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Posted Date: 19 September 2025

doi: 10.20944/preprints202509.0176.v2

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Article

Modeling Stress and Strain Energy in Battery Electrodes Under Cyclic Electrochemical Loading

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Abstract

The irreversible cyclic strain/stress in battery electrodes during ion intercalation/deintercalation drives mechanical energy dissipation, accelerating cycle life degradation. However, the lack of quantitative methods to assess stress and mechanical energy dissipation hinders a mechanistic understanding of mechanical behavior in electrochemical systems. This work aims to develop a theoretical framework to quantify stress and strain energy evolution in practical heterogeneous composite electrodes. Under assumptions of plane stress and elastic deformation, the average stress/strain energy per cycle can be derived for battery electrode during dynamic ion insertion/extraction. By considering a concentration-dependent modulus, the present framework allows for the simultaneous determination of both the bilayer stress and the apparent Young's modulus of the electrode through measurements of its curvature and intrinsic chemical strain.

Keywords: chemical stress; strain energy; chemical strain; composite electrode; ion insertion; modulus

Stress and strain energy evolution during battery cycling critically governs electrode degradation and performance decline. [1,2] During electrochemical cycling, ion concentration variations induce dimensional/volume changes in electrodes. These changes generate two distinct stress contributions: (1) diffusion-induced stresses arising from concentration gradients within active particles,[3,4] and (2) macroscopic electrode level stresses in film-substrate electrode systems caused by constraints from inactive cell components (e.g., current collectors) and geometric limitations. [2,5–9] These stresses trigger mechanical degradation through electrode fracturing, particle disintegration, and contact loss at interfaces, ultimately causing capacity fade and cell failure. [6,10–13] Meanwhile, the strain energy accumulated during deformation drives crack initiation and propagation as stored energy converts to surface energy during fracture. [14] Notably, cracks often originate at active/inactive material interfaces, emphasizing the necessity to study DIS and strain energy evolution at electrode levels.

Real-time observation techniques, such as in situ X-ray or electron-based microscopy, reveal atomic-scale deformation mechanisms but lack direct stress/strain quantification in practical porous composite electrodes composed of active particles, binders, and conductive agents. [15–17] Such investigations were made possible by the use of digital image correlation (DIC) or laser-based techniques employing Stoney's equation combined with chemo-mechanical modeling. [9,18–21] Although theoretical studies have addressed DIS and strain energy at particle scales, [14,22–25] electrode-level investigations remain limited. Bridging this gap is essential to mitigate fracture-related degradation and optimize next-generation batteries through stress-engineered electrode designs.

In this work, a theoretical framework was developed for estimating stress and strain energy generation during ion insertion/extraction in practical battery electrodes with current collector substrates. To simplify the analysis, we assumed a homogeneous ion concentration distribution within the electrode. The electrode's porous structure ensures that ion diffusion predominantly occurs across the ligaments of the porous network rather than through the bulk thickness, thereby

minimizing significant global concentration gradients. We further adopted a plane stress assumption and treated the deformation as purely elastic. Under plane stress and linear elasticity assumptions, the in-plane stress components σ_x and σ_y are expressed as:

$$\sigma_x = \frac{E}{1-\nu^2} (\epsilon_x + \nu\epsilon_y) \text{ and } \sigma_y = \frac{E}{1-\nu^2} (\epsilon_y + \nu\epsilon_x), \quad (1.1)$$

where E and ν are effective Young's modulus and effective Poisson ratio of the composite electrode, respectively. ϵ_x and ϵ_y are the strain components along the x - and y -directions. The shear stress τ_{xy} is given by:

$$\tau_{xy} = G\gamma_{xy}, \quad (1.2)$$

where $G = \frac{E}{2(1+\nu)}$ is effective shear modulus of the composite electrode, and γ_{xy} is shear strain. For isotropic battery electrodes ($\epsilon_x = \epsilon_y$ and $\tau_{xy} = 0$), σ_x and σ_y simplify to:

