Supporting information for

Mechanistic Insights into Spontaneous Redispersion of ZnO onto TiO_2 in Water-Containing Environments

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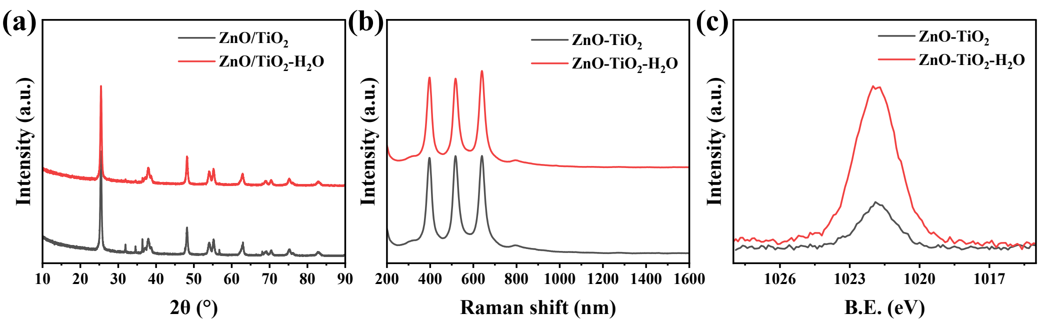

Fig. S1 (a) XRD patterns, (b) Raman spectra and (c) Zn 2*p* XPS spectra of ZnO-TiO_2 sample before and after H_2O vapor treatment.

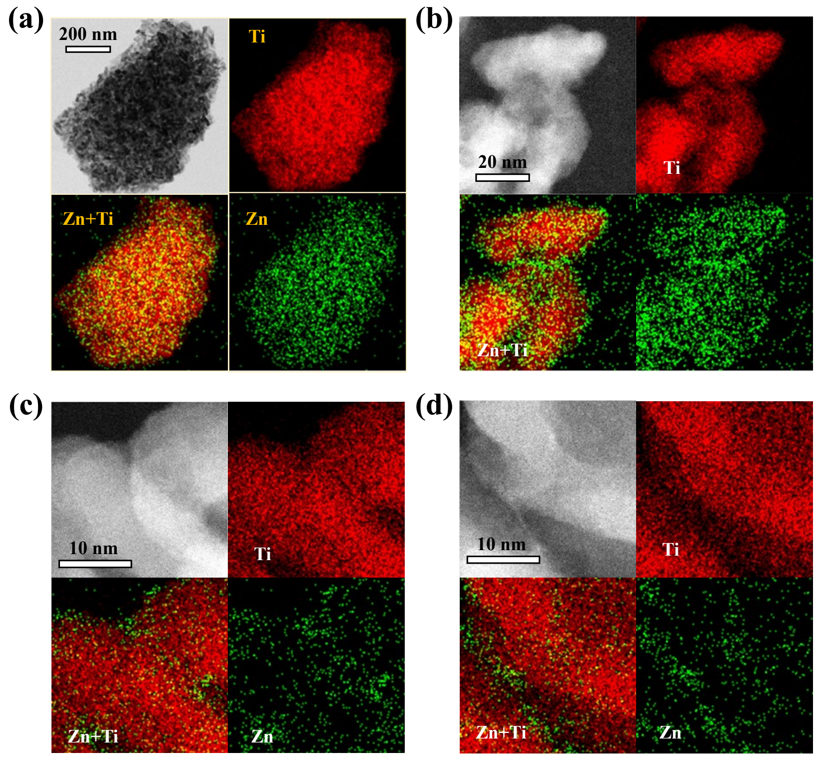

Fig. S2 STEM and EDS mapping images of ZnO-TiO_2-H_2O sample.

