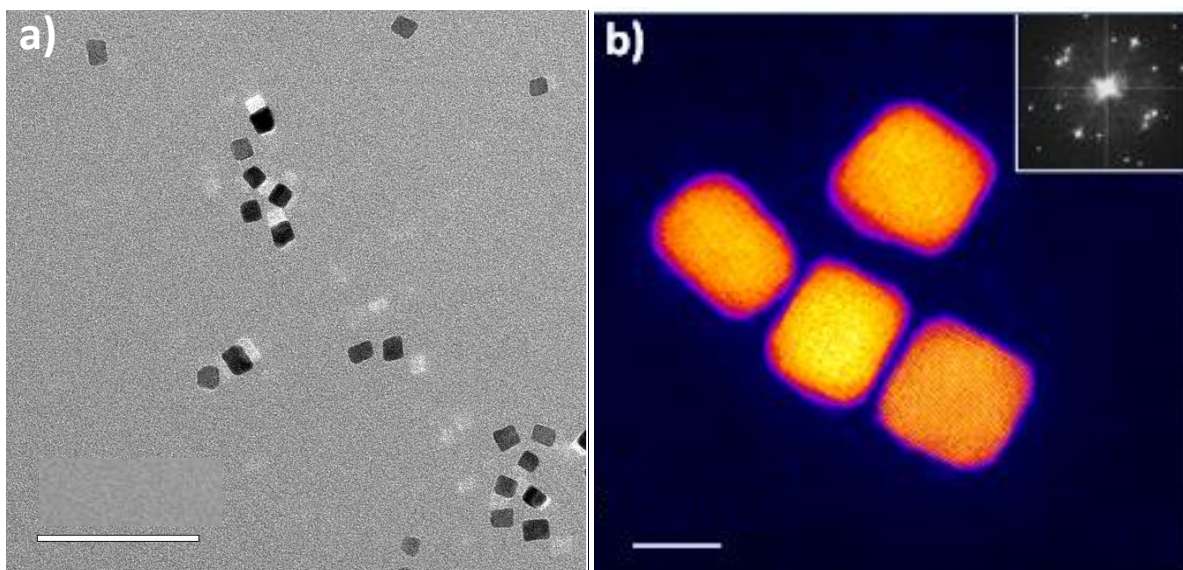
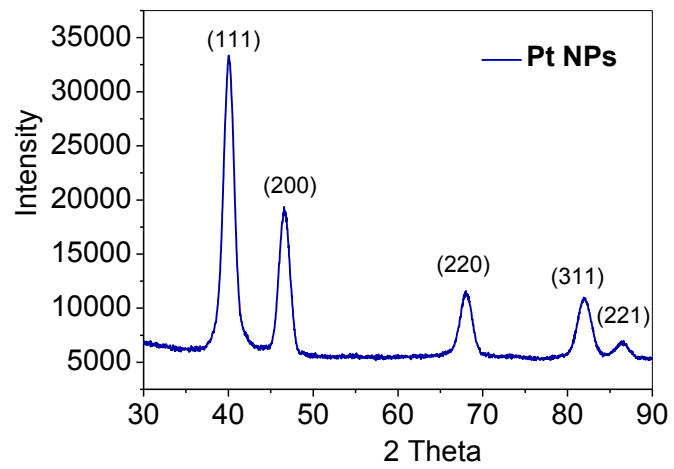


Supplementary Table 1 | Comparison between the performance of Pt/h-BNNs and literature reported catalysts.

