

## PDMS/GO 복합체 박막의 리튬 금속 표면 개질: 리튬전극의 성장 제어 및 리튬금속전지(LMB) 성능 향상

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### Surface Modification of Li Metal Electrode with PDMS/GO Composite Thin Film: Controlled Growth of Li Layer and Improved Performance of Lithium Metal Battery (LMB)

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**요 약:** 리튬금속전지(LMB)는 매우 큰 이론 용량을 갖지만 단락(short circuit), 수명 감소 등을 야기하는 덴드라이트(dendrite)가 형성되는 큰 문제점을 갖고 있다. 본 연구에서는 poly(dimethylsiloxane) (PDMS)에 graphene oxide (GO) nanosheet를 고르게 분산시킨 PDMS/GO 복합체를 합성하였고 이를 박막 형태로 코팅하여 덴드라이트의 형성을 물리적으로 억제할 수 있는 막의 효과를 이끌어내었다. PDMS의 경우, 그 자체로는 이온 전도체가 아니기 때문에 리튬 이온의 통로를 형성시켜 리튬 이온의 이동을 원활하게 하기 위하여 5wt% 불산(HF)으로 에칭하여 PDMS/GO 박막이 이온전도성을 가질 수 있도록 하였다. 주사전자현미경(scanning electron microscopy, SEM)을 통해 전면 및 단면을 관찰하여 PDMS/GO 박막의 형상을 확인하였다. 그리고 PDMS/GO 박막을 리튬금속전지에 적용하여 실시한 배터리 테스트 결과, 100번째 사이클까지 쿨롱 효율(columbic efficiency)이 평균 87.4%로 유지되었고, 박막이 코팅되지 않은 구리 전극보다 과전압이 감소되었음을 전압 구배(voltage profile)를 통해 확인하였다.

**Abstract:** Although Lithium metal battery (LMB) has a very large theoretical capacity, it has a critical problem such as formation of dendrite which causes short circuit and short cycle life of the LMB. In this study, PDMS/GO composite with evenly dispersed graphene oxide (GO) nanosheets in poly (dimethylsiloxane) (PDMS) was synthesized and coated into a thin film, resulting in the effect that can physically suppress the formation of dendrite. However, PDMS has low ion conductivity, so that we attained improved ion conductivity of PDMS/GO thin film by etching technic using 5wt% hydrofluoric acid (HF), to facilitate the movement of lithium (Li) ions by forming the channel of Li ions. The morphology of the PDMS/GO thin film was observed to confirm using SEM. When the PDMS/GO thin film was utilized to lithium metal battery system, the columbic efficiency was maintained at 87.4% on average until the 100<sup>th</sup> cycles. In addition, voltage profiles indicated reduced overpotential in comparison to the electrode without thin film.

**Keywords:** lithium metal battery, PDMS/GO thin film, ion transport, dendrite growth control

## 1. Introduction

Electronic devices such as electronic vehicles (EVs), mobile phones, and wearable devices, require batteries

with higher energy density. Currently, commercialized lithium ion battery (LIB) uses graphite which is chemically stable with good electrical conductivity as an anode material. However, graphite has a very low the-

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oretical capacity of 372 mAh/g, which limits its application to electronic devices. Among the anode materials of secondary batteries, Li metal is one of the promising alternative candidates of graphite because of its large theoretical capacity of 3,860 mAh/g and lowest reduction potential. However, there is a problem that Li dendrite is formed during charging and discharging reactions. Dendrite means that crystal nuclei are generated in the process of plating and stripping of Li ions. Afterwards, Li is accumulated in the form of branches around the nuclei. The growth of dendrite leads to infinite volume expansion of Li metal, reduced battery life, and producing dead Li. In addition, in severe cases, dendrite can penetrate the separator and cause the battery to explode[1-10].

In order to solve this problem, many studies are being conducted to physically suppress dendrite by coating a thin film on the anode. However, using the thin film to suppress dendrite has to be fulfilled the following conditions. That is, the thin film must be chemically stable to not react with the electrolyte and have sufficient mechanical strength to suppress dendrite. It must also be flexible enough to accommodate the volume expansion of Li and have sufficient Li ion conductivity[11-19].

