

Supplementary information

Bottom-up preparation of MgH₂ nanoparticles with enhanced cycle life stability during electrochemical conversion in Li-ion batteries

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Content of the supplementary information:

- (a) Dark field TEM image and the corresponding SAED electron diffraction pattern of the as-synthesized 15MgH₂@HSAG-500,
- (b) N₂ sorption isotherms at 77 K of all as-synthesized xMgH₂@HSAG-500 composites,
- (c) N₂ sorption isotherms at 77 K of 50MgH₂@HSAG-500 composite before and after ball milling,
- (d) Electrochemical characterizations of all as-synthesized xMgH₂@HSAG-500 composites for the first cycle, where x is 15, 25, 50 and 70 wt.% Mg.
- (e) Comparison between capacities of two ball milled xMgH₂@HSAG-500 composites with x = 50 and 70 wt.% Mg,
- (f) Electrochemical characterization of the pristine HSAG-500 carbon.

a) The dark field TEM image and the corresponding SAED electron diffraction pattern of the as-synthesized $15\text{MgH}_2@\text{HSAG-500}$ composite

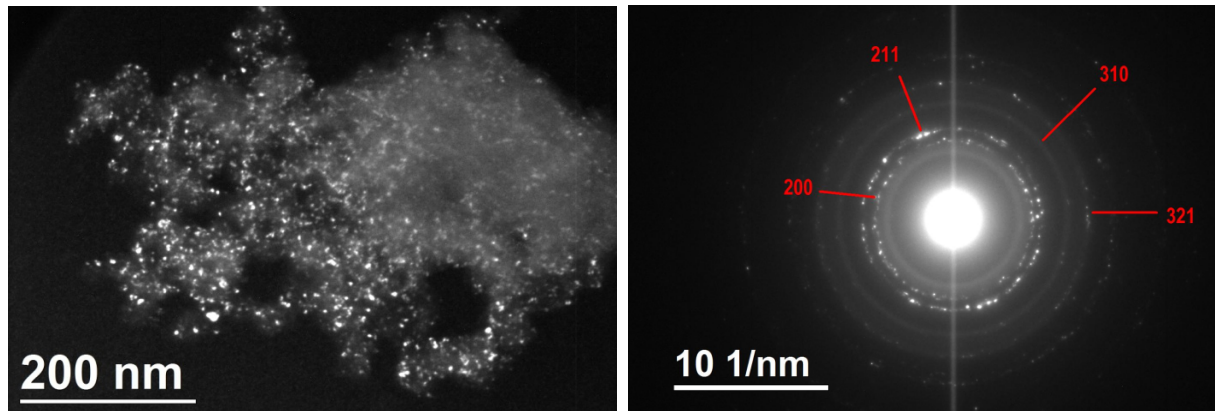


Figure SI-1. The dark field TEM image (left) and the corresponding SAED (right) of the as-synthesized $15\text{MgH}_2@\text{HSAG-500}$ composite.

All the diffraction spots in the SAED pattern are indexed in the tetragonal structure of the MgH_2 phase (JCP: 01-074-0934). Attempts have been done to index in the following structures: the cubic MgO (JCP: 3-0998) and the hexagonal Mg (JCP: 4-0770). Neither the cubic MgO nor the hexagonal Mg structures fits the diffraction spots observed presently. Therefore, neither decomposition nor oxidation occurs during TEM measurements.

b) N_2 sorption isotherms at 77 K of all as-synthesized $xMgH_2@HSAG-500$ composites, where x is 15, 25, 50 and 70 wt.% Mg

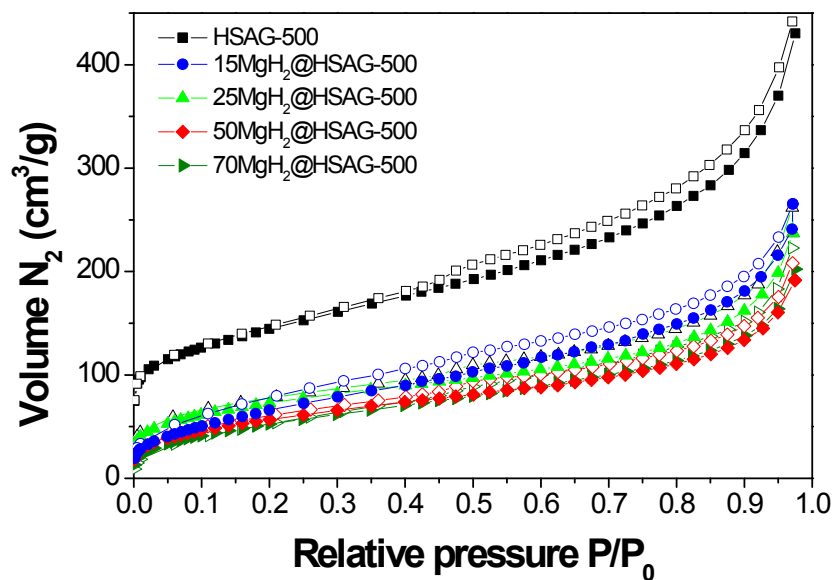


Figure SI-2. N_2 sorption isotherms at 77 K for all $xMgH_2@HSAG-500$ composites. Full and empty symbols stand for adsorption and desorption, respectively.