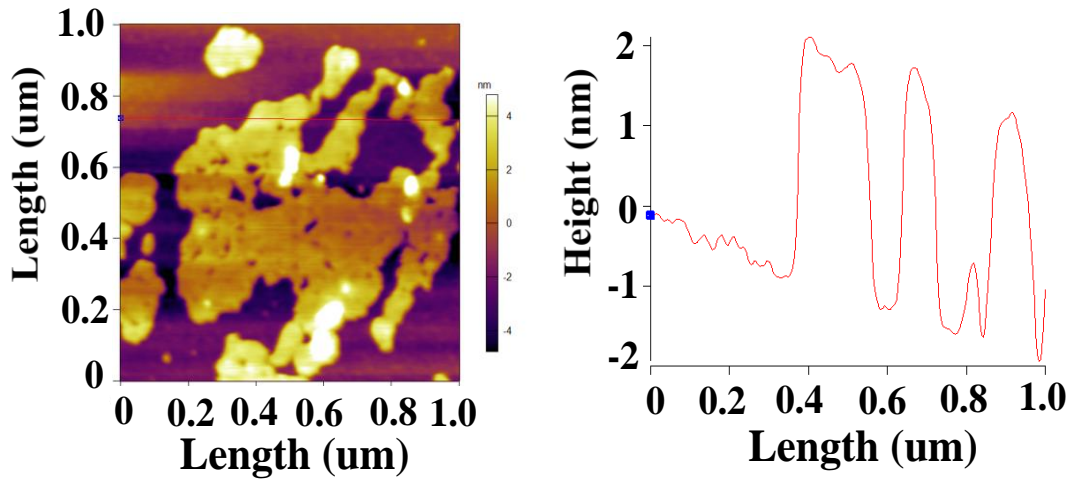
Catalysts	Pt loadings	SV(Lg ⁻¹ Pt h ⁻¹)	T ₀	T ₁₀₀	Note
Pt/SiO ₂	1 wt. %	874	150°C	200°C	[1]
Pt/CNT	1 wt. %	990	-	180°C	[2]
Pt/Al ₂ O ₃	50 g ft ⁻³	3205	80°C	150°C	[3]
Pt/Al ₂ O ₃	1.03 wt. %	-	100°C	180°C	[4]
Pt/h-BNNS	1.18 wt. %	1667	35°C	67°C	This work



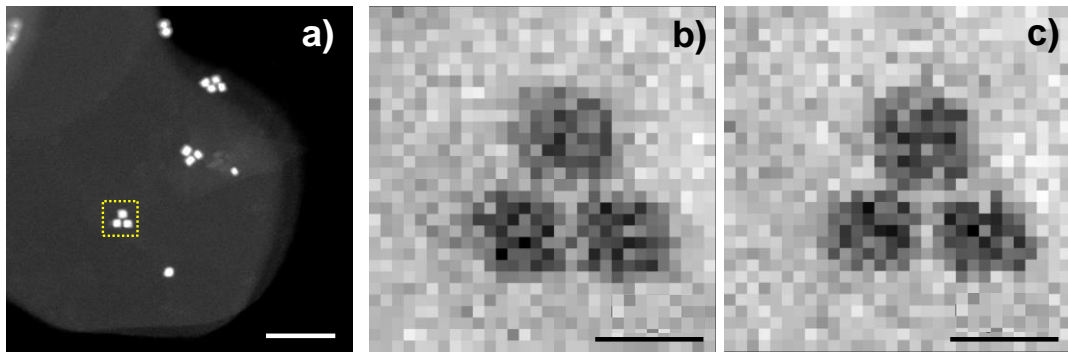
Supplementary Figure 1 | TEM (a: Scale bar, 50 nm) and STEM (b: Scale bar, 5 nm.) of Pt NPs (inset is the electron diffraction pattern of Pt NPs).



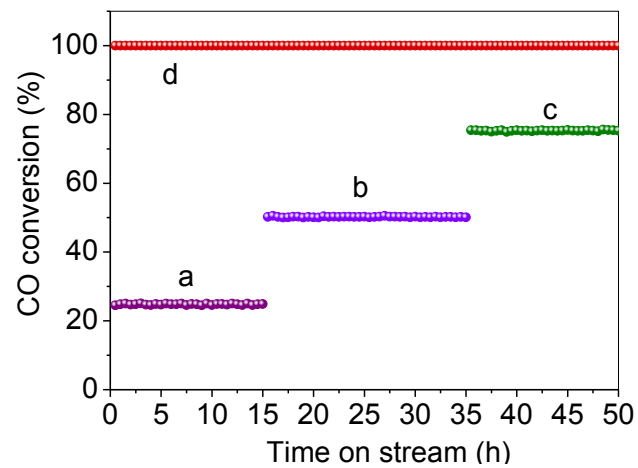
Supplementary Figure 2 | XRD of Pt NPs.



