Electronic Supplementary Information

Interfacial Competition between Borophene-Based Cathode and Electrolyte for the Multiple-Sulfide Immobilization of Lithium Sulfur Battery†

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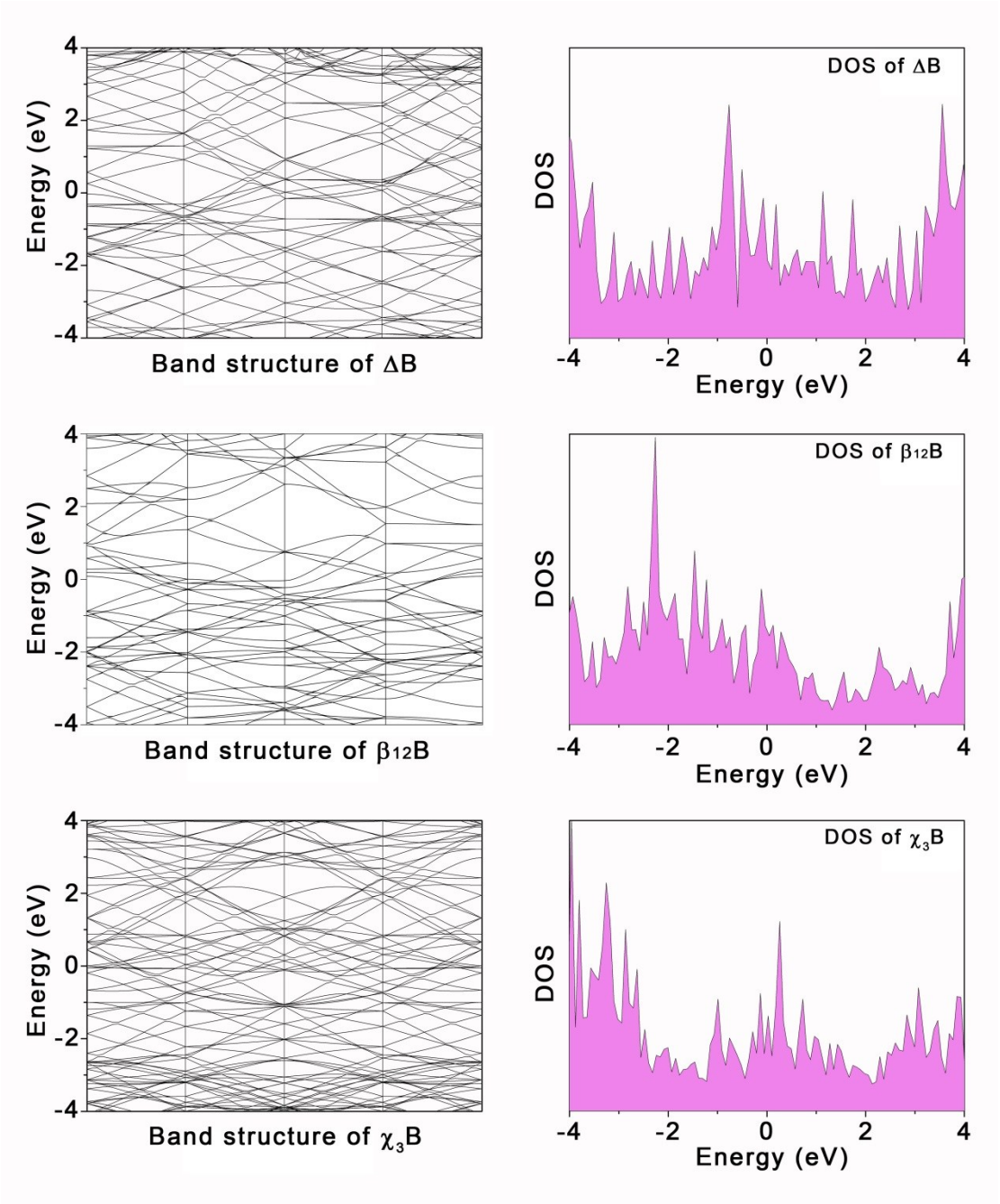

Fig. S1 Band structures and density of state (DOS) of borophene sheets. (The Fermi level has been shifted to zero).

