# Supplementary Information

## Remarkable Hydrogen Desorption Properties and Mechanisms for $Mg_2FeH_6$@$MgH_2$ Core-Shell Nanostructure

Xuezhang Xiao, Chenchen Xu, Jie Shao, Liuting Zhang, Teng Qin, Shouquan Li, Hongwei Ge, Qidong Wang and Lixin Chen*

State Key Laboratory of Silicon Materials, Key Laboratory of Advanced Materials and Applications for Batteries of Zhejiang Province, School of Materials Science and Engineering, Zhejiang University, Hangzhou 310027, (P. R. China)

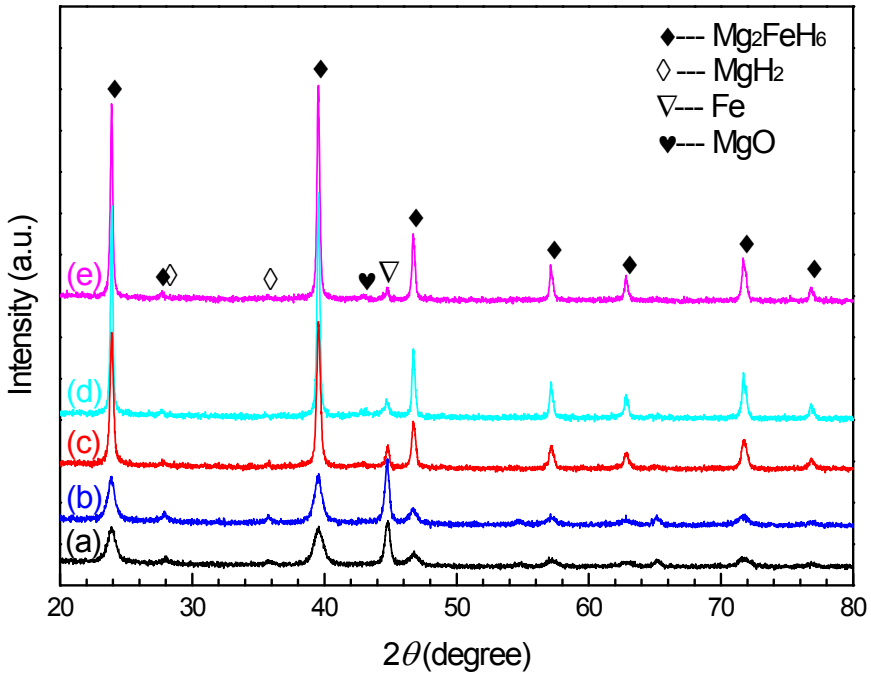


**Fig. S1** XRD patterns of the RBM 2.2~2.8Mg/Fe samples heat treated at different temperatures and $H_2$ pressures: (a) 2.2Mg/Fe, 350 ºC, 90 bar; (b) 2.2Mg/Fe, 350 ºC, 120 bar; (c) 2.2Mg/Fe, 450 ºC, 90 bar; (d) 2.2Mg/Fe, 500 ºC, 90 bar; (e) 2.8Mg/Fe, 500 ºC, 90 bar.