$$\sigma_x = \sigma_y = \frac{E}{1-\nu} \epsilon_x. \quad (1.3)$$

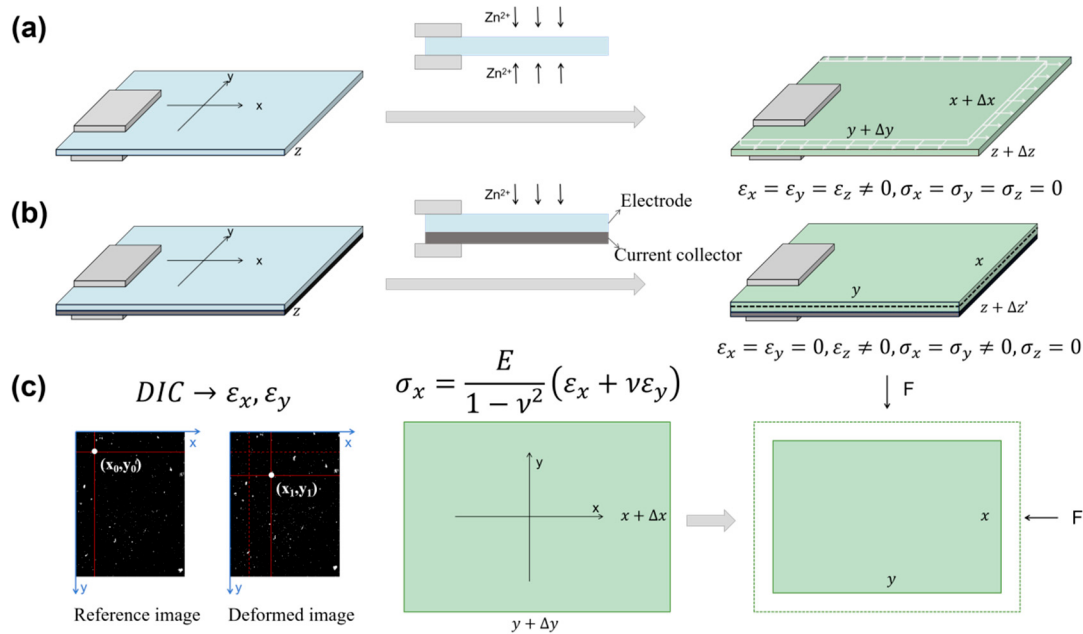


Figure 1. Illustration of (a) the chemical strain generation in freestanding electrode, (b) stress generation in practical electrode with current collector during ion insertion, and (c) the methodology to estimate stress in the practical electrode using the experimentally measured chemical strain.

In freestanding electrodes (Figure 1a), ion insertion induces free volumetric expansion, generating isotropic tensile strains ($\epsilon_x = \epsilon_y = \epsilon_z = \bar{\epsilon}_{xx}$) across all directions. This strain can be experimentally measured by the Digital-Image-Correlation (DIC)-based approach. In practical electrodes where the active layer is bonded to current collectors (Figure 1b), however, this expansion is mechanically constrained by the substrate (e.g., Cu foil) and battery casing, resulting in bilayer stresses in film-substrate electrodes. Without casing constraints, this bilayer would bend, which is the principle exploited in traditional laser curvature methods for stress measurement. Under such conditions, total strain (ϵ_x) comprises both bending strain and chemical strain (ϵ_x).

Scenario I. Film-Substrate Bilayer Electrodes under Cycling with a Constant Effective Young's Modulus and No Bending Deformation

The present study first examines film-substrate bilayer electrodes constrained within coin/pouch cells where the casing suppresses bending deformation. Consequently, the total strain equates solely

to chemical strain from ion insertion. Due to the thin-film-substrate geometry of the electrode, we assume uniform stress distribution along the thickness direction. If the bilayer electrode's initial pre-cycling state was used as reference, the in-plane strains (x, y directions), $\epsilon_{e/c}$, are approximately zero due to rigid current collector constraints, while the through-thickness strain (z -direction), ϵ_z , develops freely as the soft separator imposes negligible constraint, especially in pouch cells. This represents a reasonably valid approximation for systems with small volume change. It is also worth noting that local in-plane strains may not be strictly zero due to inhomogeneous deformation, which would affect the calculation of the spatial stress/strain energy distributions. For systems with small overall volume change, we consider in-plane strains negligible compared to freestanding electrode strains and thus exclude them from the current model.