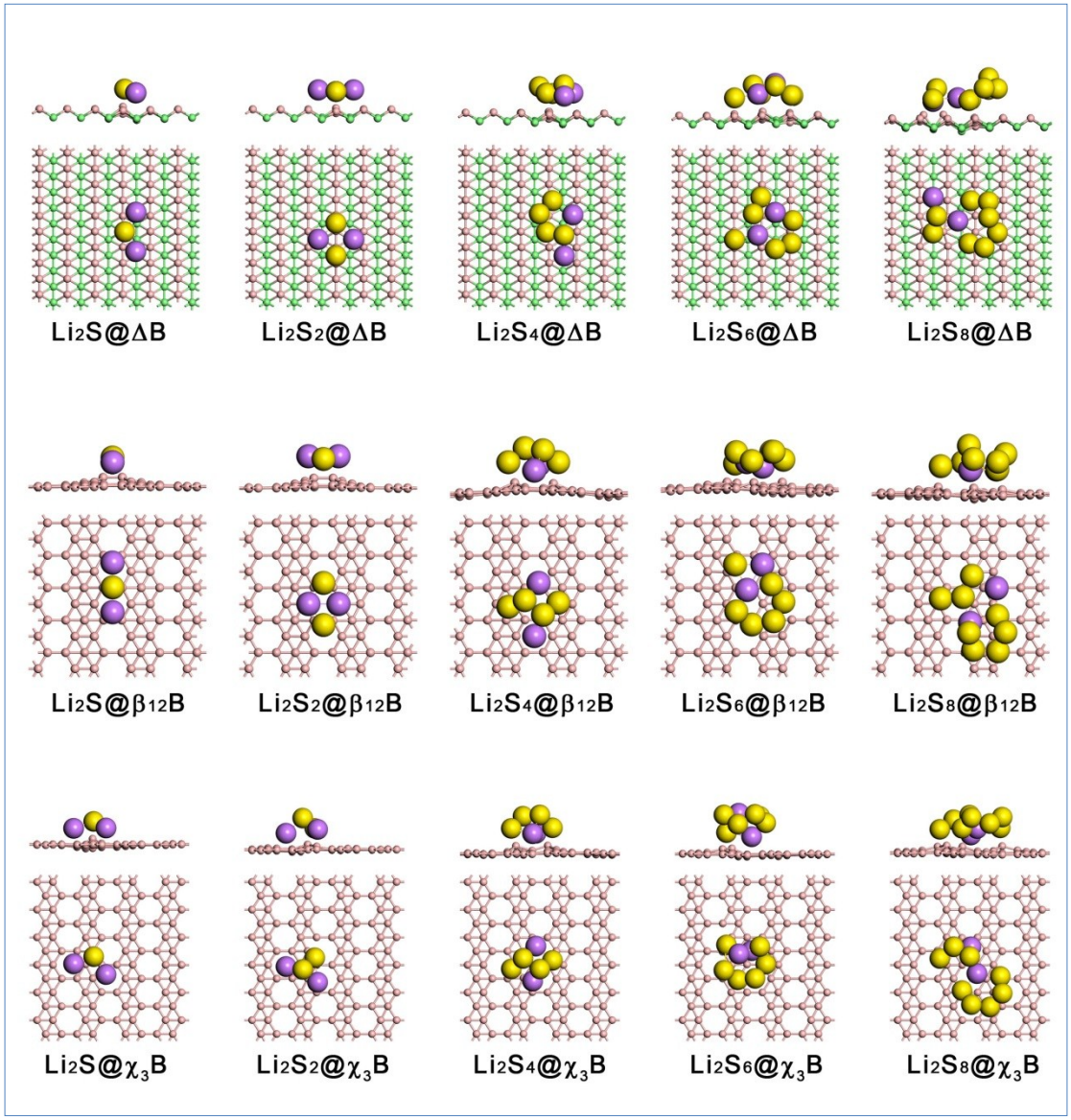

Fig. S2 Optimized structures of single Li_2S_n on borophene sheets.

