



Evaluation of Order Reduction Techniques for Porous Electrode Diffusion Equation in Lithium Ion Model

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Abstract

Models for lithium ion batteries based on electrochemical thermal principles approximate electrodes with spheres. Ion concentration in the spheres is described using Fick's second law with partial differential equations (PDE), which can be solved numerically. The model calculation time, especially the electrode ion concentration part, should be reduced as less as possible for real time control purposes. Several mathematical methods have been proposed to reduce the complexity of PDE in electrode particles which include polynomial approximation, proper orthogonal decomposition (POD), Padé approximation, Galerkin reformulation and etc. These methods are compared to each other with different input current density. Then, selected method is further integrated into a reduced order model (ROM) for a complete battery that considers Li ion concentration, potentials in electrode and electrolyte. Evaluation of simulation results reveal that the 3rd order Padé approximation serves as a better computationally efficient replacement for the diffusion equation in lithium ion battery model.

Introduction

A model based on electrochemical principle is widely used to simulate performance of lithium ion batteries [1]. The setup for the model is composed of one separator sandwiched by two composite electrodes that represent a mixture between solid electrode and electrolyte. The electrodes are made of porous particles that are modeled as spheres. In order to obtain the complete responses of a cell, the governing equation for lithium ion transport in electrode particles described as Fick's second law of diffusion need to be solved.

$$\frac{\partial C_s(r,t)}{\partial t} = D_s \frac{1}{r^2} \frac{\partial}{\partial r} \left[r^2 \frac{\partial C_s(r,t)}{\partial r} \right] \quad (1)$$

Boundary conditions:

$$\begin{aligned} \frac{\partial C_s}{\partial r} \Big|_{r=0} &= 0 \\ D_s \frac{\partial C_s}{\partial r} \Big|_{r=R_s} &= -\frac{j^{Li}}{a_s F} \end{aligned} \quad (2)$$

Numerical solution of the diffusion partial differential equation involves discretization in both the electrode thickness direction δ and the particle radial direction r . When it is assumed that the discretization in thickness direction produces M node points and the radial direction does N node points, the total states related to electrode ion concentration is about $(2/3)M*N$ for the case that the two composite electrodes occupy about $2/3$ of the thickness of a cell. For a transient calculation, the calculation time will be increasing exponentially, which impede the real time application of the model.

Therefore, simplifying the calculation of the diffusion equation in r direction becomes a crucial issue in battery control and application area. In this work, four popular model order reduction techniques are compared to each other with respect to different input current profiles as well as different model order choice. Discussion of the advantages and disadvantages of them would be useful information for choosing simplification method in lithium ion model reduction work.

1. Overview of Model Reduction Techniques

Duhamel's superposition method was the first approximation to the diffusion equation [1], but the computation time was not short enough to meet the real time requirement though the accuracy was acceptable. Wang proposed the diffusion length method [2], whose dynamic responses are not as satisfied as its steady state performances. Liu applied pseudo steady state (PSS) method to solve the diffusion equation analytically [5]. However, the large amount of coefficients presents a drawback

for simulation [3]. K. Smith used the finite element method with unequal node points in radial direction [6] while the selection of node point cannot be optimized.

Other than those method discussed above, polynomial approximation by Subramanian [4], proper orthogonal decomposition (POD) by Cai [7], Padé approximation by Forman [9] and Galerkin reformulation by Ramadesigan [8] will be compared in details in the following work.

Table 1. Parameters and Nomenclature

Faraday's constant, F (C/mol)	96487
Interfacial surface area, a_s (cm ²)	$3^* \epsilon_s / R_s$
Temperature, T (K)	298.15
Negative electrode thickness, δ_N (cm)	$52 * 10^{-4}$
Particle radius, R_s (cm)	$1 * 10^{-4}$
Electrode plate area, A (cm ²)	25530
Active material volume fraction, ϵ_s	0.51
Maximum solid phase concentration, $c_{s, \max}$ (mol*cm ⁻³)	0.0446
Stoichiometry at 0% SOC: Stoi_0	0.2249
Stoichiometry at 100% SOC: Stoi_{100}	0.8550
Initial concentration, C_0 (mol*cm ⁻³)	$1.2 * 10^{-3}$
Solid phase Li ⁺ diffusion coefficient, D_s	$2 * 10^{-12}$
Volume averaged Li ⁺ concentration inside particles, $C_{s, \text{ave}}$	
Li ⁺ concentration at the surface of the solid particles, $C_{s, \text{surf}}$	
Proper orthogonal modes in POD, Φ_i	
Eigenvalues of the diffusion equation, λ_n	

