Supplementary Material

for

Freestanding Nitrogen-doped MXene/Graphene cathode for high-performance Li-S batteries

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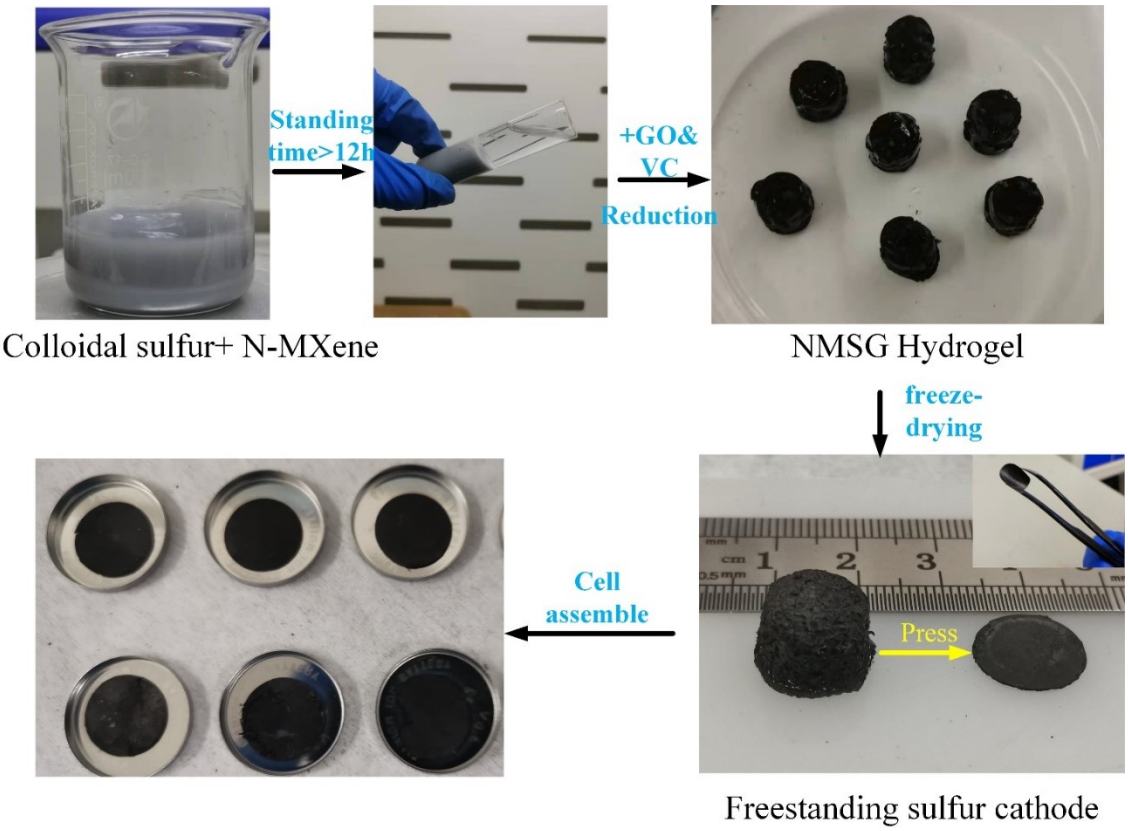

