

Supplementary Information

Structure-dependence and Metal-dependence on Atomically Dispersed Ir Catalysts for Efficient n-Butane Dehydrogenation

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Supplementary Figure 3. HAADF-STEM images of Ir_n/ND@G. (a) The image of Ir_n/ND@G at low magnification. (b) and (c) The image of Ir_n/ND@G at high magnification.

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Supplementary Figure 15. Raman spectra for the fresh and spent catalysts after 10-hours BDH. Raman spectra of (a) fresh Ir₁/ND@G, (b) spent Ir₁/ND@G, (c) fresh Ir_n/ND@G, (d) spent Ir_n/ND@G, (e) fresh IrNPs/ND@G and (f) spent IrNPs/ND@G.

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Supplementary Figure 17. HAADF-STEM images of Ir₁/ND@G after 0.5h BDH. (a) The image of Ir₁/ND@G after 0.5h BDH at low magnification. (b-d) The image of Ir₁/ND@G after 0.5h BDH at high magnification. The red circles highlighted single Ir atoms.

Supplementary Figure 18. HAADF-STEM images of Ir₁/ND@G after 20h BDH. (a) The image of Ir₁/ND@G after 20h BDH at low magnification.

(b) and (c) The image of Ir₁/ND@G after 20h BDH at high magnification. The red circles highlighted single Ir atoms.

Supplementary Figure 19. The conversion of butane and selectivity of butene over Ir₁/ND@G catalyst after several regeneration cycles. Reaction condition: atmospheric pressure, GHSV = 30000 mL · g_{cat}⁻¹ · h⁻¹, C₄H₁₀:H₂ = 1:1, He balance, 450 °C.

Supplementary Figure 20. The *in-situ* CO-DRIFTS of Ir₁/ND@G under reaction condition for 1h. Reaction conditions: 450 °C, 2%C₄H₁₀, 2%H₂, He balance.

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Supplementary Figure 27. C₄H₈-TPD profiles of the different catalysts. (a) ND@G, (b) Ir₁/ND@G, (c) Ir_n/ND@G and (d) Pt₃/ND@G. The peak at 80 °C is corresponding to physical absorption on ND@G.

Supplementary Figure 28. TOF and C₃H₆ selectivity over Ir₁/ND@G, Ir_{1+n}/ND@G, Ir_n/ND@G and IrNPs/ND@G.

Supplementary Figure 29. C₃H₈ conversion over Ir₁/ND@G, Pt₁/ND@G, Pd₁/ND@G and ND@G.

Supplementary Tables:

Supplementary Table 1. Physical structure properties of the catalysts.

Supplementary Table 2. Ir L₃-edge EXAFS fitting results for as-prepared catalysts.

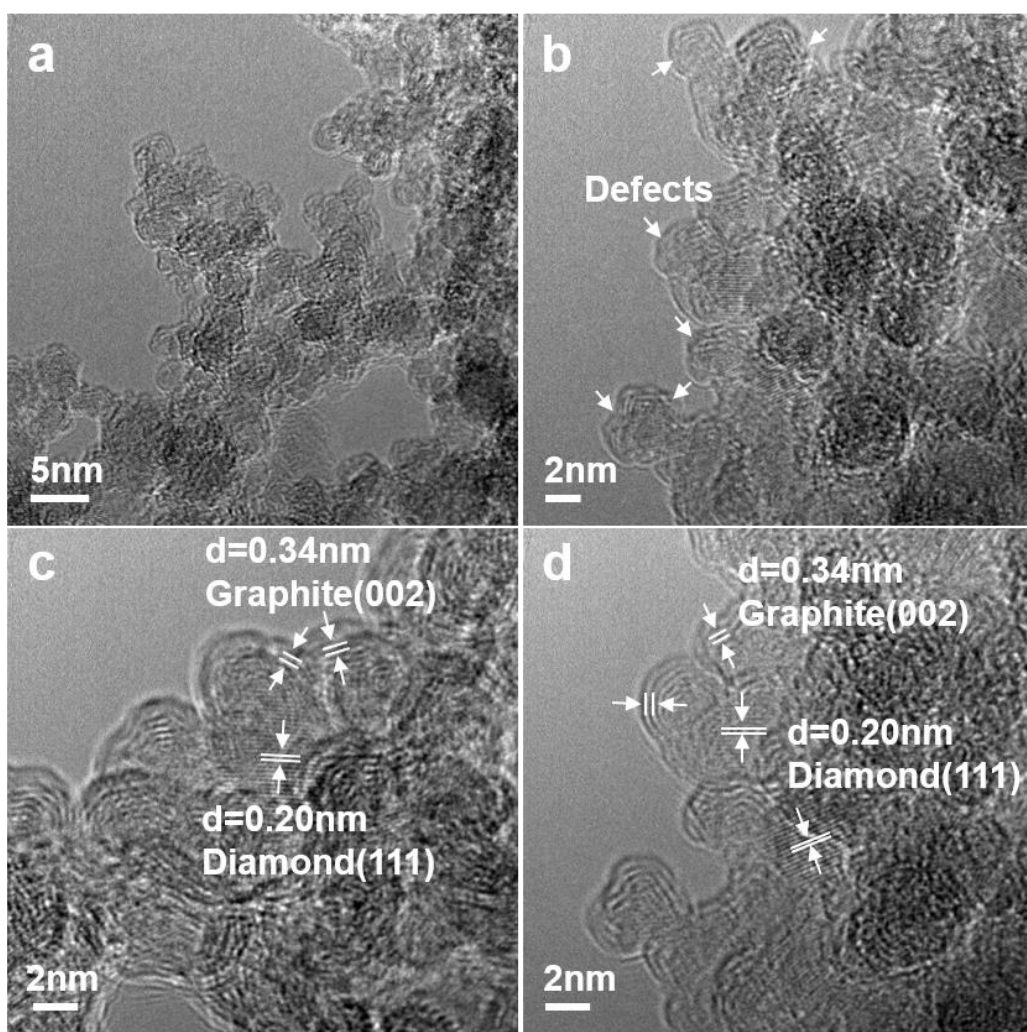
Supplementary Table 3. Catalytic performance of the catalysts for BDH.

Supplementary Table 4. Amount of carbon deposition on the catalysts for 10-hour BDH.

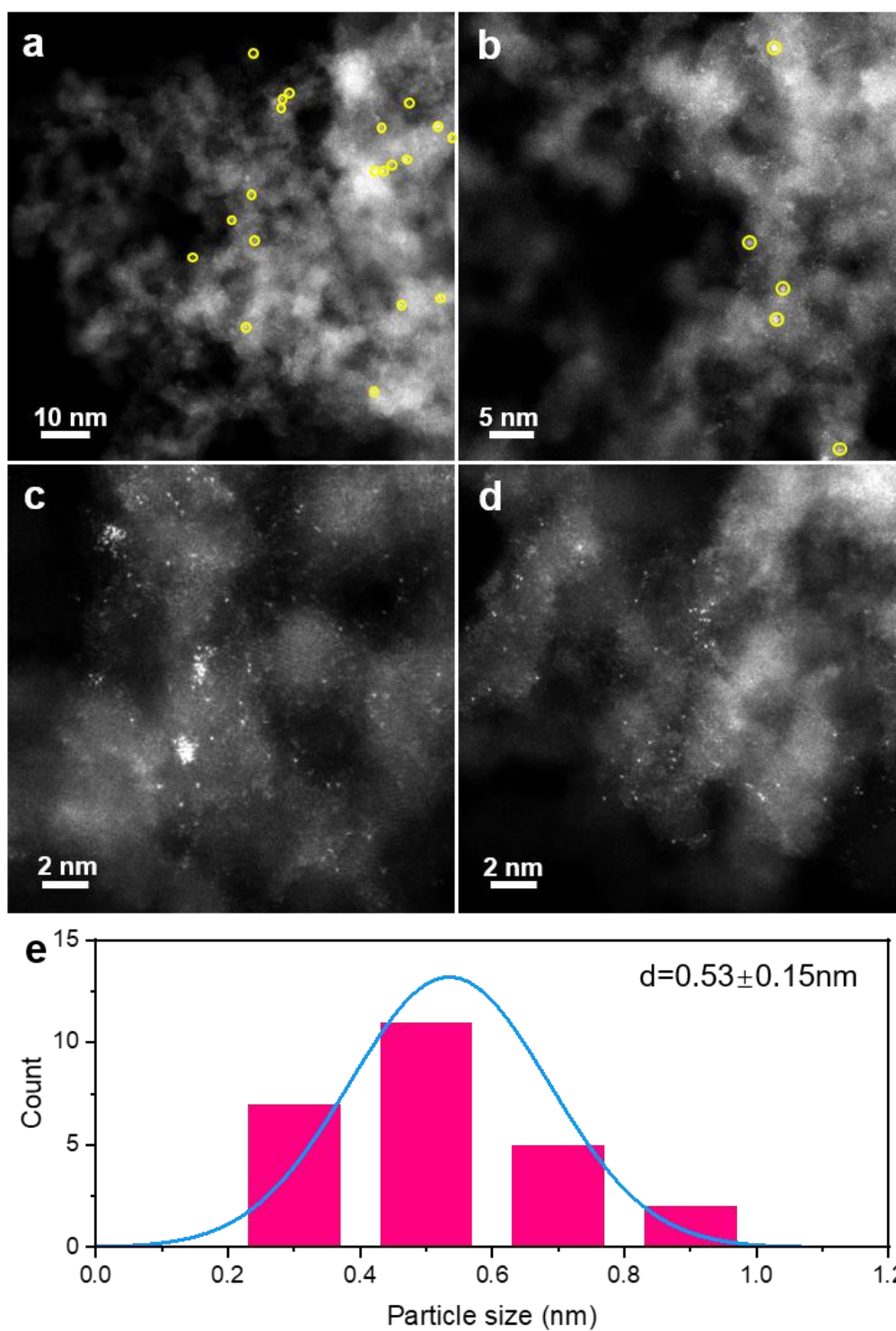
Supplementary Table 5. Catalytic performance of the catalysts for BDH.

Supplementary Table 6. Summary of the catalytic performance of various supported metal catalysts for BDH.