In this study, we designed the PDMS/GO composite thin film that meets all of the above conditions. PDMS is chemically stable, flexible and has high mechanical strength. GO nanosheet has excellent mechanical strength, thermal/chemical stability, and has affinity with Li ions due to functional groups containing oxygen atoms such as epoxy, carboxyl and hydroxyl groups. Thus, the PDMS/GO thin film with evenly distributed GO nanosheets in PDMS could suppress the formation of dendrite in an electrolytic atmosphere. However, since PDMS is not an ion conductive material, Li ions cannot pass through thin films and move onto the anode. Accordingly, the PDMS/GO thin film was etched for 10 min with 5 wt% HF to form the channel of Li ions[19-22].

In conclusion, HF-etched PDMS/GO thin film suppresses the formation of dendrite through high mechan-

ical strength and chemical resistance, and prevents direct contact between highly reactive Li metal and electrolyte, resulting in improved performance of lithium metal battery through solving the low columbic efficiency and short lifespan and increase of overpotential.

## 2. Experimental Section

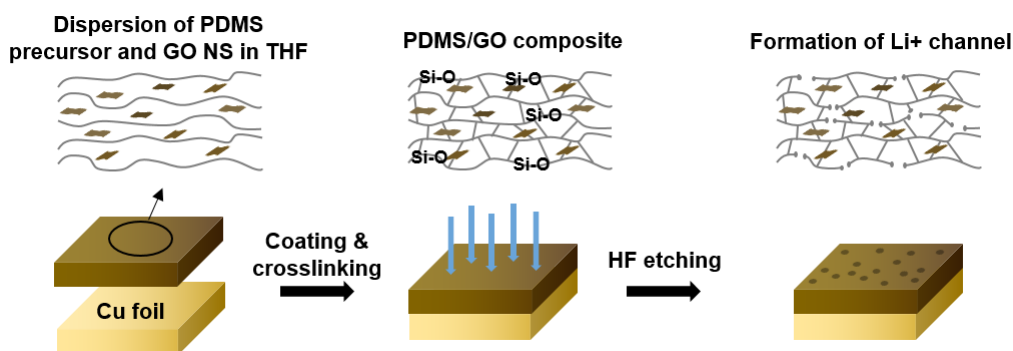
### 2.1. Materials

Sulfuric acid ( $\text{H}_2\text{SO}_4$ , 98 wt%), potassium permanganate ( $\text{KMnO}_4$ ) and hydrogen peroxide ( $\text{H}_2\text{O}_2$ , 30 wt%) were purchased from Ducusan. Phosphoric acid ( $\text{H}_3\text{PO}_4$ , 85 wt%) and Graphite powder were purchased from Daejung. These were used to synthesize GO nanosheets. PDMS elastomer (Sylgard 184A) and crosslinker (Sylgard 184B) were purchased from Dow corning and used to synthesize poly (dimethylsiloxane). Then, tetrahydrofuran (THF) was purchased from Samchun and used as a solvent for PDMS/GO composite. hydrofluoric acid (HF, 5 wt%) was used to etch the PDMS/GO composite thin film.

### 2.2. Fabrication of electrodes coated with PDMS/GO composite thin film

#### 2.2.1. Synthesis of Graphene oxide nanosheets

Graphene oxide (GO) nanosheets were synthesized by modified Hummer's method. First, the mixture of  $\text{H}_2\text{SO}_4$  and  $\text{H}_3\text{PO}_4$  was stirred in an ice bath to lower the temperature, and graphite was added into the solution.  $\text{KMnO}_4$  was slowly added while maintaining it not to exceed 20°C. The graphite was oxidized by stirring for 12 h while maintaining the temperature of the solution at 50°C to form graphite oxide. After the solution was cooled to room temperature, it was slowly added to the beaker containing ice so that the temperature did not rise sharply.  $\text{H}_2\text{O}_2$  was added to the solution until the color became bright and stirred at room temperature. By centrifugation, the graphite oxide particles were collected and re-dispersed in distilled water. The re-dispersed solution was exfoliated by sonication to obtain a GO solution having good dispersibility in distilled water[23-28].



