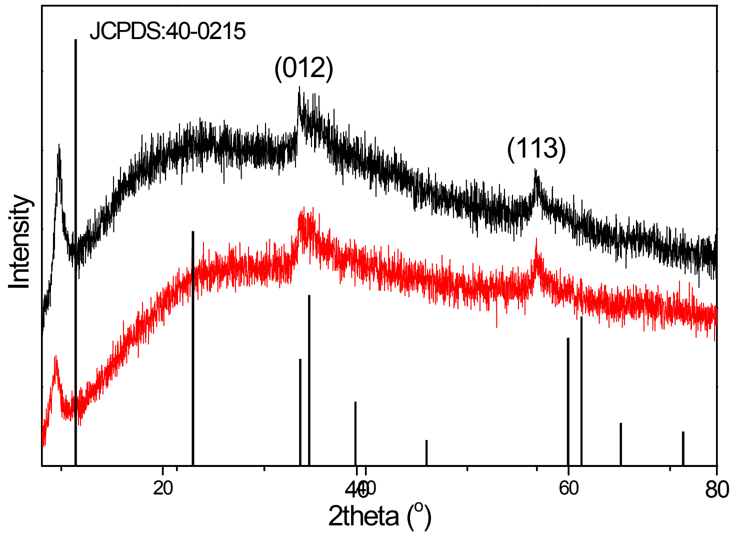


Figure S1 XRD pattern of NFLS and Pd-NFLS and the standard curve of $Ni_{0.75}Fe_{0.25}(CO_3)_{0.125}(OH)_2 \cdot 0.38H_2O$ (JCPDS: 40-0215). Upper curve was for Pd-NFLS and lower curve for NFLS. Vertical bar was for the standard XRD pattern.

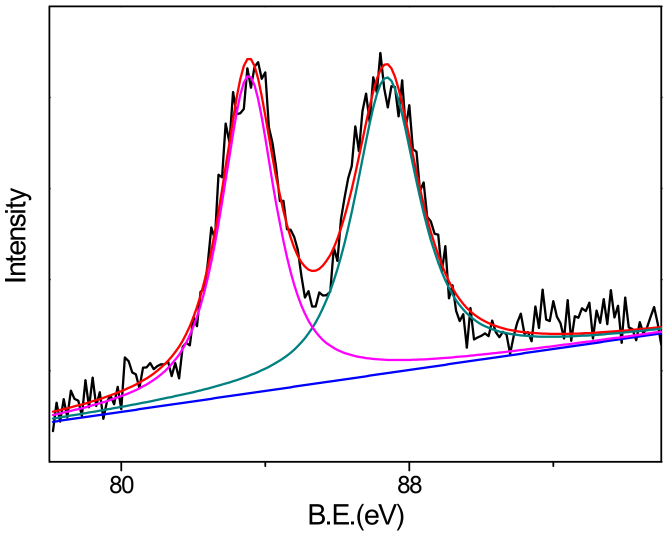


Figure S2 XPS pattern of Au 3d on Au-NFLS showed the zero valence of noble metal.

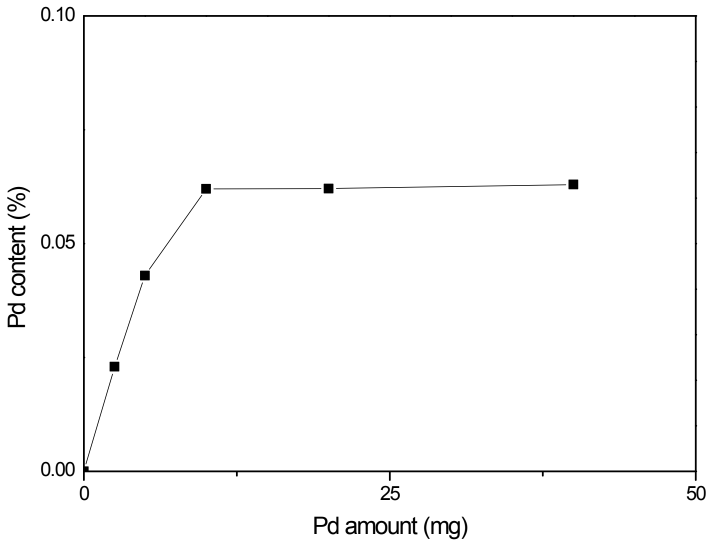


Figure S3. The content of Pd in Pd-NFLS in the different compose of soaking reactions conducted by ICP-MS. NFLS amount: 50 mg. Soaking time: 20 min.

