availability” and “Data availability” sections of the revised manuscript.