Supporting Information

Title: A High Performance Separator with Improved Thermal Stability for Li-ion Battery

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1. Experimental Section

Preparation of PPO/SiO_2 separators: 0.6 g of poly(phenylene oxide) (PPO, Sigma-Aldrich) and 0.1 g of silica nanoparticle (SiO_2, 15~20 nm, Sigma-Aldrich) were dissolved in a mixture of 4.25 mL of chloroform and 0.75 ml of butyl alcohol and subjected to vigorous stirring. The polymer solution was then cast on a glass plate, then dried at room temperature for 1 h, followed by further drying under vacuum at 60^{o}C for 12 h. The PPO separator PPOS was prepared by the same procedure, excluding SiO_2 from the polymer solution. A typical polyolefin-based separator (PE/PP, Celgard2325) was used as a standard separator.

Characterization of separators: The separator was characterized using a field emission scanning electron microscope (FE-SEM, Hitachi S-4700-II). To measure the electrolyte uptake, a separator was soaked in a liquid electrolyte composed of 1.2 M $LiPF_6$ in a mixture of ethylene carbonate (EC) and ethyl methyl carbonate (EMC) (3:7 wt%) for 12 h, and then the ratio of weight gain to the dry separator was calculated. The Li-ion conductivity was calculated from the ohmic resistance of coin cells assembled by sandwiching separators between two stainless-steel electrodes. The resistance of the separators was measured using a Solartron Multistat1470 coupled with a 1260 Frequency Response Analyzer System over a frequency range of 1 Hz to 10^6 Hz. The thermal integrity was investigated by heating a separator to 150 ^{o}C, 200 ^{o}C, and 250^{o}C for 30 min. To measure the porosity, a separator was soaked into a mineral oil for 12 h, and then the weight of separator containing the mineral oil was measured after removing the excess oil on the surface of separator. The porosity was calculated using the following equation:

$$\text{Porosity (\%)} = \frac{W_{sep+oil} - W_{sep}}{V_{sep} \times \rho_{oil}} \times 100$$

Where $W_{sep+oil}$ represents the weight of separator containing mineral oil, W_{sep} is the weight of the dry separator, V_{sep} is the volume of the separator, and ρ_{oil} is the density of mineral oil. The electrolyte uptake was evaluated by measuring the weight of the electrolyte absorbed in separator, and the weight percentage of the electrolyte in dry separator was calculated. The thermal behavior was studied by differential scanning calorimetry (DSC, Netzsch STA 449) from 50-260^{o}C at a scan rate of 10^{o}C min^{-1}. For battery performance evaluation, 2032-type coin cells were assembled using MCMB graphite as anode (Enerland), $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$ as cathode (Enerland), the prepared PSCS as separator and 1.2 M $LiPF_6$ EC/EMC (3/7 wt.) as electrolyte. Cell assembly was performed in a Argon filled glove box. The cells were tested between a voltage range of 2.7–4.2 V with a constant current of 0.1C (0.16 mA cm^{-2}) or 2C (3.2 mA cm^{-2}) charge-discharge rate at room temperature using a battery cycler (Maccor Series 4000). After 5 and 200 cycles, the Electrochemical Impedance Spectroscopy (EIS) of cells were measured using cells at the state of charge (SOC) of 50%. The harvested anode from the cycled cell was washed with EMC solvent and dried under vacuum at 60^{o}C for 12 h before the SEM measurement.

2. Effect of non-solvent and SiO_2 on Li-ion conductivity of PSCS:

To investigate the effect of non-solvent, PSCS were prepared with 15wt% SiO_2 and various volume percentage of non-solvent (butyl alcohol). The addition of non-solvent remarkably increased Li-ion conductivity of PSCS, however, the increment was not significant when the percentage is over 20% (Fig. S1a). The percentage of non-solvent was fixed as 15%, and then the effect of SiO_2 was investigated. As shown in Fig. S2b, the increase of SiO_2 content led to a higher Li-ion conductivity.

