Supporting Information

Quantitative conversion of polyethylene terephthalate (PET) and Coca-Cola bottles to p-xylene over Co-based catalysts with tailored activities for deoxygenation and hydrogenation

Yuewen Shao[a], Mengjiao Fan[a], Kai Sun[a], Guoming Gao[b], Chao Li[a], Dianqiang Li[a], Yuchen Jiang[a], Lijun Zhang[a], Shu Zhang[b], Xun Hu[a,*]

[a]School of Material Science and Engineering, University of Jinan, Jinan, 250022, P. R. China.
[b]Joint International Research Laboratory of Biomass Energy and Materials, College of Materials Science and Engineering, Nanjing Forestry University, Nanjing, 210037, P. R. China.
*Corresponding author. Tel. /fax: +86–531–89736201; E-mail: xun.hu@outlook.com (X. Hu).

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Table S1 Hydrogen consumption and reduction degree of Co-based catalysts

Entry	Catalysts	Hydrogen consumption[a] (mmol/g)	Reduction degree[a] (%)	Dispersion of Co[b] (%)	TOF (h^{-1})
1	Co-Al	9.2	75.5	10.9	5.3
2	Co-Fe-Al	9.6	56.0	4.0	147.4
3	Co-Cu-Al	10.4	77.6	17.8	22.8
4	Co-Ni-Al	12.0	86.8	15.2	0.0
5	Co-Zn-Al	6.4	52.7	10.7	32.4

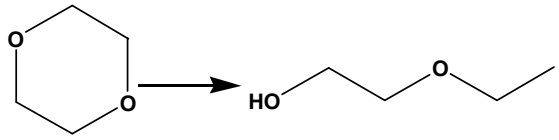
[a]The reduction degree were determined by H_2-TPR characterization.

[b]Dispersion of Co (D) was calculated following: D = 96 / particle size of Co. Particle size of Co was obtained in Table 1.

[c]TOF was calculated based on the production of xylene from TPA over Co sites on surface of Co-based catalysts, following the Eq. (1). Reaction conditions: t = 2 h; P_{H2} = 4.0 MPa (at room temperature); stirring speed = 600 rpm; catalyst loading: 80 mg; reactant: 50 mg; 1,4-dioxane: 5 mL.

$$\mathrm{TOF}_{\text{Co sites}}\left(\mathrm{h}^{-1}\right)=\frac{\text{Mole of xylene formed}}{\text{Mole of Co}\ \times\ \text{dispersion of Co}\ \times\ \text{reaction time}} \quad \text{Eq. (1)}$$

Table S2 Distribution of the products in hydrogenation of TPA[a]



1,4-dioxane 2-ethoxy-ethanol

Entry	Catalysts	T (°C)	Abundance of 2-ethoxy-ethanol (a.u.)
1	Co-Al	170	58546.0
2	Co-Fe-Al	170	0.0
3	Co-Cu-Al	170	0.0
4	Co-Ni-Al	170	295433.0
5	Co-Zn-Al	170	0.0
6	Co-Al	190	3007359.0
7	Co-Fe-Al	190	0.0
8	Co-Cu-Al	190	0.0
9	Co-Ni-Al	190	1868076.0
10	Co-Zn-Al	190	0.0
11	Co-Al	210	4006288.0
12	Co-Fe-Al	210	0.0
13	Co-Cu-Al	210	0.0
14	Co-Ni-Al	210	2538053.0
15	Co-Zn-Al	210	0.0

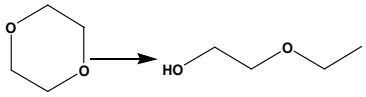
[a]Reaction conditions: t = 10 h; P_{H2} = 4.0 MPa (at room temperature); stirring speed = 600 rpm; catalyst loading: 80 mg; reactant: 50 mg; 1,4-dioxane: 5 mL.

Table S3 Distribution of the products in hydrogenation of ethylene glycol[a]

Entry	Catalysts	Con. (%)
1	Co-Al	56.6
2	Co-Fe-Al	5.2
3	Co-Ni-Al	70.6

[a]Reaction conditions: T = 190 °C; t = 2 h; P_{H2} = 4.0 MPa (at room temperature); stirring speed = 600 rpm; catalyst loading: 40 mg; reactant: 100 mg; tetrahydrofuran: 4 g.

