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# **Supporting Information**

## Design and synthesis of dinuclear cobalt(II) complexes derived from strong $\pi$ -

### acidic ligands: crystal structure and studies on the oxidation of sp3 C-H bond

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#### 1. Comparison of bond lengths of crystal structure

For complex **1** Co1A–N<sub>py</sub> is close to the 2.043(1) Å. The bond length Co1A–N<sub>py</sub> of complex **1** is good agreement with our previous reported values<sup>1</sup> and Qian Co-workers<sup>2</sup>, but smaller than the values reported by Nayab and co-workers<sup>3</sup> and larger the values reported by Batisa and co-workers.<sup>4</sup> Co1A–N<sub>im</sub> is close to 2.2008(9) Å. The bond length Co1A–N<sub>im</sub> of complex **1** is smaller than our previous reported values Qian and co-workers<sup>2</sup> and larger the values reported by Batisa and co-workers. Co1A–Cl1A and Co1A–Cl2A is close to the 2.2867(3) Å and 2.3064(4) Å is good agreement with our previous reported value<sup>5</sup> and Nayab and co-workers <sup>6</sup> but smaller than the values reported by Batisa and co-workers.<sup>2</sup> Co1A–O1A is close to the 2.0842(9)Å is good agreement with Ali and co-workers<sup>7</sup> but smaller than the values reported by Jacewicz and co-workers.<sup>8</sup> The bond length of complex **1** were consistent with reported data of high spin Co(II) complexes.<sup>5</sup>

#### 2. Data comparison with literature for the oxidation of different substrates

We compared the catalytic activity in the lights of the reported literature. In our study, catalyst **1** provided the best result for the oxidation of sp<sup>3</sup> C–H bond under optimized reaction condition. Results obtained after catalysis were compared with the literature reports (shown in Table S15). The product formation for fluorene and diphenyl methane was excellent (80%) for catalysts **1–2** and it was efficient as compared to literature reports<sup>9,10</sup> whereas less efficient for the catalytic oxidation of adamantane as compared to previous reports.<sup>11</sup> The product formation were good (>73%) for ethyl benzene as well as substituted ethyl benzene compared to previous literature reports.<sup>12</sup> Catalyst **1-2** showed good catalytic activity for cyclohexane and tetraline compared to other reports.<sup>13-14</sup> Thus, the present complexes may be placed among good catalysts known for oxidation of benzylic as well as aliphatic sp3 C–H bond.







<b>Table S1</b> : Characteristic UV-visible spectroscopic data for ligands (L <sup>1</sup> and L <sup>2</sup> ) and their cobalt (II) complexes (1- 2).								
Entry	Compound	λ <sub>max</sub> (nm)	ε (M <sup>-1</sup> cm <sup>-1</sup> )					
1	L1	227, 284, 361	11237, 7775, 33325					
2	L <sup>2</sup>	210, 241, 358	18257, 12536, 45525					
3	Complex 1	235, 304, 415	5100, 3037, 3450					
4	Complex 2	245, 362, 515	19452, 15650, 9650					



Fig. S9. FT-IR spectrum of complex 1.







Table S2: Crystal data and structural refinement parameters for complex 1				
Empirical formula	C <sub>26</sub> Cl <sub>4</sub> Co <sub>2</sub> N <sub>8</sub> HO			
Formula weight	716.20			
Temperature/K	100.0			
Color	Metallic			
Crystal system	triclinic			
Space group	P-1			
a/Å	9.1270(3)			
b/Å	9.5751(3)			
c/Å	9.8287(3)			
α/°	65.8370(10)			
β/°	70.6870(10)			
γ/°	89.6210(10)			
Volume/Å <sup>3</sup>	731.27(4)			
Z	1			
$\rho_{calc}g/cm^3$	1.626			
µ/mm⁻¹	1.536			
F(000)	364.0			
Crystal size/mm <sup>3</sup>	$0.177 \times 0.051 \times 0.021$			
Radiation	ΜοΚα (λ = 0.71073)			
20 range for data collection/°	4.716 to 61.248			
Index ranges	$-13 \le h \le 13, -13 \le k \le 13, -14 \le l \le 14$			
Reflections collected	36244			
Independent reflections	4500 [R <sub>int</sub> = 0.0299, R <sub>sigma</sub> = 0.0164]			
Data/restraints/parameters	4500/3/185			

