

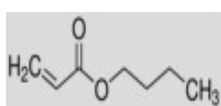
## Supporting Information

### Microgel-Assisted Assembly of Hierarchical Porous Reduced Graphene Oxide for High-Performance Lithium-Ion Battery Anodes

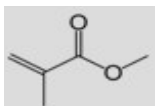
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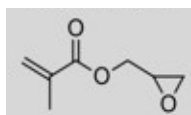
<sup>‡</sup>Department of Physics, Kansas State University, Manhattan, KS 66506, USA



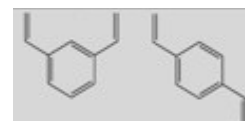
BA



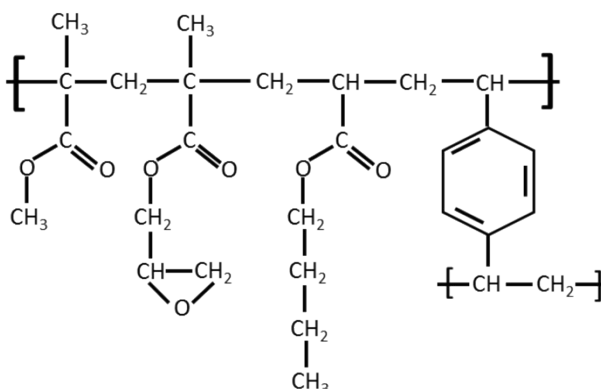
MMA



GMA



DVB



### Cross-linked P (MMA-GMA-BA)

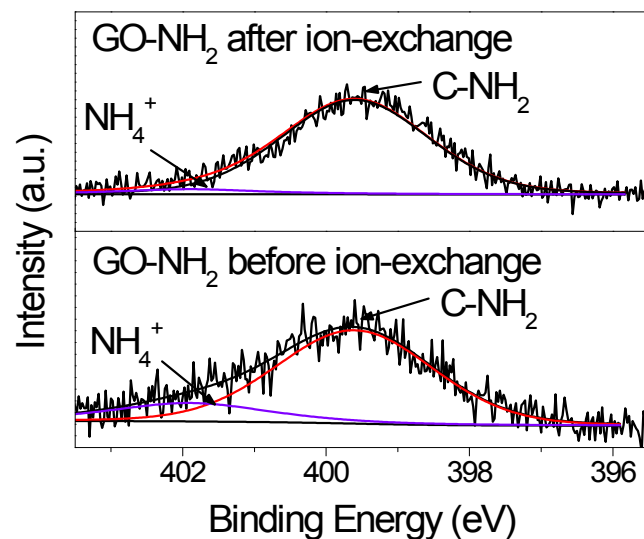
**Scheme S1.** The molecular structure of monomers used for synthesis of cross-linked P (MMA-GMA-BA) spheres and the molecular structure of P (MMA-GMA-BA) spheres.

**Table 1.** Summary of textural properties of stacked RGO and porous/wrinkled RGO obtained from physisorption; SSA and pore volume were obtained by BET and NLDT methods, respectively.