Table S4 Distribution of the products in hydrogenation of PET[a]



1,4-dioxane 2-ethoxy-ethanol

Entry	Catalysts	T (°C)	Abundance of 2-ethoxy-ethanol (a.u.)
1	Co-Al	190	4111187.0
2	Co-Fe-Al	190	0.0
3	Co-Cu-Al	190	1858770.0
4	Co-Ni-Al	190	3190875.0
5	Co-Zn-Al	190	61190.0
6	Co-Al	210	4836534.0
7	Co-Fe-Al	210	0.0
8	Co-Cu-Al	210	2332211.0
9	Co-Ni-Al	210	3239933.0
10	Co-Zn-Al	210	459132.0

[a]Reaction conditions: t = 10 h; P_{H2} = 4.0 MPa (at room temperature); stirring speed = 600 rpm; catalyst loading: 80 mg; reactant: 60 mg; 1,4-dioxane: 5 mL.

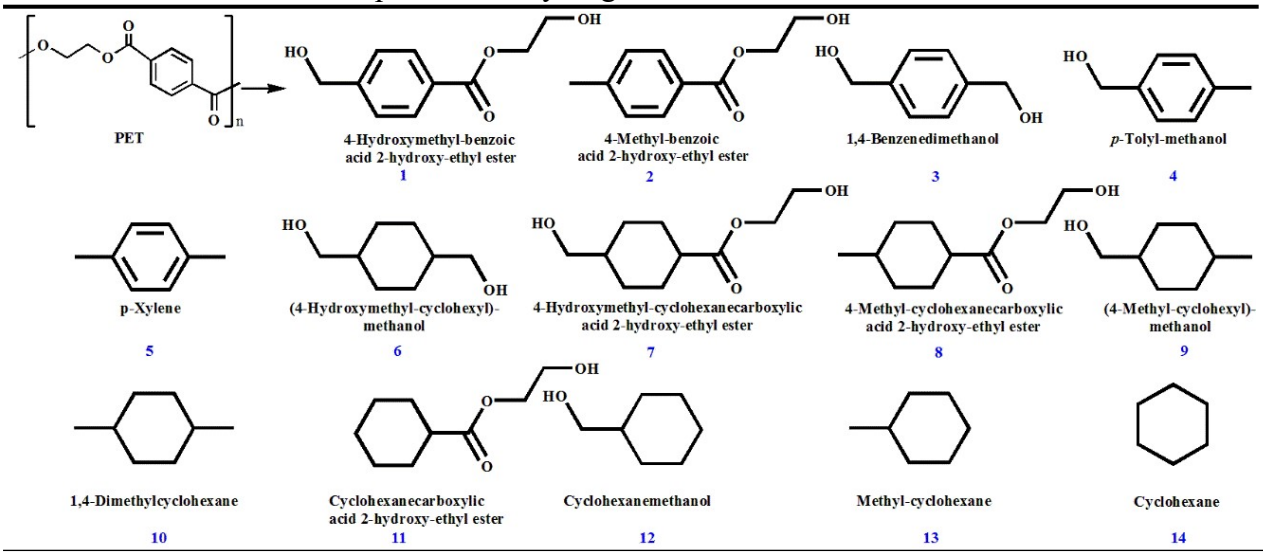
Table S5 Distribution of the products in hydrogenation of PET[a]

Entry	Catalysts	T (°C)	Yield (%)									
			2	**3[b]**	**4**	**5**	**6**	**8**	**9**	**10**	**11-14**	**EG**
1	Co-Fe-Al-700[b]	210	0.0	1.0	3.5	52.6	0.0	0.0	0.0	0.2	0.9	9.8

[a]Reaction conditions: t = 10 h; P_{H2} = 4.0 MPa (at room temperature); stirring speed = 600 rpm; catalyst loading: 80 mg; reactant: 60 mg; 1,4-dioxane: 4 mL.

[b]The product of esterification between 4-methyl-benzoic acid and ethylene glycol (4-methyl-benzoic acid-2-hydroxy-ethyl ester).