Supplementary Figure 3 | AFM image and the corresponding height profiles of *h*-BNNS

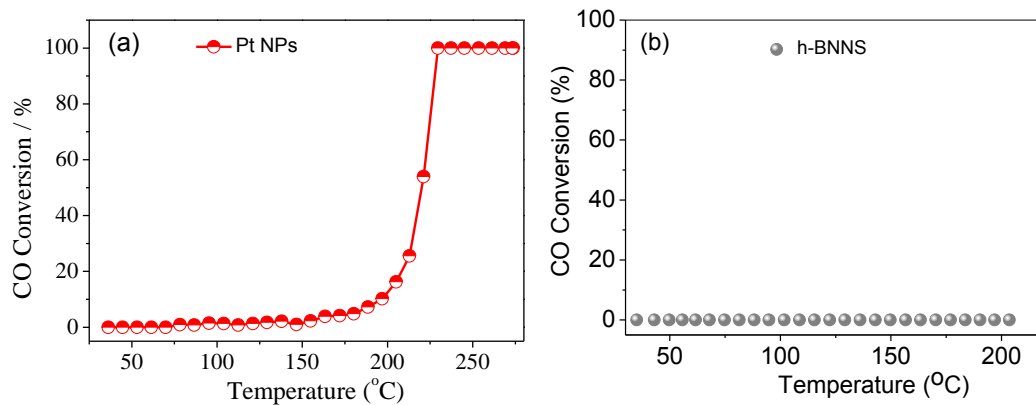


Supplementary Figure 4 | **a)** ADF image showing the distribution of bright Pt NPs in Pt/*h*-BNNS. Scale bar, 50 nm. **b)** B-K, Scale bar, 10 nm. **c)** N-K EELS elemental mapping obtained from the local area marked by a dotted square in **(a)**, Scale bar, 10 nm.