Fig.S1 Photos of the freestanding NMSG electrode synthesis process

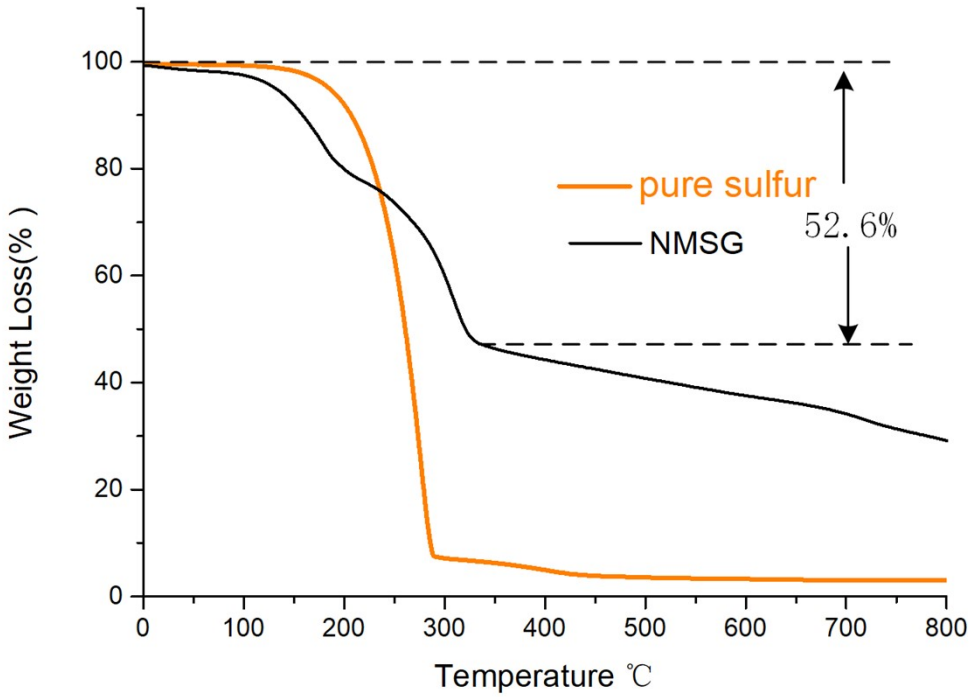

Fig.S2 TGA curve of the NMSG monolith and pure sulfur

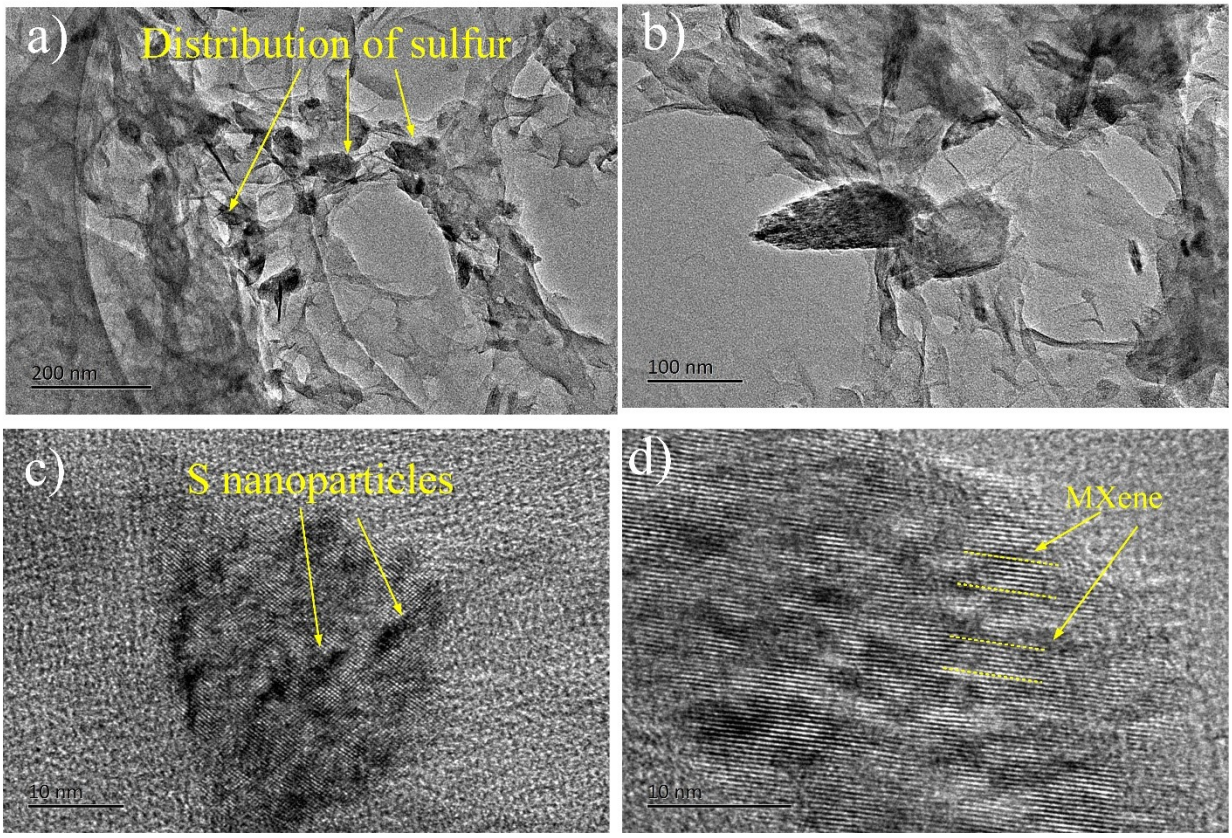

Fig.S3 HTEM images of NMSG at different magnifications

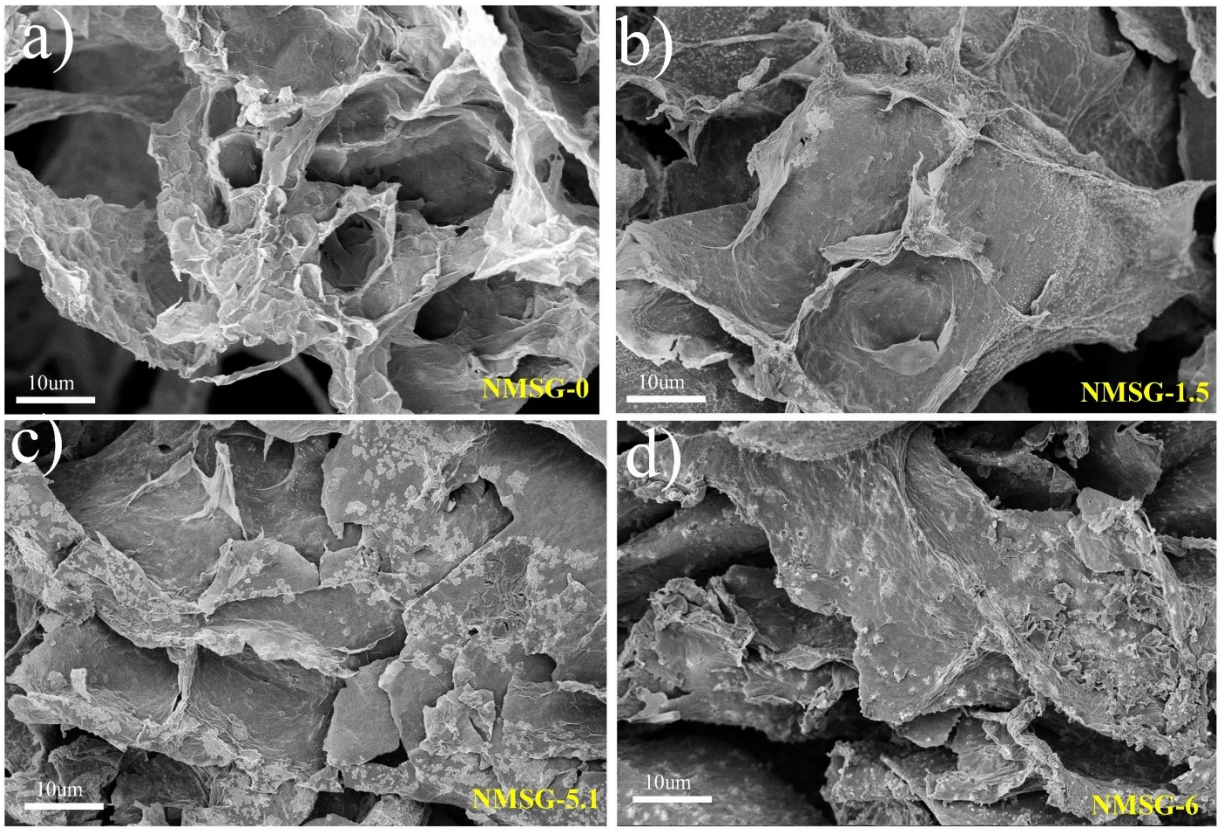

Fig.S4 SEM images of the NMSG composites with different areal sulfur loadings

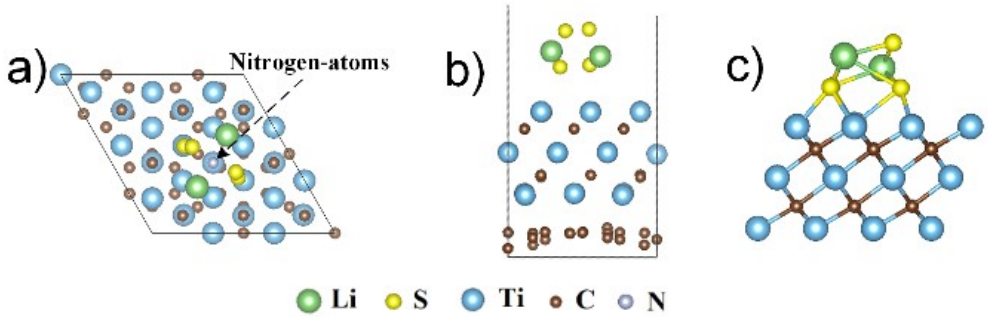

Fig.S5 a)The slab model of Li_2S_4 on the N-Ti_3C_2/graphene surfaces b) Side view of a slab model;

