Supporting Information

Depolymerization of Lignin Disassembly into Cycloalkanes over Hydrotalcite-derived NiFe Alloy Catalyst

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Table S1. PPE conversion (%) of $Ni_9Fe_1/NiAlO_z$ with different solvents and with or without H_2.
Reaction conditions: 500 mg catalyst, 500 mg PPE, 70 mL solvent, 100 °C, 3 h, with (0.8 MPa) or without H_2 pressure.

Solvent	H_2O	Methanol	Ethanol	Isopropanol	n-Dodecane
With H_2	8.5	10.1	19.8	33.4	99.8
Without H_2	-	-	-	-	-

Table S2. EXAFS fitting parameters at the Ni K-edge for various samples.

Samples	Shell	*CN*	*R* (Å)	σ^2 (Å^{-2})	ΔE_0 (eV)	*R* factor
Ni foil	Ni-Ni	12	2.5	0.006	6.3 (0.6)	0.003
NiO	Ni-O	6.1 (0.3)	2.1	0.009	-2.4 (1.1)	0.006
	Ni-Ni	11.9 (0.4)	3.0	0.008		
$Ni_9Fe_1/NiAlO_z$	Ni-O	6	2.1	0.010	2.4 (2.5)	0.01
	Ni-M_1	12	2.6	0.015		
	Ni-M_2	12	3.0	0.014		

CN, coordination number; *R*, distance between absorber and backscatter atoms; σ^2, the Mean Square Relative Displacement (MSRD); ΔE_0, energy shift interpreted as the alignment of the energy grids of the data and the theory; *R* factor indicates the goodness of the fit.

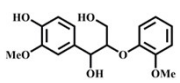
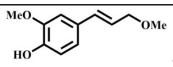
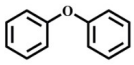
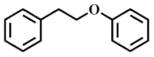
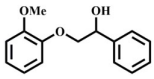
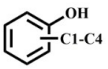
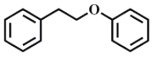
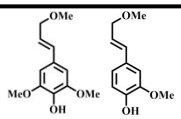
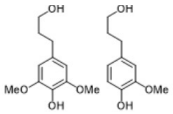
S_0^2 was fitted as 0.75 for Ni foil by fixing *CN* as the known crystallographic value. Fitting range of Ni foil is $3.0 \leq k$ (Å^{-1}) ≤ 10.5 and $1 \leq R$ (Å) ≤ 3. S_0^2 was fixed as 0.98 for the fitting of NiO. Fitting range of NiO is $3.0 \leq k$ (Å^{-1}) ≤ 10 and $1 \leq R$ (Å) ≤ 3. S_0^2 was fixed as 0.80 for the fitting of $Ni_9Fe_1/NiAlO_z$. Fitting range of $Ni_9Fe_1/NiAlO_z$ is $3.0 \leq k$ (Å^{-1}) ≤ 10.4 and $1 \leq R$ (Å) ≤ 3.

Table S3. EXAFS fitting parameters at the Fe K-edge for various samples.

Samples	Shell	*CN*	*R*(Å)	σ^2(Å^{-2})	ΔE_0(eV)	*R* factor
Fe foil	Fe-Fe	12	2.5	0.0078	5.8 (2.8)	0.005
Fe_2O_3	Fe-O_1	4.2 (1.7)	2.0	0.010	-1.3 (2.4)	0.004
	Fe-O_2	1.8	2.2			
	Fe-Fe_1	3	3.0	0.005		
	Fe-Fe_2	1	2.9			
	Fe-Fe_3	2	3.4			
	Fe-Fe_4	6	3.7	0.010		
	Fe-O_3	3	3.9	0.006		
$Ni_9Fe_1/NiAlO_z$	Fe-O_1	5.4 (0.7)	2.0	0.014	1.5 (1.2)	0.007
	Fe-M_1	6.0 (1.3)	2.6	0.023		
	Fe-M_2	5.8 (1.8)	3.0	0.013		
	Fe-O_2	4	3.3	0.028		

S_0^2 was fitted as 0.75 for Fe foil by fixing *CN* as the known crystallographic value. Fitting range of Fe foil is $3.0 \leq k$ (Å^{-1}) ≤ 11.9 and $1 \leq R$ (Å) ≤ 3. S_0^2 was fixed as 1.00 for the fitting of Fe_2O_3. Fitting range of Fe_2O_3 is $3.0 \leq k$ (Å^{-1}) ≤ 10 and $1 \leq R$ (Å) ≤ 3.5. S_0^2 was fixed as 0.85 for the fitting of $Ni_9Fe_1/NiAlO_z$. Fitting range of $Ni_9Fe_1/NiAlO_z$ is $3.0 \leq k$ (Å^{-1}) ≤ 8 and $1 \leq R$ (Å) ≤ 3.