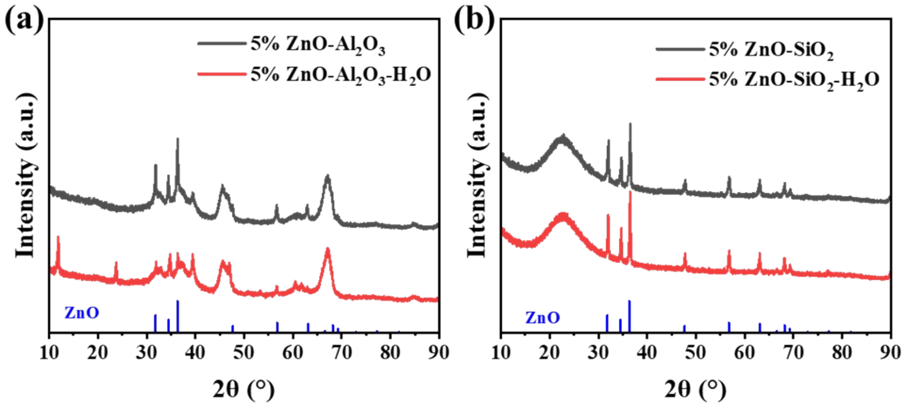

Fig. S3 XRD patterns of (a) 5 wt% ZnO-Al_2O_3 and (b) 5 wt% ZnO-SiO_2 samples before and after liquid water treatment.

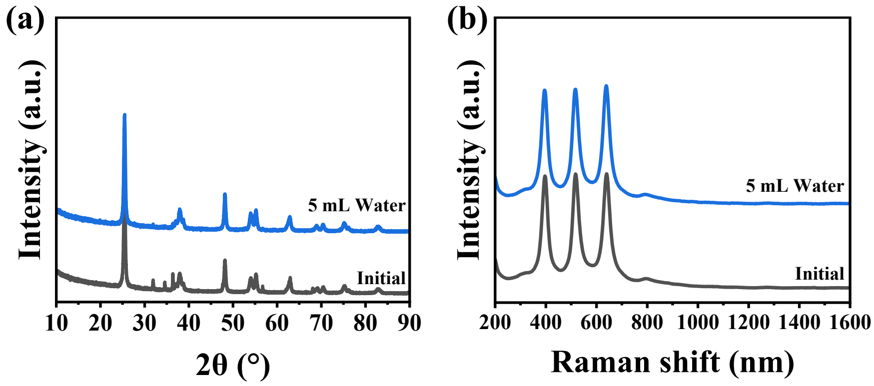

Fig. S4 (a) XRD patterns and (b) Raman spectra of ZnO-TiO_2 sample before and after liquid water treatment.

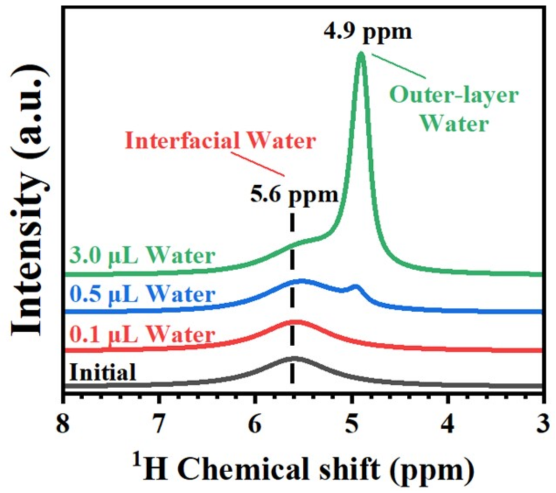

Fig. S5 In situ ^1H NMR spectra of TiO_2 with varying water content.

