

Multiscale Digital-Twin Modeling of Energy-Storage Materials by Integrating First-Principles Calculations and Machine-Learning Interatomic Potentials

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Abstract

Conventional modeling of energy-storage materials is constrained by the computational cost of first-principles calculations, limited electrochemical-state awareness in machine-learning interatomic potentials, and weak coupling between materials-scale models and online battery decision-making. This study proposes a first-principles-constrained electrochemically conditioned machine-learning interatomic potential (FP-EMLP) integrated with a multiscale digital-twin framework (FP-EMLP-DT). A multistate configuration space spanning ion concentration, temperature, defects, strain, and phase boundaries is combined with active learning based on committee uncertainty, structural novelty, and physics-based risk. The equivariant graph model incorporates electrochemical-state gating, charge-conserving projection, and differentiable long-range electrostatics, and is trained using energies, forces, stresses, equilibrium voltages, and migration barriers. Predicted diffusion coefficients, free energies, elastic tensors, and chemical expansion coefficients are transferred to an electrochemical-mechanical-thermal-damage model. The resulting model is further integrated with a physics-based reduced-order network, uncertainty-gated residual learning, online data assimilation, and degradation-aware control. FP-EMLP achieves mean absolute errors of 6.5 meV atom⁻¹, 33 meV Å⁻¹, 0.051 V, and 0.038 eV for energy, force, voltage, and migration barrier, respectively, with a 4.8% aggregate relative error across multiscale fields. Active learning reduces required first-principles labels by 33.3% while covering 91.2% of high-risk configurations. FP-EMLP-DT achieves mean absolute errors of 0.93% for state of charge and 1.21% for state of health. Degradation-aware control reduces maximum material stress and damage increment by 16.5% and 36.8%, respectively, demonstrating an effective multiscale pathway from electronic structure to online battery management.

Keywords: first-principles calculations, machine-learning interatomic potential, equivariant graph neural network, active learning, multiscale digital twin, energy-storage materials

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1. Introduction

The rapid growth of grid-connected renewable energy, electrified transportation, and distributed energy storage is transforming battery management from isolated condition

monitoring toward lifecycle prediction and control. The concept of digital twins ties together physical batteries, virtual models and real-time data, offering them a common platform in terms of state estimation, fault diagnosis and operational optimization. Semeraro et al. structured a

modeling workflow of lithium-ion battery digital twins based on a reference architecture and identified coordination between model, data, and service layers as a requirement of predictive management [1]. Issa et al. have created a vehicle-battery twin based on the Industrial Internet of Things and data-driven models, demonstrating that dynamic operating information can improve state-of-charge estimation [2]. State-of-health features of virtual cycling experiments can be derived by analyzing unstructured operation data [3], and data pre-processing combined with smart estimators further improves degradation-state recognition [4]. Aging-induced model drift is reduced through adaptive parameter updates [5], while mechanistic models enhanced by neural-network residuals balance stability and dynamic accuracy [6]. Physics-informed neural networks have demonstrated potential in operating condition modeling and real-time degradation prediction has been implemented using attention-based temporal networks [7]. Hybrid digital twins enable simultaneous estimation of voltage and state of charge [8,9] and have been expanded to energy-storage microgrid management [10], degradation-cost optimization [11], and fast-charging control [12]. Nevertheless, such approaches are mostly based on the use of terminal voltage, current, and temperature, and do not explicitly resolve ion diffusion, phase transformation, or stress-induced damage processes. These recent battery digital twins strengthen system-level state estimation and control, but they generally do not propagate atomistically resolved transport and degradation information into the online control loop.

Machine-learning interatomic potentials provide a feasible bridge between quantum-scale calculations and long-timescale materials simulation. Crystal Hamiltonian Graph Neural Network (CHGNet), a charge-informed graph model, was developed by Deng et al. and it was demonstrated that charge-informed representations improve transferability across ionic potential-energy surfaces [13]. Graph neural networks were scaled and applied to materials learning by Merchant et al., who observed that the performance of unseen structures is enhanced when data and model size are increased [14]. The stability and computational efficiency of neural force fields have been enhanced by the Equivariant Transformers, and pretraining attention-based deep potentials have made the transferability of materials stronger [15]. Higher-order equivariant message passing was used by Batatia et al [16]. to create an atomic foundation model, and it was shown that a universal pretrained potential could be used as a basis of simulations involving different classes of materials [17]. Limited targeted data may be used to enhance task specificity accuracy using fine-tuning with frozen parameters [18], and lower-cost atomistic simulation can be achieved through lightweight universal potentials [19]. Nonetheless, universal potentials continue to be unreliable with regard to phonons, surfaces and out-of-equilibrium configurations [20]. The explicit approach of long-range interactions has then emerged as a significant way to increase reliability in polar and ionic systems [21]. Collectively, these studies improve equivariance, pretraining, charge awareness, and long-range interactions, but their principal outputs

remain atomistic energies, forces, and derived material properties rather than online battery-control states.

Specialized machine-learning interatomic potentials have been applied to energy-storage materials in solid electrolytes, interfacial reactions, and ion transport [22]. Molecular dynamics simulations with long timescales can reveal structural reorganizations at lithium metal interfaces [23]. Chen et al. demonstrated that moderate anion disorder is conducive to connected Li-ion diffusion pathways when a machine-learning potential is used with Li₆PS₅Cl [24]. High-value electrolyte or material candidates could be prioritized by active learning with model feedback [25]. Sampling uncertainty minimizes the unnecessary labeling of almost-equilibrium states [26] whereas a heterogeneous ensemble enhances out-of-distribution state detection [27]. Interfacial defects and structural disorder of solid-state batteries have also been studied using machine-learning interatomic potentials [28]. The latest findings indicate that artificial intelligence approaches are currently used in materials screening, property prediction and transport analysis [29]. Nevertheless, an integrated mechanism to transfer quantum-scale results to mesoscale degradation models and system-level control has not yet been established [30]. Thus, prior studies have separately advanced atomistic material modeling and battery digital twins, whereas the cross-scale transfer of learned material parameters into degradation-aware online control is the specific gap addressed here.

Driven by these gaps, the present work proposes a first-principles-constrained electrochemically conditioned machine-learning interatomic potential (FP-EMLP) integrated into a multiscale digital-twin framework (FP-EMLP-DT). The training space is built with multistate first-principles data and active learning. Charge-conserving projection and multistate first-principles data with active learning are used in the construction of the training space. Electrochemical-state gating, charge-conserving projection, and long-range electrostatics branch are added to the equivariant graph network. The obtained free energies, diffusion coefficients, elastic tensor, and chemical expansion coefficients are then transmitted to an electrochemical-mechanical-thermal-damage coupled model. The physics-based model reduction, uncertainty-gated residual learning, online data assimilation, and degradation-aware model predictive control are subsequently combined to restore internal material states, estimate state of charge and state of health, and allow smart control of charging-discharging.

2. Multiscale Digital-Twin Model Integrating First-Principles Calculations and Machine-Learning Interatomic Potentials

In order to overcome the mismatch among computational scales, updates of state, and system-level control in the traditional models of energy-storage materials, this paper introduces FP-EMLP-DT, which is the first-principles-constrained multiscale digital-twin framework that uses electrochemically conditioned equivariant machine-learning

interatomic potential. The first-principles provide physical reference, whereas electrochemically conditioned equivariant potential acts as an atomistic-scale accelerator. The data transfer between the scales is implemented by means of a combined chemo-mechanical-thermal model, and the energy-storage system digital twin is dynamically updated using online data assimilation and model predictive control. In addition to material energies and atomic force predictions, the framework maps ion-migration barriers, diffusion

coefficients, equilibrium voltages, elastic properties, and damage evolution to the state space of the energy-storage system. Thus, it creates a direct linkage between the microscopic properties of the material and the macroscopic working conditions of the energy-storage device.

2.1 Multistate First-Principles Data Generation and Active Sampling

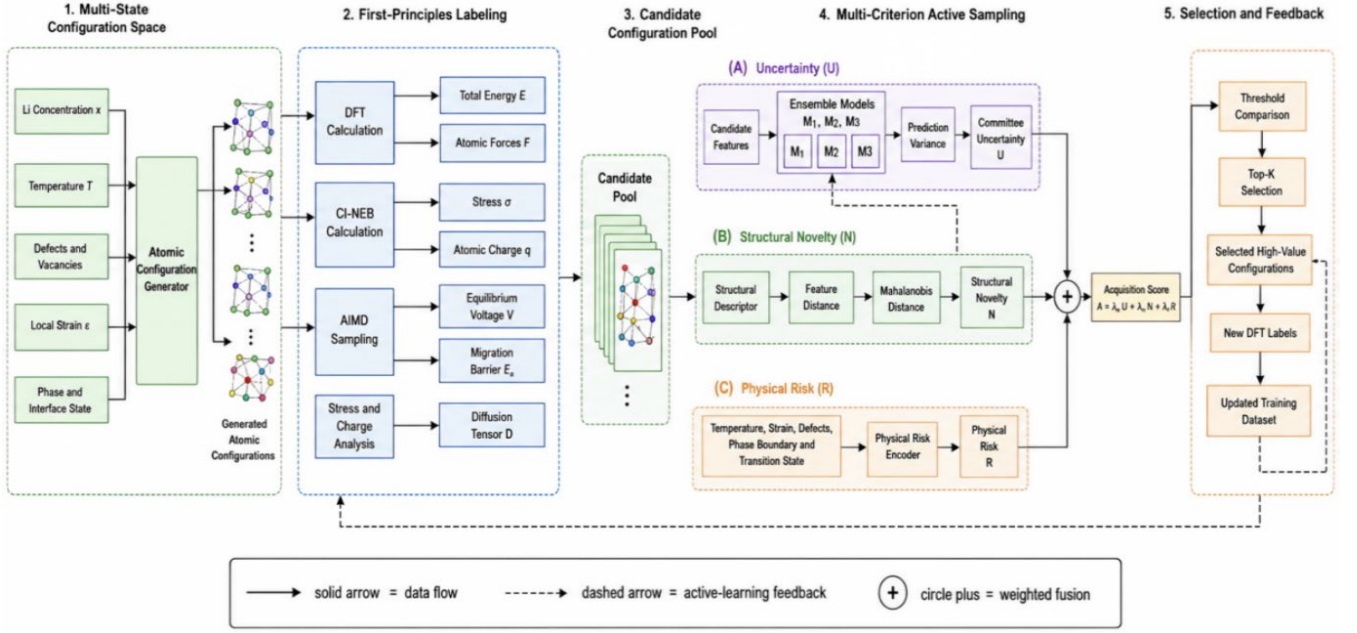


Figure 1. Multi-state data generation and active sampling framework

As illustrated in Fig. 1, the first step is constructing a first-principles configuration dataset of the energy-storage material at different ion concentrations, temperatures, defects, strains, and phases [31]. The atomic structure of the material system is given by:

$$R = \{r_i, Z_i\}_{i=1}^N \quad (1)$$

where r_i and Z represent the spatial coordinate and chemical species of the i -th atom respectively and N is the total number of atoms.

The ground-state total energy of the system at a given atomic configuration is written in density functional theory (DFT) as:

$$E_{\text{DFT}}[n, R] = T_s[n] + E_H[n] + E_{\text{xc}}[n] + \int v_{\text{ext}}(r)n(r)dr + E_{\text{ion}}(R) \quad (2)$$

where $n(r)$ is the electron density, $T_s[n]$ is the kinetic energy of noninteracting electrons, $E_H[n]$ is the Hartree energy, $E_{\text{xc}}[n]$ is the exchange-correlation energy, $v_{\text{ext}}(r)$ is the external potential produced by the ions, and $E_{\text{ion}}(R)$ is the ion-ion interaction energy.

Since energy-storing materials work at some fixed temperatures and during active charge-discharge processes, the sum of energies at zero temperature does not define their

thermodynamic properties completely. The Gibbs free energy at a finite temperature is then given by:

$$G(x, T, \epsilon) = E_{\text{DFT}}(x, \epsilon) + F_{\text{vib}}(x, T) - TS_{\text{conf}}(x) + pV \quad (3)$$

with x being the stoichiometric ratio of the working ion in the host material, T being the temperature, ϵ being the strain tensor, F_{vib} being the vibrational free energy, and S_{conf} the configurational entropy.