To model these systems, we define the freestanding electrode at a given ion concentration as the reference state. The practical electrode (with identical ion content) is then conceptualized as the freestanding electrode compressed biaxially until its in-plane dimensions match those of the constrained system (Figure 1c). Assuming $\epsilon_{e/c} = 0$, the stress generated during ion insertion can be estimated by eq. (1.3), provided ϵ_x is measured as a function of ion concentration and stress-concentration coupling effects are neglected. For cases where $\epsilon_{e/c} \neq 0$, the effective strain ($\epsilon_{\text{eff}} = \epsilon_x - \epsilon_{e/c}$) must substitute ϵ_x in eq. (1.3), necessitating experimental determination of $\epsilon_{e/c}$.

Under a plane-stress state, the strain energy density (u_e) is given by:

$$u_e = \frac{1}{2} (\sigma_x \epsilon_x + \sigma_y \epsilon_y + \tau_{xy} \gamma_{xy}), \quad (1.4)$$

For isotropic conditions ($\epsilon_x = \epsilon_y$ and $\gamma_{xy} = 0$), this simplifies to:

$$u_e = \sigma_x \epsilon_x = \frac{E}{1-\nu} \epsilon_x^2, \quad (1.5)$$

The total strain energy (U) is then calculated by integrating u_e over the electrode volume V :

$$U = \int_V u_e dV = \int_V \frac{E}{1-\nu} \epsilon_x^2 dV, \quad (1.6)$$

where V is the electrode volume and is a function of ion concentration.

The volumetric strain (e_V) during the "compression" of the freestanding electrode is expressed as:

$$e_V = \frac{\Delta V}{V_0} = \epsilon_x + \epsilon_y + \epsilon_z, \quad (1.7)$$

Under plane stress ($\sigma_z = 0$), Hooke's law yields $\epsilon_z = -\frac{\nu}{E}(2\sigma_x)$. Then, we have:

$$e_V = \frac{\Delta V}{V_0} = \frac{4\nu-2}{\nu-1} \epsilon_x. \quad (1.8)$$

The electrode volume and its differential form are given by:

$$V = V_0 + \Delta V = V_0 + V_0 e_V = V_0 \frac{5\nu-3}{\nu-1} \epsilon_x, \quad (1.9)$$

and

$$dV = V_0 \frac{5\nu-3}{\nu-1} d\epsilon_x. \quad (1.10)$$

Substituting eq. (1.10) into eq. (1.6) gives the total strain energy U as follows:

$$U = \int_{\varepsilon_x} \frac{E}{1-\nu} \varepsilon_x^2 V_0 \frac{5\nu-3}{\nu-1} d\varepsilon_x = \frac{3-5\nu}{3(1-\nu)^2} E V_0 \varepsilon_x^3 + C. \quad (1.11)$$

The boundary condition $U = 0$ at $\varepsilon_x = 0$ necessitates $C = 0$, yielding:

$$U = \frac{3-5\nu}{3(1-\nu)^2} E V_0 \varepsilon_x^3. \quad (1.12)$$

By experimentally measuring ε_x as a function of ion concentration and determining E and ν , both stress and strain energy generated during cycling of the battery electrode can be quantified using eq. (1.3) and eq. (1.12).

To determine the effective Young's modulus (E) and Poisson's ratio (ν) of the composite electrode, we can assume ν remains constant throughout the analysis, and E is bounded using micromechanical homogenization frameworks. The actual electrode is a composite comprising active materials, conductive additive (Super P), and binder PVDF. We assume that the modulus of the three components is independent of the ion concentration. For the idealized non-porous binary composite comprising conductive additive Super P and binder PVDF, the lower bound of the effective bulk modulus ($K_{m,l}$) and effective shear modulus ($G_{m,l}$) can be calculated using the iso-stress (Reuss series) homogenization framework as follows: [26]

$$\frac{1}{K_{m,l}} = \frac{\phi_{m,PVDF}}{K_{PVDF}} + \frac{\phi_{m,Super P}}{K_{Super P}}, \quad (1.13a)$$

$$\frac{1}{G_{m,l}} = \frac{\phi_{m,PVDF}}{G_{PVDF}} + \frac{\phi_{m,Super P}}{G_{Super P}}. \quad (1.13b)$$

