

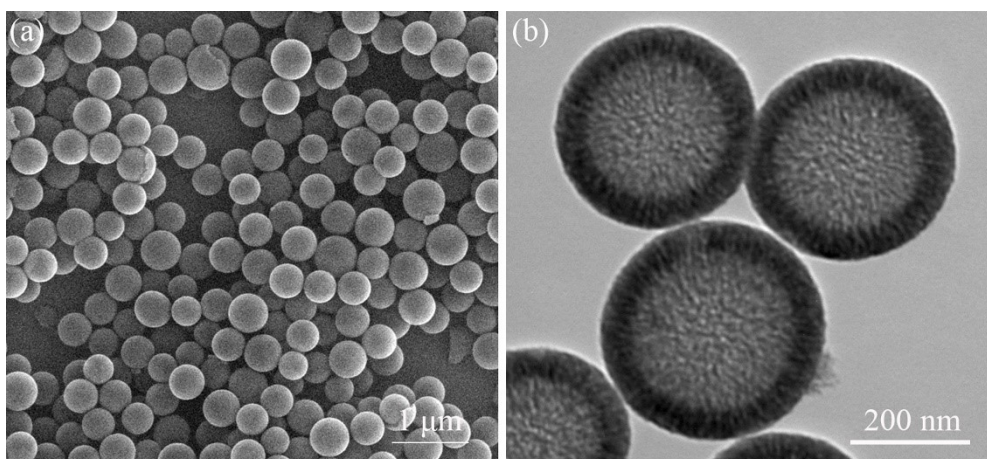
## Supplementary Information

### **Improved Hydrogen Evolution Performance by Engineering Bimetallic AuPd Loaded on Amino and Nitrogen Functionalized Mesoporous Hollow Carbon Spheres**

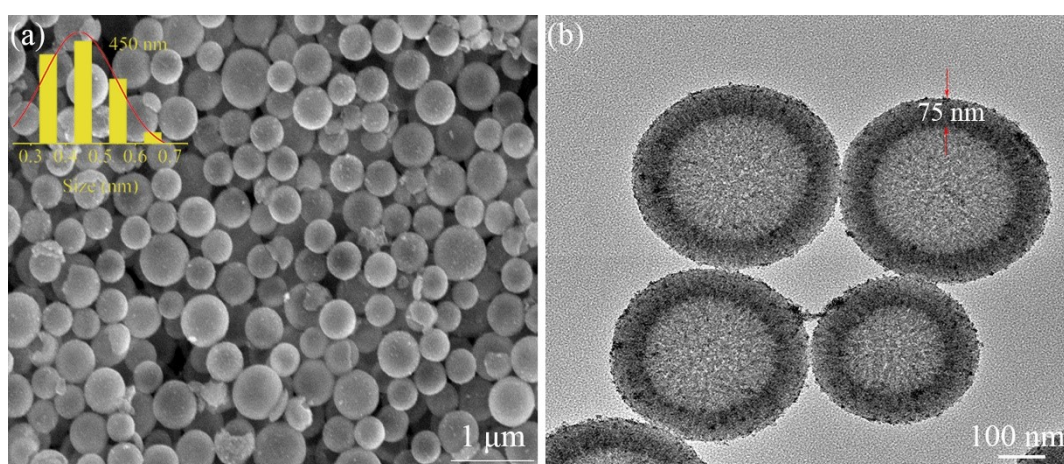
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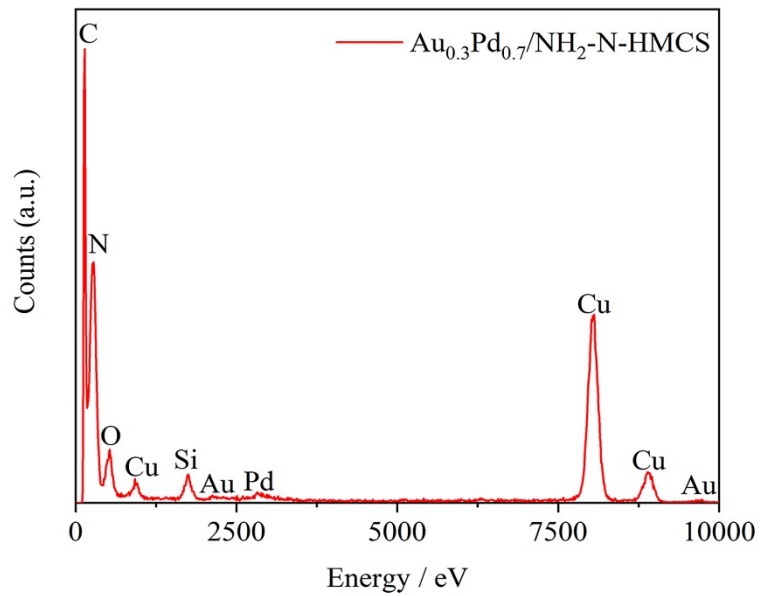
<sup>b</sup>Advanced Institute of Materials Science, Changchun University of Technology, Changchun, 130012, China. E-mail: yuechi@ccut.edu.cn