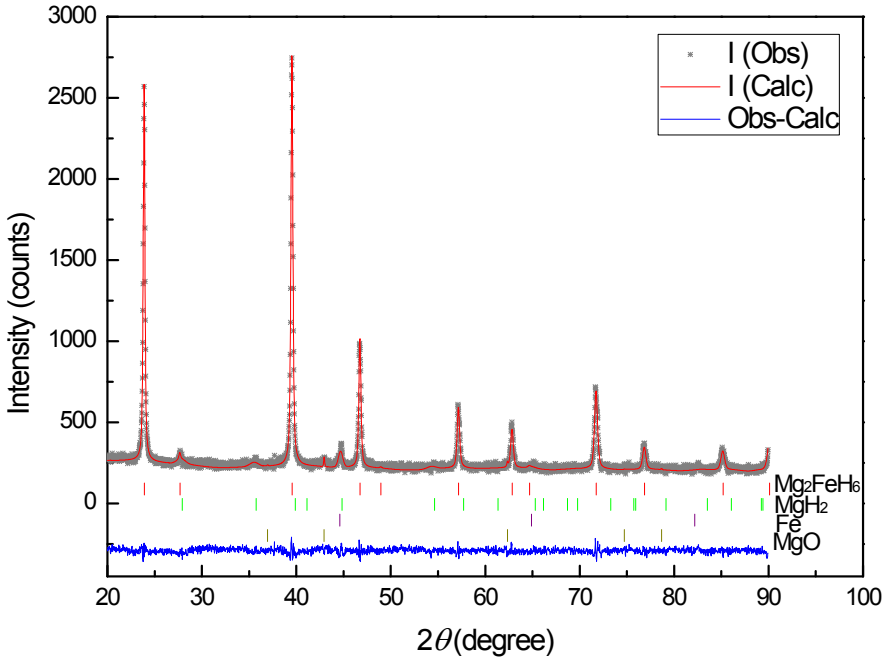


**Fig. S2** Rietveld analysis of the 2.2Mg/Fe RBM sample HTM at 500 ºC and under 90 bar $H_2$. Observed (dots), calculated (top line) and difference curves (bottom line).

The Rietveld analysis reveals that the sample is composed of the $Mg_2FeH_6$ (*Fm*-3*m*), $MgH_2$ (*P*$4_2$/*mnm*), Fe (*Im*-3*m*) and MgO (*Fm*-3*m*). The phase abundances of the $Mg_2FeH_6$, $MgH_2$, Fe and MgO are 91.4, 4.2, 2.4 and 21.1 mass%, respectively.

**Table S1** Common solid-state rate expressions for different reaction models

| Symbol | Model | Integral *f*(α) form |
|---|---|---|
| D1 | 1-D diffusion | $\alpha^2$=kt |
| D2 | 2-D diffusion | (1-α)ln(1-α)+ α=kt |
| D3 | 3-D diffusion(Jander equation) | $[1-(1-\alpha)^{1/3}]^2$=kt |
| D4 | 3-D diffusion (Ginstling-Braunshteinn equation) | $(1-2\alpha/3)-(1-\alpha)^{2/3}$=kt |
| F1 | First-order reaction | ln(1-α)=-kt |
| R2 | Contractiong area | $1-(1-\alpha)^{1/2}$=kt |
| R3 | Contracting volume | $1-(1-\alpha)^{1/3}$=kt |
| A2 | Avarami-Erofe'ev | $[-\ln(1-\alpha)]^{1/2}$=-kt |
| A3 | Avarami-Erofe'ev | $[-\ln(1-\alpha)]^{1/3}$=-kt |