1.1. 3rd Order Polynomial Approximation

The polynomial approximation is developed by assuming that the solid-state concentration inside the spherical particle can be expressed as a polynomial in the radial direction.

$$C_s(r, t) = a(t) + b(t) \left(\frac{r^2}{R_s^2} \right) + d(t) \left(\frac{r^4}{R_s^4} \right) \quad (3)$$

The time dependent coefficients $a(t)$, $b(t)$, $d(t)$ can be described by volume-average concentration, $C_{s, \text{ave}}$, volume-average concentration flux, q_{ave} and surface concentration, $C_{s, \text{surf}}$

$$\begin{aligned} a(t) &= \frac{39}{4} C_{s, \text{surf}} - 3q_{\text{ave}} R_s - \frac{35}{4} C_{s, \text{ave}} \\ b(t) &= -35 C_{s, \text{surf}} + 10q_{\text{ave}} R_s + 35 C_{s, \text{ave}} \\ d(t) &= \frac{105}{4} C_{s, \text{surf}} - 7q_{\text{ave}} R_s - \frac{105}{4} C_{s, \text{ave}} \end{aligned} \quad (4)$$

where the volume-average concentration, volume-average concentration flux and surface concentration are defines as:

$$\begin{aligned} C_{s, \text{ave}} &= \int_{r=0}^{R_s} \frac{3r^2}{R_s^3} C_s(r, t) dr \\ q_{\text{ave}} &= \int_{r=0}^{R_s} \frac{3r^2}{R_s^3} \left(\frac{\partial}{\partial r} C_s(r, t) \right) dr \\ C_{s, \text{surf}} &= C_s(R_s, t) \end{aligned} \quad (5)$$

Substituting the polynomial approximation to the diffusion equation and boundary conditions yields the following expressions for $C_{s, \text{ave}}$, q_{ave} and $C_{s, \text{surf}}$

$$\begin{aligned} \frac{d}{dt} C_{s, \text{ave}} + 3 \frac{j^{Li}}{R_s a_s F} &= 0 \\ \frac{d}{dt} q_{\text{ave}} + 30 \frac{D_s}{R_s^2} q_{\text{ave}} + \frac{45}{2} \frac{j^{Li}}{R_s^2} &= 0 \\ 35 \frac{D_s}{R_s} (C_{s, \text{surf}} - C_{s, \text{ave}}) - 8D_s q_{\text{ave}} &= -\frac{j^{Li}}{a_s F} \end{aligned} \quad (6)$$

1.2. Proper Orthogonal Decomposition (POD)

POD is a procedure to find a basis for a modal decomposition of a data set. The data applied here is the full order model solution of the diffusion equation $C_{s, \text{full}}$, which is obtained by discretization of the Fick's law in both thickness and radial direction.

$$\dot{C}_{s, \text{full}} = A \cdot C_{s, \text{full}} + b \cdot j^{Li} \quad (7)$$

Approximation of the full order variables are the linear combination of the first N proper orthogonal modes (POMs) $\Phi_i(t)$, where N is the model order we used for POD.

$$\begin{aligned} [U \ \Sigma \ V] &= \text{svd}(C_{s, \text{full}}^T \cdot C_{s, \text{full}}) \\ \Phi &= U(:, 1 : N) \end{aligned} \quad (8)$$

The linear variables are derived by substituting the POMs to the full order model that is stored along with concentration data profile before the order reduction procedure.

$$\begin{aligned} \Phi \cdot \dot{a} &= A \cdot \Phi \cdot a + b \cdot j^{Li} \\ \dot{a} &= \Phi^T A \Phi \cdot a + \Phi^T b \cdot j^{Li} \end{aligned} \quad (9)$$

Then the concentration profile can be reconstructed by linear combination of the reduced variables and POMs.

$$C_{s, \text{reduced}}(t) = \Phi \cdot a(t) \quad (10)$$

The average concentration can be evaluated by volume average the data profile while the surface concentration can be obtained directly from the end column of the data profile.

