

Supporting Information

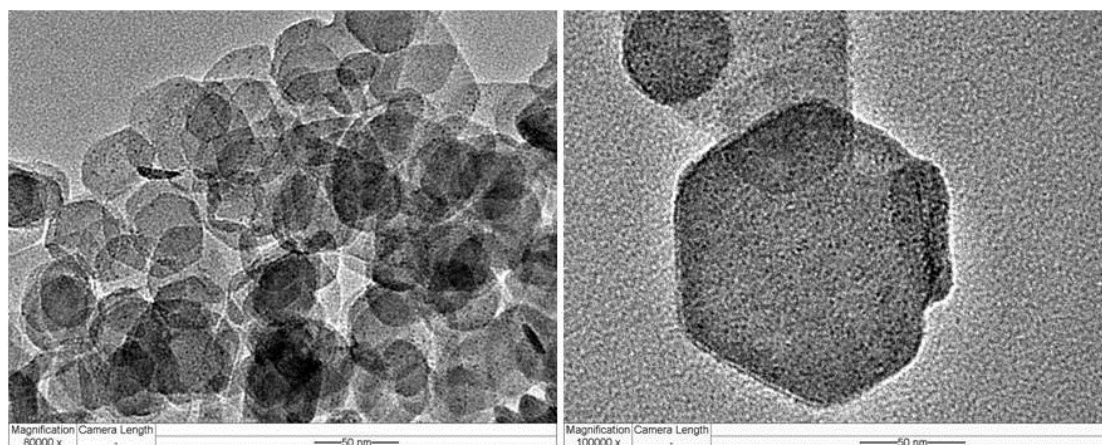
Porous Ru/RuO_x/LDH as Highly Active Heterogeneous Catalysts for Aerobic Oxidation of Alcohols

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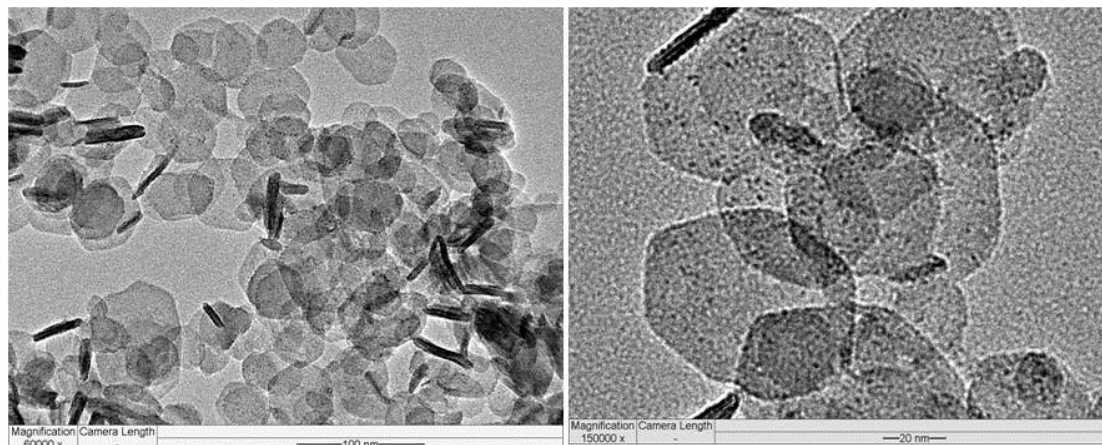
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Figure SI-1: NiAl-2:

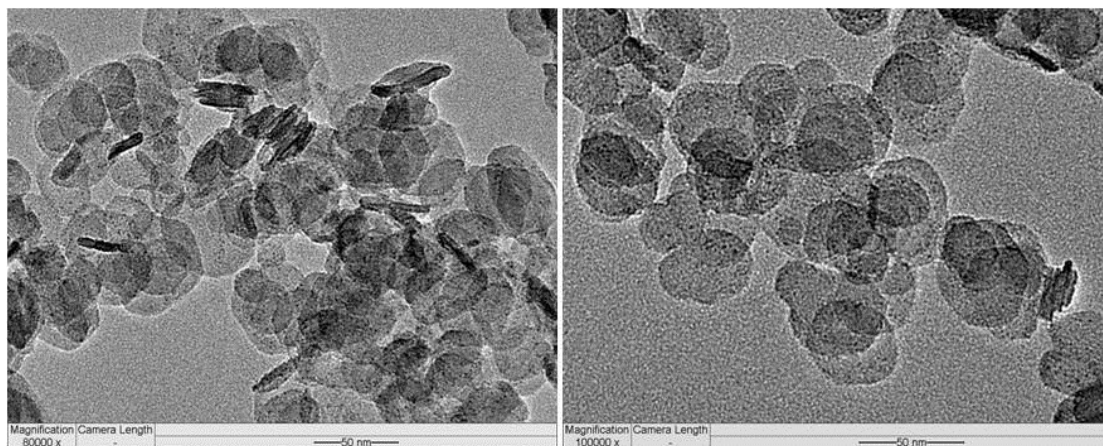


NiAl-3:

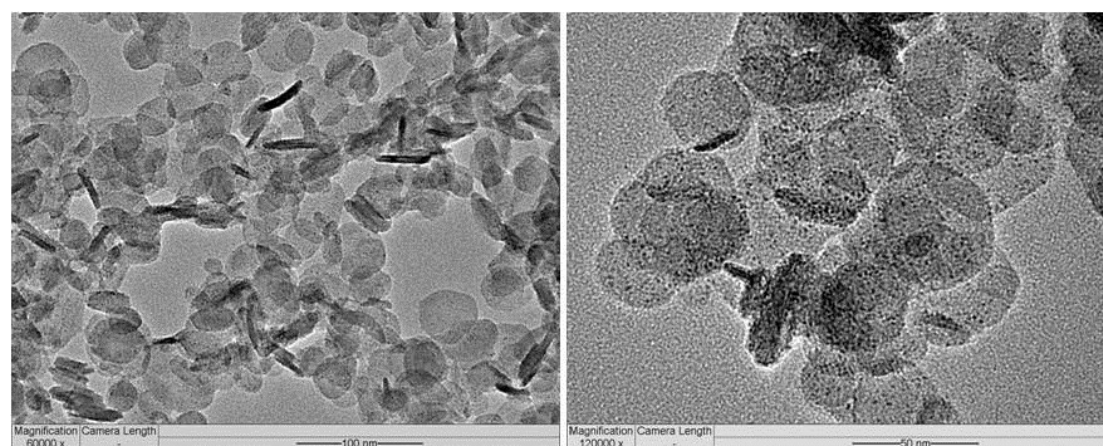


NiAl-4:

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NiAl-5:



