Supporting Information

Revealing the Promotion of Carbonyl groups on Vacancy Stabilized Pt_4/nanocarbons for Propane Dehydrogenation

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Definition of the binding energies of Pt clusters anchored on carbon nanoribbons with vacancies and OCGs

The binding energies of Pt clusters were calculated by the following equation:

$$E_b = E_{Pt/Support} - E_{Pt} - E_{Support} \quad \text{(S1)}$$

Where $E_{Pt/Support}$ was the total energy of the Pt clusters anchored on the carbon nanoribbons with various vacancies and OCGs; $E_{Pt}$was the energy of Pt clusters in vacuum; $E_{Support}$ was the total energy of the carbon nanoribbons.

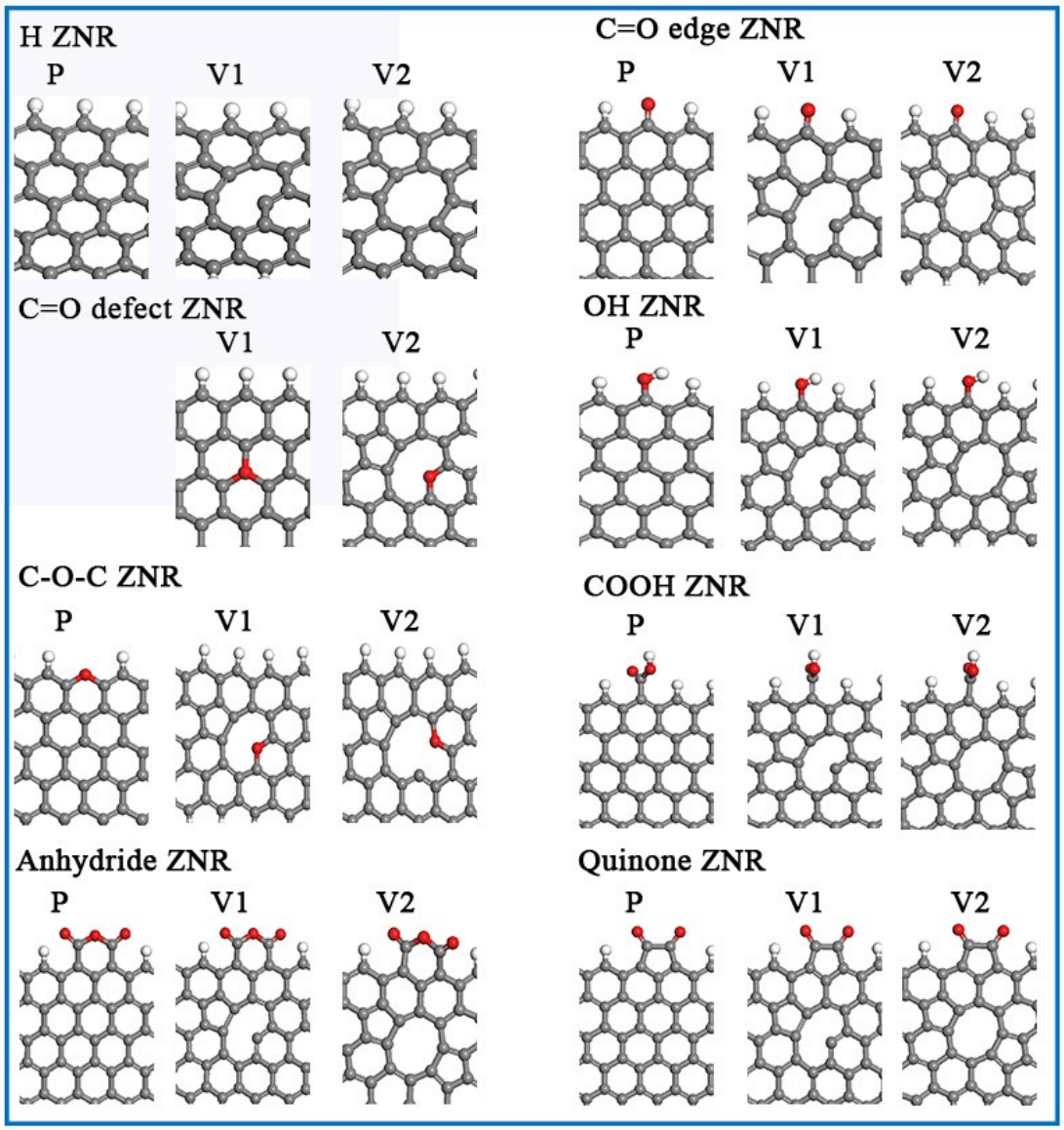

Fig. S1. The geometric configurations of different vacancies and OCGs in the carbon nanoribbons with zigzag edges.

