Journal of Energy Storage 13 (2017) 184–192

Contents lists available at ScienceDirect

Journal of Energy Storage

journal homepage: www.elsevier.com/locate/est

Modeling of thermo-mechanical stresses in Li-ion battery

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ARTICLE INFO

Article history:
Received 2 September 2016
Received in revised form 28 February 2017
Accepted 23 July 2017
Available online 1 August 2017

Keyword:
Battery Li-ion
Modeling
Mechanical
Thermal

ABSTRACT

The aim of this paper is to evaluate the thermo-mechanical behavior of a multilayer section of a lithium-ion 18650 cell during discharge. Discharging experiments with the measurements of voltage, current output and surface temperature were conducted. The model, developed in the framework of macroscopic approach, accounts for the heat generation during the battery discharge as well as the thermal expansion of each material layer of the jelly roll and can. For sake of simplicity, the mechanical behavior is assumed linear elastic and isotropic, the layers are perfectly tied and the model does not include the lithiation expansion. However, the approach helps to point out the evolution of stresses at the interfaces of the multilayer assembly during complete discharge. It introduces the question of interface cohesion/contact ageing as cycling repeats.

1. Introduction

Since the mid-'90s, Li-ion batteries are a promising energy storage system for various applications in stationary systems (smart-grids or autonomous systems) and mobile applications (Hybrid and Electric Vehicles). The advantage of lithium-ion batteries versus other storage technologies is obviously higher capacity and large working voltage (around 3.7 V or 3.2 V depending on the electrochemical couples used), leading to higher energy density, and smaller self-discharge rate.

However, during the cycling most of the materials degrade resulting in capacity fade. The reason for this capacity loss [1] can be active material volume expansion during intercalation process, modification of the composition of the passivation film on the negative electrode, and electrochemical instability of the materials or degradation of the positive electrode. Electrolyte, constituted by a mixture of different solvents as ethylene carbonate, propylene carbonate etc . . . and lithium salt and some specific additives, may undergo potential or thermal decomposition processes leading to the gases and in solution or solid species [2]. Especially at high potential or high temperature electrolyte could form gas bubbles inside the cell. For example, the thermal instability of lithium salt as LiPF6 leads to reactive gaseous species at elevated temperature [3,4]. Moreover, additive components may also decompose leading to gas generation in the cell [5]. This latter gas formation may induce internal pressure increase leading to cell degradation. Nevertheless, these phenomena are beyond the scope of this article.

As reported by Waldmann et al. [6] ageing mechanisms could be related to surrounding temperature. In particular, temperatures above 35 °C accelerate degradation reactions leading to capacity fade and internal resistance increase. Under some extreme conditions, as overcharge or exposure to very high temperature, the battery degradation can be very fast. This process is denoted as "thermal runaway".

The battery is subjected to many inter-dependent mechanisms as shown in Fig. 1. While discharging, lithium in its ionic state (Li^+) is extracted from the negative electrode (graphene planes of the graphite) and moves to the positive electrode (generally a metal oxide as $FePO_4$, or MnO_4) through the electrolyte. The movement of lithium is inverted during charging. At the electrode scale, lithium diffuses in the lattice of the electrode structure introducing structural changes and possibly significant chemical expansions leading to mechanical stresses. At the same time, heat generation takes place due to ohmic losses in the cell, charge transfer overpotentials at the interface and the mass transport limitations as well as reaction entropy. All of this induces thermal expansion leading to mechanical stresses.

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http://dx.doi.org/10.1016/j.est.2017.07.018
2352-152X/

