

Supporting information

Table S1. Performance comparison of epoxidation of α -pinene over various heterogeneous catalysts.

Entry	Catalyst	Solvent	Oxidation	Initiator	Temp. (°C)	Time (h)	Epoxide Yield (%)	Ref.
1	[Co(NH ₃) ₆]Cl ₃	DMF	40 ml/min air	–	90	5	11.1	<i>This work</i>
2	[Co(NH ₃) ₆]Cl ₃	DMF	40 ml/min air	CHP ^a	90	5	95.7	<i>This work</i>
3	7%CoOx/MOR	DMF	40 ml/min air	CHP	90	5	81.2	<i>Mol. Catal., 2019, 463, 8</i>
4	Co–MOR(L)	DMF	40 ml/min air	TBHP ^b	90	5	84.5	<i>J. Mol. Catal. A. Chem., 2015, 400, 71</i>
5	Co-ZSM-5	DMF	40 ml/min air	TBHP	90	5	79.9	<i>Catal. Commun., 2012, 21, 68</i>
6	NaCsCoY20	DMF	0.55 MPa O ₂	–	100	4	25.0	<i>J. Mol. Catal. A. Chem., 2007, 277, 72</i>
7	[Co ₄ (μ_3 -O) ₄ (μ -C ₆ H ₅ CO ₂) ₄ (4-CNpy) ₄]	Dioxane	15 ml/min air	–	100	24	20.9	<i>J. Mol. Catal. A. Chem., 2004, 223, 39</i>
8	Mn(III) acetate	DMF/toluene	50 ml/min air	–	130	6	35.0	<i>Appl. Catal. A. Gen., 2017, 546, 1-6</i>
9	NCNTs	CH ₃ CN	1.5 MPa O ₂	–	80	4	20.6	<i>Catal. Sci. Technol., 2015, 5, 3935</i>
10	MoCl ₂ O ₂ Ln/TiO ₂	CH ₃ CN	O ₂ /UV light	–	80	18	72.0	<i>Micropor. Mesopor. Mat., 2018, 340, 9</i>
11	Salen Mn ^{III} Cl–ADC	CH ₃ CN	5 ml/min O ₂	Isobutyraldehyde ^c	25	3	84.9	<i>Cellulose., 2018, 25, 1281–1289</i>
12	Biomimetic Iron Catalyst	CH ₃ CN	Air	Ethyl 2-cyclohexanone-carboxylate ^d	25	20	44.0	<i>Angew. Chem. Int. Ed., 2011, 50, 1425–1429</i>

^aCHP: Cumene hydroperoxide, ^bTBHP: *tert*-Butyl hydroperoxide, ^cisobutyraldehyde and ^d2-cyclohexanone-carboxylate: Sacrificial reagent.

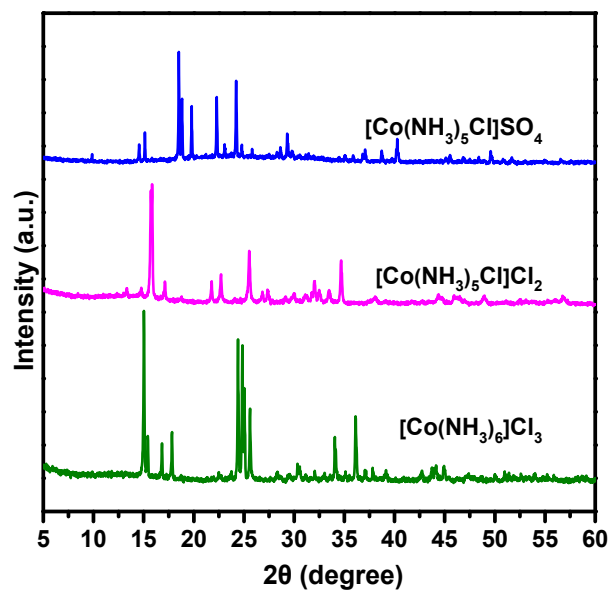


Fig. S1. The XRD patterns of different cobalt ammonia complexes.

