



A three-dimensional meso-macroscopic model for Li-Ion intercalation batteries[☆]



S. Allu^{a, *}, S. Kalnaus^a, S. Simunovic^a, J. Nanda^a, J.A. Turner^a, S. Pannala^b

^a Oak Ridge National Laboratory, Computer Science and Mathematics Division, Oak Ridge, TN 37831-6164, USA

^b SABIC, Houston, TX, USA

H I G H L I G H T S

- 3D meso-macroscopic model for Li-Ion intercalation batteries.
- Framework to incorporate closure relations of non-spherical particle morphologies.
- Unified approach to model arbitrary electrode-electrolyte-electrode topologies.
- Rigorous volume averaging with consistency between various physical processes.
- Investigated benefits of 3D interdigitated arrangements of the electrodes.

A R T I C L E I N F O

Article history:

Received 20 February 2016

Received in revised form

21 April 2016

Accepted 1 June 2016

Keywords:

Li-ion

Modeling and simulation

A B S T R A C T

In this paper we present a three-dimensional computational formulation for electrode-electrolyte-electrode system of Li-Ion batteries. The physical consistency between electrical, thermal and chemical equations is enforced at each time increment by driving the residual of the resulting coupled system of nonlinear equations to zero. The formulation utilizes a rigorous volume averaging approach typical of multiphase formulations used in other fields and recently extended to modeling of supercapacitors [1]. Unlike existing battery modeling methods which use segregated solution of conservation equations and idealized geometries, our unified approach can model arbitrary battery and electrode configurations. The consistency of multi-physics solution also allows for consideration of a wide array of initial conditions and load cases. The formulation accounts for spatio-temporal variations of material and state properties such as electrode/void volume fractions and anisotropic conductivities. The governing differential equations are discretized using the finite element method and solved using a nonlinearly consistent approach that provides robust stability and convergence. The new formulation was validated for standard Li-ion cells and compared against experiments. Its scope and ability to capture spatio-temporal variations of potential and lithium distribution is demonstrated on a prototypical three-dimensional electrode problem.

© 2016 Elsevier B.V. All rights reserved.

[☆] Notice: This manuscript has been authored by UT-Battelle, LLC under Contract No. DE-AC05-00OR22725 with the U.S. Department of Energy. The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes. The Department of Energy will provide public access to these results of federally sponsored research in accordance with the DOE Public Access Plan (<http://energy.gov/downloads/doe-public-access-plan>).

* Corresponding author.

E-mail address: allus@ornl.gov (S. Allu).

1. Introduction

Li-Ion battery packs for electric drive vehicles require energy and power densities that can only be achieved by highly energetic materials. Transport limitations of the constituent materials and safe operating conditions of the electrode-electrolyte system play an important role to meet the design requirements of these packs. A wide range of cell chemistries and additives has been developed for different applications to improve cell characteristics such as energy/power density [2] and increase safety. New 3D battery architectures [4,5], both at the cell and the pack level, are under development with a potential to achieve high power and energy

Nomenclature

a_s	specific surface area of electrode	t	time
C_e	concentration of lithium salt in liquid electrolyte phase	T	temperature
C_s	surface concentration of lithium in solid phase	t_+^0	lithium cation transference number
$C_{s,avg}$	average concentration of lithium in solid phase	U	equilibrium potential
$C_{s,max}$	maximum concentration of lithium in solid phase	α_a	anodic transfer coefficient
D_e	diffusion coefficient of lithium salt in the solution	α_c	cathodic transfer coefficient
D_e^{eff}	effective diffusivity of electrolyte	ϵ_e	volume fraction of electrolyte
D_s	diffusion coefficient of lithium in solid phase	ϵ_s	volume fraction of solid phase
D_s^{eff}	effective diffusivity of solid electrode matrix	ϕ_e	potential of liquid phase
f_+	activity coefficient of the salt in electrolyte	ϕ_s	potential of solid phase
F	Faraday's constant (96485 C mol ⁻¹)	η	surface overpotential
i_0	exchange current density	κ	ionic conductivity of electrolyte
j^{Li}	pore wall Li flux across interface	κ^{eff}	effective ionic conductivity of liquid phase in solid matrix
l_s	characteristic diffusion length in solid phase	κ_D^{eff}	effective diffusional conductivity of electrolyte
R	universal gas constant (8.3143 J mol ⁻¹ K ⁻¹)	σ	electric conductivity of solid electrode material
R_s	radius of solid electrode particle	σ^{eff}	effective electric conductivity of electrode

densities in smaller footprints compared to the two dimensional counterparts [3]. A physics-based, system level simulation capability can accelerate the battery development by filtering different choices for the electrode materials. In these system-level models one can analyze the effect of various materials and chemistry options, predict system performance, optimize material components and geometry, and address the safety and durability in an integrated fashion [6].