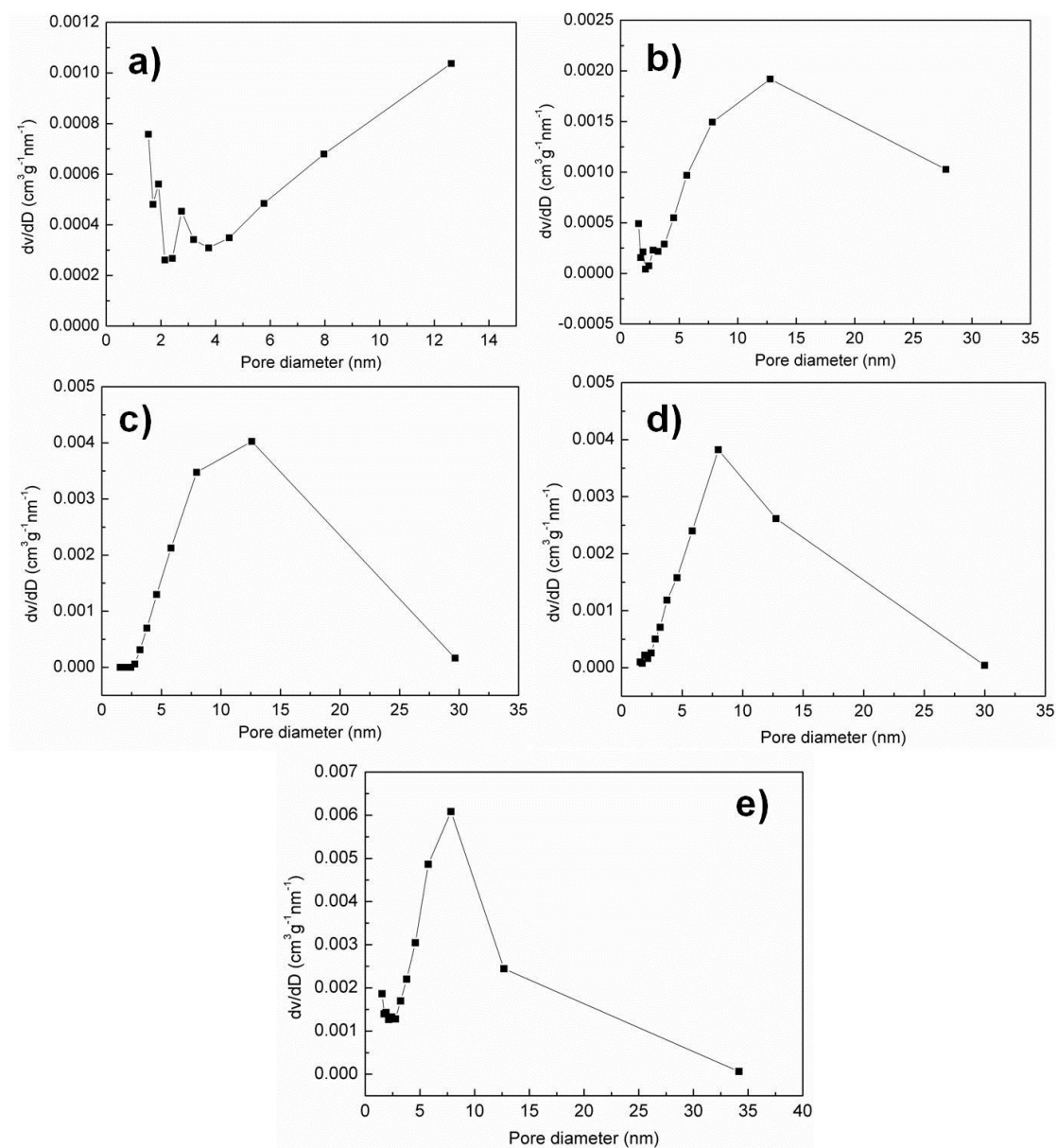


Figure SI-2. Pore size distribution of as-prepared samples (a) NiAl-1, (b) NiAl-2, (c) NiAl-3, (d) NiAl-4, (e) NiAl-5.

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TABLE SI-1: Aerobic oxidation of various alcohols catalyzed by Ru/LDH (NiAl-1) catalysts.

Entry	Substrate	Product	Conv. (%)	Sel.(%)	t (h)
1	benzyl alcohol	benzaldehyde	67.5	>99	0.5
2	1-phenylethanol	acetophenone	34.2	>99	1
3	4-methylbenzyl alcohol	4-methylbenzaldehyde	60.6	>99	0.5
4	4-methoxybenzyl alcohol	4-methoxybenzaldehyde	52.2	>99	0.5
5 ^a	4-nitrobenzyl alcohol	4-nitrobenzaldehyde	91.3	>99	0.5
6	cinnamyl alcohol	cinnamaldehyde	16.9	>99	1
7	cyclohexanol	cyclohexanone	14.3	>99	2
8	2-octanol	2-octanone	13.3	>99	2
9	1-octanol	octanal	22.3	>99	2
10	2-pyridinemethanol	2-pyridinecarboxaldehyde	16.0	>99	2

Reaction conditions: Alcohol (1 mmol), Ru/LDH (0.1 g), toluene as solvent (10 mL), 80°C, O₂ bubbling rate (20 mLmin⁻¹). ^a0.25 mmol alcohol.

