

Supporting Information

Efficient hydrogen generation from formic acid using AgPd nanoparticles immobilized on carbon nitride-functionalized SBA-15

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Calculation methods:

$$\square \quad x_a = \frac{P_{atm}V_{H2}/RT}{n_{FA}} \quad (S1)$$

Where x_a is conversion, P_{atm} is the atmospheric pressure (101325 Pa), V_{H2} is the generated volume of H_2 , R is the universal gas constant ($8.3145 \text{ m}^3 \cdot \text{Pa} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$), T is room temperature (278 K), and n_{FA} is the mole number of FA.

□

$$\square \quad TOF_{initial} = \frac{P_{atm}V_{H2}/RT}{n_{Ag+Pd}t} \quad (S2)$$

Where $TOF_{initial}$ is initial turnover frequency when x_a reaches 20%, n_{Ag+Pd} is the mole number of the Ag and Pd, and t is the reaction time when x_a reaches 20%.

□

(S1) Y. Yuan, Z. Zhao, J. Zheng, M. Yang, L. Qiu, Z. Li and Z. Zou, J. Mater. Chem., 2010, 20, 6772.

(S2) P. Chen, T. Y. Xiao, H. H. Li, J. J. Yang, Z. Wang, H. B. Yao and S. H. Yu, ACS Nano., 2012, 6, 712.

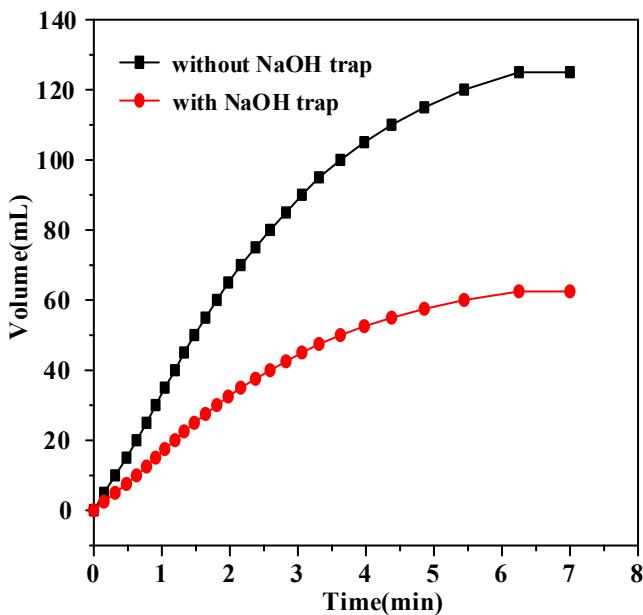


Fig. S1. Gas generation by decomposition of FA/SF vs time catalyzed by $\text{Ag}_{10}\text{Pd}_{90}/0.2\text{CND/SBA-15}$ at 323 K with and without NaOH trap (catalyst= 100 mg, $n_{\text{FA}} = 3 \text{ mmol}$, $n_{\text{SF}} = 1 \text{ mmol}$).

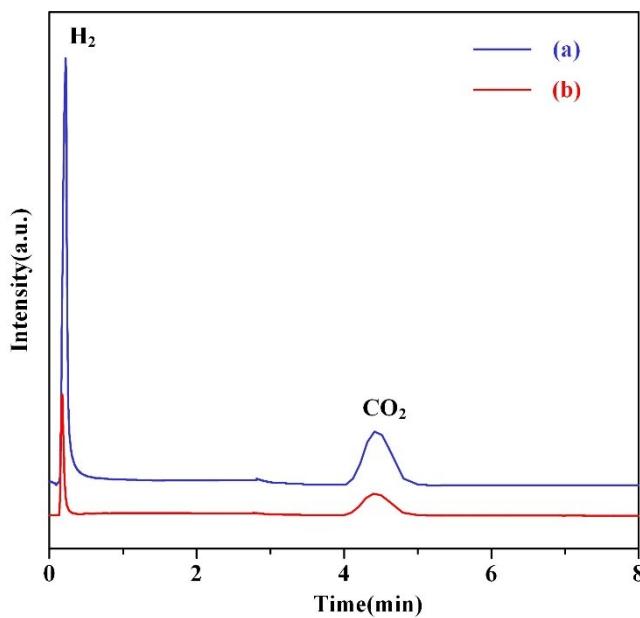


Fig. S2. GC spectrum using TCD for a) commercial mixture gas of H_2 and CO_2 , and b) evolved gas from FA/FS aqueous solution ($n_{\text{FA}} = 3 \text{ mmol}$, $n_{\text{SF}} = 1 \text{ mmol}$) over $\text{Ag}_{10}\text{Pd}_{90}/0.2\text{CND/SBA-15}$ at 323 K.