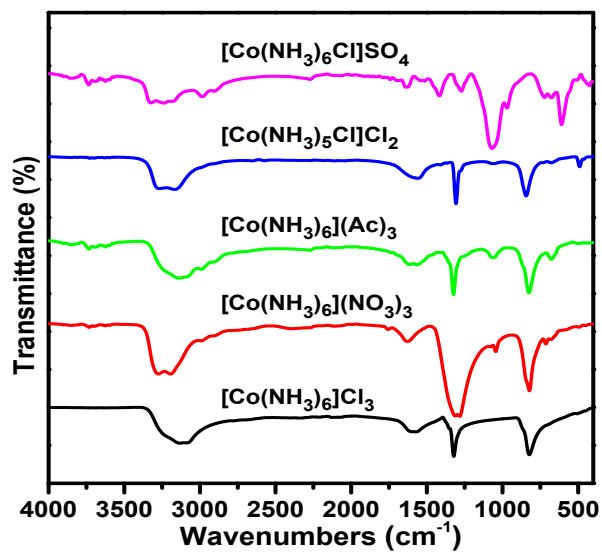


Fig. S2. The IR spectra of different cobalt ammonia complexes.

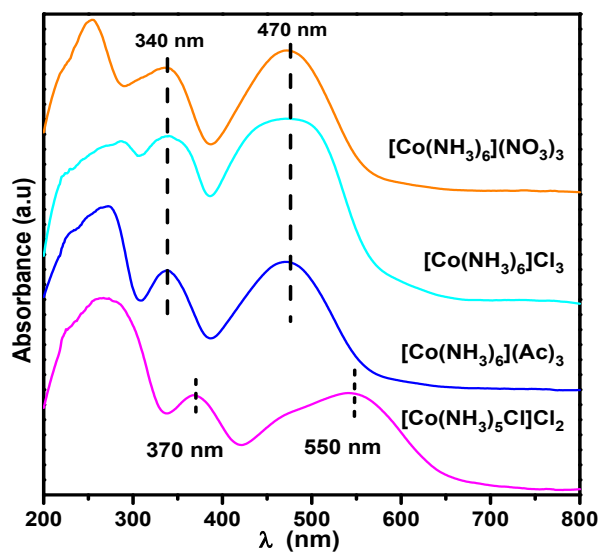


Fig. S3. UV-Vis spectra of synthesized $[\text{Co}(\text{NH}_3)_5\text{Cl}]\text{Cl}_2$, $[\text{Co}(\text{NH}_3)_6](\text{Ac})_3$, $[\text{Co}(\text{NH}_3)_6]\text{Cl}_3$ and $[\text{Co}(\text{NH}_3)_6](\text{NO}_3)_3$.

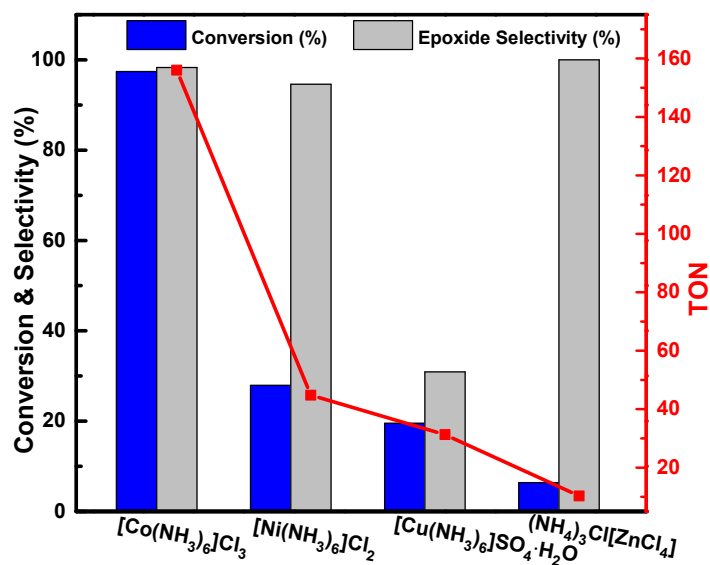


Fig. S4. The catalytic performance of different metal ammonia complexes. Conditions: catalyst, 5 mg; α -pinene, 3 mmol; solvent, DMF 10 g; initiator, CHP 0.3 mmol; temperature, 90 °C; flow rate of air, 40 ml/min; time, 5 h.