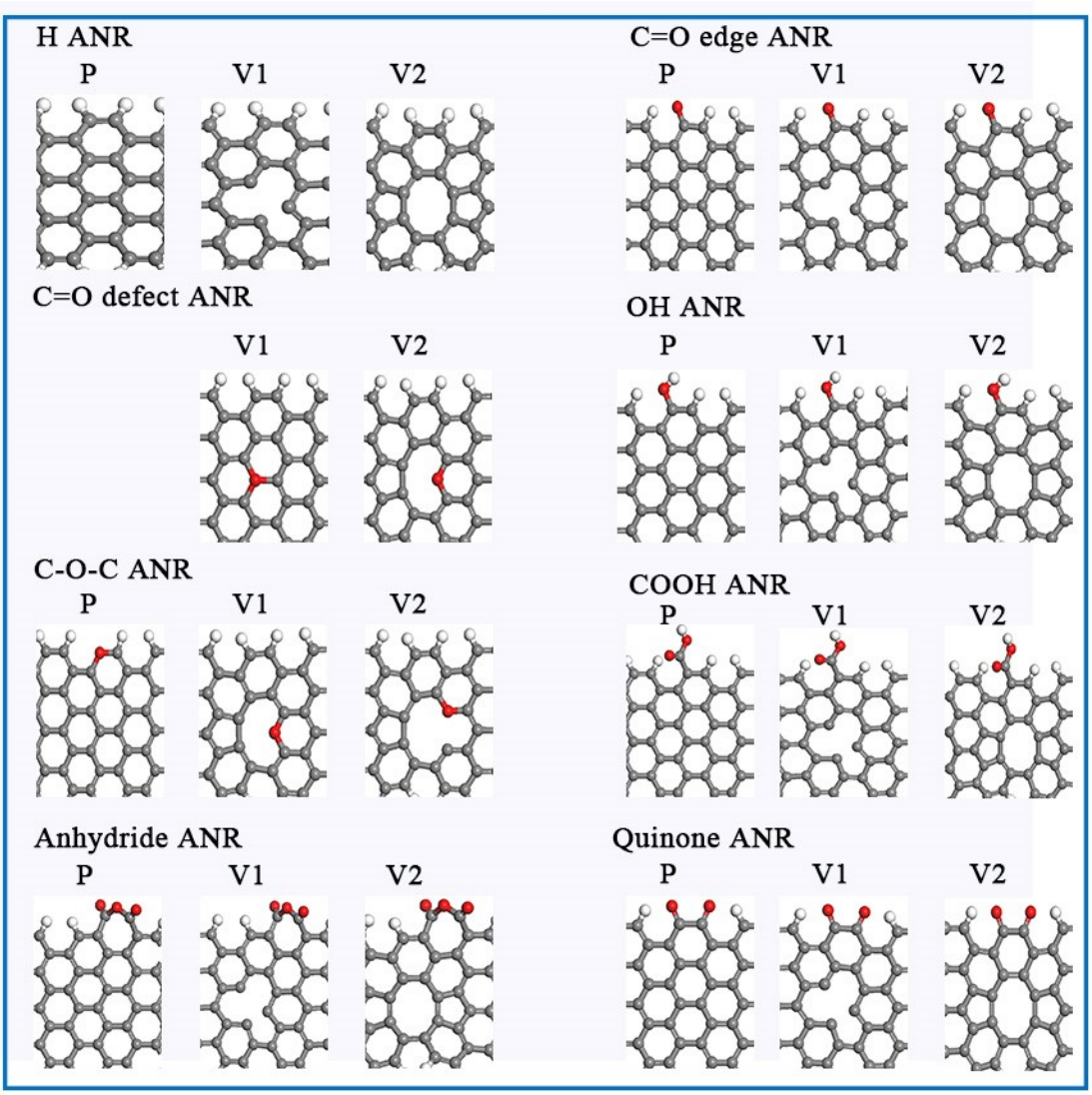

Fig. S2. The geometric configurations of different vacancies and OCGs in the carbon nanoribbons with armchair edges.