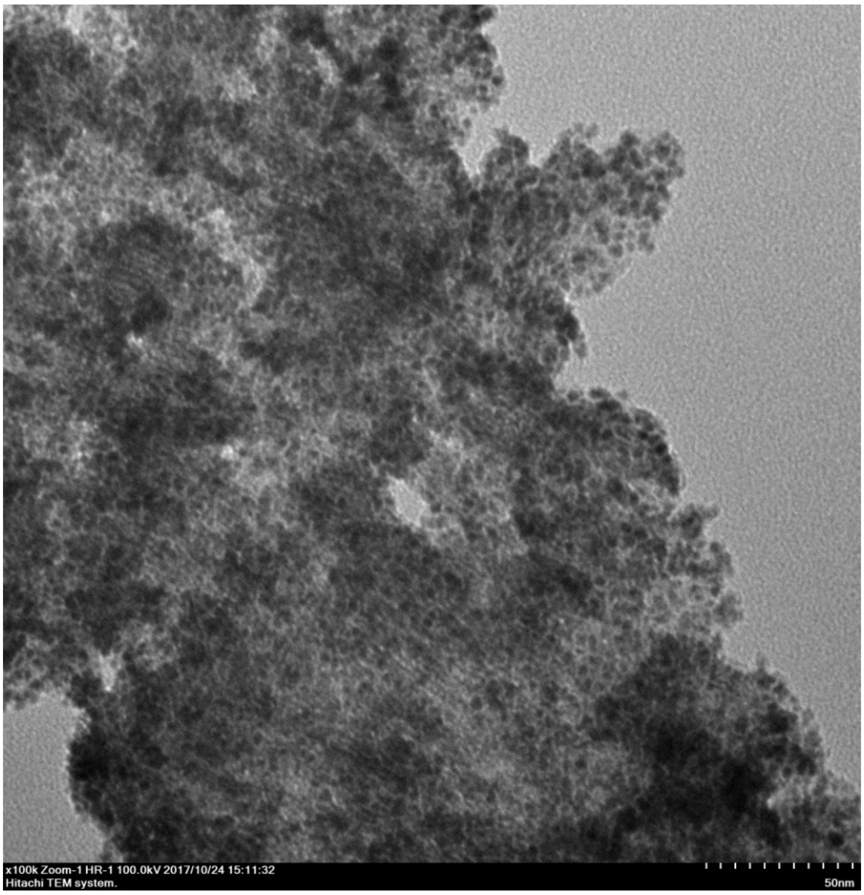
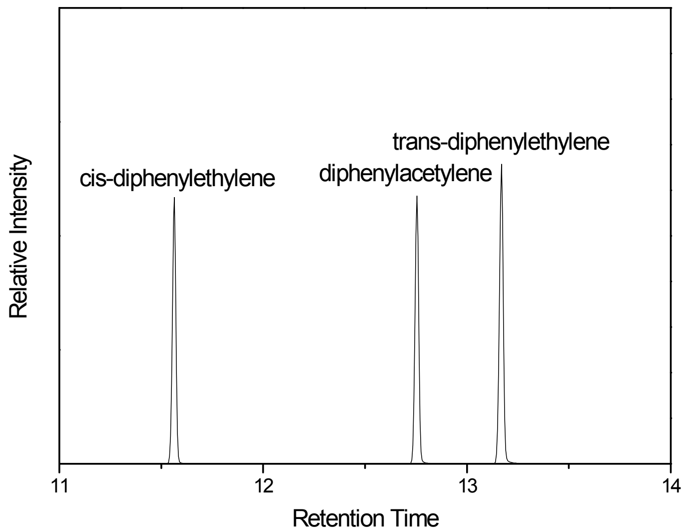


Figure S4 TEM image of Pd-NFLS with excess amount (>50 mg) of Pd resource. Pd nanoparticles were out of NFLS laminar and diameter of Pd nanoparticles was 5~10 nm.



