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Thermal analyses of LiFePO₄/graphite battery discharge processes

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HIGHLIGHTS

- We develop an electrochemical-thermal model for LiFePO₄/graphite batteries.
- Various heat sources/sinks are fully handled in this model.
- Model results provide detailed insight into the battery discharge processes.
- Comprehensive thermal analyses to discharge processes are conducted.
- Distribution and evolution of heat sources/sinks are extensively scrutinized.

A R T I C L E I N F O

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ABSTRACT

An electrochemical-thermal coupling model is developed to describe the LiFePO₄/graphite battery discharge and charge processes. Various heat generations/consumptions including Joule heat, reversible entropy heat, contact resistance heat, irreversible electrochemical reaction heat, ionic migration heat, and convective heat release to the ambient during charge or discharge processes are calculated in detail. The developed model is first validated by experimental data. Then systematic and comprehensive thermal analyses with respect to various discharge processes are performed based on the simulated results. For the specific cell considered, the irreversible electrochemical reaction heat and contact resistance heat take more proportion of the total heat generation as it is directly proportional to the squared discharge current density; the ionic migration heat is a sink with magnitude being about 1/3 of the Joule heat. The reversible entropy heat changes its sign from a negative heat sink to a positive heat source during a discharge process and its changing magnitude may be comparable to the irreversible electrochemical reaction heat for all the discharge processes of different C-rates.

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1. Introduction

Ever since reported by Padhi et al. [1] in 1997, the LiFePO₄ has been considered to be one of the most promising positive electrode materials for lithium-ion batteries. Although the gravimetric and volumetric energy density of LiFePO₄ cathode is lower than other cathode materials that are based on rare metal composites (LiCoO₂, LiNiO₂ etc.), the LiFePO₄ is a favorable choice as cathode material for lithium-ion batteries in electric vehicle (EV) and medium- to largesized energy storage applications due to its stable and safe olivine structure as well as lower cost and environment-benign essence. However, the practical utilization of LiFePO₄/graphite batteries continuously reminds us that the safety issue is still a major challenge for this type of battery. Improper design and abusive operations are identified to be the major reasons causing safety accidents. A robust and powerful mathematical—physical model that addresses the complex mechanism of relevant processes and can be used as an effective tool for thermal analyses, structural design, and thermal management design of LiFePO₄/graphite batteries is thus critically needed.

Numerical modeling is widely used and plays an important role in the research of electrochemical and thermal performance of lithium-ion battery. Various electrochemical-thermal (ECT) coupling models for lithium-ion batteries were developed and extensively discussed in public literature [2–5]. These models are based on the electrochemical (EC) model proposed by Doyle et al. [6,7] in combination of an energy conservation equation with various heat generations/consumptions formulated. During battery operations, the heat generations/consumptions contain the Joule





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Nomenclature		Tamb	ambient temperature (K)	
		$T_{\rm ref}$	reference temperature (K)	
Α	side surface area of the electrode plate (m ²)	T _{surface}	temperature at the cell surface (K)	
as	specific surface area (m ⁻¹)	t	time (s)	
С	Li^+ concentration (mol m ⁻³)	t^0_+	transference number of Li ⁺ dissolved in the electrolyte	
Cp	specific heat capacity (J kg $^{-1}$ K $^{-1}$)	U	open-circuit potential (V)	
Ď	diffusion coefficient ($m^2 s^{-1}$)	V	cell voltage (V)	
F	Faraday's constant (C mol ⁻¹)			
h	convective heat transfer coefficient (W $m^{-2} K^{-1}$)	Greek sy	reek symbols	
Ι	current load (A)	α _a	anodic transfer coefficient	
i ₀	exchange current density (A m ⁻²)	α _c	cathodic transfer coefficient	
j^{Li}	transfer current density(A m ⁻³)	ε	porosity	
L	through-plane thickness of the battery (m)	η	surface overpotential (V)	
Le	through-plane thickness of the electrode (m)	κ	ionic conductivity (S m ⁻¹)	
р	Bruggeman factor	λ	thermal conductivity (W m ⁻¹ K ⁻¹)	
q	volumetric heat generation rate (W m^{-3})	ρ	density (kg m ⁻³)	
Q	heat generation rate (W)	σ	electronic conductivity (S m^{-1})	
r	radius of solid active particles (m)	φ	electric potential (V)	
R _{c1}	contact resistance at the electrode and current			
	collector interface (Ωm^2)	Subscripts/superscripts		
R_{c2}	contact resistance at the electrolyte and solid phase	e	electrolyte phase	
	interface (Ωm^2)	S	solid phase	
R _c	the total lumped contact resistance (Ωm^2)	neg	negative electrode	
R	universal gas constant (J mol ⁻¹ K ⁻¹)	pos	positive electrode	
Т	temperature (K)	sep	separator	