Given a working ion M and a host material H , the formation free energy of the M_xH intercalated structure is defined as follows:

$$\Delta G_f(x, T) = G(M_xH, T) - G(H, T) - x\mu_M(T) \quad (4)$$

where μ_M is the chemical potential of the working ion in its reference phase. Equation (4) will be employed to determine the thermodynamic stability of the material under various ion concentrations and to supply the necessary parameters that will be used in the phase-field free-energy function which will be introduced later.

When the ion concentration changes from x_1 to x_2 , the average equilibrium voltage becomes:

$$\bar{V}(x_1, x_2) = - \frac{G(M_{x_2}H) - G(M_{x_1}H) - (x_2 - x_1)\mu_M}{zF(x_2 - x_1)} \quad (5)$$

here, z is the charge number of the working ion and F is the Faraday constant. The equilibrium voltage of intercalation-type electrode materials may be calculated based on differences in first-principles formation energies or free energies. The connection is well known in computational research on energy-storage materials.

The climbing-image nudged elastic band (NEB) method is used to calculate the minimum-energy path in order to describe how ions migrate through a crystal lattice or defect-

mediated channels. The barrier of ion migration can be described in the form below:

$$E_m = G_{TS} - G_{IS} \quad (6)$$

where G_{TS} and G_{IS} represent the free energies of the transition state and the initial stable state respectively.

Once a sufficiently long atomistic trajectory has been generated, the ionic diffusion tensor is computed using the mean-square displacement:

$$D_{\alpha\beta}(x, T) = \lim_{t \rightarrow \infty} \frac{1}{2N_M t} \sum_{i=1}^{N_M} [r_{i,\alpha}(t) - r_{i,\alpha}(0)][r_{i,\beta}(t) - r_{i,\beta}(0)] \quad (7)$$

where N_M is the number of mobile ions, and α and β are spatial directions. In contrast to a scalar diffusion coefficient, Eq. (7) describes direction-dependent ion transport of layered, tunnel-type and anisotropic energy-storing materials.

Since a thorough treatment of the potential energy surface will entail a huge amount of first-principles calculations, it can be said that an active-sampling acquisition function is built by merging predictive uncertainty with structural novelty and physically high-risk states [32]:

$$A(R) = \omega_E s_E^2(R) + \frac{\omega_F}{N} \sum_{i=1}^N \text{tr} [\Sigma_{F_i}(R)] + \omega_D d_{Mah}[\chi(R), D_{train}] + \omega_P P(R) \quad (8)$$

Where s_E^2 is the energy prediction variance between machine-learning interatomic potentials in an ensemble, Σ_{F_i} is the predictive covariance of the force experienced by atom i [33], χ is the structural descriptor, and d_{Mah} is the Mahalanobis distance between the present structure and the training set [34]. $P(R)$ is the physics-based risk score assigned to configurations with phase transition, defect formation, large strain, or high temperature.

Before weighted aggregation, the four components in Eq. (8) are normalized to comparable scales to prevent their numerical magnitudes from dominating the acquisition score. The coefficients ω_E , ω_F , ω_D , and ω_P control the relative contributions of energy uncertainty, force uncertainty, structural novelty, and physics-based risk, respectively, with $\omega_E + \omega_F + \omega_D + \omega_P = 1$. Their values were selected by a coarse sensitivity analysis around the baseline setting rather than being treated as independent physical constants. The selection criterion jointly considers prediction accuracy, coverage of high-risk nonequilibrium configurations, and the reduction in first-principles labeling workload.

In case $A(R)$ is above the defined value, the first-principles calculation is triggered by the digital twin to derive the energy, atomic forces, stress, and charge response of the new configuration. The labeled configuration is subsequently included in the training set of the machine-learning interatomic potential [35]. Such a mechanism ensures that the

training data does not become overly focused on equilibrium structures [36].

2.2 Electrochemical-State-Conditioned Equivariant Machine-Learning Interatomic Potential

Neural Equivariant Interatomic Potential (NequIP) and Message Passing Atomic Cluster Expansion (MACE) models have demonstrated that the explicit incorporation of symmetry constraints related to spatial rotations, translations and reflections into graph neural networks enhances the data efficiency of interatomic potentials and the precision of force prediction. Based on higher dimensional equivariant message passing, the proposed model also includes electrochemical state-gating and polarizable long-range electrostatics branch which can be used to model ionic energy storage materials. Model architecture is shown in Fig. 2. In the baseline formulations considered here, NequIP and MACE emphasize $E(3)$ -equivariant local message passing, whereas CHGNet additionally uses charge-informed representations; none combines the global electrochemical-state vector of Eq. (13), the explicit charge-conservation projection of Eq. (15), and the equilibrium-voltage/migration-barrier auxiliary supervision of Eq. (18) in a single model.

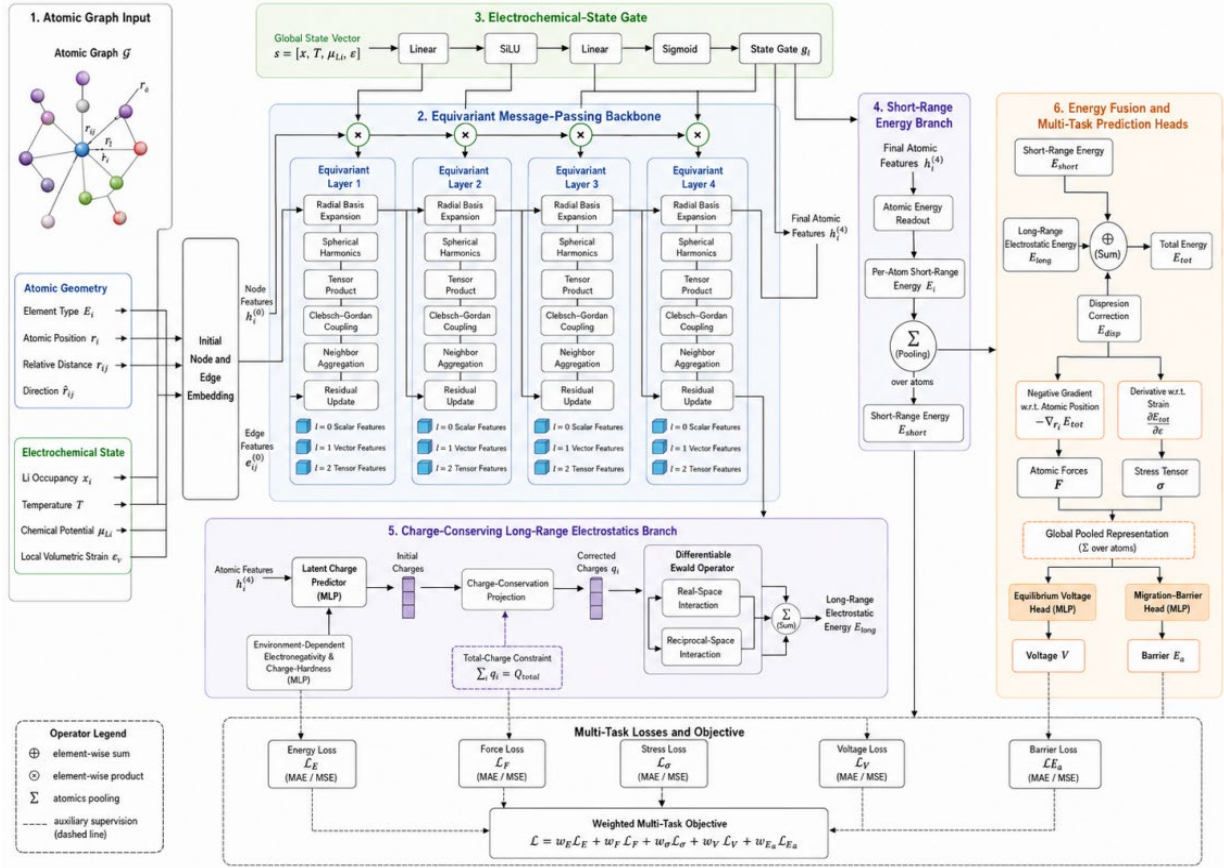


Figure 2. Architecture of FP-EMLP

The model-predicted total energy is made up of short-range local energy, long-range electrostatic energy, and a dispersion correction:

$$E_{\theta} = \sum_{i=1}^N E_{i,\theta}^{sr} + E_{\theta}^{lr} + E_{\theta}^{disp} \quad (9)$$

where $E_{i,\theta}^{sr}$ is the energy of short-range atom estimated by the equivariant graph neural network, E_{θ}^{lr} is the energy of long-range electrostatic interaction, and E_{θ}^{disp} is the dispersion correction.

Traditional machine-learning interatomic potentials normally create initial features based on elemental species and local atomic geometry only. Each of the atoms in the proposed model has all four variables, such as ion concentration, temperature, chemical potential and local strain, simultaneously incorporated into an atomic node:

$$h_i^{(0)} = \text{Emb}(Z_i) \oplus \phi_c(c_i) \oplus \phi_T(T) \oplus \phi_{\mu}(\mu_M) \oplus \phi_{\epsilon}(\epsilon_i) \quad (10)$$

where $\text{Emb}(Z_i)$ is the element-wise embedding vector, c_i is the local ionic occupancy, μ_M is the ionic chemical

potential, ϵ_i is the local volumetric strain and $\oplus$ means concatenation of features.

In the l -th network layer, the equivariant message transmitted between atoms j and i is given by:

$$m_{ij}^{(l)} = \sum_{l=0}^{l_{\max}} R_l^{(l)}(r_{ij}) [h_j^{(l)} \otimes Y_l(\hat{r}_{ij})]_{CG} \quad (11)$$

where the radial basis function $R_l^{(l)}$ is learnable, Y_l indicates the spherical harmonics, $\otimes$ means the tensor product, and $[\cdot]_{CG}$ indicates the coupling of irreducible representations by means of Clebsch-Gordan coefficients.

The aggregated message for atom i is:

$$m_i^{(l)} = \sum_{j \in N_i} f_c(r_{ij}) m_{ij}^{(l)} \quad (12)$$

with N_i being the set of neighboring atoms inside the cutoff radius of atom i , and f_c being a smooth cutoff function.

In order to enable the same atomic structure to have various effective potential-energy surfaces at varying charge-discharge states, the global electrochemical-state vector is defined by

$$s = [x, T, \mu_M, \bar{\epsilon}_{vol}, q_{cell}]^T \quad (13)$$

An electrochemical-state-gated update is then introduced:

$$g_i^{(l)} = \sigma \left[W_g^{(l)} (h_i^{(l)} \oplus s) + b_g^{(l)} \right], h_i^{(l+1)} = h_i^{(l)} + g_i^{(l)} \odot \Phi^{(l)} (h_i^{(l)}, m_i^{(l)}) \quad (14)$$

with σ as the activation function, $\odot$ denotes element-wise multiplication, and $\Phi^{(l)}$ is the equivariant feature-update function.

Equation (14) is the first significant generalization of the proposed model in comparison with the currently used equivariant machine-learning interatomic potentials. Traditional models are focused on learning a mapping between atomic structures and energies, but the suggested formulation is a joint mapping between atomic structure, electrochemical state, and energy. It allows the model to be sensitive to the interaction between atoms at various charge levels, temperatures, and strain states. Because the gate in Eq. (14) depends explicitly on the global state vector, the same local atomic geometry can yield different feature updates at different ion concentration, temperature, chemical potential, strain, or cell-charge states. This state-dependent mapping allows FP-EMLP to represent shifts of the effective potential-energy surface during intercalation/deintercalation, whereas a geometry-only potential can capture such shifts only indirectly through structural changes represented in its training data.

In order to explain charge transfer and polarization of an ionic material, the network first estimates a hypothetical latent charge q_i^0 of each atom. Constraint of total-charge conservation is subsequently applied [37]:

$$q_i = q_i^0 - \frac{1}{N} \left(\sum_{j=1}^N q_j^0 - Q_{\text{tot}} \right) \quad (15)$$

where Q_{tot} is the total charge of the simulated system.

