

MODEL ORDER REDUCTION OF ELECTROCHEMICAL BATTERIES USING GALERKIN METHOD

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ABSTRACT

This paper presents a model order reduction (MOR) method for modeling and estimation of a first-principles electrochemical Lithium-ion battery. The MOR approach combines the Galerkin method with coordinate transformation and is applied to solve the spherical diffusion problem with non-zero flux boundary conditions. The order of the reduced-order model (ROM) is carefully selected based on analysis in the frequency domain. With the reduced-order diffusion model, an enhanced single particle model which incorporates the electrolyte dynamics is developed and validated against the experimental data.

INTRODUCTION

Lithium-ion batteries are considered as the state of the art for energy storage solution in electric and hybrid vehicles. A lingering concern in the Automotive Industry lies in the degradation of the capacity and internal resistance due to usage and calendar life, which impacts reliability and warranty costs [1, 2]. Understanding and predicting the chemical and physical processes leading to aging in Li-ion cells is possible through multiscale characterization methods and first-principles electrochemical models [1, 3-5]. However, "in-situ" quantification of such processes on a vehicle is not yet achievable due to the absence of direct measurements. Hence, control-oriented modeling and model-based estimation are indispensable tools to monitor the operation of Li-ion battery packs, using the measured current and output voltage. The estimation of battery State of Charge (SOC) and State of Health (SOH) is essential to monitor the available energy and ensure safe operations. Several control-oriented models have been developed to predict SOC and SOH in automotive energy storage systems [6-9]. While these models have a simple and

intuitive structure, they do not capture the physical processes related to the transport of lithium in the solid and liquid phase, which are the foundation to an accurate description of the performance and degradation in Li-ion cells.

Porous Electrode Theory [10] has enabled the design of first-principles battery models. These models characterize the lithium intercalation in the electrodes and its influence on the cell terminal voltage as a diffusion based process. While these electrochemical models are able to accurately predict the cell terminal voltage as function of current and temperature, they require significant calibration effort and computation time, due to the presence of large scale coupled partial differential equations (PDE) and nonlinear algebraic equations, ultimately preventing their direct application from estimation and control algorithm design and verification.

As a consequence, substantial efforts need to be dedicated to the development of innovative Model Order Reduction (MOR) techniques to obtain reduced order electrochemical models derived from first principles and suitable for estimation and control applications. Therefore, several approximation methods based on model order reduction (MOR) techniques have been applied to the diffusion PDEs in the porous electrode model. Some research has focused on the approximation of the diffusion PDEs in a specific frequency range. Forman et al. [11] approximated the frequency response of the electrode surface concentration by combining a quasi-linearization and Padé approximation. Smith et al. [12] applied residue grouping method to approximate the difference of the electrode surface concentration to the average concentration. Transfer functions were represented by a truncated series of grouped residues with similar eigenvalues. Although the models reduced the complexity and numerical cost of the DFN model, they do not

provide the spatial distributions of concentrations and potentials. A good knowledge of the concentration and potential distributions could help extend the battery life by avoiding operating conditions where irreversible side reaction can occur [13,14]. Besides, the lithium distribution in the electrode particles can be directly used for SOC estimation [15,16].

For approximation of spatial concentration distributions, Subramanian et al. [17] developed a reduced order model using the polynomial approximation and volume averaging method to represent the concentration profile in the solid phase. Proper orthogonal decomposition [18] has also been applied to obtain ROMs which provides spatial concentration distributions. In recent times, the Galerkin Projection method has been applied as a MOR tool to approximate the diffusion PDEs describing the Lithium transport in the electrolyte solution. For instance, Dao et al. [19], Subramanian et al. [20] and Northrop et al. [21] used Galerkin method and reduced the PDEs that describe the concentration and potential distributions in the liquid phase but they did not extend this method to the spherical diffusion PDEs. Kehs et al. [22] represented the concentration and potential distributions by using the Galerkin projections with a series of shifted and normalized Legendre polynomials. However, due to the time-dependent boundary conditions in the solid phase, the model needs to be manually re-identified by adding the additional algebraic constraints prior to the application of the method.

In this scenario, this paper introduces a new MOR approach to extend the application of the Galerkin method in the solid phase diffusion problem. The time-dependent boundary conditions will be automatically satisfied by the transformation of coordinate. Besides, a procedure of model order selection is established by the analysis in the frequency domain.

