Supporting Information for

Oxygen Vacancy-Rich 2D TiO_2 Nanosheets: A Bridge Toward High Stability and Rapid Hydrogen Storage Kinetics of Nano-Confined MgH_2

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Supplementary Figures and Tables

Table S1 Summary of the precursors used for the synthesis of MgH_2/TiO_2 heterostructure

Samples	Amount of $MgBu_2$ (mL)	Amount of TiO_2 (mg)
blank MgH_2	1.8	0
$40MgH_2/TiO_2$	0.8	30
$50MgH_2/TiO_2$	1.2	30
$60MgH_2/TiO_2$	1.8	30
$70MgH_2/TiO_2$	2.7	30
$80MgH_2/TiO_2$	4.6	30

Note:

The amounts of MgH_2 in the prepared MgH_2/TiO_2 heterostructures are calculated according to the formula below:

$$MgBu_2 + H_2 \rightarrow MgH_2 + C_4H_{10}$$

For example, 1.8 mL of $MgBu_2$ (1.8 mmol) can be hydrogenated to obtain 46.8 mg of MgH_2. Therefore, the weight percentage of MgH_2 in the MgH_2/TiO_2 heterostructure ($m(TiO_2)$ = 30 mg) is calculated to be about 60 wt.% (46.8 mg/(30+46.8) mg).

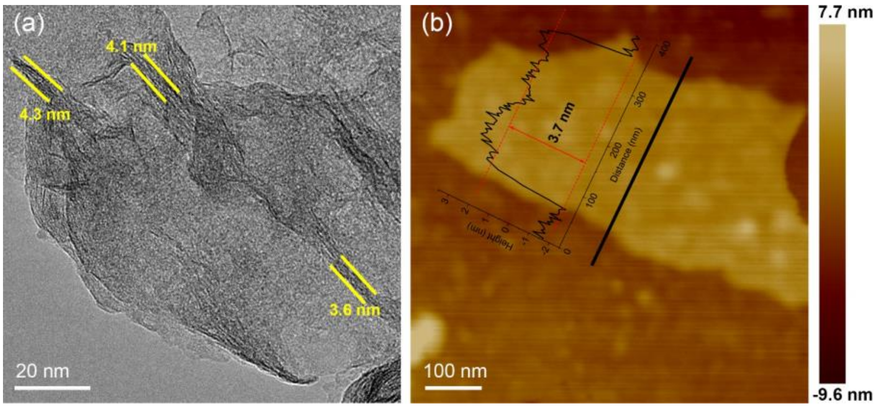

Fig. S1 a Typical TEM image showing the edge configuration of 2D TiO_2 NS. **b** AFM image demonstrating the thicknesses of a 2D TiO_2 nanosheet to be ~3.7 nm