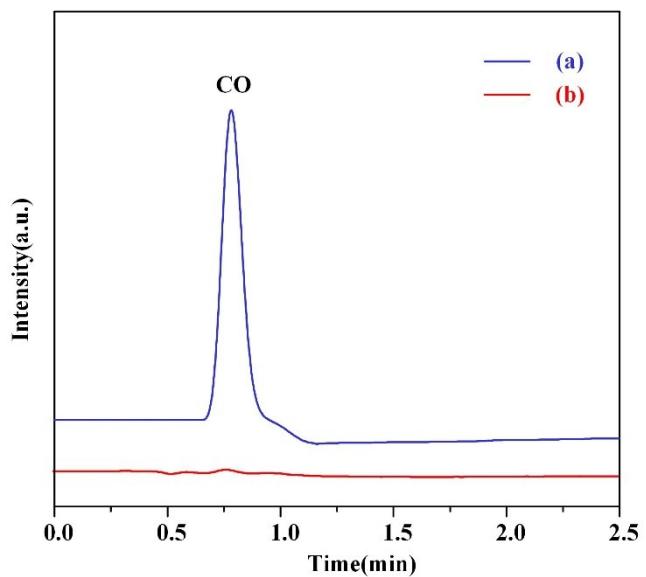


Fig. S3. GC spectrum using FID-Methanator for the a) commercial pure CO, and b) evolved gas from FA/FS aqueous solution ($n_{FA} = 3$ mmol, $n_{SF} = 1$ mmol) over $\text{Ag}_{10}\text{Pd}_{90}/0.2\text{CND/SBA-15}$ at 323 K.

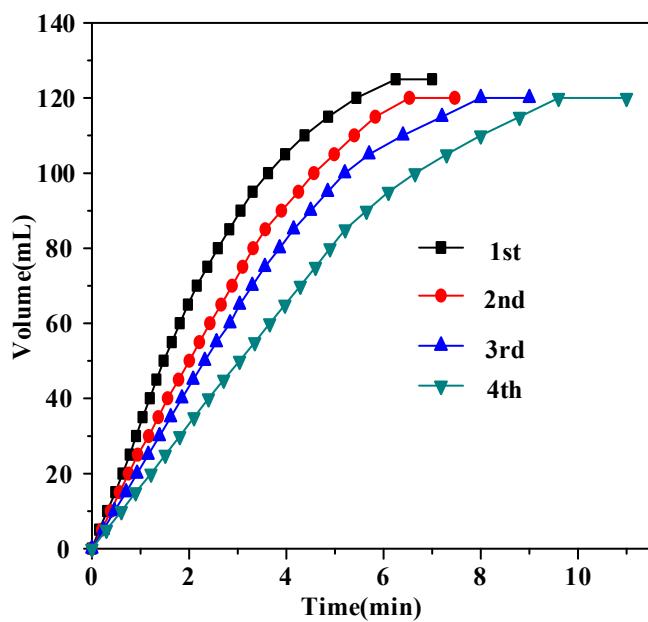


Fig. S4. Stability test on the $\text{Ag}_{10}\text{Pd}_{90}/0.2\text{CND/SBA-15}$ catalyst in the dehydrogenation of FA/SF at 323 K (catalyst= 100 mg, $n_{FA} = 3$ mmol, $n_{SF} = 1$ mmol).

