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Control-Oriented Implementation and Model Order Reduction of a Lithium-Ion Battery Electrochemical Model

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***Abstract*—The use of electrochemical models makes it computationally intractable for online implementation as the model is subject to a complicated mathematical structure including partial-differential equations (PDE). This paper is based on the single particle model with electrolyte dynamics. Methods to solve the PDEs in the governing equations are given. Model order reduction techniques are applied to the electrochemical model to reduce the order from 350 to 14. The models solved by numerical solution, residue grouping method and balanced truncation method are compared with experimental data of a coin cell for validation. The results show that the reduced order model can increase simulation time 75 times compared with the high order model and 2.3% root mean square error with the experiment result.**

***Keywords*—Lithium ion battery, single particle model with electrolyte, reduce order modeling, residue grouping, balance truncation.**

I. INTRODUCTION

Electrochemical models of batteries are typically developed from first principles and aim to accurately capture the internal battery dynamics such as diffusion, intercalation kinetics, and electric potentials. Underpinned by such models, battery internal states or parameters can be better understood and manipulated by sophisticated estimation and control approaches. However, the use of electrochemical models makes it computationally intractable for online implementation as the model is subject to a complicated mathematical structure including partial-differential equations (PDE), ordinary-differential equations (ODE) and algebraic equations. In order to devise a control-oriented model, to support real-time monitoring and control, model order reduction techniques, including residue grouping [1], balanced truncation [2], Padé approximation [3], can be used to reduce the model order while maintaining the desired level of accuracy. G.Fan in [4] compared some order reduction techniques applied to the solid-state diffusion. Although the above models have addressed the accuracy and development of reduced order models of Li-ion batteries, the implementation and comparison between different methods with real cell experiment data still need to be discussed.

This paper is based on the single particle model with electrolyte dynamics (SPMe). The PDEs in the governing equation of the SPMe are solved by finite difference method (FDM) and finite element method (FEM). After solving the PDEs, the full order model would be high (e.g. 350th order). Model order reduction techniques are used to develop a low order Li-ion battery model derivate that is suitable for real-time implementation, for example within a battery control system. Residue grouping (RG) and balance truncation (BT) are order reduction techniques for high order systems. They are applied to the SPMe model to reduce the model order from 350 to 14.

Both the full order model (FOM) and the reduced order model (ROM) have been implemented within the commercially available software Matlab and SIMULINK.

II. SINGLE PARTICLE MODEL WITH ELECTROLYTE

The SPMe model derived by Scott J. Moura in [5] is a simplification of the Newman model [6]. The schematic of the SPMe model is shown in Figure 1. The positive and negative electrodes are modelled with spherical particles surrounded by the electrolyte. The Li-ions intercalation and de-intercalation processes are performed through the surface area of these particles[7].

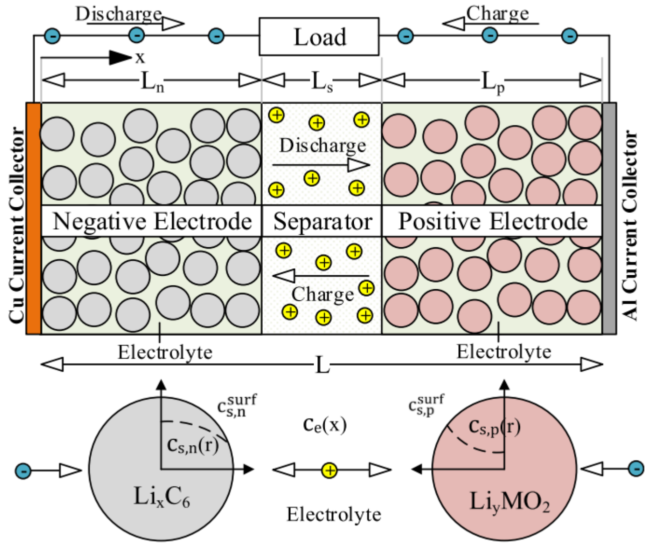

Fig. 1. Schematic of the Newman model [7]

The SPMe is derived under several assumptions [5]. These assumptions ultimately render a model consisting of two diffusion PDEs for electrodes concentrations, one diffusion PDE for electrolyte concentrations, and a nonlinear output function about the surface concentration of the solid, the boundary concentration of the electrolyte, and the current (as shown in Fig. 2).