TABLE SI-2: Aerobic oxidation of various alcohols catalyzed by Ru/LDH (NiAl-2) catalysts.

Entry	Substrate	Product	Conv. (%)	Sel.(%)	t (h)
1	benzyl alcohol	benzaldehyde	91.4	>99	0.5
2	1-phenylethanol	acetophenone	30.	>99	1
3	4-methylbenzyl alcohol	4-methylbenzaldehyde	50.3	>99	0.5
4	4-methoxybenzyl alcohol	4-methoxybenzaldehyde	69.6	>99	0.5
5 ^a	4-nitrobenzyl alcohol	4-nitrobenzaldehyde	>99	>99	0.5
6	cinnamyl alcohol	cinnamaldehyde	26.7	>99	1
7	cyclohexanol	cyclohexanone	10.0	>99	2
8	2-octanol	2-octanone	14.2	>99	2
9	1-octanol	octanal	24.1	>99	2
10	2-pyridinemethanol	2-pyridinecarboxaldehyde	24.0	>99	2

Reaction conditions: Alcohol (1 mmol), Ru/LDH (0.1 g), toluene as solvent (10 mL), 80°C, O₂ bubbling rate (20 mLmin⁻¹). ^a0.25 mmol alcohol.

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TABLE SI-3: Aerobic oxidation of various alcohols catalyzed by Ru/LDH (NiAl-3) catalysts.

Entry	Substrate	Product	Conv. (%)	Sel.(%)	t (h)
1	benzyl alcohol	benzaldehyde	>99	>99	0.5
2	1-phenylethanol	acetophenone	39.7	>99	1
3	4-methylbenzyl alcohol	4-methylbenzaldehyde	>99	>99	0.5
4	4-methoxybenzyl alcohol	4-methoxybenzaldehyde	>99	>99	0.5
5 ^a	4-nitrobenzyl alcohol	4-nitrobenzaldehyde	>99	>99	0.5
6	cinnamyl alcohol	cinnamaldehyde	51.9	>99	1
7	cyclohexanol	cyclohexanone	17.3	>99	2
8	2-octanol	2-octanone	32.1	>99	2
9	1-octanol	octanal	40.0	>99	2
10	2-pyridinemethanol	2-pyridinecarboxaldehyde	41.0	>99	2

Reaction conditions: Alcohol (1 mmol), Ru/LDH (0.1 g), toluene as solvent (10 mL), 80°C, O₂ bubbling rate (20 mLmin⁻¹). ^a0.25 mmol alcohol.

TABLE SI-4: Aerobic oxidation of various alcohols catalyzed by Ru/LDH (NiAl-4) catalysts.

Entry	Substrate	Product	Conv. (%)	Sel.(%)	t (h)
1	benzyl alcohol	benzaldehyde	>99	>99	0.5
2	1-phenylethanol	acetophenone	64.6	>99	1
3	4-methylbenzyl alcohol	4-methylbenzaldehyde	>99	>99	0.5
4	4-methoxybenzyl alcohol	4-methoxybenzaldehyde	>99	>99	0.5
5 ^a	4-nitrobenzyl alcohol	4-nitrobenzaldehyde	>99	>99	0.5
6	cinnamyl alcohol	cinnamaldehyde	80.8	>99	1
7	cyclohexanol	cyclohexanone	17.6	>99	2
8	2-octanol	2-octanone	41.8	>99	2
9	1-octanol	octanal	47.1	>99	2
10	2-pyridinemethanol	2-pyridinecarboxaldehyde	45.2	>99	2

Reaction conditions: Alcohol (1 mmol), Ru/LDH (0.1 g), toluene as solvent (10 mL), 80°C, O₂ bubbling rate (20 mLmin⁻¹). ^a0.25 mmol alcohol