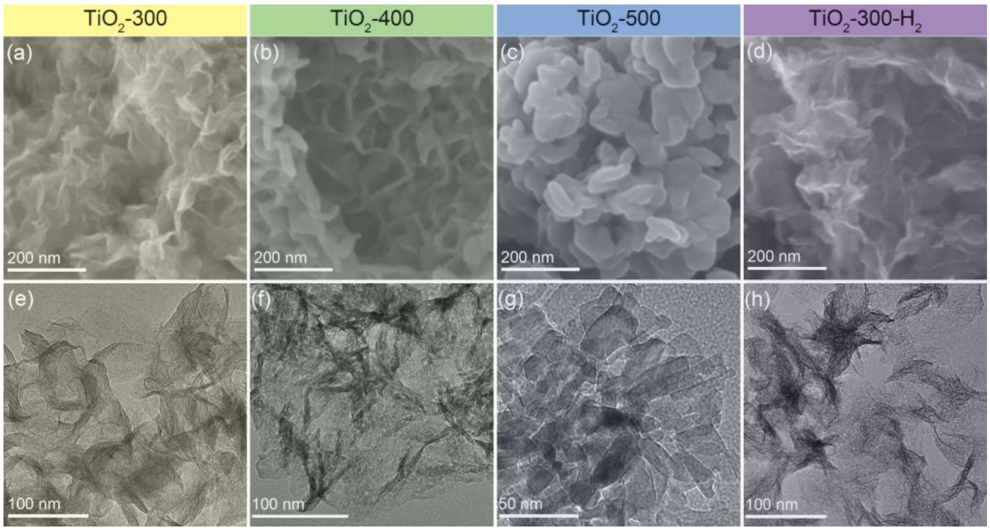

Fig. S2 Typical SEM images **a-d** and TEM images **e-h** of TiO_2 NS annealed at different temperatures

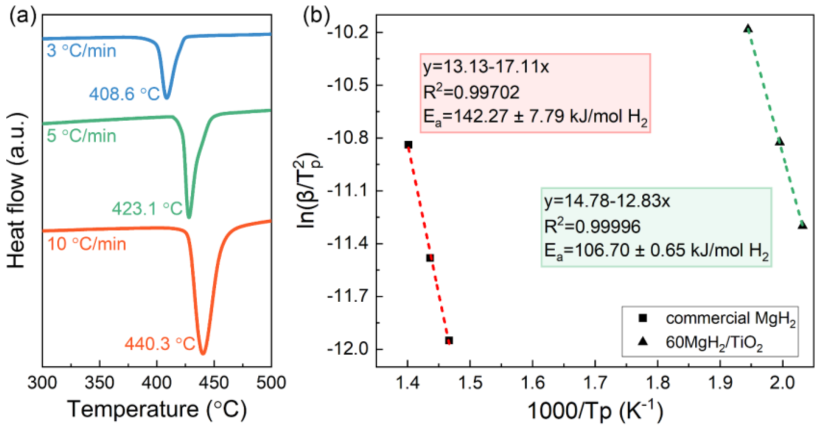

Fig. S3 a DSC curves of the commercial MgH_2. **b** Kissinger's plots of $60MgH_2/TiO_2$ and commercial MgH_2

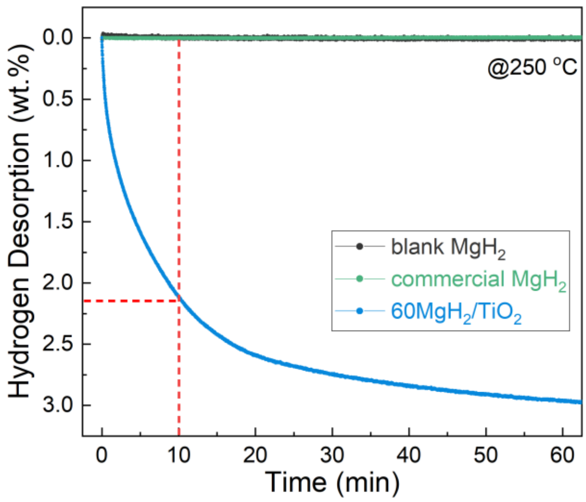

Fig. S4 The comparison of isothermal desorption behaviors of blank MgH_2, commercial MgH_2, and $60MgH_2/TiO_2$