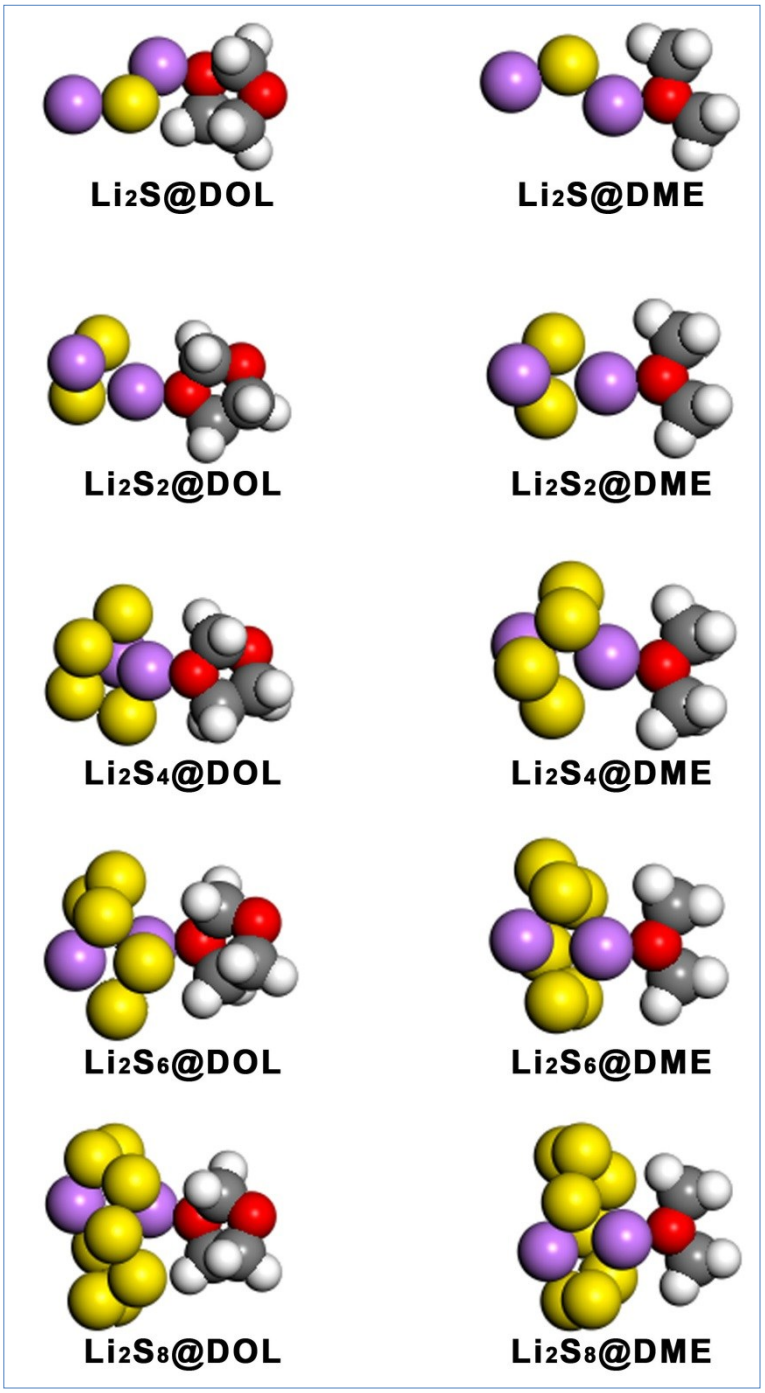

Fig. S3 Optimized structures of Li_2S_n around solvents (DOL and DME) of electrolyte.

Table S1. Binding energies of DOL and DME on borophene sheets. (unit: eV)