The long-range electrostatic energy is evaluated using a differentiable Ewald operator [38]:

$$L_{\text{MLP}} = \frac{\lambda_E}{N_c} \|\Delta E\|_2^2 + \frac{\lambda_F}{3N_c N} \|\Delta F\|_2^2 + \frac{\lambda_\sigma}{N_c} \|\Delta \sigma\|_F^2 + \lambda_V (\Delta V)^2 + \lambda_m (\Delta E_m)^2 \quad (18)$$

where N_c is the number of training configurations $\lambda_E, \lambda_F, \lambda_\sigma, \lambda_V, \lambda_m$ are the weights of the energy, force, stress, equilibrium-voltage, and migration-barrier objectives, respectively.

Similar to the traditional machine-learning interatomic potentials based only on energies, atomic forces and stresses, Eq. (18) also adds equilibrium voltage and ion-migration barrier as auxiliary control objectives. It is the second significant extension of the suggested model because it permits a potential to provide direct support in computing the properties of energy storage instead of simply enhancing the quality of atomistic trajectory reproduction.

$$E_\theta^{\text{lr}} = \frac{1}{2} q^T G_{\text{Ewald}}(R) q + \frac{1}{2} \sum_{i=1}^N J_i q_i^2 + \sum_{i=1}^N \chi_i q_i \quad (16)$$

where G_{Ewald} represents the electrostatic interaction matrix both in real-space and reciprocal-space, and χ_i and J_i represent the environment-dependent electronegativity and charge hardness, respectively.

The long-range electrostatic interactions play a very important role in machine-learning potentials of the ionic, polar as well as interfacial materials. The new models frequently add additional long-range operators to purely local machine-learning potentials by charge equilibration, latent-charge representations, or explicit operators.

Atomic forces and stresses are obtained from analytical derivatives of the total energy:

$$F_{i,\theta} = -\frac{\partial E_\theta}{\partial r_i}, \sigma_\theta = \frac{1}{V} \frac{\partial E_\theta}{\partial \epsilon} \quad (17)$$

This formulation preserves physical consistency among the predicted energy, atomic forces, and stress.

The machine-learning interatomic potential is trained using the following multitask loss function:

2.3 Atomistic–Mesoscale–System Coupling Model

Trained machine-learning interatomic potentials have been applied to large-scale molecular dynamics simulations and thermodynamic sampling to compute diffusion coefficients, chemical free energies, elastic tensors, interfacial energies, chemical expansion coefficients, and thermophysical properties. The atomistic-scale quantities are then transferred to a mesoscale coupled chemo-mechanical-thermal model [39] which is shown in Fig. 3.

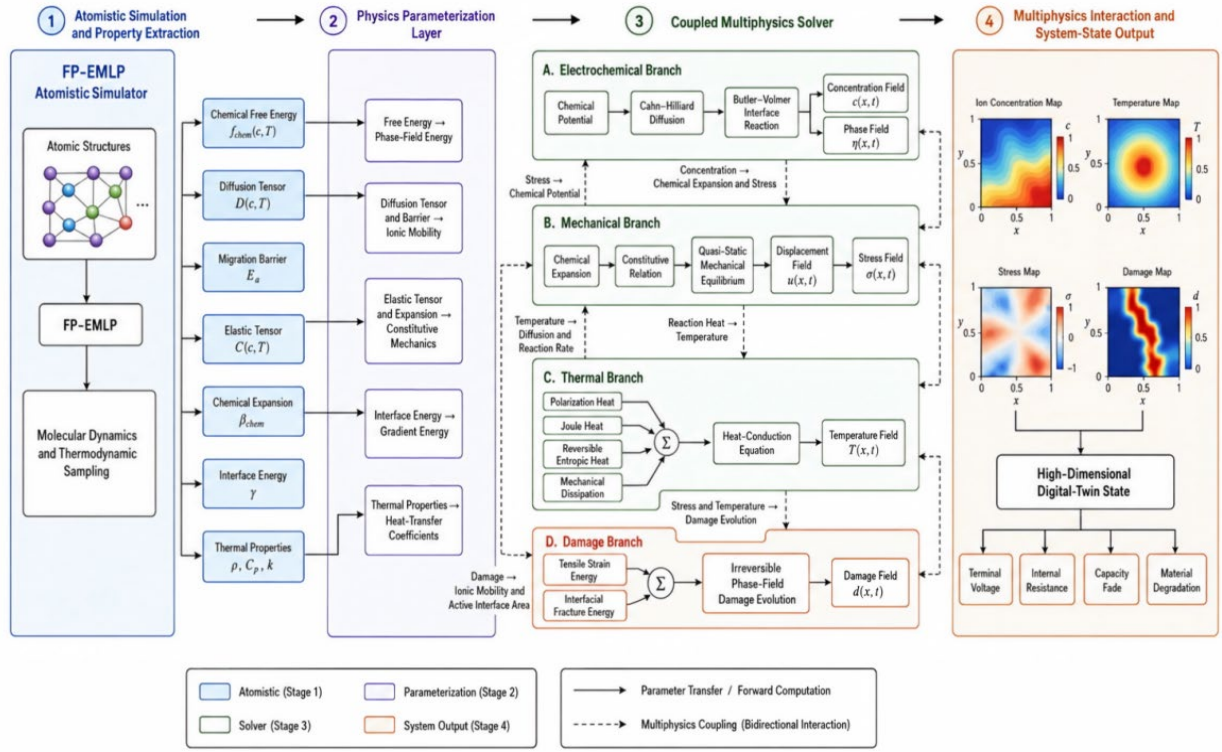


Figure 3. Atom-to-system multiscale coupling framework

In the mesoscale region Ω , the ion concentration field, phase-field order parameter $\eta(x,t)$, displacement field $u(x,t)$,

temperature field $T(x,t)$, and $d(x,t)$ damage field are defined. The entire free energy of the system can be written as:

$$G = \int_{\Omega} \left[f_{\text{chem}}^{\text{ML}}(c, \eta, T) + \frac{\kappa_c}{2} |\nabla c|^2 + \frac{\kappa_{\eta}}{2} |\nabla \eta|^2 + g(d) \psi_{\epsilon}(\epsilon - \epsilon^{\text{ch}}) + \frac{G_c}{2} \left(\frac{d^2}{l_d} + l_d |\nabla d|^2 \right) \right] d\Omega \quad (19)$$

where $f_{\text{chem}}^{\text{ML}}$ is the chemical free-energy density calculated using machine-learning interatomic potential, κ_c and κ_{η} are the gradient-energy coefficients, $g(d)$ is the stiffness degradation function, G_c is the critical fracture energy, and l_d is the damage regularization length.

This is done by taking the variational derivative of the total free energy with respect to the concentration field:

$$\mu = \frac{\delta G}{\delta c} = \frac{\partial f_{\text{chem}}^{\text{ML}}}{\partial c} - \kappa_c \nabla^2 c - \sigma : \frac{\partial \epsilon^{\text{ch}}}{\partial c} \quad (20)$$

in which the last term is the contribution of material stress to the ionic chemical potential.

$$M(c, T, d) = \frac{D^{\text{ML}}(c, T) c (1 - c)}{RT} (1 - \xi_d d), D^{\text{ML}}(c, T) = D_0(c) \exp \left[-\frac{E_m^{\text{ML}}(c)}{RT} \right] \quad (22)$$

where $D_0(c)$ is the diffusion prefactor that depends on the concentration, E_m^{ML} is the migration barrier estimated using the machine-learning interatomic potential and ξ_d is the correction coefficient that characterizes the effect of damage on the ion transport.

The evolution of the ion concentration satisfies mass conservation and a Cahn–Hilliard-type diffusion equation:

$$\frac{\partial c}{\partial t} = \nabla \cdot [M(c, T, d) \nabla \mu] + s_{\text{rxn}} \quad (21)$$

where $M(c, T, d)$ is the ionic mobility and s_{rxn} is the electrochemical reaction source term.

The ion mobility is measured by the diffusion coefficient that is calculated with a machine-learning interatomic potential:

Equation (22) allows transferring directly between atomistic migration barriers and mesoscale diffusion parameters without having to rely on the conventional method of the diffusion coefficient as an empirically constant

quantity that is unrelated to concentration, temperature or structural state.

The reaction flux at the electrode-electrolyte interface can be defined based on a modified Butler-Volmer equation [40]:

$$j=j_0(c,T,d)\left[\exp\left(\frac{\alpha_a F \eta_{ov}}{RT}\right)-\exp\left(-\frac{\alpha_c F \eta_{ov}}{RT}\right)\right] \quad (23)$$

j_0 is the exchange current density, η_{ov} is the interfacial overpotential, α_a and α_c are the anodic and cathodic charge-transfer coefficients, respectively. Letting j_0 depend on the damage field allows accounting of new reaction surfaces formed due to cracking and alteration of the local electrochemically active area.

The material satisfies quasi-static mechanical equilibrium [41]:

$$\nabla \cdot \sigma = 0, \sigma = g(d) C^{ML}(c, T) : [\varepsilon - \beta^{ML}(c - c_0) I] \quad (24)$$

$C^{ML}(c, T)$ in which denotes the elastic tensor depending on the concentration and temperature, calculated with the help of machine-learning interatomic potential, and β^{ML} is the chemical expansion coefficient.

The temperature field generated during cycling of the energy-storage material is governed by [42]:

$$\rho C_p \frac{\partial T}{\partial t} = \nabla \cdot (k_T \nabla T) + j \eta_{ov} + i^T R_{int} i + T \frac{\partial U_{eq}}{\partial T} j + Q_{mech} \quad (25)$$

where ρ is the density of the material, C_p is its specific heat and k_T is its thermal conductivity. The second, third, and fourth terms on the right-hand side are polarization heat, Joule heat, and reversible entropic heat, respectively and Q_{mech} is a

form of mechanical dissipation resulting due to the deformation and damage.

The damage field is described using an irreversible phase-field evolution equation [43]:

$$\tau_d \frac{\partial d}{\partial t} = \langle 2(1-d)H - \frac{G_c}{l_d} d + G_c l_d \nabla^2 d \rangle_+, \frac{\partial d}{\partial t} \geq 0 \quad (26)$$

where τ_d is the characteristic damage-evolution time, H is the maximal historical tensile strain-energy density, and $\langle \cdot \rangle_+$ is the positive-part operator.

Phase-field models have found widespread applications in characterizing diffusion due to lithiation, stress development, movement of phase boundaries, crack propagation and capacity fade. The main difference between the current model and previous ones is that the free energy, diffusion coefficient, elastic tensor, and chemical expansion parameters are provided by first-principles calculations and machine-learning interatomic potential instead of being fully empirical.

2.4 Online State Update and Degradation-Aware Control of the Digital Twin

The complete mesoscale model involves high dimensional concentration, temperature, stress, and damage fields, which make it computationally infeasible to monitor and control energy-storage systems in real time. Proper orthogonal decomposition or a physics-constrained autoencoder is thus used to reduce the multiphysics fields as shown in Fig. 4. The resulting reduced representation forms a reduced-order model (ROM) and supports estimation of state of charge (SOC) and state of health (SOH).

