



## Short communication

## A lithium sulfur battery with high power density



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## HIGHLIGHTS

- Sulfur content in the positive composite electrodes was 50 wt%.
- There was a correlation between surface area of the carbon and utilization of sulfur.
- Reversible capacity of over 1600 mAh g<sup>-1</sup> was obtained after 100 cycles at 1C at 25 °C.
- The positive composite electrode exhibits a power density of 11000 W kg<sup>-1</sup> at 25 °C.

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## ABSTRACT

The performance of all-solid-state lithium sulfur (Li/S) batteries is investigated for some types of conductive materials. The surface area of the conductive material is found to have a greater influence on the battery performance compared with its electronic conductivity. All-solid-state Li/S cells prepared using a positive activated-carbon-based composite electrode exhibit an excellent cycling performance with a reversible capacity of over 1600 mAh g<sup>-1</sup> after 100 cycles at 1.3 mA cm<sup>-2</sup> (1C) at 25 °C. Furthermore, the positive composite electrode exhibits a high power density of over 11000 W kg<sup>-1</sup> at 50% state of charge.

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## 1. Introduction

High-capacity rechargeable batteries are in demand for future portable electronic devices, electric vehicles, and large-scale electricity storage. Lithium sulfur (Li/S) battery is regarded as one of the most promising systems for next-generation batteries because of its high theoretical capacity [1]. In addition, sulfur is an abundant and cheap material. However, the utilization of a sulfur electrode is not sufficient to achieve the theoretical capacity because sulfur's ionic and electronic conductivities are low. Furthermore, the cycling performance rapidly decreases because polysulfides formed during battery operation are highly soluble in the liquid electrolytes [2].

Many researchers have focused on confining the sulfur materials into porous nanostructures to prevent the dissolution of lithium polysulfides during charge–discharge reactions [3]. A few

researchers have investigated ionic liquids that practically do not dissolve lithium polysulfides [4]. However, none of them has been able to completely inhibit the dissolution of polysulfides.

The use of an inorganic solid electrolyte (SE) (instead of organic liquid or ionic liquid electrolyte) is basically an effective approach to inhibit the dissolution of polysulfides. There are various inorganic SEs such as Li<sub>2</sub>S–SiS<sub>2</sub>, Li<sub>2</sub>S–P<sub>2</sub>S<sub>5</sub>, and Li<sub>4</sub>GeS<sub>4</sub>–Li<sub>3</sub>PS<sub>4</sub> with a high lithium-ion conductivity of over 10<sup>-4</sup> S cm<sup>-2</sup> at 25 °C [5–8]. All-solid-state Li/S batteries containing these SEs exhibit an excellent cycling performance because polysulfide dissolution is completely inhibited [9]. However, in many reports, the sulfur contents in the positive electrodes are still insufficient or the current density is too small for practical uses [10]. Therefore, the energy densities of these Li/S batteries are relatively low under practical current–density conditions.

In a recent report, we discussed the relationship between the P/S (number of P atoms/number of S atoms) ratio in SEs and the capacity and potential of all-solid-state Li/S batteries to improve the battery performance focusing on the reactivity of sulfur. The possibility of obtaining a high-energy-density battery was shown [11].

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In this report, we investigated the relationship between the properties of the conductive material and battery performance to further improve the efficiency of the devices. We found that the surface area of the conductive material has a greater influence on the battery performance compared with its electronic conductivity.

## 2. Experimental

### 2.1. Solid electrolyte (SE; $\text{Li}_{1.5}\text{PS}_{3.3}$ )

Reagent-grade  $\text{Li}_2\text{S}$  (Furuuchi Chem. 99.9%) and  $\text{P}_2\text{S}_5$  (Aldrich, 99%) were used as the starting materials [8]. The SE was prepared by the mechanochemical method using a planetary ball mill apparatus (Fritsch Pulverisette 7). The mechanochemical treatment was performed on a mixture of  $\text{Li}_2\text{S}$  and  $\text{P}_2\text{S}_5$  (at a molar ratio of 60:40) using a  $\text{ZrO}_2$  pot and balls.

### 2.2. Positive composite electrodes

Several positive composite electrodes containing elemental sulfur as the active material were prepared by ball milling using a planetary ball mill apparatus under an Ar atmosphere [9]. Reagent-grade sulfur (Aldrich, 99.5%), an SE ( $\text{Li}_{1.5}\text{PS}_{3.3}$ ), and the conductive materials acetylene black (AB, Denki Kagaku Kogyo), Ketjenblack (KB, Lion Co.), and activated carbon (AC, Kansai Coke and Chemicals Co., Ltd.) were used as the starting materials. The conductive materials had Brunauer–Emmett–Teller surface areas of 70, 1200, and  $3000 \text{ m}^2 \text{ g}^{-1}$ , respectively. The weight ratio of sulfur, SE, and conductive material was fixed to 50:40:10. Here AB and KB are widely known as a conductive carbon, and therefore commonly, their electronic conductivities are extremely higher than activated carbon [12,13].