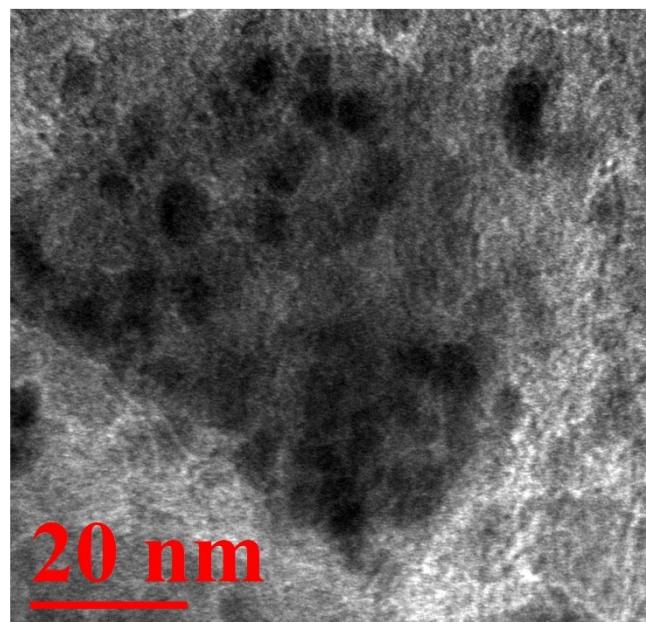


Fig. S5 TEM image of $\text{Ag}_{10}\text{Pd}_{90}/0.2\text{CND/SBA-15}$ after four runs

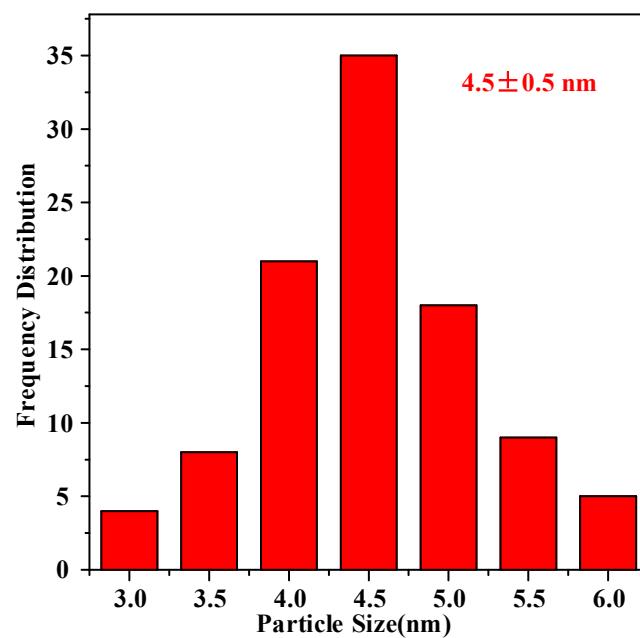


Fig. S6. $\text{Ag}_{10}\text{Pd}_{90}$ nanoparticle size distribution of $\text{Ag}_{10}\text{Pd}_{90}/0.2\text{CND/SBA-15}$, Mean size = $4.5 \pm 0.5 \text{ nm}$.

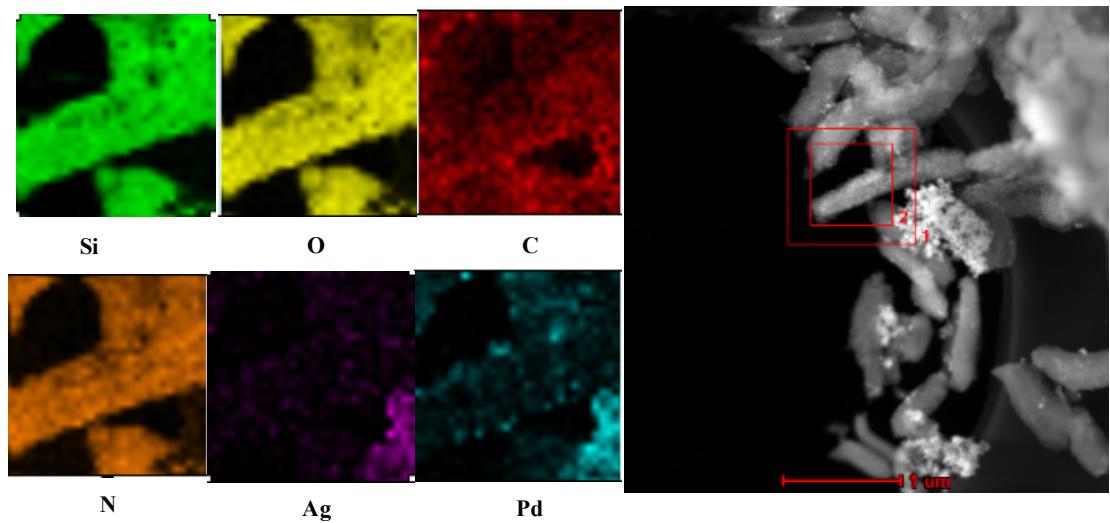


Fig.S7. The corresponding elemental mapping for Si, O, C, N, Ag and Pd elements and HAADF-STEM image of $\text{Ag}_{10}\text{Pd}_{90}/0.2\text{CND/SBA-15}$.