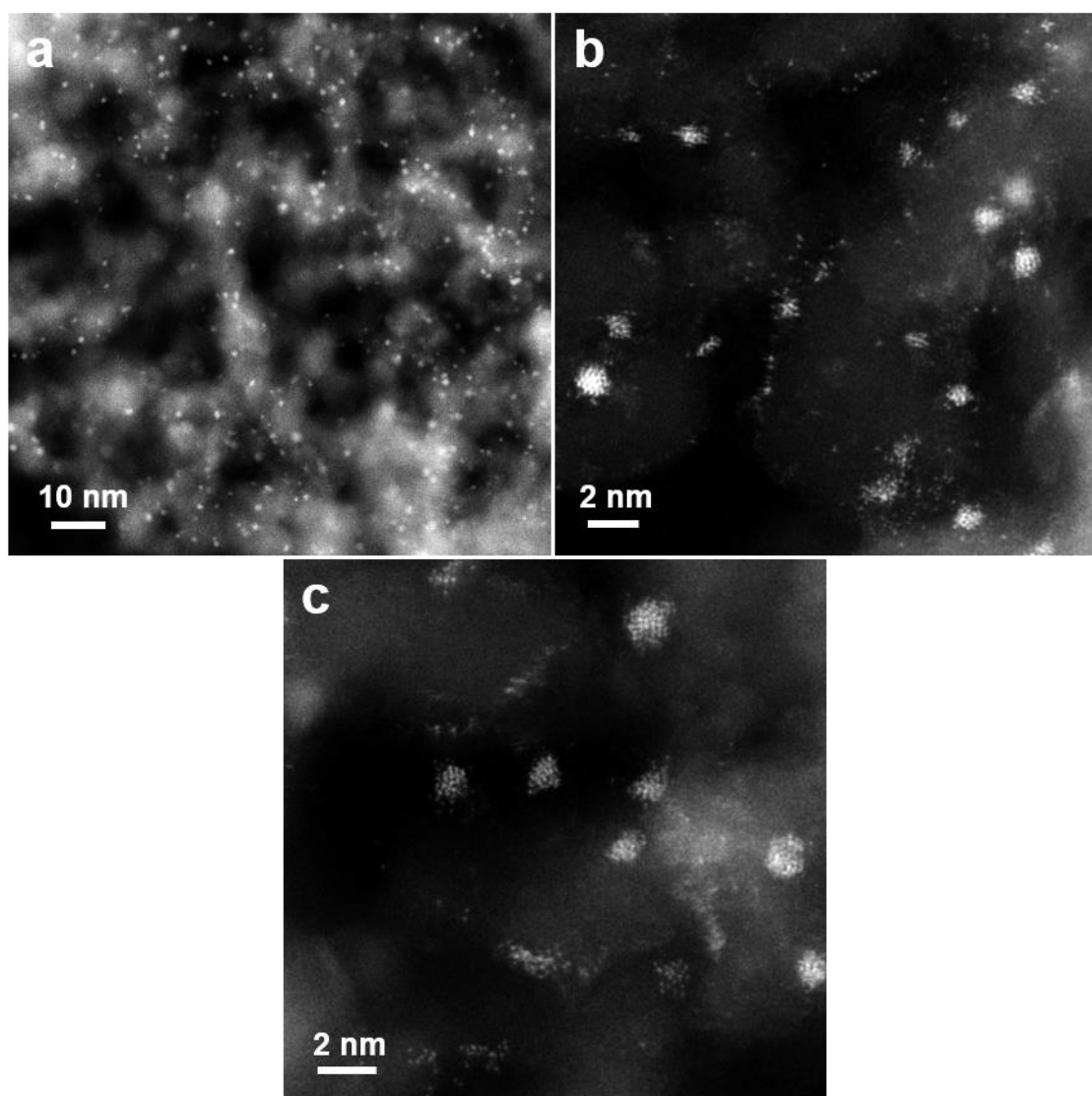
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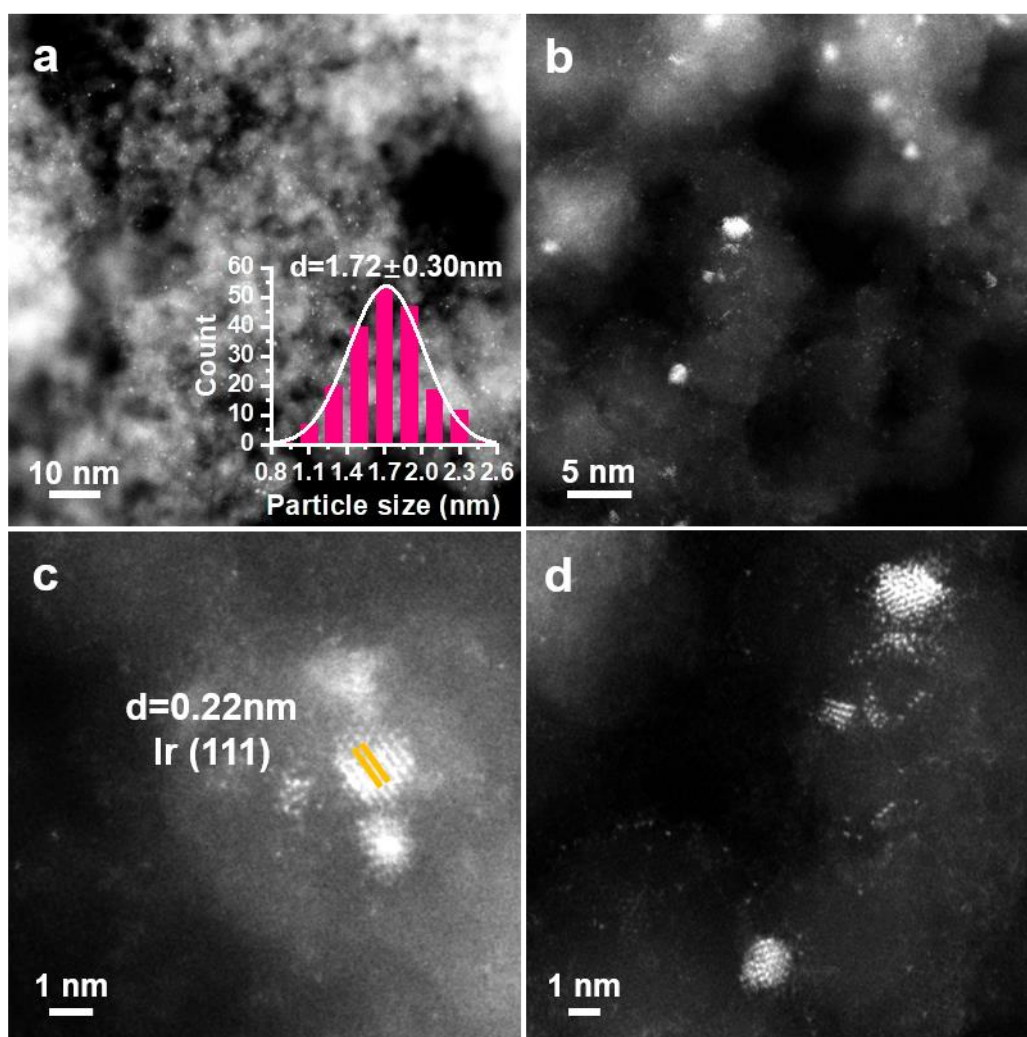
Supplementary Figure 1. HRTEM images of ND@G. (a) The surface structure at low magnification of ND@G. (b-d) The surface structure at high magnification of ND@G.



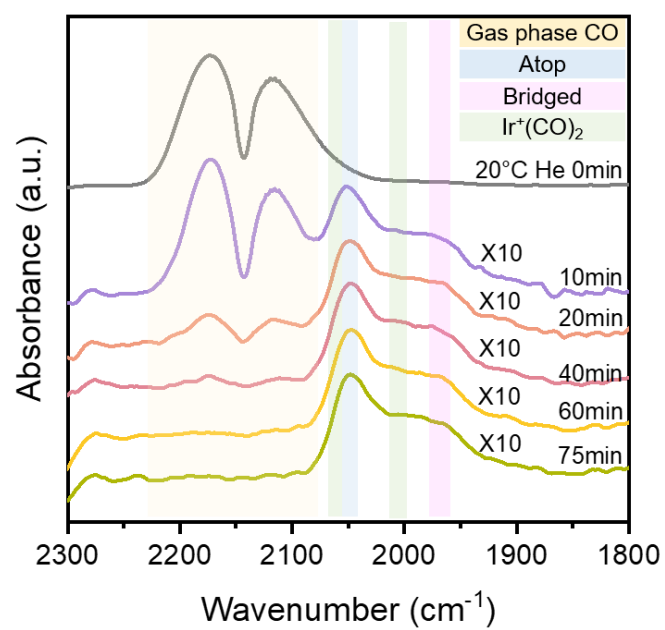
Supplementary Figure 2. Microscopic characterizations of Ir_{1+n}/ND@G. (a)-(d) HAADF-STEM images of Ir_{1+n}/ND@G. The Ir clusters were highlighted by yellow circles. (e) Particle size distribution of Ir_{1+n}/ND@G.



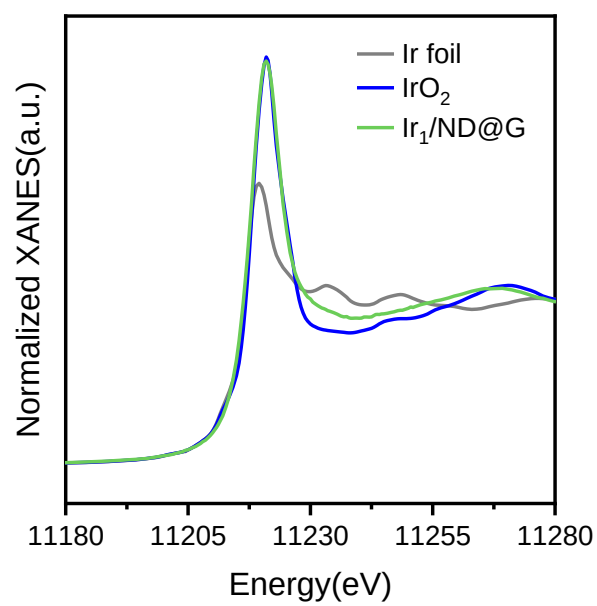
Supplementary Figure 3. HAADF-STEM images of Ir_n/ND@G. (a) The image of Ir_n/ND@G at low magnification. (b) and (c) The image of Ir_n/ND@G at high magnification.



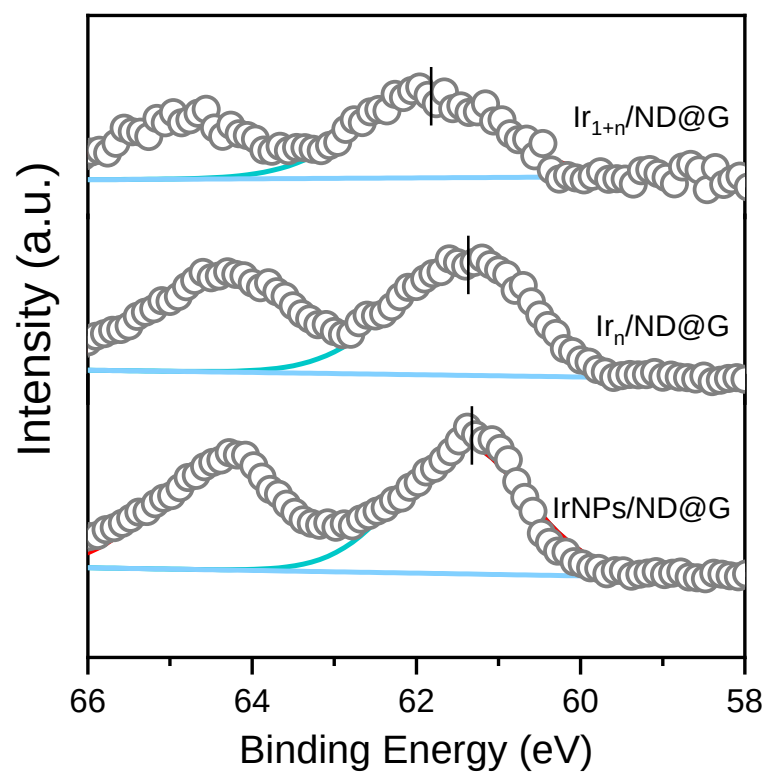
Supplementary Figure 4. HAADF-STEM images of IrNPs/ND@G. (a) The image of IrNPs/ND@G at low magnification. The inset is the particle size distribution of IrNPs/ND@G. (b-d) The image of IrNPs/ND@G at high magnification.