### 2.3. Electrochemical measurement

The electrochemical properties of the positive composite electrodes were investigated in an all-solid-state cell [14]. Two-electrode cells were assembled in a glove box filled with dry Ar using several composite electrodes as the positive electrode, Li–In alloys as the negative electrode, and  $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$  [7] as the solid electrolyte. The positive electrode powder (7.5 or 1.2 mg) and solid electrolyte powder (70 mg) were placed in a polycarbonate tube (diameter 10 mm) and pressed at 200 MPa to form a two-layered pellet. The negative electrode, formed by piling a Li foil on an In foil, was placed on the surface of the two-layered pellet, where the piled foils were in contact with the solid electrolyte. In this process, the Li and In foils formed a Li–In alloy layer. The thus obtained three-layered pellet was then sandwiched between two stainless steel rods, which served as current collectors. The Li/In molar ratio in the negative electrode was 0.79. It has been reported that if the molar ratio is kept below one, the potential of Li–In alloys remains constant at approximately 0.6 V (vs. Li) [15]. In our report, 0.6 was adopted as the potential difference between Li–In and Li.

Electrochemical tests were conducted at several constant current densities from  $0.64$  to  $39 \text{ mA cm}^{-2}$  at  $25^\circ\text{C}$  in an Ar atmosphere using a charge–discharge measuring device, ACD-01 (Asuka electronic Co. Ltd.).

The measurement of power density was conducted with reference to the method of JEVS D713. The measurements were performed at several states of charge (SOC) values at  $\pm 5.0$ , 10, 15, 20, 25, and 30 mA for 10 s.

## 3. Results and discussion

The measurements shown in Figs. 1–4 are performed on a cell containing a 7.5 mg positive composite electrode. The cut-off

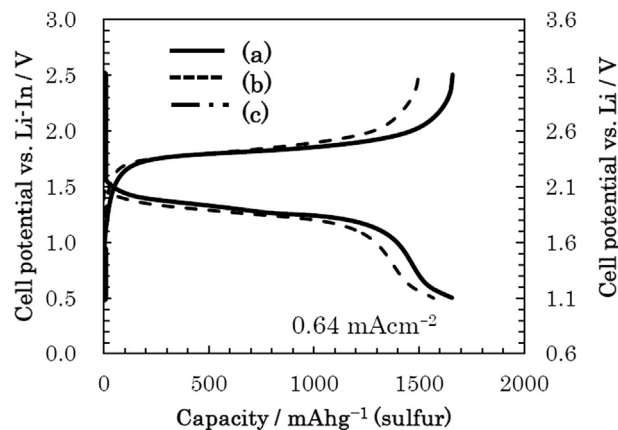


Fig. 1. Charge–discharge curves of all-solid-state lithium sulfur (Li/S) cells containing positive composite electrodes based on (a) AC, (b) KB, and (c) AB at  $0.64 \text{ mA cm}^{-2}$  at  $25^\circ\text{C}$ .

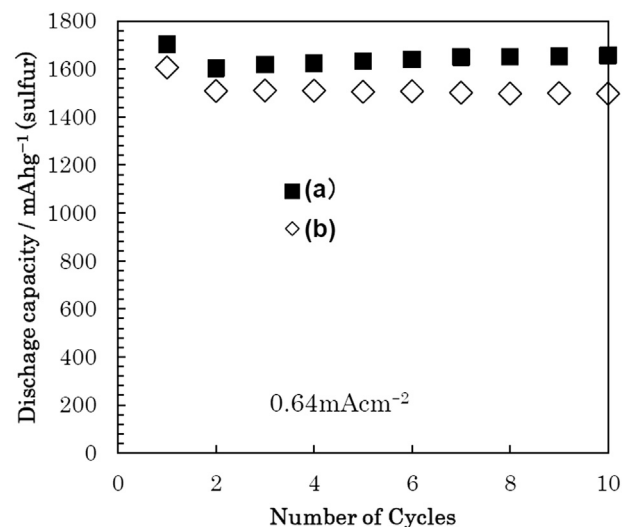


Fig. 2. Cycling performance of all-solid-state lithium sulfur (Li/S) cells containing (a) AC- and (b) KB-based positive composite electrodes at  $0.64 \text{ mA cm}^{-2}$  at  $25^\circ\text{C}$ . The weight of the positive composite electrodes is 7.5 mg. The cut-off voltage is kept between 0.5 and 2.5 V (vs. Li–In).

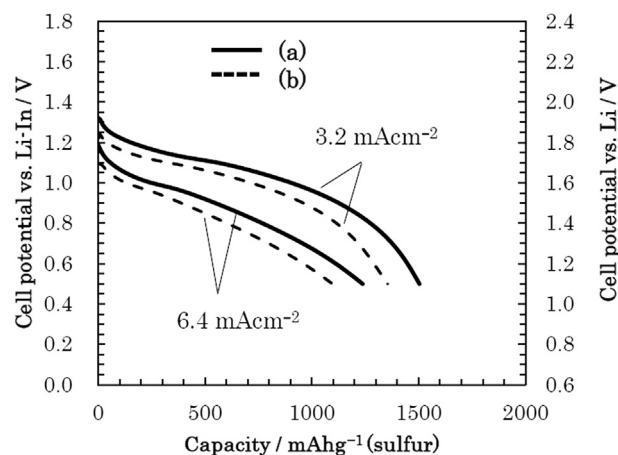


Fig. 3. Discharge curves of all-solid-state lithium sulfur (Li/S) cells containing positive composite electrodes based on (a) AC and (b) KB at current densities  $3.2$  and  $6.4 \text{ mA cm}^{-2}$  at  $25^\circ\text{C}$ . The cut-off voltage is 0.5 V (vs. Li–In).

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