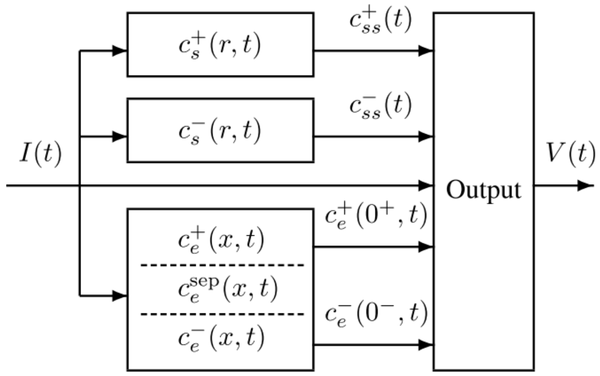

Fig. 2. Block diagram of SPMe [5].

The governing equations of SPMe are given in Table I. The nomenclatures can be found in Table II.

TABLE I. GOVERNING EQUATIONS

Governing Equations	**Eq. No.**
$\frac{\partial c_s^{\pm}}{\partial t}(r,t)=\frac{1}{r^2}\frac{\partial}{\partial r}\left[D_s^{\pm}r^2\frac{\partial c_s^{\pm}}{\partial r}(r,t)\right]$	(1)
$\frac{\partial c_e^{j}}{\partial t}(x,t)=\frac{\partial}{\partial x}\left[\frac{D_e^{eff}}{\varepsilon_e^{j}}\frac{\partial c_e^{j}}{\partial x}(x,t)\right]\mp\frac{(1-t_c^0)}{\varepsilon_e^{j}FL^{j}}I(t)$ $j\epsilon\{-,sep,+\}R_f^-$	(2)
$\kappa^{eff}\frac{\partial \phi_e^{\pm,s}}{\partial x}(x,t)=-i_e^{\pm}(x,t)+$ $\kappa^{eff}(c_e)\cdot\frac{2RT}{F}(1-t_c^0)\left(1+\frac{d\,ln\,f_{c/a}}{d\,ln\,c_e}(x,t)\right)\frac{\partial\,ln\,c_e}{\partial x}(x,t)R_f^{+}$	(3)
$\frac{\partial i_e^{\pm}}{\partial x}(x,t)=a^{\pm}Fj_n^{\pm}(x,t)$	(4)
$j_n^{\pm}(t)=\mp\frac{I(t)}{Fa^{\pm}L^{\pm}}=\frac{1}{F}i_0^{\pm}(t)\left[e^{\frac{\alpha_a F}{RT}\eta^{\pm}(t)}-e^{-\frac{\alpha_c F}{RT}\eta^{\pm}(t)}\right]$	(5)
$i_0^{\pm}(t)=k^{\pm}[c_{ss}^{\pm}(t)]^{\alpha_c}\times\left[c_e(x,t)\left(c_{s,max}^{\pm}-c_{ss}^{\pm}(t)\right)\right]^{\alpha_a}$	(6)
$\eta^{\pm}=\phi_s^{\pm}(t)-\phi_e(x,t)-U^{\pm}\left(c_{ss}^{\pm}(t)\right)-FR_f^{\pm}j_n^{\pm}(t)$	(7)

TABLE II. NOMENCLATURE

Description	**Symbol**	**Unit**
Lithium concentration in solid phase	$c_s^{\pm}$	mol/m^3
Lithium concentration in electrolyte phase	c_e^{j}	mol/m^3
Solid electric potential	$\phi_s^{\pm}$	V
Electrolyte electric potential	$\phi_e^{\pm,s}$	V
Ionic current	$i_e^{\pm}$	A/m^2
Molar ion flux	$j_n^{\pm}$	mol/m^2-s
Exchange current density	$i_0^{\pm}$	A/m^2
Overpotential	$\eta^{\pm}$	V
Lithium concentration in solid phase at particle surface	$c_{ss}^{\pm}$	mol/m^3
Apply current	I	A/m^2
Terminal voltage	V	V

III. NUMERICAL SOLUTION OF PDES

To solve the governing equations, numerical methods such as the FDM and FEM can be applied on the diffusion PDE of the SPMe in the solid and electrolyte domains separately.