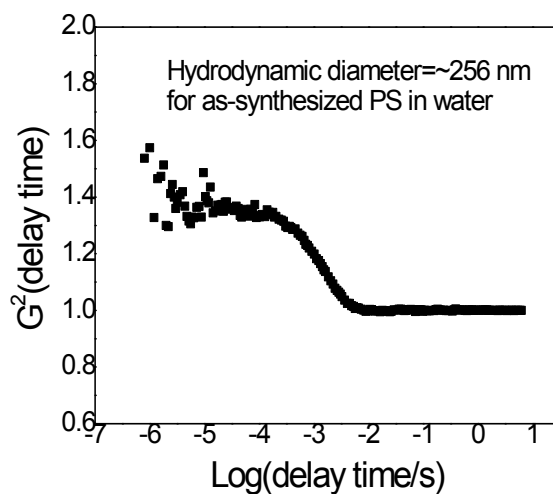
<sup>1</sup> Corresponding Author

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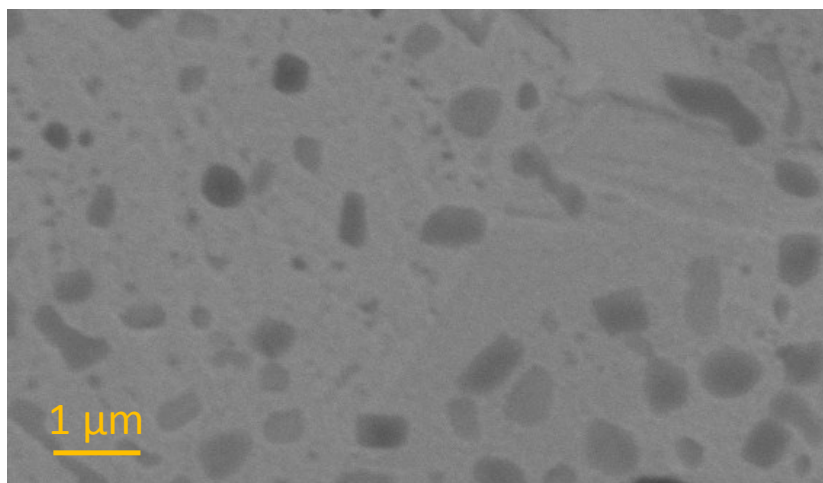
| Sample                                      | Stacked RGO | Porous/wrinkled RGO |
|---------------------------------------------|-------------|---------------------|
| SSA, m <sup>2</sup> /g                      | 96          | 276                 |
| V (2< pore size <10 nm), cm <sup>3</sup> /g | 0.27        | 0.39                |
| V (pore size <2nm), cm <sup>3</sup> /g      | ~0          | 0.0043              |



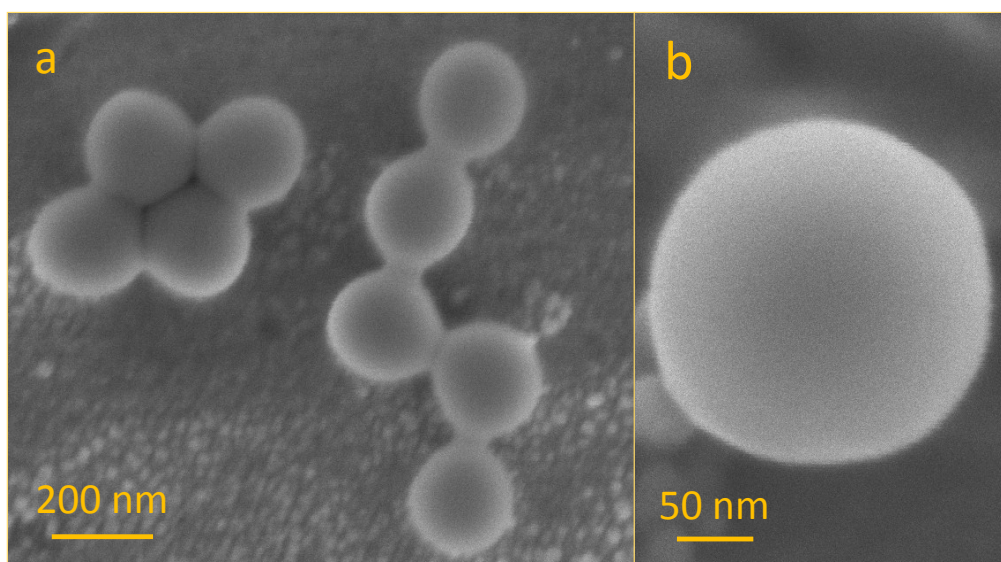
**Figure S1.** XPS N 1s spectra of GO-NH<sub>2</sub> before and after ion exchange with HCl.



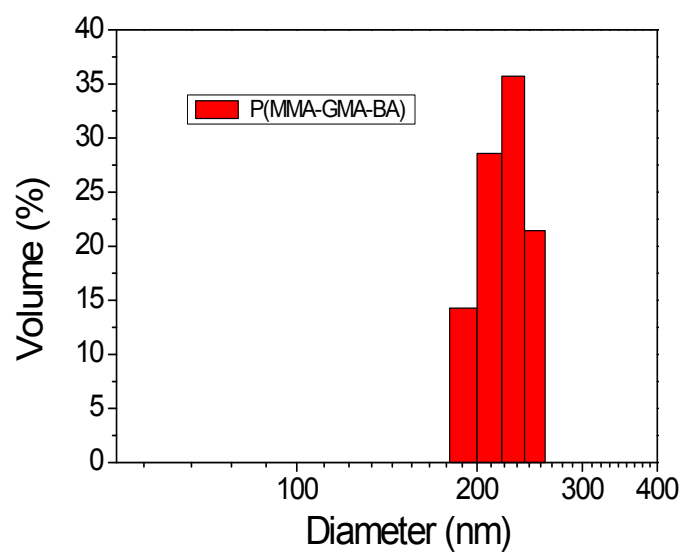
**Figure S2.** Correlation function  $G^2(t)$  measured by DLS on the diluted PS/water dispersion at 25°C.



