



Optimizing reduced graphene oxide/MXene composites as sulfur hosts for lithium-sulfur batteries: A systematic investigation

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ABSTRACT

Large volume changes, the insulating nature of sulfur, and the lithium polysulfides (LiPSs) shuttle effect significantly hinder the practical application of lithium-sulfur (Li-S) batteries. In this study, reduced graphene oxide/MXene (rGO/MXene) composites were investigated as potential sulfur host materials. Since the rGO-to-MXene ratio influences electrical conductivity, LiPSs confinement, and structural stability, MXene contents of 10 wt%, 20 wt%, and 30 wt% were systematically evaluated. The (rGO/MXene)₁₀/S₆₀ electrode exhibited the most promising performance, delivering an initial discharge capacity of 981 mAh/g and retaining 606 mAh/g after 100 cycles with Coulombic efficiency (CE) above 97 %. These results demonstrate that a 9:1 rGO-to-MXene ratio optimally enhances conductivity, structural stability, and LiPSs anchoring, making it a promising sulfur host for Li-S batteries.

1. Introduction

Lithium-sulfur (Li-S) batteries have drawn significant interest due to their high theoretical energy density (2600 Wh/kg) and sulfur's natural abundance. However, challenges such as sulfur's insulating nature, lithium polysulfides (LiPSs) shuttling, and volume expansion hinder practical implementation [1]. To overcome these limitations, conductive carbon materials and polar hosts have been explored to enhance sulfur utilization and cycling stability [2].

Porous carbon, carbon nanotubes (CNTs), and graphene derivatives improve conductivity and volume changes but fail to effectively immobilize LiPSs due to their nonpolar nature, leading to capacity fade [3]. In contrast, polar materials (metal oxides, metal sulfides, and metal nitrides) exhibit strong LiPSs interactions, serving as effective immobilizers and redox catalysts [4].

Reduced graphene oxide (rGO) and Ti₃C₂T_x MXene offer complementary advantages: rGO enhances conductivity and structural flexibility [5], while MXene improves LiPSs adsorption and redox kinetics [6]. MXene-based hosts stabilize sulfur cathodes, while rGO prevents restacking of MXene sheets, improving electrochemical performance.

This study investigates the rGO/MXene composites as a potential host material for Li-S batteries leveraging their synergistic properties to enhance electrochemical performance and advance the development of high-performance sulfur cathodes. Since the MXene to rGO ratio affects the key factors such as conductivity, LiPSs confinement, and overall electrochemical stability, this work systematically explores different MXene contents (10 wt%, 20 wt%, and 30 wt%) to determine the optimal balance for improved battery performance.

2. Experimental

A graphene oxide (GO) solution was mixed with MXene suspensions to achieve rGO:MXene ratios of 9:1, 8:2, and 7:3 designated as rGO/MXene₁₀, rGO/MXene₂₀ and rGO/MXene₃₀, respectively. Reduction and nitrogen doping were performed via a one-step hydrothermal method using ethylenediamine (EDA) (100 µL per 100 mg GO) as both a nitrogen source and reducing agent. The mixture was transferred to a Teflon-lined autoclave and heated at 120° C for 4 h. The resulting rGO/MXene hydrogel was freeze-dried to form an aerogel, while a pure rGO aerogel was prepared under the same conditions without MXene

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(Fig. S.1). The final sulfur composite electrodes were prepared by incorporating 60 wt% sulfur into each rGO/MXene aerogel via melt-diffusion, denoted as rGO_S60, (rGO/MXene10)_S60, (rGO/MXene20)_S60, and (rGO/MXene30)_S60, respectively. Detailed experimental procedures and characterization methods are provided in the supplementary information.