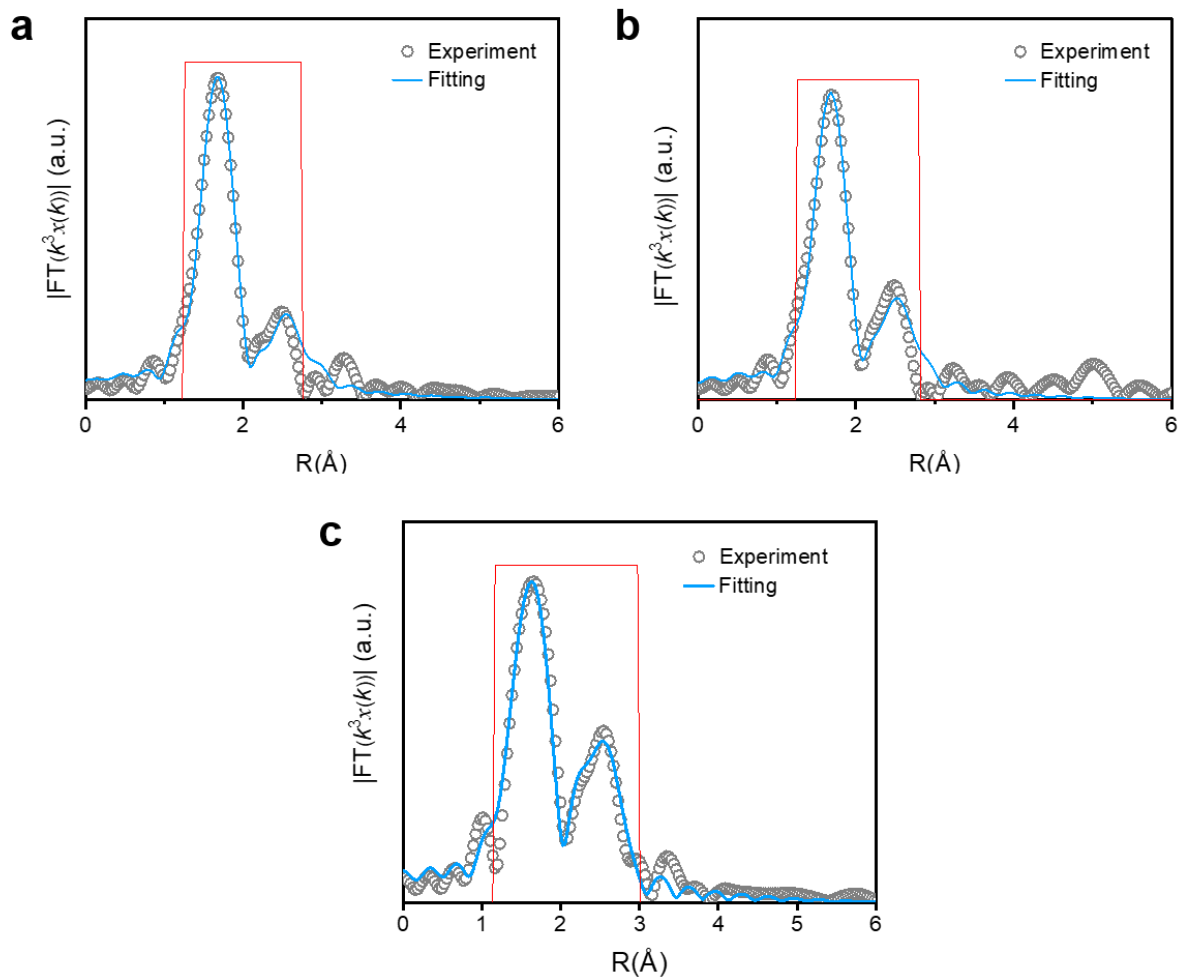
Supplementary Figure 5. *In-situ* CO-DRIFTS of IrNPs/ND@G.



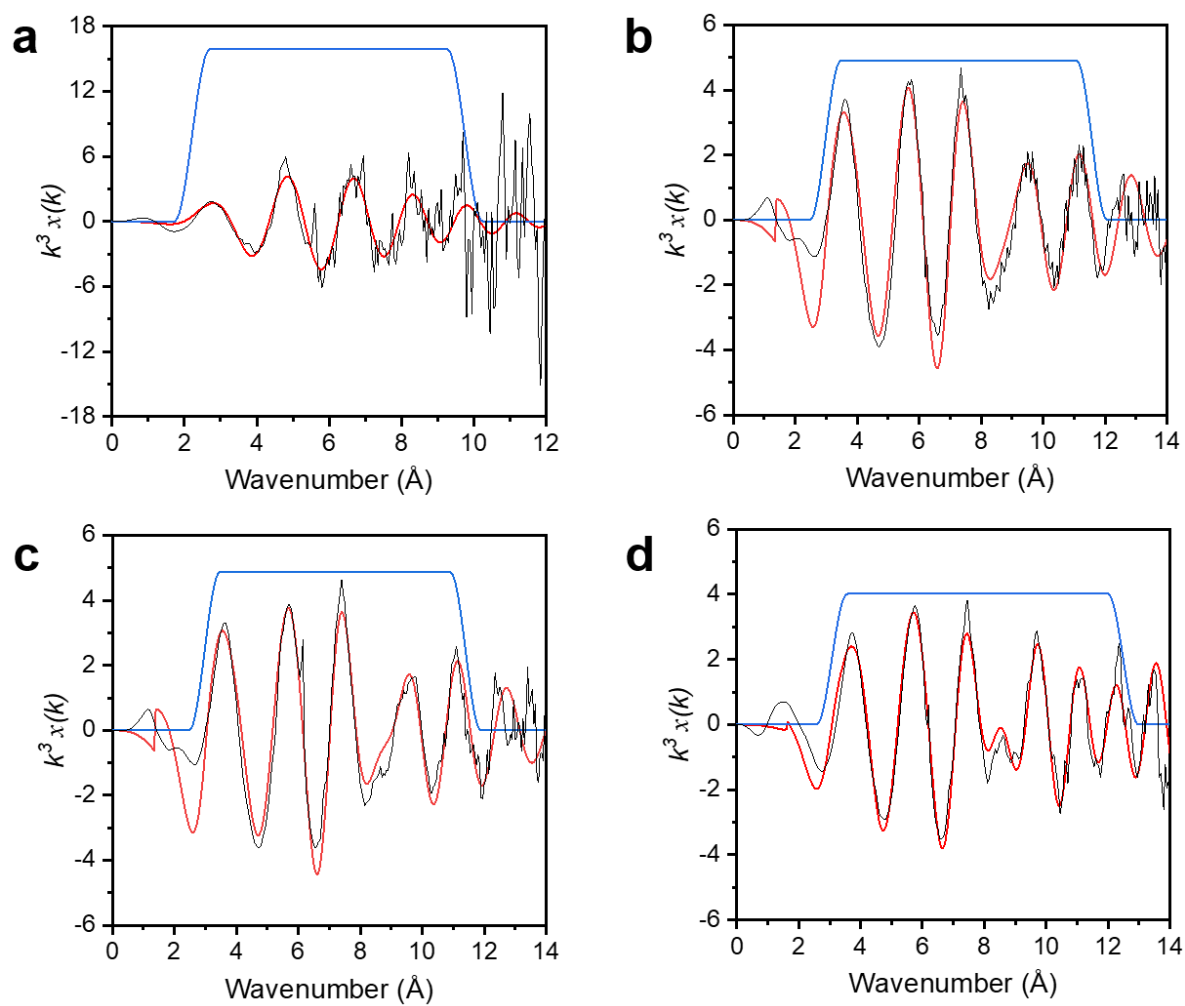
Supplementary Figure 6. Ir L_3 XANES spectra of Ir₁/ND@G, Ir foil and IrO₂.



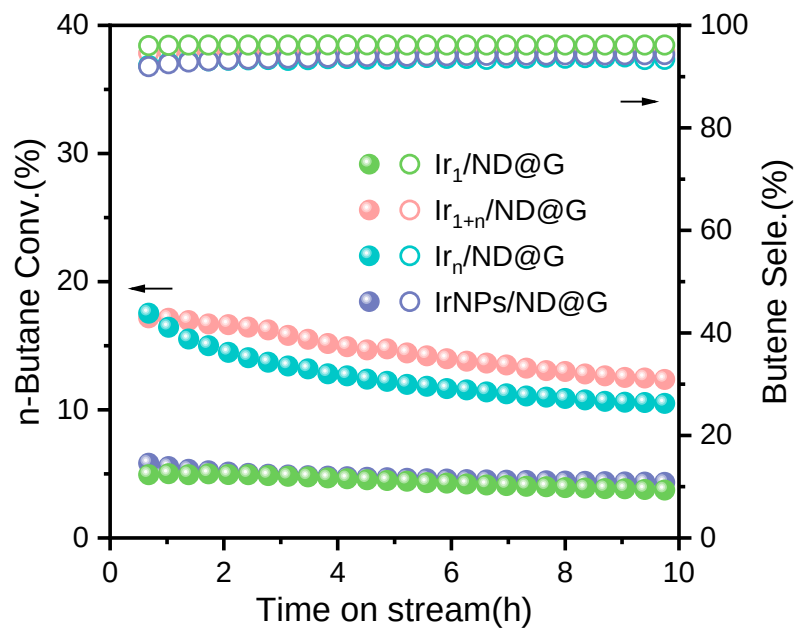
Supplementary Figure 7. Pt 4f XPS spectra of $\text{Ir}_{1+n}/\text{ND@G}$, $\text{Ir}_n/\text{ND@G}$ and $\text{IrNPs}/\text{ND@G}$.



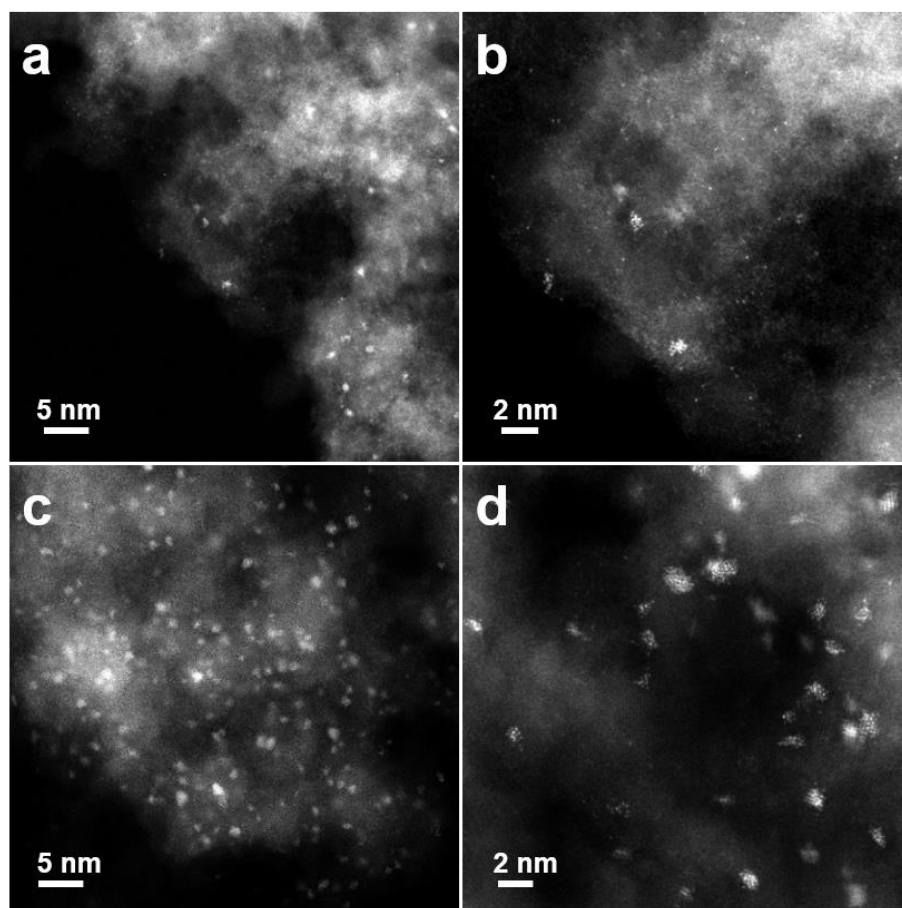
Supplementary Figure 8. EXAFS fitting results of Ir L_3 -edge in R space. (a) Ir_{1+n}/ND@G, (b) Ir_n/ND@G and (c) IrNPs/ND@G.