The remainder of the paper provides the details of the battery model and shows how the model is reduced by the combination of Galerkin method and coordinate transformation, and the procedure of model order selection. The paper then presents some simulations results of the lithium concentration and potential distributions in the solid and liquid phases. Finally, the predicted voltage by the reduced Galerkin model is compared with the experimental data.

A schematic of the Li-ion cell model is shown in Fig. 1. It consists of three domains: the negative electrode, the separator and the positive electrode. During charge, lithium ions are removed from the positive electrode and deposited into the electrolyte solution. Then the ions are transported within the liquid region by diffusion and ionic conduction, through the separator, to the surface of active material particles in negative electrode where they will be stored. Meanwhile, electrochemical reactions take place as the lithium ions are transported from/into the solid material. During discharge, the opposite process repeats. The separator helps to prevent a short circuit while allowing ions to pass through.

DESCRIPTION OF THE LITHIUM-ION CELL MODEL

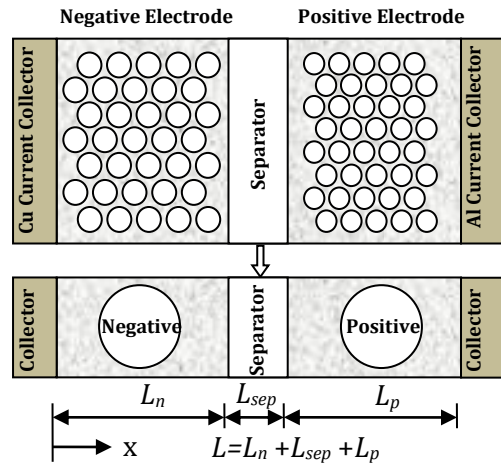


FIGURE 1. SCHEMATIC OF THE ELECTROCHEMICAL BATTERY MODEL

In this work, the model assumes that each electrode is represented by a single spherical particle with the assumption that the total surface area of the particle is equivalent to the overall area of the active material in the porous electrode. However, unlike the single-particle model, where the lithium concentration profile in the liquid phase is approximated as quasi-static and the potential changes are ignored [23], the reduced order model developed in this paper allows one to obtain the concentration and potential distributions in the whole liquid phase. Therefore, this “enhanced” single particle model could maintain sufficient accuracy even at high charge/discharge rates. The structure of the model has been used previously at the Center for Automotive Research [24,25] and is shown in Fig. 2. The parameters used in this study can be found in [26]. Besides, each of the electrode open-circuit potential curves was obtained from the half-cell experiments in [26] as well.

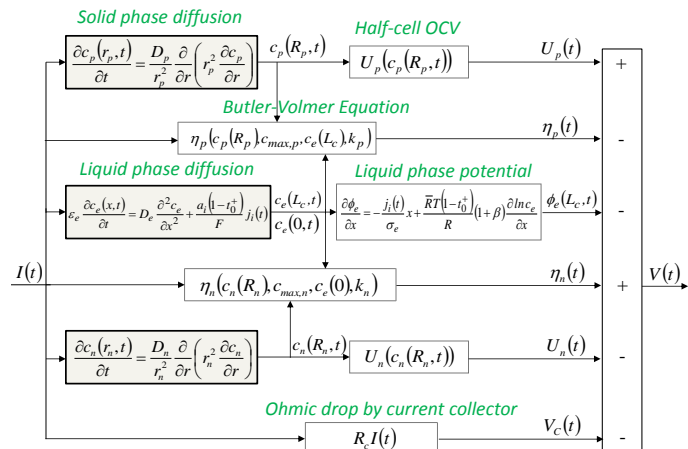


FIGURE 2. BLOCK STRUCTURE OF THE BATTERY MODEL

MODEL ORDER REDUCITON OF THE ELECTROCHEMICAL MODEL USING GALERKIN METHOD

The Galerkin method approximates the concentration by a linear combination of chosen basis functions. Application of this method in the electrolyte has already been found in [19], but the presence of the non-zero, time-dependent boundary conditions in the solid phase poses challenges to the use of Galerkin method directly. In this section, a new approach based on Galerkin method and coordinate transformation will be proposed to obtain a reduced-order solid diffusion model. In addition, the complete concentration distribution can also be obtained inside the active material particles in the electrodes.