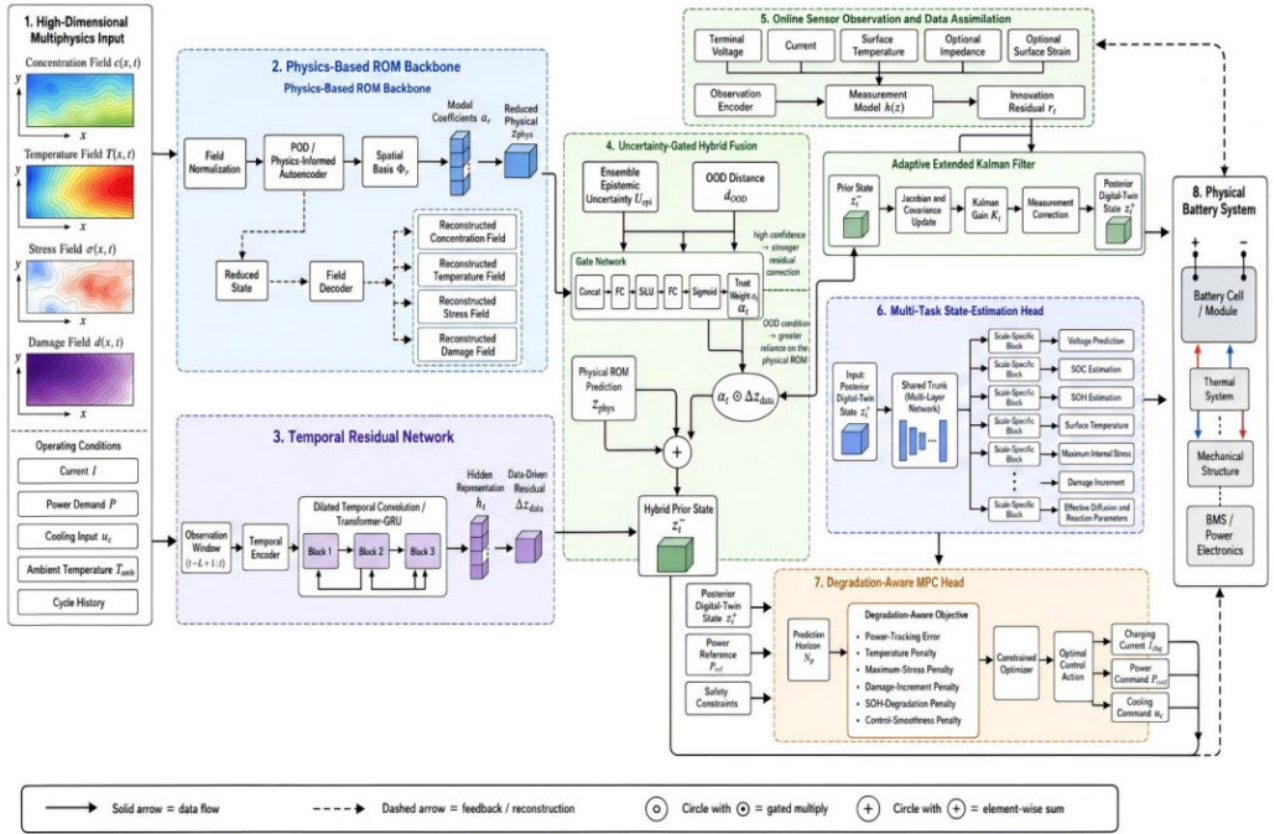


Figure 4. Online state update and degradation-aware control framework

An arbitrary physical field $\psi(x,t)$ is approximated as:

$$\psi(x,t) \approx \bar{\psi}(x) + \sum_{r=1}^{R_\psi} a_{\psi,r}(t) \phi_{\psi,r}(x), \psi \in \{c, T, \eta, d\} \quad (27)$$

Here $\bar{\psi}$ is the field mean, $\phi_{\psi,r}$ is the r -th spatial basis function, $a_{\psi,r}$ is the time dependent coefficient that corresponds to it, and R_ψ is the number of reduced-order modes kept.

The digital-twin state vector is constructed from the reduced-order coefficients as

$$\mathbf{x}_k = [a_{c,k}^T, a_{T,k}^T, a_{\eta,k}^T, a_{d,k}^T, \text{SOC}_k, \text{SOH}_k, \mathbf{R}_{\text{int},k}, \mathbf{g}_k^T]^T \quad (28)$$

where $\mathbf{g}_k^T$ is the vector of model parameters to be determined online, such as the effective diffusion coefficient, reaction-rate constant, heat-transfer coefficient, damage coefficient, and effective active area.

The combination of a physics-based reduced-order model with a residual model based on data is known as the digital-twin state transition [44].

$$\mathbf{x}_{k+1} = \mathbf{f}_{\text{ROM}}(\mathbf{x}_k, \mathbf{u}_k; \mathbf{g}_k) + \gamma_k \mathbf{r}_\omega(\mathbf{x}_k, \mathbf{u}_k; \mathbf{y}_{k-L:k}) + \mathbf{w}_k \quad (29)$$

where $\mathbf{f}_{\text{ROM}}$ is the reduced-order solver, $\mathbf{r}_\omega$ is the model residual predicted by a temporal neural network, $\mathbf{u}_k$ has the current, power, and cooling-control inputs, and $\mathbf{w}_k$ means process noise.

The confidence weight assigned to the data-driven residual is defined as:

$$\gamma_k = \exp \left[-\frac{s_{\text{ep},k}^2 + s_{\text{ood},k}^2}{\tau_s^2} \right] \quad (30)$$

where $s_{\text{ep},k}^2$ is the epistemic uncertainty of the model, $s_{\text{ood},k}^2$ measures how far the current operating condition deviates from the training-data distribution, and τ_s^2 is the uncertainty scaling parameter.

The model is capable of compensating the unmodeled dynamics when it operates in a regime with good representation, and γ_k is kept high. In out-of-distribution (OOD) situations, including high temperatures, unusual charge-discharge rates, or extreme aging, γ_k drops automatically, and the digital twin becomes more dependent on the physics-based model. Such uncertainty-dependent weighting is the third significant advancement over traditional hybrid digital-twin models.

The online sensor observation model is expressed as:

$$y_k = h(x_k, u_k) + v_k = [V_k, I_k, T_{s,k}, Z_k', Z_k'', \varepsilon_{s,k}]^T + v_k \quad (31)$$

where V_k , I_k and $T_{s,k}$ are the terminal voltage, current, and surface temperature, respectively; Z_k' and Z_k'' are the real and imaginary parts of the online impedance; $\varepsilon_{s,k}$ is the measured surface strain; and v_k represents measurement noise. Throughout the manuscript, ε denotes the mesoscale strain tensor, ε_i the local volumetric-strain feature of atom i , ε^{ch} the chemical strain in the mechanical model, and $\varepsilon_{s,k}$ the measured surface strain in the observation vector.

The framework does not imply that all sensor channels are available at the same time. If only current, voltage, and

temperature are measured, then impedance and strain channels may be disregarded. The availability of impedance or fiber-optic strain sensors allows enhancing the observability of the internal diffusion, stress and damage states. In a digital twin, battery-state estimation is normally done through synchronization between the virtual model, online operation information and a data-driven estimator [45].

The adaptive extended Kalman filter is used to jointly update the digital-twin states and parameters. After the update, degradation-aware model predictive control (MPC) is carried out:

$$\hat{x}_k^- = f_{DT}(\hat{x}_{k-1}^+, u_{k-1}) \quad (32)$$

$$P_k^- = A_{k-1} P_{k-1}^+ A_{k-1}^T + Q_k \quad (33)$$

$$K_k = P_k^- H_k^T (H_k P_k^- H_k^T + R_k)^{-1} \quad (34)$$

$$\hat{x}_k^+ = \hat{x}_k^- + K_k [y_k - h(\hat{x}_k^-, u_k)] \quad (35)$$

$$P_k^+ = (I - K_k H_k) P_k^- \quad (36)$$

$$\min_{u_{k+k+N_p-1}} J = \sum_{j=0}^{N_p-1} \left[w_P (P_{k+j} - P_{ref,k+j})^2 + w_T \|T_{k+j} - T_{max}\|_+^2 + w_\sigma \|\sigma_{max,k+j} - \sigma_{lim}\|_+^2 + w_d (d_{k+j+1} - d_{k+j})^2 + w_{SOH} (SOH_{k+j} - SOH_{k+j+1})^2 + w_u \|\Delta u_{k+j}\|_2^2 \right] \quad (37)$$

$$\text{s.t. } x_{k+j+1} = f_{DT}(x_{k+j}, u_{k+j}), I_{min} \leq I_{k+j} \leq I_{max}, V_{min} \leq V_{k+j} \leq V_{max}, T_{k+j} \leq T_{max}, \sigma_{max,k+j} \leq \sigma_{lim}, d_{k+j} \leq d_{lim} \quad (38)$$

In this case, the Jacobian matrices of the state-transition and the observation functions are denoted by A_k and H_k ; P_k denotes the state covariance matrix; Q_k and R_k denote the process- and measurement-noise covariance matrices; and N_p denotes the prediction horizon. The nonnegative weights w_P , w_T , w_σ , w_d , w_{SOH} , and w_u balance power tracking, temperature, stress, damage, SOH degradation, and input smoothness, respectively.

Control objective takes into consideration not just the power tracking, temperature regulation, and smoothness of the inputs, but also the maximum internal stress of materials, cumulative damage, and degradation of SOH. With this formulation, the results of first-principles calculations and simulation studies at the materials scale can be used directly in the operation and control of the energy-storage system instead of being merely offline tools to screen materials.

3. Experiments and Results

The proposed FP-EMLP-DT framework is assessed in terms of machine-learning interatomic-potential accuracy, cross-scale physical-field reconstruction, digital-twin state estimation, degradation-aware control, ablation behavior, and robustness.

3.1 Public Datasets and Experimental Settings

The atomistic-scale experiments are based on public materials datasets, which include Materials Project trajectory (MPtrj), Materials Project Battery, and Joint Automated Repository for Various Integrated Simulations-Density Functional Theory (JARVIS-DFT). The 120,000 energy storage material configurations in MPtrj are chosen to pretrain the machine learning interatomic potential. Material specific fine-tuning and cross-material generalization evaluation are performed with 7,840 electrode configurations obtained by Materials Project Battery. To serve as an external property test set, another set of 3,260 structures and their accompanying elastic-property information is taken out of JARVIS-DFT. The cross-scale research is focused on LiFePO₄ /FePO₄ as the main system, while LiCoO₂, LiMn₂O₄, and nickel-manganese-cobalt (NMC) architectures are utilized only to assess cross-material generalization.

In the active-learning experiments, 2,000 LiFePO₄ /FePO₄ structures produced at the same first-principles conditions have been chosen as the starting labeled set. The next step is iterative sampling of a population of 40,000 candidate structures at various concentrations of lithium, defects, strain states, phase boundaries, and thermal

perturbation. Every iteration will add 500 first-principles-labeled configurations. In eight iterations, there will be a final material-specific training set of 6,000 configurations.

LiFePO₄ battery data provided by Center for Advanced Life Cycle Engineering (CALCE) are applied to the system scale to train the digital-twins and conduct internal tests providing 52,800 time windows. National Aeronautics and Space Administration (NASA) Prognostics Center of Excellence (PCoE) battery information is used to assess the external transfer and has 18,600 time windows. Training, validation and test sets in atomistic data are separated in the proportion of 70%, 15% and 15% respectively based on

material composition, structural prototype and calculation trajectory. The partitioning of battery data by whole battery cells is done to control the leakage of data. High fidelity multiphysics models are used to generate reference concentration, stress and damage fields, whereas the public battery datasets are mainly used to evaluate measurable states such as voltage, SOC, SOH, and surface temperature. At the battery level, all time windows from a given physical cell are kept in a single subset; CALCE cells are used for model development and internal testing, whereas NASA cells are reserved for external transfer testing.

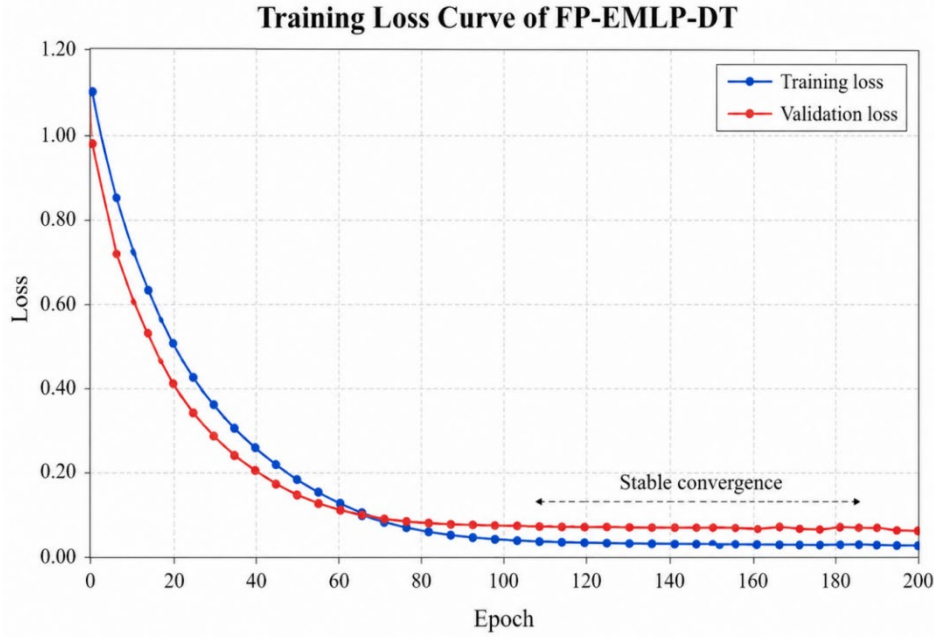


Figure 5. Training loss curves

This is demonstrated in Fig. 5 whereby training and validation losses decline sharply as the number of epochs increases and level off at about 80-100 epochs which shows that the model has effectively converged. Even though the training loss approaches a little less than the validation loss towards the end, both curves have comparable trends without a strong bounce. Such behavior indicates a consistent training

procedure and there are no signs of overfitting. The key experimental and model settings are summarized in Table 1. The density functional theory calculations use the Perdew-Burke-Ernzerhof functional with a Hubbard-U correction (PBE+U); the NEB setup and ab initio molecular dynamics (AIMD) settings are also listed for reproducibility.