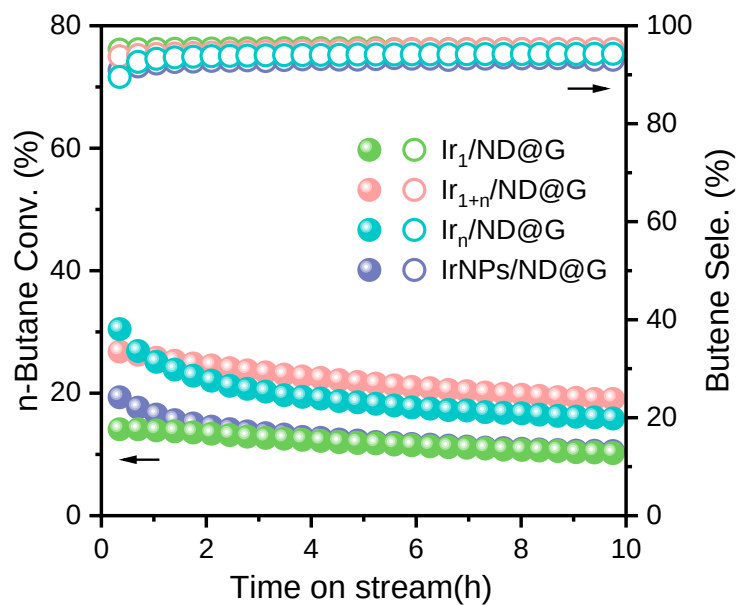
Supplementary Figure 9. EXAFS fitting results of Ir L_3 -edge in k^3 space. (a) Ir₁/ND@G, (b) Ir_{1+n}/ND@G, (c) Ir_n/ND@G and (d) IrNPs/ND@G.



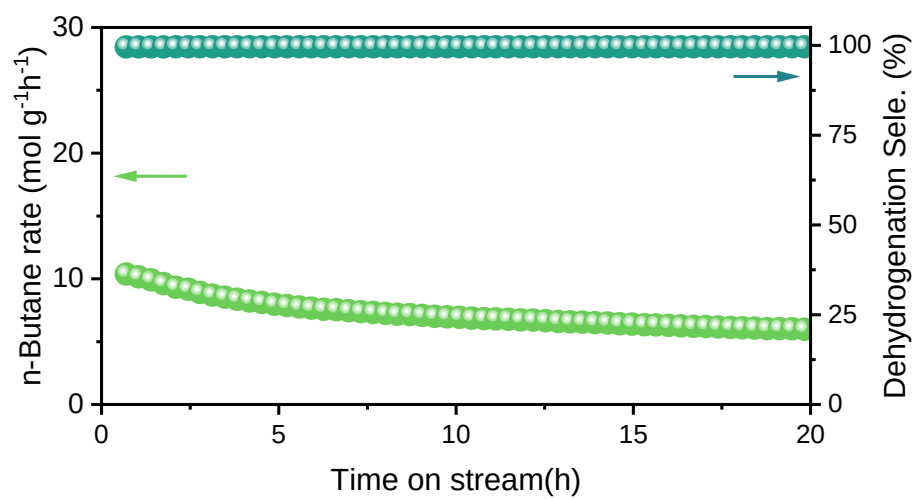
Supplementary Figure 10. The catalytic performance of as-prepared catalysts. n-Butane conversion and butene selectivity by time-on-stream during BDH at 450 °C, GHSV = 45000 $\text{mL} \cdot \text{g}_{\text{cat}}^{-1} \cdot \text{h}^{-1}$, $\text{n-C}_4\text{H}_{10} : \text{H}_2 = 1:1$ with He balance.



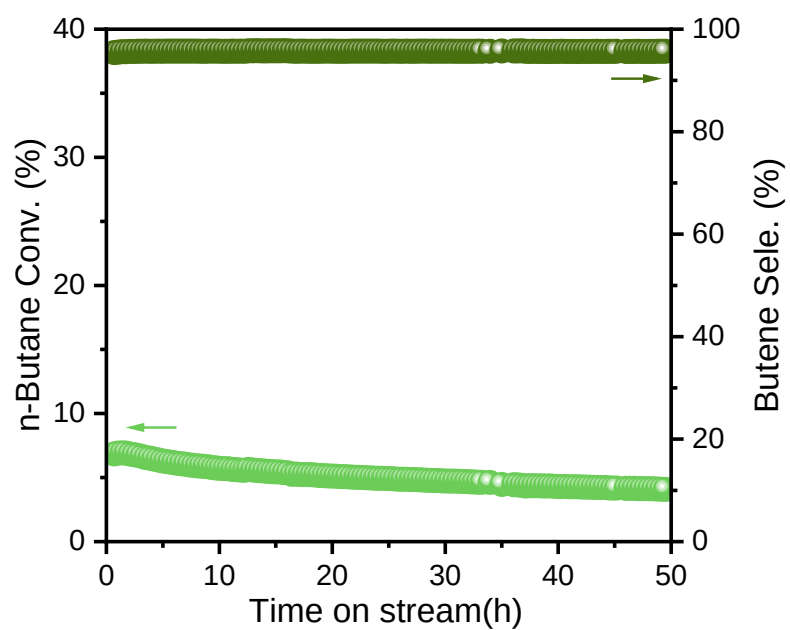
Supplementary Figure 11. Microscopic characterizations of Ir catalysts after 10h BDH. (a) and (b) HAADF-STEM images of Ir_{1+n}/ND@G. (c) and (d) HAADF-STEM images of Ir_n/ND@G.



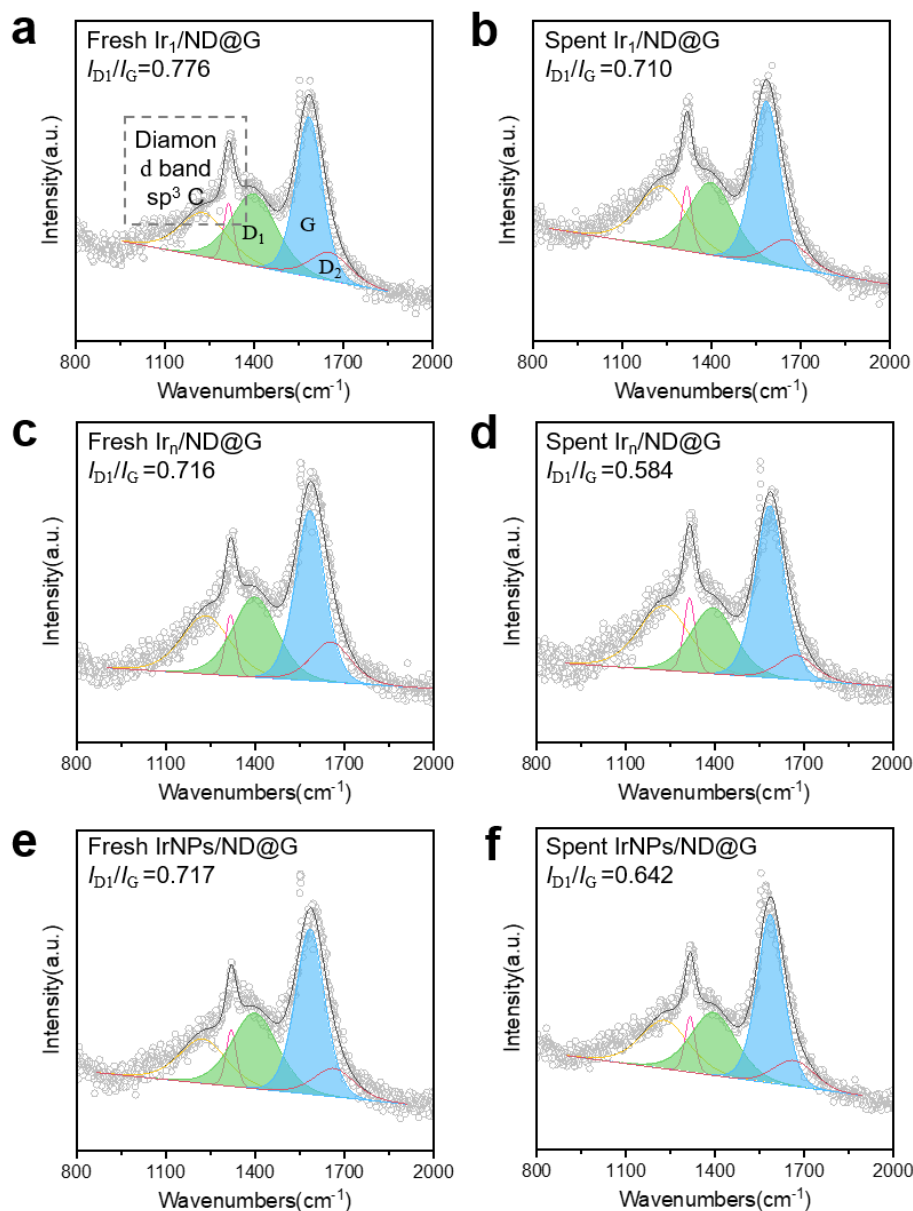
Supplementary Figure 12. The catalytic performance of 50mg as-prepared catalysts. n-Butane conversion and butene selectivity by time-on-stream during BDH at 450 °C, GHSV = $18000 \text{ mL} \cdot \text{g}_{\text{cat}}^{-1} \cdot \text{h}^{-1}$, $n\text{-C}_4\text{H}_{10} : \text{H}_2 = 1:1$ with He balance.



