Supporting information

N-coordinated Ir single atoms anchored on carbon octahedrons for catalytic oxidation of formaldehyde under ambient conditions

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Text S1.

Materials. All materials were used as received without further purification. zirconium chloride ($ZrCl_4$), 2–amino terephthalic acid ($H_2BDC\text{-}NH_2$), hydrofluoric acid solution (HF, 40%), N, N-dimethylformamide (DMF), formalin solution (38%) and glacial acetic acid were purchased from Aladdin Company. Chloroiridic acid ($H_2IrCl_6{\cdot}xH_2O$, Ir: 35 wt% in HCl) was purchased from Shanghai Macklin Biochemical Co., Ltd. Deionized water was obtained with resistivity exceeding 18 MΩ·cm.

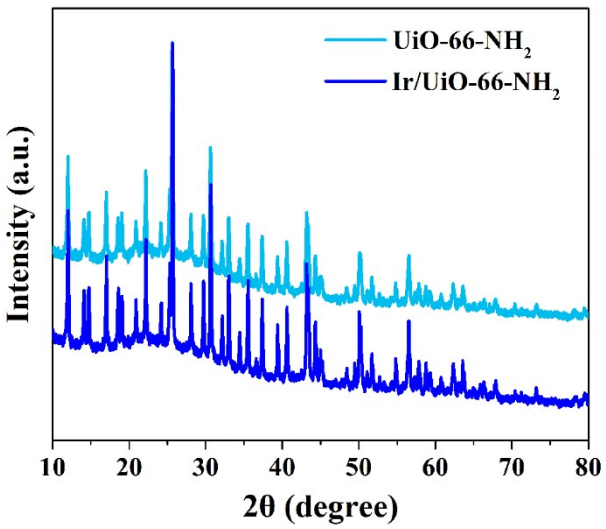

Fig. S1 XRD patterns of Ir/UiO-66-NH_2.

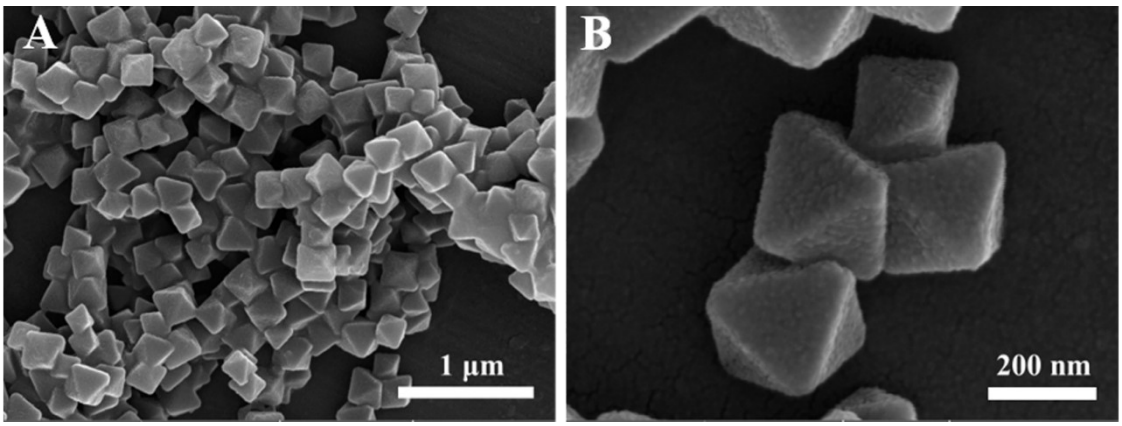

Fig. S2 SEM images of Ir/UiO-66-NH_2. Scale bars, 1 μm (A), 200 nm (B).

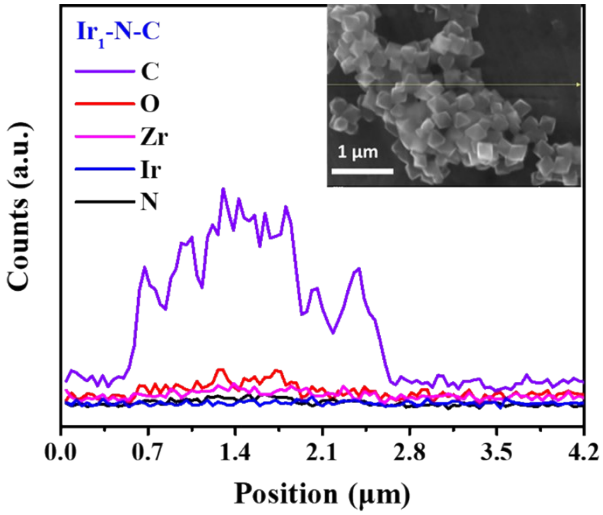

Fig. S3 EDX elemental line-scan profiles of Ir_1-N-C.