Li-ion electrolyte phase diffusion

The Li⁺ concentration in the liquid phase is governed by:

$$\varepsilon_e \frac{\partial c_e(x, t)}{\partial t} = D_e \frac{\partial^2 c_e(x, t)}{\partial x^2} + \frac{1 - t_0^+}{F} J(x, t) \quad (1)$$

The concentration can be approximated by directly applying Galerkin's approximation to Eq. (1). First, a trial solution is firstly assumed as:

$$\hat{c}_e(x, t) = c_e(x, 0) + \sum_{i=1}^N w_i(t) \phi_i(x) \quad (2)$$

where $\phi_i(x)$ are the basis functions and should be selected properly so that the assumed solution also satisfied all boundary conditions. N is the number of the basis functions. In the electrolyte phase, the zero flux boundary conditions are posed on the liquid diffusion PDE and a sinusoidal basis function is chosen in the form of:

$$\phi_i(x) = \cos\left(\frac{i\pi x}{L}\right) \quad (3)$$

The time-variant coefficients of the basis functions $w_i(t)$ are needed to be solved so that the electrolyte concentration can be approximated as a unique linear combination of the basis functions. The residual is obtained by substituting the trial solution into Eq. (1):

$$r(x, t) = \varepsilon_{e,k} \frac{\partial \hat{c}_{e,k}(x, t)}{\partial t} - D_{e,k} \frac{\partial^2 \hat{c}_{e,k}(x, t)}{\partial x^2} - \frac{1 - t_0^+}{F} J(x, t) \quad (4)$$

Note that $\varepsilon_{e,k}$, $D_{e,k}$ are spatially dependent in different domains. Applying Galerkin method to Eq. (4) gives:

$$\begin{aligned} \langle \phi_i, r \rangle &= \int_0^{L_p} \phi_i(x) r(x, t) dx \\ &+ \int_{L_p}^{L_p+L_{sep}} \phi_i(x) r(x, t) dx \\ &+ \int_{L_p+L_{sep}}^{L_{cell}} \phi_i(x) r(x, t) dx = 0 \end{aligned} \quad (5)$$

Equation (5) can be expressed using the matrix form:

$$\mathbf{M}\dot{\mathbf{w}} = \mathbf{K}\mathbf{w} + \mathbf{f} \quad (6)$$

where

$$\begin{aligned} M_{i,j} &= \varepsilon \int \phi_i(x) \phi_j(x) dx \\ K_{i,j} &= D \int \phi_i''(x) \phi_j(x) dx \\ f_i &= \frac{1 - t_0^+}{F} \int \phi_i(x) J(x, t) dx \end{aligned} \quad (7)$$

Again, the limits of the integrals in Eq. (7) are expected to be spatially dependent according to Eq. (5). After solving for the time-dependent coefficients \mathbf{w} at each time point, the concentration distribution along the cell thickness in the electrolyte can be obtained as:

$$\hat{c}_e(x, t) = c_e(x, 0) + \boldsymbol{\phi}^T \mathbf{w} \quad (8)$$

where $\boldsymbol{\phi} = [\phi_1; \phi_2; \dots; \phi_N]$.

Li-ion solid phase diffusion

The PDE that describes the lithium concentration in a spherical electrode active material particle is given below:

$$\frac{\partial c_{s,k}(r, t)}{\partial t} = D_{s,k} \frac{\partial^2 c_{s,k}(r, t)}{\partial r^2} + \frac{2D_{s,k}}{r} \frac{\partial c_{s,k}(r, t)}{\partial r} \quad (9)$$

with the boundary conditions:

$$\begin{aligned} \frac{\partial c_{s,k}(r, t)}{\partial r} \Big|_{r=0} &= 0 \\ D_{s,k} \frac{\partial c_{s,k}(r, t)}{\partial r} \Big|_{r=R_k} &= -\frac{J_k(t)}{a_k F} \end{aligned} \quad (10)$$

where $k = p$ for the positive electrode and $k = n$ for the negative electrode. The difficulty of applying Galerkin method to the solid phase diffusion lies in the time-varying boundary condition at $r = R_k$. Because the basis functions $\phi_i(r)$, such as sinusoidal functions in Eq. (3) and Legendre polynomials [22], are all time independent, it is unreasonable to expect they could automatically satisfy all the boundary conditions in the first place. However, this problem could be solved by redefining a new variable in the following way:

$$U(r, t) = c_{s,k}(r, t) + \frac{J_k(t)}{a_k F D_s} \cdot \frac{r^2}{2R_k} \quad (11)$$