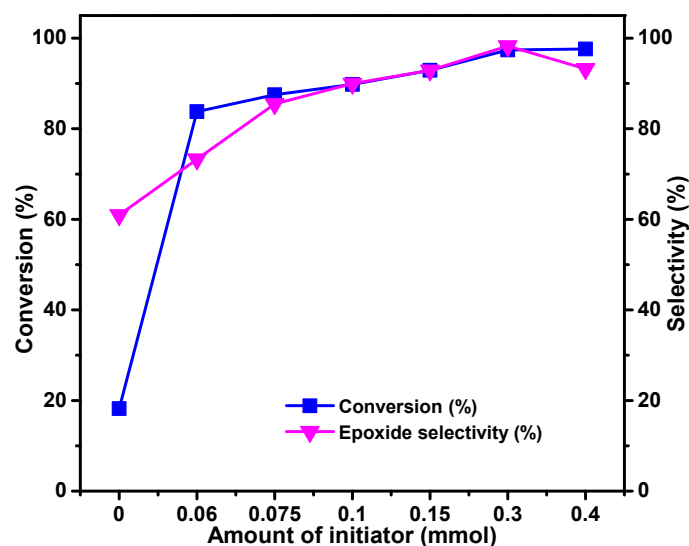


Fig. S5. The amount effect of initiator on the epoxidation of α -pinene.

Conditions: catalyst, 5 mg; α -pinene, 3 mmol; solvent, DMF 10 g; initiator, CHP; temperature, 90 °C; flow rate of air, 40 ml/min; time, 5 h.

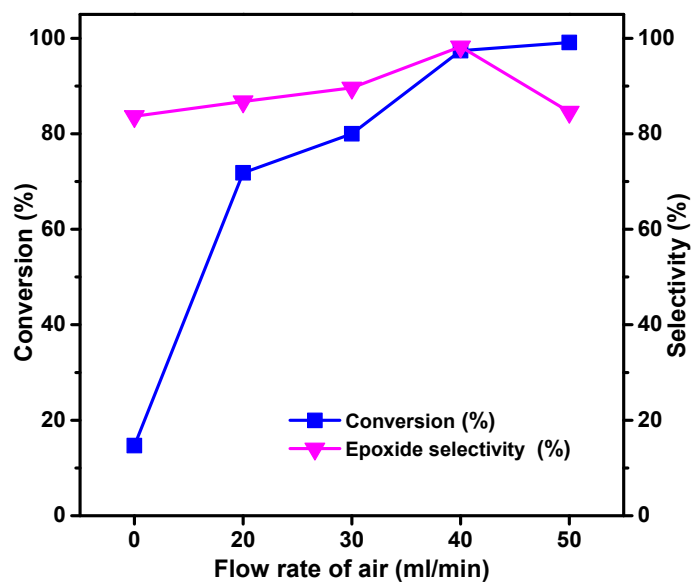


Fig. S6. The flow rate of air on the epoxidation of α -pinene.

Conditions: catalyst, 5 mg; α -pinene, 3 mmol; solvent, DMF 10 g; initiator, CHP; temperature, 90 °C; time, 5 h.