Table S6 Distribution of the products in hydrogenation of PET[a]



Entry	Catalysts	Yield (%)											
		1	2	3	4	5	6	7	8	9	10	11-14	EG
1	Co-Al	0.0	2.5	0.0	0.0	33.5	0.0	0.0	0.0	10.2	39.1	12.8	0.0
2	Co-Fe-Al	0.0	4.0	0.0	0.0	71.4	0.0	0.0	0.0	0.0	0.0	0.0	4.9
3	Co-Ni-Al	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	3.3	35.7	60.1	3.1
4	Co-Al +pyridine	0.0	0.0	0.0	0.0	76.6	0.0	0.0	0.0	0.0	15.9	4.4	0.0
5	Co-Fe-Al+pyridine	0.0	0.0	0.0	0.0	63.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0
6	Co-Ni-Al+pyridine	0.0	2.2	0.0	0.0	21.1	0.0	0.0	0.0	19.0	0.0	1.8	2.6

[a]Reaction conditions: T = 210 °C; t = 10 h; P_{H2} = 4.0 MPa (at room temperature); stirring speed = 600 rpm; catalyst loading: 30 mg; reactant: 60 mg; 1,4-dioxane: 4 mL. Pyridine loading: 50 mg.

Table S7 Distribution of the products for the hydrogenation of PET plastics over various catalysts

Entry	Catalysts	T (°C)	*t* (h)	P_{H2} (MPa)	Solvent	Con. (%)	Yield of arenes (%)	Ref.
1	Ru/Nb_2O_5	280	8	0.5	Octane	100.0	83.6	13
2	Ru/Nb_2O_5	280	12	N_2: 2.0	Water	100.0	93.3	12
3	Ru/Nb_2O_5	220	12	N_2: 2.0	Water	100.0	88.7	12
4	$CuNa/SiO_2$	210	6	N_2: 3.4	Methanol	100.0	PX: 100.0	8
5	Co/TiO_2	320	24	3.0	n-Dodecane	–	75.2	16
6	Co-Fe-Al	210	10	4	1,4-Dioxane	100.0	PX: >99.0	In this study
7	Co-Fe-Al[a]	210	10	4	1,4-Dioxane	100.0	PX: >99.0	In this study

[a]Bottle of Coca-Cola. PX: *p*-xylene.

Table S8 Kinetic study for the conversion of various reactants over Co-Fe-Al catalyst[a]

Entry	T (°C)	Reaction rate for the conversion of various reactants (k/mmol·g_{cat}^{-1}·h^{-1})					
		Terephthalic acid	**4-Hydroxymethyl benzoic acid**	***p*-Toluylic acid**	***p*-Tolualdehyde**	***p*-Tolyl-methanol**	**1,4-Benzenedimethanol**
1	170	–	16.9	8.1	82.2	10.0	16.9
2	190	18.1	57.1	32.9	269.4	42.4	96.0
3	210	40.2	205.5	70.2	495.3	203.0	364.0
4	230	156.9	–	–	–	–	–

[a]Reaction conditions: P_{H2} = 4 MPa, stirring speed = 800 rpm, catalyst: 100.0 mg, reactant: 0.8 mmol, 1,4-dioxane: 30.0 g.