Substituting Eq. (11) into Eq. (9) gives:

$$\begin{aligned} \frac{\partial U(r, t)}{\partial t} &= D_s \frac{\partial^2 U(r, t)}{\partial r^2} + \frac{2D_s}{r} \frac{\partial U(r, t)}{\partial r} \\ &- \frac{3}{aFR} J(t) + \frac{J(t)}{2RaFD_s} \cdot J(t) \end{aligned} \quad (12)$$

with a new set of boundary conditions:

$$\begin{aligned} \frac{\partial U(r, t)}{\partial r} \Big|_{r=0} &= \frac{\partial c_s(r, t)}{\partial r} \Big|_{r=0} = 0 \\ \frac{\partial U(r, t)}{\partial r} \Big|_{r=R} &= \frac{\partial c_s(r, t)}{\partial r} \Big|_{r=R} + \frac{J(t)}{aFD} = 0 \end{aligned} \quad (13)$$

The subscript k is dropped here for simplicity. Now the boundary conditions in Eq. (10) become time independent and enable us to use the sinusoidal basis functions again:

$$\phi_i(r) = \cos\left(\frac{i\pi r}{R}\right) \quad (14)$$

The trial solution $\hat{U}(r, t)$ and the residual can be written as:

$$\hat{U}(r, t) = c_s(r, 0) + \sum_{i=0}^N w_i(t) \phi_i(r) \quad (15)$$

$$r(r, t) = \frac{\partial \hat{U}(r, t)}{\partial t} - D_s \frac{\partial^2 \hat{U}(r, t)}{\partial r^2} - \frac{2D_s}{r} \frac{\partial \hat{U}(r, t)}{\partial r} + \frac{3}{aFR} J(t) - \frac{r^2}{2RaFD_s} \cdot j(t) \quad (16)$$

Applying the Galerkin method and a state space representation is obtained:

$$\dot{\mathbf{w}} = \mathbf{M}^{-1} \mathbf{K} \mathbf{w} + \mathbf{M}^{-1} \mathbf{f} \mathbf{u} + \mathbf{M}^{-1} \mathbf{g} \dot{\mathbf{u}} \quad (17)$$

where

$$\begin{aligned} M_{i,j} &= \int_0^R \phi_i(r) \phi_j(r) dr \\ K_{i,j} &= D \int_0^R \phi''_i(r) \phi_j(r) + \frac{2}{r} \phi'_i(r) \phi_j(r) dr \\ f_i &= -\frac{3}{aFR} \int_0^R \phi_i(r) \frac{1}{AL} dr \\ g_i &= \frac{1}{2RaFD} \int_0^R \phi_i(r) r^2 \frac{1}{AL} dr \end{aligned} \quad (18)$$

Here \mathbf{u} is the input current I . Due to the presence of the time derivative of the input, Eq. (17) is not a standard state space form. Thus, a new coordinate was introduced here. First of all, rewriting Eq. (17) in the following form gives:

$$\dot{\mathbf{w}} = \mathbf{A} \mathbf{w} + \mathbf{B}_1 \mathbf{u} + \mathbf{B}_2 \dot{\mathbf{u}} \quad (19)$$

where $\mathbf{A} = \mathbf{M}^{-1} \mathbf{K}$, $\mathbf{B}_1 = \mathbf{M}^{-1} \mathbf{f}$ and $\mathbf{B}_2 = \mathbf{M}^{-1} \mathbf{g}$

Then let

$$\mathbf{z} = \mathbf{w} - \mathbf{B}_2 \dot{\mathbf{u}} \quad (20)$$

Equation (37) now becomes:

$$\dot{\mathbf{z}} = \mathbf{A} \mathbf{z} + (\mathbf{B}_1 + \mathbf{A} \mathbf{B}_2) \mathbf{u} = \mathbf{A} \mathbf{z} + \mathbf{B} \mathbf{u} \quad (21)$$