1.3. Padé Approximation

Padé approximant is a rational polynomial approximation to a function centered about a point. The order of the numerator and denominator can be chosen arbitrarily.

$$P(s) = \frac{\sum_{k=0}^{Q-1} a_k s^k}{\sum_{i=1}^Q b_i s^i}, \quad b_1 = 1 \quad (11)$$

The coefficients $a_k(t)$ and b_i can be evaluated by comparing the derivatives of the original function and the approximation at $s=0$. The function used here is the transfer function between the input current density and the output average concentration or surface concentration.

$$\begin{aligned} \frac{C_{s,ave}(s)}{j^{Li}(s)} &= \frac{\int_0^{R_s} C_s(s,r)(4\pi r^2 dr)}{(4/3)\pi R_s^3} = -\frac{3D_s m}{R_s s} \\ \frac{C_{s,surf}(s)}{j^{Li}(s)} &= -\frac{(e^{2R_s\sqrt{s/D_s}} - 1)mR_s}{1 + R_s\sqrt{\frac{s}{D_s}} + e^{2R_s\sqrt{s/D_s}}\left(R_s\sqrt{\frac{s}{D_s}} - 1\right)} \\ m &= \frac{1}{D_s a_s F} \end{aligned} \quad (12)$$

The surface concentration in Padé approximation form becomes:

$$\frac{C_{s,surf}(s)}{U(s)} = \frac{a_0 + a_1 s + a_2 s^2 + \dots + a_{Q-1} s^{Q-1}}{1 + b_2 s + b_3 s^2 + \dots + b_Q s^{Q-1}}. \quad (13)$$

Then rewrite the surface concentration along with average concentration in state space form.

$$\begin{aligned} \dot{x} &= \begin{bmatrix} 0 & 1 & 0 & 0 & \dots & 0 \\ 0 & 0 & 1 & 0 & \dots & 0 \\ 0 & 0 & 0 & 1 & & 0 \\ \vdots & \vdots & \vdots & & \ddots & \vdots \\ 0 & 0 & 0 & 0 & 0 & 1 \\ 0 & -\frac{1}{b_Q} & -\frac{b_2}{b_Q} & -\frac{b_3}{b_Q} & \dots & -\frac{b_{Q-1}}{b_Q} \end{bmatrix} x + \begin{bmatrix} 0 \\ 0 \\ 0 \\ \vdots \\ 0 \\ \frac{1}{b_Q} \end{bmatrix} u \\ y &= \begin{bmatrix} C_{s,surf} \\ C_{s,ave} \end{bmatrix} x = \begin{bmatrix} \frac{a_0}{b_Q} & \frac{a_1}{b_Q} & \frac{a_2}{b_Q} & \dots & \frac{a_{Q-1}}{b_Q} \\ \frac{a_0 \cdot b_1}{b_Q} & \frac{a_0 \cdot b_2}{b_Q} & \frac{a_0 \cdot b_3}{b_Q} & \dots & \frac{a_0 \cdot b_Q}{b_Q} \end{bmatrix} \end{aligned} \quad (14)$$

The initial state is obtained by setting $C_{s,surf} = C_{s,ave} = C_{s,0}$.

$$x_0 = \begin{bmatrix} \frac{C_{s,0} \cdot b_Q}{a_0} & 0 & 0 & \dots & 0 \end{bmatrix}^T \quad (15)$$

Table 2. Padé approximation coefficients of $C_{s,surf}$.

Order	Coefficients
2nd	Num: $[-3D_s^*m/R_s, -2/7^*m^*R_s]$
	Den: $[1, R_s^2/35/D_s]$
3rd	Num: $[-3D_s^*m/R_s, -4/11^*m^*R_s, -m^*R_s^3/165/D_s]$
	Den: $[1, 3/55^*R_s^2/D_s, R_s^4/3465D_s^2]$
4th	Num: $[-3D_s^*m/R_s, -2/5^*m^*R_s, -2/195^*m^*R_s^3/D_s, -4/75075^*m^*R_s^5/D_s^2]$
	Denominator: $[1, 1/15^*R_s^2/D_s, 2/2275^*R_s^4/D_s^2, 1/675675^*R_s^6/D_s^3]$
$m=1/D_s/a_s/F$	

1.4. Galerkin Reformulation

This method is based on eigenfunctions derived by Galerkin collocation. It also approximates the diffusion equation by a polynomial expression as follows:

$$C_s(x, \tau) = a(\tau) + b(\tau)x^2 + \sum_{n=1}^{\infty} \frac{d_n(\tau) \sin(\lambda_n x)}{x} \quad (16)$$

