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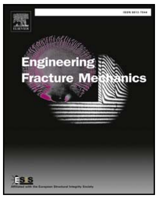

Predicting fracture evolution during lithiation process using peridynamics

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ABSTRACT

Silicon is regarded as one of the most promising anode materials for lithium-ion batteries due to its large electric capacity. However, silicon experiences large volumetric change during battery cycling which can lead to fracture and failure of lithium-ion batteries. The lithium concentration and anode material phase change have direct influence on hydrostatic stress and damage evolution. High pressure gradient around crack tips causes mass flux of lithium ions which increases the lithium-ion concentration in these regions. Therefore, it is essential to describe the physics of the problem by solving fully coupled mechanical-diffusion equations. In this study, these equations are solved using peridynamics in conjunction with newly introduced peridynamic differential operator concept used to convert partial differential equation into peridynamic form for the diffusion equation. After validating the developed framework, the capability of the current approach is demonstrated by considering a thin electrode plate with multiple pre-existing cracks oriented in different directions. It is shown that peridynamics can successfully predict the crack propagation process during the lithiation process.

1. Introduction

Performance of lithium-ion batteries mainly depends on material properties of anode, cathode and electrolyte. Several metals and compounds are selected as anode material such as cobalt, nickel, manganese and iron phosphate due to their performances in terms of thermal stability, capacity, conductivity and safety [1]. Silicon is also found to be one of the most promising anode materials in lithium-ion batteries due to its high theoretical charging capacity (3600–4200 mA Hg^{-1}) [2,3]. However, as lithium ions diffuse into silicon particles, the silicon particles experience a large volume expansion up to around 400% [2,4]. Frequent cycling of the lithium ion battery leads to stress misdistribution, degradation and delamination of the battery components which can significantly affect the battery performance.

Many studies were conducted on fracture analysis of silicon based anode. Liu et al. [5] developed a thin silicon film model to investigate the lithiation induced tensile stress and surface cracking by analytical and finite element methods. They observed a compression-traction transition zone located at lithiated and unlithiated material interface. Large volumetric expansion, plastic deformation and slow charging rate are the main factors which build up this transition zone and lead to cracks and fracture. They also argued that the magnitude and profiles of tensile stress at the surface of lithiated section depends on volumetric misfit strain, yield stress and modulus of unlithiated section.

Stamps and Huang [6] performed a mixed-mode fatigue evaluation of lithium-ion batteries by setting up a pre-cracked electrode model. They found that if a crack can propagate under certain loading when the length of a pre-existing crack is more than half size of

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Nomenclature

CCM	Classic Continuum Mechanics
FEM	finite element method
PD	peridynamics
C	current concentration
C_{avg}	average concentration of two material points (mol/m^3)
$C_{\max}$	maximum lithium ion concentration
E	elastic modulus or Young's modulus (N/m^2)
G_c	critical energy release rate (N/m)
$H_{\mathbf{x}}$	horizon of material point **x**
K	fracture toughness (Pa·m$^{0.5}$)
M	molecular mobility (m^2 J^{-1} s^{-1})
N_A	avogadro's constant
T	absolute temperature
Ω	partial molar volume (m^3 mol^{-1})
$\mathbf{b}(\mathbf{x},t)$	body force density of **x** at time t (N/m^3)
c	mechanical bond constant (N/m^6)
$dV_{\mathbf{x}'}$	volume of material particle **x**' inside the horizon of **x** (m^3)
f	pairwise force density (N/m^6)
h	thickness of electrode plate (m)
k_B	Boltzmann constant (JK^{-1})
s	bond stretch
s_c	critical bond stretch
$\ddot{\mathbf{u}}(\mathbf{x},t)$	acceleration of **x** at time t (m/s^2)
$\mathbf{u}(\mathbf{x},t)$	displacement of **x** at time t (m)
$\mathbf{u}(\mathbf{x}',t)$	displacement of **x**' at time t (m)
α	coefficient of expansion
δ	horizon size (m)
η	relative displacement of two material particles (m)
$\mu(t,\xi)$	failure parameter of bond at time t
ν	Poisson's ratio
ξ	relative position of two material points (m)
ρ	mass density (kg/m^3)
σ	cauchy stress (Pa)
$\tilde{\sigma}$	hydrostatic stress (Pa)
σ_0	first Piola-Kirchoff stress (Pa)
σ_{xx}	normal stress along x direction
σ_{yy}	normal stress along y direction
σ_{xy}, σ_{yx}	shear stress in x-y plane
$\varphi(\mathrm{x},\mathrm{t})$	damage of material point **x** at time t

the electrode width, the same loading will also cause propagation of a longer pre-existing crack in electrode. On the other hand, Ryu et al. [4] found that pressure gradient always occurs accompanied by a large volume expansion during normal cycling process and this will affect the process of lithium-ion diffusion in the electrode particles and crack evolution. Grantab and Shenoy [7] provided an investigation about pressure gradient factor on crack propagation in silicon nanowires. They used cohesive finite element method to model fracture in nanowires. In the early diffusion stage, since localised pressure around the crack tip is lower than surrounding nanowire surface region, large amount of lithium-ions moves into crack tip region which cause relatively large volume expansion and the stress around crack tip eventually reduces. On the other hand, Zuo and Zhao [8] used phase field method to study the stress evolution and crack propagation [8]. A series of damaged electrode models with different crack number and different crack orientations were considered to illustrate the evolution of fracture in an electrode. They indicated that pressure gradient factor depends on Young's modulus, partial molar volume, concentration, Poisson's ratio and the localised concentration around the crack tip region. Gao and Zhou [9] investigated the softening effects caused by lithiation induced fracture in electrode material. They developed a finite element framework and applied J-integral method to investigate the propagation of the crack tip. They also observed large amount of lithium-ion accumulation at the crack tip regions during charging process which causes a relaxation of hydrostatic stress.

As an alternative approach to finite element method (FEM), peridynamics (PD) can be utilised. Peridynamic theory is a new continuum mechanics formulation introduced by Silling [19] to overcome the problems that Classical Continuum Mechanics encountered especially for predicting crack initiation and propagation. Peridynamic theory is based on integro-diffential equations and these equations do not contain any spatial derivatives. Since its introduction, there has been a rapid progress on peridynamics. Several novel approaches have been proposed for efficient numerical solution of peridynamic equations such as dual-horizon concept [20] and adaptive refinement [21]. The technique has been applied to many different material systems [22,23] and extended for the analysis of multifield problems [24,25]. An extensive review on peridynamics can be found in Madenci and Oterkus [26].

In this study, peridynamic theory is used to investigate the fracture evolution in electrode plates of lithium ion batteries by considering pressure gradient and material phase change factors. Coupled field equations are expressed by using a combination of bond-based peridynamics [19] and peridynamic differential operator concept [10] to represent the relationships between the lithium ion concentration, hydrostatic stress and mechanical deformation. Several numerical cases with different crack numbers and crack orientations are considered and analysed.

2. Coupled diffusion-mechanical deformation mechanism

The general lithium diffusion phenomenon can be represented by using Fick's Second Law [28]. Since silicon is selected as anode material for lithium ion battery in this study, there is a large amount of volume change during lithiation and delithiation processes. Therefore, the stress induced by volume change during battery cycling should not be ignored. The stress components, σ_{ij}, can be expressed as [8].

$$\sigma_{xx} = \frac{E}{1-\nu^2}\left(\frac{\partial u_x}{\partial x} + \nu\frac{\partial u_y}{\partial y}\right) - \frac{E\Omega C_{\max}}{3(1-\nu)}C \tag{1a}$$