Electronic Supplementary Information

Nano Fe and Mg$_2$Ni derived from TMA-TM (TM=Fe, Ni) MOFs as synergetic catalysts for hydrogen storage in MgH$_2$

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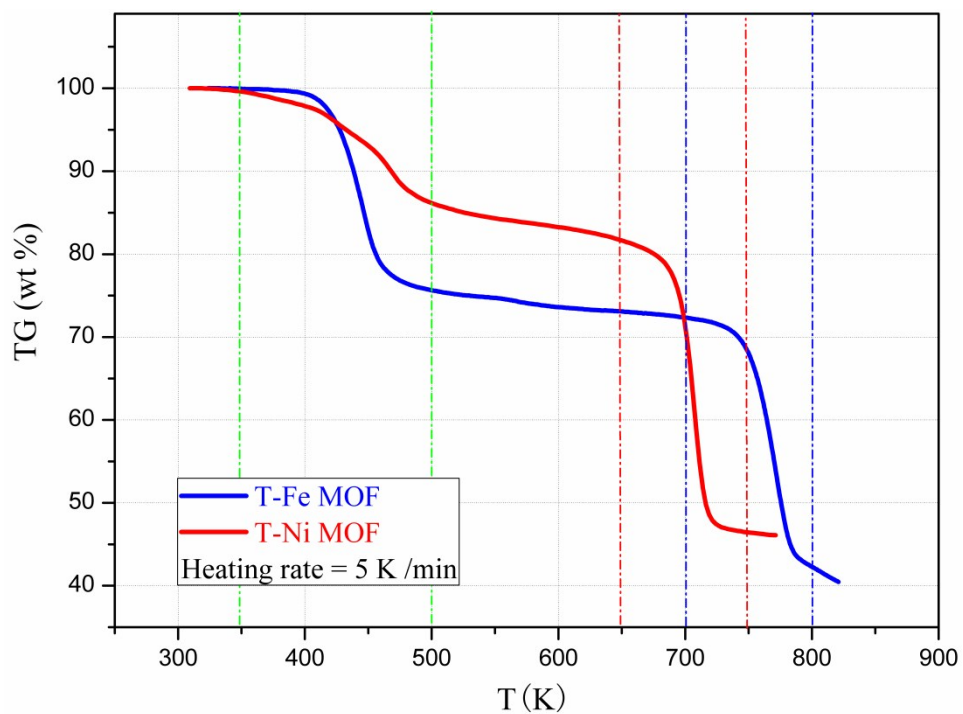
Supplementary Figures:

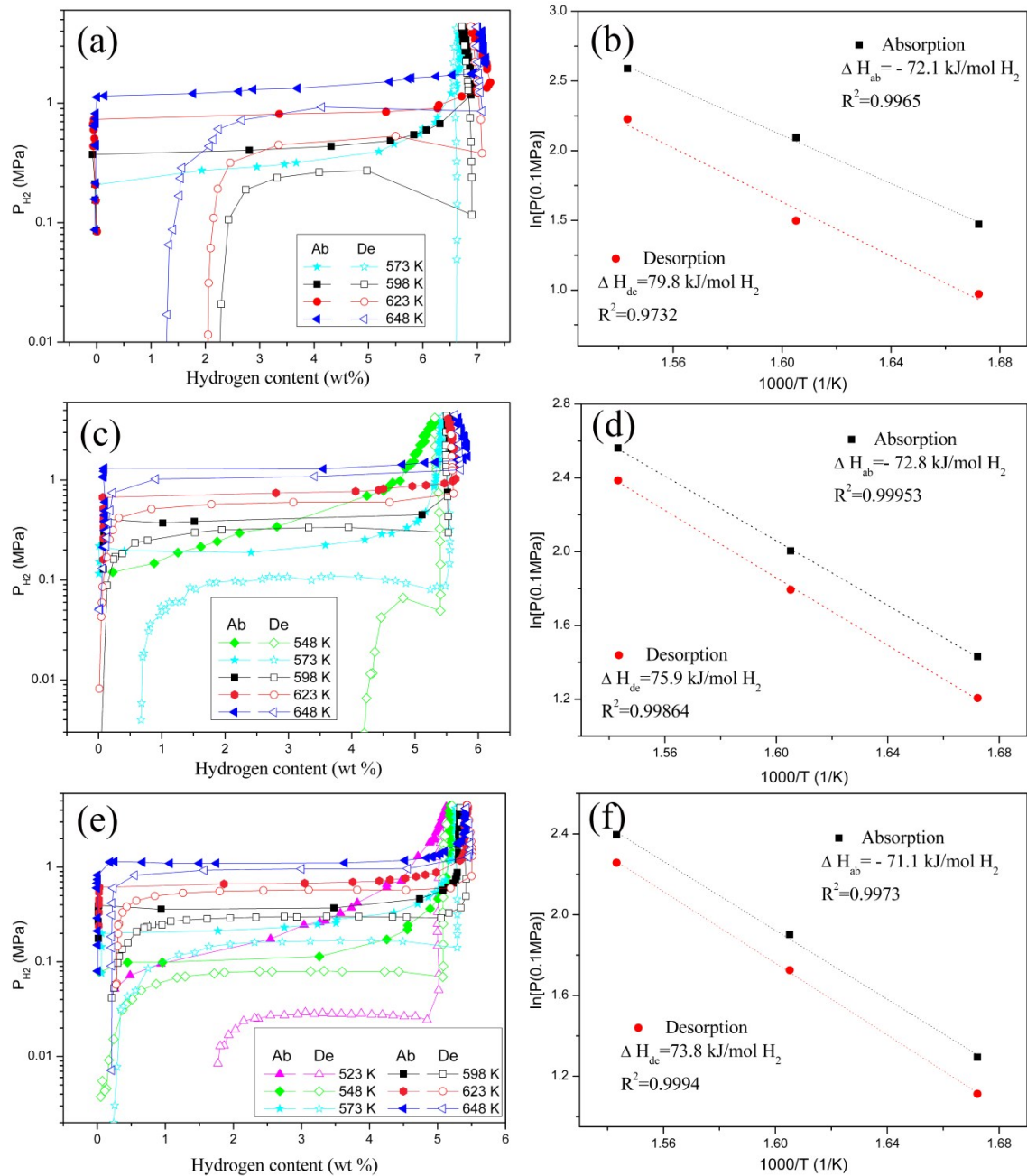**Fig. S1.** Thermogravimetric profiles of the TMA-TM MOFs.