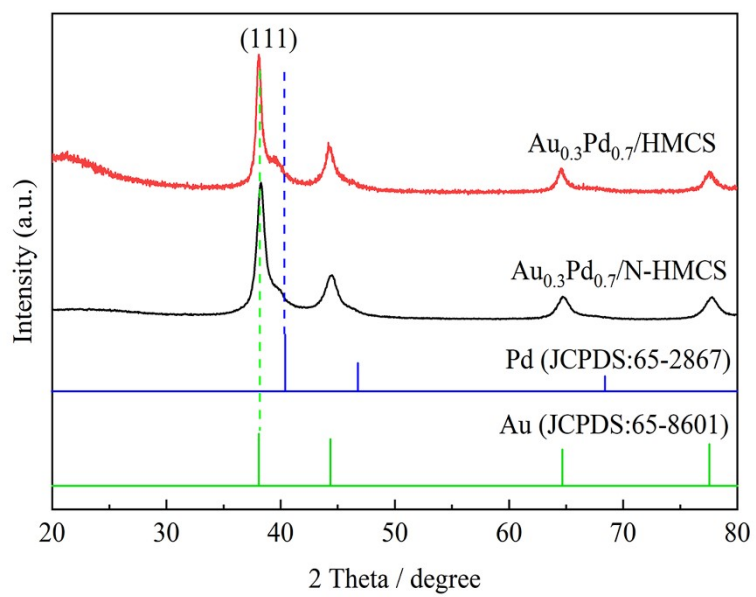
**Fig. S1.** SEM image (a) and TEM image of HMCS (b).



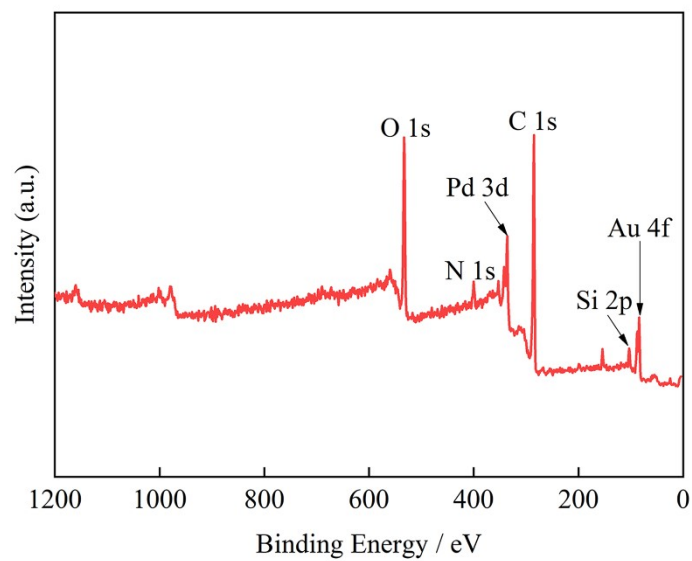
**Fig. S2.** SEM image(a) and TEM image(b) of Au<sub>0.3</sub>Pd<sub>0.7</sub>/NH<sub>2</sub>-N-HMCS.



