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Mechanical characterization and modeling for anodes and cathodes in lithium-ion batteries

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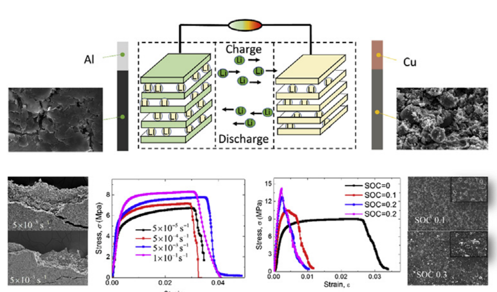
HIGHLIGHTS

- Electrode behaviors were characterized by mechanical-electrochemical factors.
- Mechanical behaviors of electrodes are strain rate, electrolyte and SOC dependent.
- Electrochemical status influences mechanical behaviors of electrodes greatly.
- Constitutive models for electrodes are established for future computation modeling.

GRAPHICAL ABSTRACT



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ABSTRACT

Mechanical properties of electrode materials have significant influence over electrochemical properties as well as mechanical integrity of lithium-ion battery cells. Here, anode and cathode in a commercially available 18650 NCA (Nickel Cobalt Aluminum Oxide)/graphite cell were comprehensively studied by tensile tests considering material anisotropy, SOC (state of charge), strain rate and electrolyte content. Results showed that the mechanical properties of both electrodes were highly dependent on strain rate and electrolyte content; however, anode was SOC dependent while cathode was not. Besides, coupled effects of strain rate and SOC of anodes were also discussed. SEM (scanning electron microscope) images of surfaces and cross-sections of electrodes showed the fracture morphology. In addition, mechanical behavior of Cu foil separated from anode with different SOC values were studied and compared. Finally, constitutive models of electrodes considering both strain rate and anisotropy effects were established. This study reveals the relationship between electrochemical dependent mechanical behavior of the electrodes. The established mechanical models of electrodes can be applied to the numerical computation of battery cells. Results are essential to predict the mechanical responses as well as the deformation of battery cell under various loading conditions, facilitating safer battery design and manufacturing.

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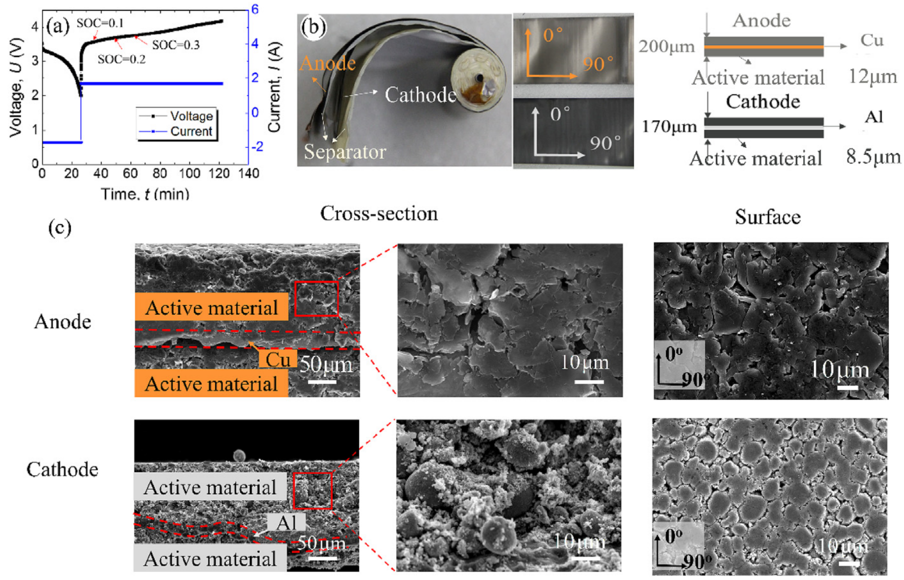

Fig. 1. Illustrations of (a) charge/discharge curves of 18650 LIB with NCA system under 0.5C charge/discharge rate; (b) unraveling of a jellyroll and three-layer composite structure of anode and cathode; and (c) The micro morphologies of anode and cathode from cross-section and surface views.

