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## COMMUNICATION

## Flexible Freestanding Sandwich-structured Sulfur Cathodes with Superior Performance for Lithium-sulfur Batteries

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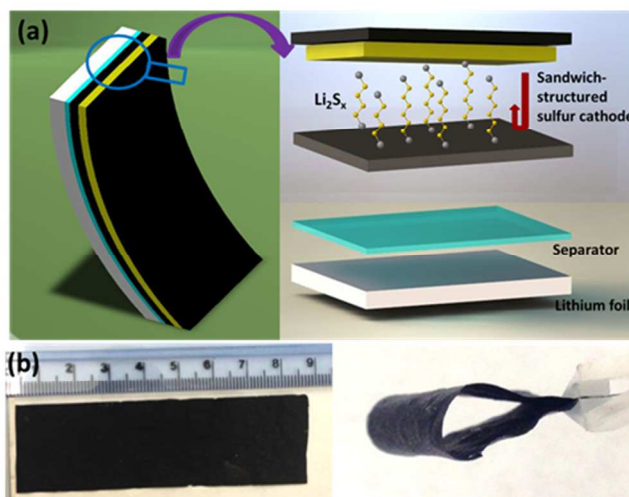
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Flexible freestanding sandwich-structured sulfur cathodes are developed for lithium-sulfur batteries, which exhibit a specific capacity around 1500 mAh/g with good cycling performance (1106 mAh/g after 100 cycles) and excellent rate capability. Moreover, high areal capacity of  $\sim 4 \text{ mAh/cm}^2$  is demonstrated based on this new cathode configuration with high sulfur loading ( $\sim 4 \text{ mg-S/cm}^2$ ).

Lithium-ion batteries (LIB) have achieved great success in the past three decades as electrochemical energy storage, especially for portable electronic devices. However, state-of-the-art LIBs still cannot meet crucial demands to reduce energy cost associated with using these batteries in emerging applications, such as electric vehicles and grid-level energy storage, due to their low energy density and high cost.<sup>1-6</sup> Recently, lithium-sulfur (Li-S) batteries have attracted increasing attention and are recognized as the next generation rechargeable battery because of their high theoretical specific capacity (1672 mAh/g) and energy density (2600 Wh/kg).<sup>7,8</sup> Additionally, low-cost, abundant, and eco-friendly nature of sulfur makes the Li-S batteries more attractive. Despite promising advantages, practical application of Li-S batteries is highly hindered by several issues. First, the electrical insulating nature of both sulfur and its discharged intermediates results in low utilization of the active materials and poor rate capability. Second, lithium polysulfides ( $\text{Li}_2\text{S}_x$ ,  $4 \leq x \leq 8$ ) generated as intermediate products during the electrochemical charge-discharge process are easily soluble into the organic electrolytes. This can lead to a loss of active material from the cathode as well as to a polysulfide shuttle phenomenon, resulting in severe capacity fading and low Coulombic efficiency.<sup>9-11</sup>

To address these aforementioned issues, many efforts have focused on developing new cathode materials to advance Li-S batteries, including porous carbon-sulfur composites cathode,<sup>11-18</sup> conducting polymer-coated sulfur cathode,<sup>19-21</sup> and new electrolyte system (new liquid electrolytes, additives, and solid electrolytes).<sup>22-25</sup> Cell configuration has recently been determined to be a crucial factor in improving Li-S batteries. Appropriate design of electrodes and/or cells can significantly improve the electrochemical performance of Li-S batteries.<sup>26-29</sup> For example, Manthiram and his co-workers<sup>28</sup> developed a novel Li-S cell configuration by inserting