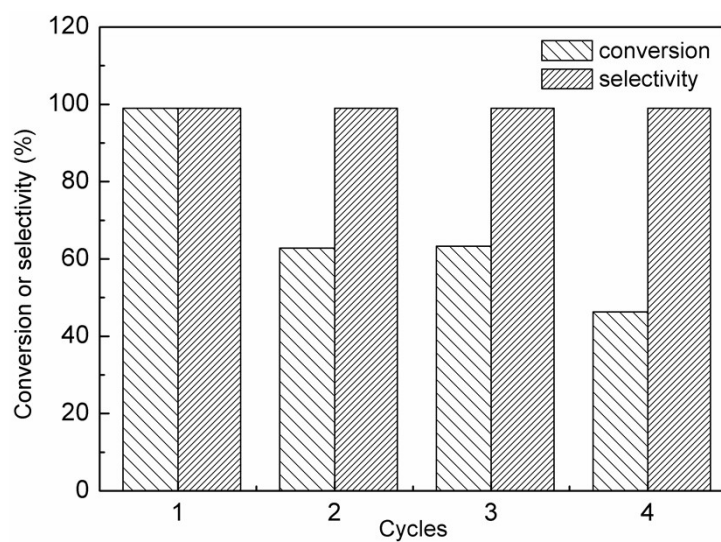
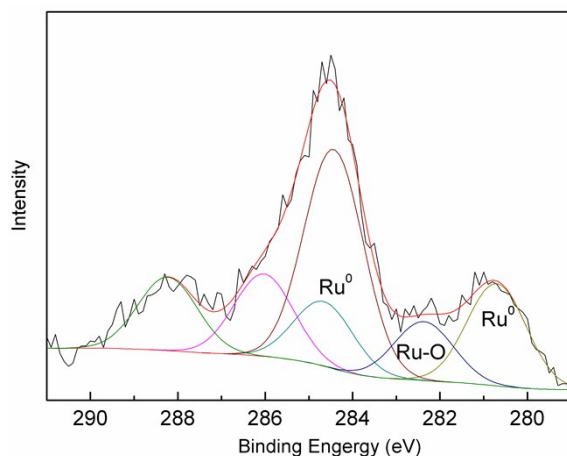


Figure SI-3. Reusability of NiAl-5 catalyst for aerobic oxidation of benzyl alcohol.

Reaction conditions: 0.1 g catalyst, 1 mmol benzyl alcohol, 10 ml toluene, O₂ flow rate, 20 ml/min; temperature, 80 °C; time, 30 min.

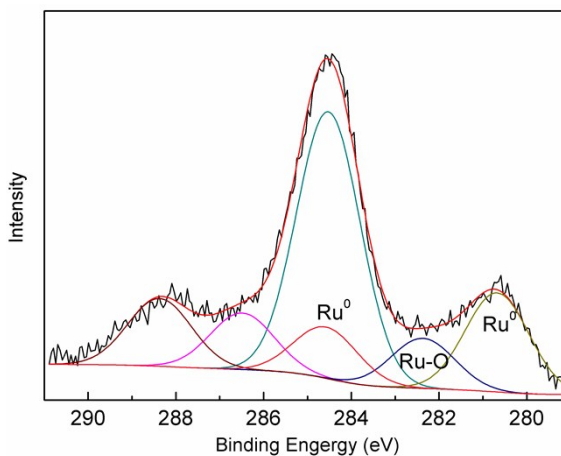
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Figure SI-4. XPS spectra of NiAl-5 catalysts.