In recent years there have been tremendous advances in research, both experimental and computational, toward improving performance and life of lithium ion batteries. Many causes for capacity loss, namely, lithium deposition, porosity change, film formation, side reactions, thermal abuse, intercalation induced stress generation and mechanical degradation have been identified. Strong improvements of the performance at the cell level have not been accompanied with commensurate improvements at the battery pack level. One of the technologies envisioned to increase battery power and energy densities is the three-dimensional form of battery electrodes. The 3D format brings up new challenges, such as manufacturing feasibility, non-uniform current distribution, stress concentrations, and complex heat dissipation, all of which need to be analyzed experimentally and computationally. A study of current density distribution in micro batteries based on periodic electrode rod elements was presented by R.W.Hart et al. [7]. Although high capacities can be achieved on a smaller footprint, many significant issues, such as capacity fade due to local lithium depletion from the electrolyte, remain [8]. It is imperative to understand the interplay between the coupled physicochemical processes (electrical, electrochemical, thermal, mechanical) that span over multiple length scales, and correlate the influence of the microscopic interactions with the macroscopic battery design features for improving battery performance.

Computational models developed for the lower-length scales (atomistic and mesoscopic) are too computationally expensive for system level analysis. The existing models at the macroscopic or system-level are primarily based on the electrical circuit models or simple 1D models [9–13]. Various mathematical formulations, such as the single-particle [14] and porous-electrode models [15], have been employed to estimate the performance of the batteries. In the single-particle method, the lithiation state of each electrode is represented by a single particle and thus changes in the concentration of electrolyte across the electrode thickness are neglected.

Such simplified approach is applicable to thin electrodes and relatively small currents. A one-dimensional model to simulate galvanostatic charge and discharge for a porous electrode and separator system was developed in Ref. [15]. The conservation of mass in the solid phase is either explicitly calculated using the Duhamel's superposition principle or solved with a pseudo second dimension i.e., the radius of the particle with rest of the differential equations. Wang et al. [16] presented a relation between the surface concentration and the average concentration of lithium in solid phase to avoid the computationally expensive solution of the solid phase diffusion equation. A comparison of approximate solutions for the solid state diffusion problem can be found in Ref. [17].

1D models are limited in their ability to capture spatial variations in permeability or conductivity and thus cannot accurately represent the multidimensional structure of recent electrode and solid electrolyte material. There have been some recent extensions to 2D [18–20] and this is still an active area of development. But these models lack the ability to capture non-uniformities in large cells. Recently, a new computational infrastructure for battery simulations [21] has been presented that can loosely couple electrochemical, thermal, and electrical models in three dimensions but the primary transport of ions in the electrochemical models studied was still limited to one direction. A reformulated porous electrode theory has been developed in Ref. [22] using phase field models to describe material transformation consistent with ion intercalation and faradaic kinetics for concentrated solutions. The formulation, however, is still not three-dimensional as the solid diffusion is one dimensional case of the idealized spherical geometry.

In this paper, we describe a system-level computational framework that solves the underlying transport and kinetics equations for arbitrary 3D cell configuration as a single domain. In the following section, we present the extension of the porous electrode theory using volume averaging technique that accounts for the spatio-temporal variations at the cell level and is informed by the constitutive models from the lower length scales. In Section 3, we describe the numerical solution method for solving the resulting Differential Algebraic Equation (DAE) system that enforces multi-physics solution consistency. Finally, in Section 4 a series of computational results are presented as verification and validation studies to demonstrate the efficiency of the modeling framework on single domain. A case study of prototype 3D cell

Download English Version:

<https://daneshyari.com/en/article/7727498>

Download Persian Version:

<https://daneshyari.com/article/7727498>

[Daneshyari.com](https://daneshyari.com)