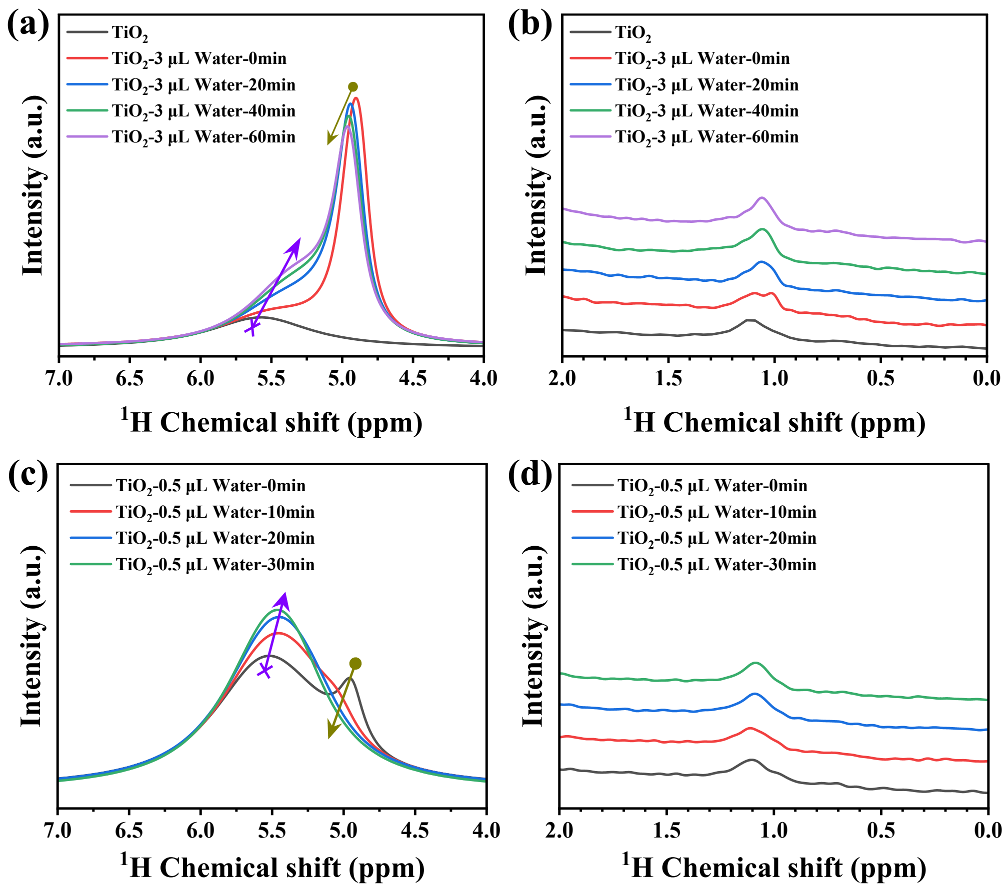

Fig. S6 In situ ^1H NMR spectra of TiO_2 after introducing water. (a-b) In situ ^1H NMR spectra of TiO_2 after introducing 3 μL water without direct contact. (c-d) In situ ^1H NMR spectra of TiO_2 after introducing 0.5 μL water without direct contact.

