



Lithium modified silica as electrolyte additive for lithium secondary batteries

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ABSTRACT

Lithium sulfonyl silica (LSS) was synthesized by replacing the surface –H group in fumed silica with (CH₂)₃SO₃Li and adopted as electrolyte additive for lithium ion battery. 3 wt% of the synthesized particles in 1 M LiPF₆ (EC/DMC = 1:1) showed improved ionic conductivity and better potential stability over the pristine electrolyte. The discharge capacity of the LiCoO₂/graphite is particularly enhanced with the addition of LSS at higher C-rates due to the enhanced ionic conductivity at room temperature. The LiCoO₂/graphite cells using 1.0 M LiPF₆/EC/DMC (1: 1) and 1.0 M LiTFSI/EC/DMC (1: 1) with LSS also showed superior performance for the self-discharge test carried out at 45 °C for 200 days. These positive impacts of LSS on LiCoO₂/graphite cells warrant its use in lithium ion batteries.

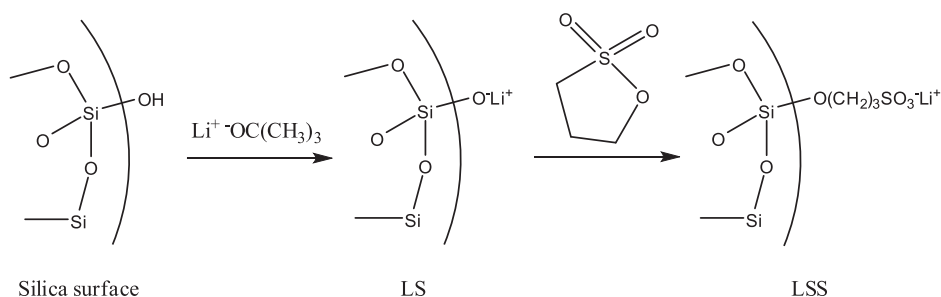
1. Introduction

Lithium secondary batteries enable charging and discharging through the intercalation and de-intercalation of lithium ions in the positive and the negative electrodes. This reaction is highly reversible and the role of electrolyte is highly important in achieving this reversibility. Recently, researchers are working tirelessly to develop new and efficient electrolytes to meet the demand of various electronic devices as well as electric vehicles which are still seeing sophisticated development [1–3]. Many literatures have reported myriad gel and solid polymer electrolytes containing ceramic additives (e.g., Al₂O₃, SiO₂, Fe₂O₃, TiO₂) [4–8]. The improvement of the electrochemical and physical properties of these electrolytes has made SiO₂ nanoparticles popular among battery researchers. Furthermore, their surfaces can easily be modified with metals and other molecules to increase the surface charge and functionality [9,10]. Compared with their polymer counterpart, SiO₂ forms gel by interacting with neighboring molecules (through –OH bonds) to form a three-dimensional network [11]. Some researchers have modified silica particles to act as both filler and origin of charge carriers [12,13]. Sun et al. and Choi et al. modified the surface of fumed silica by replacing the surface groups with lithium using lithium tert-butoxide as a source of lithium. Both authors used the lithium modified silica in fabricating polymer electrolyte and observed an improvement in lithium ion conductivity of the polymer electrolyte

and the performance of lithium ion battery using this polymer electrolyte [14,15]. Yongxing et al. also prepared nanocomposite gel by dispersing different forms of silica into liquid electrolytes. They observed a small decrease in ionic conductivity of the composite gel but improved cycle performance over the liquid electrolyte [16]. Again, Bhattacharyya et al. introduced small concentrations of different oxide fillers into liquid electrolyte and their composite liquid electrolyte showed enhanced ionic conductivity compared to the pristine electrolyte [17,18]. Furthermore, lithium modified silica nanoparticles synthesized from hydrophobic silica have been suggested as additive in regular liquid electrolyte [19]. The cycle performance at –20 °C and room temperature of graphite/LiCoO₂ cells employing the electrolyte with the silica nanosalt was particularly enhanced compared to the bare electrolyte [20]. Lithium hexafluorophosphate (LiPF₆) has attracted a lot of attention in lithium ion batteries, due to its superior characteristics such as high ionic conductivity, excellent electrochemical stability, but poor thermal stability and the side reaction in the presence of water discourages its usage. Researchers have proven that, the performance of LiPF₆ can be improved by adding an auxiliary salt. The inclusion of lithium bis(oxalato)borate (LiBOB) salt into LiPF₆ liquid electrolyte promoted the formation of the solid electrolyte interface film on graphite anode. Again, the combination of LiPF₆ and lithium difluoro(oxalato)borate (LiDFOD) saw an improvement in thermal stability of graphite/LiFePO₄ cells [21,22].

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Scheme 1. The simplified synthesis process of lithium sulfonyl silica (LSS) [14].

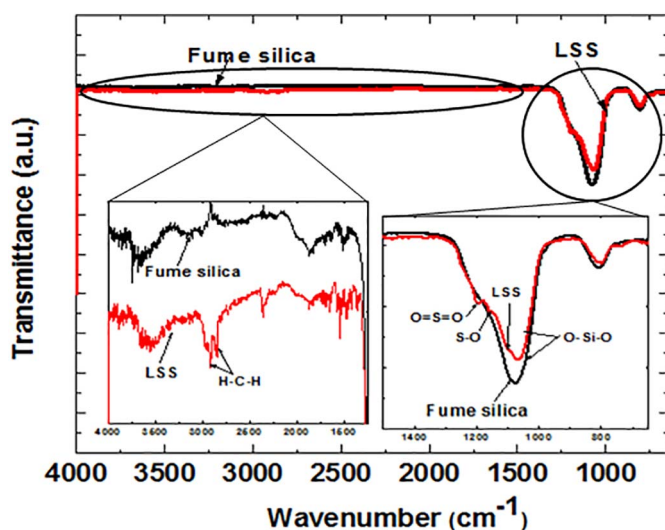


Fig. 1. TGA of LSS and the bare fume silica.