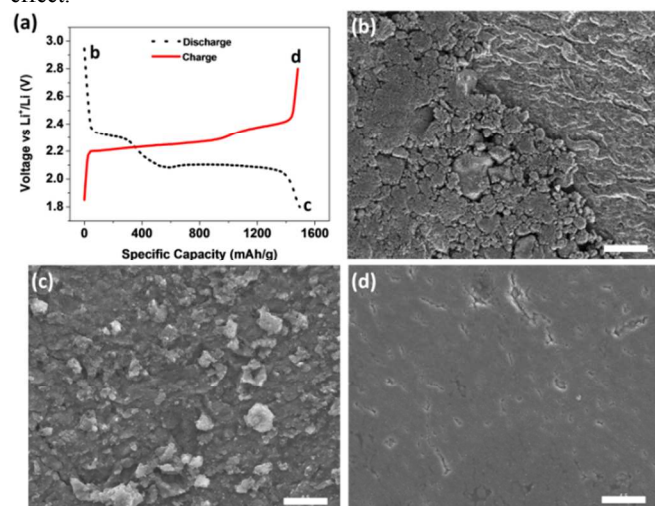
**Figure 1.** Schematic illustration of a Li-S cell with sandwich-structured cathode. (b) Photo images of sandwich-structured electrodes at tiled (left) and bended state (right).

a carbon interlayer between the cathode and separator. This highly conductive interlayer significantly decreased the resistance of the electrode and enhanced the utilization of sulfur by trapping the soluble lithium polysulfides within the carbon interlayer. Zhang *et al.*<sup>27</sup> constructed a dual layer sulfur cathode that consisted of a highly porous carbon cloth as the cathode current collector and a porous sulfur paper as the source of active material. An extremely high sulfur loading up to  $10 \text{ mg/cm}^2$  was achieved. Although the sulfur utilization is low, this cathode design is still attractive for high energy density cells that necessitate high sulfur loading.

Herein, we report a novel sulfur cathode composed of a layer of sulfur active materials sandwiched by two flexible porous carbon membranes, as shown in Figure 1a. This novel sandwich-structured cathode design exhibits many advantages. First, the porous carbon membrane can effectively capture soluble lithium polysulfides into its highly porous structure during charge-discharge process, resulting in much improved cycling stability and Coulombic efficiency. Second, the charge transfer resistance is significantly decreased compared to conventional sulfur cathodes not only due to reduced

contact resistance between sulfur cathodes and current collector but also due to intimate contact between sulfur and two layers of highly conductive carbon. This attribute makes it potentially viable to fabricate high-performance, high-sulfur-loading cathodes with significantly improved sulfur utilization and rate capability. Third, this carbon-supported sandwich-structured cathode is flexible, which can potentially extend Li-S batteries to other applications such as wearable devices, and implantable biomedical devices.

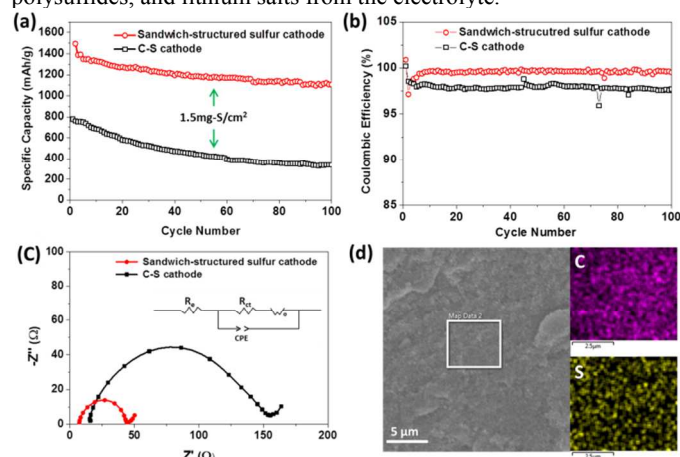
The flexible freestanding carbon membranes were fabricated by milling–assistant dispersion of porous carbon (Ketjen black carbon) in PTFE latex (See detail in Supporting Information), followed by roll press and drying. In this manner, a large piece of flexible carbon membrane (e.g., 2 cm × 8 cm) was obtained (Figure 1b). Ball-milled sulfur/carbon black (7:3, weight ratio) was placed in the middle of two pieces of porous carbon membranes, giving a flexible sandwich-structured sulfur cathode. The Ketjen black carbon has a large Brunauer–Emmett–Teller (BET) surface area of 1326.84 m<sup>2</sup>/g and high Barrett–Joyner–Halenda pore volume of 2.43 cm<sup>3</sup>/g (Figure S1). The use of the high-surface-area, high-pore-volume carbon can promote high capacity physical adsorption of soluble lithium polysulfides, mitigating the loss of active materials and “shuttle” effect.<sup>28</sup>