Goodness-of-fit on F <sup>2</sup>	1.065			
Final R indexes [I>=2σ (I)]	$R_1 = 0.0245$ , $wR_2 = 0.0630$			
Final R indexes [all data]	$R_1 = 0.0262, wR_2 = 0.0644$			
Largest diff. peak/hole / e Å <sup>-3</sup>	0.65/-0.29			
<sup>a</sup> GOF = $[\Sigma[w(Fo^2-Fc^2)^2]/M-N)]^{1/2}$ (M = number of reflections, N = number of parameters refined). <sup>b</sup> R1=				
$\Sigma \  Fo  -  Fc   / \Sigma  Fo , {}^{c}wR_{2} = [\Sigma [w(Fo^{2} - Fc^{2})_{2}] / \Sigma [w(Fo_{2})_{2}]]^{1/2}$				

Table S3: Selected bond distances and bond angles of complex 1							
Bond Lengths (Å)		Bond Angl	es(°)				
Co1A—Cl1A	2.2867(3)	Cl1A—Co1A—Cl2A	130.07(1)				
Co1A—Cl2A	2.3064(4)	O1A—Co1A—Cl1A	94.55(3)				
Co1A—O1A	2.0842(9)	N2 —Co1A —Cl1A	86.42(3)				
Co1A—N2	2.2008(9)	Cl1A—Co1A—N1	111.04(3)				
Co1A—N1	2.043(1)	Cl2A—Co1A—O1A	90.01(3)				
		Cl2A—Co1A—N2	93.75(3)				
		Cl2A —Co1A —N1	117.38(3)				
		01A Co1A N2	174.19(4)				
		01A Co1A N1	98.38(4)				
		N2 —Co1A —N1	75.95(4)				

Oxidant, Catalyst

Solvent, Temp., Time



Scheme S1: Oxidation of fluorene

<sup>a</sup>All reactions were carried out in CH<sub>3</sub>CN at 60 °C. **Catalysts** (0.04 mmol), alkanes (1 mmol), and TBHP (8 mmol).

<sup>b</sup>TON = mmol of product/mmol of catalyst loading

<sup>c</sup>TOF = TON / time (in hours).

Table S4: Oxidation of fluorene using different type						
complex	complexes (1-2)					
Complex	% Yield					
$Co_2(L^1)(H_2O)_2Cl_4$ (1)	94					
$Co_2(L^2)(H_2O)_2Cl_4(2)$	91					
Ligands	Trace					
CoCl <sub>2</sub> .6H <sub>2</sub> O	Trace					



Fig. S15. Oxidation of fluorene using different type complexes (1-2)

Table S5	Table S5: Effect of different oxidants on the oxidation of fluorene using Complex 1.								
Entry	Substrate		Amount (mm	ol)	Solvent	% Yield			
		Fluorene	Complex 1	Oxidant					
1	Fluorene	1	0.04	TBHP(8)	CH₃CN	94			
2	Fluorene	1	0.04	H <sub>2</sub> O <sub>2</sub> (8)	CH₃CN	41			
3	Fluorene	1	0.04	mCPBA(8)	CH₃CN	20			



Table S6: Varying amount of Complex 1.								
Entry	Substrate	Amount (mmol)			Solvent	% Yield		
		Fluorene	Complex 1	ТВНР	•			
1	Fluorene	1	0.01	8	CH₃CN	18%		
2	Fluorene	1	0.02	8	CH₃CN	42%		
3	Fluorene	1	0.03	8	CH₃CN	58%		
4	Fluorene	1	0.04	8	CH₃CN	94%		
5	Fluorene	1	0.05	8	CH₃CN	94%		
	Note: All the data was recorded after 12 h							

Fig. S16. Effect of different oxidants on the oxidation of fluorene using Complex 1



Fig. S17. Varying amount of Complex 1.