Table S4 The performance comparison of $Ni_9Fe_1/NiAlO_z$ with previously reported catalysts for model compound and lignin depolymerization.

Catalysts	Subtrate	Reaction condition	Major products and selectivity		Ref.
Mo_1Al/MgO		200 °C, 4h, 1 MPa N_2	 58 %	 64%	J. Am. Chem. Soc. 2023, 145, 12884-12893
Ni-Fe/MCS		250 °C, 4 h, 5 MPa H_2	 49.1%	 49.9%	Appl. Catal. B-Environ. 2019, 253, 348-358
Br-Ru/SiO_2		120 °C, 6 h, 0.5 MPa H_2	 44.7%	 45.6%	Angew. Chem. Int. Ed. 2021, 60, 12513-125233
Ni/Ni-PS		160 °C, 2 h, 1 MPa H_2	 33.8%	 4 7.6%	Green Chem. 2022, 24, 846-857
NiFe(3)/TiO_2-HT		250 °C, 1 h, 5 MPa H_2	 71.8%		Chem. Eng. J. 2022, 446 136578
RuRe/H-Beta		250 °C, 10 h, 0 MPa H_2	 26.1%	 43.8%	J. Energy Chem. 2022, 67, 492-49
$Ni_9Fe_1/NiAlO_z$		100 °C, 3 h, 0.8 MPa H_2	 40.5%	 56.8%	This work
Mo_1Al/MgO	Eucalyptus wood	200 °C, 8 h, 1 MPa N_2		92%	J. Am. Chem. Soc. 2023, 145, 12884-12893
RuRe/H-Beta	Alkaline lignin	200 °C, 2 MPa H_2, 10 h		54%	J. Energy Chem. 2022, 67, 492-49
Pd/C	Extracted lignin from birch	200 °C, 15 h, 4 MPa H_2		90%	Angew. Chem.Int. Ed. 2018, 57, 1356.
$Ru/NbOPO_4$[a]	Kraft lignin	310 °C, 40 h, 0.5 MPa H_2		68%	Chem, 2019, 5, 1521-1536,
$Ni_9Fe_1/NiAlO_z$[a]	Enzymatic hydrolysis lignin	300 °C, 6 h, 3 MPa H_2		100%	This Work

[a] Breaking the limit of lignin monomer production

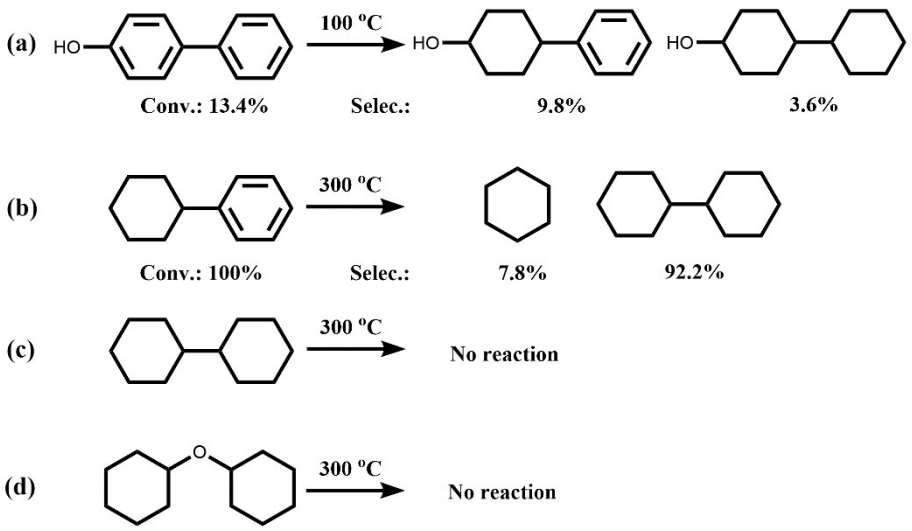


Fig. S1. Product distributions for the conversion of (a) 4-phenylphenol, (b) phenylcyclohexane, (c) bicyclohexyl and (d) cyclohexyloxy-cyclohexane.