3. Results and discussion

The MAX phase exhibits a layered structure with smooth surfaces (Fig. 1a), while etched MXene adopts an accordion-like morphology, confirming the removal of aluminum (Al) (Fig. 1b) [7]. When filtered onto a PVDF membrane, MXene nanosheets align into stacked layers (Fig. 1c), with wrinkles and wavy structure indicating suitable flexibility for electrode application. rGO sheets display a wrinkled, porous surface morphology (Fig. 1d), which can enhance sulfur confinement. The rGO/MXene10 composite forms a porous, conductive network (Fig. 1e), favorable for sulfur immobilization and electrolyte access. SEM imaging

of (rGO/MXene10)_S60 (Fig. 1f) reveals a porous structure with a uniform sulfur coating. TEM images show fragmented, glass-like MXene flakes (Fig. 1g) and ultrathin, wrinkled rGO sheets (Fig. 1h). Both features are visible in Fig. 1i, where the glass-like MXene and tissue-like rGO structures clearly co-exist.

The XRD pattern of the MAX phase (Fig. 2a) shows characteristic peaks at 9.5° , 19.7° , 34.0° , 38.9° , and 41.7° , corresponding to the (002), (004), (101), (104), and (105) planes [7]. After etching, the (002) peak shifts to 6.3° , increasing the d-spacing from 0.93 nm to 1.4 nm, confirming Li^+ intercalation and surface functionalization [8,9]. The weakened (104) peak at 38.8° indicates partial Al removal. GO exhibits a (001) peak at 11.6° , indicating expanded interlayer spacing due to oxygen functional groups [10]. Upon reduction by EDA, rGO exhibits a broad peak in-between 19° - 29° , reflecting oxygen removal and partial graphitic structure restoration [9]. A weak 11.5° peak in rGO suggests incomplete reduction, leaving residual oxygen groups. The XRD pattern of rGO/MXene10 closely resembles rGO, indicating well-dispersed MXene, reduced restacking, and enhanced exfoliation.