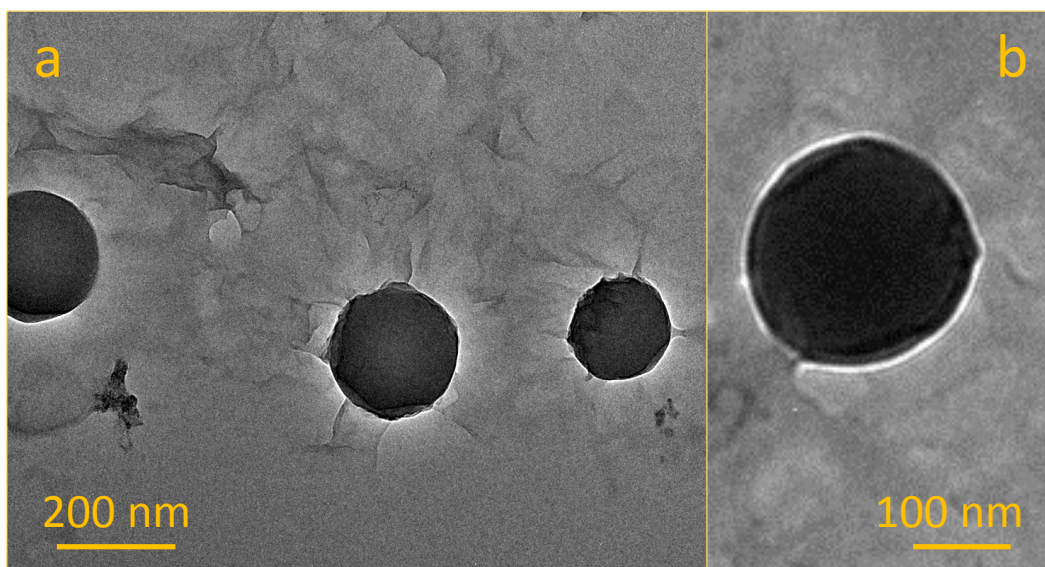
**Figure S3.** The SEM images of GO-NH<sub>2</sub> flakes.



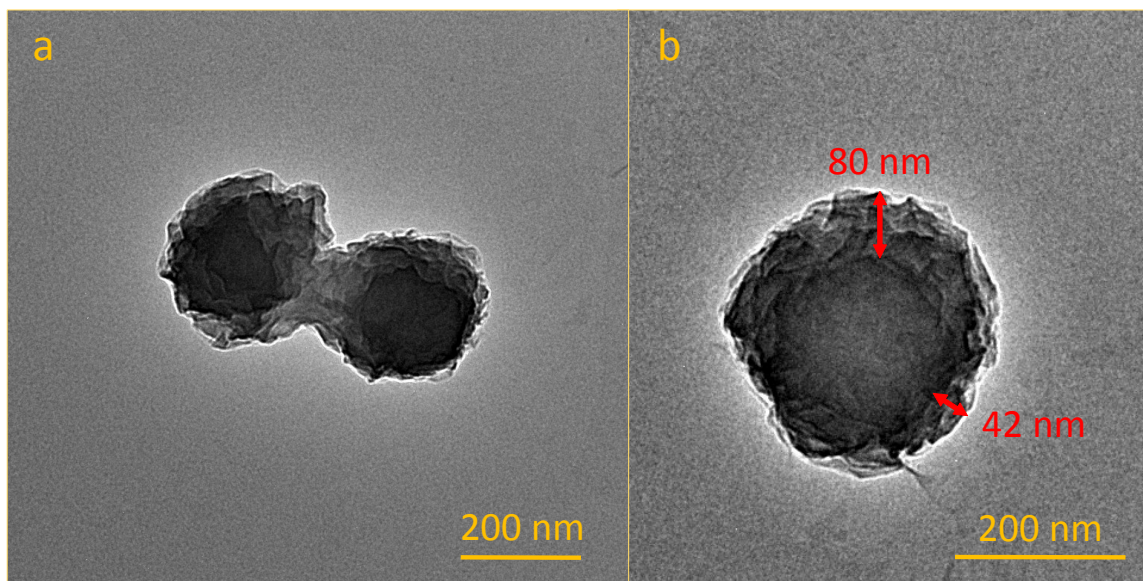
**Figure S4.** (a) and (b) SEM images of as-synthesized PS (0.1wt% DVB).