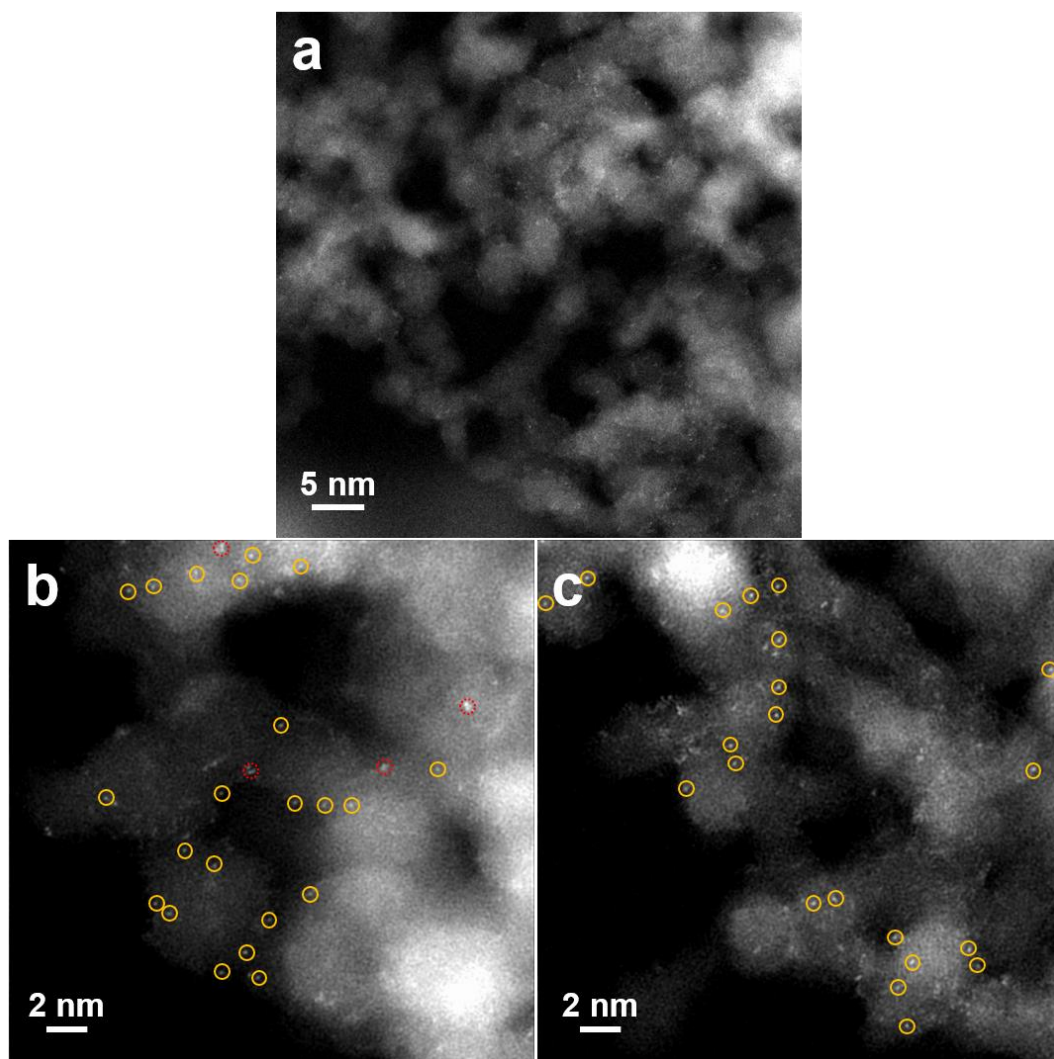
Supplementary Figure 13. Stability test over Ir₁/ND@G at 450 °C for 20 hours.



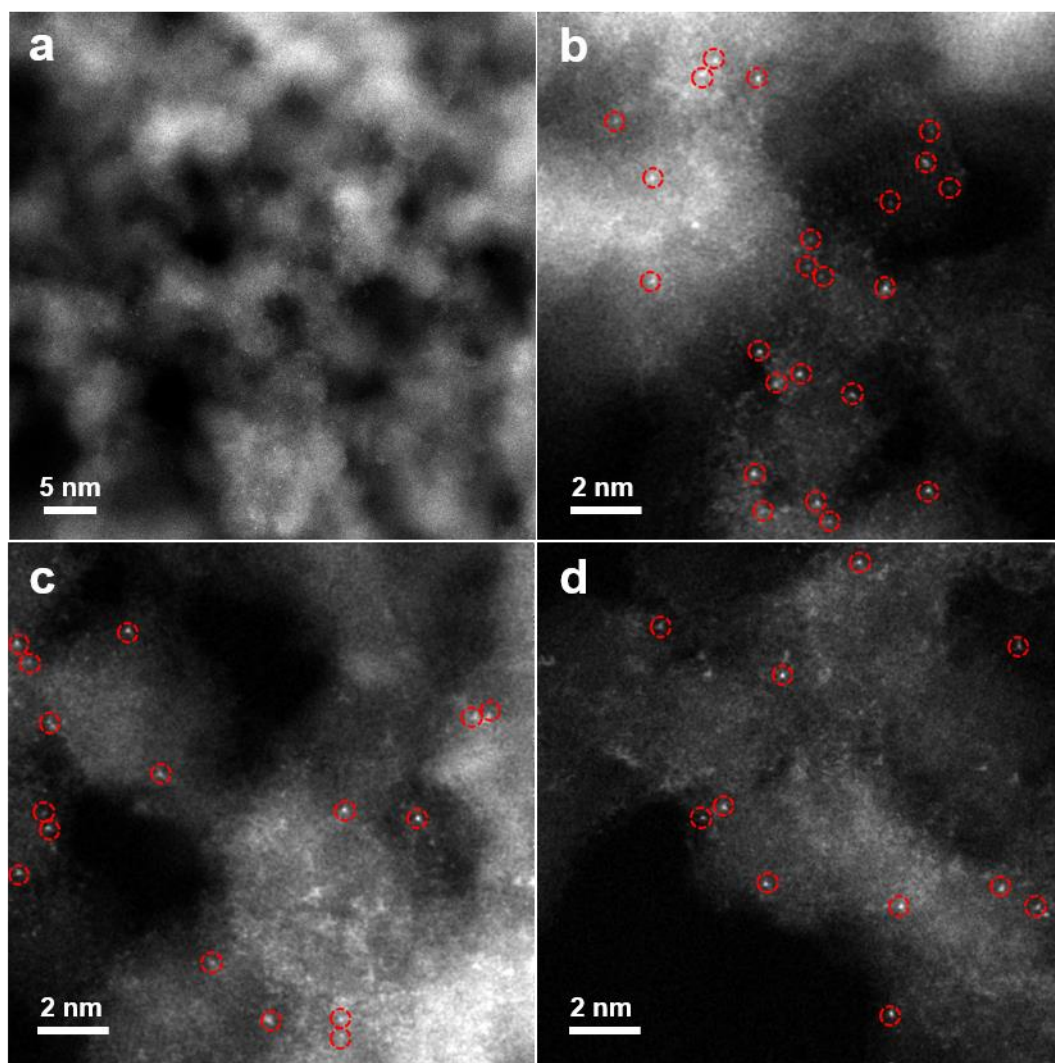
Supplementary Figure 14. Stability test over Ir₁/ND@G at 450 °C for 50h. Reaction condition: GHSV = 30000 mL · g_{cat}⁻¹ · h⁻¹, n-C₄H₁₀:H₂ = 1:1 with He balance.



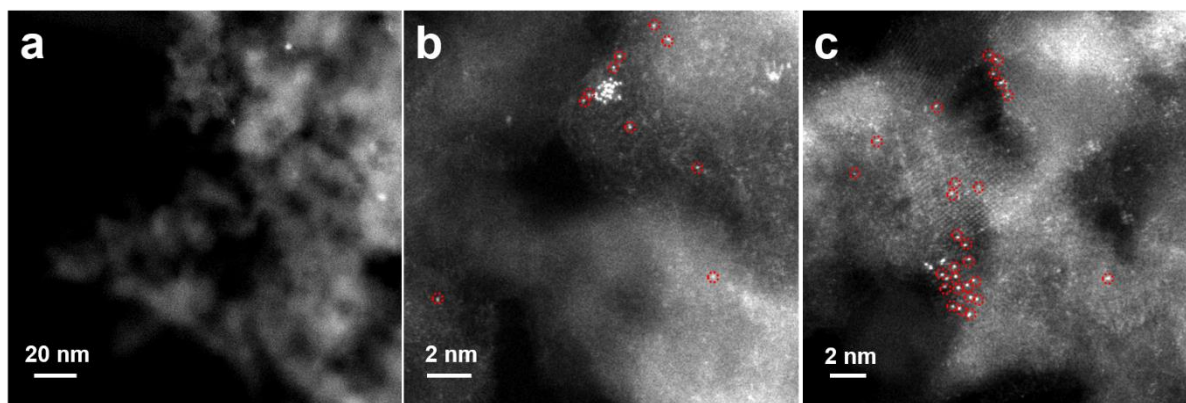
Supplementary Figure 15. Raman spectra for the fresh and spent catalysts after 10-hours BDH. Raman spectra of (a) fresh Ir₁/ND@G, (b) spent Ir₁/ND@G, (c) fresh Ir_n/ND@G, (d) spent Ir_n/ND@G, (e) fresh IrNPs/ND@G and (f) spent IrNPs/ND@G. The peak at approximately 1400 cm⁻¹ could be assigned to the disordered graphitic lattice (D1). The peak at approximately 1580 cm⁻¹ could be assigned to the well-ordered nanocrystalline graphite (G). I_{D1}/I_G (the integral areas values of D1-band and G-band) are used to qualitatively evaluate the degree of coke formation. Comparing I_{D1}/I_G ratio between fresh and spent catalysts, the lower I_{D1}/I_G ratio on spent catalysts suggests more graphitic coke deposited during BDH.



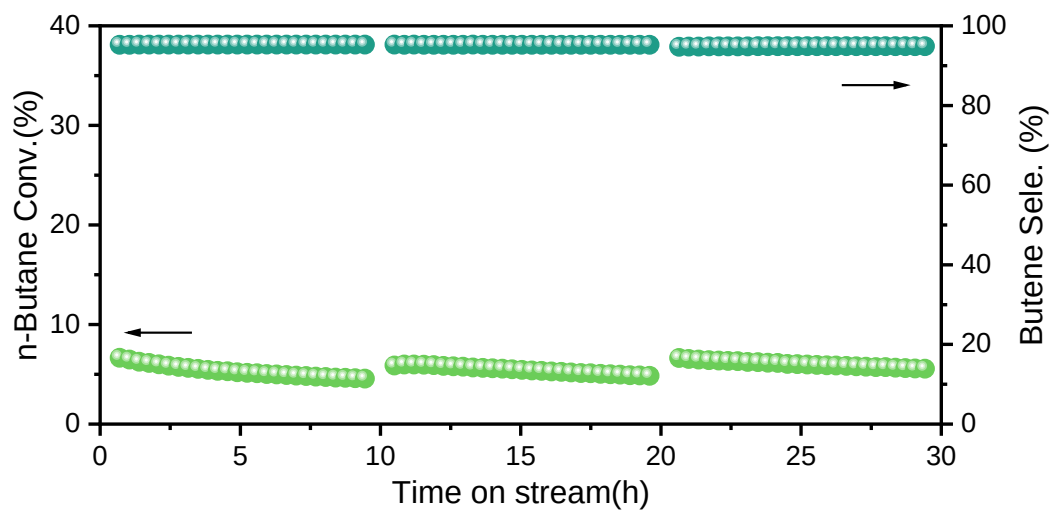
Supplementary Figure 16. HAADF-STEM images of Ir₁/ND@G after 10h BDH. (a) The image of Ir₁/ND@G after 10h BDH at low magnification. (b) and (c) The image of Ir₁/ND@G after 10h BDH at high magnification. The red circles highlighted the small Ir clusters. The yellow circles highlighted single Ir atoms.