Fig. S2. PC isotherms and the van't Hoff plots of pure MgH_2 (a-b), the MgH_2-Fe MOF (c-d) and the MgH_2-TM MOFs (TM=Fe, Ni) (e-f) composites tested at different temperatures (Ab, absorption; De, desorption).
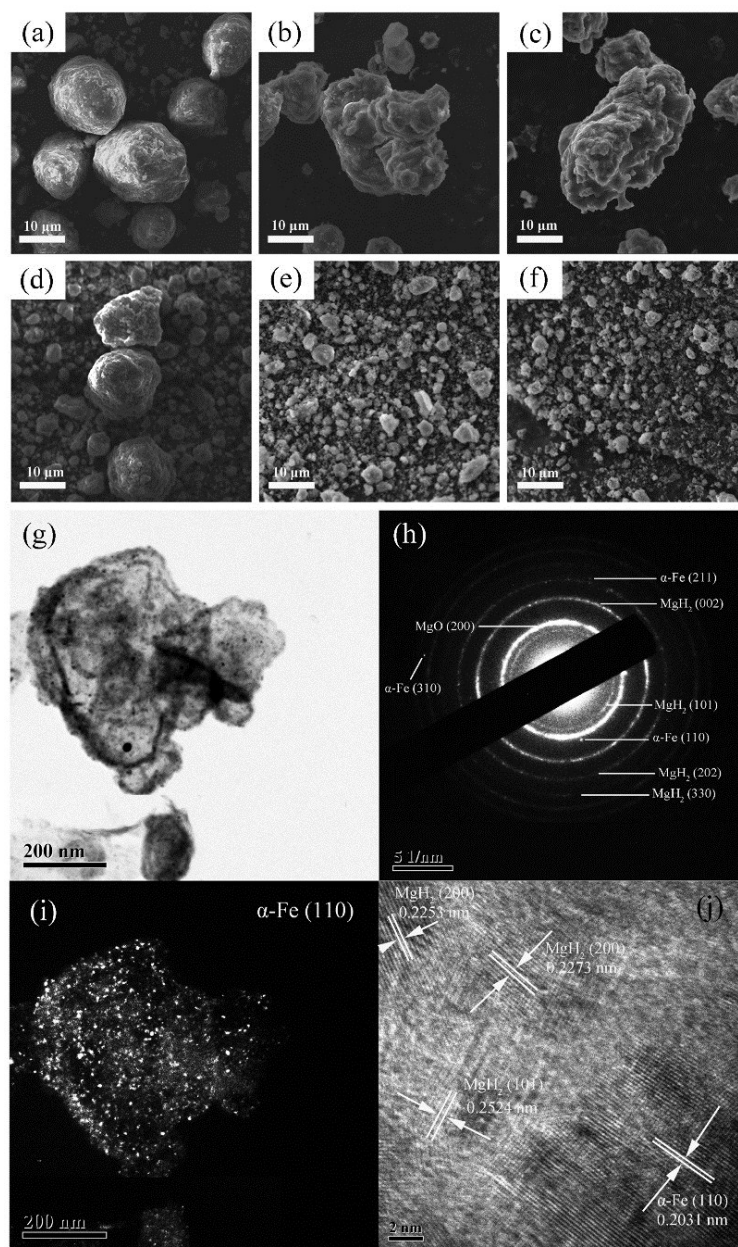
Fig. S3. SEM images of the as milled, rehydrogenated and dehydrogenated of pure MgH$_2$ (a-c) and MgH$_2$-Fe MOF composites (d-f), typical bright field TEM micrographs (g), the corresponding SAED patterns (h), the dark field micrographs (i) contributed by α-Fe (110) and the HRTEM image (j) of the hydrogenated MgH$_2$-Fe MOF composite sample.