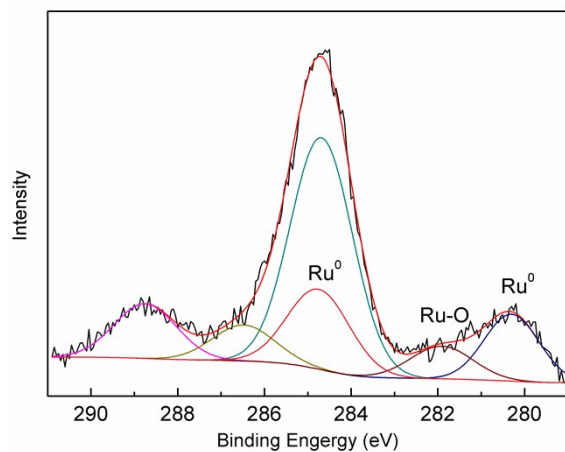
Before reaction:

$$\text{Ru}^0/\text{Ru-O} = 1415.96/815.26 = 1.74$$



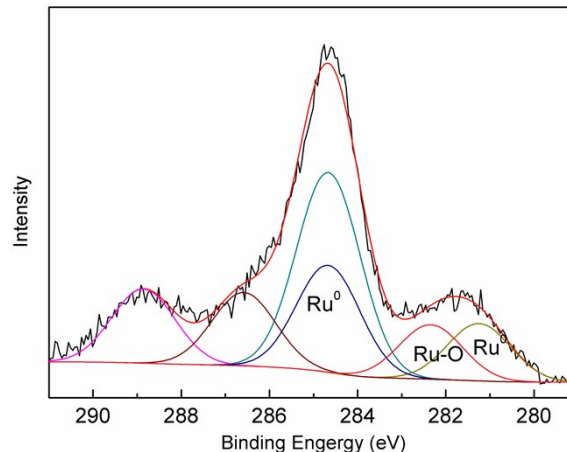
After four cycles:

$$\text{Ru}^0/\text{Ru-O} = 1155.30/582.52 = 1.98$$



In H₂ at 200 °C for 4h:

$$\text{Ru}^0/\text{Ru-O} = 866.40/478.54 = 1.81$$



In O₂ at 200 °C for 4h:

$$\text{Ru}^0/\text{Ru-O} = 1068.50/1014.58 = 1.05$$

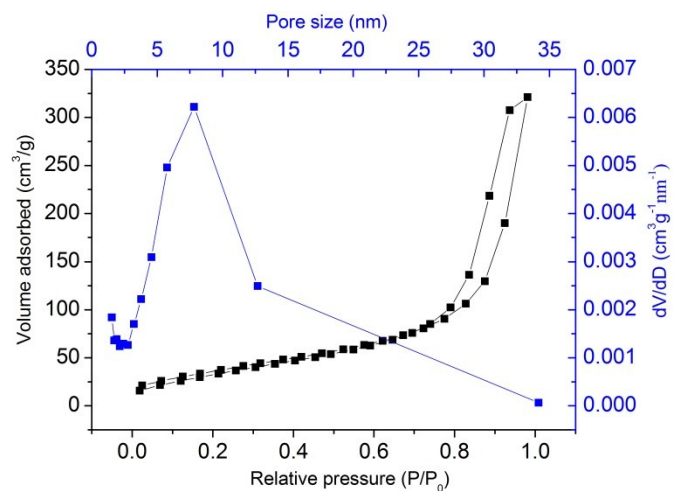


Figure SI-5. Nitrogen physisorption (adsorption/desorption) isotherms and pore size distribution curves of spent NiAl-5 catalyst after four cycles.

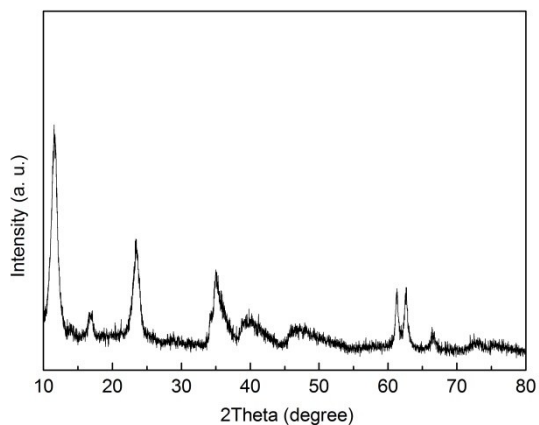


Figure SI-6. Typical XRD pattern of spent NiAl-5 catalyst after four cycles.