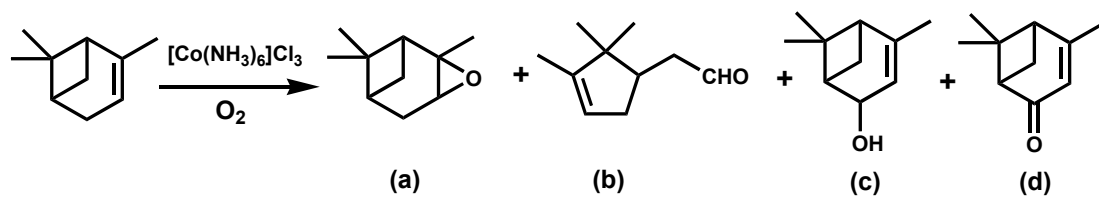


Fig. S7. Schematic illustration for the epoxidation of α -pinene. (a) α -pinene oxide (b) Camphorenal aldehyde (c) Verbenol (d) Verbenone

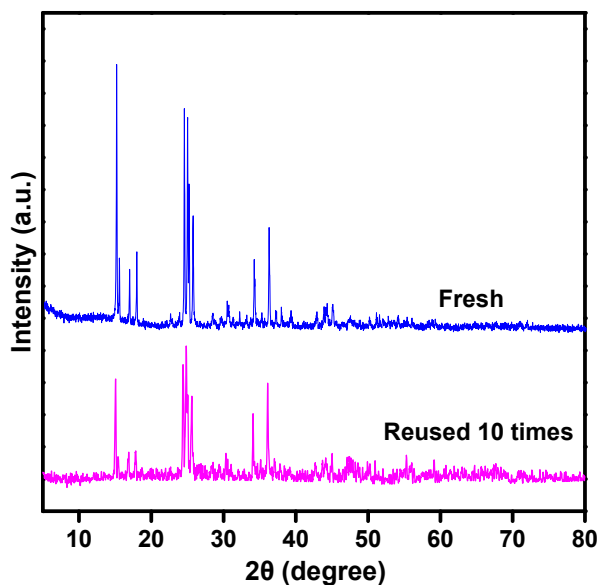


Fig. S8. The XRD patterns of fresh $[\text{Co}(\text{NH}_3)_6]\text{Cl}_3$ and reused 10 times.

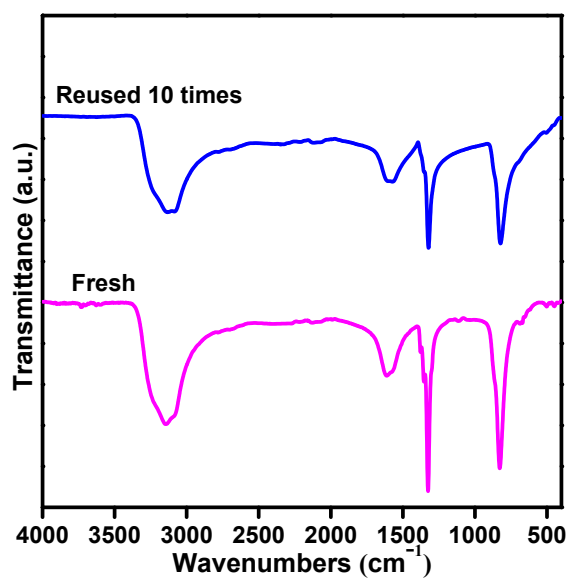


Fig. S9. The IR spectra of fresh $[\text{Co}(\text{NH}_3)_6]\text{Cl}_3$ and reused 10 times.