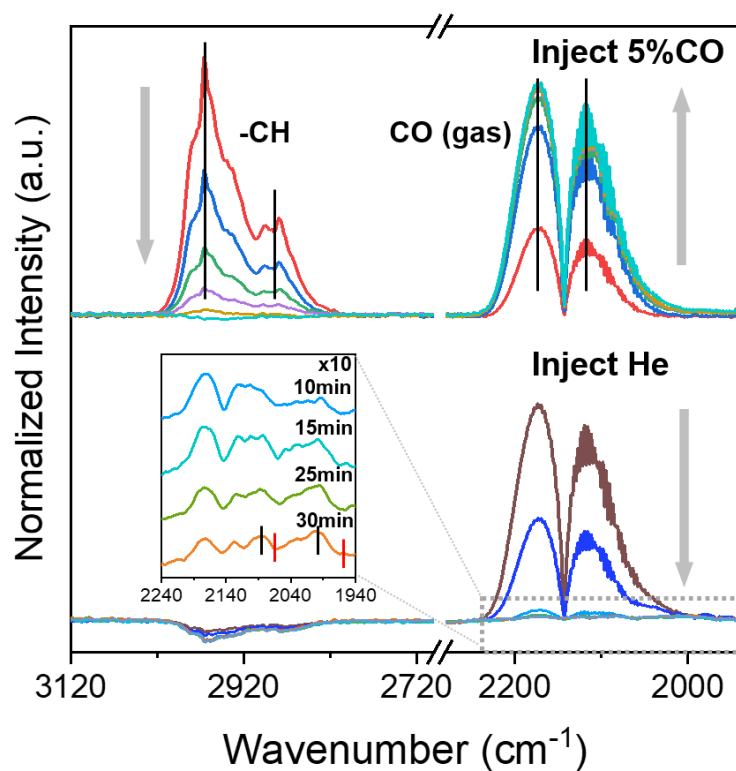
Supplementary Figure 17. HAADF-STEM images of Ir₁/ND@G after 0.5h BDH. (a) The image of Ir₁/ND@G after 0.5h BDH at low magnification. (b-d) The image of Ir₁/ND@G after 0.5h BDH at high magnification. The red circles highlighted single Ir atoms.



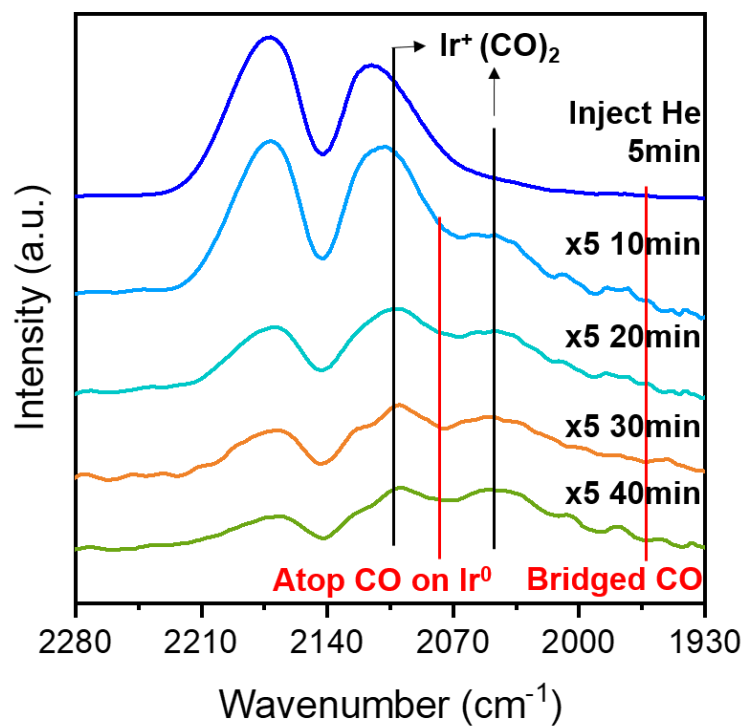
Supplementary Figure 18. HAADF-STEM images of Ir₁/ND@G after 20h BDH. (a) The image of Ir₁/ND@G after 20h BDH at low magnification. (b) and (c) The image of Ir₁/ND@G after 20h BDH at high magnification. The red circles highlighted single Ir atoms.



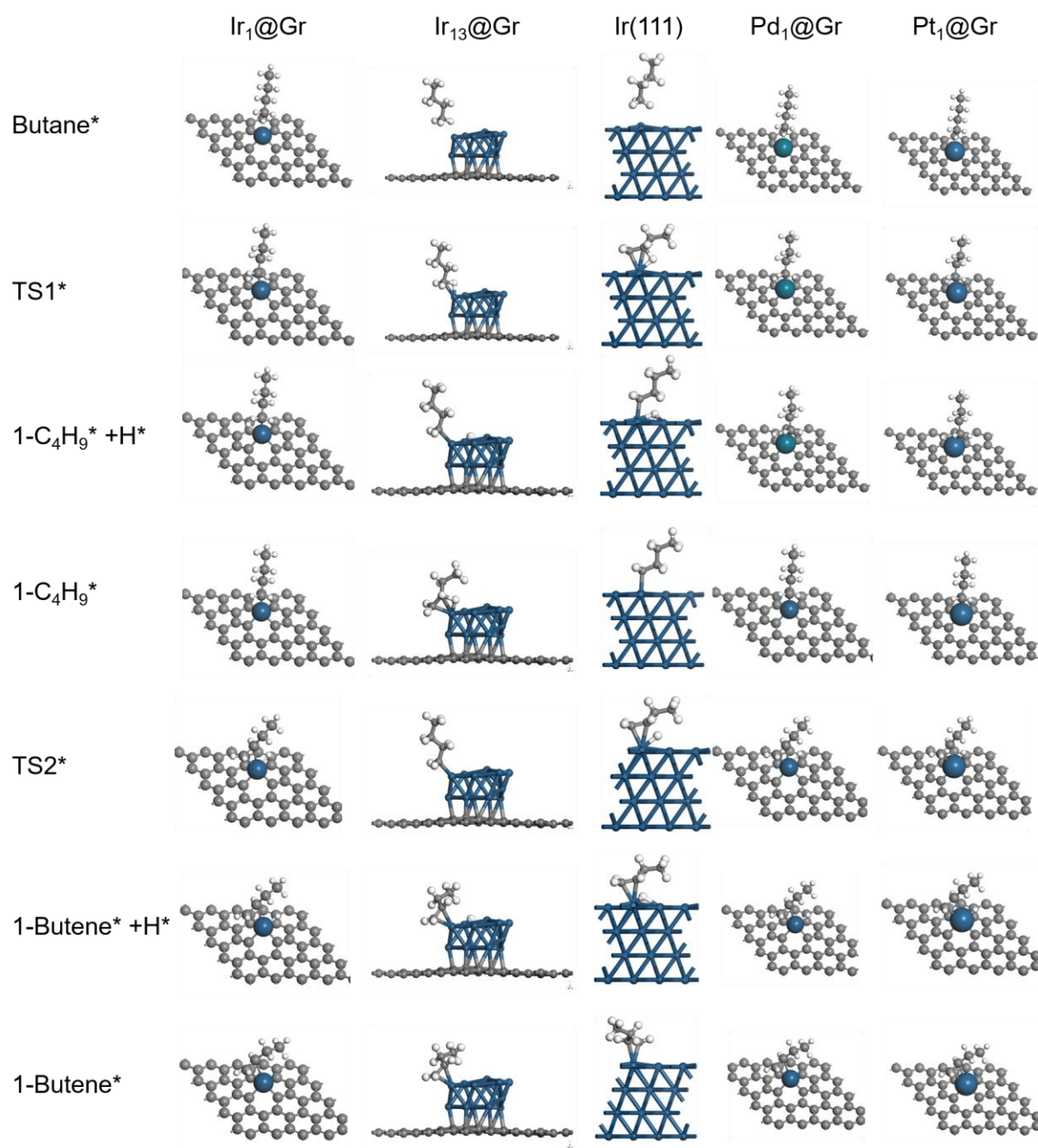
Supplementary Figure 19. The conversion of butane and selectivity of butene over Ir₁/ND@G catalyst after several regeneration cycles. Reaction condition: atmospheric pressure, GHSV = 30000 mL · g_{cat}⁻¹ · h⁻¹, C₄H₁₀:H₂ = 1:1, He balance, 450 °C.



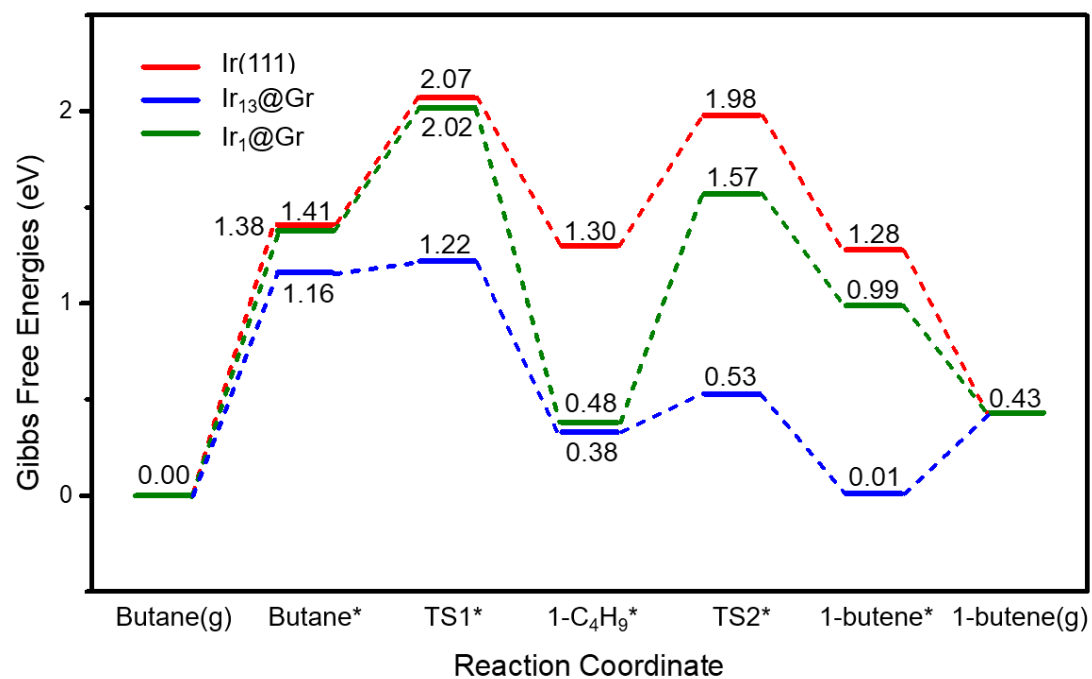
Supplementary Figure 20. The *in-situ* CO-DRIFTS of Ir₁/ND@G under reaction condition for 1h. Reaction conditions: 450 °C, 2%C₄H₁₀, 2%H₂, He balance.



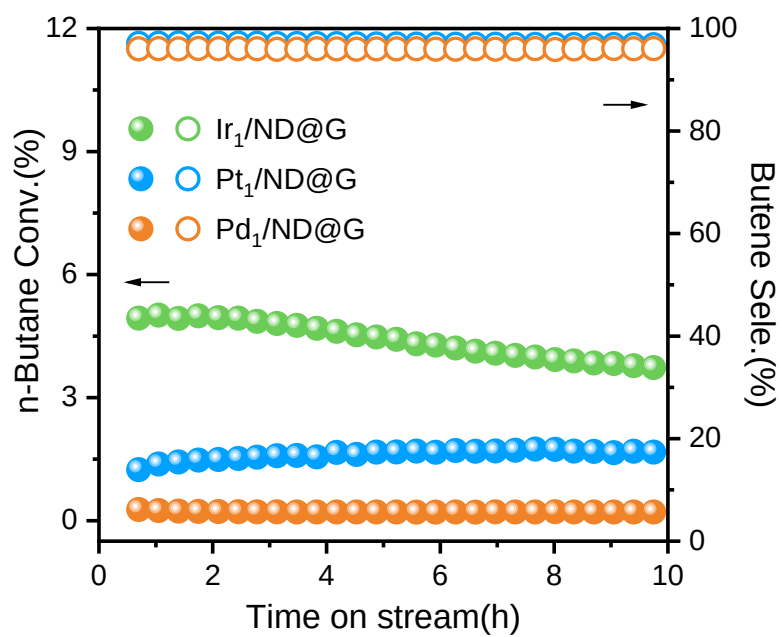
Supplementary Figure 21. The *in-situ* CO-DRIFTS of Ir₁/ND@G under reduction condition for 1h. Reduction conditions: 450 °C, 10%H₂/He.