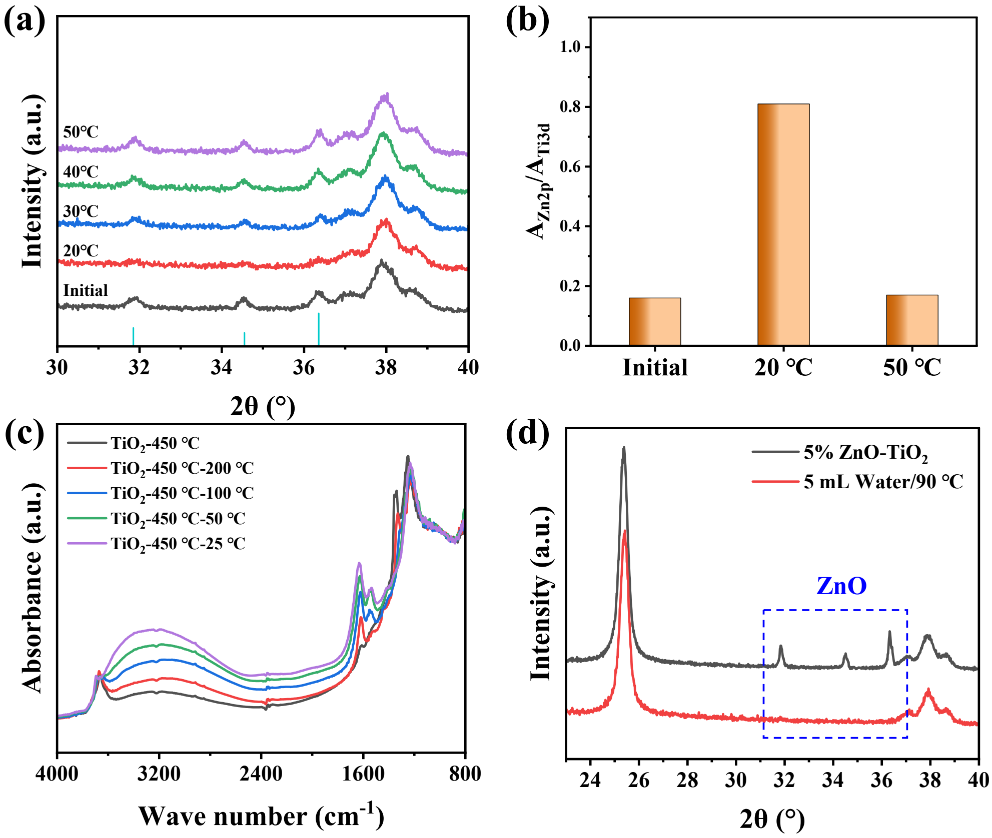

Fig. S7 Effect of temperature on ZnO redispersion. (a) XRD patterns of ZnO-TiO_2 sample after treatment in 3.2% H_2O/O_2 at different temperatures. (b) Zn2*p*/Ti3*d* XPS peak area ratio of ZnO-TiO_2 sample after treatment in 3.2% H_2O/O_2 at different temperatures. (c) DRIFTS spectra of TiO_2 at different temperatures. (d) XRD patterns of ZnO-TiO_2 sample before and after treatment in liquid water at 90 °C.

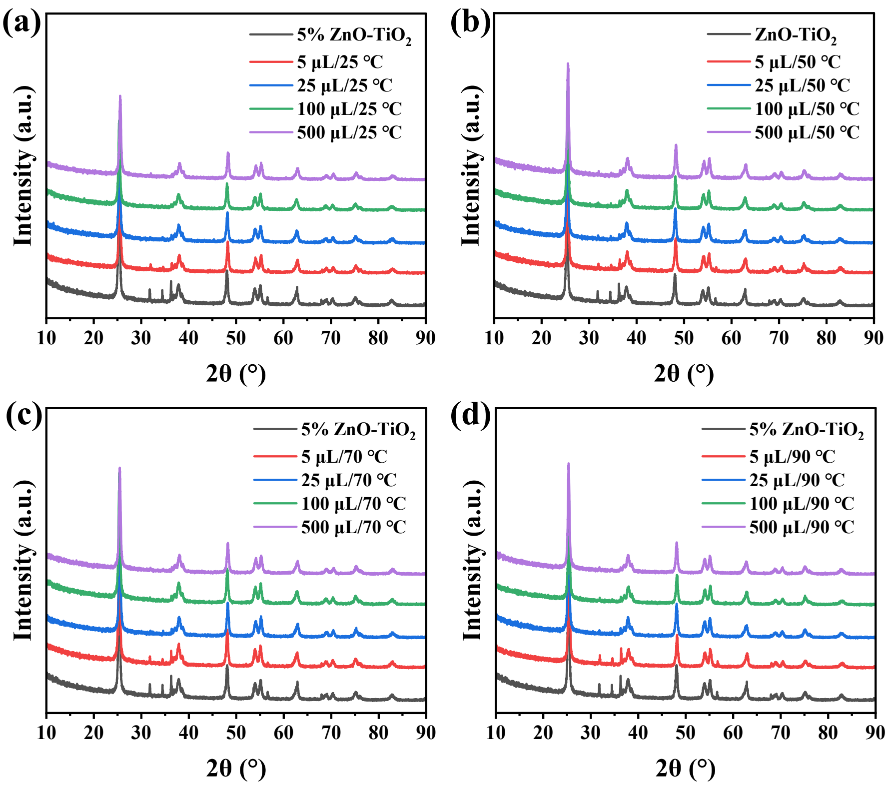

Fig. S8 Effect of temperature and water content on ZnO redispersion. XRD patterns of $ZnO\text{-}TiO_2$ before and after water treatment without contacting at (a) 25 °C, (b) 50 °C, (c) 70 °C, and (d) 90 °C.