The upper bound of the effective bulk modulus ($K_{m,u}$) and effective shear modulus ($G_{m,u}$) can be calculated using the iso-strain (Voigt parallel) homogenization framework as follows: [27]

$$K_{m,u} = \phi_{m,PVDF} K_{PVDF} + \phi_{m,Super P} K_{Super P}, \quad (1.14a)$$

$$G_{m,u} = \phi_{m,PVDF} G_{PVDF} + \phi_{m,Super P} G_{Super P}, \quad (1.14b)$$

where $\phi_{m,Super P} = \frac{V_{Super P}}{V_{PVDF} + V_{Super P}}$ and $\phi_{m,PVDF} = \frac{V_{PVDF}}{V_{PVDF} + V_{Super P}}$ are the volume fractions of Super P and PVDF in the non-porous binary composite. $K_{Super P}$ and K_{PVDF} denote the bulk moduli of Super P and PVDF, respectively, while $G_{Super P}$ and G_{PVDF} represent their corresponding shear moduli. $V_{Super P}$ and V_{PVDF} are the volume of the Super P and PVDF, respectively.

To account for the inherent porosity of practical electrodes, the effective bulk modulus (K_{pm}) requires porosity-dependent corrections: [28,29]

$$K_{pm} = \left(\frac{1}{3(1-2\nu_{pm})} \right) \left(\frac{9K_m G_m}{3K_m + G_m} \right) \left(\frac{\rho_{pm}}{\rho_m} \right)^2, \quad (1.15)$$

where $\rho_m = \phi_{m,Super P} \cdot \rho_{Super P} + \phi_{m,PVDF} \cdot \rho_{PVDF}$, and $\rho_{pm} = \phi_{pm,Super P} \cdot \rho_{Super P} + \phi_{pm,PVDF} \cdot \rho_{PVDF}$, representing the densities of the fully dense and porous Super P/PVDF binary composite matrix. $\phi_{m,Super P}$ and $\phi_{m,PVDF}$ are the global volume fractions of Super P and PVDF within the fully dense matrix. $\phi_{pm,Super P}$ and $\phi_{pm,PVDF}$ are the global volume fractions of Super P and PVDF within the porous matrix. ν_{pm} corresponds to Poisson's ratio of the porous matrix. Substituting K_m and G_m in eq. (1.15) with $K_{m,l}$, $G_{m,l}$ or $K_{m,u}$, $G_{m,u}$ yields the lower bound and upper bound of the effective bulk modulus (K_{pm}) of porous Super P/PVDF binary composite matrix.

The effective bulk modulus K_e of the whole porous electrode is given by: [28]

$$K_e = \frac{K_{pm}(1+\phi_a\theta\Omega)}{1-\phi_a\varphi\Omega}, \quad (1.16)$$

where $\Omega = \frac{K_a - K_{pm}}{K_a + \theta K_{pm}}$, $\varphi = 1 + \frac{\phi_a \phi_{pm} (1 - \gamma \phi_{pm}) (K_a - K_{pm}) (\theta' - \theta)}{K_a + \theta' (\phi_a K_a + \phi_{pm} K_{pm})}$, $\theta' = \frac{2(1 - 2\nu_a) K_a}{(1 + \nu_a) K_{pm}}$, $\theta = \frac{2(1 - 2\nu_{pm})}{(1 + \nu_{pm})}$, $\gamma = \frac{2\lambda - 1}{\lambda}$, and $\lambda = \frac{2}{3}$. ϕ_a is the volumetric fraction of active material within the porous composite electrode. K_a and ν_a are bulk modulus and Poisson ratio of the active material. Then, the effective Young's modulus of the whole porous electrode can be calculated by:

$$E = \frac{K_e}{3(1 - 2\nu)}, \quad (1.17)$$

Using the above theoretical framework and in situ DIC-measured chemical strain data, the stress and strain energy in practical battery electrodes under operational cycling conditions can be calculated for a film-substrate bilayer electrode with a constant effective Young's modulus and without bending deformation.

Scenario II. Cycling of a Film-Substrate Bilayer Electrode under Bending with Concentration-Dependent Modulus

During cycling, volumetric changes in the electrode film are constrained by the substrate, resulting in a strain mismatch between the two layers. This mismatch generates stress, leading to bending deformation of the bilayer electrode and consequent changes in curvature if there is no constraints from the battery cases along the thickness direction.