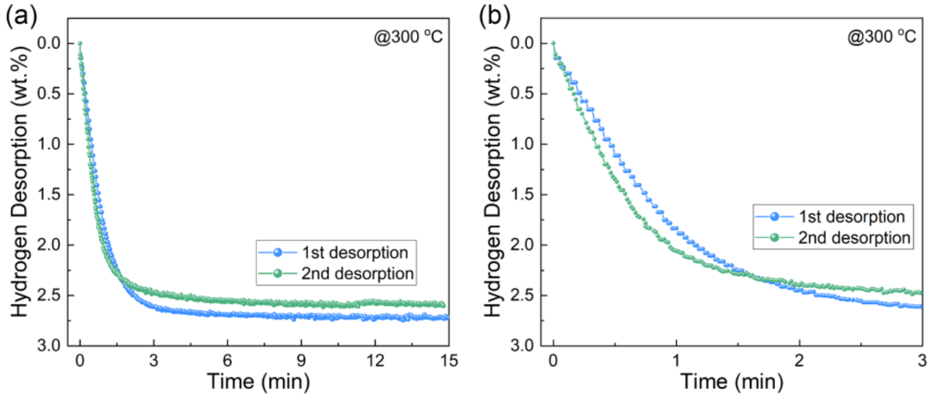

Fig. S5 **a** The comparison of isothermal desorption behaviors of 60MgH_2/TiO_2 in the first two cycles at 300 °C and **b** the corresponding enlarged view for the first three minutes

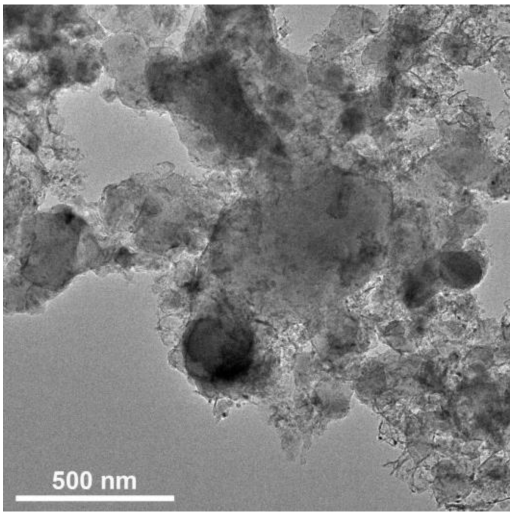

Fig. S6 Typical TEM image of blank MgH_2 synthesized without TiO_2 NS after several de/re-hydrogenation cycles

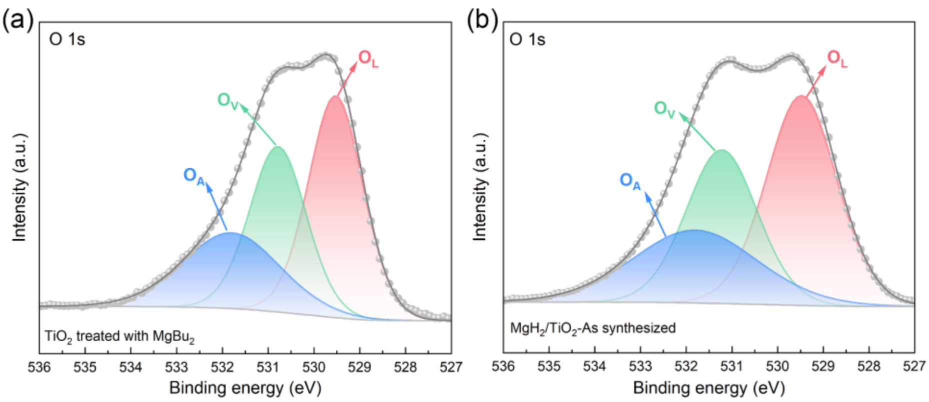

Fig. S7 O 1s XPS spectra of **a** the TiO_2 nanosheets treated with $MgBu_2$ and **b** the as-synthesized MgH_2/TiO_2 heterostructure (O_A: Adsorbed oxygen; O_V: Oxygen vacancy; O_L: Lattice oxygen)

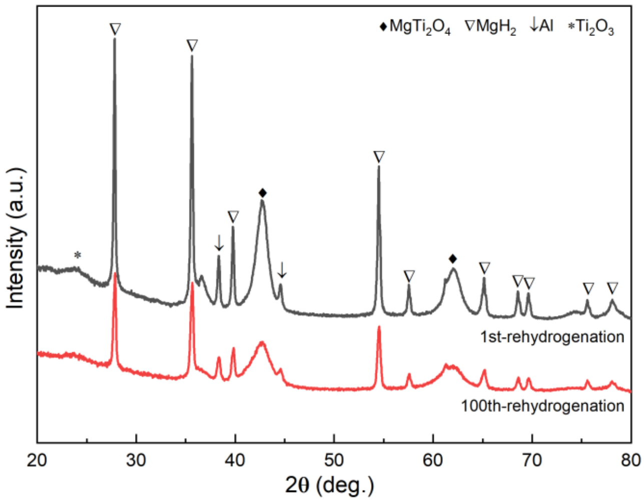

Fig. S8 XRD patterns of the re-hydrogenated MgH_2/TiO_2 heterostructure after 1 de/re-hydrogenation cycle and 100 de/re-hydrogenation cycles