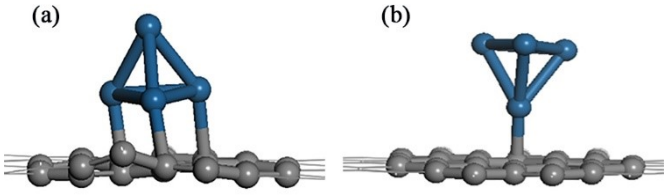

Fig. S3. The anchored models of (a) top tetrahedral (b) bottom tetrahedral of Pt clusters anchored on carbon nanoribbon.

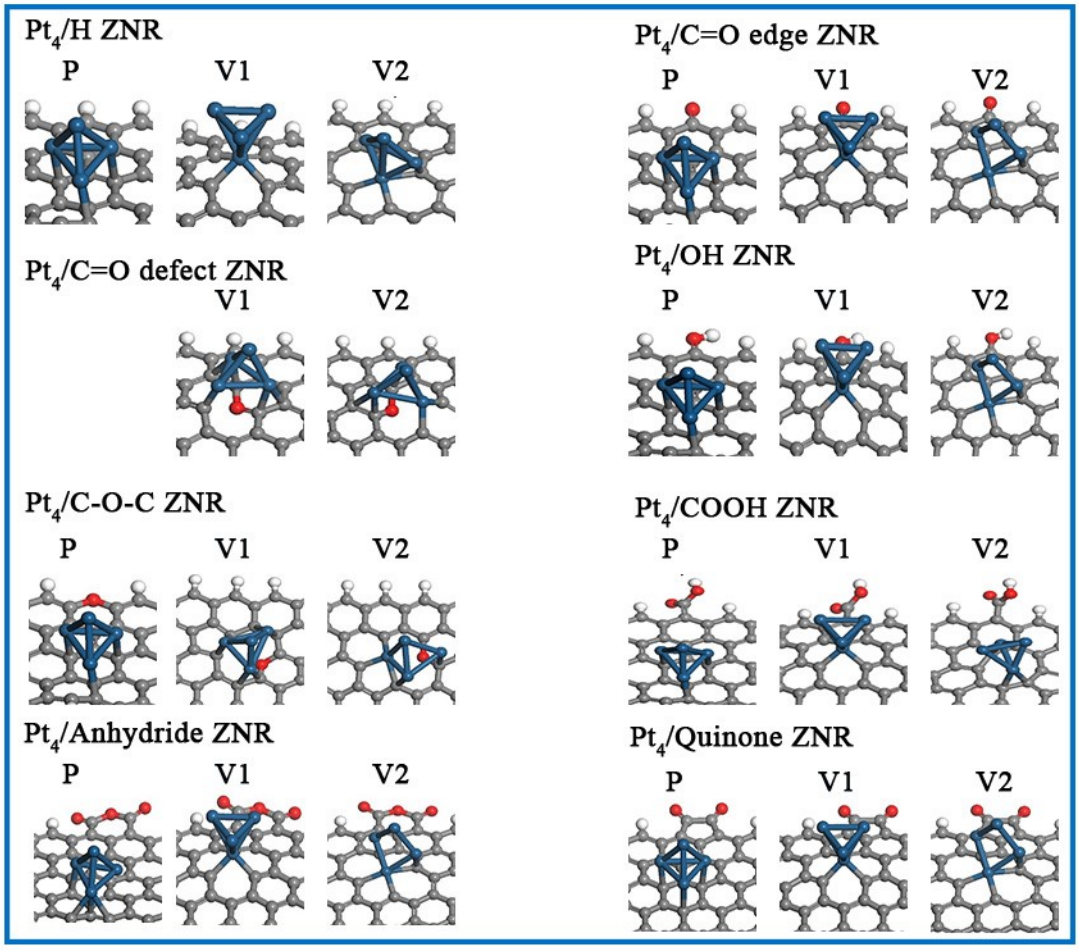

Fig. S4. The optimized configurations of Pt_4 anchored on the carbon nanoribbons with zigzag edges.