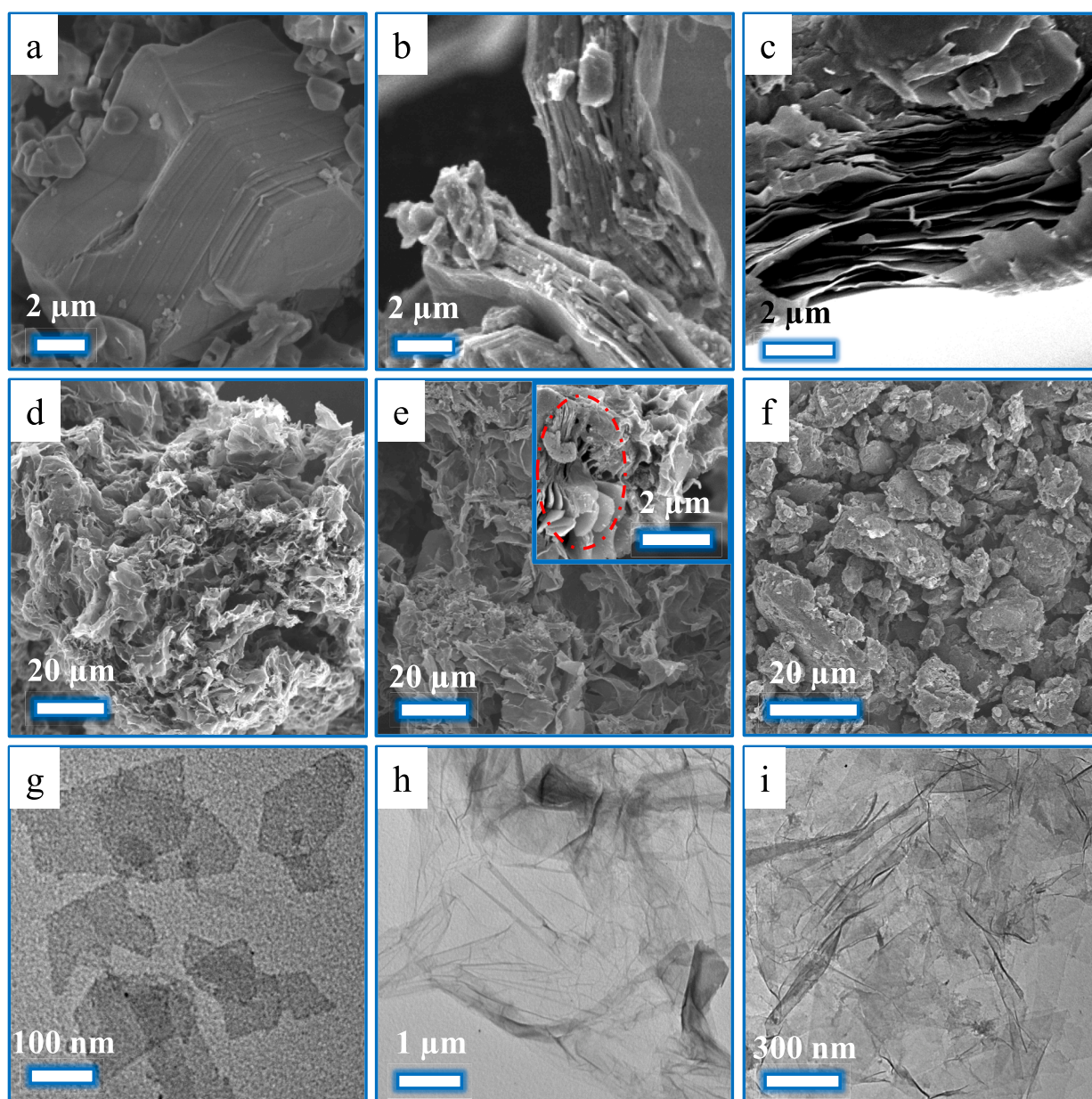


Fig. 1. SEM images: (a) MAX phase, (b) multilayer MXene, and (c) cross-sectional view of MXene sheets filtered on a PVDF membrane, (d) rGO, (e) rGO/MXene10 composite, and (f) (rGO/MXene10)_S60 composite; TEM images: (g) MXene flakes, (h) rGO sheets, and (i) rGO/MXene10 composite.