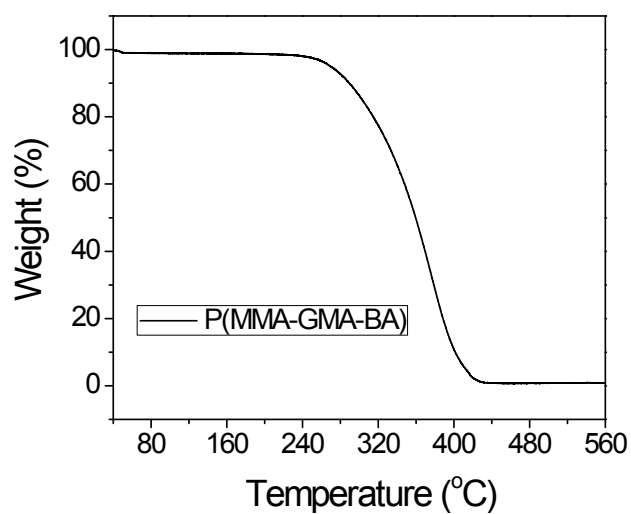
**Figure S5.** Particle size distribution of as-synthesized PS (0.1 wt% DVB).



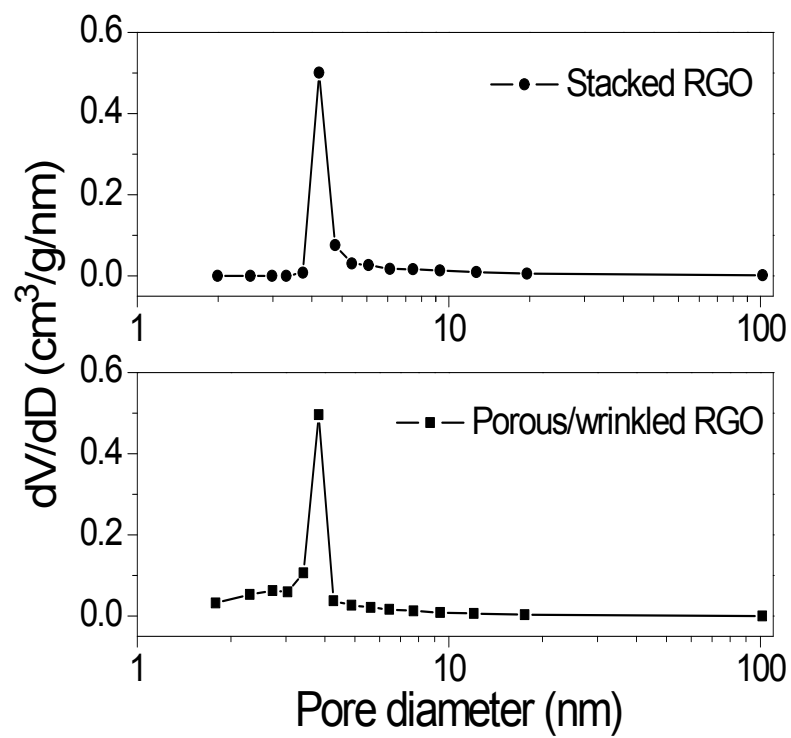
**Figure S6.** (a) and (b) TEM images of GO wrapped as-synthesized PS (0.1wt% DVB) with smooth GO surfaces.



