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Electrochemical and morphological properties of separator induced by different stretching ratio

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The polyethylene (PE) based separator for lithium-ion rechargeable batteries has been investigated and compared with conventional triple layer (PP/PE/PP) Celgard 2340 separator using layered $Li[Ni_{1/3}Co_{1/3}Mn_{1/3}]O_2$ as a cathode material. The morphology, contact angle and electrochemical behavior of the 180% and 300% stretched PE separators were evaluated. The electrochemical result revealed that the initial discharge capacity of PE 180% and 300% stretched separator had 146.28 mAh/g and 133.43 mAh/g at C/5 rates, respectively. On the other hand, 300% stretched separator possesses better capacity retention ratio than the others and therefore demonstrated a good cycleability with less capacity fading than that of 180% stretched separator and conventional Celgard 2340 separator.

Keywords: Lithium-ion batteries, Separator, Microporous, Cathode material, Specific capacity

Introduction

The remarkable development of portable electronic devices has led to increasing demand on Li-ion battery (LIB) with high energy densities and capacities to facilitate compact and lighter portable equipment [1-4]. LIB consists of cathode, anode, electrolyte and separator. The separator is a membrane, which is the key component for LIB, and used for preventing battery safety failures because its primary function is to secure electrical isolation between a cathode and an anode of a battery [5-6]. Many experimental works have been published on inorganic separator [7-8], polyethylene terephthalate (PET) nonwoven inorganic composite separator [9] and nanofiber based separators.

In this study, we investigated the electrochemical properties depending on stretching ratio of the separator. Two different stretching ratios are allowed to manufacture the separators, but all these are in same chemical composition. 2032-type coin cells were assembled using attempted separators in argon atmosphere and their electrochemical properties such as potential profile, capacity and AC impedance were evaluated.

Experimental

The electrode of the cathode fabrication is started with mixing of 94 wt% of layered $Li[Ni_{1/3}Co_{1/3}Mn_{1/3}]O_2$ active material, 3 wt% of conductive carbon agent and 3 wt% of polyvinylidene fluoride (PVdF) which acts as a binder. The 2032-type coin cells were assembled for the 180 % and 300 % stretched separators (CS Tech), respectively, commonly using the prepared cathode, Li metal as an anode and 1.3M $LiPF_6$ EC/EMC (3/7 vol%). These were compared with cells adopted by the Celgard 2340 separator. The properties of each used separators are listed in Table 1. Stretching of PE separator was performed using uniaxial roll-to-roll stretching machine but the gap between rolls was maintained by same thickness. Therefore, 180 % and 300 % stretched samples had the same thickness. The physical contact angle of the separators at room temperature was measured by the KSV Sigma 701 Tensiometer (Finland). The morphology analysis of the each separator was observed using a field emission-scanning electron microscope (FE-SEM, FLeo Supra 55, Genesis 2000, Carl Zeiss). The electrochemical impedance spectroscopy (EIS) measurements were performed by IVIUM technologies instruments. Also, cycling test was conducted using a

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Table 1. The properties of 180%, 300% stretched PE separators and Celgard 2340 separator

	Celgard 2340 separator	180% stretched PE separator	300% stretched PE separator
Process	Dry	Dry	Dry
Composition	PP/PE/PP	PE	PE
Thickness	38 μm	20 μm	20 μm
Gurley	780 sec/100cc	400 sec/100cc	250 sec/100cc

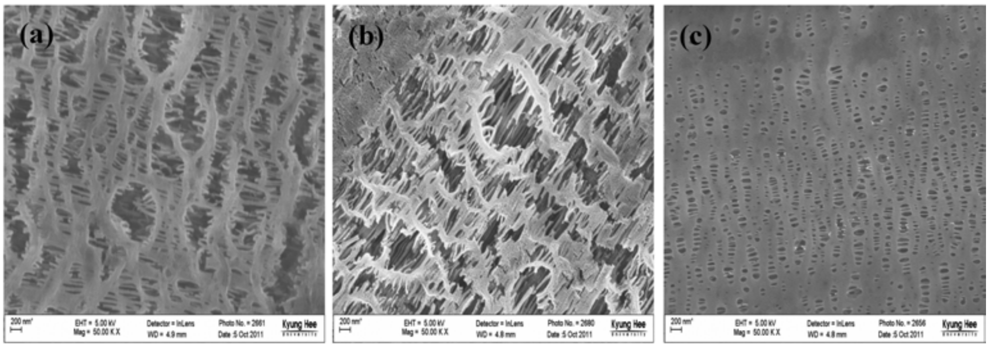

Fig. 1. FE-SEM surface morphology images of (a) PE monolayer separator, 180% stretched (b) 300% stretched and (c) Celgard 2340 separator.