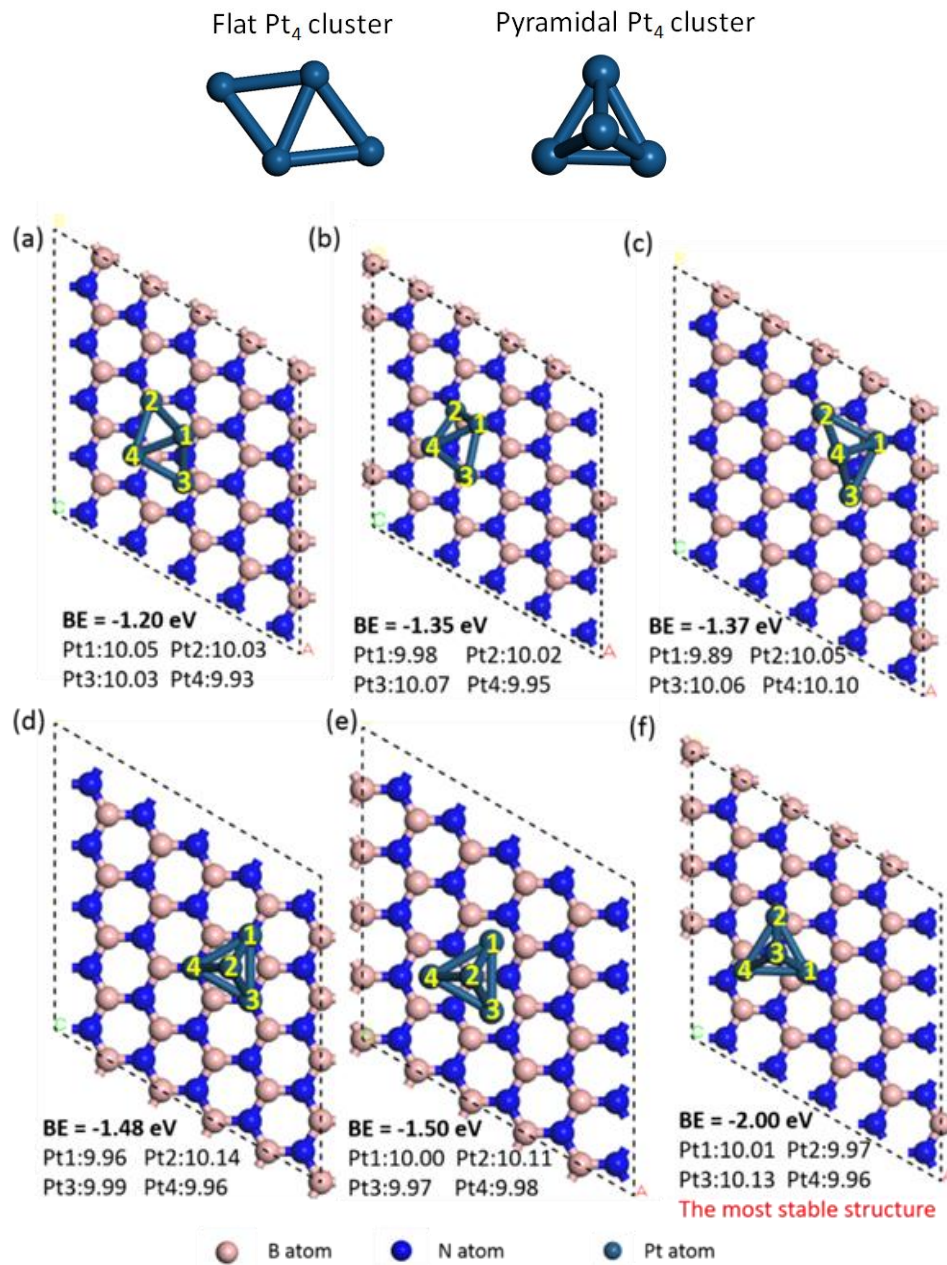


Supplementary Figure 5 | CO conversion of Pt/h-BNNS at different temperatures a: 39°C

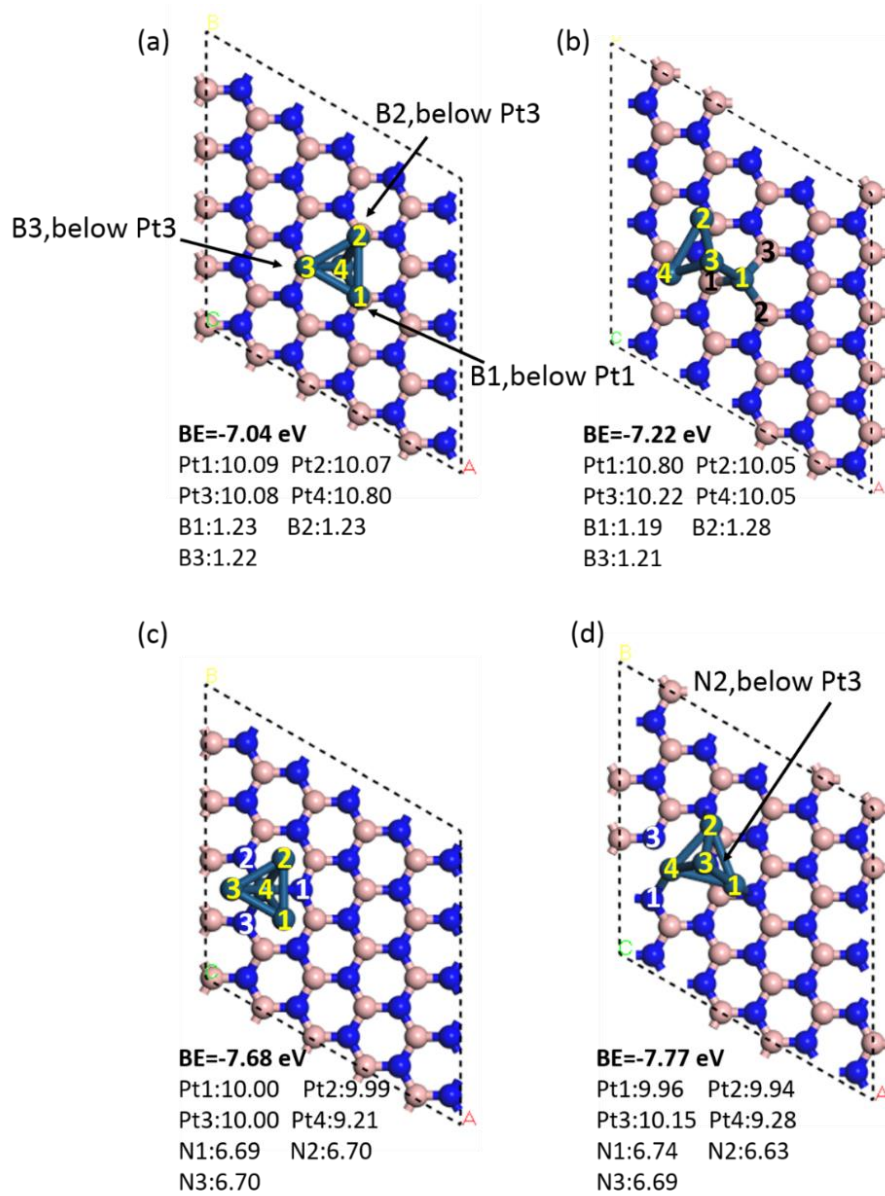
b:45 °C c:49 °C d:75 °C, m (catalyst) =30 mg Pt content= 1.18 wt.%



