## **Electronic Supplementary Information**

## **Regulating Effect of Heterojunction on Electrocatalytic Oxidation of**

## Methanol for Pt/WO<sub>3</sub>-NaTaO<sub>3</sub> Catalyst Regenerated CO

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**Fig. S1.** XRD patterns of  $Pt/WO_3$ ,  $Pt/NaTaO_3$ , and  $Pt/WO_3$ -NaTaO\_3 composites with different molar ratio (W:Ta = 5:1, 3:1, 1:1, 0.5:1, 0.3:1, 0.2:1, 0.1:1).



Fig. S2. Full XPS spectrum of (a) pure WO<sub>3</sub>, (b) bare NaTaO<sub>3</sub>, (c) Pt/WO<sub>3</sub>-NaTaO<sub>3</sub> composites with W:Ta =3:1, (d) Pt/WO<sub>3</sub>-NaTaO<sub>3</sub> composites with W:Ta =0.2:1.



Fig. S3. TEM and HRTEM images of the commercial Pt/C (20 wt.%).

**Table S1.** The electrochemically active surface area (ECSA) calculated by the cyclic voltammetric curves in Fig. 7 and the onset potential, peak potential, current density and the tolerance  $(I_f/I_b)$  of commercial Pt/C catalyst and three as-prepared Pt/WO<sub>3</sub>-NaTaO<sub>3</sub> composite catalysts in acid.

Type of Pt/C catalyst	ECSA/m <sup>3</sup> ·g <sup>-1</sup> Pt	Oneset potential/V	Peak potential/V	Current density /mA·cm <sup>-2</sup>	I <sub>f</sub> /I <sub>b</sub>
$Pt/WO_3$ -NaTaO <sub>3</sub> W:Ta = 5:1	16.8	0.42	0.64	55	0.81
W.Ta = 5.1 Pt/WO <sub>3</sub> -NaTaO <sub>3</sub> W:Ta = 3.1	40.6	0.35	0.69	120	1.19
$Pt/WO_3$ -NaTaO <sub>3</sub> $W:T_2 = 1:1$	31.88	0.36	0.68	100	1.12
W.Ta = 1.1 Pt/WO <sub>3</sub> -NaTaO <sub>3</sub>	23.41	0.35	0.62	85	1.09
W:Ta = 0.5:T $Pt/WO_3-NaTaO_3$ W:Ta = 0.3:T	21.6	0.45	0.64	70	1.06
$Pt/WO_3$ -NaTaO <sub>3</sub> W·Ta = 0.2·1	18.3	0.41	0.63	60	0.98
$Pt/WO_3$ -NaTaO <sub>3</sub> W·Ta = 0 1·1	13.2	0.44	0.64	30	0.92
commercial Pt/C	14.4	0.41	0.64	40	0.88
Pt/WO <sub>3</sub>	14.3	0.43	0.65	35	0.86
Pt/NaTaO <sub>3</sub>	13.6	0.46	0.65	30	0.79

**Table S2.** The electrochemically active surface area (ECSA) calculated by the cyclic voltammetric curves in Fig. 8 and the onset potential, peak potential, current density and the tolerance  $(I_f/I_b)$  of commercial Pt/C catalyst and three as-prepared Pt/WO<sub>3</sub>-NaTaO<sub>3</sub> composite catalysts in alkali.

Type of Pt/C catalyst	ECSA/m <sup>3</sup> ·g <sup>-1</sup> Pt	Oneset potential/V	Peak potential/V	Current density /mA·cm <sup>-2</sup>	I <sub>f</sub> /I <sub>b</sub>
$Pt/WO_3$ -NaTa $O_3$ W:Ta = 5:1	13.7	-0.36	-0.14	28	0.86
W.Ta = 5.1 Pt/WO <sub>3</sub> -NaTaO <sub>3</sub> W.Ta = 3.1	22.51	-0.37	-0.09	48	1.15
$Pt/WO_3$ -NaTaO <sub>3</sub>	24.3	-0.38	-0.09	55	1.09
W.Ta = 1.1 Pt/WO <sub>3</sub> -NaTaO <sub>3</sub>	26.3	-0.44	-0.06	60	1.02
W.Ta = 0.51T Pt/WO <sub>3</sub> -NaTaO <sub>3</sub>	29.1	-0.45	-0.05	75	0.99
$W: Ia = 0.3:I$ $Pt/WO_3-NaTaO_3$	31.9	-0.51	-0.1	90	0.93
W:1a = $0.2:1$ Pt/WO <sub>3</sub> -NaTaO <sub>3</sub> W:Ta = $0.1:1$	13.2	-0.36	-0.094	23	0.89
commercial Pt/C	13.5	-0.37	-0.131	28	0.81
Pt/WO <sub>3</sub>	7.17	-0.32	-0.178	18	0.78
Pt/NaTaO <sub>3</sub>	14.2	-0.38	-0.092	30	0.88



**Fig. S4.** Schematic drawing of supercell model for WO<sub>3</sub>(020) surface: (a) before relaxation, (b) after relaxation.

Adsorption location	$\Delta E_{chem}(eV)$
W-top	-0.19
W-W	0.77
hollow	0.89

**Table S3.** Chemical adsorption energy ( $\Delta E_{chem}$ ) of CH3OH molecule on different location forWO3(020) surface.



Fig. S5. Density of state for CH<sub>3</sub>OH molecule adsorbed on W-top site of WO<sub>3</sub>(020).