Electronic Supplementary Material (ESI) for Nanoscale.

Electronic Supplementary Information

One-Pot Pyrolysis of Lithium Sulfate and Graphene Nanoplatelet Aggregates: In situ Formed Li_2S/Graphene Composite for Lithium–Sulfur Batteries

*Zhe Li, Shiguo Zhang, Ce Zhang, Kazuhide Ueno, Tomohiro Yasuda, Ryoichi Tatara, Kaoru Dokko, and Masayoshi Watanabe**

Department of Chemistry and Biotechnology, Yokohama National University, 79-5 Tokiwadai, Hodogaya-ku, Yokohama 240-8501, Japan
E-mail: mwatanab@ynu.ac.jp

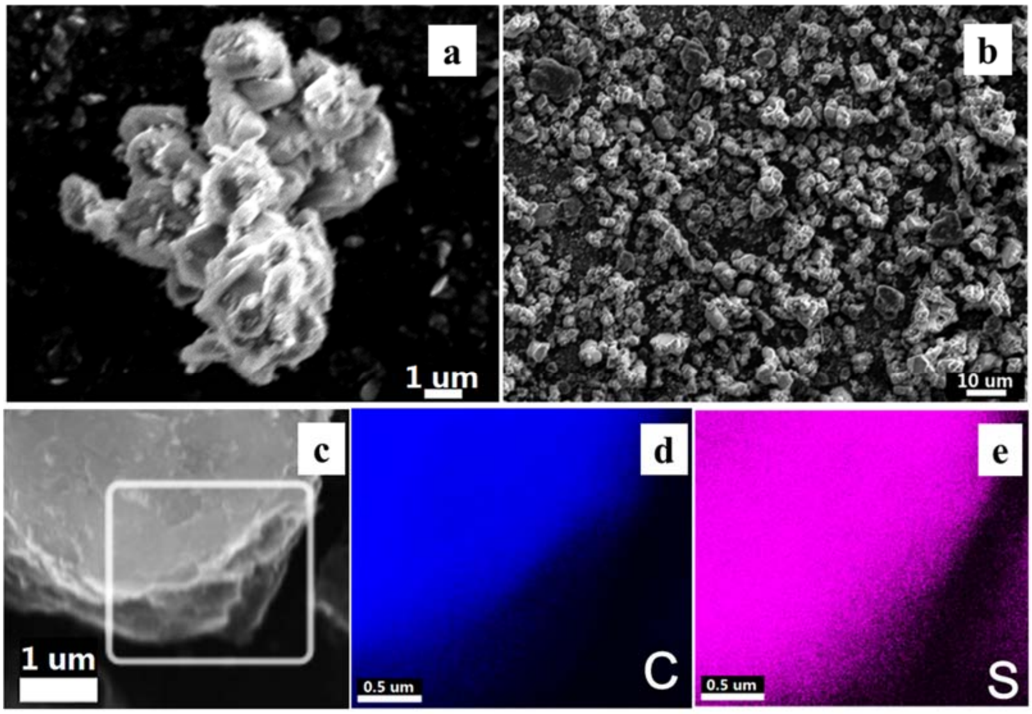

Figure S1. FESEM images of (a) GNAs and (b) LS-GNAs C. (c) Enlarged FESEM image of LS-GNAs C particle and the corresponding energy-dispersive X-ray spectroscopy (EDS) elemental mappings of (d) carbon and (e) sulfur. GNAs consist of agglomerations of thick plates. The resultant LS-GNAs C particles have irregular shapes and particle sizes of 1.0–10.0 µm. Deposition of Li_2SO_4 results in a uniform distribution of Li_2SO_4 on the surface of the GNAs, as indicated by the homogeneous EDS signals of carbon and sulfur.