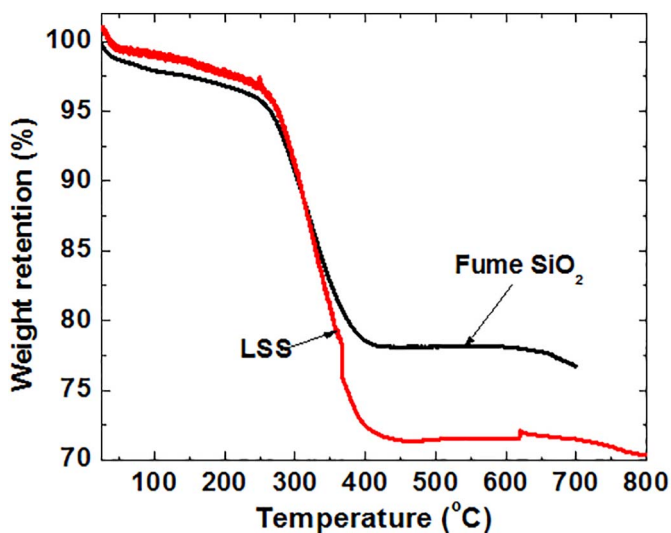


Fig. 2. FT-IR spectra of the LSS and the bare silica.

In this study, based on the synthesis procedure of reference 12, we synthesized $\text{SiO}_2\text{-Li}$. The $\text{SiO}_2\text{-Li}$ prepared was named lithium sulfonyl silica (LSS) and used throughout this paper. The LSS obtained is investigated as an electrolyte additive to augment the electrochemical performance of LiPF_6 -based electrolyte in attempt to improve rate performance, and thermal stability of graphite/ LiCoO_2 cells adopting this composite electrolyte.

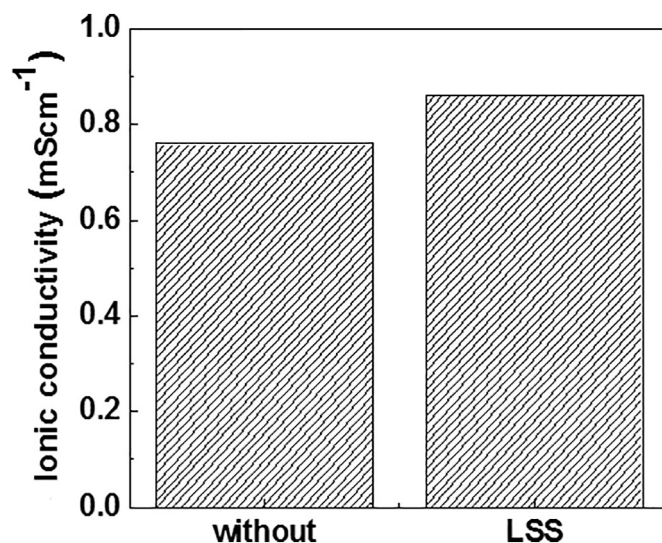


Fig. 3. Ionic conductivity of the electrolyte with and without LSS.

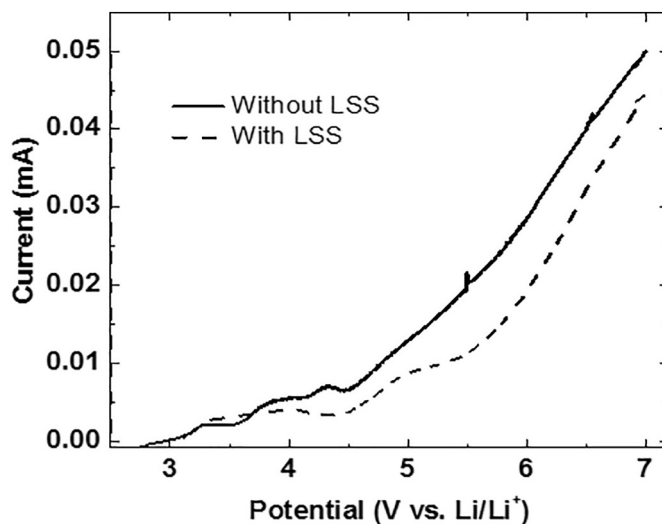


Fig. 4. Oxidation potential of the electrolyte with and without LSS at a scan rate of 1 mV/s, from 2.5–6 V vs. Li/Li^+ .

2. Experimental

2.1. Synthesis of LSS

SiO_2 (12 nm, Degussa, Aerosil®, Evonik) was first vacuum dried at 300 °C for 24 h to remove moisture. A mixture of 15 mL of 1.0 M lithium tert-butoxide (> 99.9% Aldrich) and 3 g of SiO_2 was stirred in 100 mL of THF (> 99.9% Aldrich) for 2 h. After 2 h, 4 g of 1,3-propanesultone (> 99.9% Aldrich) was added with continuous stirring at 70 °C for 48 h.

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