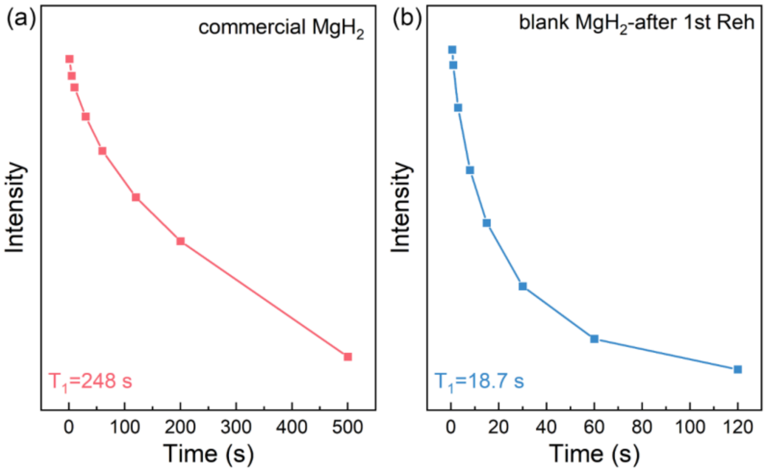

Fig. S9 NMR spin-lattice relaxation curves of **a** commercial MgH_2 and **b** blank MgH_2 after the 1st re-hydrogenation