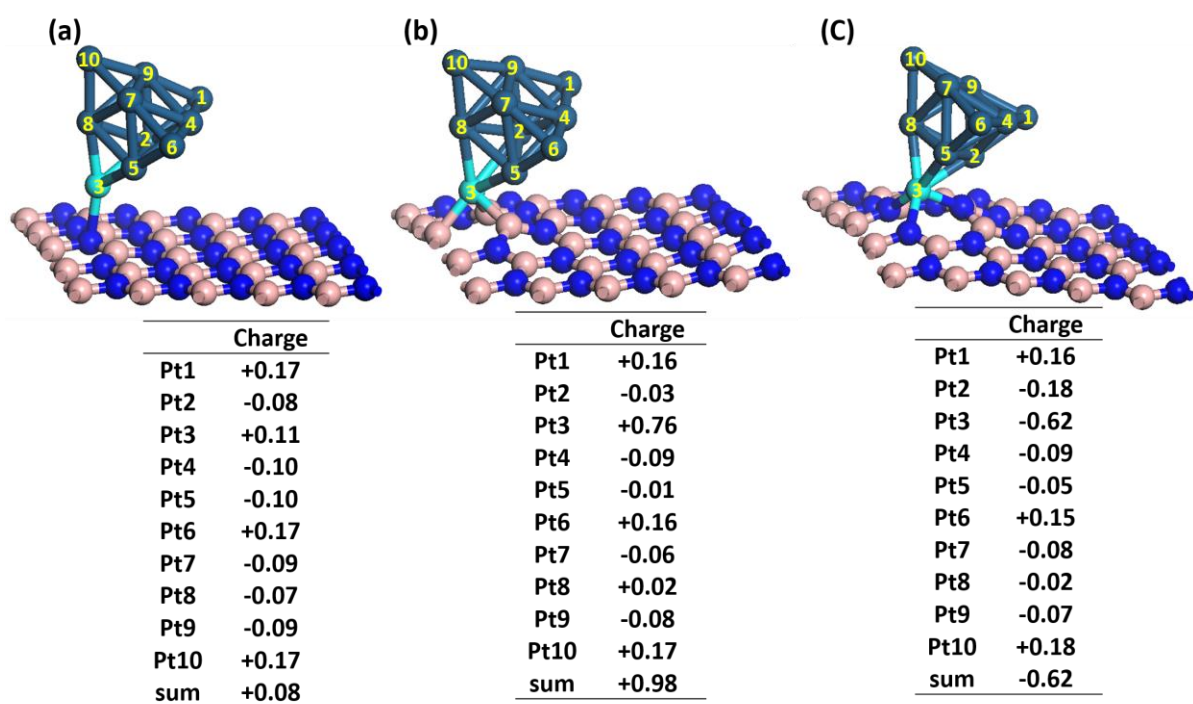
Supplementary Figure 6 | CO oxidation activity of Pt NPs (a) and *h*-BNNS (b) alone. The mass of the catalyst equals with the amount of Pt NPs and *h*-BNNS in 30 mg Pt NPs/*h*-BNNS, respectively. CO flow rate 10 mL min⁻¹;