The Li-ion concentration in the solid phase can be obtained in the end:

$$\begin{aligned} c_s(r, t) &= c_s(r, 0) + \sum_{i=0}^N w_i(t) \phi_i(r) - \frac{J(t)}{aFD} \cdot \frac{r^2}{2R} \\ &= c_s(r, 0) + \mathbf{C} \mathbf{z} + \mathbf{D} \mathbf{u} \end{aligned} \quad (22)$$

where

$$\mathbf{A} = \begin{bmatrix} -\frac{(0 \cdot \pi)^2 D}{R^2} + a_{00}/2 & a_{01}/2 & \cdots & a_{0n}/2 \\ a_{10} & -\frac{(1 \cdot \pi)^2 D}{R^2} + a_{11} & \cdots & a_{1n} \\ \vdots & \vdots & \ddots & \vdots \\ a_{n0} & a_{n1} & \cdots & -\frac{(n \cdot \pi)^2 D}{R^2} + a_{nn} \end{bmatrix}$$

$$\mathbf{B}_1 = -\frac{3}{aFRAL} \begin{bmatrix} 1 \\ 0 \\ \vdots \\ 0 \end{bmatrix}, \quad \mathbf{B}_2 = \frac{2R}{aFDAL} \begin{bmatrix} \frac{1}{12} \\ \frac{\cos(2\pi)}{(1\pi)^2} \\ \vdots \\ \frac{\cos(n\pi)}{(n\pi)^2} \end{bmatrix}, \quad \mathbf{C} = \boldsymbol{\phi}^T, \quad \mathbf{D} =$$

$$\boldsymbol{\phi}^T \mathbf{B}_2 - \frac{1}{aFDAL} \cdot \frac{r^2}{2R}$$

and $a_{ij} = -\frac{2j\pi D (\text{Si}(j\pi - i\pi) + \text{Si}(j\pi + i\pi))}{R^2}$. $\text{Si}(x)$ is the *sine integral function* and defined as:

$$\text{Si}(x) = \int_0^x \frac{\sin(t)}{t} dt$$

Selection of the model order

Before the implementation of the ROM for simulations, the order of ROM should be determined. However, since there is no standard procedure for order truncation in ROMs, this section attempts to explore a possible approach to select the model order for ROMs.

One way to determine the appropriate order for a ROM is to compare each transfer function of the ROMs with the one of the PDE-based model. Thus, the truncation order can be decided by setting a trade-off between the model order and the ROM's ability to capture the frequency response of the PDE-based model. In this work, the transfer functions are obtained for the electrode surface concentration based on the fact that the surface concentration governs many respects of the cell behavior.

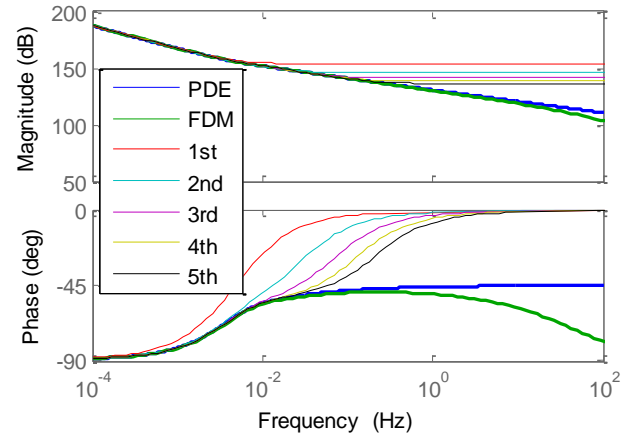


FIGURE 3. FREQUENCY RESPONSE OF SURFACE CONCENTRATION FROM SOLID DIFFUSION PDE AND GALERKIN METHOD

Figure 3 compares the first to fifth-order Galerkin ROMs with the PDE-based model. Results show that all frequency responses at different orders agree very well with the PDE-based model in the low frequency range. Besides, the accuracy of the ROMs increases with the growth of number of states. It should also be noted that all the Galerkin ROMs will be sensitive to high frequency noise, because, the transfer functions of the Galerkin models are non-strictly proper systems. However, as shown in Fig. 4, 90% of the frequency distribution of the battery input profile is within frequencies less than 0.4 Hz. Using this information, a fifth order Galerkin method model is chosen as qualified candidates for the solid phase ROM based on the values of parameters in this work. A similar procedure is also applied to the electrolyte diffusion phase, and a third order truncation is selected.