To determine the stress and effective Young's modulus of the electrode film during cycling, we developed a mechanical model of the bilayer structure comprising the active electrode and current collector. This model represents the first coupled framework that integrates the evolution of active electrode thickness during cycling with in situ curvature measurements and chemical strain data. Similar to Scenario I, the model is founded on the assumptions of isotropic linear elasticity and an equi-biaxial stress state, and uniform distribution of ion concentration within the electrode. The active layer is perfectly bonded to the current collector, ensuring strain continuity across the interface. The interface between the current collector and the active layer is defined as the midplane of the system. A Cartesian coordinate system is established with its origin o located at the centroid of this midplane. The z -axis is normal to the electrode plane (along the thickness direction), while the x - and y -axes lie within the midplane, extending along the lateral and transverse directions, as illustrated in Figure 2.

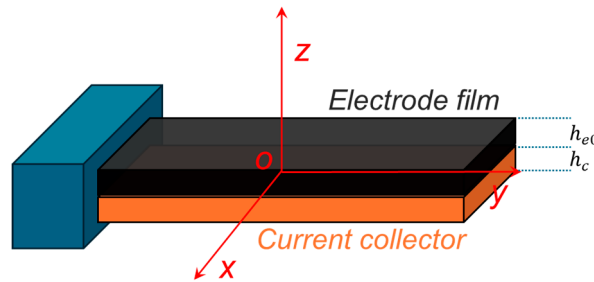


Figure 2. Schematic illustration of the bilayer electrode and establishment of the Cartesian coordinate system.

The thickness of the current collector is denoted as h_c , the initial active electrode thickness as h_{e0} , and the lithiated electrode thickness as h_e . Within classical beam theory, the in-plane strain in a bending electrode varies linearly through the thickness direction. The in-plane strains of the active electrode (ϵ_{xx}^e and ϵ_{yy}^e) and current collector (ϵ_{xx}^c and ϵ_{yy}^c) can be given by:

$$\begin{cases} \epsilon_{xx}^e = \epsilon_{yy}^e = \epsilon_0 + \kappa z - \epsilon_x^{\text{chem}}, & 0 \leq z \leq h_e \\ \epsilon_{xx}^c = \epsilon_{yy}^c = \epsilon_0 + \kappa z, & -h_c \leq z \leq 0 \end{cases} \quad (2.1)$$

z is the distance from the midplane, and ϵ_0 is the midplane strain at $z = 0$. κ is the curvature of the electrode. ϵ_x^{chem} is the intrinsic chemical strain of the active electrode ($\epsilon_x^{\text{chem}} = \bar{\epsilon}_{xx}$), which exists only in the active electrode portion. Given that the electrode's thickness is much smaller than

its in-plane dimensions, the electrode can be treated as being under a generalized plane stress state during electrochemical processes. Based on the generalized Hooke's law, the in-plane elastic stresses are derived as follows:

$$\begin{cases} \sigma_{xx}^e = \sigma_{yy}^e = \frac{E_e}{1 - \nu_e} (\varepsilon_0 + \kappa z - \varepsilon_x^{chem}), 0 \leq z \leq h_e \\ \sigma_{xx}^c = \sigma_{yy}^c = \frac{E_c}{1 - \nu_c} (\varepsilon_0 + \kappa z), -h_c \leq z \leq 0 \end{cases} \quad (2.2)$$

σ_{xx}^e , σ_{yy}^e , σ_{xx}^c and σ_{yy}^c are the in-plane elastic stresses of the active electrode and the current collector, respectively. E_e and E_c are the effective Young's modulus of the active electrode and current collector. E_c is a constant value, while E_e varies with the Li concentration. ν_e and ν_c are Poisson's ratios of the active electrode and the current collector. Due to the absence of mechanical constraint along the z -direction, lithiation leads to thickening of the electrode film in the bilayer structure.

To determine the thickness of the electrode film during cycling, we first consider the freestanding electrode as the reference, with an initial thickness denoted as h_{e0} . Upon lithiation at a given lithium ion concentration, the freestanding electrode expands freely in all three directions. Under the assumption of isotropy, it develops isotropic chemical strains, denoted as $\varepsilon_{xx} = \varepsilon_{yy} = \varepsilon_{zz} = \varepsilon_x^{chem}$. Thus, the thickness of the freestanding electrode after expansion can be expressed as