Where x and r are dimensionless variables corresponding to radial r and time t . λ_n are the eigenvalues of the diffusion equation. The coefficients $a(\tau)$ and $b(\tau)$ are obtained from evaluating the average concentration by the polynomial expression.

$$\begin{aligned} a(\tau) &= C_{s,ave}(\tau) + \frac{3\delta(\tau)}{10} \\ b(\tau) &= -\frac{1}{2}\delta(\tau) \end{aligned} \quad (17)$$

Where δ is dimensionless input current density. The coefficient $d_n(\tau)$ is solved by introducing an intermediate variable Q_n .

$$d_n(\tau) = \frac{d}{dt} Q_n(\tau) \quad (18)$$

The two interested variables, average concentration and surface concentration are expressed as follows.

$$\begin{aligned} \frac{dC_{ave}}{d\tau} &= -3\delta(\tau) \\ C_{surf} &= C_{ave} - \frac{\delta(\tau)}{5} + 2\delta(\tau) \sum_{n=1}^m \frac{1}{\lambda_n^2} - \sum_{n=1}^m Q_n \lambda_n^2 \sin(\lambda_n) \\ \frac{dQ_n}{d\tau} + \lambda_n^2 Q_n &= 2 \frac{\delta(\tau)}{\lambda_n^2 \sin(\lambda_n)} \\ \lambda_n &= \tan(\lambda_n) \end{aligned} \quad (19)$$

2. Comparison Analysis

Before integrating these methods into a complete reduced order model for batteries, we compared the concentration responses of them corresponding to different input current density j^{Li} and different model order number N . The simulation results of the first part show the steady state and dynamic

performances of the approximations when the battery were operated at different input conditions while the second part discuss the effects of model order number to the accuracy of the methods. Simulation results of each method are compared to the full order model results, which were calculated by finite volume method (FVM) with 25 grid points in the radial direction. This full order model results are considered as exact solution to the diffusion equation in this work.

Other than concentration distribution along radial direction in a solid particle, people have more interests on average concentration $C_{s,ave}$ and surface concentration $C_{s,surf}$ of the particle since they represent two key variables in battery application, state of charge (SOC) and equilibrium potential (U_{equ}).

$$SOC = \frac{1}{L_-} \int_0^{L_-} \frac{(C_{s,ave} - C_{s,max} * Stoi_0)}{C_{s,max} * (Stoi_{100} - Stoi_0)} dx$$

$$U_{equ} = f(C_{s,surf} / C_{s,max}) \quad (20)$$

$Stoi_0$ and $Stoi_{100}$ are stoichiometry number at 0% SOC and 100% SOC respectively.

Average concentration in electrode particles is evaluated based on mass balance principles that applied to all these methods. Therefore, no big differences appeared in the simulation results for these approximations. However, surface concentrations display big discrepancy to each other. They are compared with exact solution in the following figures.

The parameters used in the simulation are based on carbon anode and $LiMn_2O_4$ cathode materials and the ambient temperature is 25°C. All methods were coded in Matlab running under Windows with Intel Core i7-2600 CPU at 3.4GHz.

2.1. Comparison of Different Input Current Density j^{Li}

The following comparisons in this part are simulated using 3rd order model for each method. They are comparable to the 3rd order polynomial approximation introduced before. Average current density j^{Li} applied here is calculated by dividing the input current (10A) to electrode volume for a certain grid point in thickness direction.

Figure 1 is the surface concentration response of constant discharge current and rest input profile. The plot of errors reveals that there are overshoot at the beginning of discharge and rest part of the four models. This is due to the response time of the methods. Apparently, the green circle-dash curve and the blue dash-dash curve that represents Padé

approximant and Galerkin reformulation have relatively less error in these two regions, which indicate faster response speed of the models to the changing of input conditions. The errors of the middle part of the constant current discharge are tolerable, which means these four methods all have acceptable steady state performances.