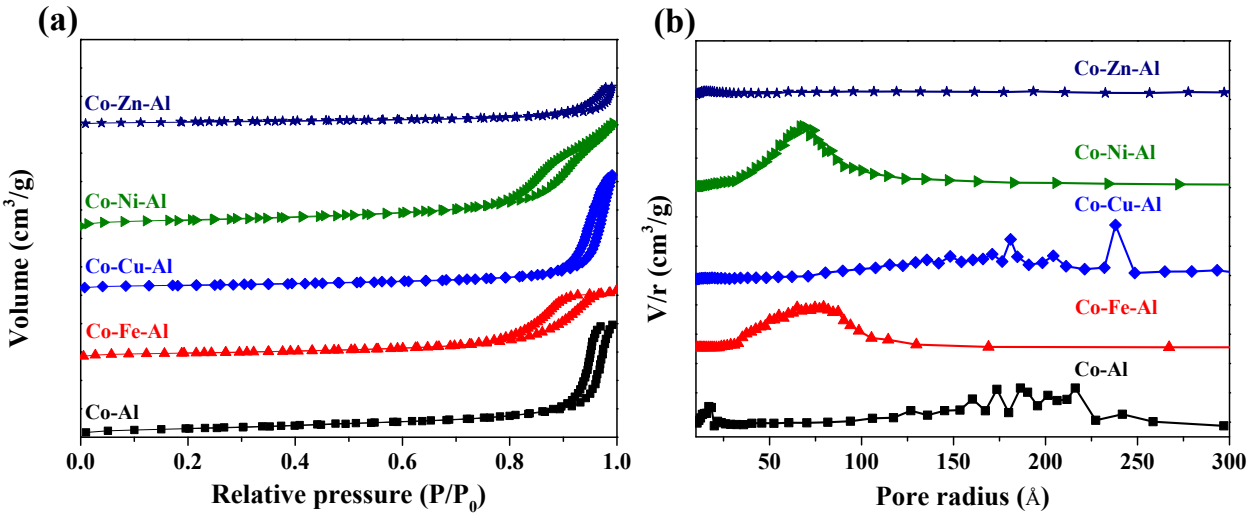


Fig. S1 (a) Nitrogen adsorption-desorption isotherms and (b) corresponding pore-size distribution curves of the calcined Co-based catalysts.

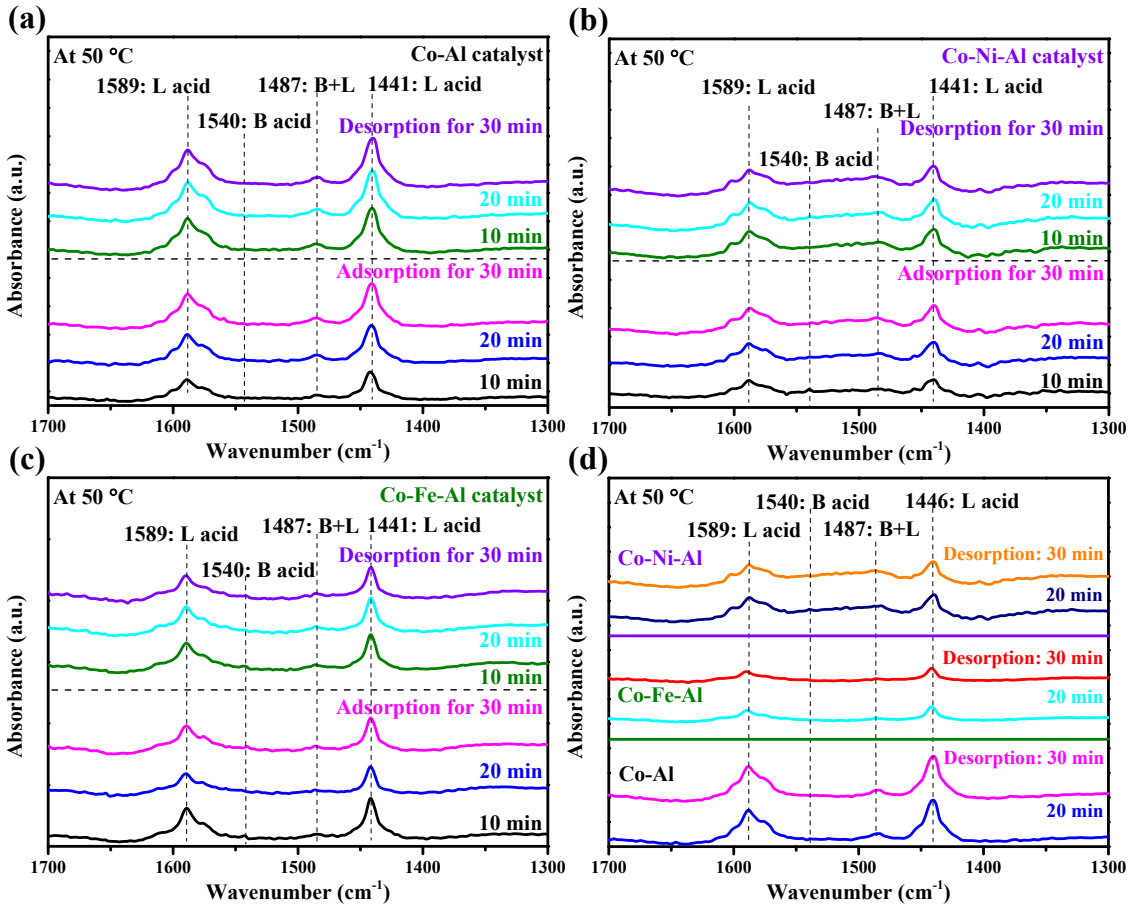


Fig. S2 In-situ FTIR spectra of pyridine of various Co-based catalysts: (a) Co-Al; (b) Co-Ni-Al; (c) Co-Fe-Al; (d) comparison of various Co-based catalysts.

