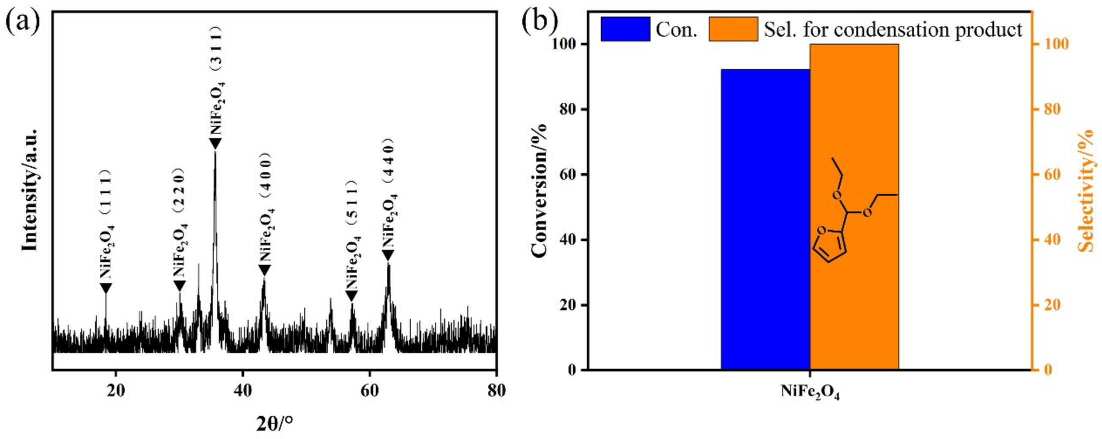


Fig. S1 XRD pattern of $NiFe_2O_4$ catalyst(a) and furfural hydrogenation reaction result with $NiFe_2O_4$ catalyst(b).

Table S1 Desorption peak temperature and hydrogen adsorption quantity of 2NaNi, 2NaFe, 0NaNiFe and 2NaNiFe

Sample	Temperature at maximum/℃	Quantity/mmol/g
2NaNi	86.3	0.20887
	289.4	$3.696*10^{-2}$
	382.8	$6.678*10^{-2}$
2NaFe	114.7	0.18650
	716.2	2.16829
0NaNiFe	106.4	0.12948
	318	$2.697*10^{-2}$
	384.6	$9.408*10^{-3}$
	464	$99.370*10^{-2}$
2NaNiFe	136.6	0.30649
	246.7	0.1383
	322.8	$3.880*10^{-2}$
	490.3	0.11348

Table S2 The ratio of Ni and Fe on liner scan, mapping scan and ICP-MS of 2NaNiFe

Test Measure	Ni/%	Fe/%	Ni:Fe
Liner scan	62.68[a]	15.36[a]	4.08
Mapping	11.68[a]	2.74[a]	4.26
ICP-MS	12.50[b]	2.89[b]	4.33

a: the value indicates the atomic percent of element in catalyst

b: the value indicates the element concentration of catalyst, mmol/g.

Table S3 The BE on Na 1s, O 1s, Ni 2p and Fe 2p of 2NaNiFe under Ex XPS and In XPS condition

Entry	Na 1s	O 1s		Ni 2p		Fe 2p	
	Na	$Ni(OH)_2$	$NiFe_2O_4$	Ni^0	$NiFe_2O_4$	Fe^0	Fe_2O_3
Ex XPS	1071.42 [1]	531.6	529.9	852.54	855.4	706.7	711.4
In XPS	1071.95	531.7[2]	530[3]	852.78[4]	855.5($Ni(OH)_2$)[5]	706.5[6]	711.6[7]

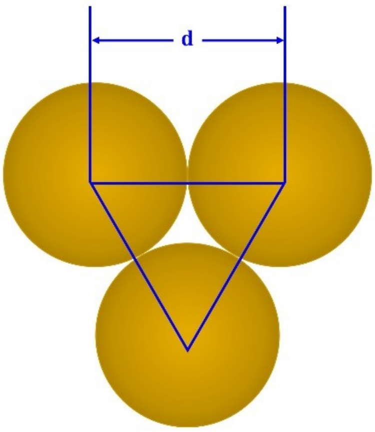
Table S4 Catalytic performance of recently developed catalysts for the hydrogenation reaction of furfural

Entry	Catalyst	Reduction before reaction	Reaction condition							Ref.
			$m_{furfural}$ /mg	$m_{catalyst}$ /mg	Tem. /℃	Pre. /MPa	Time /h	Con. /%	Sel. /%	
1	Ir_1/mpg-TiO_2	250℃, 2h	96	120	140	1	6	99	99	8
2	Pt/m-CN-2	No	252	29	70	1	2	99.3	98.5	9
3	Pd/MnO_2	No	1000	10	180	2	5	83	60	10
4	Pt_1Cu_{20}	300℃, 0.5h	20	30	50	2	7	97	99	11
5	Ni@OMC	No	290	100	180	3	12	92.3	98.6	12
6	CuNi/*rss* AC	$NaBH_4$, 70℃, 12h	250	25	140	1.5	4	100	84.4	13
7	Ni-Fe(2)CeO_2 HT-573	No	106	50	150	1	0.5	96	95	14
8	NiCoZn@CN-600	400℃, 2h	245	25	160	2	3	100	85.6	15
9	CuCo/Zn@NPC-600	400℃	245	25	140	2	4	100	100	16
10	2NaNiFe	No	700	25	130	1	1	98.21	98.39	This work

Turnover frequency (TOF) calculations[17]

Turnover frequency (TOF) can unveil the consumption rate of per number of exposed Ni_3Fe_1 sites on the catalyst. TOF should be evaluated under a stable furfural consumption state at low conversion. The Ni_3Fe_1 alloy distribution and structure of 2NaNiFe catalyst had been confirmed by XRD and HRTEM.

We supposed that the (111) plane of Ni_3Fe_1 phase was bounded on the spherical nanoparticles and defined the lattice distance and the particle diameter as d and $d^{'}$, respectively. It can be known that only face-centered cubic Ni_3Fe_1 alloy phase excited in 2NaNiFe catalyst, so the space utilization of closet packing is 74.05%.



a. Area of single Ni_3Fe_1 unit= $2 \times \frac{1}{2}d \times \frac{\sqrt{3}}{2}d$ =$0.866d^2$

Superficial area of one spherical nanoparticle= $\pi d'^2$

N_{sa} (Number of surface atoms)= $\pi d'^2/0.866d^2$=$3.626\pi d'^2/d^2$

b. Volume of single atom= $\frac{\pi d^3}{6}$

Volume of one spherical nanoparticle= $\frac{\pi d'^3}{6}$

N_{san} (Number of surface atoms of one spherical nanoparticle) = $\frac{\pi d'^3}{6} \times 74.05\%/\frac{\pi d^3}{6}$ = $0.7405 \times d'^3/d^3)$

D (Dispersion) = N_{sa}/ N_{san} = $(3.626\pi d'^2/d^2)/(0.7405 \times d'^3/d^3)$ = $4.90 \times d/d'$

The TOF value of 2NaNiFe catalyst was calculated as follow: $n_{furfural}$ = 700/96.08 =

7.2856 mmol, $Conversion_{15min}$= 49.57%, n_{metal} = (12.5028+2.8863) × 0.025=0.3847 mmol, D_{metal} = 4.9×0.205/11.2 = 0.0897, t = 0.25 h.

$TOF_{2NaNiFe}$ (h^{-1}) = 7.2856×49.57%/(0.3847×0.0897×0.25) = 418.63 h^{-1}

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