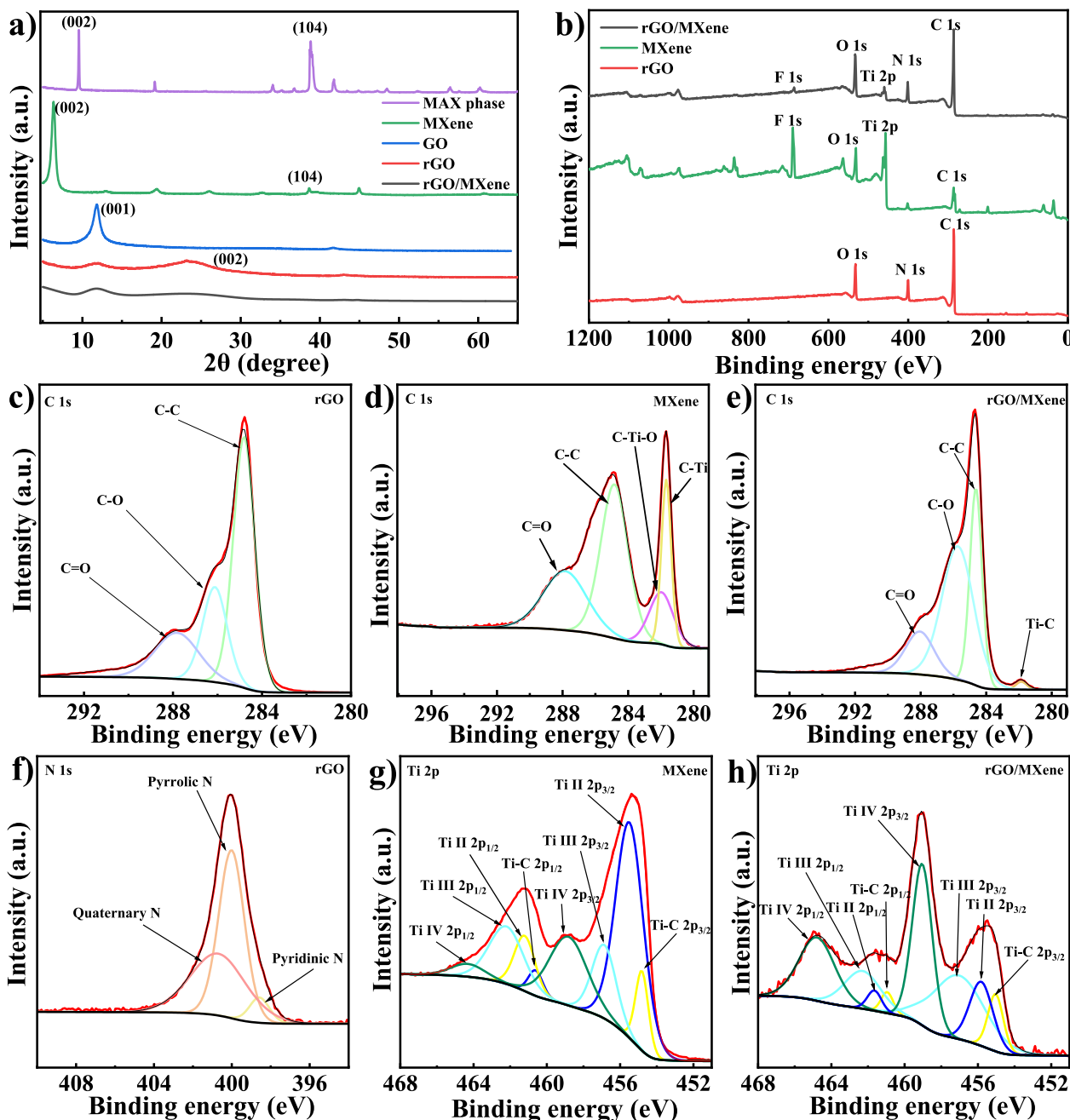


Fig. 2. (a) XRD patterns of MAX phase, MXene, GO, rGO, and rGO/MXene10 composite; (b) Low-resolution XPS survey spectra of MXene, rGO, and rGO/MXene10 composite; High-resolution XPS spectra of (c) C 1s for rGO, (d) C 1s for MXene, (e) C 1s for rGO/MXene10 composite, (f) N 1s for rGO, (g) Ti 2p for MXene, and (h) Ti 2p for rGO/MXene10 composite.

The surface elemental composition and functional groups of the composites were analyzed by X-ray photoelectron spectroscopy (XPS). The full-scan spectrum (Fig. 2b) confirms the successful nitrogen doping in rGO via EDA, as indicated by the presence of N 1s alongside C 1s and O 1s, while MXene exhibits F 1s, O 1s, Ti 2p, and C 1s signals. The presence of abundant oxygen- and fluorine-containing groups on etched MXene confirms successful surface modification. The low-resolution XPS spectrum of rGO/MXene10 verifies the co-existence of rGO and MXene, evidenced by F and Ti from MXene and N from rGO. The high-resolution C 1s spectra (Fig. 2c-e) display five peaks at 281.7, 282.0, 284.7, 286.0, and 288.0 eV, corresponding to C-Ti, C-Ti-O, C-C, C-O, and C=O bonds, respectively [11,12]. In the rGO/MXene10 composite, a decrease in C-Ti and an increase in C-O indicate enhanced oxidation. As shown in Fig. 2f, the N 1s spectrum consists of pyridinic N (398.6 eV),

pyrrolic N (400.0 eV), and quaternary N (401.0 eV) [13]. These nitrogen-containing groups have been reported to improve the affinity of carbon for LiPSs, enhancing Li-S battery performance [13].