Table 1. Key experimental and model settings (dataset partitioning follows Section 3.1)

Module	Parameter	Setting
DFT	Exchange-Correlation Functional	PBE+U
DFT	Plane-Wave Cutoff Energy	520 eV
NEB	Number of Intermediate Images	7
AIMD	Temperature Range	300–900K
DFT-AIMD	Time Step / Trajectory Length	1 fs/15 ps
AIMD	Time Step / Trajectory Length	1 fs/200 ps
FP-EMLP	Equivariant Layers	4
FP-EMLP	Hidden Dimension	128

Module	Parameter	Setting
FP-EMLP	Cutoff Radius	5.0Å
FP-EMLP	Maximum Harmonic Order	lmax=2
Training	Optimizer / Learning Rate	AdamW/1.0×10 ⁻³
Training	Batch Size / Maximum Epochs	16/500
Digital Twin	Latent-State Dimension	64
Digital Twin	Observation Window	60 time steps
Digital Twin	Sampling Interval	1 s
MPC	Prediction Horizon	20 steps
Reproducibility	Random Seeds	5

3.2 Machine-Learning Interatomic Potential and Cross-Scale Physical Performance

Firstly, the errors in prediction of the various models are compared on the basis of total energy, atomic force, average voltage, and ion-migration barrier. In order to assess the quality of transferring atomistic parameters to the mesoscale,

the mean relative errors in the concentration, temperature, stress, and damage fields are also computed with respect to the high-fidelity multiphysics reference model. Prediction accuracy is reported primarily using mean absolute error (MAE). The compared baselines include Directional Message Passing Neural Network (DimeNet++), Polarizable Atom Interaction Neural Network (PaiNN), NequIP, CHGNet, and MACE.

Table 2. Performance comparison of different machine-learning potentials (all models use the same held-out atomistic test partition defined in Section 3.1)

Model	Energy (meV atom ⁻¹)	MAEForce (meV Å ⁻¹)	MAEVoltage (V)	MAE Migration-Barrier MAE (eV)	Field Relative Error (%)
SchNet	15.8	73	0.118	0.091	12.7
DimeNet++	12.9	61	0.102	0.082	10.8
PaiNN	11.4	54	0.094	0.074	9.6
NequIP	8.7	42	0.078	0.061	7.4
CHGNet	8.3	41	0.069	0.053	6.8
MACE	7.9	39	0.071	0.056	6.6
FP-EMLP	6.5	33	0.051	0.038	4.8

The equivariant machine-learning interatomic potentials are found to be superior to the continuous-filter model and the angular-message-passing model in general, as is depicted in Table 2. The lowest error among all evaluation metrics is obtained by FP-EMLP. Relative to MACE, FP-EMLP lowers the energy MAE from 7.9 to 6.5 meV atom⁻¹, a 17.7% decrease. The force MAE decreases from 39 to 33 meV Angstrom⁻¹, an improvement of 15.4%.

FP-EMLP lowers the voltage error of 0.069 to 0.051 V and the migration-barrier error of 0.053 to 0.038 eV in case of properties associated with energy-storage performance compared to CHGNet. Such findings suggest that electrochemical-state gating, the long-range electrostatics branch, and auxiliary supervisory measures with voltage and migration barriers enhance the description of ion insertion and diffusion.

These cross-scale findings also indicate that the FP-EMLP-parameterized model has relative errors of 4.1%, 3.7%, 5.2% and 6.1% in the concentration, temperature, stress and damage fields respectively as well as a cumulative field error of 4.8%. In contrast, the total error of the multiphysics model based on fixed empirical parameters is

greater than 10%. Here are some examples: the results indicate that diffusion coefficients, elastic tensors, chemical expansion coefficients, and migration barriers calculated by the machine-learning interatomic potential increase the precision of mesoscale field-evolution predictions.

The evaluation of the considered active-learning strategy is made by comparing random sampling, farthest-point sampling, committee-uncertainty sampling and the proposed joint sampling method on the same initial set of 2,000 labeled configurations and the same pool of candidates (40,000). All the methods add 500 first-principles-labeled configurations in each iteration and have the same FP-EMLP architecture, optimizer and number of training epochs. The equilibrium crystals, thermally perturbed structures, vacancy and antisite defects, strained configurations, phase boundaries, and ion-migration transition states form the fixed test set. Such a design would make sure that active learning cannot be biased towards equilibrium configurations.

With an increasing number of labeled configurations by a factor of three (2,000 to 6,000), all four methods demonstrate a steady decrease in energy, force and migration-barrier error.

Nevertheless, the suggested approach is faster. At 6,000 labeled configurations, random sampling provides energy, force, and migration-barrier MAEs of 9.0 meV atom⁻¹, 44 meV Å⁻¹, and 0.059 eV respectively. The respective errors obtained by farthest point sampling are 8.0 meV atom⁻¹, 40 meV Å⁻¹, and 0.050 eV. The committee-uncertainty sampling gives 7.1 meV atom⁻¹, 36 meV Å⁻¹, and 0.041 eV. The proposed joint sampling strategy also minimizes these errors down to 6.5 meV atom⁻¹, 33 meV Å⁻¹, and 0.038 eV which is in agreement with the full FP-EMLP results in Table 2.

Regarding the labeling efficiency, the suggested approach minimizes the force MAE to less than 40 meV Å⁻¹ by employing merely 4,000 labeled configurations, in contrast to

farthest-point sampling, which takes about 6,000 configurations to achieve the same precision. The proposed approach will thus decrease the need to add further first-principles labels by about 33.3%. The approach also covers 91.2 percent of high-risk configurations such as high temperature states, defects, phase boundaries and migration transition states. The value is larger compared to values of 52.3, 68.7 and 79.4 that were recorded with random sampling, farthest-point sampling and uncertainty-only sampling respectively. The findings indicate that a combination of committee uncertainty, structural novelty, and physics-based risk avoids the re-selection of geometrically similar configurations with high uncertainties and enhances the coverage of rare nonequilibrium states.

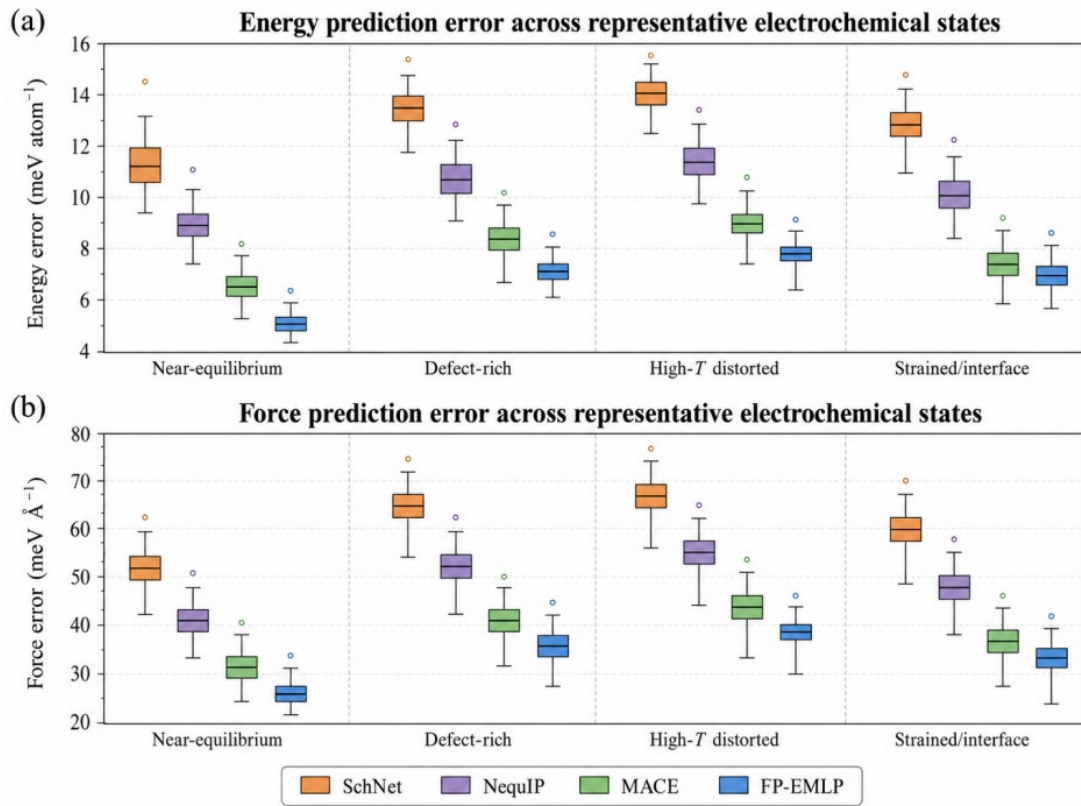


Figure 6. Prediction errors across electrochemical states

Comparing the energy and force error distributions in near-equilibrium, defect-rich, high-temperature-distorted, and strained/interfacial systems allows evaluating predictive stability at various electrochemical states. This is demonstrated in Fig. 6 where FP-EMLP has smaller median errors and less spread in all four state categories. In more complicated nonequilibrium systems with defects, high

temperature and interfaces it still has an advantage. Conversely, the other competing models have wider error distributions, which means that electrochemical-state conditioning and long-range interaction modeling enhances the generalization in more complicated areas of the potential-energy surface.

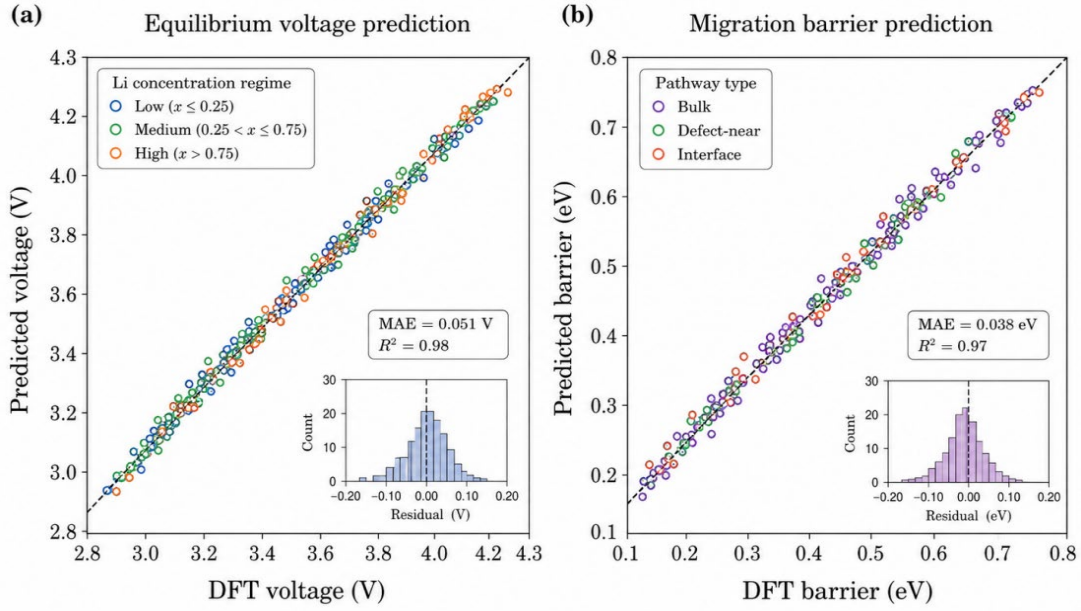


Figure 7. Voltage and migration-barrier prediction

Figure 7 is a comparison of the equilibrium voltages and ion-migration barriers obtained through FP-EMLP and their respective DFT reference values. Both groups of predictions are close to the identity line with no significant systemic bias at various lithium-concentration ranges or types of migration pathways. The residuals are approximately equal to zero, which is an indication that the model gives constant forecasts of the main electrochemical characteristics in different lithiation conditions and diffusion conditions.