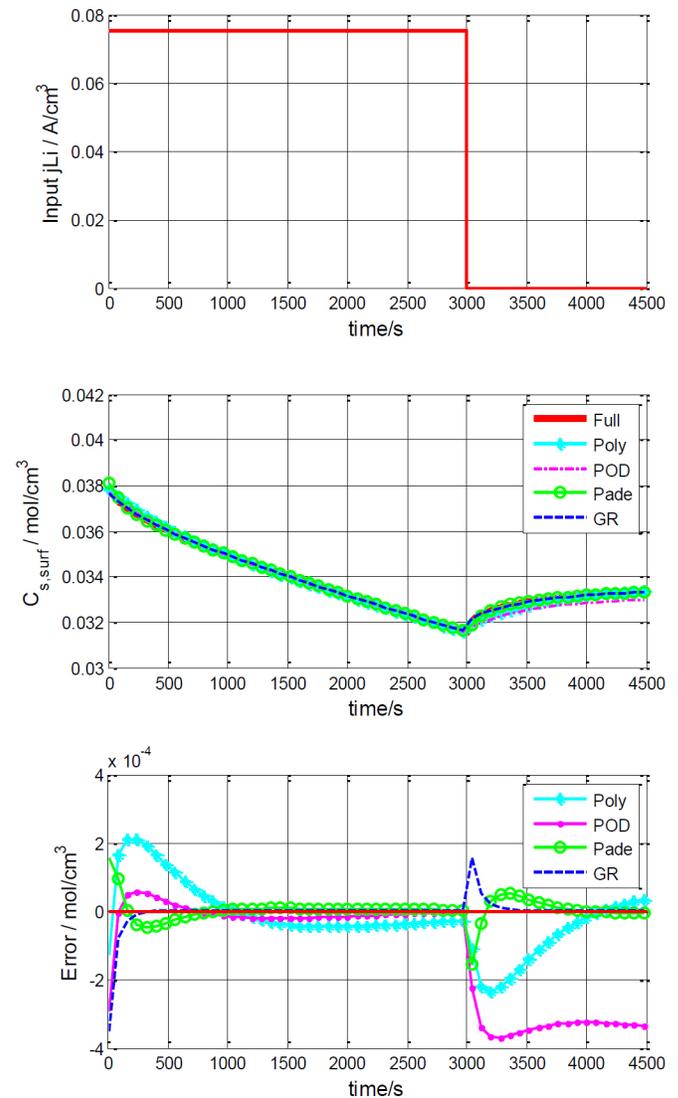


Figure 1. Surface concentration responses with constant current-rest input profile. Star-line: polynomial approximation; dot-dash: POD; circle-line: Padé approximant; dash-dash: Galerkin reformulation.

More dynamic responses are plotted in Figure 2 and Figure 3. Figure 2 is the surface concentration responses corresponding to constant current discharge, rest, constant current charge input profile, which represents pulse operating condition. Compared to Figure 1, this input profile requires faster response speed of the models to follow the frequent changing of the input command. The 3rd order POD with purple dot-dash

line has more errors compared to other three methods. This is due to the discrepancy of the input current profile to the full order model when calculating the reference concentration data, and the input current profile to the simulation here.

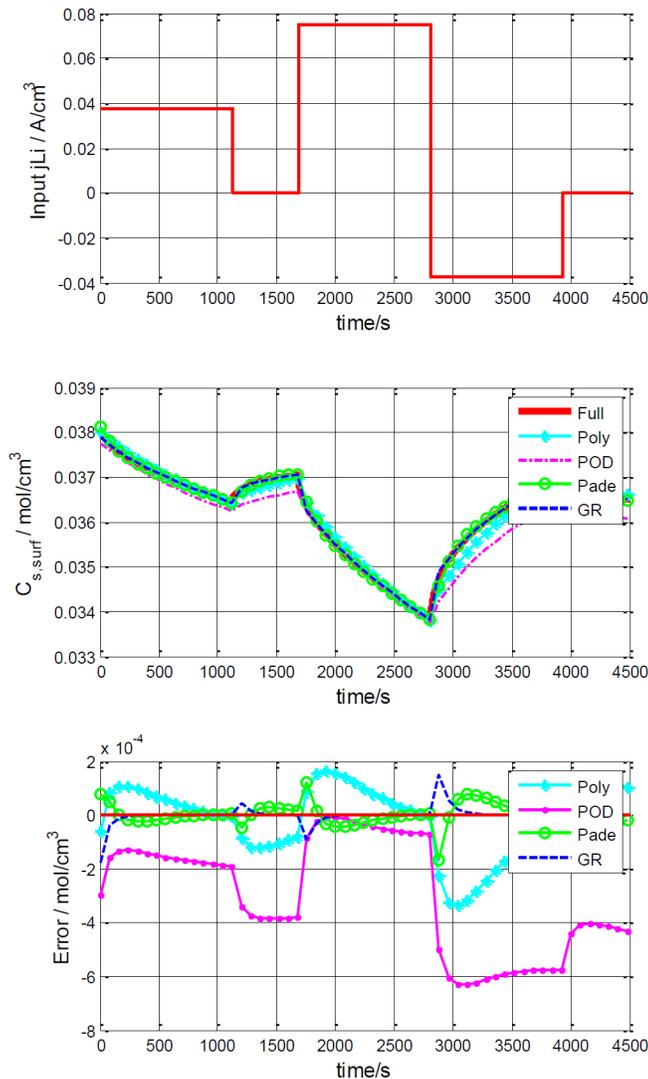


Figure 2. Surface concentration responses with pulse current input profile. Star-line: polynomial approximation; dot-dash: POD; circle-line: Padé approximant; dash-dash: Galerkin reformulation.

