Supporting Information

From photocatalysis to photon-phonon co-driven catalysis for methanol reforming to hydrogen and valuable by-products

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Table S1 Representative thermocatalysts for methanol-involved hydrogen production.

Catalyst	T (°C)/P (atm)	CO selectivity (%)	Conversion (%)	H_2 selectivity (%)	H_2 activity (mmol g^{-1} h^{-1})	S/M ratio	Ref.
$Cu/ZrAl_{0.4}$	270/1	1.23 (300 °C)	96	-	460.1 (220 °C)	1.5	[1]
Ru/TiO_2/monolith	300/1	5.4	98.9	97.8	-	2	[2]
$15Pt/15In_2O_3/CeO_2$	350/1	-	99.9	-	-	1.4	[3]
1Cu1Zr/AZ	400/-	3.2	90.6	71.3	655.1	1.2	[4]
AlCuFe oxide	220/-	0.0	89.86	95.7	-	1.2	[5]
Pt/CeO_2-NR	350/1	6.2	100	75	-	-	[6]
$Cu\text{-}1In/SiO_2$	260/1	0.08	71.2	-	301	1.3	[7]
$Cu/ZnTiO_3$	300/1	0.0	99.9	>90	5.46	2	[8]
0.5Zn-Pt/MOC	160/1	-	65.9	-	106.9	3	[9]
CuZnAl-5Mg	200/1	-	-	-	172.6	1	[10]
CuCe-MC (IMP)	250	15	68	>95	-	3	[11]
PdZn(Al)O/MCM-48-2	230/-	0.6	73.8	99	281.3	1	[12]
CuPd-A	220/1	3	85	-	86.3 (220 °C)	1.5	[13]
$Pd/In_2O_3/CeO_2$-r	250/-	<1	96	-	-	1.4	[14]
$0.1\%Pd/ZnAl_2O_4$	280/-	2	73	-	41 (20 °C)	1.1	[15]
CuZn/MCM-41 (COMCM)	300	<2	88	91	-	3	[16]

Table S2 Selected representative photocatalysts without a cocatalyst for methanol based H_2 production operated at room temperature.

Catalysts	**CH_3OH concentration**	**Light source**	**H_2 activity ($mmol^{-1}$ g^{-1} h^{-1})**	**Ref.**
TiO_2 nanosheets	30%	300 W Xe lamp	0.895	[17]
In_2TiO_5	33.3%	450 W Hg Lamp	0.44	[18]
Cu_2O	76%	UV–Vis 300 W Xe	0.42	[19]
ZnO	76%	UV–Vis 300 W Xe	3.1	[19]
ZnS	30%	Hg (290 nm)	1.67	[20]
C_3N_4	76%	UV–Vis 300 W Xe	1.5	[19]
MgO	100%	1000 W Hg lamp	0.328	[21]
MoS_2	20%	300 W Xe lamp	62	[22]
$SrTiO_3$	100%	300 W Xe lamp	0.164	[23]
UiO-66(NH_2)	25%	Xe-Hg Lamp	0.308	[24]

Table S3 Representative heterojunction or cocatalysts/photocatalyst structure for methanol based H_2 production and the apparent quantum efficiency (AQE).

	Photocatalyst	Cocatalysts	CH_3OH concentration	Light source	H_2 activity ($mmol^{-1}\ g^{-1}\ h^{-1}$)	AQE	Ref.
Heterojunction without cocatalysts	$TiO_2/BiYO_3$	/	20%	300 W Xe lamp	0.159	13.4% (380 nm)	[25]
	Cu_2O/TiO_2	/	30%	full-acr Xe lamp	11.0	15.1% (365 nm)	[26]
	TiO_2–Co_3O_4	/	25%	300W Xe lamp	2.9	/	[27]
	gC_3N_4/TiO_2	/	25%	500 W Xe lamp	0.76	/	[28]
	$NiSe/TiO_2$	/	10%	300 W Xe lamp	1.1	/	[29]
	$TiO_2@SiO_2$	/	25%	300 W Xe arc lamp	0.4	/	[30]
	MoS_2/TiO_2	/	10%	300W Xe-arc lamp	2.1	6.4% (360 nm)	[31]
	MoS_2/CdS	/	76%	300 W Xe-lamp, λ ≥ 420 nm	12.0	5% (450 nm)	[19]
	Anatase/rutile TiO_2nanofiber	/	20%	300 W Xe lamp	6.48	20.9 (365 nm)	[32]
	TiO_2	i-Pt	20%	Xe lamp	19.7	8.6% (375 nm)	[33]
	Ultrathin TiO_2	$_{1\%}$Pt	30%	300 W Xe lamp	17.9	28.5% (350 nm)	[17]
	TiO_2-ZSM	$_{2\%}$Pt	10%	300 W Xe lamp	10.0	12.6% (365 nm)	[34]
	Black TiO_2	Pt	50%	Xe lamp AM 1.5	10.0	/	[35]
	$NYTiO_2$	$_{0.188\%}$Pt	50%	200 W Hg lamp	20.9	/	[36]