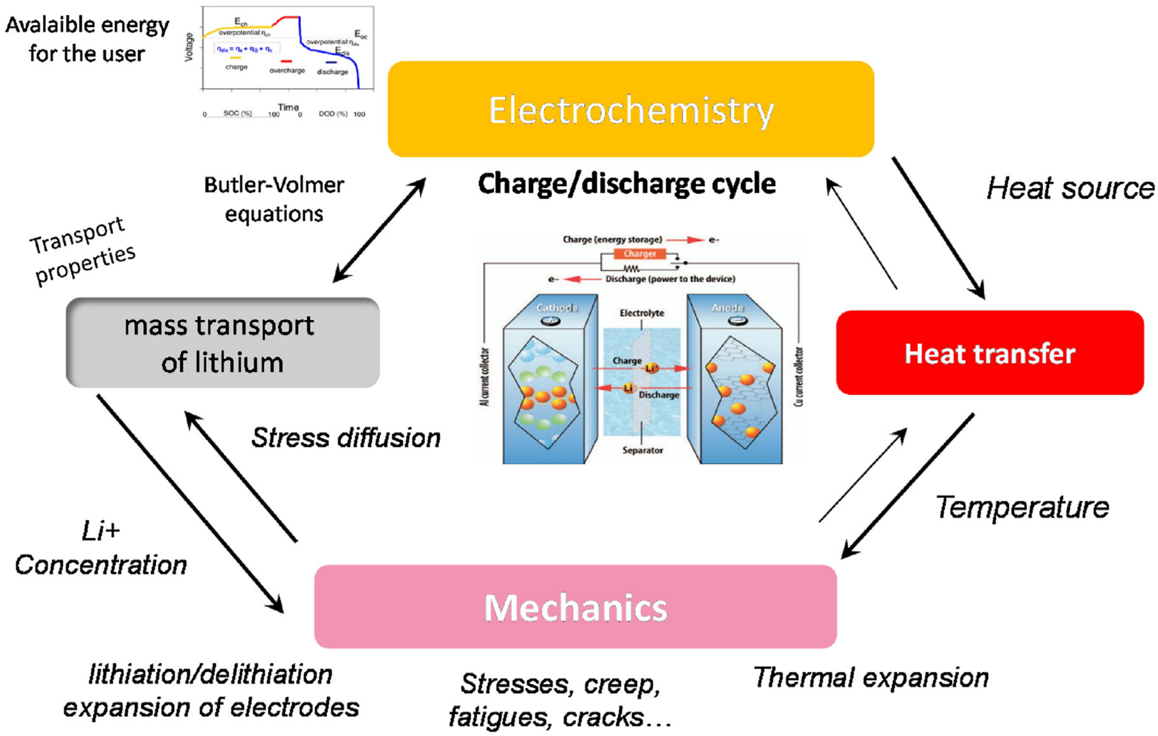

Fig. 1. Main coupling phenomena.

As the battery suffers mechanical deformations during cycling by both thermal and chemical ways, thermo-electro-mechanical models are crucial in understanding internal degradation in the cell. A summary of various modeling approaches, including mechanical aspect, is given in Table 1. The studies could be classified into three scales: particle-scale, electrode-scale and whole battery-scale.

In addition, Table 2 proposes a review of mechanical behaviors in the different parts of a battery [29–33]: cathode, anode, current collector and separator.

The aim of this paper is to underline the complexity of the mechanical loading in the multilayer section of an 18650 battery at macroscopic scale. The thermal behavior of a $LiFePO_4$/graphite lithium-ion 18650 battery has been investigated at ambient conditions during high C-rate discharges. Then, a thermo-mechanical model is developed to predict the thermal gradient inside the battery as well as the stress along the battery and especially at the material interfaces.

2. Experiments

Commercial $LiFePO_4$/graphite lithium-ion 18650 cells have been tested as they have better chemical stability and are less hazardous in case of abusive conditions [34]. The mean characteristics of the $LiFePO_4$/graphite batteries used in this study are: Nominal capacity = 1100 mAh; Dimensions = 65 × 18.1 × 18.1 mm; weight = 41 g and Nominal voltage = 3.2–3.3 V. The electrolyte is a mixture of ethylene carbonate (EC), propylene carbonate (PC),

Table 1
Review of models.

Modeling scale	Physical coupling	Ref.	Keywords	Software
Particle	Electro-chemo-mechanics Stress diffusion	[7,14]	Elastic-viscoplastic material, insertion induced cracking,	Matlab, Abaqus
		[9,26]	Porous electrode, intercalation induced stress	Matlab, Abaqus, Comsol
		[13]	Deformation-induced degradation, lithium concentration	Matlab
		[8,10–12,40]	Crack propagation during lithiation-delithiation	Abaqus
Sandwich or layer	Electro-chemo-mechanics	[17]	Capacity fade due to creep in separator	Matlab
		[18]	Mechanical stresses and degradation during intercalation, multiscale model	Comsol
		[16]		–
	Electro-thermo-chemo-mechanics	[19]	Multiscale model	–
		[15]	Mesoscale model	Matlab, Abaqus
Complete battery	Mechanics	[25,27]	Mechanical testing simulation, chock, impact, large deformation	Ls dyna
		[24]		Abaqus
		[22]		Code MMM (Volkswagen)
		[20]		ESI
	Electro-thermal	[21]	Heat source, Thermal modelling	Matlab
		[28,23,41]		Abaqus, Comsol