	DOL	DME
ΔB	**-0.3492**	**-0.2521**
β_{12}B	**-0.3779**	**-0.2793**
χ_3B	**-0.3075**	**-0.247**

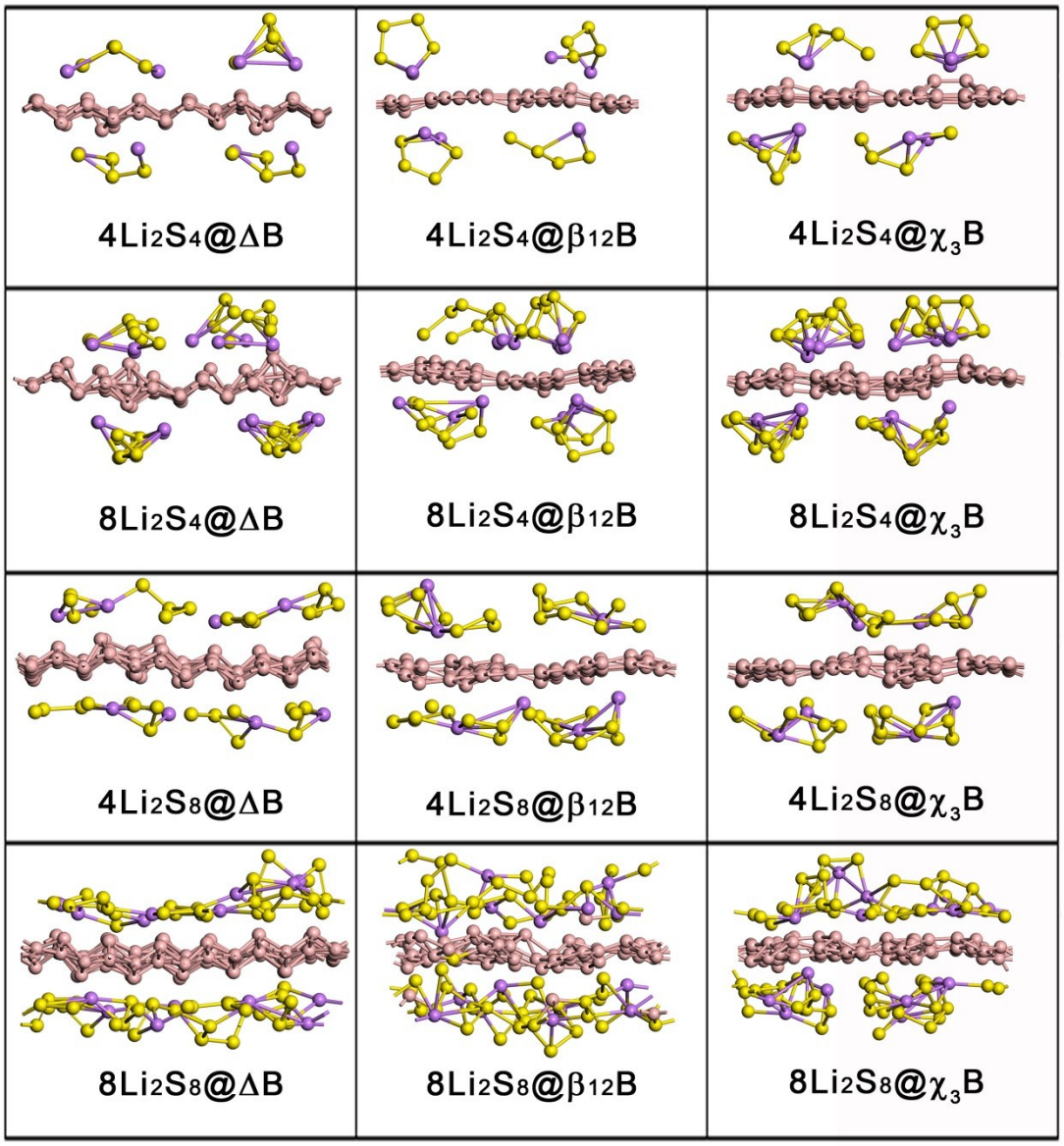

Fig. S4 Optimized structures for multi-Li_2S_n on borophene sheets.

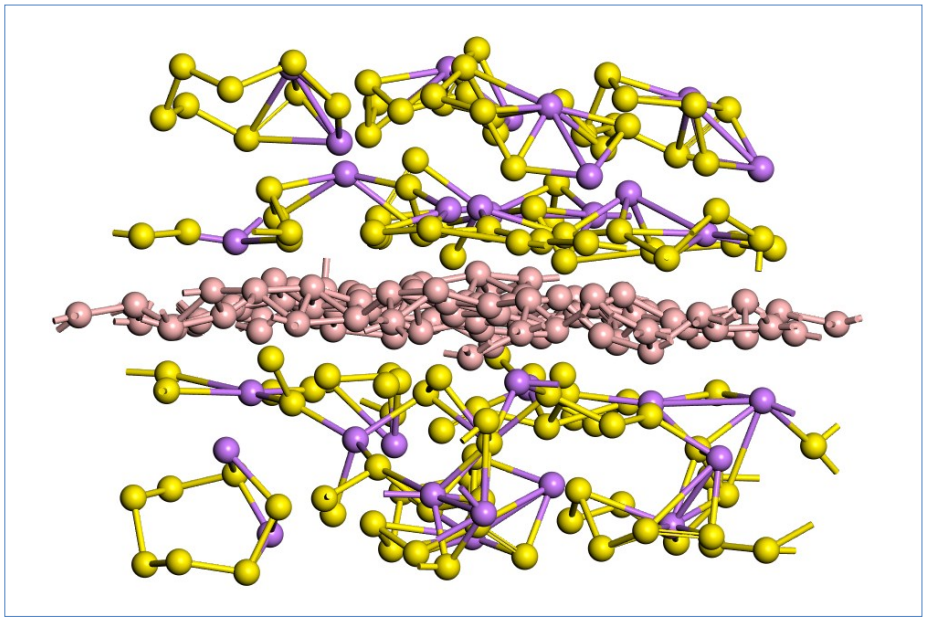

Fig. S5 The optimized structure of 16 Li_2S_8 on χ_3B.

