Supplementary information

Bottom-up preparation of MgH$_2$ 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 15MgH$_2$@HSAG-500,
- (b) N$_2$ sorption isotherms at 77 K of all as-synthesized $x$MgH$_2$@HSAG-500 composites,
- (c) N$_2$ sorption isotherms at 77 K of 50MgH$_2$@HSAG-500 composite before and after ball milling,
- (d) Electrochemical characterizations of all as-synthesized $x$MgH$_2$@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 $x$MgH$_2$@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 15MgH$_2$@HSAG-500 composite

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

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\textsubscript{2} sorption isotherms at 77 K of all as-synthesized \(x\text{MgH}_2@\text{HSAG-500}\) composites, where \(x\) is 15, 25, 50 and 70 wt.% Mg

Figure SI-2. N\textsubscript{2} sorption isotherms at 77 K for all \(x\text{MgH}_2@\text{HSAG-500}\) 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.