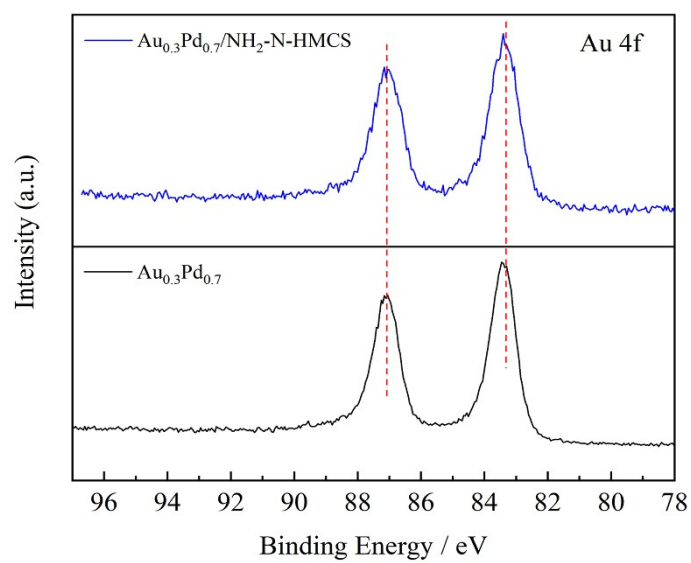
**Fig. S3.** EDX spectrum of  $\text{Au}_{0.3}\text{Pd}_{0.7}/\text{NH}_2\text{-N-HMCS}$



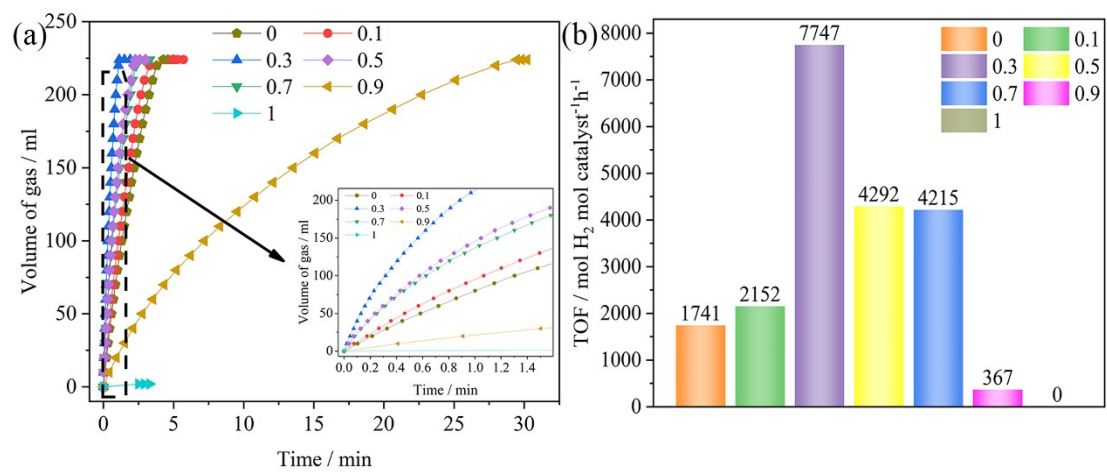
**Fig. S4.** The XRD patterns of  $\text{Au}_{0.3}\text{Pd}_{0.7}/\text{HMCS}$  and  $\text{Au}_{0.3}\text{Pd}_{0.7}/\text{N-HMCS}$ .



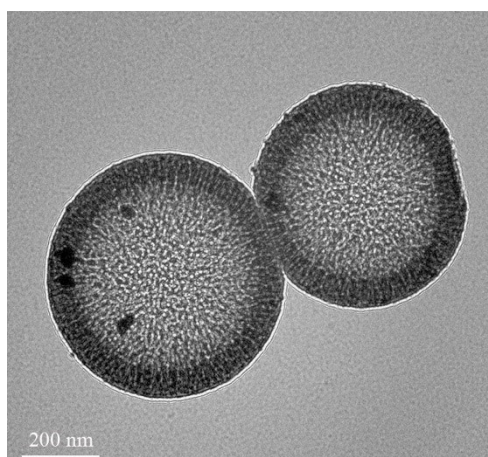
**Fig. S5.** The XPS spectrum of  $\text{Au}_{0.3}\text{Pd}_{0.7}/\text{NH}_2\text{-N-HMCS}$ .