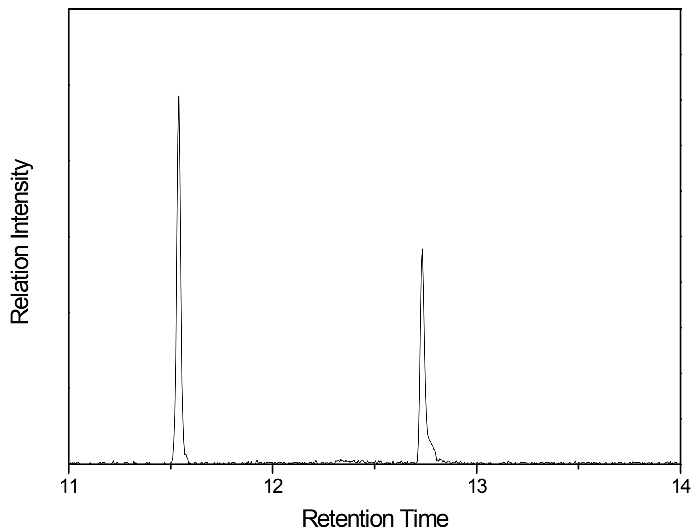


Figure S5. (a)The GC-MS pattern of the mixture of diphenylacetylene, cis-1,2-diphenylethylene and trans-1,2-diphenylethylene.

The concentration of those three reagents are equal, thus the peak area can be used to calculate the amount of each component in catalysis process. (b) A typical GC-MS pattern of semihydrogenation. Pd-NFLS works as catalyst in these reactions. No trans- product appears in the GC-MS pattern, which reveals excellent stereoselectivity of PD-NFLS catalyst.

GC-MS was carried out on Thermo Scientific DSQ GC/MS (QP2010, Shimadzu). 50 μl product was dispersed in 950 μl ethanol and He was used as carrier gas.

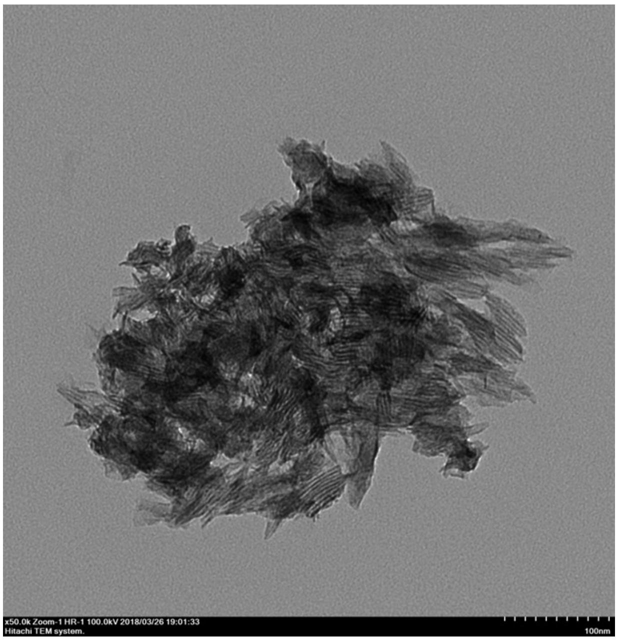


Figure S6 TEM image of Pd-NFLS after catalyst process still remain the periodic laminar structure.