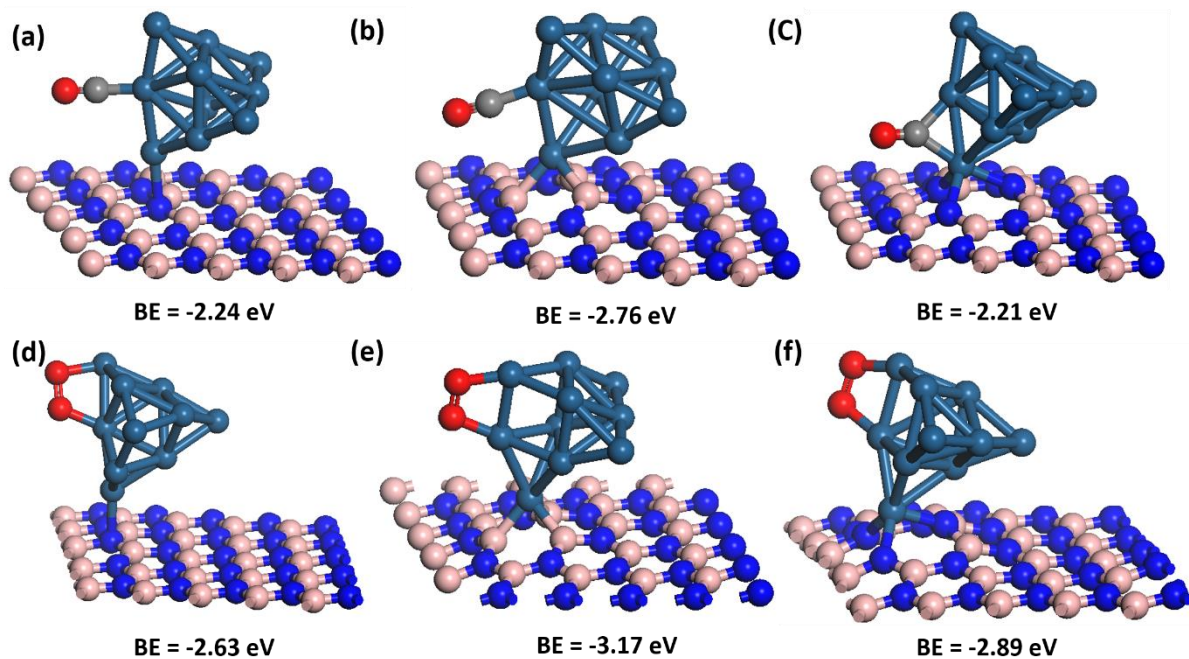
Supplementary Figure 7 | Optimized structure and corresponding Bader charge of a Pt₄ cluster on clean *h*-BNNS considered in this study. **(a-c)** Flat Pt₄ cluster on *h*-BNNS. **(d-f)** Pyramidal Pt₄ cluster on *h*-BNNS.



Supplementary Figure 8 | Optimized structure and corresponding Bader charge of a Pt₄ cluster on (a) Nv (b) Nv (c) Bv (d) Bv *h*-BNNS considered in this study

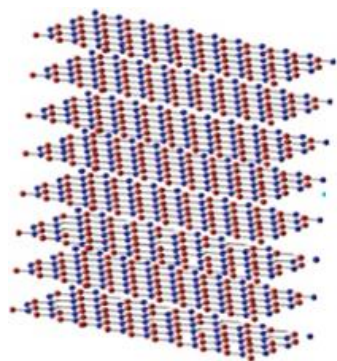
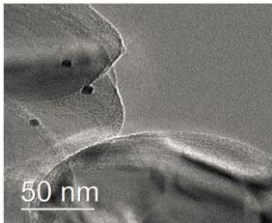


Supplementary Figure 9 | Optimized structures and Bader charges of Pt₁₀ cluster on *h*-BNNS. (a) Pt₁₀ cluster on clean, vacancy-free *h*-BNNS, (b) Pt₁₀ cluster *h*-BNNS with N_v, and (c) Pt₁₀ cluster on *h*-BNNS with B_v. The sum here is the total valence electrons for Pt₁₀ cluster. The Pt atoms with the most charge transfer on *h*-BNNS with N_v and B_v are highlighted in blue. '+' sign represents charge accumulation; while '-' sign represents charge depletion.