3.3 Digital-Twin State Estimation and Degradation-Aware Control

The state estimation experiments of digital twins are used to measure the prediction error of terminal voltage, SOC, SOH and surface temperature and the inference time per time step. Test data consists of internal CALCE test cells and external NASA test cells. Performance is quantified using root mean square error (RMSE), MAE, and per-step inference time, as summarized in Table 3. The baselines include an equivalent-circuit model with an extended Kalman filter (ECM-EKF), long short-term memory (LSTM), temporal convolutional network (TCN), Transformer, and a physics-based ROM.

Table 3. Digital-twin state-estimation performance

Method	Voltage (mV)	RMSE SOC (%)	MAE SOH (%)	MAE Temperature (°C)	RMSE Inference Time per Step (ms)
ECM-EKF	31.4	2.18	2.72	1.31	4.2
LSTM	24.8	1.62	2.10	1.07	5.8
TCN	22.1	1.43	1.88	0.94	4.9
Transformer	20.5	1.31	1.72	0.89	9.7
Physics-Based ROM	21.7	1.49	1.94	0.91	6.1
Hybrid Model without Uncertainty Gate	18.6	1.17	1.52	0.78	8.4
FP-EMLP-DT	15.8	0.93	1.21	0.64	9.1

The typical ECM-EKF is inexpensive to compute on average but gives high error rates in the estimates of voltage, SOC, and SOH. LSTM, TCN and Transformer models are good at predicting dynamics over short term intervals, but their errors sum up to longer term degradation. The physics-based reduced-order model offers stable forecasts but it cannot be

compared with the hybrid physics-data models in terms of total accuracy since unmodeled dynamics are not completely canceled.

The overall FP-EMLP-DT has a voltage RMSE of 15.8 mV, an SOC MAE of 0.93 percent, an SOH MAE of 1.21 percent, and a temperature RMSE of 0.64 degrees Celsius.

The FP-EMLP-DT compared to the hybrid model without uncertainty gating decreases the SOC and SOH errors by 20.5% and 20.4, respectively. Its inference time per step is 9.1 ms which is significantly lower than the 1 s sampling interval and thus meets the conditions of online operation.

The control experiments have 2C fast charging, 25 degrees Celsius ambient temperature, and initial SOH of 90 percent

as the base conditions. Various control methods are analyzed in relation to the time to charge, maximum temperature, maximum stress, material degradation, and energy efficiency. The evaluated strategies include constant-current-constant-voltage (CC-CV) charging and several MPC-based controllers.

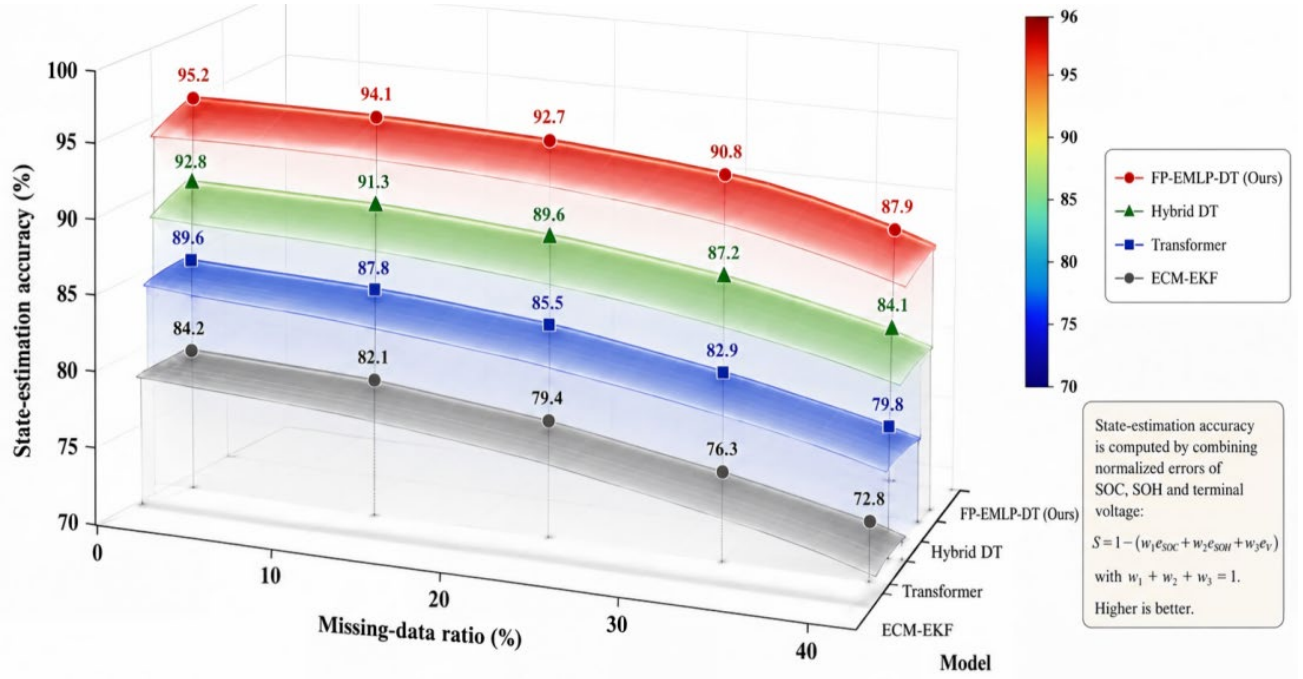


Figure 8. State-estimation robustness of FP-EMLP-DT under increasing missing-data ratios

In order to investigate how stable state estimation is with incomplete observations, the percentage of missing input data is slowly raised and the results of various models are tested. Figure 8 demonstrates that the accuracy of state estimation of all methods decreases with the increasing missing data ratio.

FP-EMLP-DT is the most accurate, however, it has a lower performance drop. The finding suggests that the physics-based reduced-order branch and uncertainty gating can be used to successfully control the negative impact of missing observations on the online state updates.

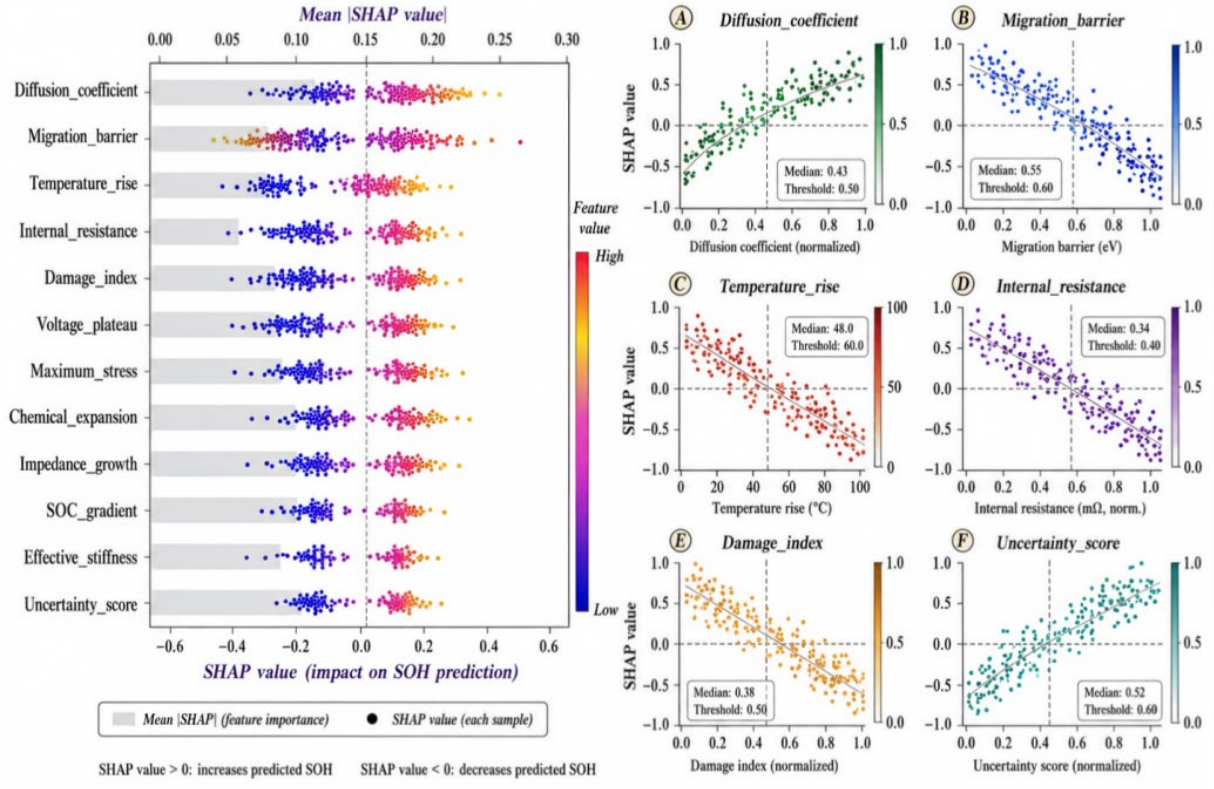


Figure 9. Feature-importance and dependence analysis for SOH prediction in FP-EMLP-DT

SHapley Additive exPlanations (SHAP) analysis is applied in order to ascertain what extent multiscale physical states impact SOH estimation, i.e., the importance and direction of influence of digital-twin characteristics. These parameters, namely, the diffusion coefficient, migration barrier, temperature rise, internal resistance, and damage index are highly significant towards predicting the SOH as illustrated in Fig. 9. Increased diffusion capability has been found to be a positive contributor to the estimated SOH and increased levels of migration barrier, temperature rise, internal

resistance, and damage level have been found to be mostly negative contributors. These findings indicate that the model interprets the variability of battery-health across different angles such as the flow of ions, thermal status, and the breakdown of materials. The uncertainty score also has a strong correlation with the SHAP values which means that the model identifies how prediction confidence affects health-state estimation. The charging-control strategies and their quantitative performance are summarized in Table 4.

Table 4. Performance comparison of different charging-control strategies

Control Strategy	Charging Time (min)	Maximum Temperature (°C)	Maximum Stress (MPa)	Normalized Damage Increment (dimensionless)	Energy Efficiency (%)
CC-CV	48.0	46.8	182	1.00	91.8
Temperature-Threshold Control	52.6	43.1	169	0.84	92.4
Conventional MPC	45.7	42.6	158	0.76	93.1
SOH-Aware MPC	47.4	41.9	149	0.63	93.0
Degradation-Aware MPC	46.8	40.7	132	0.48	93.4

The CC-CV control is easy to apply but the peak temperature and stress levels are rather large. The temperature-threshold control restricts the level of temperature increase, although charging time grows to 52.6 minutes. Traditional MPC has

the least charging time with no explicit limitation on material stress or damage.

On the other hand, degradation aware-MPC has a 2.4 percent lower increase in the charging time compared to the traditional MPC, which has the maximum temperature of 42.6, the maximum stress of 158 MPa and the normalized damage increment of 0.76 reduced to 40.7, 132 MPa and 0.48 respectively. The findings indicate that it is possible to reduce material degradation without significantly affecting the rate of charging through the control objective by including the internal temperature, stress and damage states.

3.4 Ablation, Robustness, and Computational Efficiency

In order to measure the role of each component, the electrochemical-state gate, long-range electrostatics branch, voltage and migration-barrier auxiliary losses, physics-based reduced-order branch, and uncertainty gate are eliminated separately. The rest of the training conditions do not change.