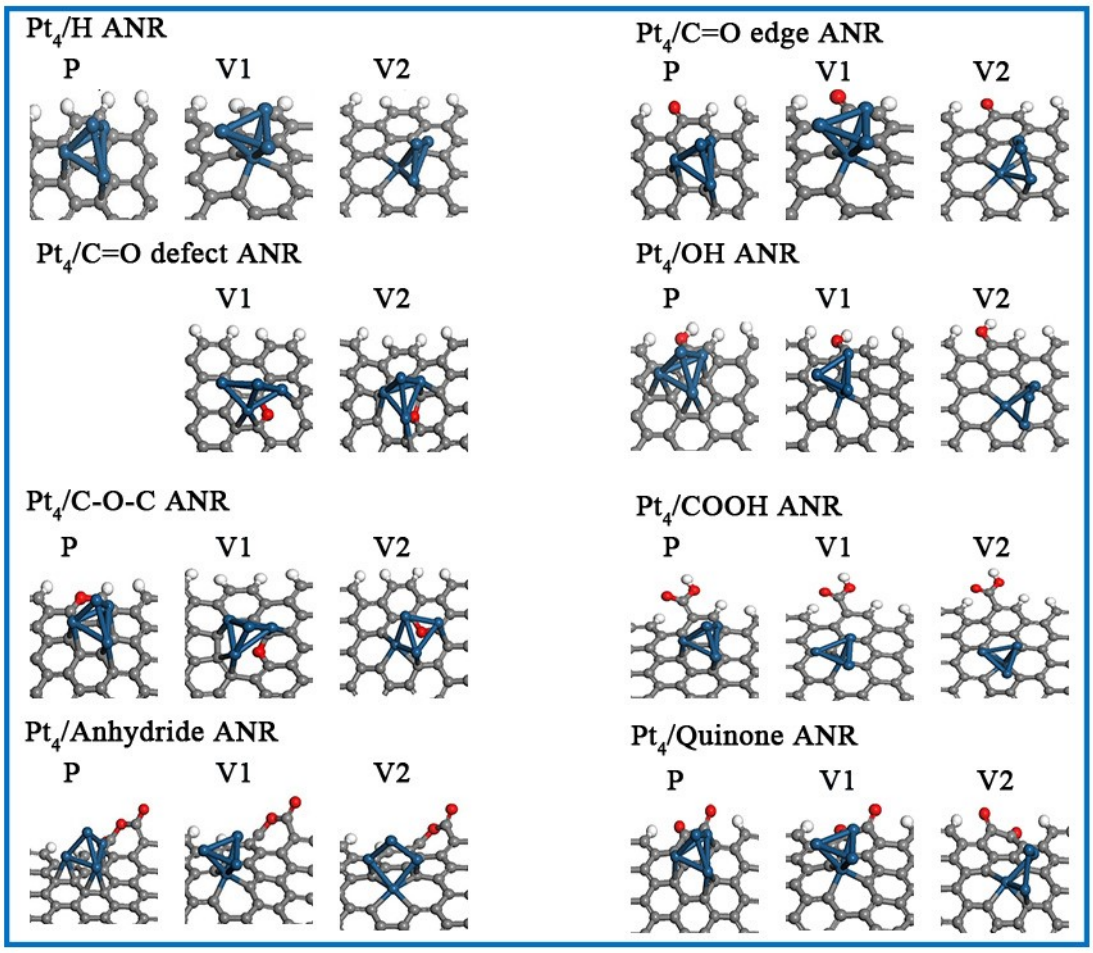

Fig. S5. The optimized configurations of Pt_4 anchored on the carbon nanoribbons with armchair edges.