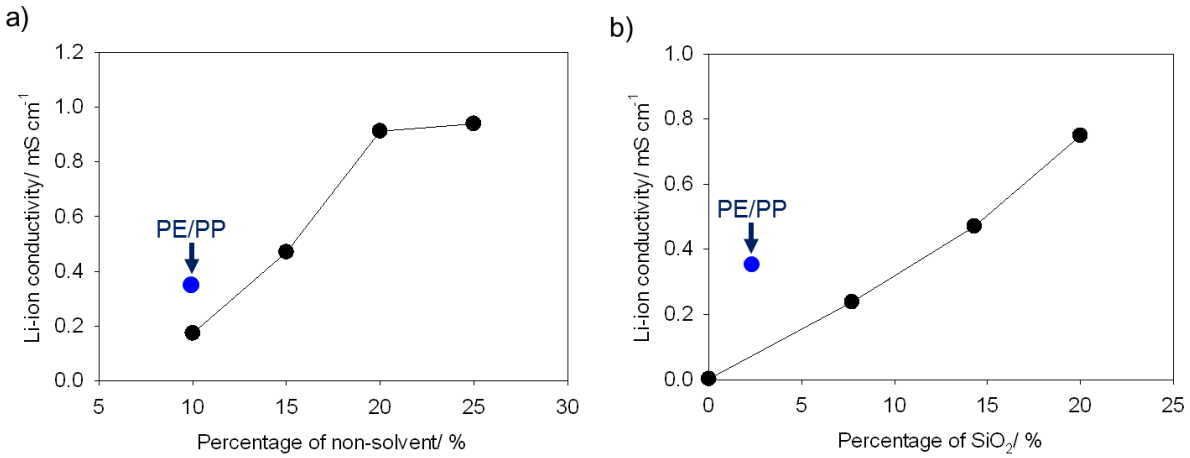

Fig. S1. Dependence of Li-ion conductivity of PSCS on: (a) the percentage of non-solvent and (b) SiO_2.

3. Properties of separators:

Typical properties of separators were evaluated and the results are listed in Table S1.

Table S1. Properties of separators

Separator	Thickness (μm)	Porosity (%)	Electrolyte Uptake (%)	Conductivity (mS cm^{-1})
PE/PP	25	54.1	110	0.380
PPOS	20	-	18	0.005
PSCS	21	60.4	162	0.470

4. Electrochemical stability of PSCS:

The electrochemical stability of the separators was evaluated using coin cells comprising of a stainless-steel working electrode, separator filled with liquid electrolyte and lithium foil as counter and reference electrode by a linear sweep voltammetry experiment with a scan rate of 1.0 mV s^{-1}.

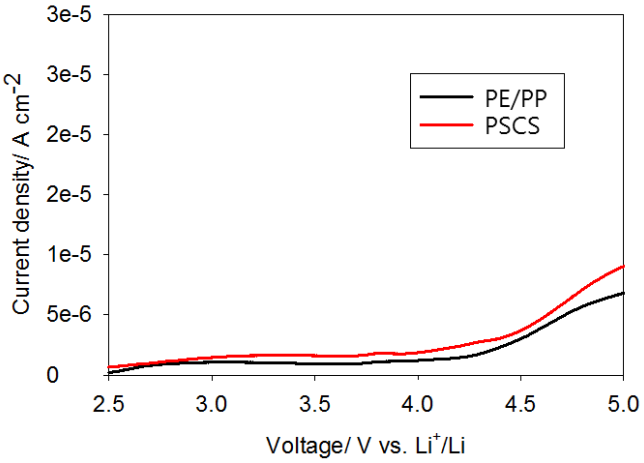

Fig. S2. Linear sweep voltammograms of PE/PP and PSCS.

5. Mechanical properties of PSCS:

Tensile strengths of separators (1" x 5") were evaluated by using the universal testing machine (Instron 3343) with a strain rate of 15 mm min^{-1}.