Table 5. Ablation results of the proposed FP-EMLP-DT framework

Model Variant	Energy MAE (meV atom ⁻¹)	Migration-Barrier MAE (eV)	SOC MAE (%)	OOD SOC MAE (%)	Field Error (%)	Relative
Without Electrochemical-State Gate	7.4	0.052	1.05	1.73	5.7	
Without Long-Range Electrostatics	7.1	0.058	1.01	1.65	5.5	
Without Voltage and Barrier Auxiliary Losses	6.9	0.064	0.99	1.57	5.3	
Without Physics-Based ROM Branch	6.5	0.038	1.28	2.11	8.7	
Without Uncertainty Gate	6.6	0.039	0.97	1.92	5.1	
Complete FP-EMLP-DT	6.5	0.038	0.93	1.42	4.8	

The ablation results indicate that the various modules have different functional roles as indicated in Table 5. Deletion of the electrochemical-state gate causes an increase in energy MAE of 6.5-7.4 meV atom⁻¹ which implies that conditioning by concentration, temperature and strain enhances the representation of potential-energy surfaces across electrochemical states. Deleting the long-range electrostatics branch or the voltage and migration-barrier auxiliary losses raises the migration-barrier MAE to 0.058 and 0.064 eV respectively. This confirms the fact that nonlocal interactions and multitask physical supervision are especially relevant to predict the migration of ions. In particular, removing the long-range electrostatics branch increases the migration-barrier MAE from 0.038 to 0.058 eV, corresponding to a 52.6% increase. Because a migration barrier is the energy difference between an initial state and a transition state, nonlocal charge redistribution and polarization along the migration path can alter this difference even when local

geometries are similar; omitting the electrostatic branch therefore degrades barrier prediction disproportionately.

Physics-based reduced-order branch mainly influences internal-field reconstruction and out-of-distribution prediction. The elimination thereof causes the field-variable error to rise by 4.8-8.7%. Stability in non-normal operating conditions is significantly enhanced with uncertainty gating, and the absence of it causes the OOD SOC MAE to grow by 1.42-1.92%. Complete model shows a SOC MAE of 1.18 and 1.36 at 2% Gaussian noise and 20% missing data respectively, indicating that it can tolerate noisy and incomplete observations.

The FP-EMLP has a computational speed of about 104 times that of the first-principles method in computing atomic forces. Simplified digital twin is around 86 times faster compared to the full three-dimensional multiphysics model. State estimation and MPC control computation time averages at 37 ms which meets the demands of online monitoring and real-time control.

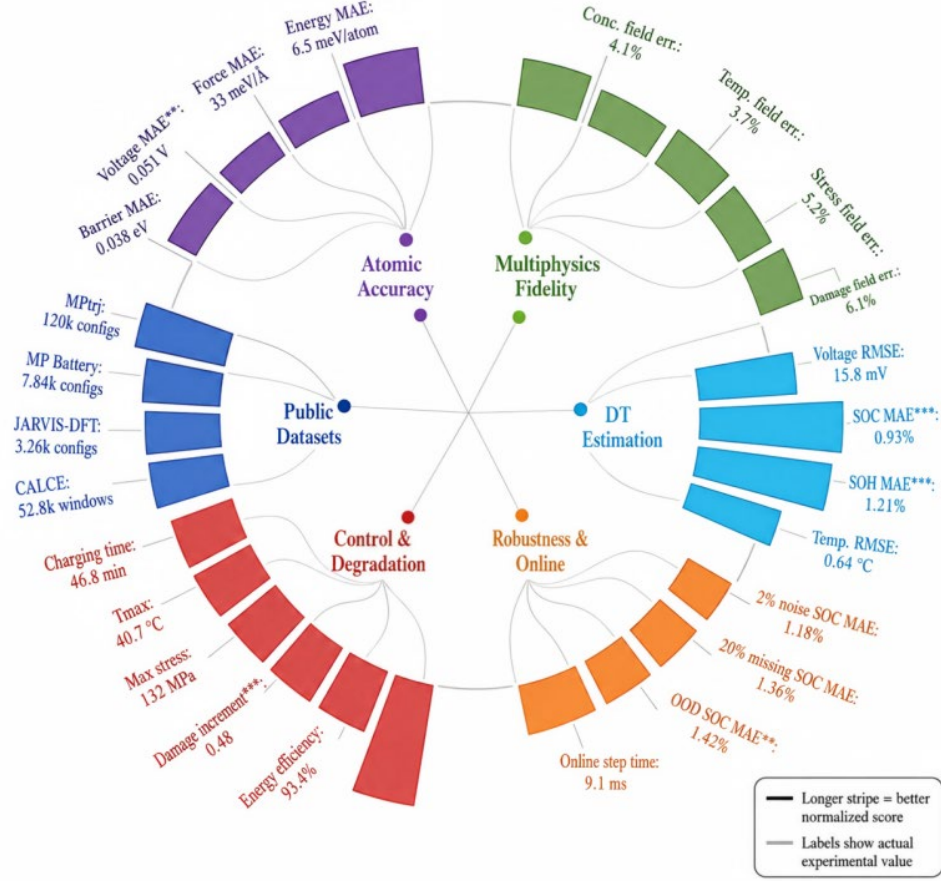


Figure 10. Circular summary of the multiscale experimental results of FP-EMLP-DT

In order to present a summary of the performance of the FP-EMLP-DT at various scales, Figure 10 shows the results on the publicly available datasets, atomistic prediction, multiphysics-field reconstruction, digital-twin state estimation, robustness, and degradation-aware control. The

suggested architecture is highly effective in atomistic precision, multi-scale field reconstruction, on-line state estimation, and degradation alleviation and forms a continuous validation sequence between material modeling and system-level operation.

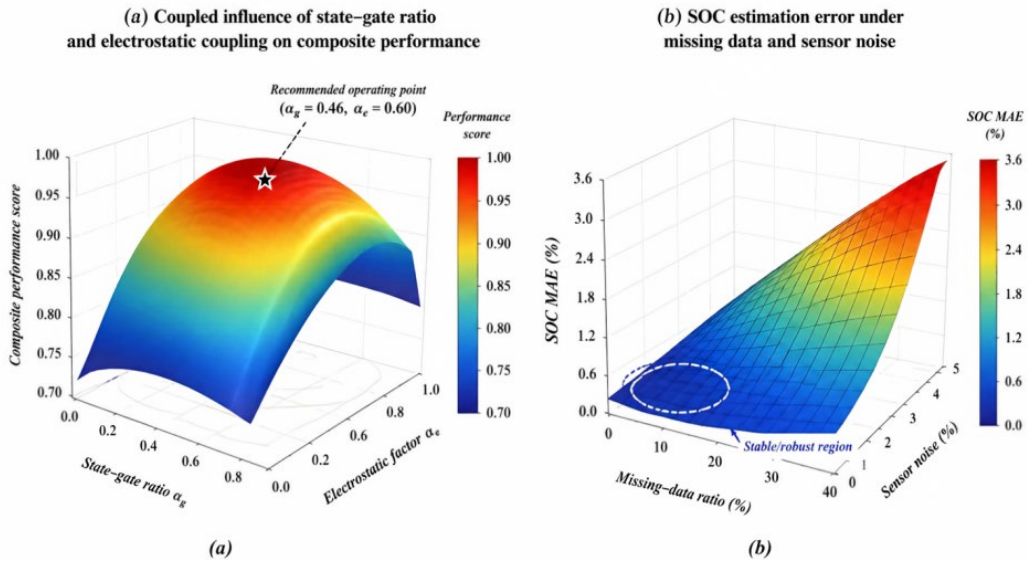


Figure 11. Parameter sensitivity and robustness analysis of FP-EMLP-DT

In order to further explore the impact of key model parameters and incomplete data, the state-gating coefficient, electrostatic coupling coefficient, missing-data ratio, and

sensor-noise level are changed. In this case, α_g and α_e represent the relative scaling coefficients of state-gating and long-range electrostatics branches, respectively. Composite performance score is calculated through normalization of atomistic prediction, cross-scale field-reconstruction, and state-estimation measures.

As it is demonstrated in Fig. 11(a), the composite performance does not vary in a linear fashion depending on

the two coupling strengths and achieves a favourable area close to $\alpha_g \sim 0.46$ and $\alpha_e \sim 0.60$. The outcome of this suggests that the balance between electrochemical state conditioning and long range electrostatic modeling is necessary. In Figure 11(b), it can be seen that the SOC estimation error also rises when the missing data proportion and sensor noise level are increased at the same time. However, even under low to moderate perturbations, the model is fairly stable and confirms that uncertainty gating and physics based reduced order branch enhances robustness to incomplete observations.

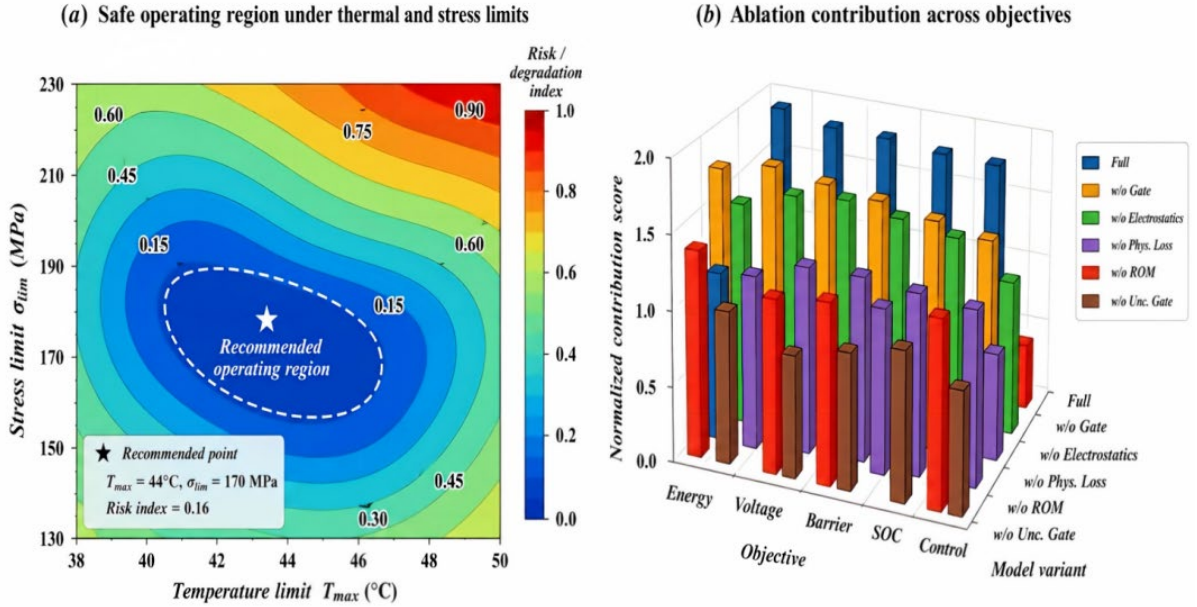


Figure 12. Control trade-off and ablation analysis of FP-EMLP-DT

In order to study further the impact of control constraints and key modules on the overall performance, it is shown in Fig. 12 that both temperature and stress limits are the joint determinants of the system-level risk and degradation. A moderately constrained regime produces a relatively stable low-risk operating region. The individual modules also

perform in different ways in different tasks. State-gating and long-range electrostatics branches have the strongest influence on atomistic and electrochemical behavior, while physics-based reduced-order and uncertainty-gating branches are more influential with respect to state estimation and control.

Table 6. Sensitivity analysis of active-learning weights

Setting	$\omega_E, \omega_F, \omega_D, \omega_P$	Energy MAE (meV atom ⁻¹)	Force MAE (meV Å ⁻¹)	High-risk coverage (%)	DFT-label reduction (%)
S1	(0.40, 0.20, 0.20, 0.20)	6.4	35	86.8	31.0
S2	(0.20, 0.40, 0.20, 0.20)	6.8	32	87.5	31.8
Baseline	(0.25, 0.25, 0.20, 0.30)	6.5	33	91.2	33.3
S4	(0.30, 0.30, 0.30, 0.10)	6.6	33	82.4	35.0
S5	(0.20, 0.20, 0.15, 0.45)	6.8	35	94.5	27.8

Table 6 presents the sensitivity of the active-learning strategy to different weight combinations. Increasing the energy or force uncertainty weight slightly improves the corresponding prediction metric but reduces the coverage of high-risk configurations. A lower physics-risk weight increases labeling efficiency but decreases high-risk coverage, whereas an excessively large physics-risk weight improves high-risk coverage at the expense of prediction accuracy and labeling efficiency. The baseline setting (0.25,0.25,0.20,0.30) therefore provides a balanced trade-off among prediction accuracy, high-risk configuration coverage, and DFT-labeling efficiency.