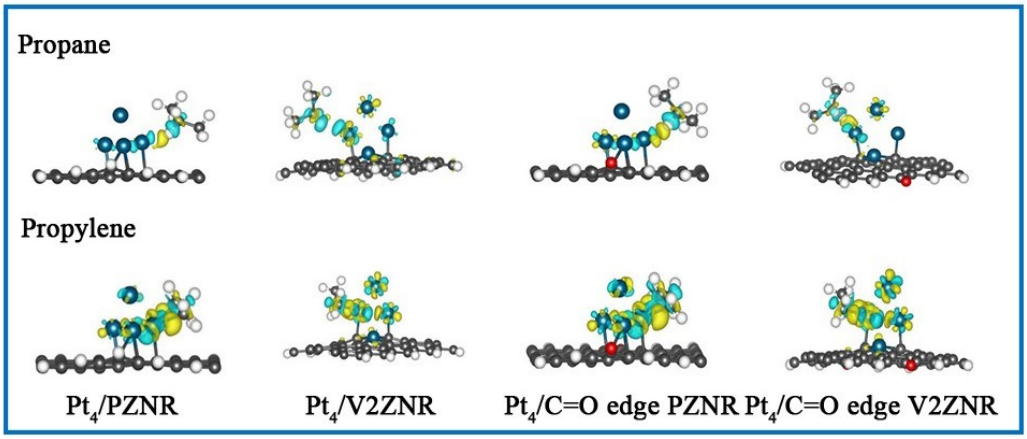

Fig. S6. Charge density differences of propane and propylene adsorbed on the catalysts, the charge accumulation and depletion were colored cyan and yellow, with the isosurface value being 0.003 a.u.

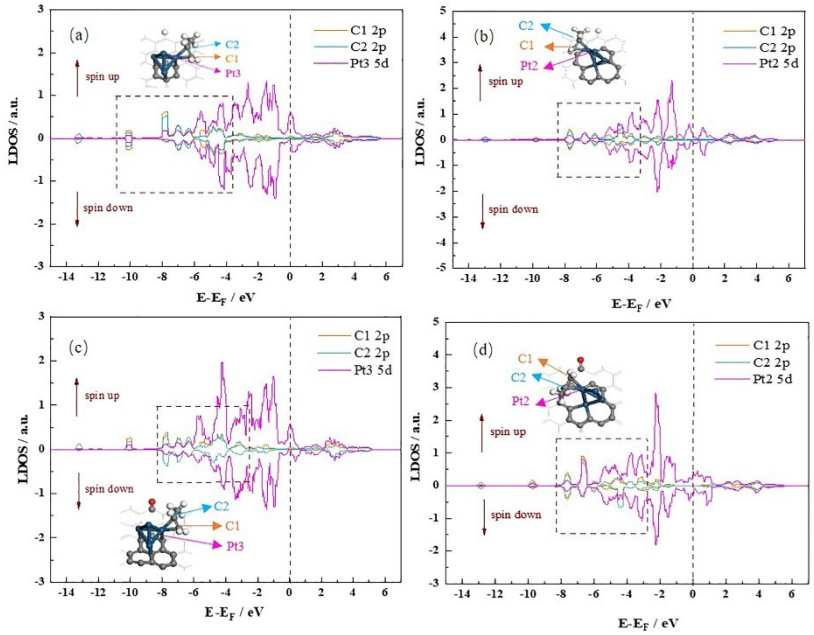

Fig. S7. The partial density of orbit states (PDOS) of the d-orbit in Pt atoms and p-orbit in C atoms of propylene adsorbed in (a) Pt_4/PZNR, (b) Pt_4/V2ZNR, (c) Pt_4/C=O edge PZNR, (d) Pt_4/C=O edge V2ZNR.

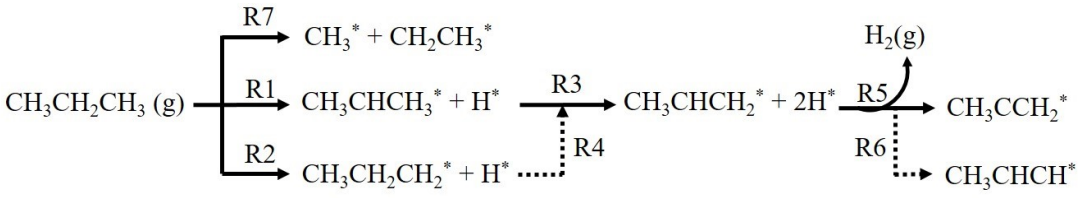

Fig. S8. The elementary reaction pathways involved in PDH.