The first principles calculations are performed using DFT in conjunction with projector augmented wave (PAW). The electron structure was calculated using the Vienna Ab initio Simulation Package (VASP)[1], and the interaction between nucleus and electron was described using a projector enhanced wave (PAW) model with perdew-Burke-Ernzerhof (PBE) function[2]. Then, the brillouin region of all systems was sampled using monkhorst-pack grids centered on gamma-points, and the k-point sampling method setup of 3×3×1Monkhorst Pack was used for flat plate geometry optimization. The convergence criteria for force and energy are 0.02 EVA^{-1} and 10^{-5} eV, respectively. Fig.S5) depict the slab model and side view of a representative polysulfide Li_2S_4 on the surface of N-Ti_3C_2/graphene heterojunction.

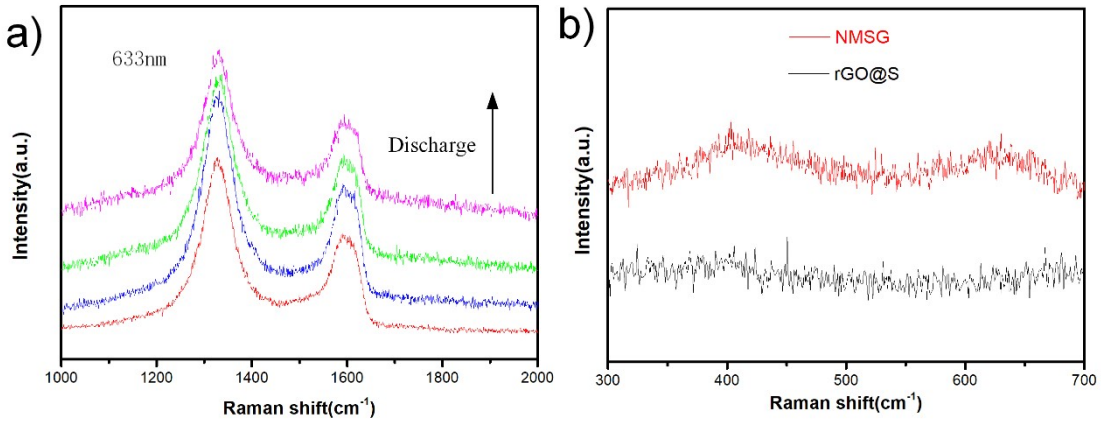

Fig.S6 Normalized Raman spectra of NMSG undergo different discharge depths.