**Fig. 1.** Schematic illustration of the formation of PDMS/GO thin film on the Cu foil.

#### 2.2.2. Synthesis of PDMS/GO composite and preparation of electrodes

For the synthesis of PDMS/GO composite, PDMS elastomer was added to THF and stirred. Then, the GO solution was sonicated and dispersed in THF for 2 h, and then poured in the PDMS elastomer dispersion and sonicated again to disperse the PDMS elastomer and GO nanosheets in THF. Then, crosslinker was added and stirred to synthesize the PDMS/GO composite in which GO nanosheet particles were evenly dispersed on PDMS[29-31]. Cu foil was pretreated with HCl to remove foreign matters, and the previously prepared composite was blade coated. Then, the composite was then vacuum dried at 70°C for 5 h to cure and remove the solvent and etched with HF for optimum time.

#### 2.3. Material characterization

Scanning electron microscope (FE-SEM, HITACHI S4800) was used to confirm the uniform formation of PDMS/GO film and ion channels with HF etching time.

#### 2.4. Electrochemical characterization

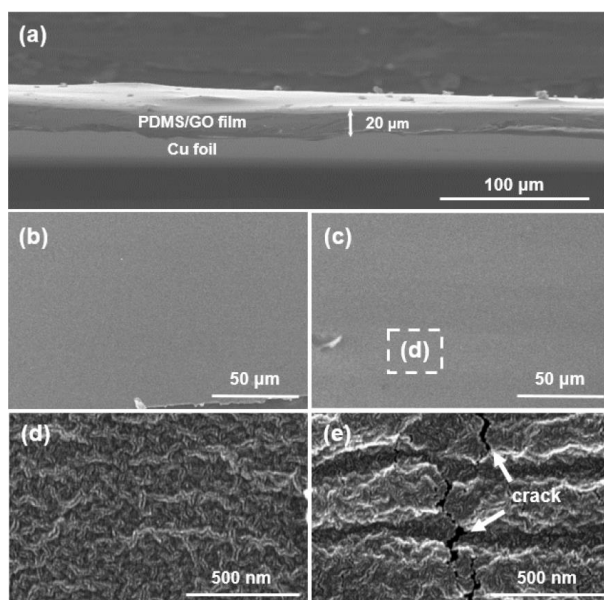
In order to confirm electrochemical performance, CR-2032-Type Coin Half-Cells were fabricated using Li foil as a counter electrode and PDMS/GO thin film-coated electrode as a working electrode. As electrolyte, LiTFSI dissolved in 1,3-dioxolane (DOL)/1,2-dimethoxyethane (DME) was used. Coin cell was manufactured in an argon filled glove box. Confirmation of the electrochemical performance was performed by using a battery tester (Wonatech Co, Ltd, WBCS3000).

Electrochemical performance was measured by plating up to 1 mAh/cm<sup>2</sup> and stripping up to 1 V, at current densities of 0.5 mA/cm<sup>2</sup> upto 10<sup>th</sup> cycle for stable formation of initial Li nuclei. After that, the measurement was performed at a current density of 1 mA/cm<sup>2</sup>.

### 3. Results and Discussion

Fig. 1 shows the formation of PDMS/GO thin film on Cu foil. As described above, the PDMS/GO composite was synthesized and blade-coated on the Cu foil to form a thin film. It was vacuum-dried to cure the film and remove the solvent, then etched with 5% HF to form Li ion channels[19,29-31].

SEM analysis was performed to confirm the morphology of synthesized PDMS/GO composite thin film (Fig. 2). Fig. 2 (a) shows a cross-section SEM image of the thin film formed by blade-coating. The PDMS/GO composite cured through vacuum-drying. During curing the thickness of the PDMS/GO composite thin film was uniformly formed to 20  $\mu$ m. Fig. 2 (b) is a top-view SEM image of the PDMS/GO composite thin film before etching treatment. Fig. 2 (c), (d) and (e) indicate top-view SEM images of the PDMS/GO composite thin film by etching with HF to form the Li ion channels. Fig. 2 (c) and (d) show the SEM images of PDMS/GO composite thin film etched for 10 min with 5% HF. Fig. 2 (d) exhibits the high-resolution SEM image of the PDMS/GO composite with etching for 10 min. Fig. 2 (e) is a high-resolution SEM image of the thin film etched for 15 min. Compared to the PDMS/



**Fig. 2.** (a) Cross-section SEM image of PDMS/GO thin film, (b) top-view SEM image of PDMS/GO thin film (before etching), (c) top-view SEM image of PDMS/GO thin film (etching for 10 min), (d) top-view high-resolution SEM image of PDMS/GO thin film (etching for 10 min), (e) top-view high-resolution SEM image of PDMS/GO thin film (etching for 15 min).