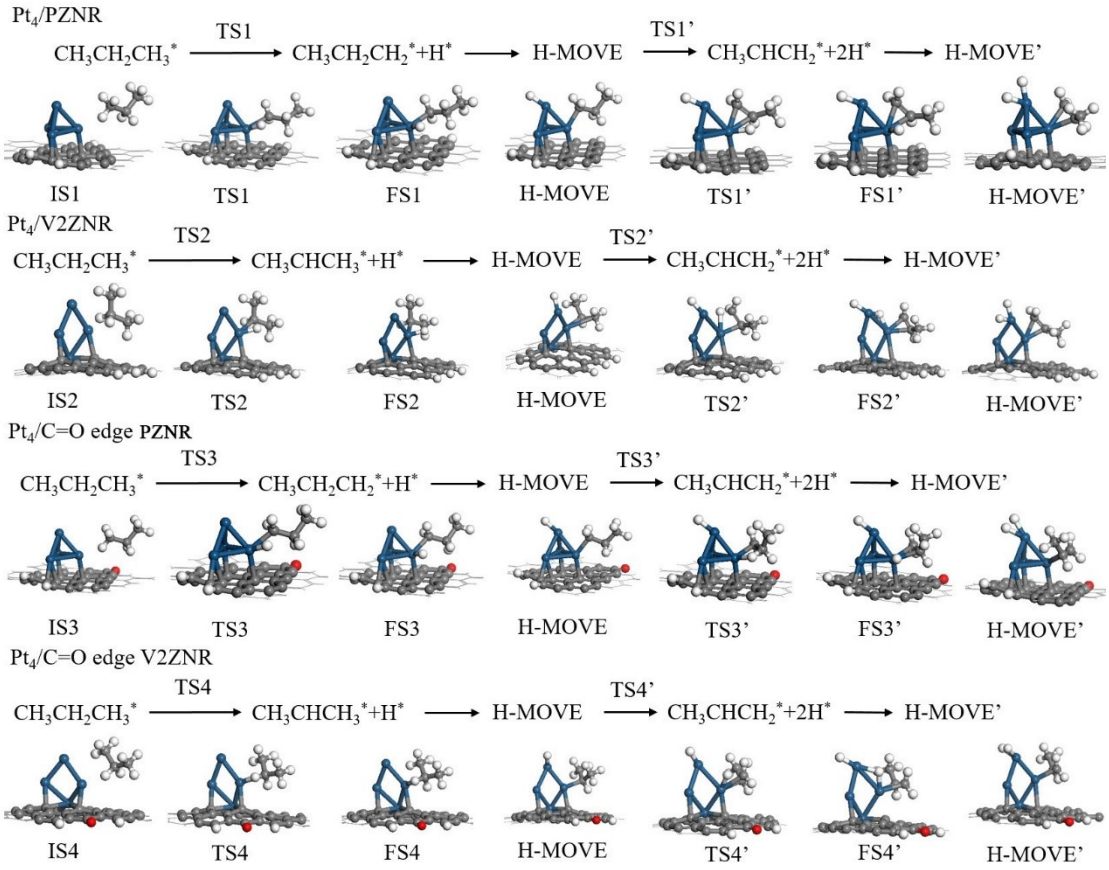

Fig. S9. The geometric configurations of the key reactive intermediates involved in the PDH.

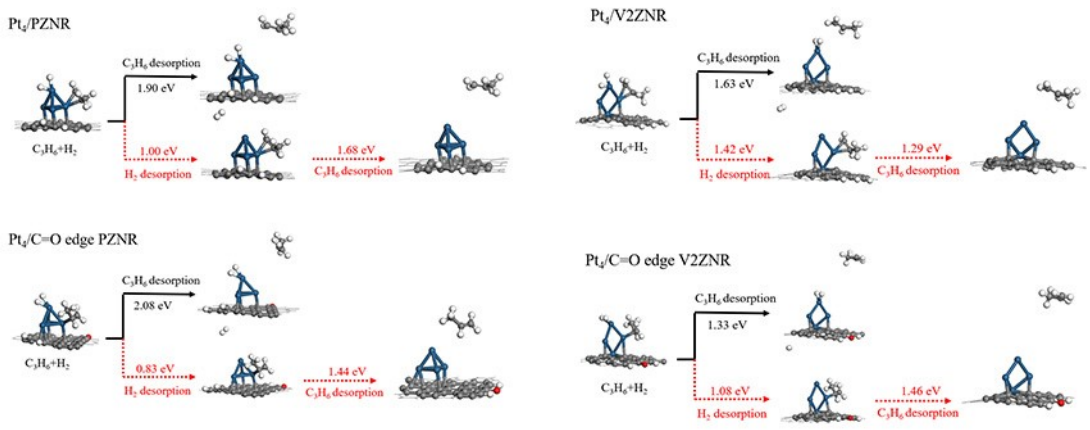

Fig. S10. The competition desorption of propylene and hydrogen during the regeneration of the active site.