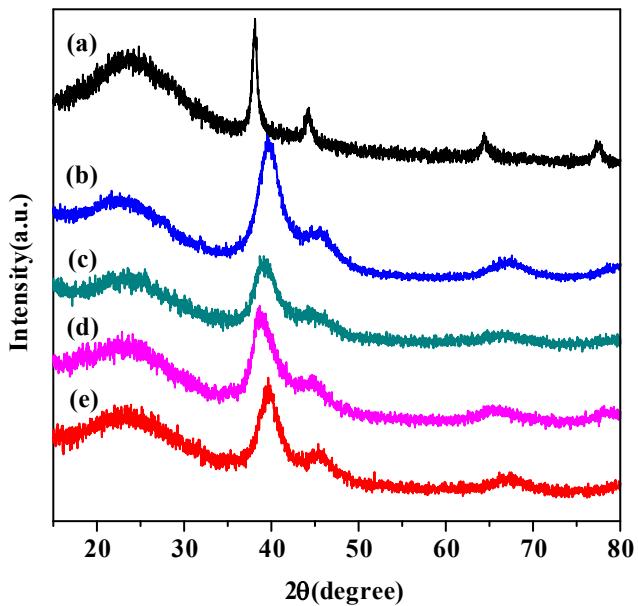


Fig.S8. Wide-angle XRD patterns for (a) $\text{Ag}/0.2\text{CND/SBA-15}$, (b) $\text{Ag}_{10}\text{Pd}_{90}/0.2\text{CND/SBA-15}$, (c) $\text{Ag}_{20}\text{Pd}_{80}/0.2\text{CND/SBA-15}$, (d) $\text{Ag}_{30}\text{Pd}_{70}/0.2\text{CND/SBA-15}$ and (e) $\text{Pd}/0.2\text{CND/SBA-15}$.

Table S1. ICP-AES results and specific surface area of AgPd/mCND/SBA-15 catalysts

Catalyst	Ag (wt%)	Pd (wt%)	Ag-Pd initial composition	Ag-Pd final composition	Final Metals/Catalyst (mmol/100 mg)	$S_{BET}(m^2 g^{-1})$
Ag ₁₀ Pd ₉₀ /0.2C ND/SBA-15	0.93	9.26	10:90	9:91	0.096	548
Ag ₂₀ Pd ₈₀ /0.2C ND/SBA-15	1.95	8.16	20:80	19:81	0.095	543
Ag ₃₀ Pd ₇₀ /0.2C ND/SBA-15	3.25	6.78	30:70	32:68	0.094	541
Ag ₁₀ Pd ₉₀ /SBA- 15	1.09	8.68	10:90	11:89	0.092	565
Ag ₁₀ Pd ₉₀ /0.1C ND/SBA-15	0.81	9.17	10:90	8:92	0.094	555
Ag ₁₀ Pd ₉₀ /0.3C ND/SBA-15	1.21	8.68	10:90	12:88	0.093	542
Ag ₁₀ Pd ₉₀ /0.4C ND/SBA-15	1.14	9.06	10:90	11:89	0.096	535

Table. S2 Comparison of AgPd/mCND/SBA-15 catalyzed decomposition of FA/SF solution (n_{FA} =3 mmol, n_{SF} = 1 mmol)

Catalyst	T/ K	Conversion/ %	TOF _{initial} / h ⁻¹	H ₂ /CO ₂ ratio
Ag ₁₀ Pd ₉₀ /0.2CND/SBA-15	323	100	893	1:1
Ag ₂₀ Pd ₈₀ /0.2CND/SBA-15	323	96.0	609	1:1
Ag ₃₀ Pd ₇₀ /0.2CND/SBA-15	323	92.0	478	1:1
Ag ₁₀ Pd ₉₀ /SBA-15	323	92.0	304	1:1
Ag ₁₀ Pd ₉₀ /0.1CND/SBA-15	323	96.0	638	1:1
Ag ₁₀ Pd ₉₀ /0.3CND/SBA-15	323	96.0	558	1:1
Ag ₁₀ Pd ₉₀ /0.4CND/SBA-15	323	92.0	343	1:1

TOF_{initial} is calculated when x_a reaches 20%.