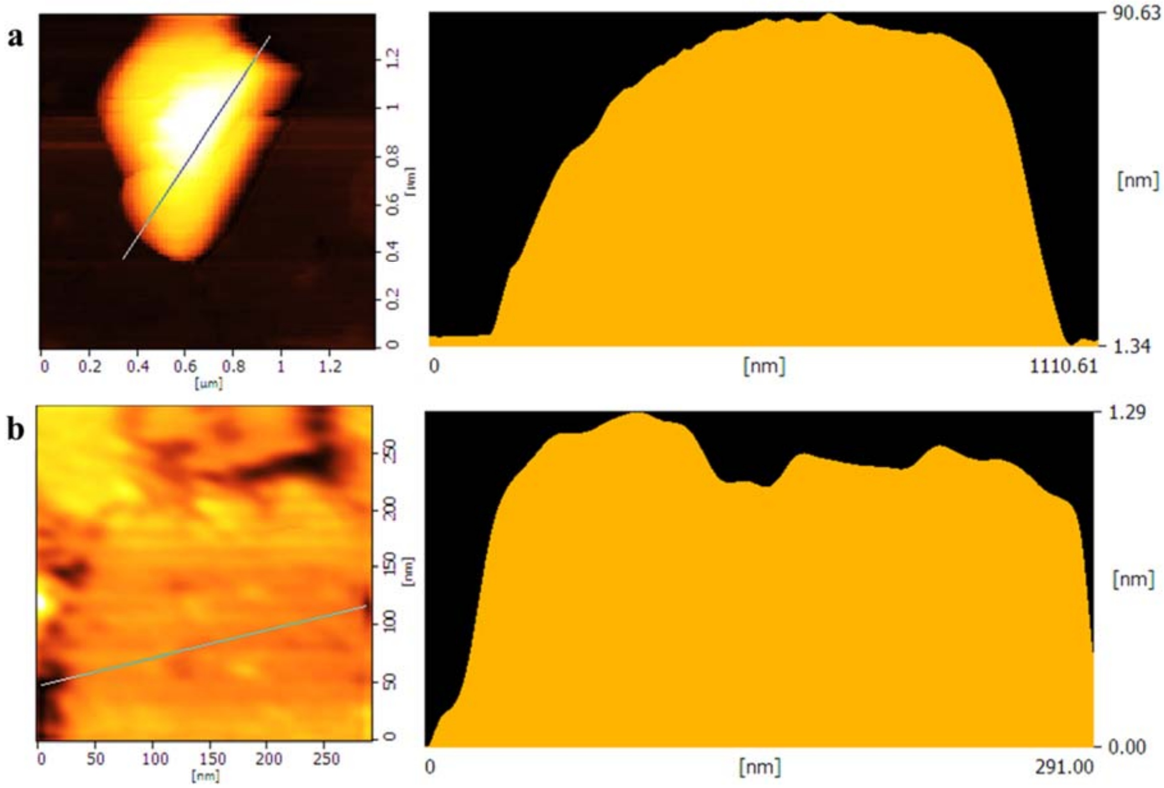

Figure S2. Tapping-mode atomic force microscopy (AFM) images with the corresponding height profiles: (a) GNAs and (b) LS-GNAs H-W. Figure S2a reveals that the thickness of the GNAs is around 89 nm, which is due to intrinsic agglomeration. Figure S2b shows that the thicknesses of LS-GNAs H-W is between 1.0 and 1.3 nm, suggesting that the graphene sheets in this sample are thinner than those in the GNAs.