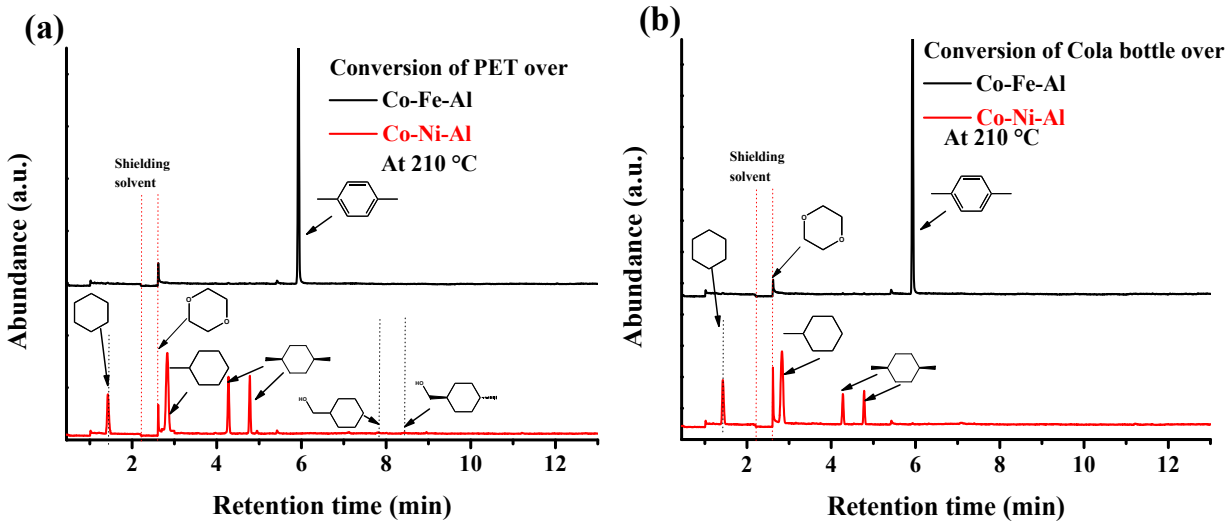


Fig. S3 GC-MS spectra for the conversion of (a) PET or (b) Cola bottle over Co-based catalysts.

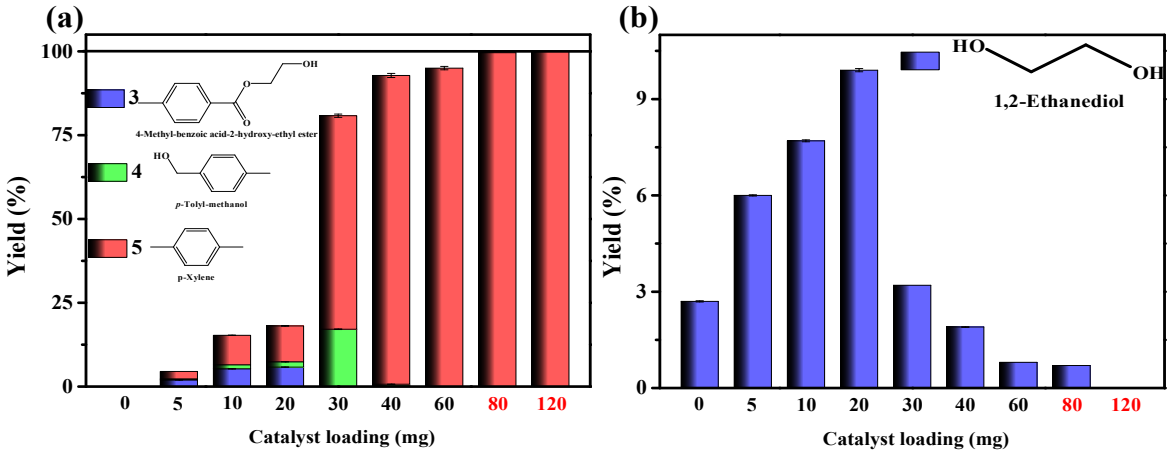


Fig. S4 Catalytic conversion of PET over Co-Fe-Al catalyst and corresponding products distribution versus various catalyst loading. Reaction conditions: T = 210 °C; t = 10 h; P_{H2} = 4.0 MPa (at room temperature); stirring speed = 600 rpm; reactant: 60 mg; 1,4-dioxane: 4 mL.