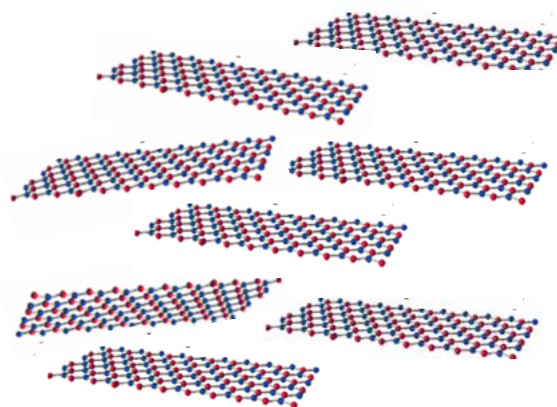
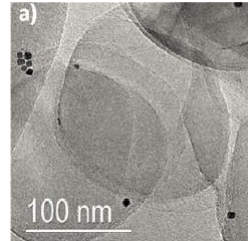
Supplementary Figure 10 | The most stable configuration of CO (a-c) and O₂ (d-f) adsorption and BEs on Pt₁₀ cluster. (a) Pt₁₀ cluster on clean, vacancy-free *h*-BNNS, (b) Pt₁₀ cluster *h*-BNNS with N_v, and (c) Pt₁₀ cluster on *h*-BNNS with B_v, (d) Pt₁₀ cluster on clean, vacancy-free *h*-BNNS, (e) Pt₁₀ cluster *h*-BNNS with N_v, and (f) Pt₁₀ cluster on *h*-BNNS with B_v.

Low surface area ($10 \text{ m}^2 \text{ g}^{-1}$)



bulk h-BN

High surface area ($278 \text{ m}^2 \text{ g}^{-1}$)

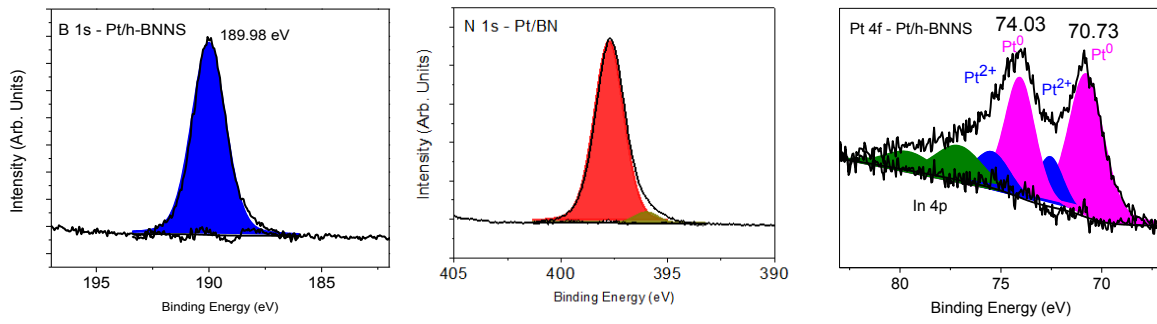


h-BNNS

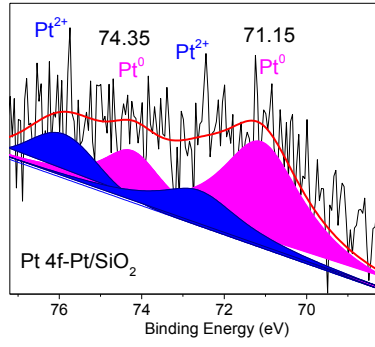
exfoliation



Supplementary Figure 11 | Pt NPs supported on bulk BN and *h*-BNNS and the schematic illustration of the exfoliation of bulk BN to *h*-BNNS.