A. Finite-Difference Method

The principle of FDM is very similar to numerical methods used to solve ODEs. In this method the differential operator is approximated by replacing it with the derivatives in the equation using differential quotients [8].

Central difference of the first order derivatives of FDM can be applied to solve the diffusion PDE of the SPMe. The numerical solution of the solid phase with finite difference approach would be:

$$\frac{\partial c_s^{\pm}}{\partial t}(Z,t)=\left[\frac{D_s^{\pm}}{Z{R_s^{\pm}}^2\Delta x}+\frac{D_s^{\pm}}{{R_s^{\pm}}^2(\Delta x)^2}\right]c_{s\,(n+1)}^{\pm}+\left[\frac{D_s^{\pm}}{{R_s^{\pm}}^2(\Delta x)^2}-\frac{D_s^{\pm}}{Z{R_s^{\pm}}^2\Delta x}\right]c_{s\,(n-1)}^{\pm}+\left[\frac{-2D_s^{\pm}}{{R_s^{\pm}}^2(\Delta x)^2}\right]c_{s\,(n)}^{\pm}$$

where $Z=\frac{r}{R_s^{\pm}}$, Δx is the distance between each two nodes, and the boundary conditions are:

$$\frac{\partial c_s^{\pm}}{\partial r}(0,t)=0$$

$$\frac{\partial c_s^{\pm}}{\partial r}(R_s^{\pm},t)=\pm\frac{1}{D_s^{\pm}Fa^{\pm}L^{\pm}}I(t)$$

B. Finite-Element Method

The FDM is the simplest and most commonly used approach to the solution of the diffusion equations found in battery models. But FDM ignored the concentrations between each two nodes. The finite element method (FEM) add in admissible functions and generates equations for the nodal dynamics that can be realized in state-variable or transfer-function forms. More information and details on the FEM method can be found in [9, 10].

In the simulation part of this paper, the SPMe model is implemented in Matlab/SIMULINK. The full order model (FOM) is a numerical solution by FDM with 350 states in total. The solid phase is mashed in 50 nodes for each electrode and the electrolyte is mashed in 250 nodes across the three domains.

IV. MODEL ORDER REDUCTION TECHNIQUES

Solving the PDEs by FDM and FEM would produce a high order model. The full order would be 350th order. This full order model is computationally intractable for online implementation. Therefore, model order reduction techniques can be used to reduce the model order while maintaining the desired level of accuracy. Residue grouping and balance truncation methods are applied to in this paper.

A. *Residue Grouping Method*

To apply residue grouping method analytically, we need to obtain a transcendental transfer function with an infinite number of poles. Transcendental transfer functions are commonly characterised by numerous closely spaced poles with similar residues. The transcendental transfer function of the positive electrode solid diffusion equation is [11]:

$$\frac{c_{ss}{}^{+}(s)}{I(s)} = Z + \sum_{k=1}^{\infty} \frac{Res_k s}{s - p_k}$$

where $c_{ss}{}^{+}(s)$ is the surface concentration after Laplace transform, p_k represent the poles, Res_k represent the residues, and Z represents the steady state solution. The derivative of the transcendental transfer function and application of the residue grouping method on the electrode solid diffusion equation can be found in [1].

The poles are $p_k = -D_s\left(\frac{\xi_k}{R_s}\right)^2$ where ξ_k are roots of $\tan(\xi_k) = \xi_k$ not including $\xi_0 = 0$.The residues are $Res_k = \frac{-2}{a_s F R_s p_k}$. The direct term is $Z = \frac{-R_s}{5 a_s F D_s}$.

From [1], we take a cut-off frequency of 10 Hz to approximate the solid state diffusion dynamics and neglecting the high frequency dynamics above the cut-off frequency. Then plot the poles versus residues as Fig.3 shows.

Residue grouping method is to partition the frequency range of interest into d “bins” and lump together modes within each bin. The grouping procedure yields the d-th order transfer function as a reduced order transfer function.