**Figure 2.** (a) Typical initial charge/discharge voltage profiles for the sandwich-structured sulfur cathode at C/5 (1C=1672 mAh/g) between 1.8 V and 2.8 V vs. Li/Li<sup>+</sup>. As shown, two discharge plateaus at ~2.3 V and ~2.0 V are observed. This discharge profile coincides with observation in the literature using conventional sulfur cathodes, in which the higher voltage plateau corresponds to the reduction of sulfur to higher-order lithium polysulfides (Li<sub>2</sub>S<sub>n</sub>, 6 ≤ n ≤ 8) and the lower plateau is ascribed to the reduction of higher-order lithium polysulfides to lower-order lithium polysulfides (Li<sub>2</sub>S<sub>n</sub>, 2 ≤ n < 6).<sup>9, 30</sup> The charge curve also shows two voltage plateaus around 2.3 V and 2.4 V vs. Li/Li<sup>+</sup>, which corresponds to the oxidation of the lower-order polysulfides and oxidation of the high-order polysulfides, respectively. Cyclic voltammetry (CV) was further performed to verify this redox reaction. Two cathodic peaks (2.0 V and 2.3 V) as well as the corresponding anodic peaks (2.3 and 2.4 V) (Figure S2) resemble the discharge/charge plateaus in the above-mentioned voltage profiles (Figure 2a). The nearly overlapped redox peaks in the following four-cycle scanning represent a good cycling stability of this sandwich-structured cathode.

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The morphology change of active sulfur materials within the sandwich-structured cathode in the first cycle was investigated by scanning electron microscopy (SEM). Figure 2b, 2c, and 2d illustrate the morphology of sulfur on fresh electrode, its discharged products on electrodes at 1.8 V vs Li/Li<sup>+</sup>, and its charged product on electrodes at 2.8 V vs Li/Li<sup>+</sup>, respectively. Sub-millimeter and micro-sized of sulfur particles is clearly observed on the carbon membrane in the fresh electrode. Upon deep discharge to 1.8V, a coarse surface containing many irregular particles are observed, which are considered to be Li<sub>2</sub>S<sub>2</sub> and Li<sub>2</sub>S on carbon black particles.<sup>31</sup> Noticeably, the electrode after the 1<sup>st</sup> charge shows a relatively smooth surface coated with melt-like materials. These materials are most likely formed by sulfur, higher ordered lithium polysulfides, and lithium salts from the electrolyte.<sup>26</sup>



**Figure 3.** (a) Cycling stability and (b) Coulombic efficiency of sandwich-structured and conventional C-S electrodes at C/5 (1C=1672 mAh/g) between 1.8-2.8V vs Li/Li<sup>+</sup>. The capacity values were calculated based on the mass of sulfur; (c) Nyquist plots of the Li-S cells measured before cycling; (d) SEM image and corresponding carbon (purple) and sulfur (yellow) EDS mapping of upper carbon membrane of sandwich-structured sulfur cathode after 20 cycles.

The battery performance was studied in two-electrode CR2016 coin cells using a galvanostatic charge-discharge process with 1M lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) and 0.2M lithium nitrate (LiNO<sub>3</sub>) in 1,3-dioxolane/dimethoxy ethane (DOL/DME, 1:1 volume ratio) as the electrolyte. All capacity values were calculated based on the mass of sulfur. The sulfur cathodes were fabricated with the same ratio of sulfur to carbon via conventional coating method and were used as control samples, noted as C-S for simplicity. Figure 3a and b shows the cycling stability and Coulombic efficiency of sandwich-structured sulfur cathode and the C-S cathode, respectively. The sandwich-structured sulfur cathode delivered an initial capacity of 1494.7 mAh/g at C/5, corresponding to ~90% of theoretical sulfur utilization. This is almost two times higher than that of the conventional C-S cathode which only showed an initial capacity of 777.2 mAh/g. Moreover, the sandwich-structured cathode exhibited exceptional cycling performance. After 100 extended cycles, the capacity was still above 1100 mAh/g, which is strikingly better than the conventional C-S cathode. The Coulombic efficiency of the sandwich-structured sulfur cathode in the first cycle was calculated to be ~100 % and 99.5 % after 100 cycles. The conventional C-S cathode still shows a relatively lower Coulombic efficiency (97-98%) compared with sandwich-structured sulfur cathode even when LiNO<sub>3</sub> was adopted to protect the lithium anode to improve Coulombic efficiency.<sup>32</sup> The enhanced electrochemical performance of sandwich-structured electrodes can be ascribed to both retardation of polysulfide migration by physical adsorption in two carbon layers and enhanced