4. Conclusions

The paper fills the gap in unification of first-principles calculations and machine-learning interatomic potentials with multiphysics modeling and online battery management by introducing the FP-EMLP-DT multiscale digital-twin framework. This is what can be taken as the general conclusion.

(1) A machine-learning interatomic potential based on electrochemical-state-conditioned equivariance was created. The system utilizes the variables of ion concentration, temperature, chemical potential and local strain to gate atomic properties and has an added charge-conserving projection and differential long-range electrostatics. It is trained by multitask supervision on energy, atomic forces, stress, equilibrium voltage, and migration barriers. FP-EMLP gives energy and force MAEs of 6.5 meV atom⁻¹ and 33 meV Å⁻¹ respectively as well as voltage and migration-barrier MAEs of 0.051 V and 0.038 eV. These numbers are less than the graph-neural-network potentials that are used in comparison. The active-learning strategy, i.e., model uncertainty combined with structural novelty and physics-based risk reduces the required first-principles labels by around 33.3 percent and maximizes the coverage of high-risk nonequilibrium states to 91.2 percent.

(2) An AI-assisted atomistic-mesoscopic-system parameter-transfer method has been developed. Electrochemical-mechanical-thermal-damage coupling model is based on the chemical free energies, diffusion coefficients, elastic tensors, and chemical expansion parameters calculated using the machine-learning interatomic

potentials. It allows the digital twin to reconstitute the internal states that cannot be measured directly, such as concentration, temperature, stress, and damage fields. The FP-EMLP-parameterized multiphysics model has an aggregate relative field error of 4.8. Following the incorporation of physics-based reduced-order model, uncertainty-gated time residual learning, and online data assimilation, FP-EMLP-DT has a voltage RMSE of 15.8 mV and an error of 0.93%, 1.21% and 0.64 °C in SOC, SOH, and temperature respectively. It can be seen that physics-based modeling combined with data-driven residual correction leads to high prediction accuracy, understandable inner states, and consistent behavior in out-of-distribution conditions.

(3) Smart operational control that involved degradation of materials information was attained. The proposed degradation-aware MPC simultaneously optimizes power tracking, battery temperature, material stress, damage accumulation, and SOH loss. Compared with conventional MPC, the proposed approach minimizes the maximal stress by 16.5 percent and the normalized damage increment by 36.8 percent, whereas the charging time increases by 2.4 percent only and the energy efficiency remains at 93.4 percent. The model will have little error in SOC estimation in the presence of sensor noise and missing data and state estimation and control will take on average 37ms. Such results suggest that the proposed AI-powered digital twin can be highly promising in terms of online monitoring, degradation diagnosis and real-time battery management. The present study is limited by the absence of a systematic sensitivity calibration for the active-learning acquisition weights, the lack of edge-BMS and large-array deployment benchmarks, and validation centered on selected public battery datasets with LiFePO₄/FePO₄ as the principal material system. Future work will address acquisition-weight calibration, lightweight parallel deployment, and broader cross-chemistry validation.

Acknowledgements

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References

- [1] Semeraro C, Aljaghoub H, Abdelkareem MA, et al. Guidelines for designing a digital twin for Li-ion battery: A reference methodology[J]. *Energy*, 2023, 284: 128699.
- [2] Issa R, Badr MM, Shalash O, et al. A data-driven digital twin of electric vehicle Li-ion battery state-of-charge estimation enabled by driving behavior application programming interfaces[J]. *Batteries*, 2023, 9: 521.
- [3] Schmitt J, Horstkötter I, Bäker B. State-of-health estimation by virtual experiments using recurrent decoder–encoder based lithium-ion digital battery twins trained on unstructured battery data[J]. *Journal of Energy Storage*, 2023, 58: 106335.
- [4] Safavi V, Bazmohammadi N, Vasquez JC, et al. Battery state-of-health estimation: A step towards battery digital twins[J]. *Electronics*, 2024, 13: 587.
- [5] Fu K, Hamacher T, Perić VS. Development of self-adaptive digital twin for battery monitoring and management system[J]. *Electric Power Systems Research*, 2024, 234: 110698.
- [6] Lyu C, Xu S, Li J, et al. Digital twin modeling method for lithium-ion batteries based on data-mechanism fusion driving[J]. *Green Energy and Intelligent Transportation*, 2024, 3: 100162.
- [7] Li W, Li Y, Garg A, et al. Enhancing real-time degradation prediction of lithium-ion battery: A digital twin framework with CNN-LSTM-attention model[J]. *Energy*, 2024, 286: 129681.
- [8] Wang F, Zhai Z, Zhao Z, et al. Physics-informed neural network for lithium-ion battery degradation stable modeling and prognosis[J]. *Nature Communications*, 2024, 15: 4332.
- [9] Ji C, Jin G, Zhang R. State of charge estimation for lithium-ion batteries based on a digital twin hybrid model[J]. *Energy Reports*, 2025, 13: 2174-2185.
- [10] Salaorni D, Bianchi F, Colnago S, et al. A novel digital twin for battery energy storage systems in micro-grids[J]. *Journal of Energy Storage*, 2025, 132: 117745.
- [11] Safavi V, Mohammadi Vaniar A, Bazmohammadi N, et al. A battery degradation-aware energy management system for agricultural microgrids[J]. *Journal of Energy Storage*, 2025, 108: 115059.
- [12] Qasem M, Stubblefield J, Qandil M, et al. Real-time AI-enabled digital twin for battery health estimation and fast charging using partial-discharge data[J]. *eTransportation*, 2026, 27: 100528.
- [13] Deng B, Zhong P, Jun K, et al. CHGNet as a pretrained universal neural network potential for charge-informed atomistic modelling[J]. *Nature Machine Intelligence*, 2023, 5: 1031-1041.
- [14] Merchant A, Batzner S, Schoenholz SS, et al. Scaling deep learning for materials discovery[J]. *Nature*, 2023, 624: 80-85.
- [15] Frank JT, Unke OT, Müller KR, et al. A Euclidean transformer for fast and stable machine learned force fields[J]. *Nature Communications*, 2024, 15: 6539.
- [16] Zhang D, Bi H, Dai FZ, et al. Pretraining of attention-based deep learning potential model for molecular simulation[J]. *npj Computational Materials*, 2024, 10: 94.
- [17] Batatia I, Benner P, Chiang Y, et al. A foundation model for atomistic materials chemistry[J]. *Journal of Chemical Physics*, 2025, 163: 184110.
- [18] Radova M, Stark WG, Allen CS, et al. Fine-tuning foundation models of materials interatomic potentials with frozen transfer learning[J]. *npj Computational Materials*, 2025, 11: 237.
- [19] Mazitov A, Bigi F, Kellner M, et al. PET-MAD as a lightweight universal interatomic potential for advanced materials modeling[J]. *Nature Communications*, 2025, 16: 10653.
- [20] Loew A, Sun D, Wang HC, et al. Universal machine learning interatomic potentials are ready for phonons[J]. *npj Computational Materials*, 2025, 11: 178.
- [21] Kabylda A, Frank JT, Suárez-Dou S, et al. Molecular simulations with a pretrained neural network and universal pairwise force fields[J]. *Journal of the American Chemical Society*, 2025, 147: 33723-33734.
- [22] Wang R, Guo M, Gao Y, et al. A pre-trained deep potential model for sulfide solid electrolytes with broad coverage and high accuracy[J]. *npj Computational Materials*, 2025, 11: 266.
- [23] Lai G, Zhang R, Fang C, et al. Machine-learning-accelerated mechanistic exploration of interface modification in lithium metal anode[J]. *npj Computational Materials*, 2025, 11: 245.
- [24] Chen J, Fang M, Wu Q, et al. Insights into the atomic mechanism of lithium-ion diffusion in Li6PS5Cl via a machine learning potential[J]. *Chemistry of Materials*, 2025, 37: 591-599.
- [25] Ma P, Kumar R, Wang KH, et al. Active learning accelerates electrolyte solvent screening for anode-free lithium metal batteries[J]. *Nature Communications*, 2025, 16: 8396.
- [26] van der Oord C, Sachs M, Kovács DP, et al. Hyperactive learning for data-driven interatomic potentials[J]. *npj Computational Materials*, 2023, 9: 168.
- [27] Liu K, Wei Z, Gao W, et al. Heterogeneous ensemble enables a universal uncertainty metric for atomistic foundation models[J]. *npj Computational Materials*, 2026, 12: 34.
- [28] Kim K, Adelstein N, Dive A, et al. Probing degradation at solid-state battery interfaces using machine-learning interatomic potential[J]. *Energy Storage Materials*, 2024, 73: 103842.
- [29] Xu G, Jiang M, Li J, et al. Machine learning-accelerated discovery and design of electrode materials and electrolytes for lithium ion batteries[J]. *Energy Storage Materials*, 2024, 72: 103710.
- [30] Mortazavi B. Recent advances in machine learning-assisted multiscale design of energy materials[J]. *Advanced Energy Materials*, 2025, 15: 2403876.
- [31] Poul M, Huber L, Neugebauer J. Automated generation of structure datasets for machine learning potentials and alloys[J]. *npj Computational Materials*, 2025, 11: 174.
- [32] Kulichenko M, Barros K, Lubbers N, et al. Uncertainty-driven dynamics for active learning of interatomic potentials[J]. *Nature Computational Science*, 2023, 3: 230-239.
- [33] Perez D, Subramanyam A P A, Swinburne T D. Uncertainty quantification for misspecified machine learned interatomic potentials[J]. *npj Computational Materials*, 2025, 11: 263.
- [34] Qi J, Ko T W, Wood B C, et al. Robust training of machine learning interatomic potentials with dimensionality reduction and stratified sampling[J]. *npj Computational Materials*, 2024, 10: 43.
- [35] Zaverkin V, Holzmüller D, Christiansen H, et al. Uncertainty-biased molecular dynamics for learning uniformly accurate interatomic potentials[J]. *npj Computational Materials*, 2024, 10: 83.
- [36] Subramanyam A P A, Perez D. Information-entropy-driven generation of material-agnostic datasets for machine-learning interatomic potentials[J]. *npj Computational Materials*, 2025, 11: 218.
- [37] Shaidu Y, Pellegrini F, Küçükbenli E, et al. Incorporating long-range electrostatics in neural network potentials via variational charge equilibration from shortsighted ingredients[J]. *npj Computational Materials*, 2024, 10: 47.

- [38] Cheng B. Latent Ewald summation for machine learning of long-range interactions[J]. *npj Computational Materials*, 2025, 11: 80.
- [39] Bistri D, Di Leo C V. A continuum electro-chemo-mechanical gradient theory coupled with damage: Application to Li-metal filament growth in all-solid-state batteries[J]. *Journal of the Mechanics and Physics of Solids*, 2023, 174: 105252.
- [40] Taghikhani K, Huber W, Weddle P J, et al. Modeling coupled electro-chemo-mechanical phenomena within all-solid-state battery composite cathodes[J]. *Journal of the Mechanics and Physics of Solids*, 2025, 198: 106060.
- [41] Chen W X, Allen J M, Rezaei S, et al. Cohesive phase-field chemo-mechanical simulations of inter- and trans-granular fractures in polycrystalline NMC cathodes via image-based 3D reconstruction[J]. *Journal of Power Sources*, 2024, 596: 234054.
- [42] Gao X, Zhang Y, Liew K M. Electro-chemo-thermo-mechanical phase-field model for lithium penetration in solid electrolytes[J]. *International Journal of Mechanical Sciences*, 2025, 307: 110913.
- [43] Zeng Q, Rezaei S, Carrillo L, et al. Chemomechanical damage prediction from phase-field simulation video sequences using a deep-learning-based methodology[J]. *iScience*, 2024, 27(9): 110822.
- [44] Wang X, Li J, Wang R, et al. Lithium battery digital twin model with incremental learning[J]. *CHAIN*, 2024, 1(3): 249-262.
- [45] Zhao K, Liu Y, Zhou Y, et al. Digital twin-supported battery state estimation based on TCN-LSTM neural networks and transfer learning[J]. *CSEE Journal of Power and Energy Systems*, 2025, 11(2): 567-579.