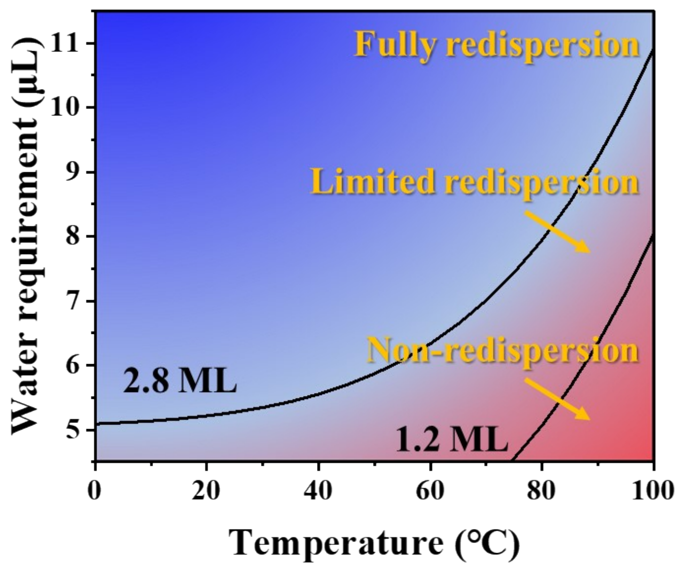

Fig. S9 The water requirement for ZnO redispersion at various temperatures.

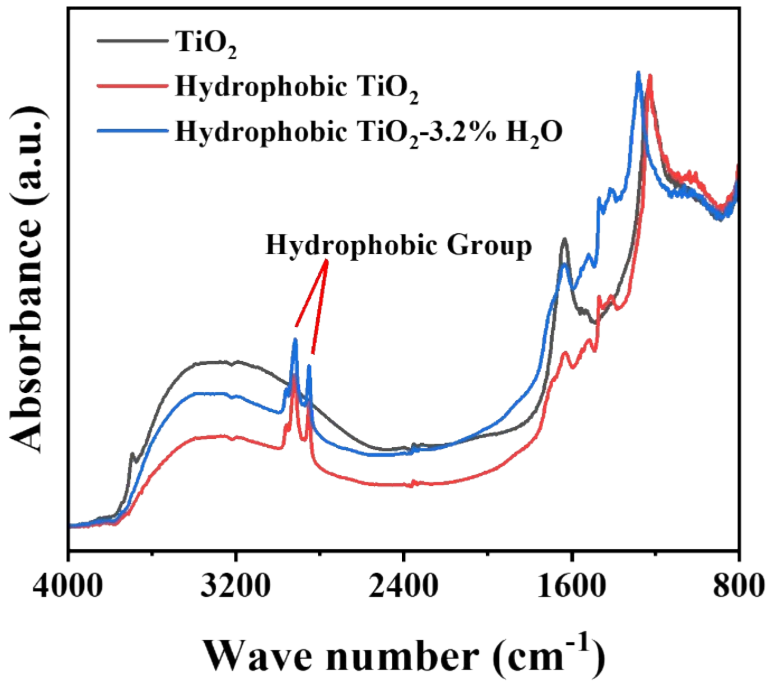

Fig. S10 DRIFTS spectra of TiO_2 and hydrophobic TiO_2 before and after treatment in 3.2% H_2O/O_2.

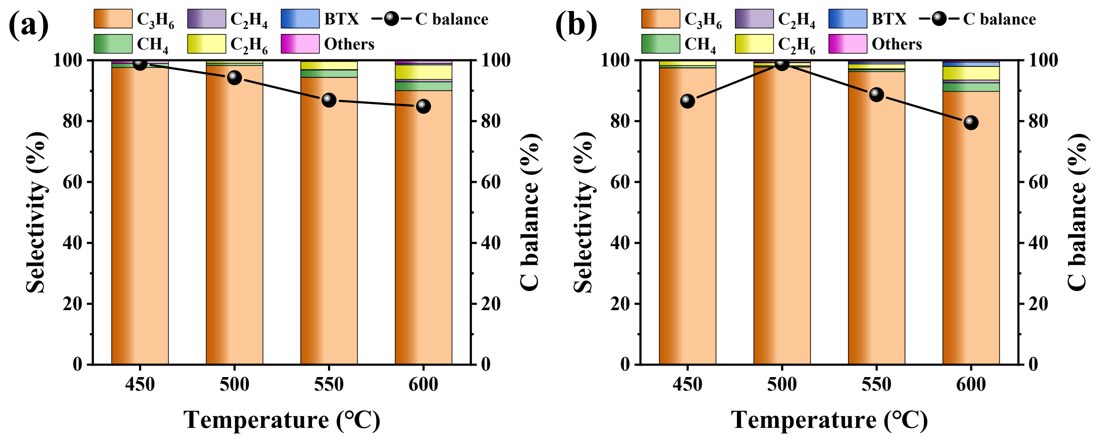

Fig. S11 C_3H_6 selectivity of ZnO-TiO_2 (a) before and (b) after water treatment in PDH.