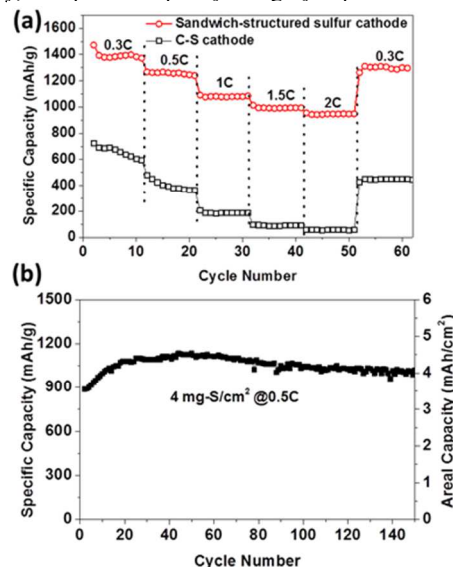
electron transfer across the electrode. Porous carbon membranes enable the electrolyte to permeate the carbon layer, allowing the electrochemical reactions to proceed in the cathode. More importantly, the tortuous pores in the membrane can localize the polysulfide species diffusing from the cathode. In addition, both the top and bottom carbon membranes act as pseudo current collectors, which can facilitate electron transfer from both sides of the sandwich-structured cathode. This parallel electron conducting circuit along carbon membranes can effectively reduce the area current density of active materials.

To prove these hypotheses, electrochemical impedance spectroscopy (EIS) was conducted on the fresh cells based on sandwich-structured cathodes and C-S cathodes, and the results are illustrated in a modified Randle circuit, as shown in Figure 3c. The Nyquist plots of these cells are composed of a depressed semicircle from the high frequency region to mid-frequency region and a sloping line in the low-frequency region.  $R_0$  in the equivalent circuit represents the ohmic resistance contribution of two-electrode cell resulting from electrolyte, current collectors and cell contact.<sup>33</sup> One can observe from the EIS that  $R_0$  is decreased in the sandwich-structured cathode than in conventional C-S cathodes. This decreased ohmic resistance is most possibly due to the sandwich-like structure, where the flexible carbon membrane can provide the large effective contact area and conformal contact between the highly porous carbon pseudo current collector and the active material particles as well as the cell shells.<sup>28, 34-36</sup> The diameter of the semicircle in Figure 3c represents the charge transfer resistance ( $R_{ct}$ ), which is mainly generated at the interface between the electrode and the electrolyte. The reduced  $R_{ct}$  for sandwich-structured sulfur cathode confirmed this structure can greatly facilitate the charge transfer due to increased contact surface area between carbon membrane, carbon black, and sulfur particles.<sup>35, 37, 38</sup> Moreover, flexible free-standing sandwich-structured network a conformal electric contact of sulfur and its discharged product with conducting matrix.<sup>37</sup> SEM and corresponding element mapping were further tested on the carbon membranes disassembled from sandwich-structured sulfur cathode after 20 cycles. As shown in Figure 3d, the sulfur element was found all over the carbon paper and distributed homogeneously, which indicates that the soluble lithium polysulfides were captured and remained by the highly porous carbon paper. This enables the re-utilization of the active material rather than diffusion to the anode sides.

