Materials and Design 95 (2016) 319–328

Contents lists available at ScienceDirect

Materials and Design

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

# Coupled effect of strain rate and solvent on dynamic mechanical behaviors of separators in lithium ion batteries

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ARTICLE INFO

*Article history:*
Received 30 September 2015
Received in revised form 16 December 2015
Accepted 9 January 2016
Available online 26 January 2016

*Keywords:*
Battery separator
Mechanical property
Anisotropy
Strain rate effect
Viscoelasticity

ABSTRACT

Mechanical properties of separators have a great impact on the electrochemical performance of lithium ion battery, such as capacity, charge/discharge behavior, charging cycles and among others. In the present study, two typical widely commercialized separators, Celgard 2400 and Celgard 2340, are the investigation objects with single layer and three-layer structures respectively. Firstly, to investigate material anisotropy and strain rate effects, tensile tests conducted on MTS and Instron at various strain rates from 0.01 to 50 $s^{-1}$ are carried out with samples prepared at three different directions: transverse direction (TD, 0°), machine direction (MD, 90°) and 45°. The failure strain decreases while failure stress increases with the strain rate for materials in all three directions. Material anisotropy is observed within the porous structure with nanofiber reinforced in the polymer matrix. Secondly, the environmental solvent effect is also examined: dimethyl carbonate (DMC) solution has a negative effect on the mechanical property of the separator while water may exert a positive effect. Furthermore, DMA Q800 is employed to study the viscoelasticity of separators at various temperatures and frequencies. A simple viscoelastic model based on Kelvin–Voigt model is proposed for the two types of separators. This research may serve as a solid step towards the comprehensive understanding of LIB separator and shed light on the future research of unveiling physical relation between mechanical properties and electrochemical performance of LIB.

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## 1. Introduction

Generally, the current widely commercialized separator in lithium-ion batteries (LIBs) is a type of porous polymer material placed between cathodes and anodes to separate the two electrodes and thus to avoid short circuit while providing channels for the transportation of lithium ions [1–5]. Separators usually consist of a polymeric membrane with one or multiple microporous layers [2,3] and they are required to be chemically and electrochemically stable as well as mechanically robust during the electrochemical reaction [6]. Regarding the mechanical properties, the separator should be strong enough to withstand the possible tension and compression of winding operation during LIB assembling [1]. Catastrophic consequences could follow with the failure of the battery separator such as short circuits, fires and even explosions [7]. Moreover, mechanical failure or deformation of separator is critical for evaluating the electrochemical performances including cell life and capacity degradation [8]. Therefore, mechanical properties of the separator have attracted much research attention with respect to the LIB safety. Previous studies have mainly focused on the essential properties of the separator such as porosity [1,2,9–11], pore size [1,2], thickness [1,2], chemical stability [1,2,12], permeability [1,2], mechanical strength [1,2,8,9,12], wettability [1,2,13,14], thermal stability [1,2, 9] and thermal shutdown [1,2,9], and which factors dominate are also discussed. A comprehensive and detailed review can be referred to Ref [1,2]. On the other hand, other researches [15–18] investigated the fabrication process with various composite materials and the physical, electrochemical, and thermal properties of some new materials were thoroughly discussed. Moreover, the composite polymer with various fillers as inert ceramic oxides ($SiO_2$ and $TiO_2$), ferroelectric material ($Al_2O_3$), super acid oxides, Nunes-Pereira et al. [19], single polymers, composites and polymer blends based on PVDF and its copolymers [20], whose main properties were summarized and compared with corresponding pristine polymer to find a method of increasing the performance. Well-designed experiments to explore the mechanical properties of the separators were reported in 2011 by Xiao et al. [8]. They conducted creep and frequency sweep experiment on single layer polypropylene separator and discovered the anisotropy of the separator due to the complexity of the material itself with oriented reinforcement fibers. To study the mechanical behaviors of the separator under compression, Arnold et al. [21] designed compression tests with

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http://dx.doi.org/10.1016/j.matdes.2016.01.082
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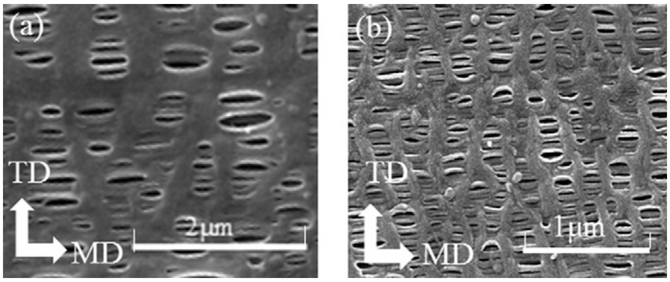


**Fig. 1.** Field emission scanning electron microscope image of (a) Celgard 2400 and (b) Celgard 2340.

**Table 1**
Experiment designs for coupling of strain rate, anisotropy and environment solvent of Celgard 2400 and Celgard 2340.

| Experimental types | Environment | Direction (°) | Strain rate (/s) loading stress (MPa) |
|---|---|---|---|
| Quasi-static | Dry | 0/45/90 | 0.01/0.1/1 |
| | Water | 0/45/90 | 0.01/0.1/1 |
| | DMC | 0/90 | 10/50 |
| Dynamic | Dry/water | 0/45/90 | 10/50 |
| Frequency scan | Dry | 0 | Scanning frequency 0.01–100 Hz |
| Temperature scan | Dry | 0 | Scanning temperature −120 °C–120 °C |
| Creep | Dry | 0 | 10/20/40 |

multiple layers of sample stacking and studied the mechanical behaviors at low strain rates. More research has concentrated on the influence of solvent [3,22] to reproduce the effect of the actual environment for the LIB separator. Chen [23] explored LIB's failure and safety that is associated with the mechanical behavior of separators. It was discovered that LIB's capacity is attenuated under compression with the influence of the closed pores of the separator. Recently, Yan et al. [24] explained the influence of environment solvent on the separator on a molecular scale using molecular dynamics simulation.