Table S1. The activation barriers and reaction energies of C-H bond cleavage in CH_4 catalyzed by Pt_4 anchored on carbon nanoribbons.

Catalyst	E_a / eV	ΔE / eV	Catalyst	E_a / eV	ΔE / eV
Pt_4/PZNR	0.19	-0.14	Pt_4/PANR	0.28	-0.12
Pt_4/V1ZNR	0.18	-0.43	Pt_4/V1ANR	0.20	-0.53
Pt_4/V2ZNR	0.12	-0.18	Pt_4/V2ANR	0.24	-0.25
Pt_4/C=O edge PZNR	0.17	-0.01	Pt_4/C=O edge PANR	0.13	-0.43
Pt_4/C=O edge V1ZNR	0.17	-0.44	Pt_4/C=O edge V1ANR	0.19	-0.25
Pt_4/C=O edge V2ZNR	0.09	-0.21	Pt_4/C=O edge V2ANR	0.23	-0.09
Pt_4/C=O defect V1ZNR	0.47	-0.06	Pt_4/C=O defect V1ANR	0.24	0.01
Pt_4/C=O defect V2ZNR	0.43	0.06	Pt_4/C=O defect V2ANR	0.34	-0.02
Pt_4/OH PZNR	0.17	-0.32	Pt_4/OH PANR	0.17	-0.48
Pt_4/OH V1ZNR	0.17	-0.34	Pt_4/OH V1ANR	0.18	-0.54
Pt_4/OH V2ZNR	0.12	-0.18	Pt_4/OH V2ANR	0.13	-0.24
Pt_4/C-O-C PZNR	0.17	-0.17	Pt_4/C-O-C PANR	0.14	-0.42
Pt_4/C-O-C V1ZNR	0.23	-0.10	Pt_4/C-O-C V1ANR	0.22	-0.53
Pt_4/C-O-C V2ZNR	0.32	-0.16	Pt_4/C-O-C V2ANR	0.26	-0.25
Pt_4/COOH PZNR	0.15	-0.12	Pt_4/COOH PANR	0.20	-0.18
Pt_4/COOH V1ZNR	0.26	-0.04	Pt_4/COOH V1ANR	0.22	-0.56
Pt_4/COOH V2ZNR	0.18	0.00	Pt_4/COOH V2ANR	0.37	-0.08
Pt_4/Anhydride PZNR	0.35	-0.09	Pt_4/Anhydride PANR	0.10	-0.49
Pt_4/Anhydride V1ZNR	0.22	-0.40	Pt_4/Anhydride V1ANR	0.22	-0.45
Pt_4/Anhydride V2ZNR	0.12	-0.16	Pt_4/Anhydride V2ANR	0.21	-0.17
Pt_4/Quinone PZNR	0.26	-0.31	Pt_4/Quinone PANR	0.19	-0.22
Pt_4/Quinone V1ZNR	0.17	-0.48	Pt_4/Quinone V1ANR	0.18	-0.39
Pt_4/Quinone V2ZNR	0.11	-0.18	Pt_4/Quinone V2ANR	0.16	-0.21

Table S2. The activation barriers and reaction energies of C-H bond cleavage from propane to propylene catalyzed by different active site in Pt_4/C=O edge V2ZNR.

Active site in Pt_4/C=O edge V2ZNR / Reactions	Pt_4 cluster		C=O edge groups	
	E_a / eV	ΔE / eV	E_a / eV	ΔE / eV
$CH_3CH_2CH_3^* \rightarrow CH_3CHCH_3^* + H^*$	0.40	0.02	1.02	-0.16
$CH_3CHCH_3^* + H^* \rightarrow CH_3CH{=}CH_2^* + 2H^*$	0.37	-0.28	1.13	0.16