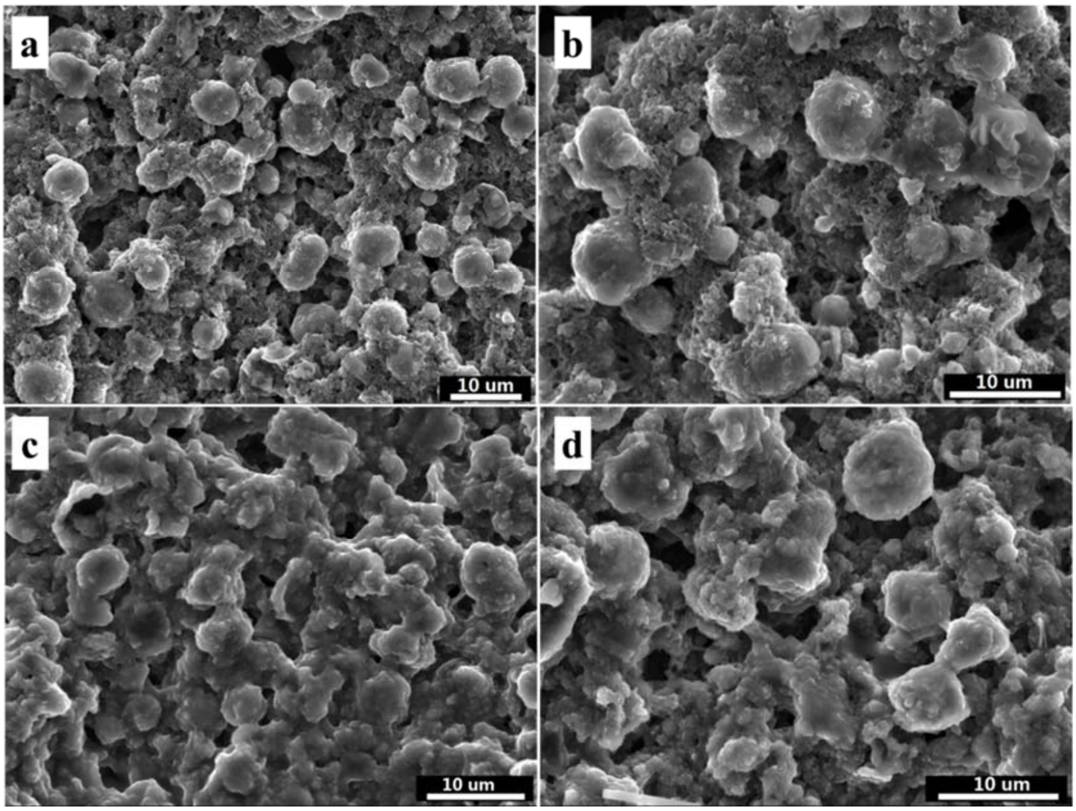

Figure S3. FESEM images of (a, b) a fresh LS-GNAs H electrode before cycling and (c, d) the electrode after 40 cycles. For the electrode after 40 cycles, no large-sized Li_2S agglomerations were observed, and abundant porous spaces were still preserved, which could enable the electrolyte channel for electrolyte immersion to be unblocked.

Table S1. Content of different components in Li_2S/graphene composite-based electrode.

Composite	Electrode			
Li_2S Content (wt%)	Li_2S (wt%)	Carbon (wt%)		PVP (wt%)
		Graphene	Carbon Black	
69.5	60.0	26.3	3.7	10.0
78.3	60.0	16.6	13.4	10.0
82.4	60.0	12.8	17.2	10.0
86.4	60.0	9.4	20.6	10.0

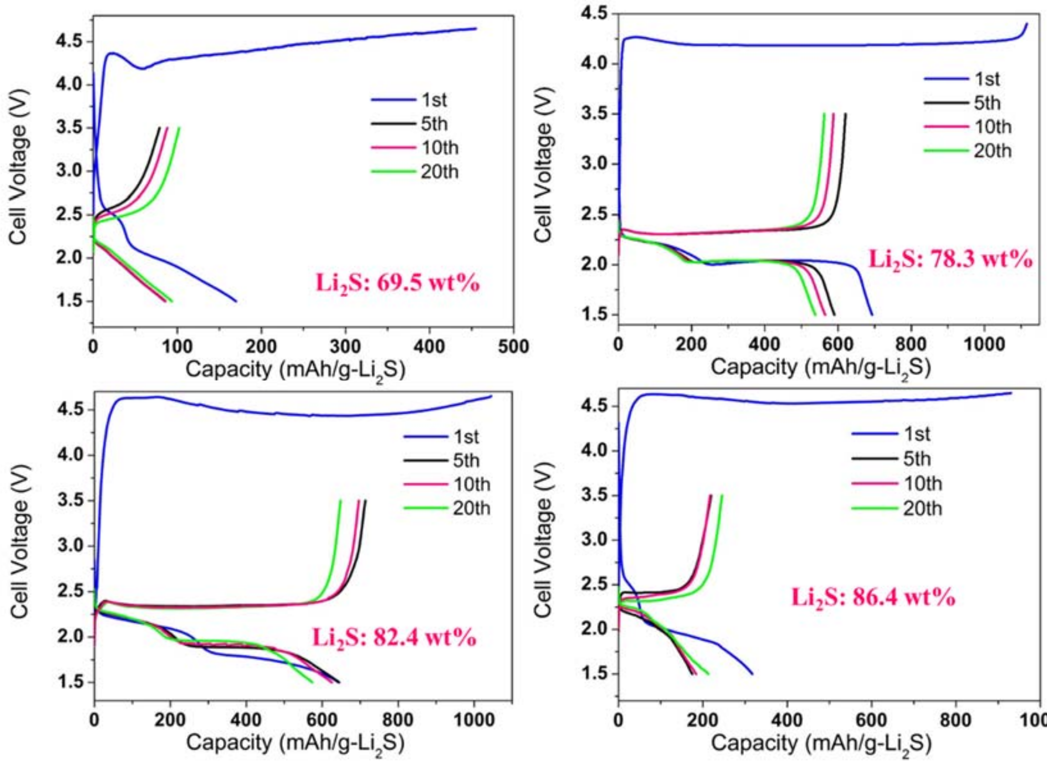

Figure S4. Galvanostatic charge–discharge profiles of different Li_2S/graphene composites in the 1st, 5th, 10th, and 20th cycles at a current rate of 1/12 C (1 C = 1166 mA/g). Graphene can enhance electrical and mechanical connectivity between the individual Li_2S, while carbon black would improve the conductivity of the whole electrode. When the content of graphene is low (e.g., the case of 86.4 wt% Li_2S content in composite), some Li_2S in the composite may agglomerate together, which require more energy to overcome the barrier of lithium extraction and lead to a poor utilization of Li_2S. Meanwhile, Li_2S/graphene composite with Li_2S content of 69.5 wt%, demonstrated a very sluggish initial activation process of Li_2S with a lower charge capacity of 455 mAh/g, due to the lack of carbon black (only 3.7 wt% of carbon black in the electrode).