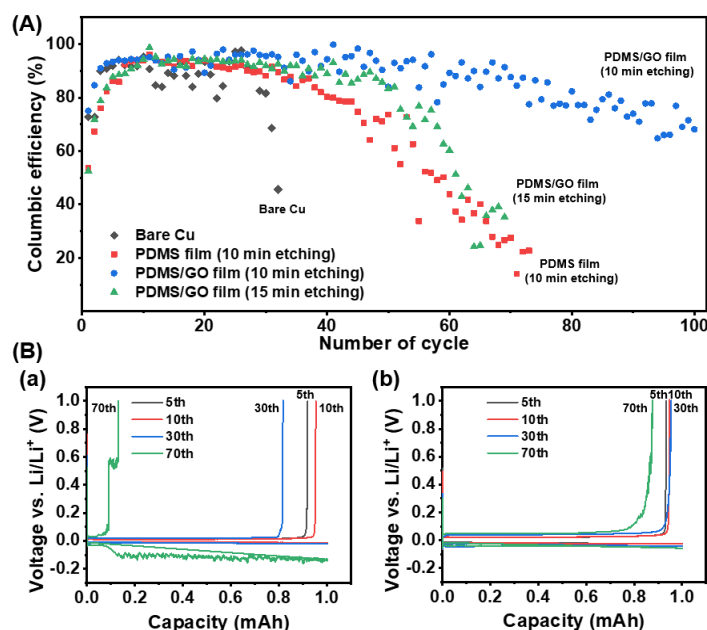
GO composite thin film etched for 10 min, the PDMS/GO composite thin film etched for 15 min was found to have excessively large ion channel size formed by HF etching, and the thin film was damaged.

The synthesized PDMS/GO composite was used as a thin film and applied to lithium metal battery system by conducting electrochemical performance test. Fig. 3 (A) shows coulombic efficiency with cycles of bare Cu (untreated one) electrode, PDMS etched for 10 min, and the PDMS/GO composite thin film electrode etched for 10 min and 15 min, respectively. Coulombic efficiency can be defined as the ratio of the amount of charge during stripping and plating at each electrode, which is considered an important parameter of lifespan of a battery. In the bare Cu electrode, the coulombic efficiency was drastically lowered due to the formation of dendrite before the 30<sup>th</sup> cycle. This low efficiency is due to less stripping and the less Li resulting from non-uniform Li deposition on the electrodes, which generates of inert Li[18-19]. In contrast, the coulombic

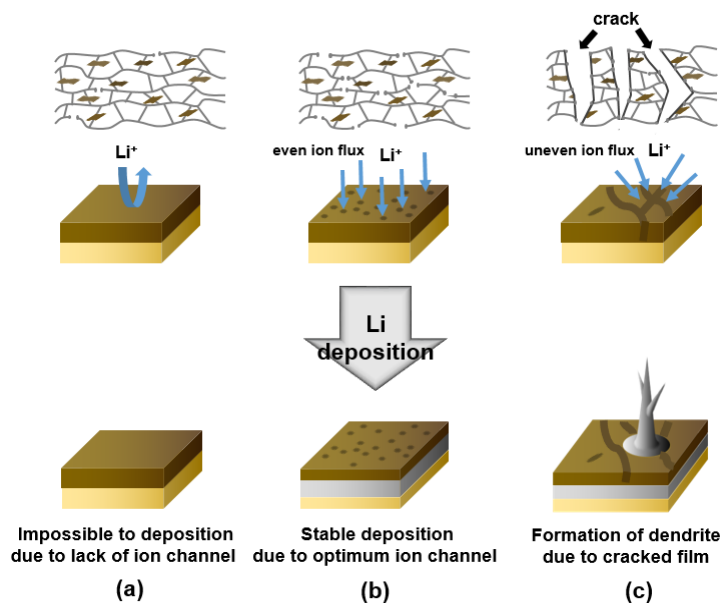
efficiency of PDMS thin film electrode etched for 10 min was maintained about 85.4% on average up to 50<sup>th</sup> cycles, and the coulombic efficiency of PDMS/GO thin film electrode etched for 10 min was maintained at 87.4% on average up to 100<sup>th</sup> cycles, which is beneficial to the function of GO nanosheets. The PDMS/GO thin film electrode etched for 15 min was measured to confirm the effect of the HF etching time. The PDMS/GO thin film electrode etched for 15 min showed a sharp decrease in coulombic efficiency at the 50<sup>th</sup> cycle due to the damaged thin film, as shown in Fig. 2 (c). Therefore, it was confirmed that the optimum HF etching time was 10 min.