$$h_e = h_{e0}(1 + \varepsilon_x^{chem}) \quad (2.3)$$

When the electrode film is bonded to the current collector, its expansion is mechanically constrained by the substrate. This restriction induces biaxial stress in the film and causes the electrode to bend. In the following step, we use the freestanding electrode at lithiated state as the reference state. The bilayer electrode (with identical ion content) is then conceptualized as this freestanding electrode subjected to biaxial compression until its in-plane dimensions match those of the constrained system. Due to the Poisson effect, this compression leads to an increase in the thickness of the electrode film. The strain (ε_z^e) at each point through the thickness of the active electrode film arises primarily from the Poisson effect and can be expressed as:

$$\varepsilon_z^e = -\frac{\nu_e}{E_e} (\sigma_{xx}^e + \sigma_{yy}^e) = -\frac{2\nu_e}{E_e} \sigma_{xx}^e = -\frac{2\nu_e}{1 - \nu_e} (\varepsilon_0 + \kappa z - \varepsilon_x^{chem}), 0 \leq z \leq h_e \quad (2.4)$$

The average strain through the thickness direction ($\bar{\varepsilon}_z^e$) is obtained by integrating over the initial thickness and can be expressed as:

$$\bar{\varepsilon}_z^e = \frac{1}{h_{e0}} \int_0^{h_{e0}} \varepsilon_z^e dz = \frac{2\nu_e}{h_{e0}(\nu_e - 1)} \left(\varepsilon_0 h_{e0} + \frac{1}{2} \kappa h_{e0}^2 - \varepsilon_x^{chem} h_{e0} \right) \quad (2.5)$$

Then, the total thickness of the electrode film after lithiation can be expressed as:

$$h_e = h_{e0}(1 + \varepsilon_x^{chem})(1 + \bar{\varepsilon}_z^e) \quad (2.6)$$

From equations (2.3), (2.5), and (2.6), we can obtain:

$$h_e = h_{e0}(1 + \varepsilon_x^{chem}) \left[1 + \frac{2\nu_e}{h_{e0}(\nu_e - 1)} \left(\varepsilon_0 h_{e0} + \frac{1}{2} \kappa h_{e0}^2 - \varepsilon_x^{chem} h_{e0} \right) \right] \quad (2.7)$$

As the system is not subjected to any external forces, it must satisfy the conditions of both force and moment equilibrium: [30]

$$\int_{-h_c}^0 \sigma_{xx}^c dz + \int_0^{h_e} \sigma_{xx}^e dz = 0 \quad (2.8)$$

$$\int_{-h_c}^0 \sigma_{xx}^c z dz + \int_0^{h_e} \sigma_{xx}^e z dz = 0 \quad (2.9)$$

Substituting equation (2.2) into equations (2.8-2.9), we have:

$$\frac{E_c}{1-\nu_c} \left(\epsilon_0 h_c - \frac{1}{2} \kappa h_c^2 \right) + \frac{E_e}{1-\nu_e} \left(\epsilon_0 h_e + \frac{1}{2} \kappa h_e^2 - \epsilon_x^{\text{chem}} h_e \right) = 0 \quad (2.10)$$

$$\frac{E_c}{1-\nu_c} \left(\frac{1}{3} \kappa h_c^3 - \frac{1}{2} \epsilon_0 h_c^2 \right) + \frac{E_e}{1-\nu_e} \left(\frac{1}{2} \epsilon_0 h_e^2 + \frac{1}{3} \kappa h_e^3 - \frac{1}{2} \epsilon_x^{\text{chem}} h_e^2 \right) = 0 \quad (2.11)$$

By combining equations (2.7), (2.10), and (2.11), the electrode thickness, midplane stress in the active electrode ($\sigma_{xx,z=0}^e$), and the Young's modulus of the electrode can be determined as functions of Li concentration during cycling.

Summary

In conclusion, a theoretical framework was developed to quantify stress and strain energy evolution in practical battery electrodes with current collectors during ion insertion/extraction cycles. The model integrates in situ strain data from freestanding electrodes, measured via DIC, under plane stress conditions and linear elastic assumptions. When the electrode modulus was assumed constant, iso-stress (Reuss) and iso-strain (Voigt) homogenization models were used, along with porosity-dependent corrections, to estimate bounds for the effective Young's modulus of the composite electrodes. Conversely, when the modulus was treated as concentration-dependent, both the stress and the apparent Young's modulus could be determined simultaneously by measuring the electrode curvature and the intrinsic chemical strain. This work establishes a foundational methodology for assessing stress evolution and mechanical energy dissipation during practical battery cycling.

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