heat (ionic + electronic ohmic heat), ionic migration heat, irreversible EC reaction heat, reversible entropy heat, electric contact resistance heat at the current collector/electrode interface and that caused by the SEI layer covering the solid phase, and heat release to the ambient. All of these heats vary with time and space. Analyzing the spatial distribution and temporal evolution of these heats is very important to the design, operation and thermal management of batteries.

Srinivasan et al. [8], Zhang [9], and Ye et al. [10] analyzed the thermal behavior of LiMn₂O₄ battery. However they did not consider the heat generation due to electric contact resistance and a comprehensive thermal analysis was generally not conducted. For LiFePO₄/graphite batteries, the differences of physicochemical property such as, the temperature characteristics of open circuit potential that dictate heat generations, between LiFePO₄ and other cathode materials may lead to completely different thermal behaviors. Thermal analyses of LiFePO₄/graphite battery discharge/ charge processes have been seldom reported to date. In the present work we perform a comprehensive and systematical thermal analysis with respect to various discharge processes (1C, 3C and 5C) of a LiFePO₄/graphite battery based on the simulated results from a self-developed ECT coupling model. The ECT model solves the intercoupled charge (ion in the electrolyte and electron in the solid phase), species (Li⁺ in the electrolyte and lithium in electrode active materials) and energy conservation equations. Modeling results are validated by experimental data first. Then various heat generations/consumptions including the Joule heat, reversible entropy heat, contact resistance heat, irreversible EC reaction heat, ionic migration heat, and convective heat release to the ambient during discharge processes are calculated and analyzed in detail.

2. Model

The developed model is 3-dimensional, consisting of an electrochemical model and a thermal model, termed as ECT coupling model.

2.1. Electrochemical model

Fig. 1 schematically shows the physical model of a LiFePO₄/ graphite battery considered in the present ECT model. The cell consists of a copper current collector, a negative electrode, a separator, a positive electrode and an aluminum current collector. Both the electrodes and the separator are all porous. The electrode is composed of active material, electrolyte, polymer binder and conductive filler. The active materials of anode and cathode are commonly graphite mesocarbon microbeads and Li_yFePO₄ (LFP), respectively. The electrolyte is commonly lithium hexafluorophosphate (LiPF₆) in a mixture of propylene carbonate, ethylene carbonate and dimethyl carbonate. The porous separator is composed of liquid electrolyte and polymer matrix. Illustrated also in Fig. 1 is the assumed Li transport model in solid particles of anode and cathode active materials.

During charge/discharge processes, the electrochemical reactions occurring in electrodes can be expressed as follows.

in the anode composite electrode,

$$\operatorname{Li}_{x}C_{6} \xrightarrow{\operatorname{discharge}}_{\operatorname{charge}} 6C + x\operatorname{Li}^{+} + xe^{-}$$
(1)

in the cathode composite electrode,

$$\operatorname{Li}_{y-x}\operatorname{FePO}_4 + x\operatorname{Li}^+ + xe^- \xrightarrow[\text{charge}]{\text{discharge}} \operatorname{Li}_y\operatorname{FePO}_4 \tag{2}$$

The electrochemical model for LiFePO₄/graphite battery developed in this work is based on porous electrode and concentrated solution theories [6,7]. Major assumptions are described as the following.

(1) Electrode active materials are considered to be spherical particles of uniform size. Download English Version:

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