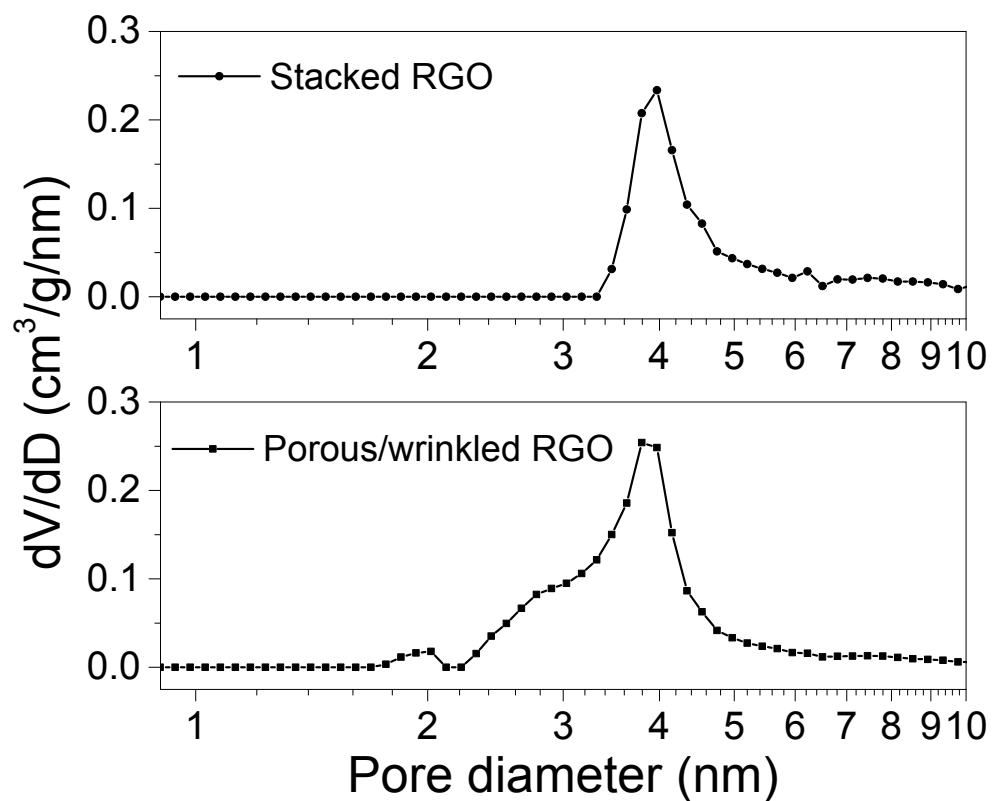
**Figure S7.** (a) and (b) TEM images of wrinkled GO@shrunk PS with wrinkled GO surfaces.



**Figure S8.** Weight loss vs. temperature curve by TGA analysis of cross-linked P (MMA-GMA-BA) microspheres (0.1 wt%DVB).



**Figure S9.** Pore size distributions modeled by the BJH method for stacked and porous/wrinkled RGO.



**Figure S10.** Pore size distributions modeled by the NLDT method for stacked and porous/wrinkled RGO.

### Calculation of Mass Ratio of Polymer Spheres (PS) to GO

Mass ratio of polymer spheres (PS) to GO is ~2:1. The mass ratio was estimated as follows:

$\text{Volume (PS)} * \text{Density(PS)} = \text{Mass ratio (PS/GO)} * \text{Surface area (PS gel)} * \text{Thickness(GO)} * \text{Density(GO)}.$

$\text{Volume (PS)} = \sim 8780076 \text{ nm}^3$  (DLS diameter = 256 nm);

$\text{Density (PS)} = \sim 1.2 \text{ g/cm}^3,$

$\text{Surface area (PS gel)} = \sim 1216768 \text{ nm}^2$  (DLS diameter = 623nm);

$\text{Density (GO)} = \sim 1.9 \text{ g/cm}^3;$

$\text{Thickness (single layer GO)} = \sim 0.6\text{-}1.4 \text{ nm};$

So, mass ratio is dependent on the thickness of GO. On the basis of TEM characterization and the stable dispersion of GO even at high centrifugation speed, we assume that the synthesized GO has few layers (2-6 layers).<sup>6</sup>

The estimated mass ratio is 0.54-3.8. We have used an average value of  $\sim 2.0$  for our paper. Multiple TEM images (Figure S7) show that most of GO sheets wrap around polymer spheres with few free GO sheets left.