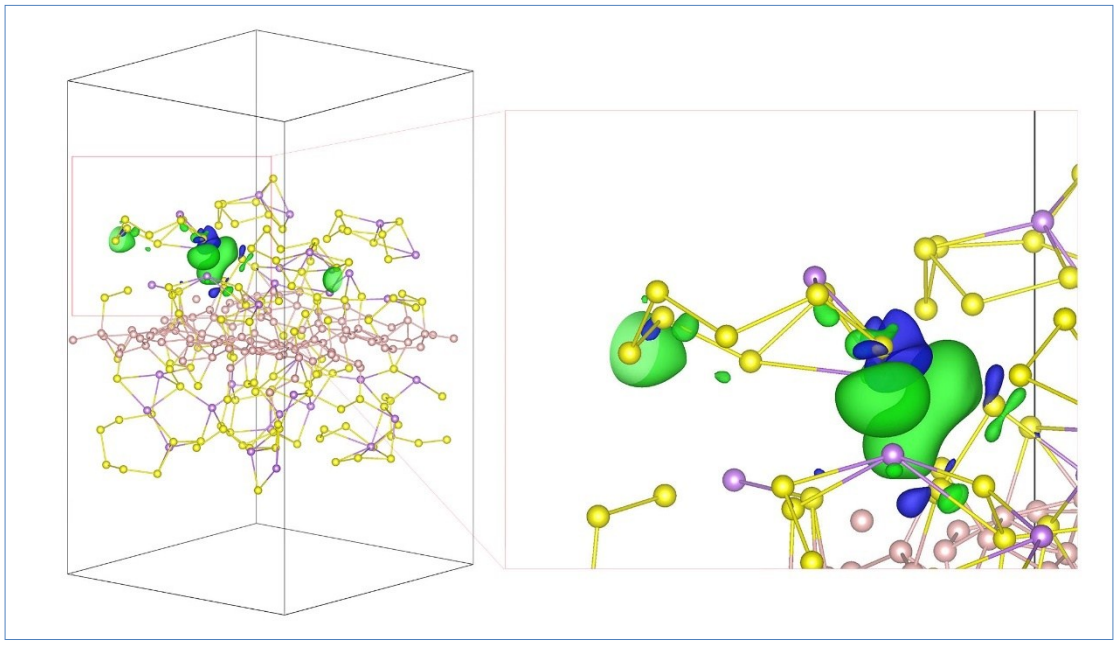

Fig. S6 The charge re-distribution among Li_2S_8 molecules. Green areas: charge accumulation, blue areas: charge depletion.