Table S1 Catalytic performances of other reported semihydrogenation catalysts

Catalyst	Sub.[a]	Temp.[b]	Red.[c]	Conv.[d]	TON[e]	Ref.
Au NPs	Phenylacetylene et al.	80 °C	H_2(g)	~99%	5000 h^{-1}	10.1021/acscatal.6b03441
Au_{25} clusters	Phenylacetylene et al.	100 °C	H_2(g)	~99%		10.1021/ja503724j
Au@CeO_2	Phenylacetylene	Rt.	H_2(g)	~99%		10.1021/jacs.5b07521
Cu/SiO_2	Diphenylacetylene et al.	60 °C	H_2(g)	~99%		10.1021/jacs.6b10817
Au_xAgNPore	Diphenylacetylene et al.	90 °C	H_2(g)	86%		10.1021/jacs.6b06569
PdNPore	Diphenylacetylene et al.	Rt.	H_2(g)	90%		10.1021/acscatal.7b02915
PdIn/$MgAl_2O_4$	Phenylacetylene	90 °C	H_2(g)	97%		10.1021/jacs.7b01471
Pd-MDPC	1-octyne	Rt.	H_2(g)	62%		10.1002/cctc.201501283
Ni SAs	Acetylene	Rt.	H_2(g)	90%		10.1039/c7cc04820c
Pd@Ru Ns	1-octyne	Rt.	H_2(g)	81%		10.1002/adma.201604829
Pd_1/ND@G	acetylene	180 °C	H_2(g)	90%	230 h^{-1}	10.1021/jacs.8b07476
Pd-POM	Diphenylacetylene	Rt.	$PhMe_2SiH$	91%	2000 h^{-1}	10.1039/C5SC03554F
Pd-Pb NCs	Diphenylacetylene	Rt.	H_2(g)	96%		10.1002/anie.201503148
Pd^0-AmP-HSN	Phenylacetylene	Rt.	H_2(g)	95%		10.1002/cctc.201501112
Co(PDI)	4-phenyl-1-butene	40 °C	$(EtO)_2MeSiH$	97%		10.1021/acscatal.6b02272
$Cu(OAc)_2\cdot H_2O$	1-phenyl-1-allylene	Rt.	$PhMe_2SiH$	80%		10.1002/adsc.201200200
Pd/Zn-Ti	Phenylacetylene	45 °C	H_2(g)	93%		10.1016/j.apsusc.2018.06.091
pre-NiCu/MMO	Phenylacetylene	100 °C	H_2(g)	86%		10.1016/j.jcat.2018.01.009
NiMgGaLDH	Phenylacetylene	50 °C	H_2(g)	80%		10.1002/cctc.201300813

a Sub., substrate. b Temp., temperature. c Red., reductant. d Conv., conversion (from alkyne to alkene). e TON, turnover number.

Table S2 Selective hydrogenation of different α,β-unsaturated aldehydes by different kinds of LDH based nanoreactors.

Entry	Catalysts	Reductants	Time (h)	Conv. (%)*	Selectivity (%) **		
					A	B	C
Substrate:	cinnamaldehyde						
1	Pd-NFLS	$NaBH_4$	8	98.1	94.5	0	5.5
2	Au-NFLS	$NaBH_4$	8	43.1	83.0	1.7	15.3
3	Pt-NFLS	$NaBH_4$	8	69.7	82.1	7.5	10.4
4	Pd NPs	$NaBH_4$	8	37.8	56.3	7.4	36.3
5	Pd-NFLS	$PhSiHMe_2$	24	46.7	18.6	24.7	56.7
6	Au-NFLS	$PhSiHMe_2$	24	47.5	22.2	37.0	40.8
7	Pt-NFLS	$PhSiHMe_2$	24	94.8	1.5	84.3	14.2
8	Pd NPs	$PhSiHMe_2$	24	7.8	17.7	19.6	62.7
9	NFLS	$NaBH_4$	8	0	0	0	0
10	NFLS	$PhSiHMe_2$	24	0	0	0	0
Substrate: furfural							
11	Pd-NFLS	$NaBH_4$	1	98.5	100	0	0

12	Au-NFLS	$NaBH_4$	1	96.1	100	0	0
13	Pt-NFLS	$NaBH_4$	1	94.9	100	0	0
Substrate: 3-methyl-2-butenal							
14	Pd-NFLS	$NaBH_4$	16	54.3	94.2	2.3	3.5
15	Au-NFLS	$NaBH_4$	16	31.2	88.2	3.9	7.9
16	Pt-NFLS	$NaBH_4$	16	34.6	90.1	2.1	7.8

*Conv., conversion. Conversion of substrates and selectivity for specific products were tested by gas chromatography-mass spectrometry

** Selectivity, A, α,β-unsaturated alcohol; B, saturated aldehyde; C, saturated alcohol.