Figure 3 shows the comparison with sinusoid input current profile. This input profile is a pure discharging-charging procedure without resting period. The loading are changing continuously but more frequently compared to the previous two inputs. The plots show that the dynamic response characteristics of Padé approximant and Galerkin reformulation are relatively better than polynomial approximation and POD of the model order three. The results demonstrate that Padé and Galerkin principle based approximation can better representing the dynamic behavior of the diffusion equation described in equation (1).

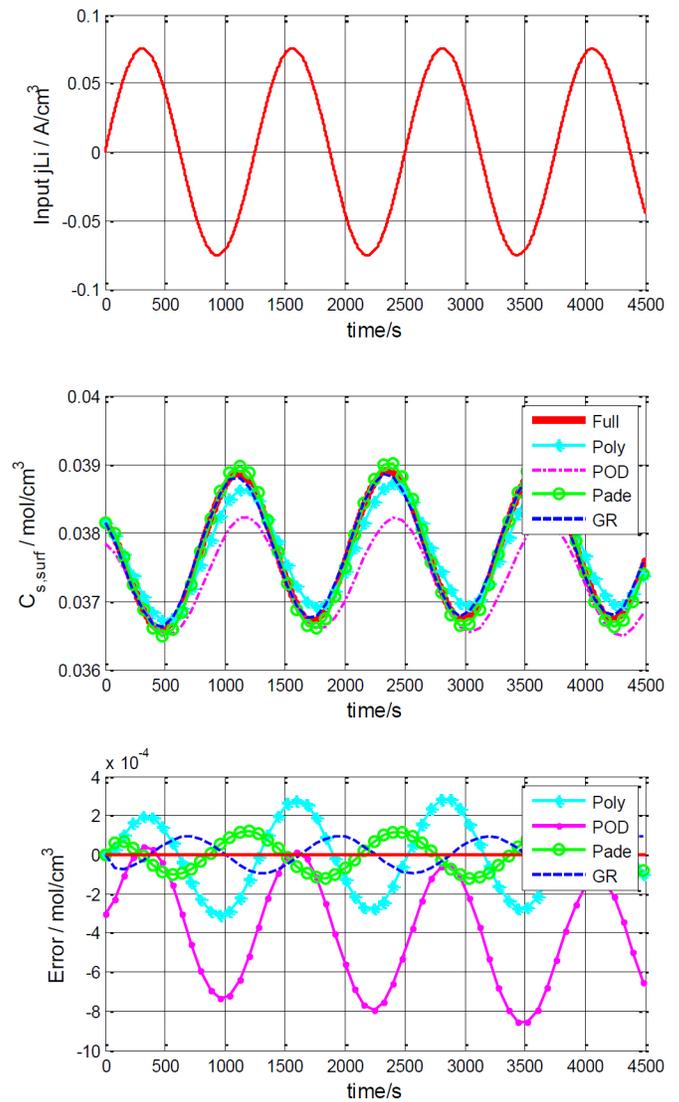


Figure 3. Surface concentration responses with sine current input profile. Star-line: polynomial approximation; dot-dash: POD; circle-line: Padé approximant; dash-dash: Galerkin reformulation.

2.2. Comparison of Different Model Order Number N

Figure 4 and Figure 5 display the effects of model order number to the accuracy of the calculation. Figure 4 is the simulation results of 2nd order POD, Padé approximant and Galerkin along with the original 3rd order polynomial while Figure 5 shows the results of the 5th order. Pulse current input plotted in Figure 2 is applied in this set of comparison. Since it is unable to increase model order number of polynomial approximation automatically in the same expression, only the original 3rd order polynomial shown in equation (6) is compared in this part. Other three methods can be set a desired model order once the algorithm has built.