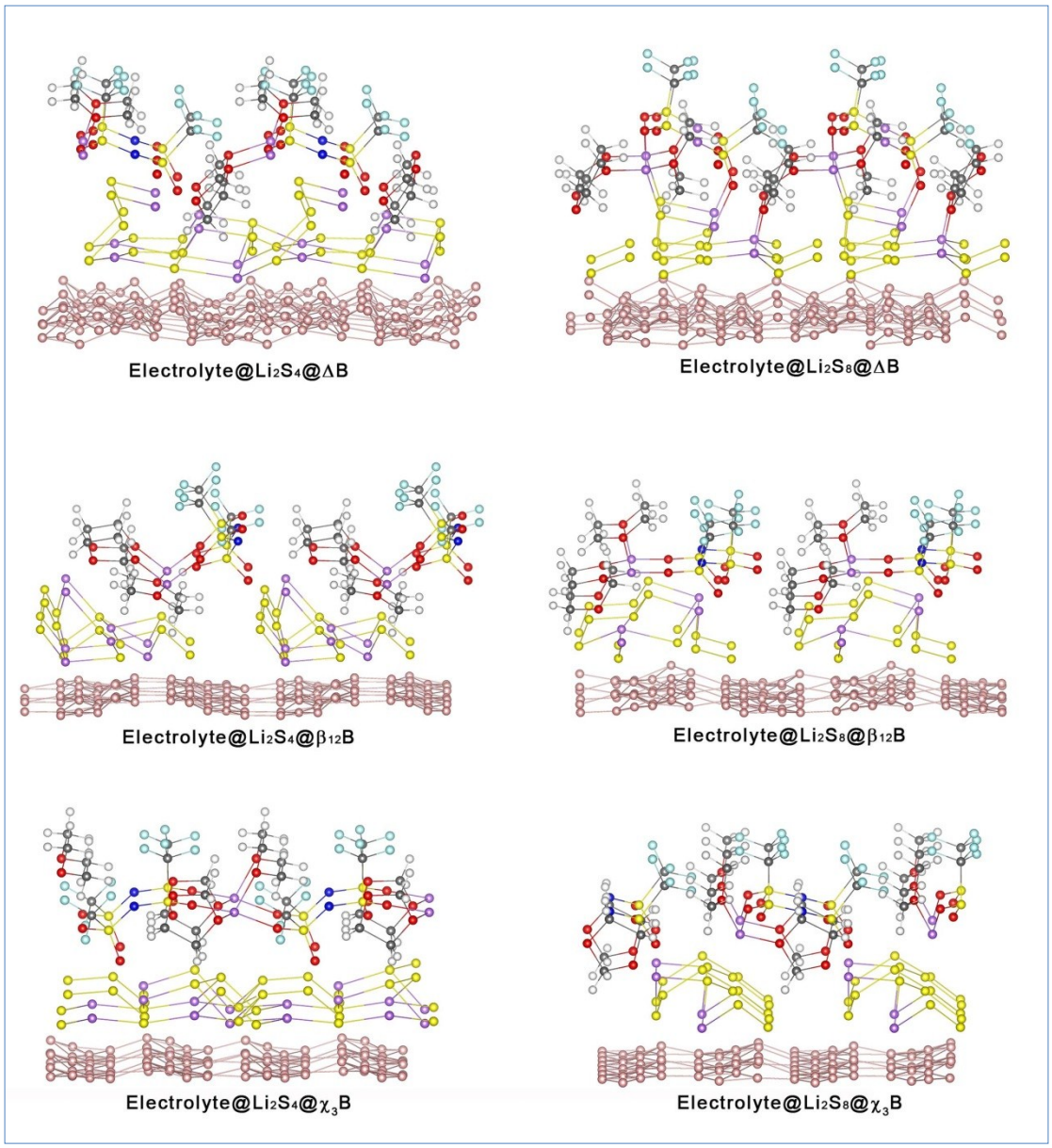

Fig. S7 The geometric structures of multiple Li_2S_4 and Li_2S_8 on borophene nanosheets after 25 ps FP-MD simulations at 300 K with both DOL and DME molecules included.