The Ti 2p spectra of MXene and rGO/MXene10 (Fig. 2 g, h) exhibit peaks corresponding to Ti-C (2p_{3/2} at 455.0 eV, 2p_{1/2} at 461.0 eV), Ti(II) (2p_{3/2} at 455.8 eV, 2p_{1/2} at 461.6 eV), Ti(III) (2p_{3/2} at 457.1 eV, 2p_{1/2} at 462.3 eV), and Ti(IV) (2p_{3/2} at 459.0 eV, 2p_{1/2} at 464.6 eV), consistent with previous studies [12,14]. Ti(II) and Ti(III) are assigned to surface-terminating groups, while Ti(IV) corresponds to TiO₂. The rGO/MXene composite shows no significant changes in titanium's core chemical state compared to pristine MXene, except for an increased Ti(IV) signal, which is attributed to surface oxidation induced by rGO interaction and probe sonication during composite preparation [12].

Fig. S.2 presents TGA curves of various samples measured under a N₂

atmosphere at $10\text{ }^{\circ}\text{C min}^{-1}$ heating rate to determine sulfur content. The results confirm that all samples contain $\sim 60\text{ wt\%}$ sulfur, aligning with the initial sulfur loading, verifying the effectiveness of the melt diffusion method. A gradual weight loss beyond $330\text{ }^{\circ}\text{C}$ suggests the decomposition of residual oxygen-containing functional groups from GO [5].

The electrochemical performance of Li-S cells was evaluated through charge–discharge cycling at 0.1C (Fig. 3a). The (rGO/MXene10)_S60 cell exhibited the comparatively better performance, achieving an initial discharge capacity of 981 mAh/g and retaining 606 mAh/g after 100 cycles (76 % retention) with a Coulombic efficiency (CE) above 97 %, indicating high reversibility and stable sulfur conversion. Similarly, the (rGO/MXene20)_S60 cell demonstrated capacity retention of 604 mAh/g after 100 cycles. However, unlike the stable cycling of (rGO/MXene10)_S60, it exhibited a gradual decline in later cycles, suggesting that higher MXene content is not favorable for Li-S batteries. In contrast, rGO_S60 and (rGO/MXene30)_S60 showed lower initial discharge capacities (808 and 831 mAh/g , respectively) with more pronounced fading. The rapid decline of the rGO_S60 cell suggests insufficient LiPSs confinement, while the (rGO/MXene30)_S60 did not improve performance, likely due to excessive MXene reducing the availability of active sites for sulfur confinement. The promising performance of the (rGO/MXene10)_S60 suggests that a 9:1 rGO-to-MXene ratio optimally balances conductivity, stability, and LiPSs confinement via rGO's physical trapping and MXene's chemical anchoring.

Fig. 3b presents the galvanostatic charge–discharge profiles of the (rGO/MXene10)_S60 cell at different cycles. The higher-voltage plateau ($2.4\text{--}2.1\text{ V}$) corresponds to the formation of soluble LiPSs (from S_8 to Li_2S_4), while the lower plateau ($\sim 2.1\text{ V}$) represents the transition to insoluble $\text{Li}_2\text{S}_2/\text{Li}_2\text{S}$ [15]. The retention of prominent and well-defined charge and discharge plateaus over multiple cycles further highlights the structural stability and efficient sulfur conversion in the (rGO/MXene10)_S60 cathode. Additionally, long-term cycling at 1C (Fig. 3c)

shows the (rGO/MXene10)_S60 cell retains 514 mAh/g after 365 cycles with a CE of 97.7 %, highlighting its stability and reversibility.