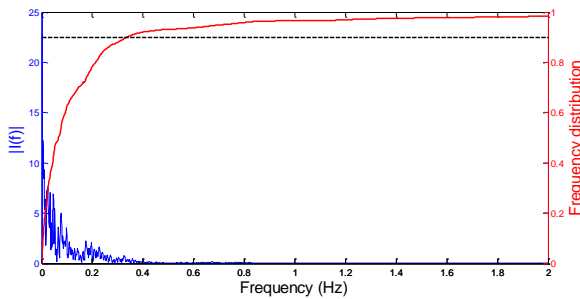


FIGURE 4. FREQUENCY CONTENT OF BATTERY CURRENT PROFILE USING A FAST FOURIER TRANSFORM

RESULTS AND DISCUSSION

Figure 5 compares the concentration distribution obtained by the fifth-order Galerkin model and finite difference method (FDM). The battery was subjected to part of the United States Advanced Battery Consortium (USABC) PHEV dynamic charge-depleting duty cycle profile. As shown in Fig. 5, the fifth-order reduced Galerkin model agrees very well with the FDM in the whole diffusion domain. The maximum error between the predicted concentration by the Galerkin model and FDM is only 1.56%.

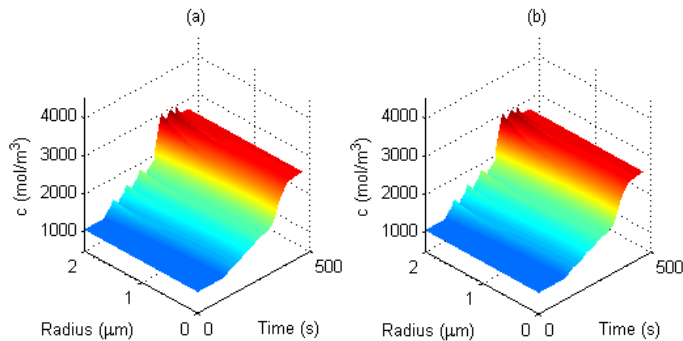


FIGURE 5: COMPARISON OF THE CONCENTRATION DISTRIBUTION AT THE POSITIVE ELECTRODE PREDICTED BY FDM AND GALERKIN METHOD (A) FDM (B) GALERKIN METHOD (C) CONCENTRATION DIFFERENCES

The concentration distribution inside the negative electrode during the whole simulation time is shown in Fig. 6. For clarity, the comparison of the Galerkin model with the FDM is skipped. As shown in Fig. 6, during the charge-depleting duty cycle, lithium ions were gradually moving out of the negative electrode into positive electrode. Although the input current became negative sometimes and the battery was charged for a short while, a decreasing trend in the concentration at the negative electrode can be observed. In addition, the concentration profile along the radius of the active material particles displays as a parabolic shape. When the lithium ions de-intercalate from the negative electrode particles, the concentration would first decrease at the surface of the particles. As the diffusion process continues, lithium ions are depleted gradually inside the particles and then an overall reduction of the concentration can be found in the negative electrode.

Figure 7 shows the lithium concentration in the electrolyte and the concentration overpotential caused by the difference of concentration between electrode surfaces and the bulk solution. The concentration overpotential profile is obtained by calculating the potential difference between $x = 0$ and other locations along the cell thickness. The potential at $x = 0$ is set to be 0 V since the only the potential differences are relevant to the calculation of the cell terminal voltage [24]. The results show that when the cell is discharged or charged with large currents, neither the concentration nor potential changes in the electrolyte is negligible. Particularly, when the discharge current reaches almost 10 C at $t = 340$ seconds, the concentration overpotential reaches as high as -0.1 V, which contributes a lot to the drop of the cell terminal voltage.

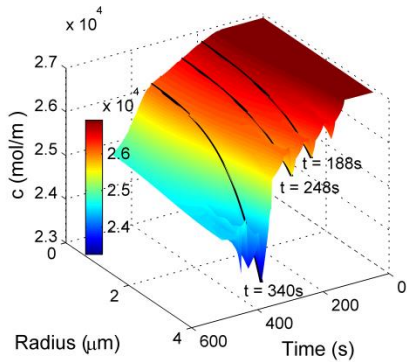
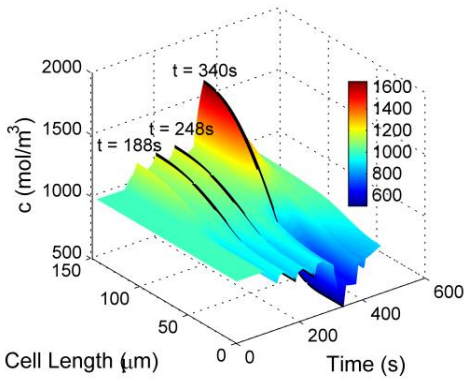
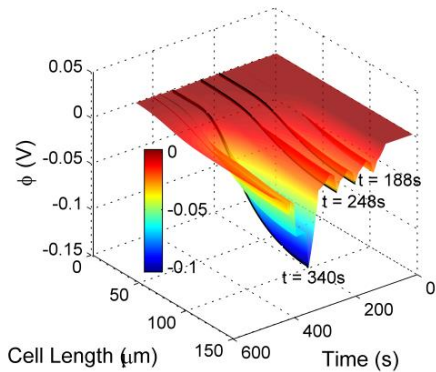


