Influence of iron oxide-based mesoporous graphene frameworks for lithium-sulfur battery separator

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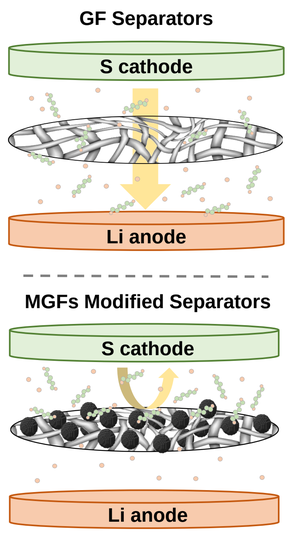
Introduction

Through the development of portable electronics and electric vehicles industries, the importance of energy storage systems that have greater battery capacity and are more environmentally friendly than conventional Li-ion batteries elevated for next-generation batteries. Lithium-Sulfur Batteries (LSBs) have been attention to their high theoretical specific capacity (1675 m Ah g^{-1}).

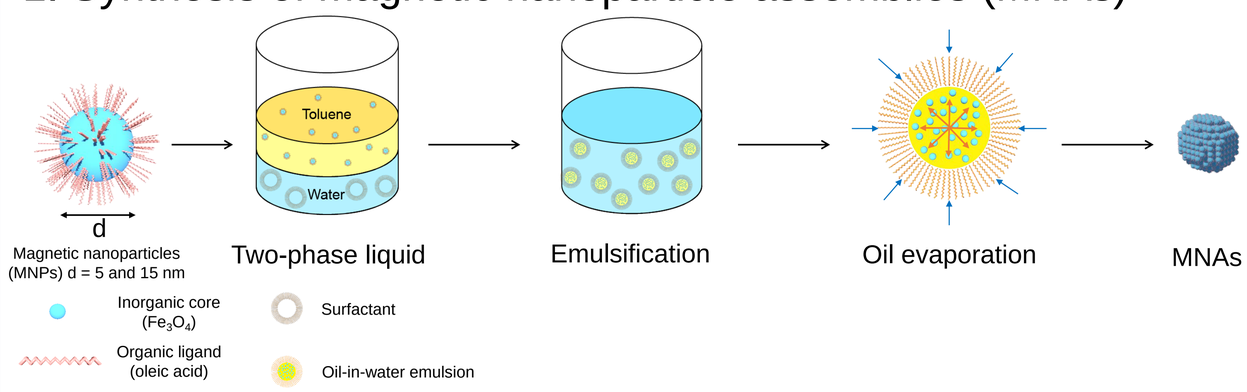
Despite of their advantages, diffusion of intermediate high-order lithium polysulfide species (LiPSs) in charge / discharge process impedes LSBs performance by the corrosion in lithium anode and deactivation of electrodes. Separators, modified with mesoporous graphene frameworks (MGFs), may effectively limit the diffusion of lithium polysulfide by its physical interaction with long-range ordered mesoporous carbon network with distinctive pore sizes. The modified separator with MGFs might be one of the promising approaches to suppress diffusion of high-order LiPSs.

Here, we fabricated modified separator with MGFs with different pore sizes (5 nm and 15 nm), evaluated diffusion rate over the time.

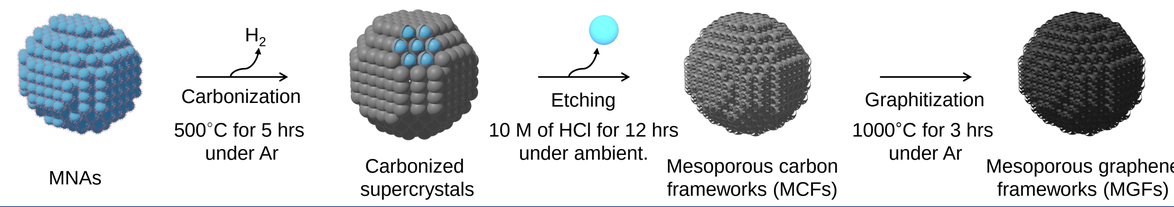


Experimental Methods

1. Synthesis of magnetic nanoparticle assemblies (MNAs)



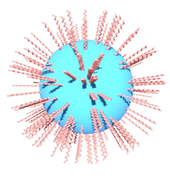
2. Synthesis of MGFs



Results

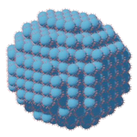
1. Synthesis of MGFs

(a) MNPs



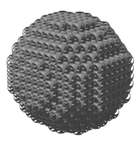
Monodispersed MNPs synthesized by nucleation and growth mechanism