c) N_2 sorption isotherms at 77 K and pore size distribution of $50MgH_2@HSAG-500$ composite before and after ball milling

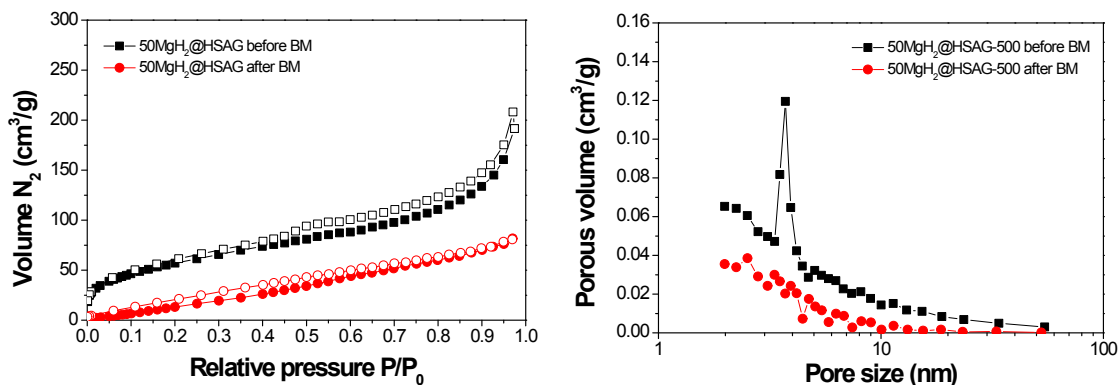


Figure SI-3. N_2 sorption isotherms at 77 K (left) and pore size distribution of $50MgH_2@HSAG-500$ composite (right) before and after ball milling. Full and empty symbols stand for adsorption and desorption, respectively.

The ball milling strongly decreases the textural properties of the composite $50MgH_2@HSAG-500$. This composite shows almost negligible microporosity (see also Table 1). The pore size distribution, as determined by Barrett-Joyner-Halenda analysis on the desorption branch, reveals a strong decrease of porosity.

d) Electrochemical characterizations of all as-synthesized $x\text{MgH}_2@\text{HSAG-500}$ composites during the first cycle (x is 15, 25, 50 and 70 wt.% Mg)