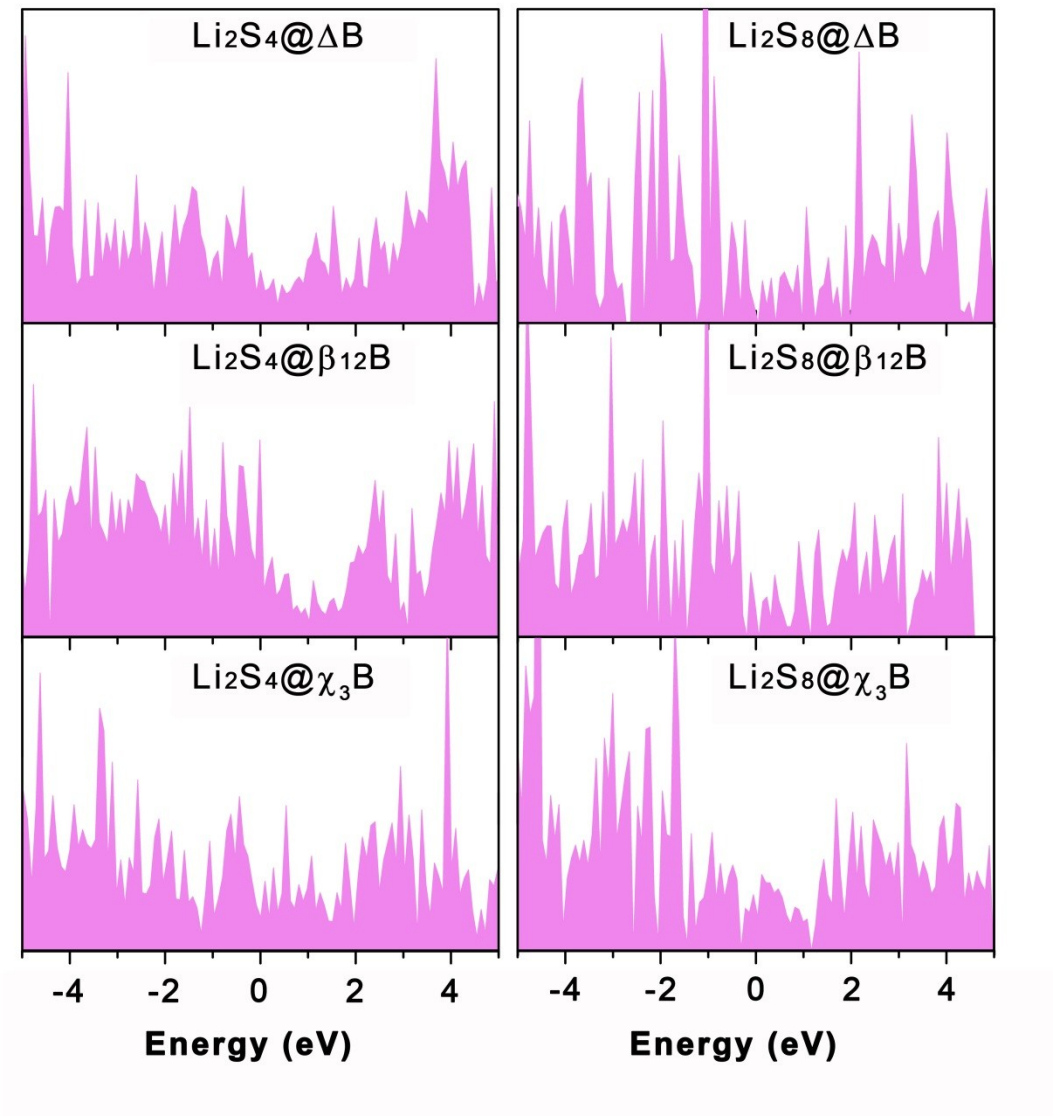

Fig. S8 Density of state for Li_2S_n (n=4,8) on borophenes.