(b) MNAs



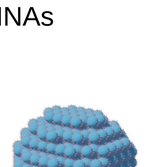
MNAs synthesized by emulsification showing supercrystal structure

(c) MCFs

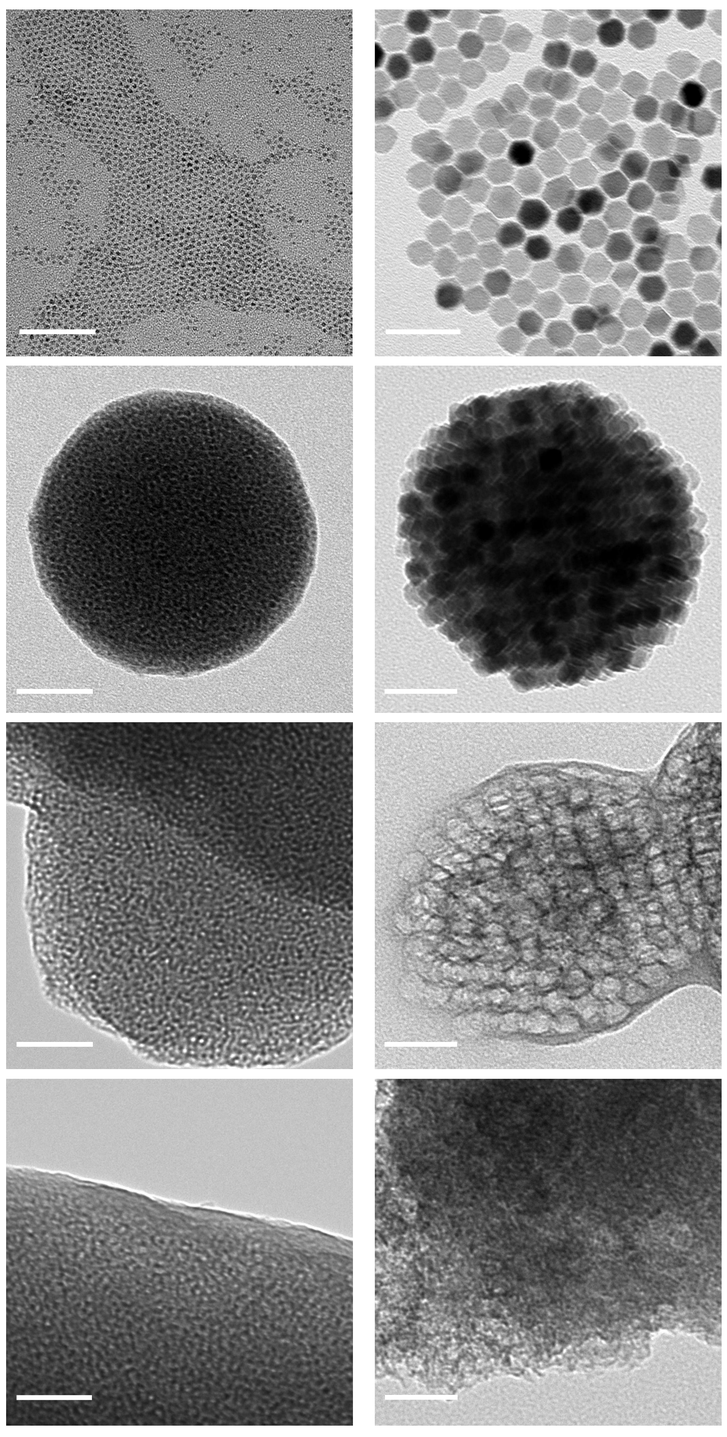


Bright areas surrounded by dark carbon layers showing porous structure

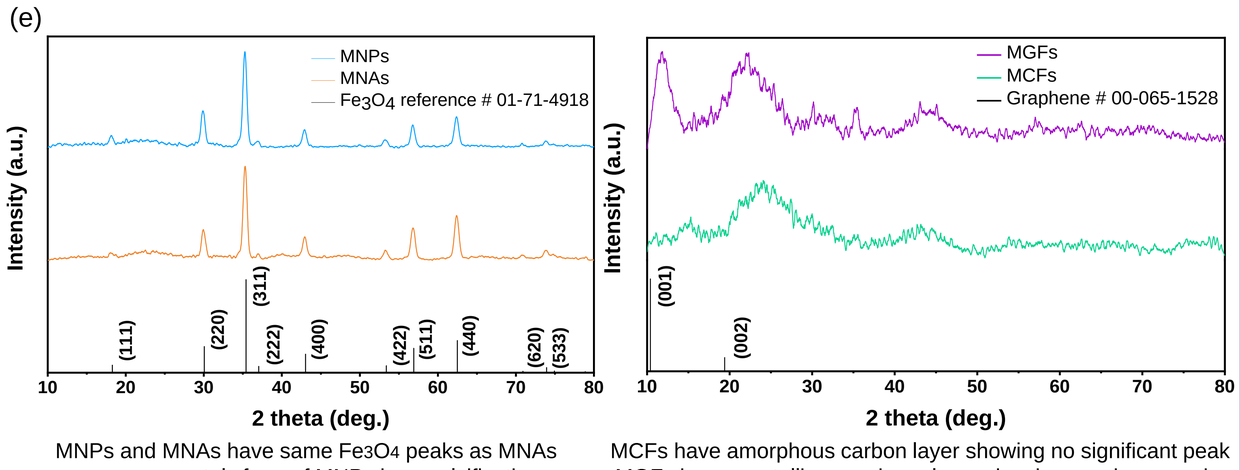
(d) MGFs



Porous structure maintained after graphitization



(e)



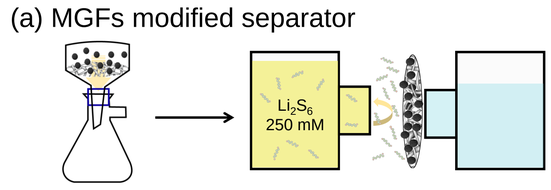
MNPs and MNAs have same Fe_3O_4 peaks as MNAs are supercrystals form of MNPs by emulsification

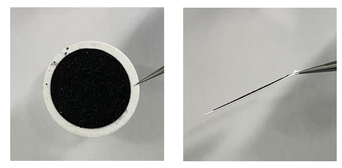
MCFs have amorphous carbon layer showing no significant peak MGFs have crystalline graphene layer showing graphene peaks

Fig. 1. (a) TEM images of 5 nm and 15 nm of Fe_3O_4 based MNPs, (b) TEM images of 5 nm and 15 nm of Fe_3O_4 based MNAs, (c) TEM images of 5 nm and 15 nm pore sized MCFs, (d) TEM images of 5 nm and 15 nm pore sized MGFs, all scale bars are 50 nm scale (e) XRD data for MNPs, MNAs, MCFs and MGFs