Supplementary Figure 12 | XPS of Pt/h-BNNS



Supplementary Figure 13 | XPS of Pt/SiO₂

Supplementary Methods

Preparation of Pt/bulk BN catalysts. A certain volume of Pt NPs hexane solution was blow-dried by N₂, weighing the mass of Pt NPs. A certain amount of bulk BN (mass ratio of Pt NPs and bulk BN is 1:11) was dispersed in the mixture of ethanol (5 mL) and hexane (5 mL) under sonication. Pt NPs were re-dispersed in the hexane (5 mL) under sonication and dropped slowly into the bulk BN solution, sonicating for 1 h. The as-prepared sample was separated via centrifugation, washed with ethanol three times and dried in vacuum at 50°C for further use. Pt content in Pt/bulk BN was 1.21 wt.% by ICP-OES.

Preparation of Pt/TiO₂ catalysts. A certain volume of Pt NPs hexane solution was blow-dried by N₂, weighing the mass of Pt NPs. A certain amount of P25 (mass ratio of Pt NPs and P25 is 1:11) was dispersed in the mixture of ethanol (5 mL) and hexane (5 mL) under sonication. Pt NPs were re-dispersed in the hexane (5 mL) under sonication and dropped slowly into the P25 solution, sonicating for 1 h. The as-prepared sample was separated via centrifugation, washed with ethanol three times and dried in vacuum at 50°C for further use. Pt content in Pt/TiO₂ was 1.15 wt.% by ICP-OES.

Preparation of Pt/SiO₂ catalysts. A certain volume of Pt NPs hexane solution was blow-dried by N₂, weighing the mass of Pt NPs. A certain amount of SiO₂ (mass ratio of Pt NPs and SiO₂ is 1:12) was dispersed in the mixture of ethanol (5 mL) and hexane (5 mL) under sonication. Pt NPs were re-dispersed in the hexane (5 mL) under sonication and dropped slowly into the SiO₂ solution, sonicating for 1 h. The as-prepared sample was separated via centrifugation, washed with ethanol three times and dried in vacuum at 50°C for further use. Pt content in Pt/SiO₂ was 1.16 wt.% by ICP-OES.

Preparation of Pt/C catalysts. A certain volume of Pt NPs hexane solution was blow-dried by N₂, weighing the mass of Pt NPs. A certain amount of acetylene black (mass ratio of Pt NPs and acetylene black is 1:12) was dispersed in the mixture of ethanol (5 mL) and hexane (5 mL) under sonication. Pt NPs were re-dispersed in the hexane (5 mL) under sonication and dropped slowly into the acetylene black solution, sonicating for 1 h. The as-prepared sample was separated via centrifugation, washed with ethanol three times and dried in vacuum at 50°C for further use. Pt content in Pt/C was 1.18 wt.% by ICP-OES.

Supplementary REFERENCES

- [1] Jung, C. *et al.* Catalytic activity of Pt/SiO₂ nanocatalysts synthesized via ultrasonic spray pyrolysis process under CO oxidation, *Appl. Catal. B-Environ.*, **154–155**, 171–176 (2014).
- [2] Jardim, E. O., Gonçalves, M., Rico-Francés, S., Sepúlveda-Escribano, A., & Silvestre-Albero, J. Superior performance of multi-wall carbon nanotubes as support of Pt-based catalysts for the preferential CO oxidation: Effect of ceria addition, *Appl. Catal. B- Environ.*, **113–114**, 72–78 (2012).
- [3] Hazlett, M. J., & Epling, W. S. Spatially resolving CO and C₃H₆ oxidation reactions in a Pt/Al₂O₃ model oxidation catalyst, *Catal. Today*, **267**, 157–166 (2016).

[4] Ivanova, A.S. *et al.* Metal–support interactions in Pt/Al₂O₃ and Pd/Al₂O₃ catalysts for CO oxidation, *Appl. Catal. B-Environ.*, **97**, 57–71 (2010).