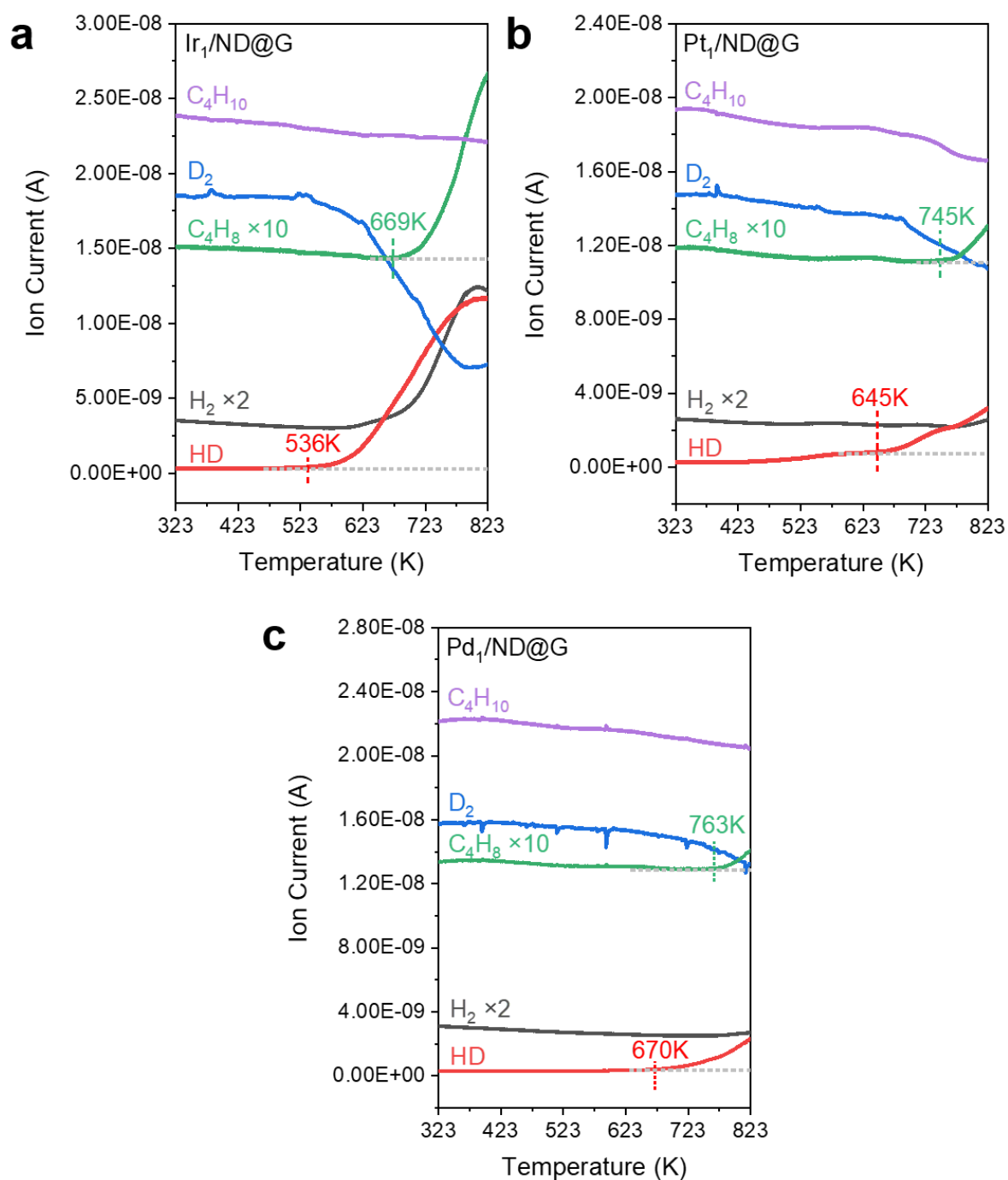
Supplementary Figure 22. The optimized configurations of surface intermediates on $\text{Ir}_1@\text{Gr}$, $\text{Ir}_{13}@\text{Gr}$, $\text{Ir}(111)$, $\text{Pd}_1@\text{Gr}$ and $\text{Pt}_1@\text{Gr}$.



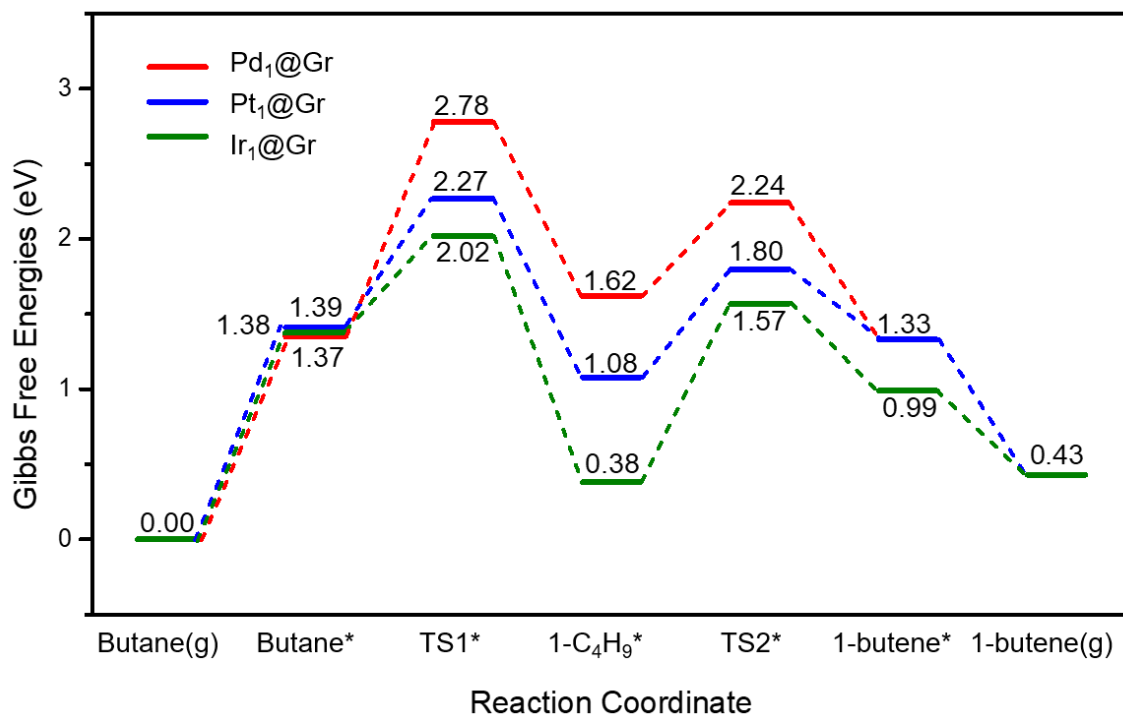
Supplementary Figure 23. Gibbs free energies profiles of BDH on Ir(111), Ir₁₃@Gr and Ir₁@Gr.



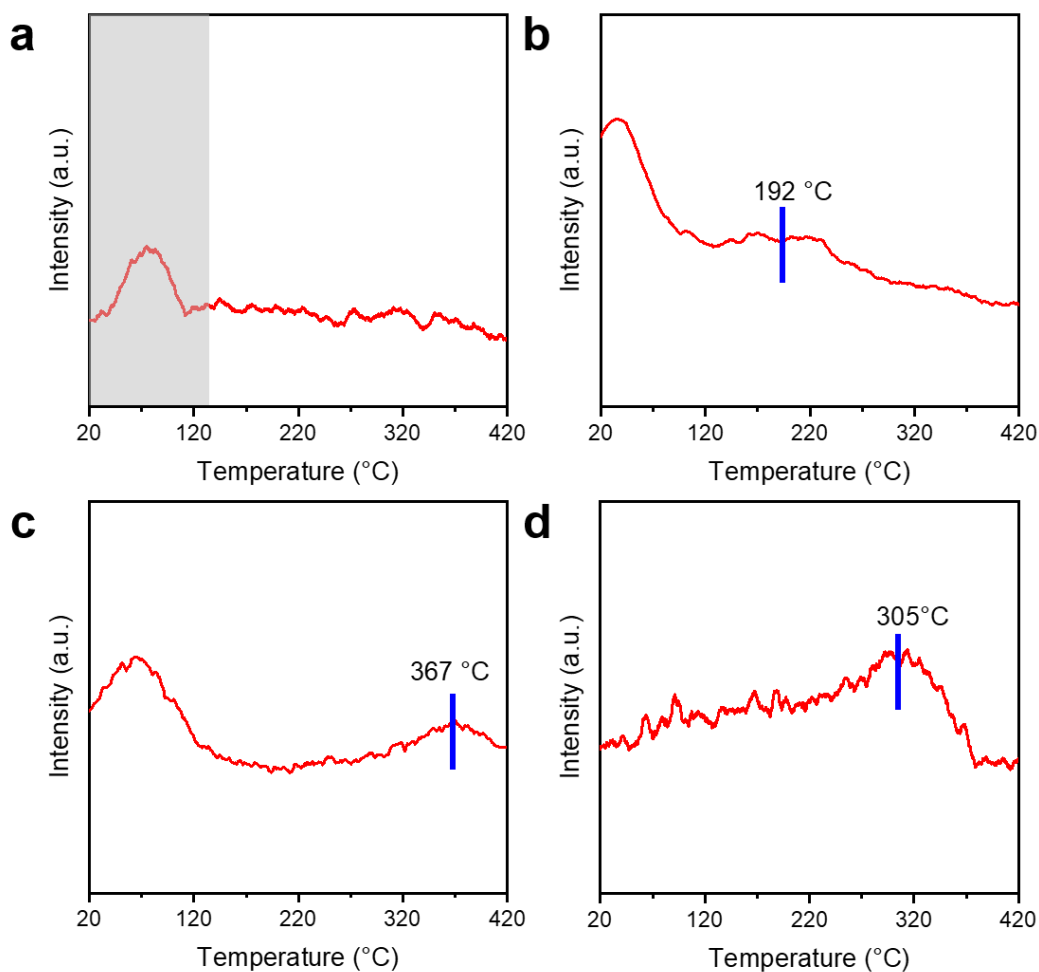
Supplementary Figure 24. n-Butane conversion and butene selectivity of Ir₁/ND@G, Pt₁/ND@G and Pd₁/ND@G.