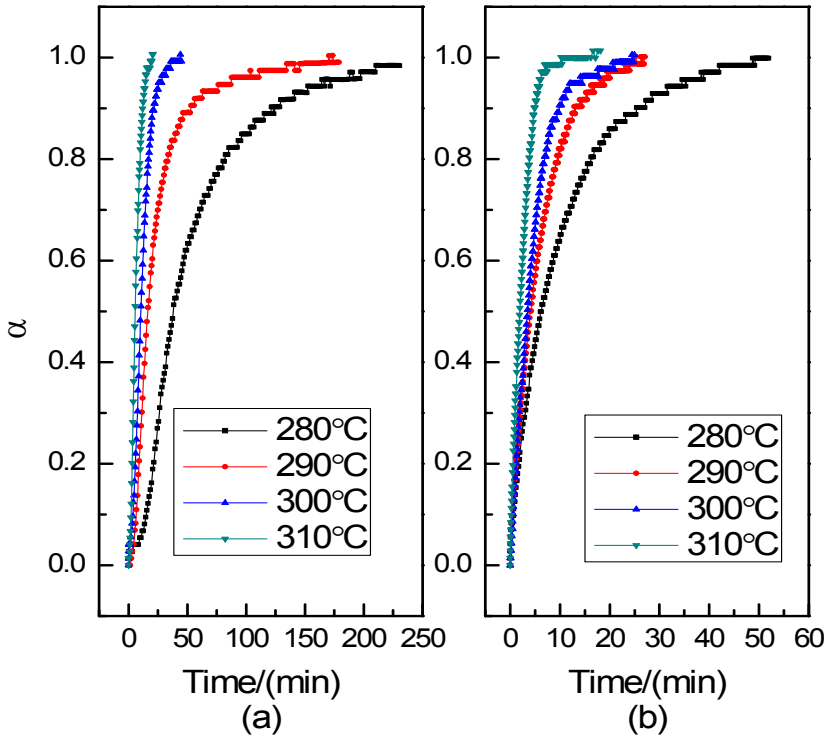


**Fig. S3** Reaction friction against the time: (a) $Mg_2FeH_6/MgH_2$ MP, (b) $Mg_2FeH_6$@$MgH_2$ CSNP.

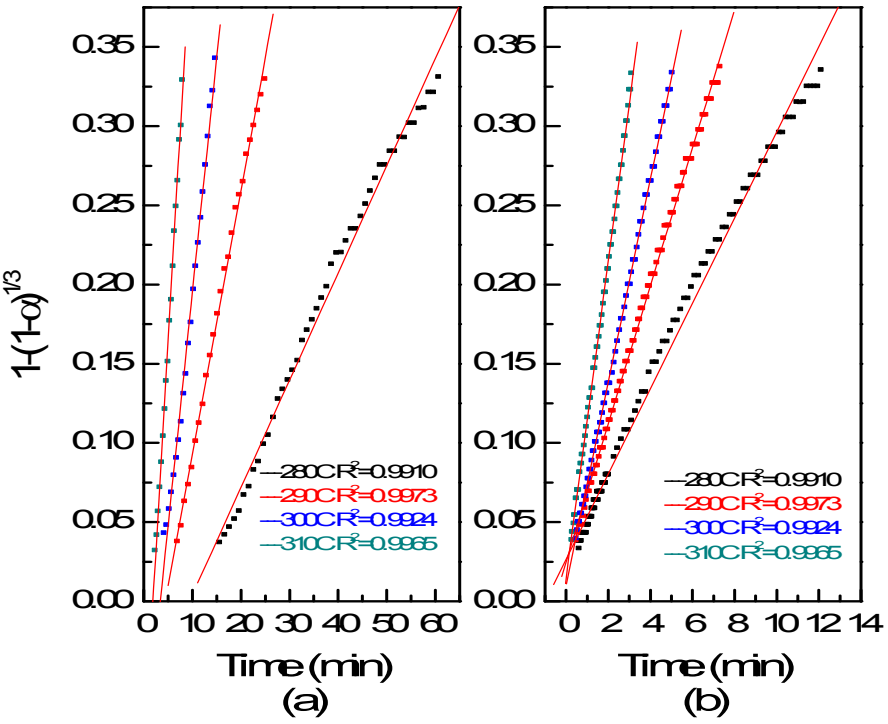


**Fig. S4** Time dependence of R3 equation for hydrogen decomposition at different temperatures ($0.1<\alpha<0.7$):
(a) $Mg_2FeH_6/MgH_2$ MP, (b) $Mg_2FeH_6$@$MgH_2$ CSNP.

