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Strain distribution and failure mode of polymer separators for Li-ion batteries under biaxial loading☆

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HIGHLIGHTS

- Biaxial mechanical behavior of separators studied in detail.
- Critical first principal strain at failure was determined by strain mapping.
- Finite element simulations correlated well with experiments.

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ABSTRACT

Deformation of polymer separators for Li-ion batteries has been studied under biaxial tension by using a dome test setup. This deformation mode provides characterization of separator strength under more complex loading conditions, closer representing deformation of an electric vehicle battery during crash event, compared to uniaxial tension or compression. Two polymer separators, Celgard 2325 and Celgard 2075 were investigated by deformation with spheres of three different diameters. Strains in separators were measured *in situ* by using Digital Image Correlation (DIC) technique. The results show consistent rupture of separators along the machine direction coinciding with areas of high strain accumulation. The critical first principal strain for failure was independent of the sphere diameter and was determined to be approximately 34% and 43% for Celgard 2325 and Celgard 2075 respectively. These values can be taken as a criterion for internal short circuit in a battery following an out-of-plane impact. A Finite Element (FE) model was built with the anisotropic description of separator behavior, derived from tensile tests in orthogonal directions. The results of simulations predicted the response of separator rather well when compared to experimental results for various sizes of rigid sphere.

1. Introduction

With the invention of new higher energy and power density electrode materials and advancements in manufacturing cost reduction, electrochemical energy storage got firmly established itself in automotive industry as a source of power for electric vehicles (EVs). Majority of EVs currently on the market are powered by lithium-ion battery packs. While the primary function of an EV battery pack is to deliver power to the drive train, the safety of the battery is as important as its power/energy performance characteristics. Such safety requirements drive the design of the battery packs in terms of thermal management and state of health monitoring as well as in terms of protection against external impact. The latter results in increase in weight of the pack due to necessity for building protective metallic enclosures and thus lowers the vehicle range due to that additional mass. Better understanding of the response of battery to external mechanical loads

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would lead to improved predictive simulation tools which in turn will aid design of advanced and safer battery packs that may require less, or differently arranged, protective armor.

Automotive lithium-ion batteries consist of multiple electrochemical cells, arranged into modules and connected electrically together in series or parallel. The basic building block is thus a cell, which usually is either a rectangular pouch containing stacked electrodes, or it has a wound configuration, as in case of cylindrical cells. Inside the cell, the charge is stored and released by moving lithium ions between the electrodes; the charge transported by ions is then transferred to electronic current that flows to external circuit. The physical separation of positive (cathode) and negative (anode) electrodes is achieved by placing a porous membrane which is made of material that is insulator for electrons; the pores provide pathway for ionic current through liquid electrolyte. This membrane is termed separator and is usually made of polyolefin materials, such as polypropylene (PP) and polyethylene (PE) [1,2]. The primary role of separator is to prevent contact between positive and negative electrodes while providing ionic conduction via electrolyte. Separator is therefore the most critical safety component inside the cell which prevents electrode contact and internal short circuits. Such critical role translates to requirements for thermal and mechanical properties of separators. The former is usually dictated by low thermal shrinkage, - as a typical requirement less than 5% shrinkage is allowed after 1 h at 90°C [3]. The general guiding principle for such requirement is prevention of exposure of electrode edges due to reduction of separator dimensions allowing these edges to contact each other and induce short circuit.

Mechanical properties are usually assessed by the puncture strength (ASTM D4830 [4]) as well as mix penetration strength parameter [3]. The latter is designed to characterize the resistance of separator to particle penetration, and is performed on a single cell sandwich (cathode/separator/anode) positioned on a rigid surface and being pressed by a rigid sphere from the top. During such test ("ball crush test" [3]) the cell sandwich is electrically connected to multi-meter, which registers drop in resistance due the internal short circuit. A conservative requirement for the mix penetration load is at least 100 kgf/mil (39 kN/mm), where apparently the length measure refers to the separator thickness. The puncture strength measurement is complementary to the mix penetration test, since the puncture strength was found to have correlation to the mix penetration strength [3]. In addition the test could provide information related to nail penetration test [5,6]. The puncture strength of separator should be at least 300 gf/mil (116 N/mm) [3]. Finally, the mechanical strength of separator is established in the direction where the tension from the winding machine during battery assembly is applied and is based on 1000 kgf/cm^2 (100 MPa) Young's modulus [1] or 2% offset strain corresponding to 1000 psi (6.9 MPa) [2].