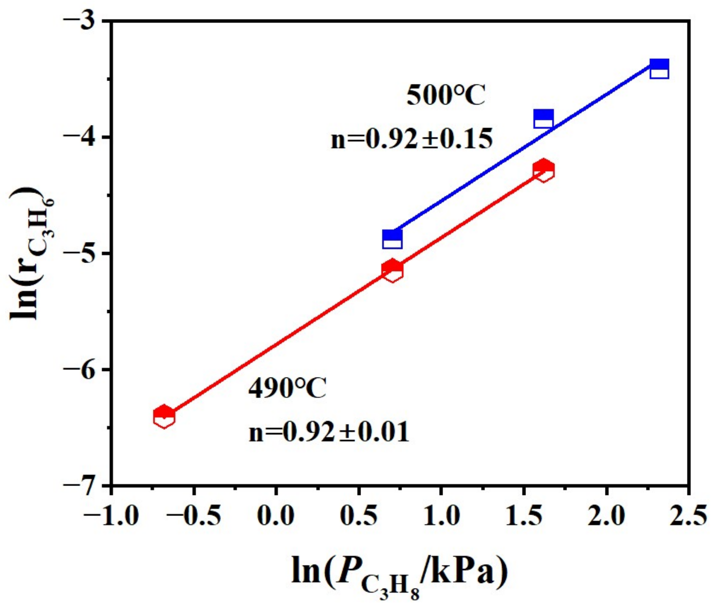

Fig. S12 Kinetic fitting plot of the reaction order for C_3H_8 in propane dehydrogenation reaction.

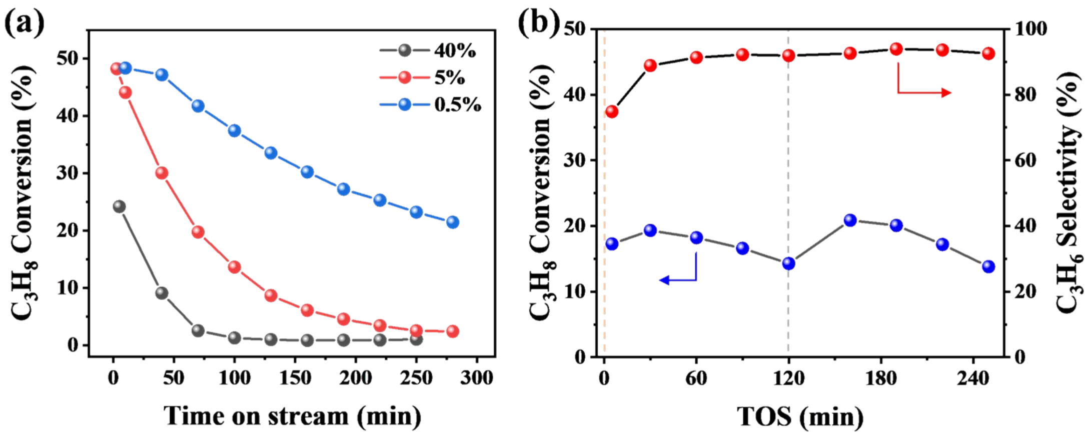

Fig. S13 (a) Stability test of ZnO-TiO_2-H_2O catalysts in the PDH reaction under different C_3H_8 concentrations. (b) Stability test of 5% ZnO/TiO_2 sample prepared by impregnation method in the PDH reaction after pre-oxidation and regeneration (orange dashed line: pre-oxidation process, heating to 550 °C in 40 mL/min O_2; gray dashed line: regeneration process, treatment in 40 mL/min O_2 at 550 °C for 10 min).

Table S1 Quantitative calculation of H_2O coverage according to Fig. 2d.