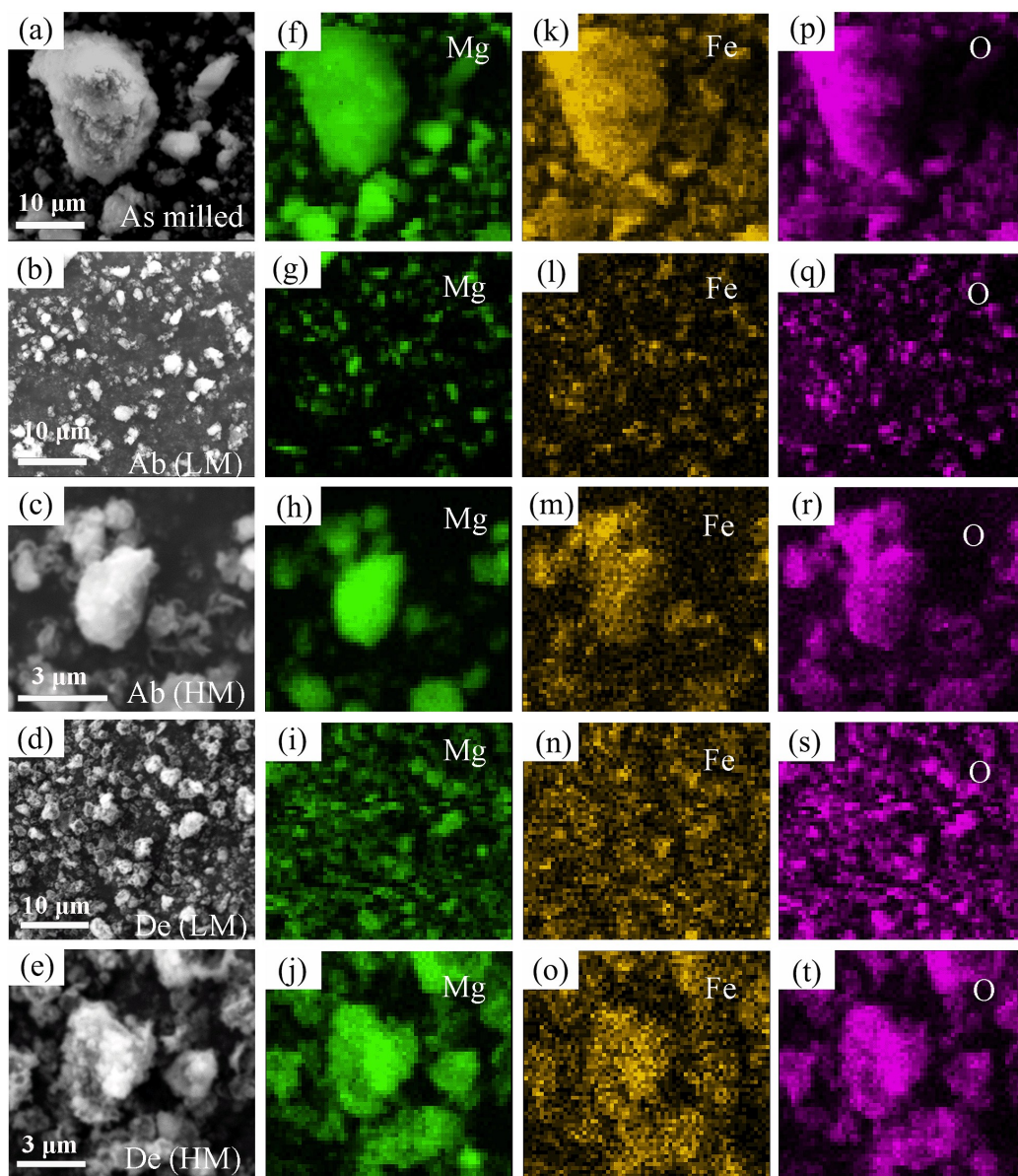
Fig. S4. SEM micrographs of the as milled (a), re-hydrogenated (b, c) and dehydrogenated (d, e) MgH2-Fe MOF composite in low magnification (LM) and high magnification (HM), the corresponding elemental mappings.
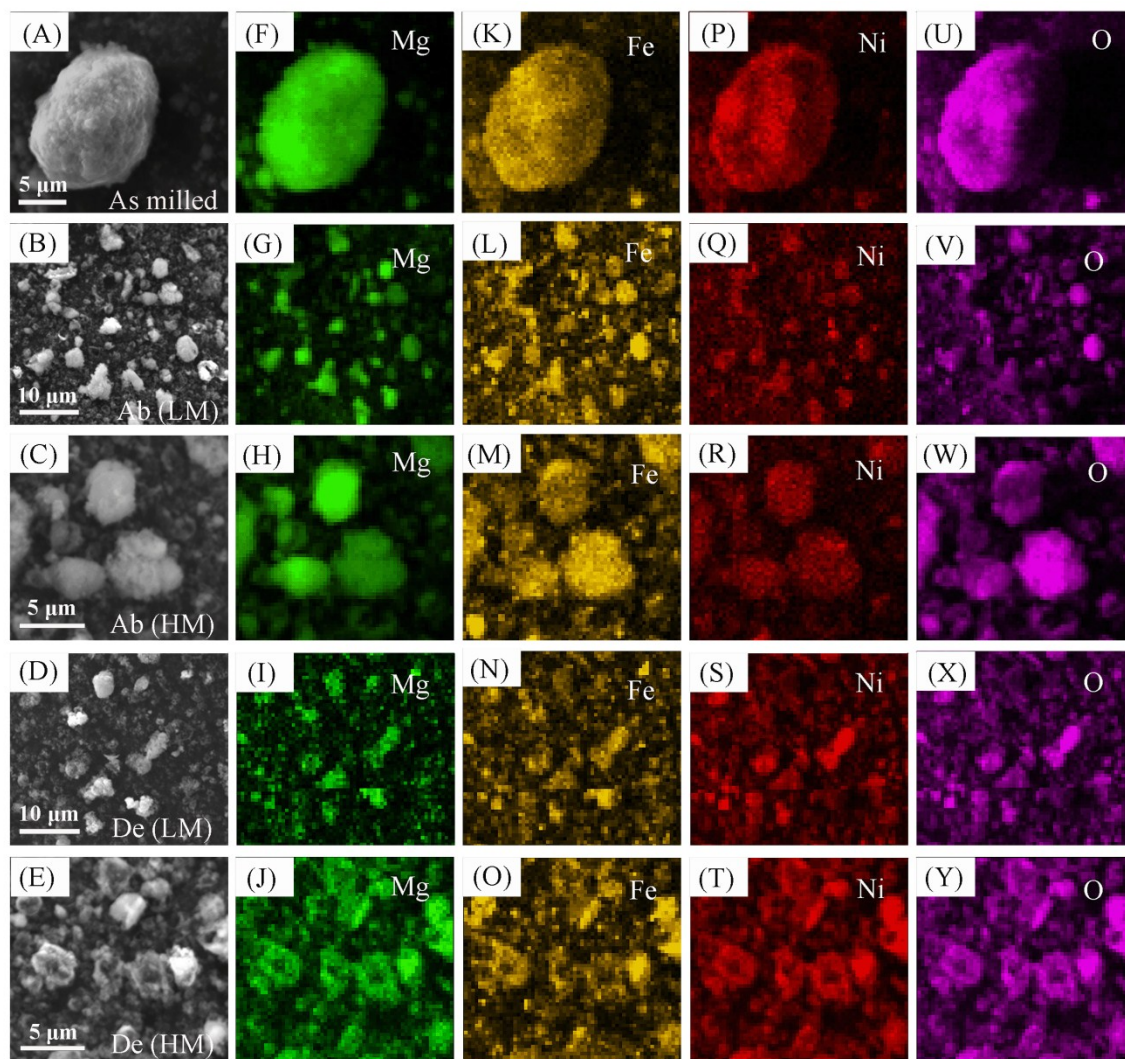
Fig. S5. SEM micrographs of the as milled (A), re-hydrogenated (B, C) and dehydrogenated (D, E) ternary MgH$_2$-TM MOFs (TM=Fe, Ni) composite in low magnification (LM) and high magnification (HM), the corresponding elemental mappings.