Significant deviation of 2nd order POD and Padé approximant to the exact solution are observed in the 2nd order model results plotted in Figure 4. However, when the model order number increases to five, errors of these two methods drop dramatically. Especially the Padé approximant that obtains the highest accuracy compared to other three methods shown in Figure 5. This phenomenon illustrates that the dependency of approximation to the number of representing terms of the methods is relatively high for Padé and POD. The feature of these two methods, which is representing the problem using the first several dominant terms, is the main reason of the effects shown here.

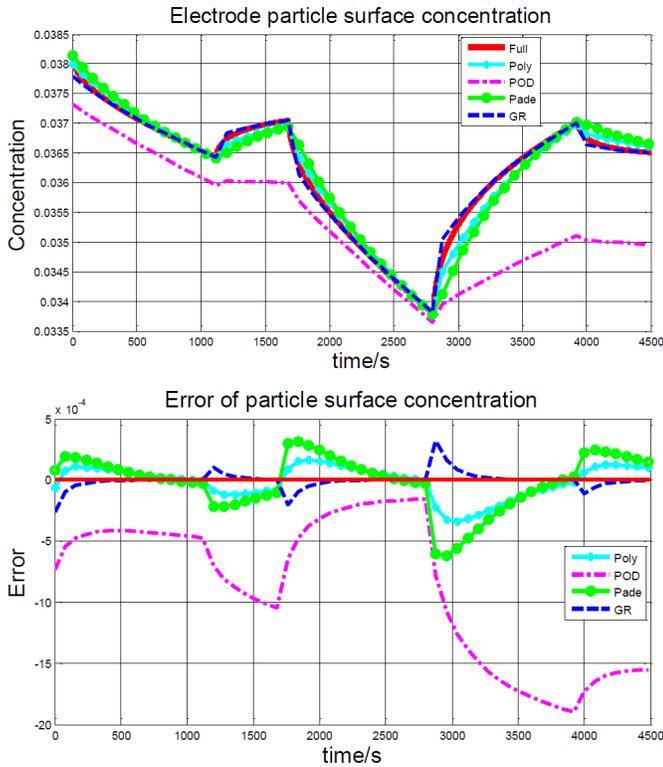


Figure 4. Surface concentration responses of 2nd order reduced order techniques. Star-line: polynomial approximation; dot-dash: POD; circle-line: Padé approximant; dash-dash: Galerkin reformulation.

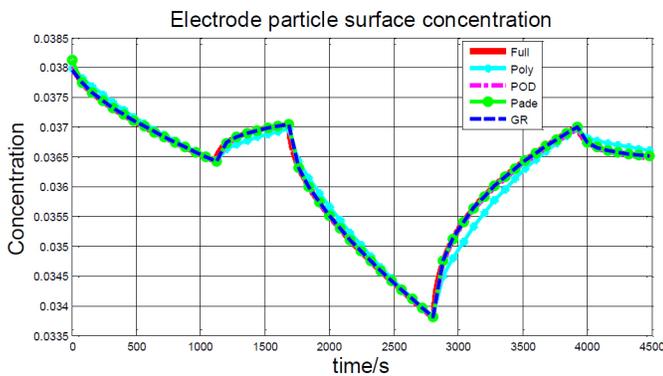


Figure 5.

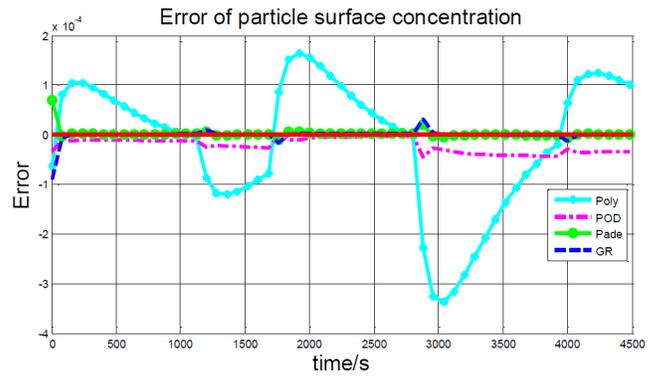


Figure 5. (cont.) Surface concentration responses of 5th order reduced order techniques. Star-line: polynomial approximation; dot-dash: POD; circle-line: Padé approximant; dash-dash: Galerkin reformulation.

2.3. Computational Time and Error Analysis

Computational time and errors with respect to different model order numbers are shown in Figure 6. Pulse current input is used in this simulation and the errors are norm values at each model order numbers.