Processing Conditions	Sample Weight (mg)	Peak Area	Hydrogen Content (mol)	H_2O Coverage (ML)
Dry	88	2.9E+10	4.1E-05	0.2
1.6% H_2O/O_2	95	1.8E+11	2.5E-04	1.2
2.3% H_2O/O_2	90	2.4E+11	3.3E-04	1.7
3.2% H_2O/O_2	97	4.2E+11	5.8E-04	2.8
7.4% H_2O/O_2	93	4.6E+11	6.4E-04	3.2

Polydimethylsiloxane (PDMS) was added to the test sample as an internal standard due to its defined structure, which facilitates the calculation of hydrogen atom amount based on mass. By comparing the ^{1}H peak area of PDMS at 0.16 ppm and ^{1}H peak area of adsorbed water in the range of 5.1 to 5.7 ppm, the hydrogen atom amount in the surface adsorbed water can be determined. Subsequently, the coverage of the surface adsorbed water is calculated using the specific surface area and weight of TiO_2 sample.

Table S2 Calculation of water consumption by evaporation in 10 mL bottle at different temperatures.

Temperature (°C)	Saturated vapor pressure of water (kPa)	Evaporated water volume (μL)
25	3.2	0.2
50	12.3	0.8
70	31.2	2.0
90	70.1	4.2

Table S3 Comparison of PDH performance of Zn-based catalysts in this work and other works.

Temperature (°C)	Catalyst	Reaction Gas	WHSV ($g_{C3H8}·h^{-1}·g_{cat.}^{-1}$)	X(C_3H_8)	S(C_3H_6)	STY ($g_{C3H6}·h^{-1}·g_{cat.}^{-1}$)	Specific Activity ($mol_{C3H6}·h^{-1}·mol_{Zn}^{-1}$)	Ref.
525	10Zn0.1Pt/HZ	C_3H_8/N_2=5/95	0.239	56.2%	79.0%	0.101	1.58	[59]
550	Zn/ZSM-5(20)-IE(16 h)-(0.17)	C_3H_8/He=5/95	10.8	29.0%	69.0%	2.06	356	[60]
550	Zn/ZSM-5(20)-CVD(DMZ)/RED/OX	C_3H_8/He=5/95	10.8	42.0%	62.0%	2.68	102	[60]
550	Zn/ZSM-5(20)-IWI-(0.70)	C_3H_8/He=5/95	10.8	50.0%	42.0%	2.16	96.0	[60]
550	3c-ZnO-ZSM-5	C_3H_8/Ar=5/95	1.09	54.0%	26.0%	0.146	2.52	[61]
550	2c-ZnO-Y	C_3H_8/Ar=5/95	0.996	15.0%	83.0%	0.118	0.917	[61]
550	ZnO-S-1_3	C_3H_8/N_2=40/60	7.89	31.0%	87.0%	2.03	49.4	[12]
550	ZnO//ZnO-S-1_3	C_3H_8/N_2=40/60	1.44	37.0%	94.7%	0.480	1.73	[12]
550	6ZnO/S-1(0.5Mg)	C_3H_8/N_2=40/60	2.15	29.0%	90.0%	0.483	18.8	[13]
600	Znβ-10	C_3H_8/N_2=5/95	0.359	53.3%	92.9%	0.170	2.64	[62]
600	10%Zn/250HZSM-5	C_3H_8/N_2=5/95	0.538	72.9%	67.0%	0.251	3.91	[63]
600	10%Zn/80HZSM-5	C_3H_8/N_2=5/95	0.538	92.3%	12.0%	0.057	0.886	[63]
600	15Zn0.1Pt/Al_2O_3	$C_3H_8/H_2/N_2$=28/28/44	3.01	35.0%	94.0%	0.772	9.11	[64]
600	5%ZnO/HZSM-5(650)	$C_3H_8/CO_2/N_2$=2.5/5/92.5	0.269	54.3%	54.7%	0.076	2.38	[65]
600	Zn_1Co_1/NC	C_3H_8/H_2/He=5/5/90	1.61	20.0%	92.0%	0.284	83.3	[66]
550	ZnO-TiO_2-H_2O	C_3H_8/Ar/N_2=5/5/90	0.538	48.2%	96.3%	0.211	8.66	This work
550	ZnO-TiO_2-H_2O	C_3H_8/Ar=40/60	4.30	24.2%	97.1%	0.966	39.6	This work