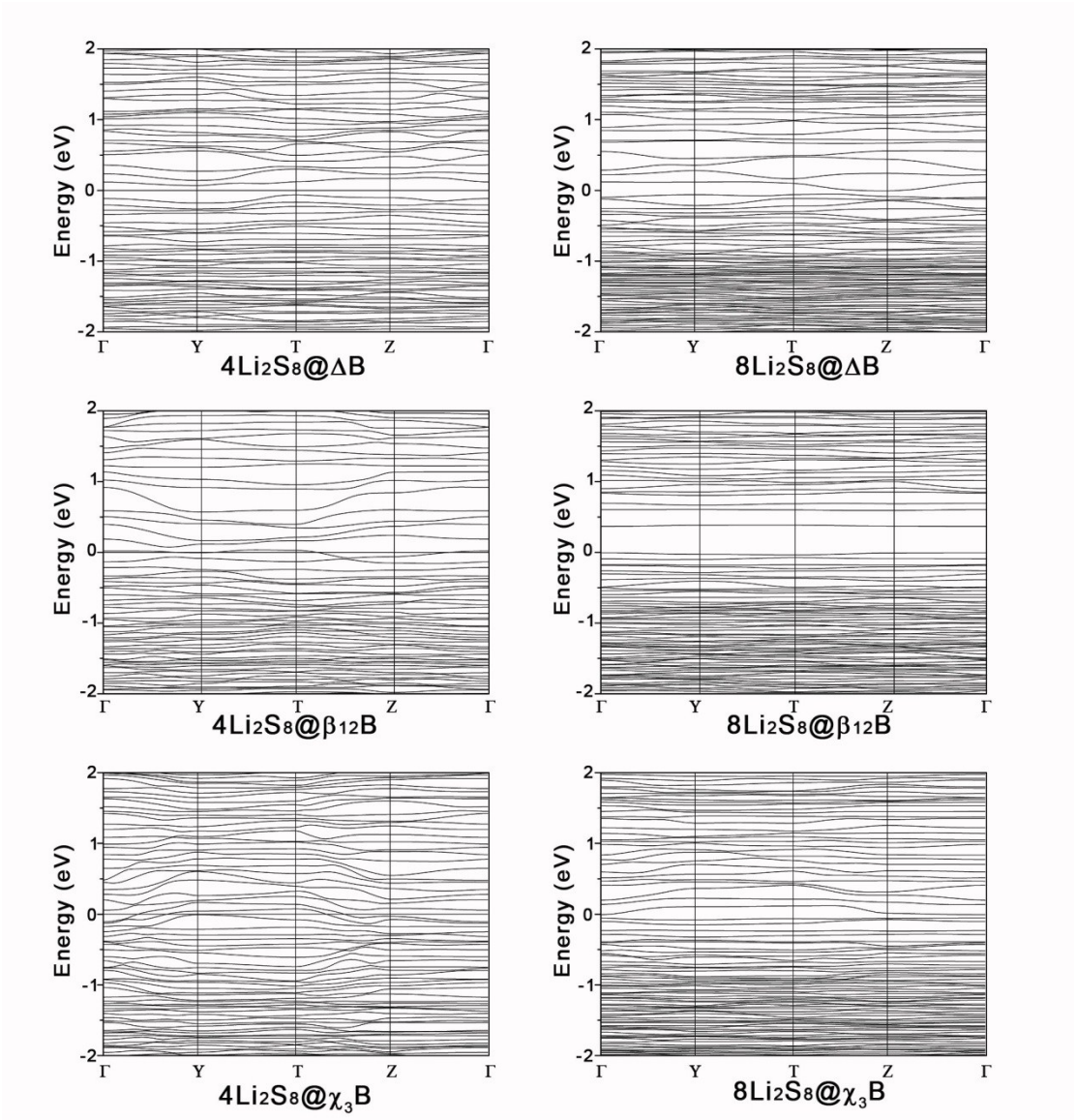

Fig. S9 Band structures of $m$$Li_2S_8$ (m=4,8) on borophenes.

The Mass Loading of Sulfur on graphene

To test the method of mass loading, we firstly optimized 8 Li_2S_8 on graphene, as in Fig. S10, from which from which we know that graphene cannot hold such an amount of sulfides, and lot of sulfides are clustered and leave away from graphene. And then, we found that 4 Li_2S_8 can be effectively adsorbed on both side of graphene for the average binding energy slightly higher (~0.05 eV) than Li_2S_8 to solvents. In optimized $4Li_2S_8$@graphene, the total sulfur loading is ~40 wt%. More calculation demonstrated that one Li_2S_8 cannot be efficiently adsorbed on graphene for the lower binding energy of one Li_2S_8 on graphene than that to solvents. Therefore, the utilized sulfur on graphene should be 33.36 wt% for effectively used molecules of 3 Li_2S_8.

We only find few experimental works about pure graphene cathodes of Li-S battery. Li et al. [*Chem. Commun.*, 2012, **48**, 4106–4108] prepared graphene-sulfur mixture as the cathode of Li-S battery, in which the mass of pure sulfur is 67 wt%. Based on their measurement data, the initial discharge capacity is 767 mAh g^{-1}, indicating that the sulfur utilization is 30.7 wt% of mixture (67 wt% × 767/1675 = 30.68 wt%, where the 1675 represents the theoretical capacity of sulfur of 1675 mAh g^{-1}) , only slightly lower than our predicted result. Although, lot of factors are neglected or simplified, our result is still closer to experimental data, which can be attributed to some positive affect from practical conditions, including attraction from lithium salts, porous surfaces and additives. Moreover, the experimental data of the mass loading of sulfur is evaluated based on pure sulfur mixed with host materials, which is much higher than real sulfur utilization. Meanwhile, our calculated sulfur mass loading can represent the theoretical initial utilization of sulfur. Hence, our method should be more scientific to qualitatively evaluate potential hosts.

Overall, our method about sulfur loading should be reasonably used to evaluate host materials.

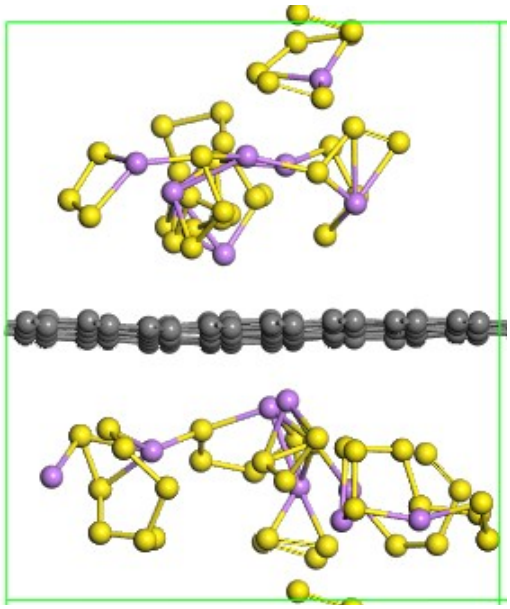

Fig. S10 The optimized structure of $8Li_2S_8$ on monolayer graphene