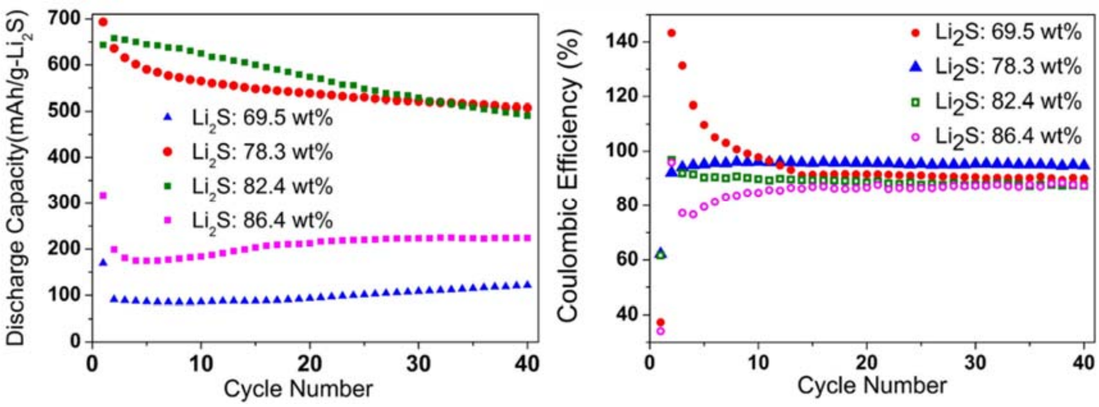

Figure S5. Cycling performance and Coulombic efficiency of different Li_2S/graphene composites over 40 cycles at a current rate of 1/12 C.

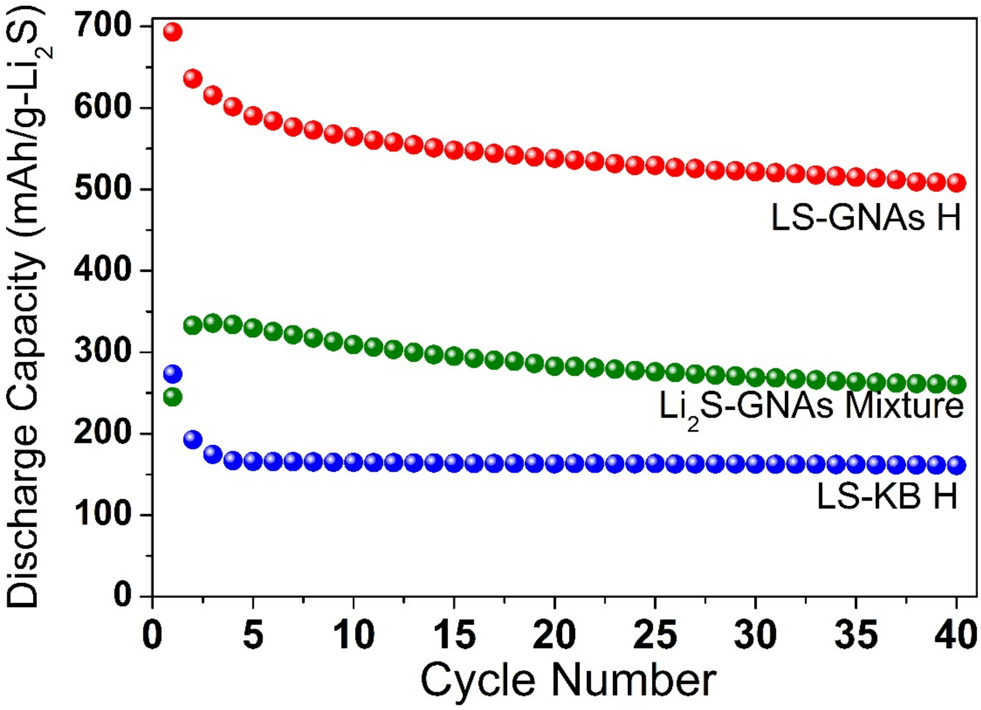

Figure S6. Cycling performance of LS-GNAs H, Li_2S–GNAs mixture, and LS-KB H over 40 cycles at a current rate of 1/12 C.