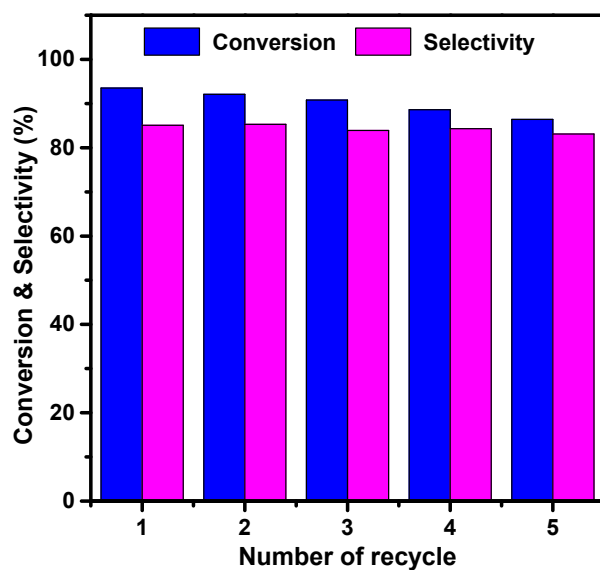


Fig. S10. Results of recyclable performance of $[\text{Co}(\text{NH}_3)_5\text{Cl}]\text{Cl}_2$ catalyst. Conditions: catalyst, $[\text{Co}(\text{NH}_3)_5\text{Cl}]\text{Cl}_2$; α -pinene, 3 mmol; solvent, DMF 10 g; initiator, CHP 0.3 mmol; temperature, 90 °C; flow rate of air, 40 ml/min; time, 5 h.

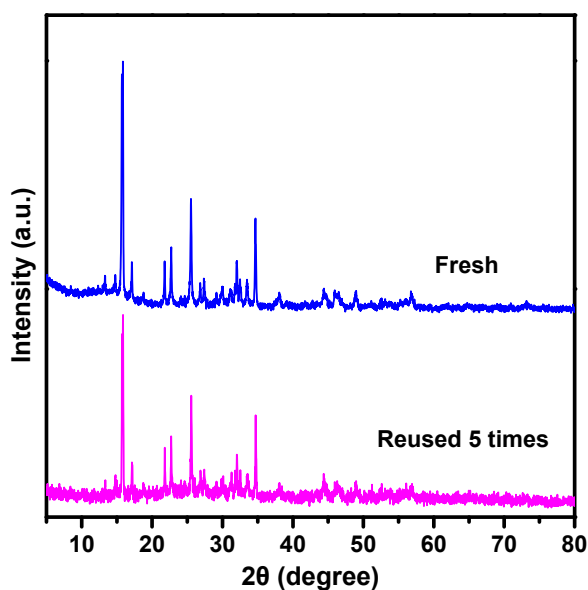


Fig. S11. The XRD patterns of fresh $[\text{Co}(\text{NH}_3)_5\text{Cl}]\text{Cl}_2$ and reused 5 times.

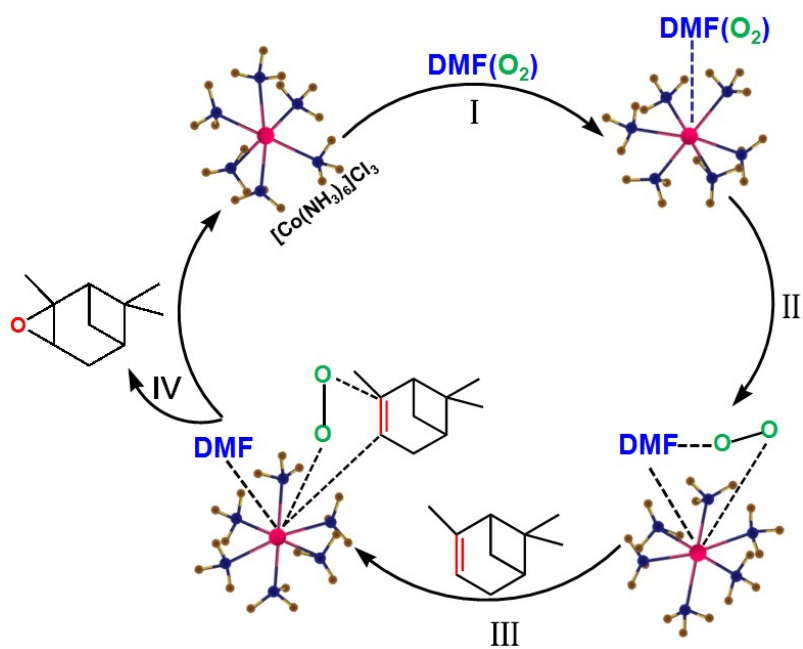


Fig. S12. Proposed mechanism for air activation and epoxidation.