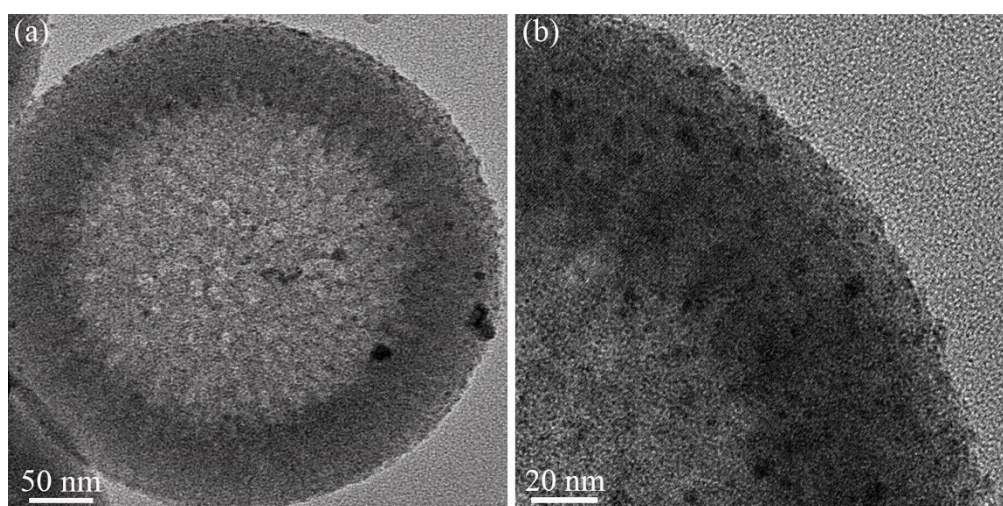
**Fig. S6.** The high-resolution XPS spectra of Au 4f for  $\text{Au}_{0.3}\text{Pd}_{0.7}/\text{NH}_2\text{-N-HMCS}$ .



**Fig. S7.** Time-course plots for the dehydrogenation of FA (1.0 M, 5.0 mL) catalyze by Au<sub>x</sub>Pd<sub>1-x</sub>/NH<sub>2</sub>-N-HMCS (x= 0, 0.1, 0.3, 0.5, 0.7, 0.9 and 1) at 298 K (a) and the related initial TOF values (b).