1. Introduction

Safety issues of lithium-ion batteries (LIBs) have been bottlenecks in further application of LIBs in electric vehicles [1–4]. The internal components (i.e. the separator, anode and cathode) would fail or fracture once the battery cell suffers from mechanical abusive loadings, triggering the irreversible internal short-circuit [5–7]. Then a large amount of heat is generated during the internal short-circuit and serious chemical decomposition reactions are ignited, followed by a great quantity of venting gas. This is referred to as the thermal runaway process, which causes the internal pressure to increase rapidly, eventually leading to fire or explosion [8–10]. Therefore, a full understanding of loading-dependent and electrochemical-dependent mechanical behaviors of lithium-ion battery components is essential and urgent in battery safe design, optimization and evaluation.

Pioneering studies on mechanical behavior of lithium-ion battery components were conducted from both micro- and macro-length scales. From micro-length level, stress evolution, distribution and fracture analysis of particles on electrode current collectors with the diffusion of lithium ions during charging and discharging were investigated [11–14]. The expansion of cathode/anode particles would cause grain fracture and thus lead to battery capacity deterioration or sudden failure [15,16]. Also, the charge/discharge would cause the exfoliation and failure of SEI membrane [17] as well as the adhesive failure due to electrode volume change [18,19]. In the meantime, volume change of electrodes due to intercalation/de-intercalation can also introduce extra stress within the battery [20]. From macroscale perspective, the mechanical properties of different types of separators, such as single layer [21] and composite polymer layers [22,23], were studied in respects of strain rate [24–26], temperature [24], solvent [27,28] and anisotropy [29] based on the mechanical experiments and numerical simulations. Xiao et al. [30] conducted creep test and frequency sweep, discovered the anisotropy of the separator. Arnold et al. [28] studied the *in-situ* mechanical behavior of polymer separators in solvent environment and found that the mechanical properties of separator would be weaken by solvent. Further, Xu et al. [31] performed comprehensive mechanical tests of both two types of separators, considering both the anisotropy and environment solvent effects coupled with the effect of strain rate. In the meantime, the in-plane tensile properties [32,33] and out-plane compression performances [34] of electrodes were studied with LIBs in $LiCoO_2$/$LixC_6$ system. Sahraei et al. [33] found that the active material particle would influence the mechanical properties of current collector. In addition, the SOC dependency of anode on out-plane compression properties was first discovered by Xu et al. [34]. Recently, Ma et al. [35] established an electrochemical-irradiated plasticity model of metallic electrodes and studied their yield strengths affected by SOC. Besides, the constitutive models were developed to describe the stress-strain response of electrodes, the most simple and general form is $\sigma = E\varepsilon$ (where σ is stress matrix, E is elastic constants matrix and ε is strain matrix), and the failure criterion is also established [36].

Since LIB cells with NCA cathodes are widely applied in electric vehicles due to high energy and power density, however, little is known about the mechanical behaviors of LIB cell and its component materials. To bridge this critical gap, in this paper, mechanical behavior of electrodes in the NCA cells is comprehensively investigated in terms of mechanical loading conditions (i.e. anisotropicity and strain rate dependency) and electrochemical status of the cells (i.e. SOC and solvent dependency). Furthermore, simple and effective constitutive models for both electrodes are established, facilitating further mechanical analysis and numerical simulation of LIB study.

2. Experimental

2.1. Materials

Material samples were taken out from fresh (discharged to cut-off voltage then charged to a specific SOC value) 18650 lithium-ion batteries based on a $Li_x(Ni_{0.80}Co_{0.15}Al_{0.05})O_2$ (NCA) cathode, whose charging and discharging characteristic curves are shown in Fig. 1a with the SOC values noted. The jellyroll of the lithium-ion battery shown in Fig. 1b was rolled in separator-cathode-separator-anode sequence. The electrode is a current foil coated with the particulate coating on both sides with thickness of 200 μm and 170 μm in anode and cathode, respectively. Specifically, the anode was 12 μm Cu foil coating with $Li_{0.008}C_6$ while the cathode was 8.5 μm Al foil coating with $Li_{0.925}(Ni_{0.80}Co_{0.15}Al_{0.05})O_2$. Here, the axial and rolling directions of battery are defined as 0° and 90°, respectively. Electrodes samples were prepared as square shapes for the following tensile tests, and one layer of electrode was tested in each tensile test.

Scanning electron microscope (JSM 6010L) was also employed to observe the microstructure of two electrodes, results are shown in Fig. 1c. Three-layer composite structure was observed for both anode