FIGURE 6. CONCENTRATION DISTRIBUTION IN THE NEGATIVE ELECTRODE



(a)



(b)

FIGURE 7. CONCENTRATION AND POTENTIAL DISTRIBUTION IN THE ELECTROLYTE PHASE DURING A PHEV CHARGE DEPLETING CYCLE: (A) ELECTROLYTE CONCENTRATION, (B) ELECTROLYTE POTENTIAL

The cell voltage is then obtained by combining all the above results. Numerical simulations are run in MATLAB on a PC with a 2.60 GHz i5-3320M CPU and 4 GB RAM. Comparison of predicted voltage by Galerkin model with the experimental voltage is shown in Fig. 8. The root-mean-square error of the differences between the predicted voltage and experimental data is only 2.4 mV. Meanwhile, the simulation

time for this model is less than 1.5 seconds using 0.01 s time steps, which is 300 times faster than the real time. With a high level of accuracy as well as computational efficiency, the proposed Galerkin method will be suitable for real-time simulation, control, and estimation applications.

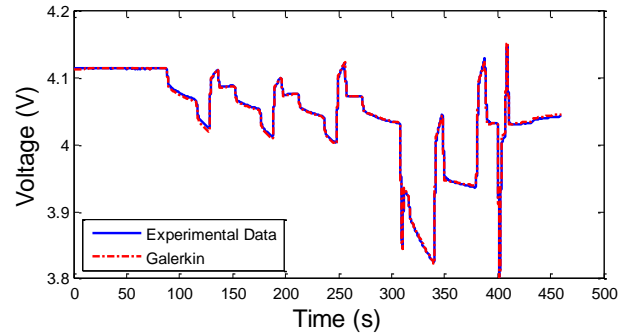


FIGURE 8. COMPARISON OF PREDICTED VOLTAGE WITH EXPERIMENTAL DATA

Furthermore, the Galerkin Models are also compared with the well-established MOR approach by balanced truncation (BT). The maximum concentration errors between different ROMs and FDM are listed in Table 1. It has been shown that the two methods will give comparable RMS error at higher truncation orders. BT is viewed as the “gold standard” of MOR [27], but it should be mentioned that the PDE needs to be discretized before the application of BT, and therefore the truncation error is included from the FDM. On the other hand, the main advantage of Galerkin method over BT is the fact that the resulting ROM is fully physical, for example, the parameters of the ROM are directly related to the physical constants of the system (described by the PDEs). This enables one to conduct state and parameter estimation directly on the ROM. From the computational standpoint, the computation cost for BT method is much higher, especially for very large systems since this method requires the diagonalization of the controllability and observability Gramians.

TABLE 1. MAXIMUM CONCENTRATION ERROR BETWEEN DIFFERENT ROMS AND FDM

Model order	Maximum error between ROMs and FDM (%)	
	Balanced truncation	Galerkin method
1st order	20.1%	12.1%
2nd order	10.3%	5.5%
3rd order	5.2%	3.1%
4th order	2.1%	2.0%
5th order	1.1%	1.1%

CONCLUSIONS

A new MOR approach based on Galerkin method and coordinate transformation is proposed in this paper. This approach has the advantage of reducing the number of diffusion states greatly and predicting accurate concentration profiles at the same time. The model order truncation is determined by

analysis in the frequency domain. The resulting concentration distribution is compared with the one obtained by the FDM and the predicted cell voltage is validated against the experimental data. The RMS error is only 2.4 mV and the simulation time turns out to be 300 times faster than the real time.

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