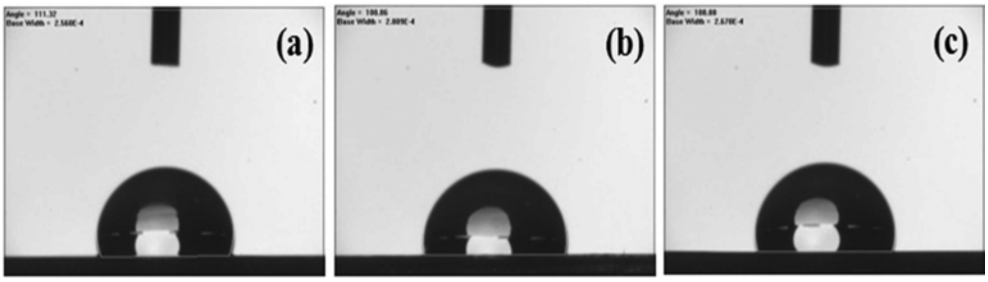

Fig. 2. Physical contact angle performance of images (a) PE monolayer separator, 180% stretched, (b) 300% stretched and (c) Celgard 2340 separator.

cycler (BT2000, Arbin) at current rate of C/10, C/5 and cut-off potential between 2.5 and 4.3 V vs. Li/Li^+. All electrochemical characterizations were carried out at room temperature.

Results and Discussion

Surface morphologies

Fig. 1 shows the surface morphologies of the three types of separators. 300 % stretched PE separator had larger pores and it contained more fibers behind of the first phase than 180 % stretched separator. This indicates that the indentation of 300 % stretched PE separator was higher than that of 180 % stretched PE separator. Fig. 1(c) shows the conventional Celgard 2340 separator with smaller pore size and thickness of 38 μm, which is thicker than PE monolayer separator (20 μm). Moreover, the PE separators had larger pores than that of the Celgard 2340 separator and maybe easily communicate with electrolyte between anode and cathode.

The wettability of the separator is crucial variable because separator with good wettability can contain the electrolyte effectively. It had been expected that the wettability of separators with large pores would be better. However, Fig. 2 shows the 180 %, 300 % stretched separator and conventional Celgard 2340 separator all exhibit hydrophobic nature from the contact angle measurement and those results of the contact angle did not match with our expectation. It might be attributed that the stretched separators had large pores but those had lower gurley values in comparison with the Celgard 2340 separator.

Electrochemical performance

Initial specific discharge capacity of 180 % stretched PE separator was 146 mAh/g and 300 % stretched PE separator had 133 mAh/g at C/5 rates and 2.5-4.3 V of cut-off voltage as shown in Fig. 3. Fig. 4 shows the capacity retention ratio of layered $Li[Ni_{1/3}Co_{1/3}Mn_{1/3}]O_2$ as the cathode material with different separators like PE stretched separators and Celgard 2340 separator at C/10 rates. It was interestingly found that the 300% stretched separator shows good capacity retention ratio, probably due to the limited ion transport of larger pores, whereas it shows the lowest capacity. Fig. 5 shows the AC impedance spectra of 180 %, 300 % stretched PE separator compared with Celgard 2340 separator. In this work, the impedance result confirms that the each 2032-type coin cells of separators (180 %, 300 % stretched and Celgard 2340) shows only one semicircle in the high and intermediate frequency and a 45° line in the low frequency. The charge transfer resistance of the 180% stretched separator is quite low. However, the 300% stretched separator had higher charge transfer resistance rather than the other two separators. It has been also correlated with the lowest specific capacity as shown in Fig. 3.