Supplementary Table:

Table S1. Pore characteristic parameters of the TMA-TM MOFs (TM=Fe, Ni).

Samples	BET surface area (m^2/g)	BJH pore size (nm)	Pore volume (cm^3/g)
TMA-Fe MOF	1015.49	3.02	0.5317
TMA-Ni MOF	346.22	7.40	0.1736

Table S2. PCT data of the pure MgH_2, MgH_2-Fe MOF and ternary MgH_2-TM MOFs (TM=Fe, Ni) composite samples obtained at different temperatures.

Samples	Temperature (K)	Maximum H-absorption (wt %)	Reversible H_2 sorption capacity (wt %)	Absorption plateaus (MPa)	Desorption plateaus (MPa)
Pure Mg	648	7.02	5.71	1.33	0.91
	623	6.89	4.85	0.81	0.48
	598	6.73	4.51	0.41	0.26
	573	6.68	—	0.30	—
MgH_2-Fe MOF composite	648	5.64	5.63	1.29	1.09
	623	5.52	5.51	0.74	0.61
	598	5.51	5.50	0.42	0.33
	573	5.44	4.76	0.19	0.11
	548	5.32	1.13	—	—
MgH_2-TM MOFs composite	648	5.42	5.27	1.12	0.96
	623	5.43	5.24	0.68	0.57
	598	5.32	5.19	0.36	0.30
	573	5.26	5.07	0.23	0.17
	548	5.19	5.16	0.11	0.08
	523	5.13	3.35	—	0.03