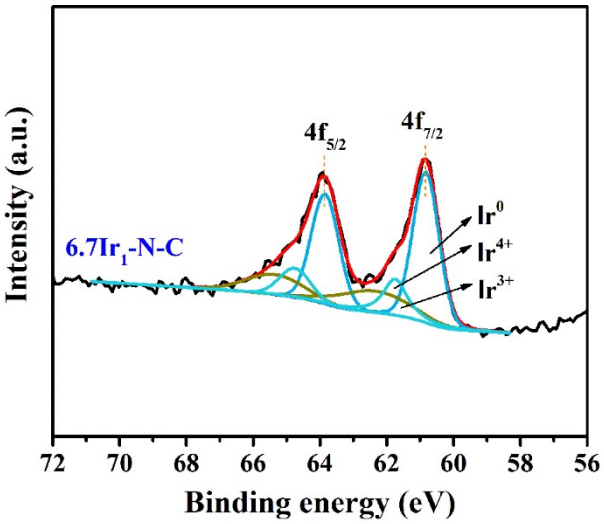

Fig. S4 High-resolution of Ir 4f XPS spectra of 6.7Ir_1-N-C.

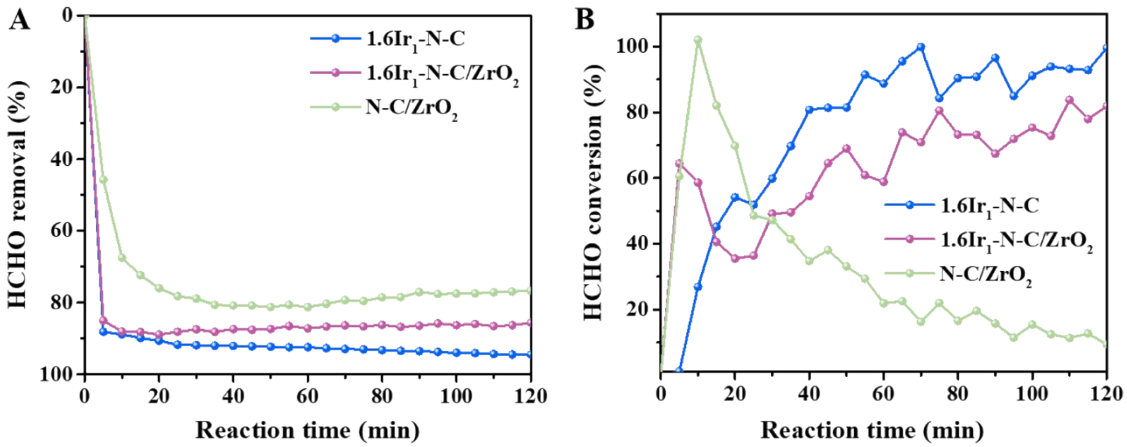

Fig. S5 (A) HCHO removal as a function of time over 1.6Ir_1-N-C, 1.6Ir_1-N-C/ZrO_2 and N-C/ZrO_2 within 120 min. (B) The corresponding HCHO conversion as a function of time. Reaction conditions: 20 °C, 20% O_2, and N_2 balance, RH = 30%, WHSV = 60,000 mL h^{-1} g_{cat}^{-1}.

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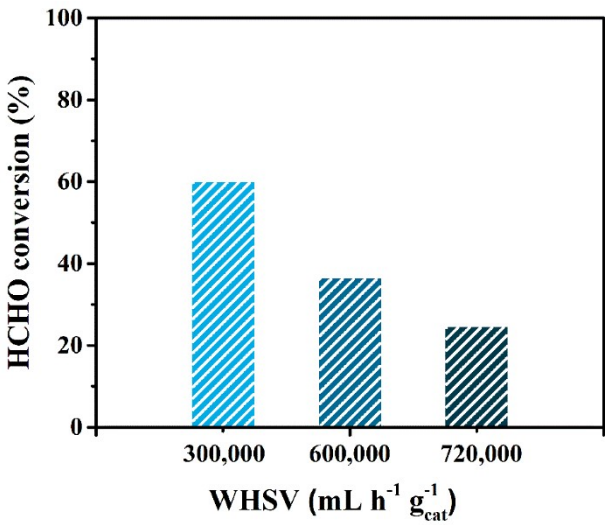


Fig. S6 HCHO conversion as a function of time over 1.6Ir_1-N-C under WHSV of 300,000, 600,000 and 720,000 mL h^{-1} g_{cat}^{-1}.