Conclusions

In this work, the 180 % and 300 % stretched PE monolayer separators were investigated and compared

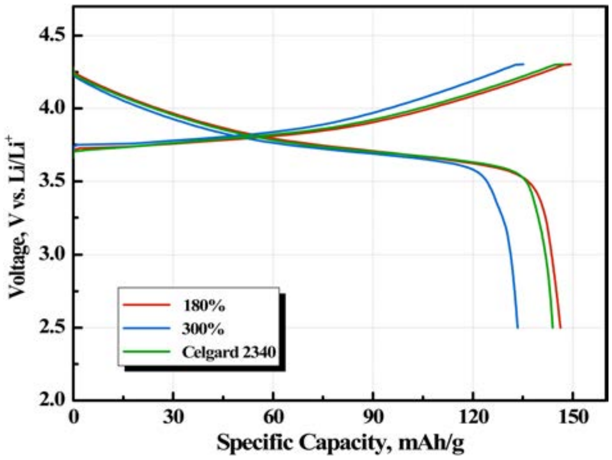

Fig. 3. The initial charge-discharge curves of PE monolayer separators and conventional Celgard 2340 separator at C/5 rates.

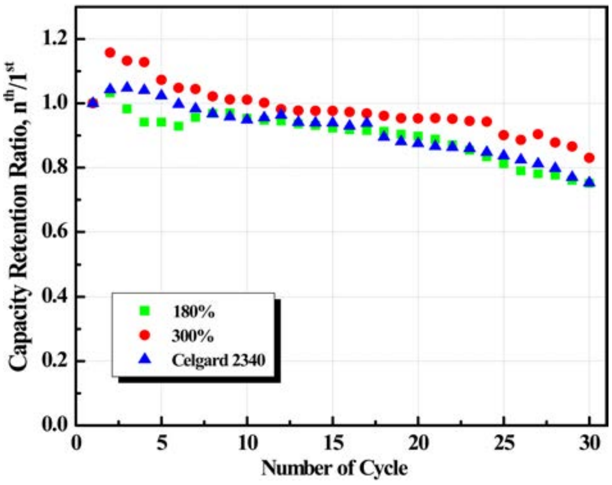

Fig. 4. Comparison of capacity retention of PE monolayer separators and conventional Celgard 2340 separator at C/10 rates.

with conventional PP/PE/PP layer Celgard 2340 separator using layered $Li[Ni_{1/3}Co_{1/3}Mn_{1/3}]O_2$ cathode material. The 300 % stretched PE separator had higher charge transfer resistance than the 180 % stretched PE separator and also the initial discharge capacity of the 300 % stretched separator was found to be lowest at 133 mAh g^{-1}. However, its capacity fading was less than 180% stretched separator and Celgard 2340 separator during 30 cycles. Therefore, 300% stretched separator delivers lower specific capacity at initial

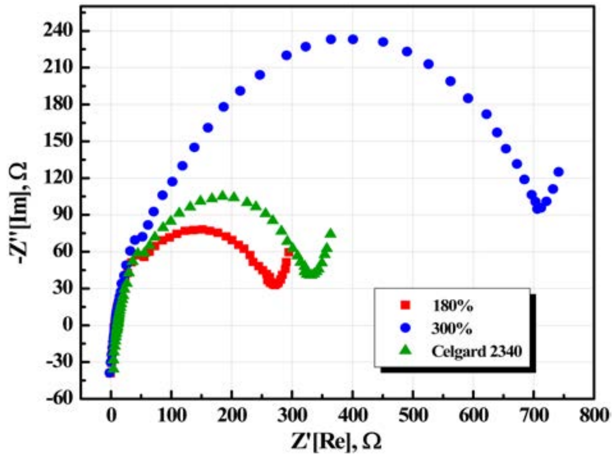

Fig. 5. The Nyquist plot of electrochemical impedance spectra of CS tech PE separator (300%, 180% stretched) and conventional Celgard 2340 separator.

cycling but it seems to be relatively in a good cycling stability when compared with the 180% stretched and Celgard 2340 separators.

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