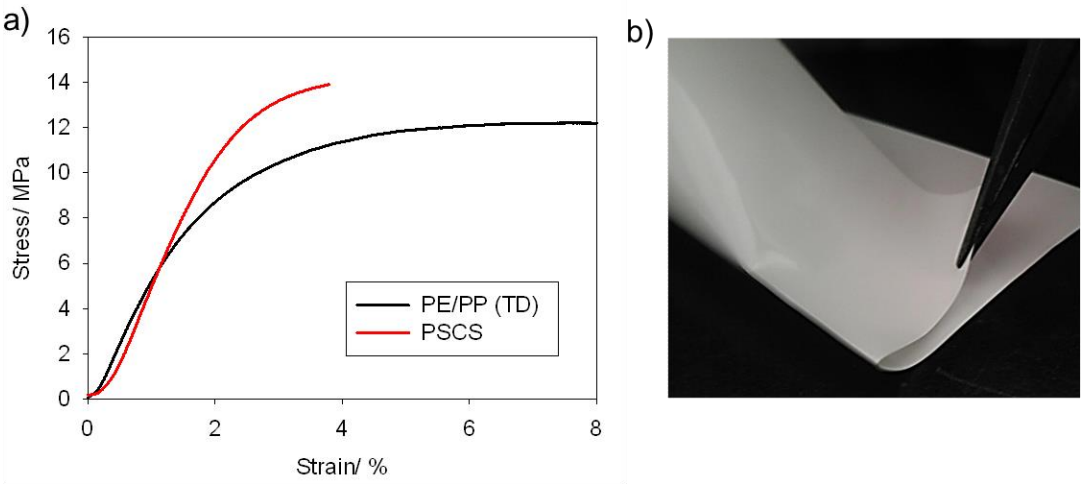

Fig. S3. (a) Stress-strain curves of PE/PP and PSCS and (b) Folded PSCS separator without crack.

6. C-rate performance of PSCS cells

After five formation cycles at 0.1C-rate, the PE/PP and PSCS cells were cycled at 0.2 C, 1 C, 2 C, 3 C, 4 C, and 5 C charge-discharge rate for 3 cycles and the data were recorded in Fig S4.

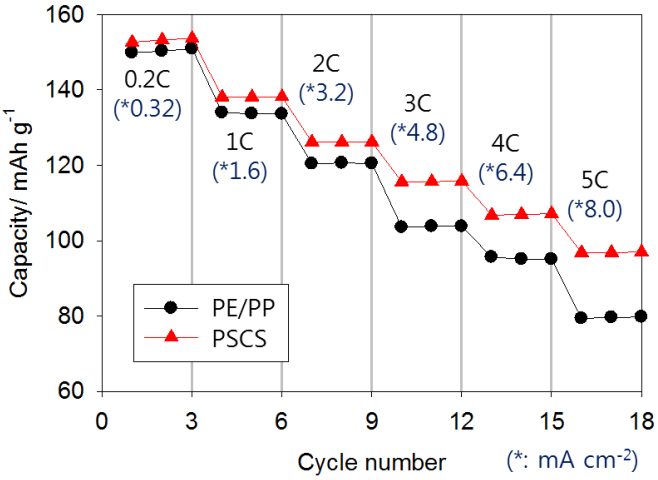

Fig. S4. Comparison of C-rate performance between PE/PP cell and PSCS cell, where charge-discharge C-rate are varied from 0.2 C to 5 C.

7. Electrochemical impedance spectroscopy (EIS)

EIS data were obtained from the PE/PP and PSCS cells at the state of charge (SOC) of 50% after 5 and 200 cycles.

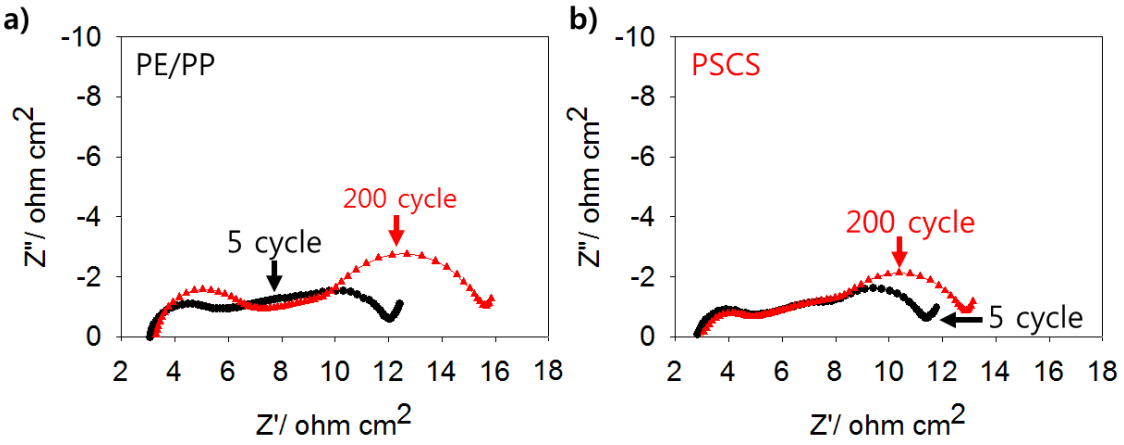

Fig. S5. EIS profiles of (a) PE/PP cell and (b) PSCS cell at the 5th and the 200th cycle.