Given its critical role in battery functionality and safety, battery separator warrants detailed investigation, specifically when its mechanical properties are concerned. While the majority of commercial separators are based on similar materials (PE and PP), there is a significant number of variations in terms of thickness, porosity or combinations of PE and PP membranes into multi-layered structure. A number of studies on mechanical behavior of Li-ion battery separators has been published relatively recently [7–13]. The reports mostly study tensile and/or compressive behavior of separators with or without liquid [9,11] and under different strain rates [8,12]. Rather comprehensive study has been published in Ref. [10], where properties of separators from three different manufacturers were compared in terms of uniaxial deformation at a fixed strain rate and in terms of resistance to tear; the mechanical experiments were complimented with atomic force microscopy (AFM) studies of microstructure. Analysis of uniaxial and biaxial deformation of two proprietary types of polymer separators was performed in Ref. [13] and two modes of internal short circuit were proposed depending on the separator failure pattern.

Previous work [12] has demonstrated use of digital image correlation (DIC) technique to map the strains in separators under uniaxial deformation. In the current investigation we report on biaxial deformation of two types of commercial polymer separators that are commonly used in battery manufacturing: a single PP layer and triple-layer PP/PE/PP separator designed for thermal shutdown. Deformation was studied under conditions of a dome test, where separator constrained at the perimeter is deformed by a rigid sphere. The strains in separator were mapped using DIC and the critical principal strain for failure has been measured. A finite element (FE) model has been built to simulate the biaxial deformation with material properties taken from the previous work on uniaxial behavior [12]. The results of FE analysis match the experimental observations well.

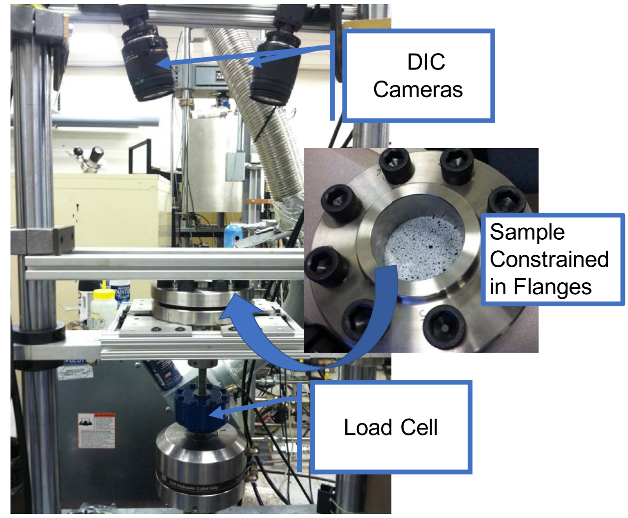

Fig. 1. Setup for biaxial testing.

2. Experiments

In order to gather more information on separator behavior, biaxial mechanical experiments were performed representing conditions closer to those involved in cell or battery deformation under out-of-plane impact by a rigid object. The following setup was assembled for this purpose (Fig. 1). The load frame was equipped with 100 lbs load cell (445 N). The circular sample of separator was placed between two stainless-steel 304L flanges. The edges of the inner flange opening were chamfered and smoothed; in addition rubber gaskets were placed between the flanges to avoid any tearing of the separator by steel. The nuts holding the flanges were hand tightened to avoid applying excess pressure to the perimeter of the sample. This provided enough force to keep the separator from pulling out of the flanges and yet to avoid over-tightening.

The deformation was imparted on the specimen by moving the polished hardened steel ball upward thus creating biaxial stretch of the separator sample. The approach is reminiscent of the deep drawing tests (or dome tests) done in sheet metal forming research. Spheres of three different diameters were used: 1 inch (25.4 mm), 2 inch (50.8 mm) and 2.5 inch (63.5 mm). The steel balls were mirror polished by the supplier (McMaster-Carr Supply Co). In addition they were sprayed by Teflon anti-friction coating. The ball was supported in the loading setup by the concave surface of the coupler; the latter was threaded to the load cell. There was no rigid attachment between the sphere and the load train of the machine, - this arrangement avoided any bending or torsion. The speed of the ball was maintained at 0.008 inch/s (0.2 mm/s) in all of the experiments. 24 samples were tested in total with two different types of commercial separators: Celgard 2325 and Celgard 2075. The mechanical properties of these separators have been reported previously [12]. The basic properties provided by the supplier are arranged in Table 1.