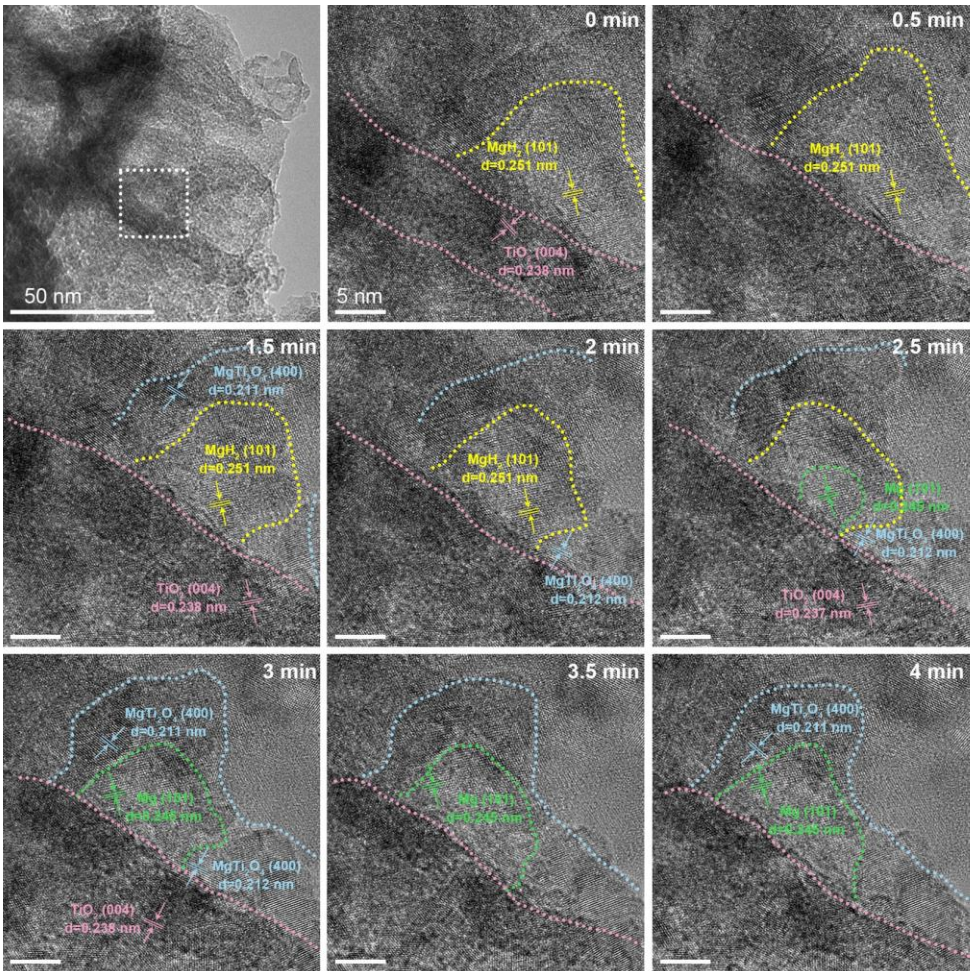

Fig. S10 Typical TEM and HRTEM images of the hydrogenated MgH_2/TiO_2 composites under electron beam radiation during the hydrogen desorption process

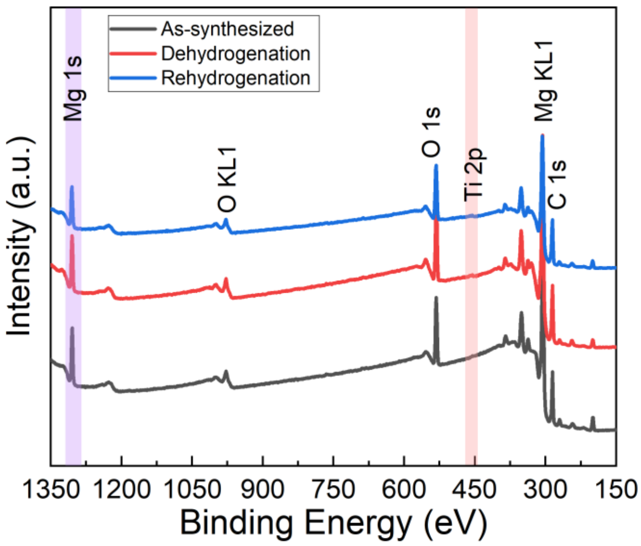

Fig. S11 XPS spectra of the MgH_2/TiO_2 heterostructure at different states

Table S2 Comparison of hydrogen storage performances of MgH_2/TiO_2 heterostructure in the present work with other MgH_2 based systems

Scaffolds /catalysts [a]	Loading capacity (wt.%)	System gravimetric capacity (wt.%)	T_{onset} (°C) (hydrogen desorption)	T_{peak} (°C) (heating rate: 5 °C min^{-1})	Initial dehydrogenation rate at 300 °C (wt% min^{-1})	Refs
Graphene	60	4.5	250	340	0.7	[S1]
CMK-3	40-60	2.4-3.7	250	300-260	None	[S2]
Carbon aerogels	10	1.5 [b]	220	280	None	[S3]
TiO_2@C [a]	90	6.5	205	259 (6 °C min^{-1})	1.12	[S4]
Carbon aerogels	18.2	1.14	200	385 (3.6 °C min^{-1})	None	[S5]
3D TiO_2 [a]	95	6.7	199.2	245.4	None	[S6]
2D TiO_2 (B) [a]	90	6.29	200	227.6	None	[S7]
BCNT	78	5.79	237.5	276.7	0.75	[S8]
TiO_2@Gr [a]	90	6.5	270	290 (10 °C min^{-1})	0.38	[S9]
CoS	60	3.1	297	314	0.255	[S10]
Ni-MOF	50	2.7	314.5	330.4	None	[S11]
TiO_2	60	3.4	180	228.7	2.116	This work

Note:

[a] The synthesis method is high-energy ball milling.

[b] Calculation of the gravimetric capacity does not include the weight of the scaffold.

Supplementary References

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