Other than polynomial approach, it is observed that the errors of POD, Padé approximant and Galerkin reformulation converge to zero while the model order number is increasing as well as the simulation time. Galerkin reformulation shows the best accuracy but the computational time is the longest. POD cannot have satisfied accuracy until the model order number up to five. Padé approximant has a comparatively better performance when both the simulation speed and accuracy are considered. As shown in Figure 6, error of the 3rd order Padé approximant converges to a tolerable region while the simulation time is relatively short. Performance of the POD method can be further improved by applying various input current profiles to build the full order model solution but it takes much more simulation time.

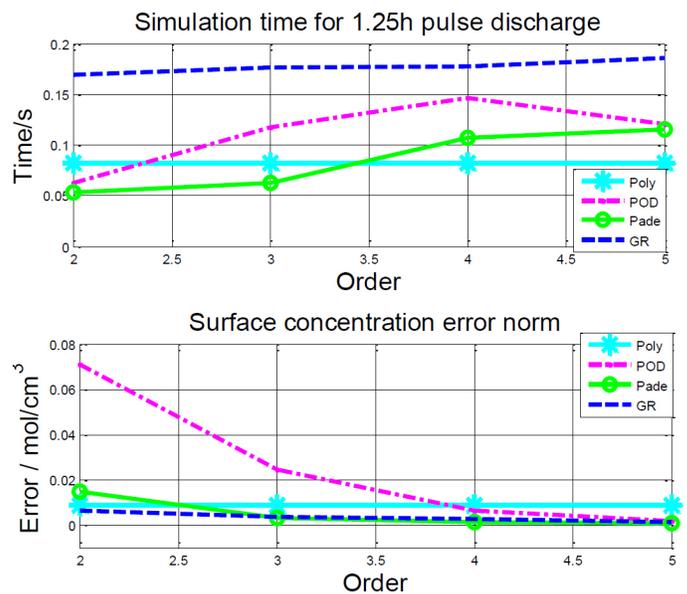


Figure 6. Computational time and error with pulse current input profile. Star-line: polynomial approximation; dot-dash: POD; circle-line: Padé approximant; dash-dash: Galerkin reformulation.

3. Electrode Particle Model in Battery Model

Electrochemical principle based lithium ion battery model is governed by four PDEs describing Li ion concentrations and cell potentials in electrode and electrolyte. The four variables $C_{s,surf}$, C_e , Φ_s and Φ_e are coupled by the Butler-Volmer kinetic expression. Linearization of B-V equation, applying constant conductivities and neglecting the effects of electrolyte concentration to the charge conservation simplify the model to the following form:

$$\begin{aligned}\frac{\partial C_s(r,t)}{\partial t} &= D_s \frac{1}{r^2} \frac{\partial}{\partial r} \left[r^2 \frac{\partial C_s(r,t)}{\partial r} \right] \\ \frac{\partial(\varepsilon_e c_e)}{\partial t} &= \frac{\partial}{\partial x} \left(D_e^{eff} \frac{\partial c_e}{\partial x} \right) + \frac{1-t_+^0}{F} j^{Li} \\ \frac{\partial}{\partial x} \left(\frac{\partial \phi_{se}}{\partial x} \right) &= j^{Li} \left(\frac{1}{\sigma^{eff}} + \frac{1}{\kappa^{eff}} \right) \\ j^{Li} &= \frac{a_s i_0 F}{RT} (\phi_{se} - U).\end{aligned}\tag{21}$$

Calculation of electrolyte concentration C_e can be further simplified by applying residue grouping method [10].

The optimized order reduction technique for diffusion equation in electrode particles is integrated with other parts described in [equation \(21\)](#) to estimate the battery states such as SOC and terminal voltage. The performances of the model compared to experimental data will be shown in future work.

Summary

Model order reduction techniques are often used to simplify the calculation of diffusion equation for porous electrode. Polynomial approximation, Padé approximant, POD and Galerkin reformulation are the four representing methods. The methods are evaluated by responses of different current input profiles and different model order numbers. Simulation results show that only high order POD is adequate to represent the transient behavior of the concentration distribution in electrode particles. Galerkin reformulation has high accuracy even at low model order, but the computational time is extensive. 3rd order polynomial has acceptable speed and accuracy, but model upgrade will increase the complexity of the algorithm. The comparison work indicates that the 3rd order Padé approximant could be the first choice for ROM of lithium ion batteries needed for real time application.

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