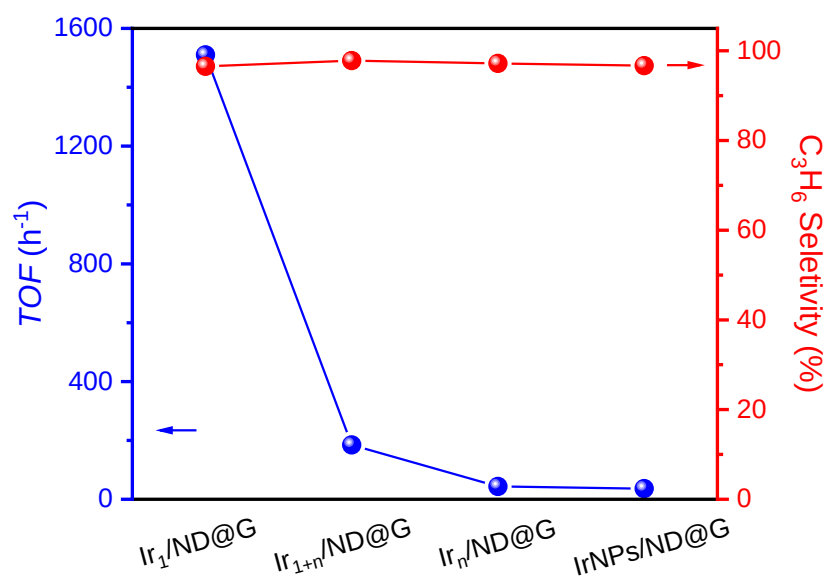
Supplementary Figure 25. TPSR profiles of the mixture of n-butane and D_2 on the different catalysts. (a) $\text{Ir}_1/\text{ND@G}$, (b) $\text{Pt}_1/\text{ND@G}$ and (c) $\text{Pd}_1/\text{ND@G}$.



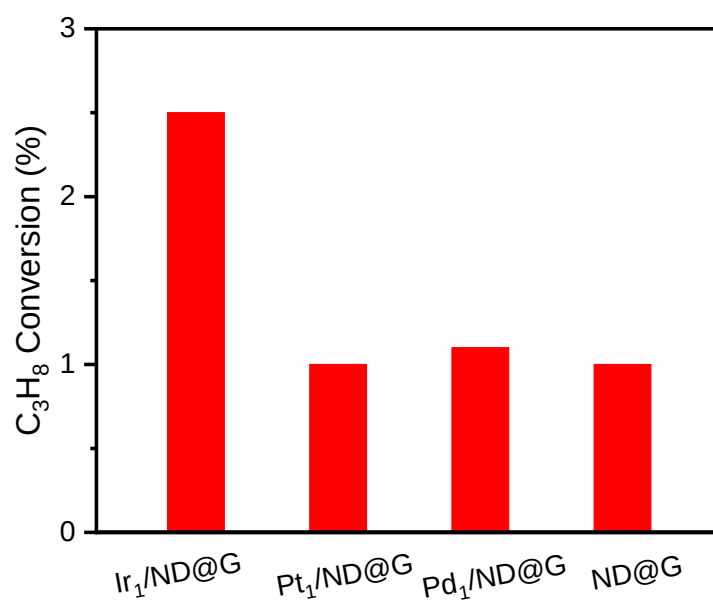
Supplementary Figure 26. Gibbs free energies profiles of BDH on Pd₁@Gr, Pt₁@Gr and Ir₁@Gr.



Supplementary Figure 27. C₄H₈-TPD profiles of the different catalysts. (a) ND@G, (b) Ir₁/ND@G, (c) Ir_n/ND@G and (d) Pt₃/ND@G. The peak at 80 °C is corresponding to physical absorption on ND@G.



Supplementary Figure 28. TOF and C_3H_6 selectivity over $\text{Ir}_1/\text{ND@G}$, $\text{Ir}_{1+n}/\text{ND@G}$, $\text{Ir}_n/\text{ND@G}$ and $\text{IrNPs}/\text{ND@G}$.



Supplementary Figure 29. C_3H_8 conversion over $Ir_1/ND@G$, $Pt_1/ND@G$, $Pd_1/ND@G$ and $ND@G$.

Supplementary Table 1. Physical structure properties of the catalysts.

Catalysts	Ir loading ^a (wt%)	S _{BET} ^b (m ² /g)	V _{pore} ^c (cm ³ /g)	D _{pore} ^d (nm)	Average particle diameter (nm)	Ir dispersion ^e (%)
ND@G	—	350.3	1.4	14.5	—	100
Ir ₁ /ND@G	0.021	353.4	1.5	13.6	—	100
Ir _{1+n} /NDG	0.43	333.0	1.5	15.3	0.53	100
Ir _n /ND@G	1.3	331.4	1.4	14.9	0.77	100
IrNP/ND@G	1.5	354.9	1.5	14.7	1.72	58.1

^adetermined by ICP-OES; ^bBET method; ^cVolume of N₂ at $p/p_0 = 0.98$; ^dN₂ desorption; ^eIr dispersion=1/average particle diameter.

Supplementary Discussion: As shown in **Supplementary Table 1**, not all samples have the dispersion of 100 %. The average particle diameter of clusters in Ir_{1+n}/ND@G or Ir_n/ND@G was less than 1 nm. Calculated by the equation (Ir dispersion=1/average particle diameter), the Ir dispersion was 100 %, indicating Ir species was atomically dispersed and fully exposed on ND@G. For IrNPs/ND@G, the average particle diameter of Ir NPs was 1.72 nm. Calculated by the equation, the Ir dispersion was 58.1 %. The difference in Ir dispersion has been considered during TOF calculations.

Supplementary Table 2. Ir L₃-edge EXAFS fitting results for as-prepared catalysts.

Samples	Shell	R (Å) ^a	C.N. ^b	σ^2 (Å ²) ^c	E ₀ shift (eV) ^d	R-factor (*10 ⁻³)
Ir foil	Ir-Ir	2.71	12	0.003	7.9	3.5
IrO ₂	Ir-O	1.98	2	0.003	11.3	5.4
Ir ₁ /ND@G	Ir-C/O	1.98	3.6	0.007	-8.2	8.0
Ir _{1+n} /ND@G	Ir-C/O	2.04	1.4	0.005	8.3	10.5
	Ir-Ir	2.67	2.6	0.010		
Ir _n /ND@G	Ir-C/O	2.03	1.2	0.005	8.2	12.3
	Ir-Ir	2.67	3.5	0.008		
IrNPs/ND@G	Ir-C/O	2.00	3.3	0.007	9.8	18.9
	Ir-Ir	2.69	3.6	0.006		

^aR is interatomic distance (the bond length between Ir central atoms and surrounding coordination atoms).

^bC.N. is the coordination number.

^c σ^2 is Debye-Waller factor (a measure of thermal and static disorder in absorber scatter distances).

^dE₀ shift is edge energy shift (the difference between the zero kinetic energy value of the sample and that of the theoretical model).

Supplementary Table 3. Catalytic performance of the catalysts for BDH.

Catalysts	Mass (mg)	Time (h)	Conversion (%)	Selectivity (%)			Rate (mol/(g _{Metal} ·h))	TOF (s ⁻¹)
				C ₄ olefins	Butene	Others		
Ir ₁ /ND@G	20	0.35	4.6	99.3	95.6	4.2	8.8	0.48
		9.75	3.7	99.4	96.2	3.8	7.1	
Ir _{1+n} /NDG	20	0.35	17.1	97.4	93.9	6.1	1.6	0.17
		9.75	12.4	98.7	94.9	5.1	1.2	
Ir _n /ND@G	20	0.35	14.7	97.1	91.7	8.3	0.59	0.025
		9.75	9.1	98.4	93.4	6.6	0.32	
IrNPs/ND@G	20	0.35	6.0	96.7	90.7	9.3	0.17	0.014
		9.75	4.3	98.5	94.3	5.7	0.12	
Pt ₁ /ND@G	50	0.35	1.2	99.5	97.1	3.9	0.20	0.0077
		9.75	1.7	99.5	96.8	3.2	0.26	
Pd ₁ /ND@G	20	0.35	0.33	100.0	95.8	4.2	0.013	0.0031
		9.75	0.21	100.0	96.1	3.9	0.0085	

Supplementary Table 4. Amount of carbon deposition on the catalysts for 10-hour BDH.

Catalysts	Carbon deposition (wt %)
Ir ₁ /ND@G	0.10
Ir _n /ND@G	2.08
IrNPs/ND@G	2.12
Pt ₃ /ND@G	0.64
ND@G	0.06

Supplementary Table 5. Catalytic performance of the catalysts for BDH.

Catalysts	Conversion (%)	Selectivity (%)			K_d (h^{-1})
		C ₄ olefins	Butene	Others	
Ir ₁ /ND@G	13.6/10.2	99.0/99.3	94.8/94.9	4.1/4.3	0.0326
Ir _{1+n} /NDG	26.8/19.0	97.3/98.7	93.7/95.3	4.0/3.4	0.0445
Ir _n /ND@G	30.4/15.9	94.7/98.5	89.5/94.2	5.3/4.2	0.0837
IrNPs/ND@G	19.3/10.5	96.3/98.0	91.0/93.0	5.3/5.0	0.0712

Supplementary Table 6. Summary of the catalytic performance of various supported metal catalysts for BDH.

Catalyst	Mass (g)	Temp. (°C)	Feed	GHVS (mL g ⁻¹ h ⁻¹)	TOS (h)	Conv. (%)	Butene Sele. (%)	TOF (s ⁻¹)	K _d (h ⁻¹)	Ref
Pd-Cu/Al	0.25	550	C ₄ H ₁₀ : H ₂ : N ₂ = 1:3:6	24000	10h	34/17	86.5	0.84	0.0922	1
Pd-Pt/Al						47/39.7	85.5	1.18	0.0297	2
PtSn/Sp-Zn-C	0.2	530	H ₂ /n-C ₄ H ₁₀ = 1.25	5400	2h	28/23	98/98	0.75	0.1319	3
Pt/B/SiO ₂	0.05	550	1% n-C ₄ H ₁₀ /Ar	2700	25h	1.6/1.5	75/75	0.034	0.0042	4
PtSn/MgAl ₂ O ₄	0.2	530	H ₂ /n-C ₄ H ₁₀ = 1.25	5400	2h	31/29	93.8/95	0.71	0.0476	5
PtSnIn(0.5)/γ-Al ₂ O ₃	0.2	530	H ₂ /n-C ₄ H ₁₀ = 1.25	5400	2h	30/28	95	-	0.0486	6
Pt-Sn/SiO ₂	0.05	550	20% C ₄ H ₁₀ /N ₂	24000	13h	33.6/26	99% (total C ₄)	9	0.0281	7
PtSn/Al ₂ O ₃	0.03	575	H ₂ :N ₂ :n-C ₄ H ₁₀ =1:1:1	72,000	10h	37.3/33.8	91.7/91.3	10.2	0.0152	8
Pt/Mg(In)(Al)O	0.005	530	H ₂ /C ₄ H ₁₀ = 2.5	124800	2h	13/7.5	97/97	5.53	0.3057	9
Pt/Sn/Zn/γ-Al ₂ O ₃	0.5	550	C ₄ H ₁₀ : N ₂ =1:1	600	6h	76.0/59.1	62.4/79.1	-	0.1307	10
Pt-TS-1	0.1	500	C ₄ H ₁₀ : H ₂ : Ar = 5: 1: 94	1500	10h	19/20	97 (total C ₄)	0.193	-	11
PtMn/SiO ₂ -Reduction	0.005	500	20% C ₄ H ₁₀ /N ₂	24000	100 h	16/13	99	-	0.002	12
PtMn/SiO ₂ -RWGS	0.055					50/18	99	-	0.015	
Pt _{1.7} Sn/ND@G	0.05	450	C ₄ H ₁₀ :H ₂ = 1:1, He balance	18000	10h	35.4/30.9	96.6/96.7	0.12	0.0223	13
Ir ₁ /ND@G	0.02	450	C ₄ H ₁₀ :H ₂ = 1:1, He balance	45000	10h	4.6/3.5	95.6/96.2	0.48	0.0241	The work
Ir _n /ND@G						14.7/9.1	91.7/94.3	0.025	0.0749	
IrNPs/ND@G						6.0/4.3	90.7/94.3	0.014	0.041	

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