The rate capability of sandwich-structured sulfur cathode was further evaluated by increasing the C-rate from 0.3C to 2C, as shown in Figure 4a. As the current density increases, a specific capacity of 1380 mAh/g, 1260 mAh/g, 1070 mAh/g, 990 mAh/g was obtained at 0.3C, 0.5C, 1C, and 1.5C, respectively. Even at 2C, it still can deliver a high reversible capacity of  $\sim 950$  mAh/g. This rate performance is overwhelmingly better than that of the C-S cathode. When the rate restored to 0.3C after 50 cycles of rate testing, the specific capacity in the sandwich-like sulfur cathode returned to 1316 mAh/g, close to 1380 mAh/g from the initial 0.3C trial. This indicates that the cathode charge/discharge was reversible even at high current density. The rate performance is closely related to the electrical conductivity of the carbon matrix, which provide electron transport pathway from the current collector to the sulfur atoms during the electrochemical redox reaction. The facilitated electron transfer behavior – manifest as improvements in the rate capability – can likely be attributed to the electrical conductivity gained from the sandwich-like structure, as demonstrated by EIS analysis mentioned above.

For practical applications, areal capacity ( $\text{mAh}/\text{cm}^2$ ) is also quite important, which is commonly calculated as specific capacity ( $\text{mAh}/\text{g}$ ) of sulfur  $\times$  sulfur mass loading ( $\text{g sulfur}/\text{cm}^2$  of electrode

face area). Based on this, it is clear that achieving a high areal capacity necessitates high sulfur mass loading in the electrodes. Commonly, the specific capacity is highly dependent on the sulfur



**Figure 4.** (a) The rate capability of sandwich-structured sulfur and C-S cathodes; (b) the specific capacity and areal capacity of sandwich-structured sulfur cathode as function of cycling with a sulfur loading of 4.0  $\text{mg}/\text{cm}^2$ .

loading, and it will drop with increasing sulfur loading because of the higher polarization of the electrodes due to ohmic resistance from both electrodes and electrolyte.<sup>14</sup> Most of the high-specific-capacity cathodes for Li-S batteries reported in the literature were obtained at a low sulfur loading (below 2  $\text{mg S}/\text{cm}^2$ ).<sup>8</sup> This loading is insufficient to achieve a sulfur cathode with areal energy density greater than that of present Li-ion cathodes.<sup>8, 15</sup> Benefiting from the unique design, the sandwich-structured sulfur cathode with sulfur loading of 4.0  $\text{mg-S}/\text{cm}^2$  was able to be fabricated. Figure 4b shows the specific capacity and corresponding areal capacity as a function of cycle number tested at a rate of 0.5C for a high-sulfur-loading sandwich-structured cathode. The specific capacity of sandwich-structured cathode progressively increased in the first several cycle's activation, and then stabilized around 1000  $\text{mAh}/\text{g}$  within the extended 150 cycles. More importantly, the calculated areal capacity is enhanced to a level as high as  $\sim 4$   $\text{mAh}/\text{cm}^2$ , making this new cathode promising in the real application.

## Conclusions

In summary, we successfully developed a novel sandwich-structured sulfur cathode composed of active sulfur materials sandwiched by two porous carbon layers for lithium-sulfur batteries. This design has many advantages over conventional C-S cathodes. First, the diffusion of soluble lithium polysulfides is effectively suppressed by two layers of porous carbon "barrier". Second, charge transfer is significantly improved by this novel design. Third, the flexible sulfur cathode can potentially extend the application of lithium-sulfur batteries into flexible devices. A high capacity and excellent cyclability were successfully demonstrated by using this sandwich-structured design. In addition, a high areal capacity of  $\sim 4$   $\text{mAh}/\text{cm}^2$  up to 150 cycles were achieved at electrode level. Taking the advantages of facile fabrication process and excellent performance, the novel configuration proposed would be of interest for scalable production and fabrication for high-energy Li-S batteries.

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## Notes and references

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Flexible freestanding sandwich-structured sulfur cathodes are developed for lithium-sulfur batteries, which exhibit excellent cycling stability and rate capability. Moreover, high areal capacity of  $\sim 4$  mAh/cm<sup>2</sup> is demonstrated based on this new cathode configuration with high sulfur loading ( $\sim 4$  mg-S/cm<sup>2</sup>).