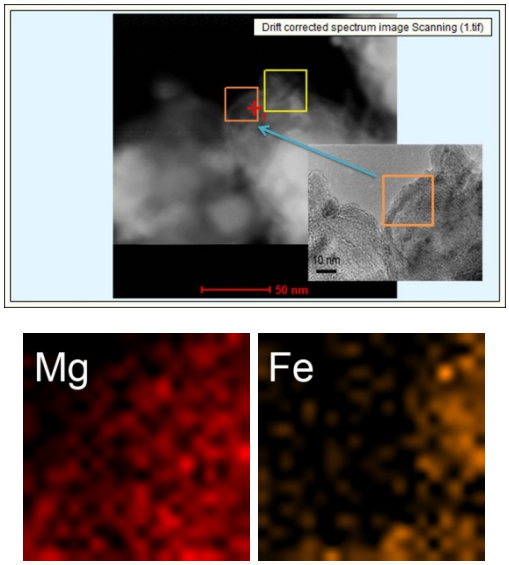


**Fig. S5** STEM-HADDF images and the corresponding element mapping of the rehydrogenated $Mg_2FeH_6@MgH_2$ sample

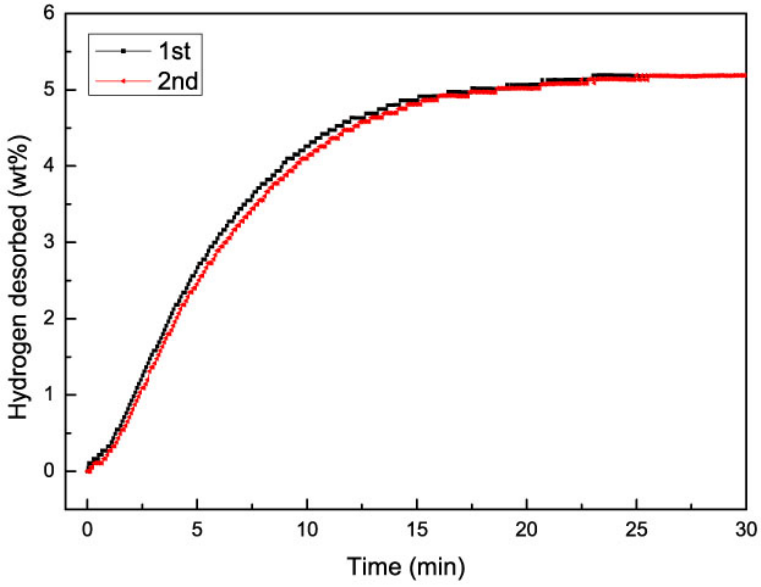


**Fig. S6** Isothermal hydrogen desorption curves of $Mg_2FeH_6@MgH_2$ CSNP at 300 ºC under 0.01bar in the first and second cycles.