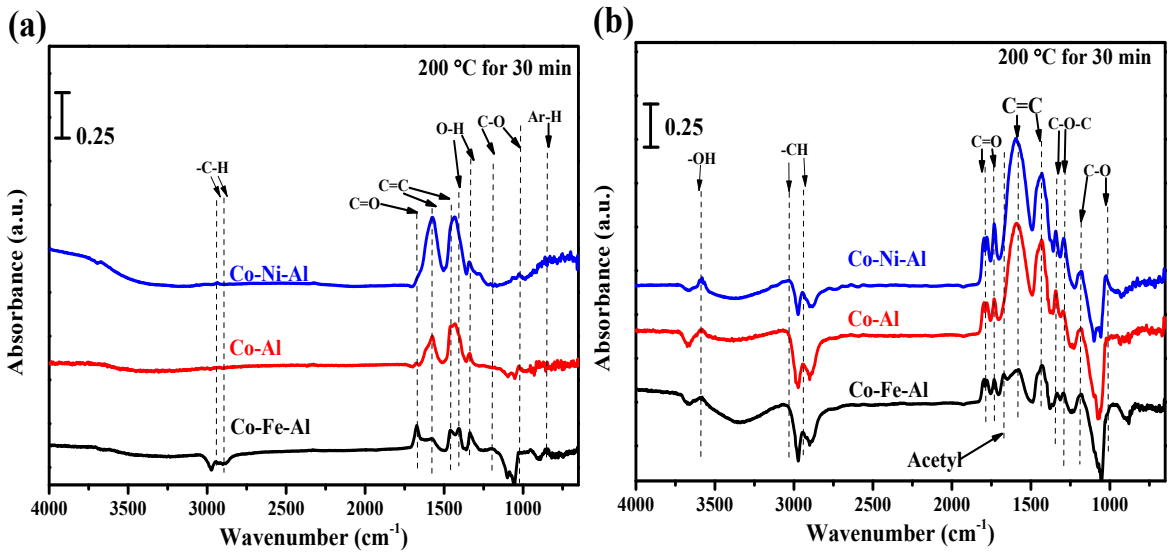


Fig. S5 (a) In-situ DRIFTS spectra for the hydrogenation of benzyl alcohol at 200 °C for 30 min over Co-Fe-Al, Co-Al, and Co-Ni-Al catalysts. (b) In-situ DRIFTS spectra for the hydrogenation of acetic acid at 200 °C for 30 min over Co-Fe-Al, Co-Al, and Co-Ni-Al catalysts.

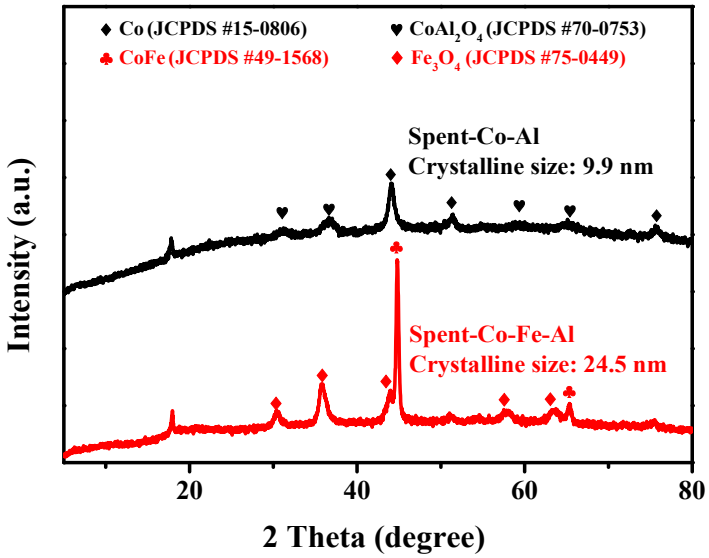


Fig. S6 The spent Co-Al and Co-Fe-Al catalysts after reuse for five times.