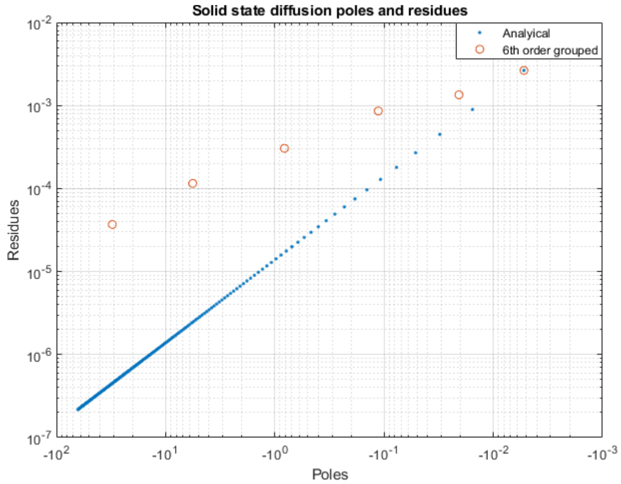

Fig. 3. Solid state diffusion poles and residues.

In Fig. 3 the blue dots are the analytical solution of the residues versus poles. After group the points into 6 “bins”, the reduced order solution would be a 6th order transfer function with the input of apply current $I(t)$ and output the solid state surface concentration$c_{ss}{}^{+}(t)$.

The same process can be applied on the negative electrode. For the electrolyte phase, the residue grouping method can be applied numerically based on FEM. The transfer function of $\Delta c_e(t)$ vs $I(t)$ would be got using the method in [1].

B. *Balanced Truncation Method*

Balanced truncation method is applied by minimising the system's maximum error in the frequency domain [2]. A balanced truncation uses Hankel singular values instead of eigenvalues to discard states and reduce the model order. With the proper choice of a threshold for state truncation, the model order can be reduced significantly without affecting the performance a lot. Fig. 4 show the plot of Hankel Singulars in different states for order reduction of the SPMe model with parameters in Table III.

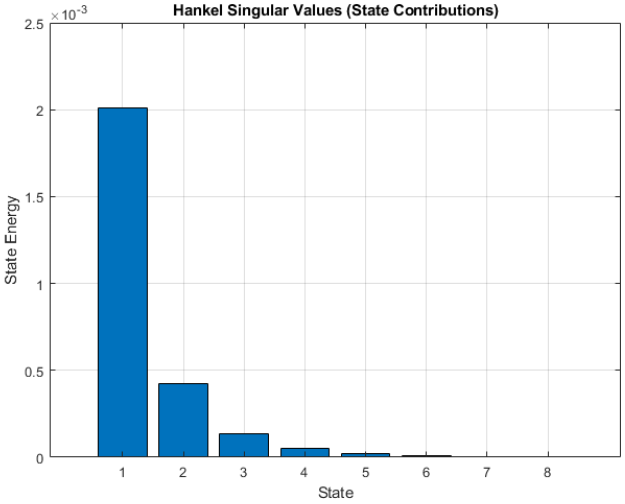

Fig. 4. Hankel Singular Values (state contributions).

The implementation of balanced truncation in Matlab can simply by using function “*balred()*” to capture the reduced order model’s transfer function.

The model order reductions in this paper, use a 6th order transfer function for both the positive and negative electrode diffusion equation and the electrolyte diffusion as a 2nd order. In total, the reduced order model is a 14th order for both implementations of residue grouping and balanced truncation.

V. SIMULATION AND VALIDATION

To compare the different model order reduction techniques, both full order model and reduced order models based on SPMe model are implemented in SIMULINK. The parameters (Table III) are obtained by fitting the experimental data of a coin cell.

The validation of the model is performed using a discharge current of 0.5 C until the voltage reaches the minimum value, followed by a charge of 0.5C. The terminal voltage of the experimentally measured, the full order model (FOM), residue grouping reduced order model (RG) and balanced truncation reduced order model (BT) are compared. The comparison is shown in Fig. 5 below.

TABLE III. PARAMETERS

Parameters	**Symbol**	**Value**
Thickness of anode	L^{-}	22 µm
Thickness of separator	L^{s}	20 µm
Thickness of cathode	L^{+}	35 µm
Radius of solid particles in anode	R_s^{-}	11.5 µm
Radius of solid particles in cathode	R_s^{+}	14.5 µm