Fig. 3 (B) (a) and (b) show the voltage profile of the bare Cu electrode and the PDMS/GO thin film electrode. Voltage profiles of the 5, 10, 30, and 70<sup>th</sup> cycles were analyzed. In the case of bare Cu electrode, its overpotential increases due to resistance according to non-uniform solid-electrolyte interface (SEI) layer and dendrite formation. On the other hand, the PDMS/GO composite thin film electrode shows the voltage reached to 1 V at constant capacity as compared to the bare Cu electrode in the 5, 10, 30<sup>th</sup> cycles. In the 70<sup>th</sup> cycle, a slight overpotential caused the voltage to rise at a slightly lower capacity than the previous cycles, resulting in a more stable voltage profile. The voltage of both electrodes reached to 1 V at a lower capacity at the 5<sup>th</sup> cycle than the 10<sup>th</sup> cycle, which can be seen as a result of the formation of the SEI due to the side reaction between the electrolyte and Li in the initial cycle[16,19].

Fig. 4 shows the effect of between HF etching time and Li deposition behavior in the PDMS/GO composite, by comparing the Li ion transport behavior in electrodes with different etching time (5~15 min). As shown in Fig. 4 (a), if the etching time is shorter than 10 min, the channels where Li ions cannot pass sufficiently are formed. Therefore, Li ions may not be transferred enough to the Cu foil because they couldn't pass through the PDMS polymer. As shown in Fig. 4 (b), 10 min is found to be the optimum time to prepare PDMS/GO composite thin film since the ion channels



**Fig. 3.** Electrochemical characteristics; (A) Coulombic efficiency of bare Cu (untreated one), PDMS coating, PDMS/GO coating electrode, (B) (a) voltage profiles of Bare Cu electrode, (b) voltage profiles of PDMS/GO film electrode.



**Fig. 4.** Schematic illustration of PDMS/GO based Li metal electrodes prepared with various etching time (a) PDMS/GO/Li metal electrode with insufficient ion channel (etching time < 10 min), (b) PDMS/GO/Li metal electrode with optimized ion channel (etching time ~10 min), (c) PDMS/GO/Li metal electrode with cracks (etching time > 10 min).

which Li ions can be sufficiently passed while the dendrite can be suppressed. In addition, Li is stably deposited due to the uniform Li ion flux. In contrast, when the etching time is longer than 10 min, the etch-

ing is excessive as shown in Fig. 4 (c), the PDMS/GO composite thin film is damaged and Li ion flux is concentrated into an excessively large ion channel, and thus dendrite is formed easily[16,18,19].

## 4. Conclusions

In this study, the PDMS/GO composite based on Li metal electrode was designed to improve the performance of Li metal battery by effectively suppressing the growth of Li dendrite. The composite of PDMS/GO film was prepared by mixing exfoliated GO nanosheets and PDMS. In succession, the PDMS/GO based Li metal electrodes were prepared by blade-coating on Cu foil with the solution of PDMS/GO composite. Since Li ions are hard to penetrate across the PDMS/GO thin film, the ion channels for allowing Li ions were formed by HF etching to facilitate the smooth movement of Li ions. Electrochemical test results show that columbic efficiency of PDMS/GO composite thin film electrode etched for 10 min was maintained at 87.4% on average up to 100<sup>th</sup> cycles compared to bare Cu electrode and PDMS thin film electrode. Furthermore, the voltage profiles of the 5, 10, 30, and 70<sup>th</sup> cycles exhibits the reduction of overpotential of electrodes.

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