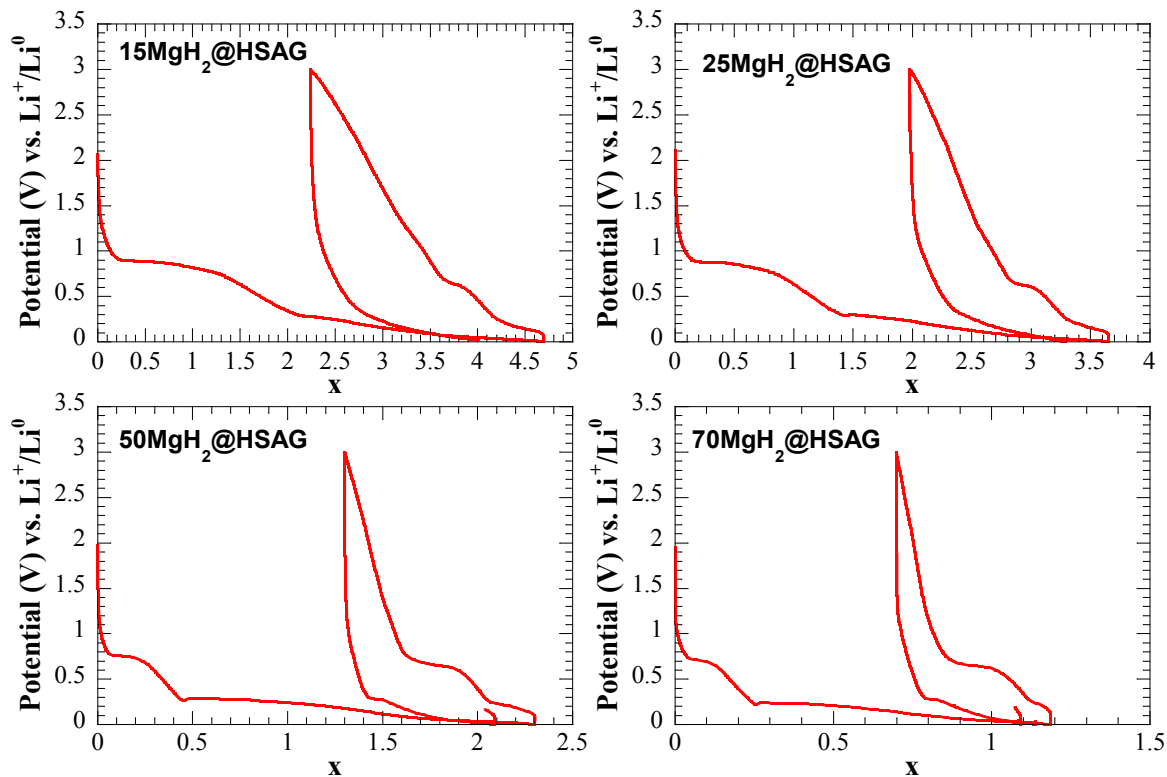


Figure SI-4. Potential/composition profiles of all as-synthesized $x\text{MgH}_2@\text{HSAG-500}$ composites with $x = 15, 25, 50$ and 70 wt.% Mg.

e) Comparison between capacities of two ball milled composites $x\text{MgH}_2@\text{HSAG-500}$ with $x = 50$ and 70 wt.% Mg over 10 cycles.

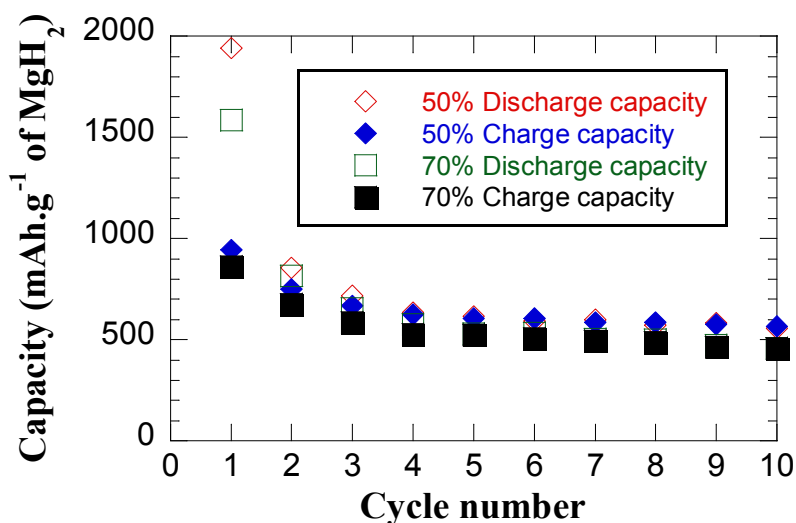


Figure SI-5. The variation of capacity of two ball milled composites $x\text{MgH}_2@\text{HSAG-500}$ with $x = 50$ and 70 wt.% Mg over 10 cycles.

These two composites with different carbon content have been prepared in the same way (bottom-up synthesis followed by ball milling under Ar for 10 h). The overall capacity expressed by gram of MgH_2 is comparable over 10 cycles. This suggests that the carbon contribution is negligible for these composites and supports our hypothesis of the calculation of the overall capacity relative to solely MgH_2 content.

f) Electrochemical characterization of pristine HSAG-500 carbon

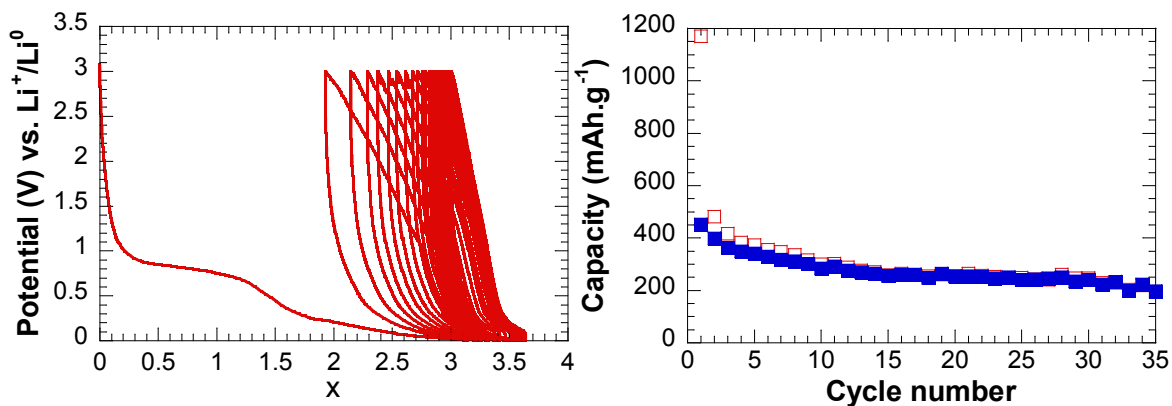


Figure SI-6. Potential/composition profiles of the pristine HSAG-500 carbon during 35 discharge/charge cycles.

The pristine HSAG graphite shows a rapid fading of the electrochemical capacity from approximately 400 mAh.g⁻¹ in the first cycle to almost stable reversible capacity of 200 mAh.g⁻¹ after 35 cycles.