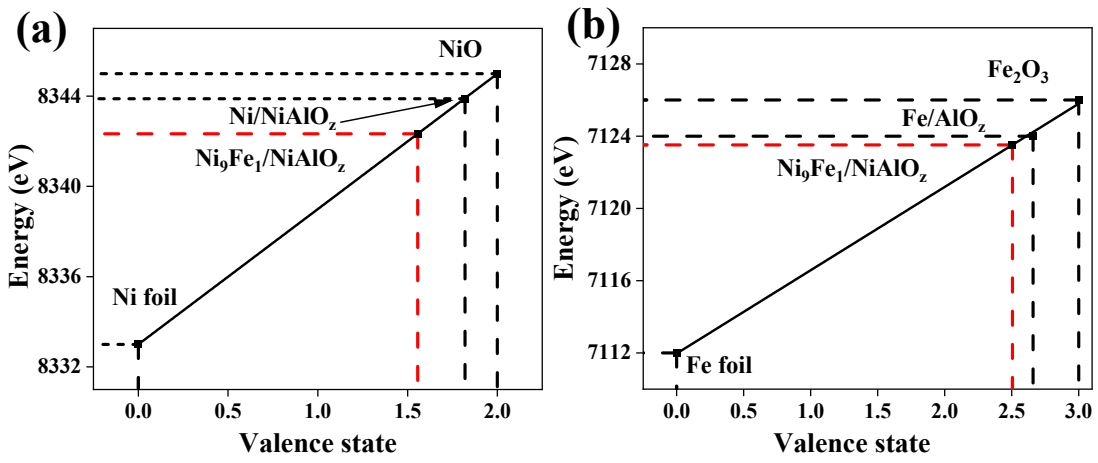


Fig. S2. The valence result of Ni (a) and Fe (b).

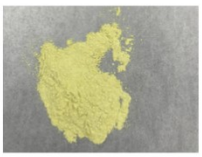
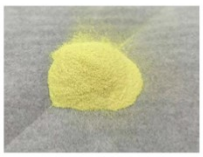
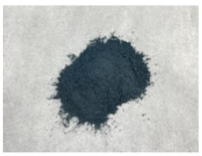
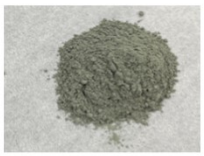
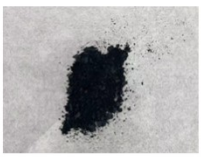
	$Ni_9Fe_1/NiAlO_z$	$Ni/NiAlO_z$	Fe/AlO_z
0 S			
30 S			
180 S			

Fig. S3. Color change photographs of the mixtures.

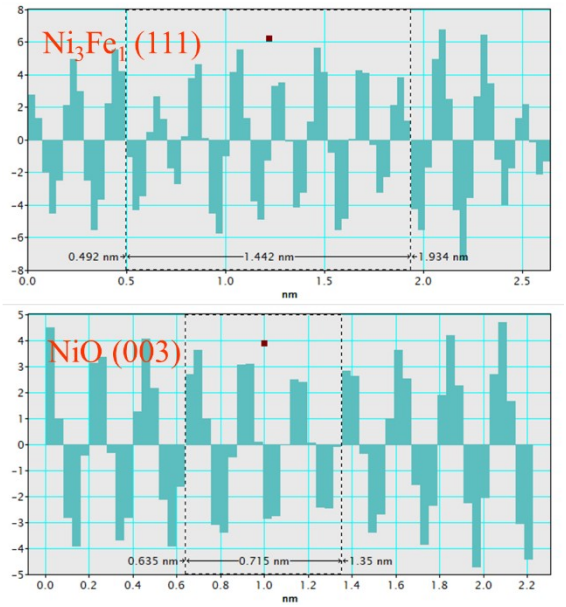


Fig. S4. Lattice stripe calculation for HRTEM.