Fig.S6a) shows the normalized Raman spectra of NMSG undergo different discharge depths.Using electrode in the disassembled cell, we tested the cell for every 0.3V voltage drop during the 2.8V discharge process, the The D and G peaks of graphene stability indicate the structural integrity of the NMSG during electrochemical reduction. Fig.S6b) shows Raman spectra of NMSG and rGO@S. The NMSG sample

presented two main characteristic MXene Raman shifts at the peak of 415 and 625 cm^{-1} , indicating the presence of MXene in the NMSG.

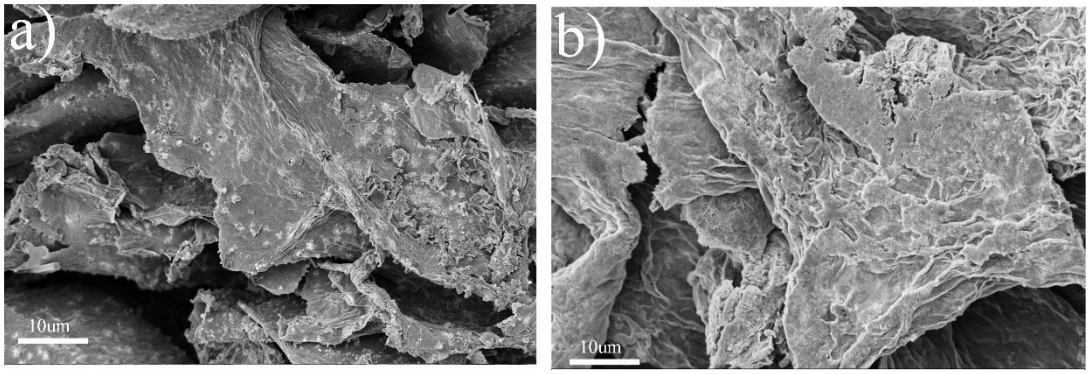

Fig.S7 SEM images of the NMSG-6 electrode a)before cycling and b)after 100 cycles at 0.2C.

Table S1. Comparison of other MXene/S/Graphene ternery hybrids electrode for Li-S battery

	Material	Adsorption Mechnism	Areal sulfur loadng [mg/cm²]	Rate	Fatigue resistance			Ref.
					Cycle	Capacity	Fade ratio per cycle	
1	N-$Ti_3C_2T_x$/S	Chemical	1.5	0.2C	200	950	0.085%	[1]
			5.1	0.2C	500	588	0.046%	
2	$Ti_3C_2T_x$/rGo fibers	Chemical	1.4	0.2C	100	942	0.092%	[2]
			-	-	-	-	——	
3	$Ti_3C_2T_x$/rGo/S	Chemical	1.5	1C	500	596	0.073%	[3]
			6	0.1C	30	879	——	
4	$Ti_3C_2T_x$/rGo/S	Chemical	1.5	0.5C	300	878	0.077%	[4]
			-	-	-	-		
5	**N-$Ti_3C_2T_x$ @S/rGO**	Chemical &Physical	1.5	1C	300	721	0.067%	Our work
			5.1	0.2C	200	611	0.061%	

Reference

[1] Bao W , Liu L , Wang C , et al. Facile Synthesis of Crumpled Nitrogen‐Doped MXene Nanosheets as a New Sulfur Host for Lithium–Sulfur Batteries . Advanced Energy Materials, 2018, 8(13):1702485.
[2] Li H , Shao F , Wen X , et al. Graphene/MXene fibers-enveloped sulfur cathodes for high-performance Li-S batteries. Electrochimica Acta, 2021:137838.
[3] Song J , X Guo, Zhang J , et al. Rational design of free-standing 3D porous MXene/rGO hybrid aerogels as polysulfide reservoirs for high-energy lithium–sulfur batteries[J]. Journal of Materials Chemistry A, 2019.
[4] Bao W , Xie X , Xu J , et al. Confine sulfur in 3D flexible hybrid MXene/reduced graphene oxide nanosheets for lithium sulfur battery[J]. Chemistry, 2017.