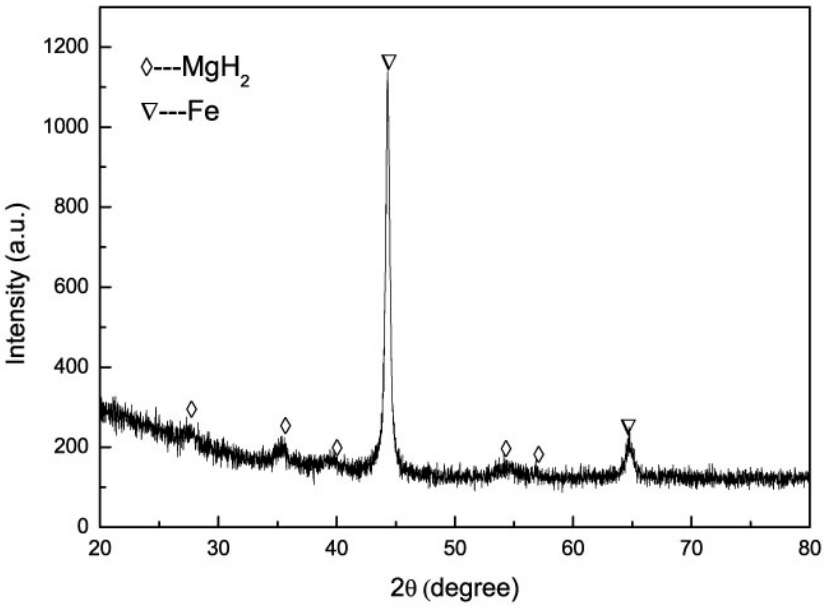


**Fig. S7** XRD patterns of the 2.2Mg/Fe sample ball-milled for 20h.

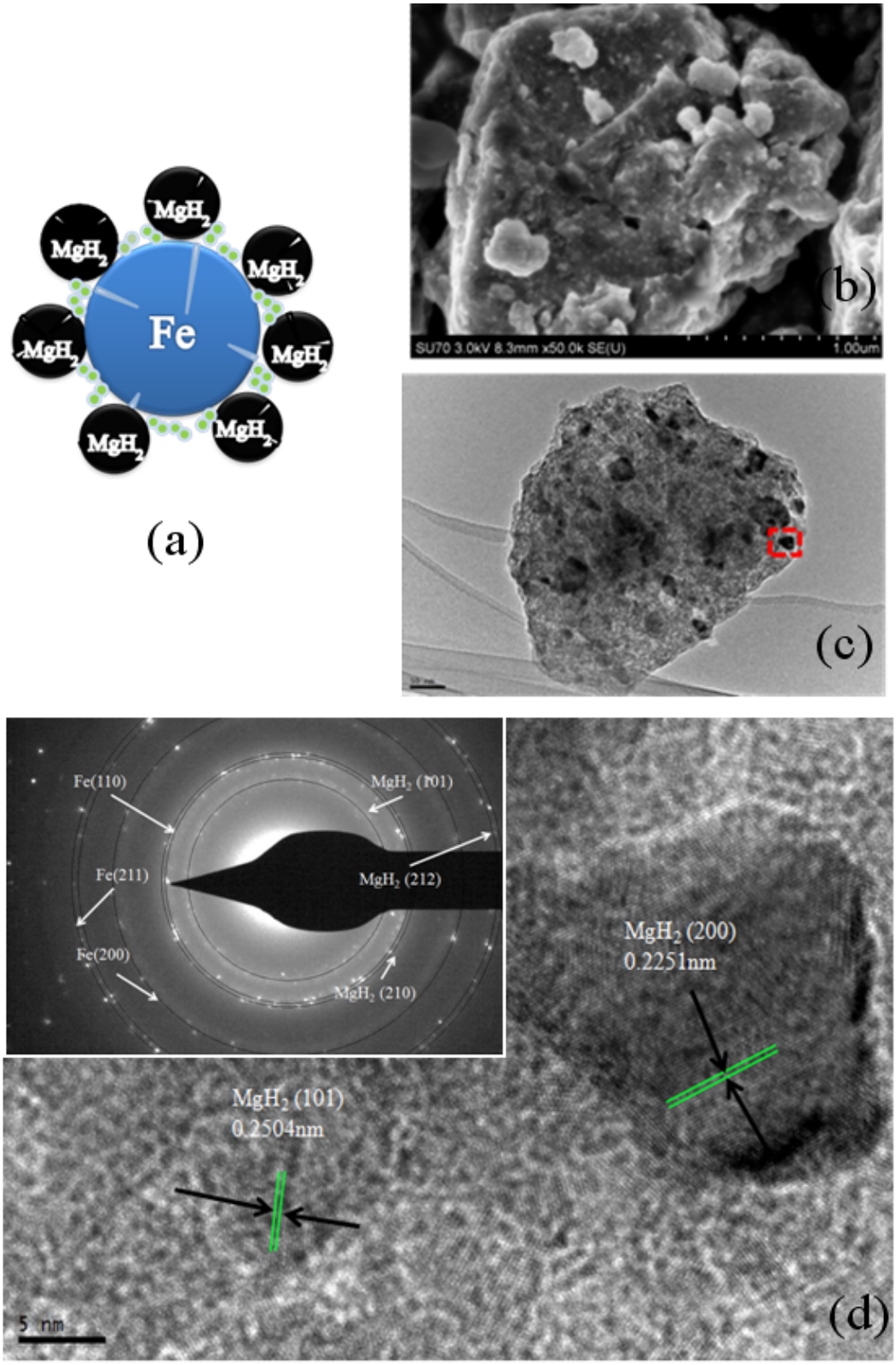


**Fig. S8** images of the 2.2Mg/Fe sample ball-milled for 20h:
(a) one process in Scheme 1;(b) SEM image;(c) TEM image;(d) HRTEM and SAED images of the red rectangle in Fig. S8(c).