These pioneering investigations lay a concrete foundation for us to understand the mechanical behaviors of the separators made from specific materials at ambient conditions. However, the dynamic mechanical behaviors [25] of the separator still remain blank whereas the battery mechanical integrity usually fails [26,27]. Furthermore, the strain rate effect under ambient conditions and the anisotropy in both mechanical properties and morphologies need to be studied. This would help us to completely resolve the mechanical responses of the separator under dynamic loadings [28], e.g., vibrations or impacts which would accelerate the degradation of electrochemical performance, and on the other hand, the description of mechanical behavior at extreme loading conditions of LIB as a whole remains unsolved. Therefore, the current study mainly focuses on the investigations of the dynamic mechanical behaviors of the separator. Dynamic strain rates from 0.01/s to 50/s are chosen and mechanical properties from stress–strain curves are analyzed. Furthermore, the anisotropy and environment solvent effects are also considered, and are coupled with the effect of strain rate. This may reveal the mechanical behaviors in a more realistic way. Finally, the viscoelasticity of the separator are studied with temperature and frequency scanning sweep experiment on a DMA Q800 machine. The acquired information may help to build a deep understanding of mechanical behaviors of the separator and the corresponding LIB performance.

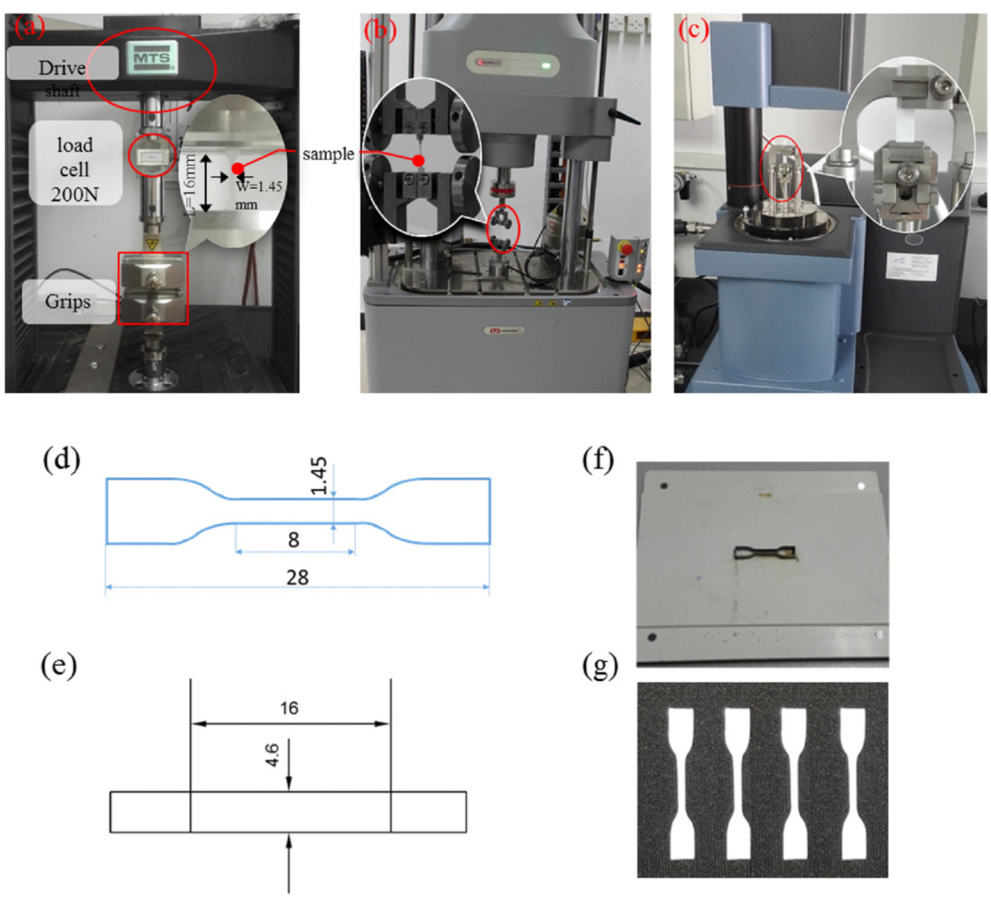


**Fig. 2.** (a) MTS criterion for quasi-static tensile test; (b) Instron E10000 for dynamic tensile test; (c) DMA Q800 for dynamic mechanical test; (d) the sample dimension of tensile test [30]; (e) the sample dimension of DMA; (f) Cutting tool; (g) the samples cut by cutting tool.

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