4. Conclusion

The study highlights the potential of rGO/MXene composites as sulfur hosts for Li-S batteries, with the (rGO/MXene10)_S60 electrode exhibiting the most favorable overall performance, delivering 981 mAh/g initially and retaining 606 mAh/g after 100 cycles. Although (rGO/MXene20)_S60 achieved a comparable final capacity of 604 mAh/g , its gradual fading suggests lower cycling stability. In contrast, the rGO_S60 and (rGO/MXene30)_S60 exhibited poorer performance due to limited LiPSs retention and fewer active sites. The optimized 9:1 rGO-to-MXene ratio effectively enhances conductivity, LiPSs anchoring, and overall electrochemical performance. Furthermore, (rGO/MXene10)_S60 retains 514 mAh/g after 365 cycles at 1C with 97.7 % CE, confirming its stability. This work provides valuable insights into the role of MXene content in Li-S batteries, guiding the development of next-generation sulfur cathodes.

CRediT authorship contribution statement

Mukhammed Kenzhebek: Writing – review & editing, Writing – original draft, Validation, Methodology, Investigation, Data curation. **Almagul Mentbayeva:** Writing – review & editing, Supervision. **Zha-zira Supiyeva:** Resources, Funding acquisition. **Asset Kabyshev:** Writing – review & editing, Supervision. **Fail Sultanov:** Writing – review & editing, Supervision, Methodology, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial

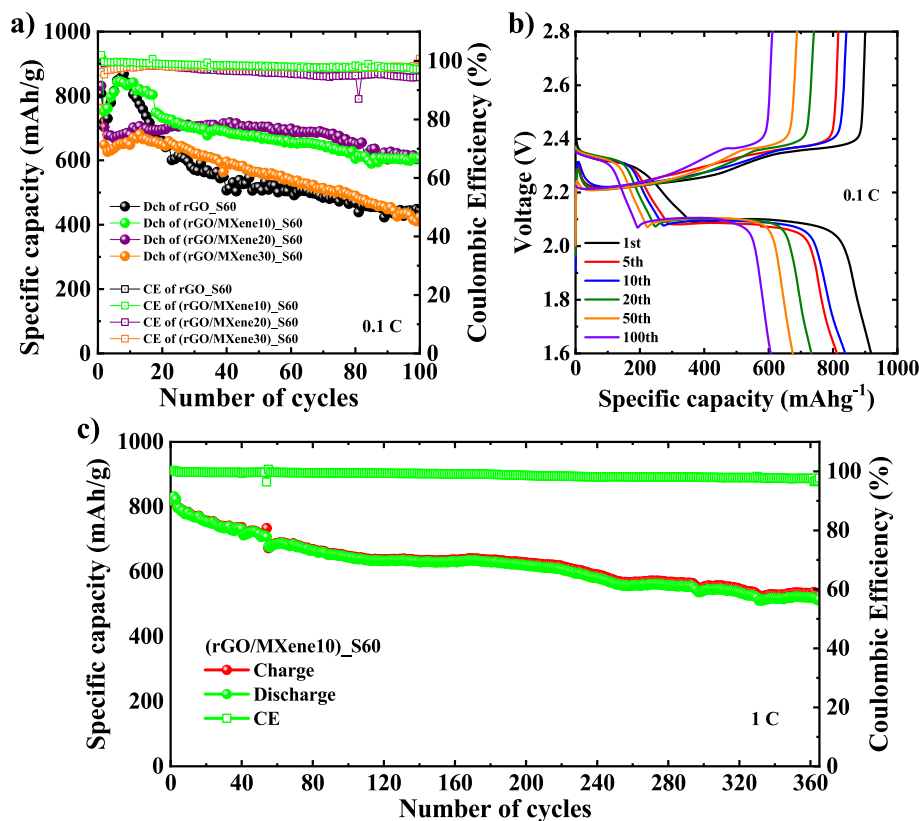


Fig. 3. (a) Charge-discharge cycling performance of Li-S cells at 0.1C , (b) corresponding galvanostatic charge–discharge curves and (c) long-term cycling performance of Li-S cell with (rGO/MXene10)_S60 cathode.

interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.matlet.2025.138773>.

Data availability

Data will be made available on request.

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