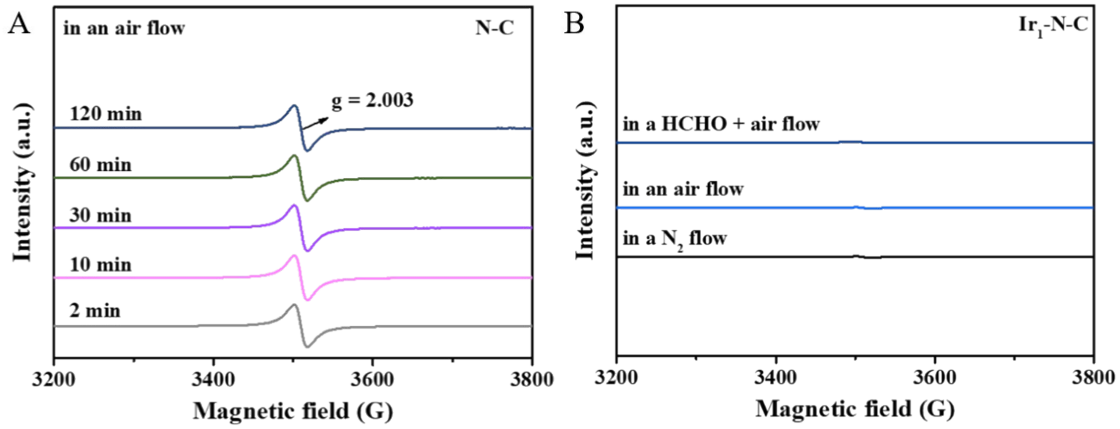


Fig. S7 (A) ESR spectra of N-C in an air flow within 120 min. (B) ESR spectra of Ir_1-N-C samples in a N_2 flow, in an air flow and in a HCHO + air flow, respectively.

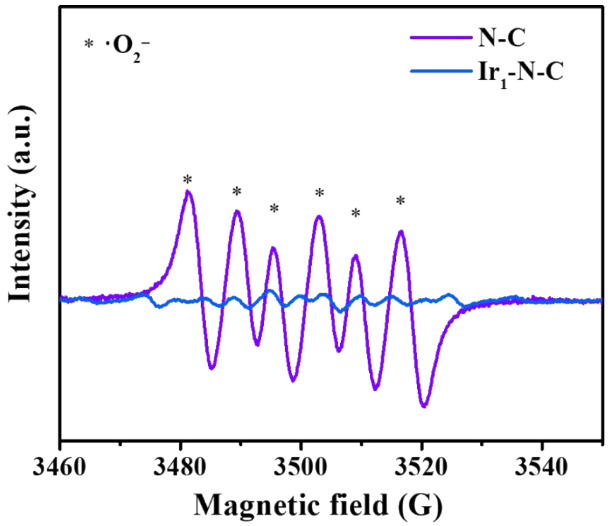


Fig. S8 DMPO spin-trapping ESR spectra of $\bullet O_2^-$ in methanol solution.

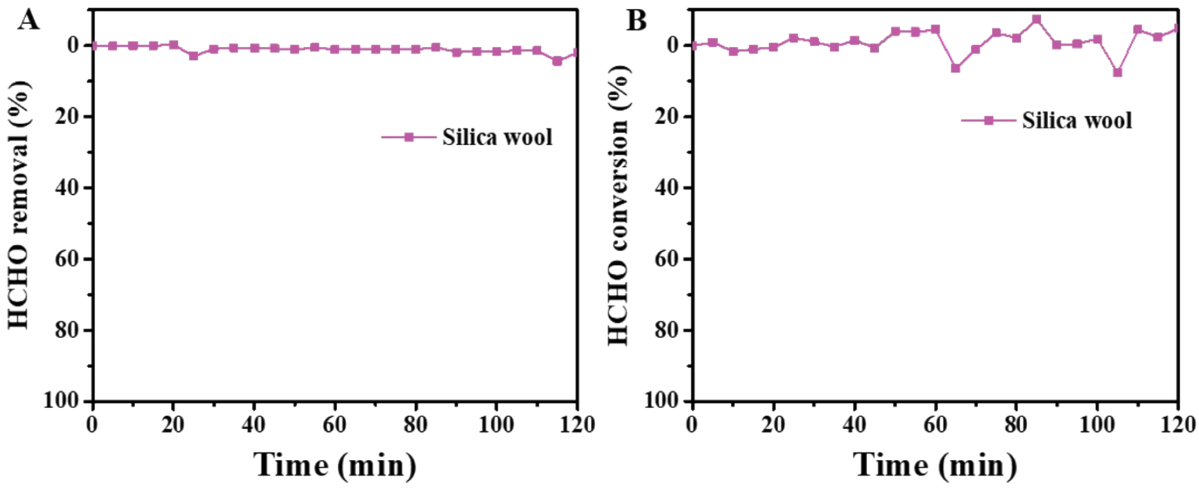


Fig. S9 (A) HCHO removal, (B) HCHO conversion as a function of time over silica wool.