**Fig. S8.** TEM image of Au<sub>0.3</sub>Pd<sub>0.7</sub>/HMCS.

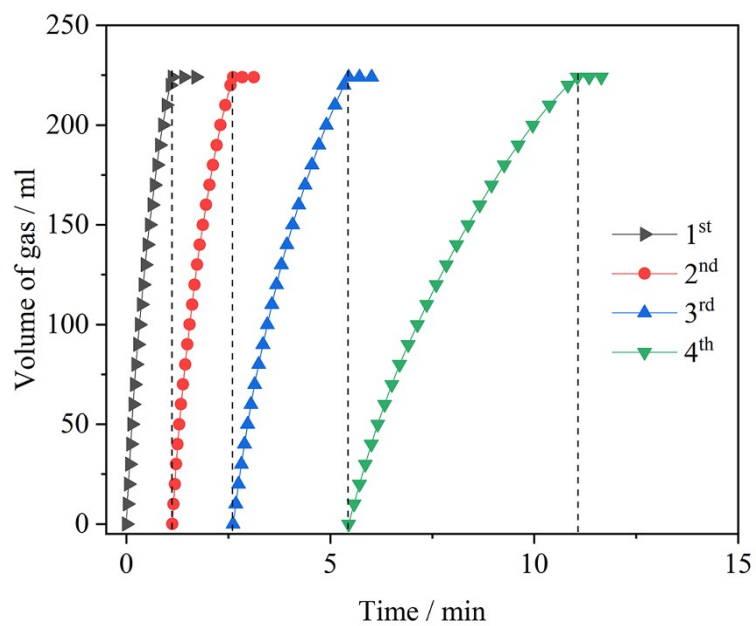


**Fig. S9.** TEM images of Au<sub>0.3</sub>Pd<sub>0.7</sub>/N-HMCS.

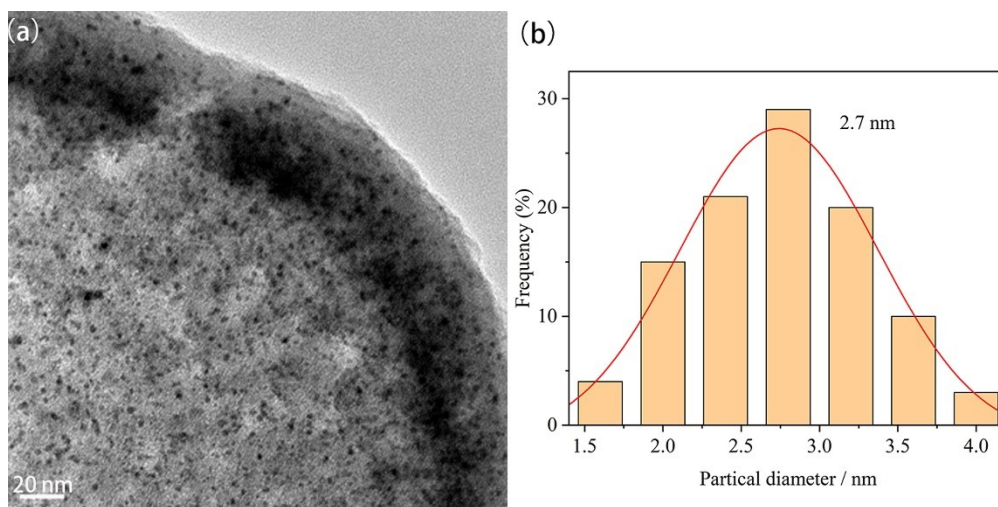
**Table S1.** Comparisons of catalytic activities for the dehydrogenation of FA catalyzed by previously reported heterogeneous catalysts with the as-synthesized Au<sub>0.3</sub>Pd<sub>0.7</sub>/NH<sub>2</sub>-N-HMCS in this work.

Catalyst	Temp. (K)	Additive	TOF (h <sup>-1</sup> )	Ref
Au@Pd/UiO-66(Zr <sub>85</sub> Ti <sub>15</sub> )	303	None	200	3
Pd/CN <sub>0.25</sub>	298	None	752	17
Au <sub>1</sub> Pd <sub>1.5</sub> /MIL-101-NH <sub>2</sub>	298	None	526	40
NiPd/NH <sub>2</sub> -N-rGO	298	None	954.3	33
Au <sub>0.75</sub> Pd <sub>0.25</sub> /C-L-7.5	298	HCOONa	718	11
Pd/S-1-in-K	298	HCOONa	856	9
(Co <sub>6</sub> )Ag <sub>0.1</sub> Pd <sub>0.9</sub> /RGO	323	HCOONa	2739	41
Au <sub>2</sub> Pd <sub>3</sub> @(P)N-C	303	HCOONa	5400	6
Au <sub>0.3</sub> Pd <sub>0.7</sub> /NH <sub>2</sub> -N-HMCS	298	None	7747	This work





**Fig. S10.** Durability test of  $\text{Au}_{0.3}\text{Pd}_{0.7}/\text{NH}_2\text{-N-HMCS}$  towards the dehydrogenation of FA.



**Fig. S11.** (a) TEM image and (b) the corresponding particle size distribution of  $\text{Au}_{0.3}\text{Pd}_{0.7}/\text{NH}_2\text{-N-HMCS}$  after the 4<sup>th</sup> run.