Noble metal cocatalysts	TiO_2/CNT	$_{0.5\%}$Pt	50%	200 W Hg lamp	40.6	/	[37]
	Co_3O_4-TiO_2	$_{1\%}$Pt	25%	300 W Hg lamp	5.28	11.3% (365 nm)	[27]
	$CaNbO_6$	Rh	10%	500 W Xe lamp	76.96	65% (300 nm)	[38]
	Na-ZnO	$_{2\%}$Pt	10%	300 W Xe lamp	1.38	13.5 % (350 nm)	[39]
	g-C_3N_4	$_{3\%}$Pt	10%	300 W Xe lamp	0.66	6.8 % (420 nm)	[40]
	g-C_3N_4	Rh	10%	Xe Arc Lamp (visible)	0.015	/	[41]
	CuO	$_{1\%}$Pt	25%	300 W Xe lamp	6.8	/	[42]
	TiO_2	$_{1\%}$Pd	50%	400 W Hg	1.20	/	[43]
	TiO_2/WO_3	Au	36%	300 W Xe-lamp	5.39	/	[44]
	g-C_3N_4	Ag NPs	100%	300 W Xe-lamp, 350−780 nm	0.15	/	[45]
	$SrTiO_3$	$_{2\%}$Ag	50%	UV low pressure Hg lamp	9.26	/	[46]
	TiO_2	$_{1\%}$PtO	30%	300 W Xe lamp	4.4	/	[47]
	$BaTiO_3$	AgCl/Ag	10%	300 W Xe lamp	4.53	/	[48]
	TiO_2	$_{0.75\%}$Cu	67%	300 W Xe lamp	101.7	56% (365nm)	[49]
	TiO_2	CuNWs	25%	UV-LED lamp	5.1	17.2% (365 nm)	[50]
	TiO_2	$_{0.75\%}$Cu	33%	Xe lamp(MAX-302)	16.6	45.5% (340 nm)	[51]
	TiO_2	Cu	50%	Xe lamp(MAX-302)	17.8	16.4% (365 nm)	[52]
	TiO_2	$_{0.5\%}$Ni	10%	UV SB-100P/F Lamp	15.7	22.2% (365 nm)	[53]

Non-noble metal cocatalysts	CdS	$_{0.5\%}Ni$	100%	300 W Xe-lamp, λ ≥ 420 nm	7.7	/	[54]
	TiO_2	$_{0.4\%}Ni_2P$	50%	300 W Xe lamp	9.38	11.6 % (360 nm)	[55]
	CdS	Ni_2P	100%	300 W Xe lamp, λ ≥ 420 nm	0.58	/	[56]
	$Zn_2In_2S_5$	CoP	76%	300W Xe lamp	5.5	/	[57]
	TiO_2	$Ni(OH)_2$	25%	UV-LEDs (3 W, 365 nm)	3.1	12.4% (365 nm)	[58]
	MoS_2	Cd	20%	300 W Xe-lamp, λ ≥ 420 nm	33.2	/	[22]
	TiO_2	$_{3\%}$ Li	20%	300 W Xe lamp	41.8	/	[59]
	CeO_2	Fe	10%	125 W Hg lamp	12.82	9.1 % (420 nm)	[60]
Bimetallic cocatalysts	TiO_2	Rh-Cu	20%	UV-pen ray Hg lamp 365 nm	5.5	/	[61]
	TiO_2	Ag-Au	25%	UV-300 W Hg lamp	0.7	3.3% (420 nm)	[62]
	TiO_2	Ni_1Pd_{10}	50%	400 W mercury arc (Eiko-sha 400)	4.2	41 % (355 nm)	[63]
	TiO_2	PtCu	70%	LED 365nm	476.8	99.2 % (365 nm)	[64]

Table S4 The oxidation products' selectivity on the representative photocatalysts operated at different temperatures.

Catalysts	CH_3OH concentration	Light source	Product	H_2 activity ($mmol\ g^{-1}\ h^{-1}$)	Selectivity	Ref.
0.5%Pt/TiO_2/10 %CNT	40%	Fe-halogen-Hg	HCHO	9.0	31%	[65]
0.5%Pt/$^{P25}TiO_2$	40%	Fe-halogen-Hg	HCHO	7.0	80%	[65]
1%Pt/$^{P25}TiO_2$	40%	Hg lamp	HCHO	7.16	61%	[66]
1%Au/ $^{P25}TiO_2$	40%	Hg lamp	HCHO	5.2	64%	[66]
1%Ag/ $^{P25}TiO_2$	40%	Hg lamp	HCHO	0.81	86%	[66]
PtCu- TiO_2	70%	365 LED lamp	HCHO	476.8	99%	[64]
$^{P25}TiO_2$	40%	Hg lamp	HCHO	0.48	77%	[66]
FP-$_{0.5\%}$Pt	40%	Hg arc lamp	HCHO	17.38	--	[19]
ZnO	76%	300W Xe lamp	HCHO	3.0	91%	[19]

ZnS	76%	300W Xe lamp	HCHO	2.2	43%	[19]
MoS_2 foam/CdS	76%	300W Xe lamp	HCHO	2.5	10%	[19]
g-C_3N_4	76%	300W Xe lamp	HCHO	0.79	64%	[19]
Bi_2WO_6	75%	300W Hg lamp Vis	HCHO	0.008	--	[67]
MoS_2 foam/CdS	76%	300 W Xe-lamp,	$HOCH_2CH_2OH$	11	90%	[19]
ZnS	76%	300W Xe lamp	$HOCH_2CH_2OH$	1.3	54%	[19]
CdS	76%	300W Xe lamp	$HOCH_2CH_2OH$	0.46	71%	[57]
CoP/$Zn_2In_2S_5$	76%	300W Xe lamp	$HOCH_2CH_2OH$	1.1	84%	[57]
Ag/TiO_2	100%	500 W Hg lamp 365nm	$HCOOCH_3$	7.3	80%	[68]
Au/TiO_2	100%	500 W Hg lamp 365nm	$HCOOCH_3$	5.9	75%	[68]
Cu/TiO_2	100%	500 W Hg lamp	$HCOOCH_3$	4.4	55%	[69]
CuO/CuZnAl	100%	500 W Hg lamp	$HCOOCH_3$	5.8	60%	[70]

GaN	100%	300 W Xe lamp for	CH_3CH_2OH	4.05	100%	[23]

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