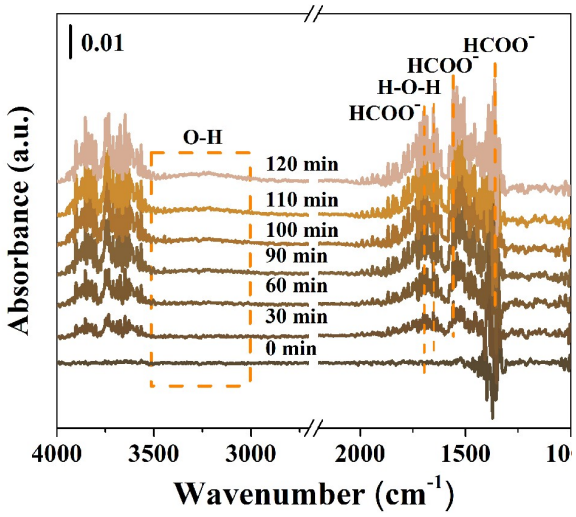


Fig. S10 In situ DRIFTS of silica wool. Reaction conditions: 50 ppm of HCHO, 30 mL min^{-1} of the total flow, 20% O_2 and N_2 as balance gas, T = 20 °C.

Table S1 Structural information and fitting parameters from Ir L3-edge EXAFS spectra of Ir_1-N-C.

Catalyst	shell	CN [a]	R(Å) [b]	σ^2 [c]	ΔE_0 [d]	R factor
Ir_1-N-C	Ir-N	4.3±0.4	2.05±0.01	0.0021	12.6±1.7	0.0171

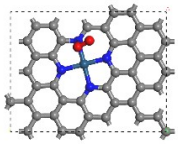
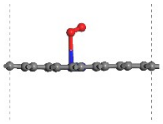
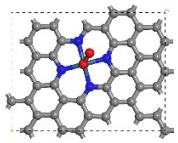
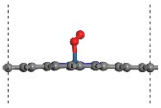
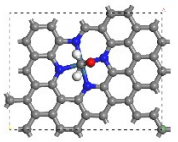
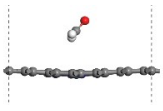
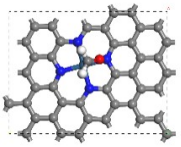
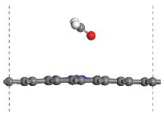
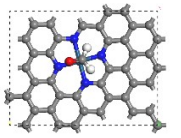
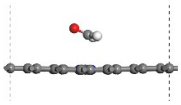
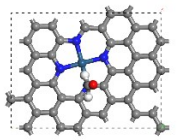
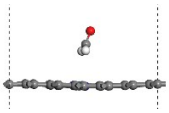
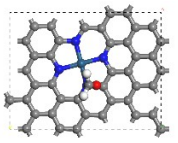
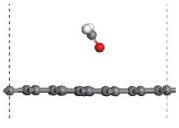
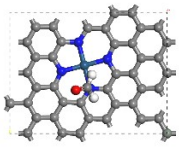
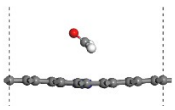
a CN: coordination numbers; b R: bond distance; c σ^2: Debye-Waller factors; d ΔE_0: the inner potential correction. R factor: goodness of fit. S_0^2 was set to 0.84, according to the experimental EXAFS fit of IrO_2 reference by fixing CN as the known crystallographic value.

Table S2 Catalytic activity of Ir_1-N-C and reported supported Ir catalysts.

Samples	Ir loading [a] (wt.%)	*T* (°C)	Reaction condition	Specific rate (mmol g_{Ir}^{-1} h^{-1})	Note
Ir_1-N-C	1.6	20	100 ppm HCHO, 20% O_2, N_2 balance, RH = 30%	401.8	This work
Ir/Al_2O_3	1.5	20	180 ppm HCHO, 20% O_2, He balance, RH = 50%	79.2	Ref.[1]
Ir/Al_2O_3	1.8	20	180 ppm HCHO, 20% O_2, He balance, RH = 50%	43.5	Ref.[1]
Na-Ir/TiO_2	0.95	25	120 ppm HCHO, 20% O_2, He balance, RH = 50%	159.0	Ref.[2]

a Determined by ICP-OES; b Calculated at 20 °C at which HCHO conversion reached 20%.

Table S3 The possible adsorption configurations of O_2 and HCHO on Ir_1-N-C, respectively.

Species	Adsorption configurations		ΔE_{ad} (eV)
	Top view	Side view	
O_2			-0.01
			-0.43
HCHO			0.34
			0.38
			0.25
			0.35
			0.37
			0.35

Notes and references

1. X. Sun, J. Lin, Y. Wang, L. Li, X. Pan, Y. Su and X. Wang, *Appl. Catal., B*, 2020, **268**, 118741.

2. Y. Li, X. Chen, C. Wang, C. Zhang, H. He, *ACS Catal*. 2018, **8**, 11377.