2. High-order LiPSs Li_2S_6 diffusion test

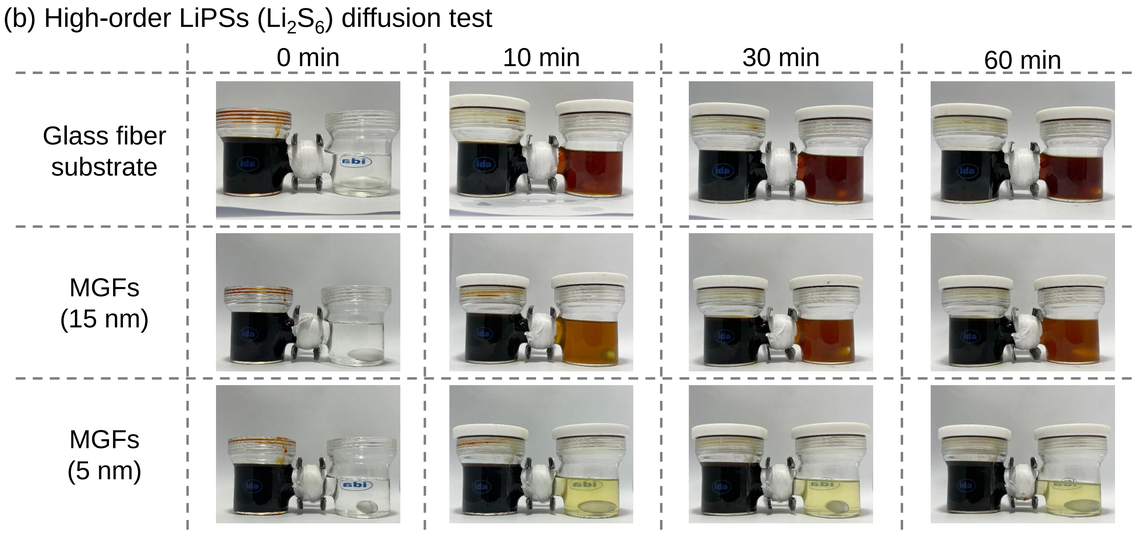
(a) MGFs modified separator

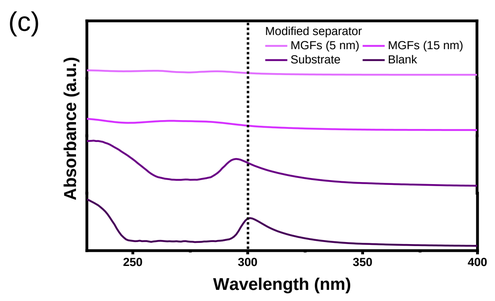




Modified separators with different pore sized MGFs were fabricated by vacuum filtration approach Li_2S_6 solution, 250 mM in Dimethoxy ethane and 1,3-dioxolane solvent (v : v = 1 : 1)

(b) High-order LiPSs (Li_2S_6) diffusion test





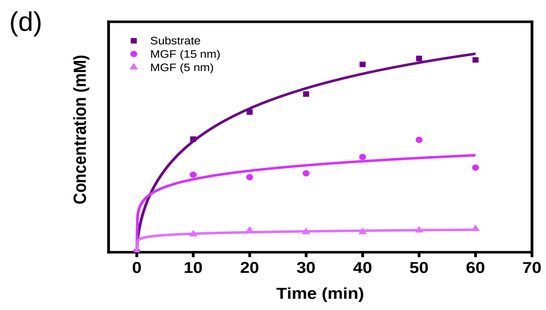


Fig. 2. Li_2S_6 adsorption test: (a) photographs of front and side views of modified separators with MGFs, (b) photographs of H-type cell with Li_2S_6 (left) and DOL/DME solution (right) with modified separators (5 nm and 15 nm MGFs and substrate) after 0/10/30/60 minutes, (c) UV-vis absorption spectra (50 min) of modified separators after 50 minutes (d) concentrations of Li_2S_6 over time by modified separators with dilution factors of 100

Conclusion & Further Study

Modified separators with different pore sized MGFs have been fabricated to effectively suppress diffusion of LiPSs through physical confinement of LiPSs. To control the pore size of MGFs, we used 5 nm and 15 nm MNPs as building blocks. Glass fiber separators are modified with different pore sized MGFs for Li_2S_6 diffusion test to evaluate their ability to suppress LiPSs. Experimental results indicated that modified separators with 5 nm pore sized MGFs most effectively suppress diffusion of high-order LiPSs and glass fiber separators suppress the least. These results were validated through UV-vis absorption spectra where separators coated with 5 nm pore size of MGFs showed the lowest concentration at 60 minutes.

For further studies, chemical adsorption of LiPSs can be an alternative solution to suppress LiPSs diffusion. FeS is known for its excellent ability to adsorb LiPSs, which could be used to chemically suppress LiPSs diffusion. We will synthesize MCFs with FeS core, which will chemically adsorb LiPSs through FeS core and physically confine diffusion of LiPSs. To synthesize FeS core, iron oxide cores will be partially etched. Then, we will convert O to S through anion exchange, forming FeS cores.

Reference

[1] A. Dong et al., *Nano Research* **2017**, 7, 2495-2507 [2] A. Dong *et al.*, *Chem. Mater.* **2016**, *28*, 3823-3830 [3] T. Hyeon *et al.*, *Nature Materials* **2004**, *3*, 891-895 [4] T. Hyeon *et al.*, *J. Am. Chem. Soc.* **2011**, *133*, 12624-12631 [5] Y. Cui *et al.*, *Energy Storage Materials* **2018**, *13*, 241-246