Volume fraction of solid in anode	ε_s^-	0.635
Volume fraction of solid in cathode	ε_s^+	0.476
Volume fraction of electrolyte in anode	ε_e^-	0.295
Volume fraction of electrolyte in separator	ε_e^s	0.44
Volume fraction of electrolyte in cathode	ε_e^+	0.443
Diffusion coefficient of solid in anode	D_s^-	0.1 $\mu m^2 s^{-1}$
Diffusion coefficient of solid in cathode	D_s^+	0.8 $\mu m^2 s^{-1}$
Diffusion coefficient of electrolyte	D_e	278.8 $\mu m^2 s^{-1}$
Conductivity of solid in anode	σ^-	100 $\Omega^{-1} m^{-1}$
Conductivity of solid in cathode	σ^+	100 $\Omega^{-1} m^{-1}$
Bruggeman porosity	b	1.5
Ambient temperature	T	298.15 K
Transference number	t_c^0	0.363
Kinetic reaction rate of anode	k^-	5 x 10^{-6} $(A/m^2)(mol^3/mol)^{(1+\alpha)}$
Kinetic reaction rate of cathode	k^+	7 x 10^{-6} $(A/m^2)(mol^3/mol)^{(1+\alpha)}$
Charge transfer coefficients	α	0.5 Ωm^{-2}
Anode-electrolyte resistivity	R_f^-	0 Ωm^{-2}
Cathode-electrolyte resistivity	R_f^+	0 Ωm^{-2}
Contact resistance/Current collector Resistance	R_c	0 Ωm^{-2}
Conductivity of electrolyte	κ	0.95 Sm^{-1}
Faraday’s constant	F	96485.33289 $Cmol^{-1}$
Universal gas constant	R	8.314472 $Jmol^{-1}K^{-1}$
Fixed electrolyte concentration	c_e	1000 $molm^{-3}$
Cell maximum voltage	V_{max}	4.115 V
Cell minimum voltage	V_{min}	2.5 V
Maximum concentration of anode	$c_{s,max}^-$	31,389 $molm^{-3}$
Maximum concentration of cathode	$c_{s,max}^+$	36,292 $molm^{-3}$

The real running time, peak error and root mean square error (RMSE) are given in Table IV. From the comparison figure and table it can be found, FOM has similar accuracy with the reduced order model RG and BT by comparing with the experiment results. However, the reduced order models’ computation time are much shorter than the FOM with the same simulation time (14000 s) and step length (0.1 s).

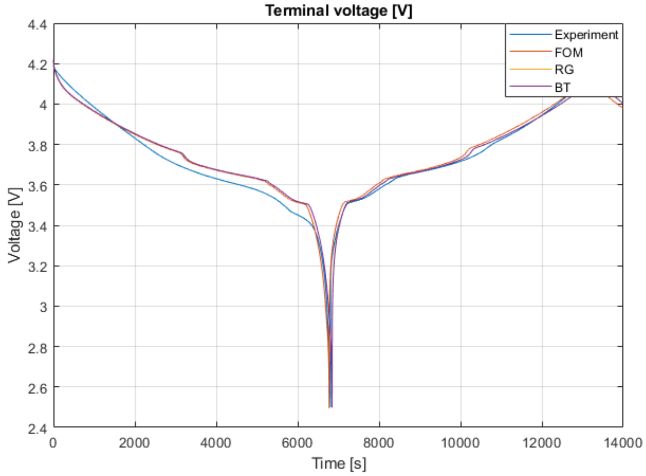

Fig. 5. Comparison of the terminal voltage among experiment result, full order model, residue grouping reduced order model, and balance truncation reduced order model.

TABLE IV. MODELS COMPARISON

Model	Computation time	RMSE
FOM	2711.46 s	2.8185%
RG	76.28 s	2.3312%
BT	75.34 s	2.3334%

The residue grouping and balanced truncation method were implemented with the same reduced order in the simulation model. The residue grouping method appears very similar to the balanced truncation method but achieves slightly better accuracy in the whole cycle.

VI. CONCLUSION

This paper applied three methods to solve the PDEs in the electrochemical models of li-ion battery for control oriented implementation. The proposed model is most similar to those developed in [5], yet we analyse the solver of the PDEs in the governing equations and the model order reduction methods applied on the model. The reduced order single particle model, implemented in SIMULINK, was validated with experiment data of a coin cell. Accuracies of 97.67% was obtained for the reduced order models (compared with 97.18% for a full order model) when validated with constant current charge and discharge experiment. In addition, a model based observer can be subsequently designed and tested in the future.

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