Entry	Substrate	A	mount (mmol)		Solvent	% Yield
		Fluorene	Complex 1	TBHP		
1	Fluorene	1	0.04	2	CH₃CN	15%
2	Fluorene	1	0.04	4	CH₃CN	34%
3	Fluorene	1	0.04	6	CH₃CN	72%
4	Fluorene	1	0.04	8	CH₃CN	94%
5	Fluorene	1	0.04	10	CH₃CN	94%



Fig. S18. Varying amount of oxidant TBHP with constant amount of Complex 1.

Table S8: Effect of temperature on the oxidation of fluorene using Complex 1								
Entry	Substrate	Amount (mmol)			Temp	% Yield		
		Fluorene	Complex 1	ТВНР	- °C			
1	Fluorene	1	0.04	8	0	0		
2	Fluorene	1	0.04	8	20	5		
3	Fluorene	1	0.04	8	40	45		
4	Fluorene	1	0.04	8	60	61		
5	Fluorene	1	0.04	8	80	94		



Fig. S19. Effect of temperature on the oxidation of fluorene using Complex 1

Table S9: Effect of solvent on the oxidation of fluorene using Complex 1.

Entry	Substrate	Amount (mmol)			Solvent	% Yield
		Fluorene	Complex 1	ТВНР	-	
1	Fluorene	1	0.04	8	CH₃CN	94
2	Fluorene	1	0.04	8	CH₃OH	20
3	Fluorene	1	0.04	8	Xylene	38
4	Fluorene	1	0.04	8	Benzene	47
5	Fluorene	1	0.04	8	Toluene	58
6	Fluorene	1	0.04	8	DMSO	52
7	Fluorene	1	0.04	8	DMF	42
8	Fluorene	1	0.04	8	THF	12
9	Fluorene	1	0.04	8	DCM	22



Fig. S20. Effect of solvent on the oxidation of fluorene using Complex 1.

Table S10: Oxidation of fluorene at different time using Complex 1.					
Entry	Substrate	Time (h)	Solvent	% Yield	
1	Fluorene	1	CH₃CN	8	
2	Fluorene	2	CH₃CN	12	
3	Fluorene	3	CH₃CN	16	
4	Fluorene	4	CH₃CN	22	
5	Fluorene	5	CH₃CN	26	
6	Fluorene	6	CH₃CN	32	
7	Fluorene	7	CH₃CN	44	
8	Fluorene	8	CH₃CN	51	
9	Fluorene	9	CH₃CN	57	
10	Fluorene	10	CH₃CN	72	
11	Fluorene	11	CH₃CN	82	
12	Fluorene	12	CH₃CN	94	





















Table S11: Data analysis for oxidation of cyclohexane<sup>a</sup>

Complex	% Conve	ersion	TON⁵	۲OF(h <sup>-1</sup> )	
	Α	В			
Co <sub>2</sub> (L <sup>1</sup> )(H <sub>2</sub> O) <sub>2</sub> Cl <sub>4</sub> ( <b>1</b> )	20	15	5	0.4	
$Co_2(L^2)(H_2O)_2Cl_4(2)$	31	18	7	0.6	

<sup>a</sup>All reactions were carried out in  $CH_3CN$  at 60 °C. Cyclohexane (1 mmol), **Complex** (0.04 mmol), and TBHP (8 mmol).

% Conversion = mmole of product/ mmole of reactant

<sup>b</sup>TON = mmol of product/mmol of catalyst loading

<sup>c</sup>TOF = TON / time (in hours).

Table S12: Data analysis for oxidation of phenylacetonitrile <sup>a</sup>					
Complex	% Conversion	TON <sup>b</sup>	°TOF (h⁻¹)		
Co <sub>2</sub> (L <sup>1</sup> )(H <sub>2</sub> O) <sub>2</sub> Cl <sub>4</sub> ( <b>1</b> )	51	13	1		
$Co_2(L^2)(H_2O)_2CI_4(2)$	48	12	1		
<sup>a</sup> All reactions were carried out in CH <sub>3</sub> CN at 60 °C. Phenylacetonitrile (1 mmol), <b>Complex</b> (0.04 mmol), and TBHP (8 mmol).					
% Conversion = mmole of product/ mmole of reactant					
<sup>b</sup> TON = mmol of product/mmol of catalyst loading					
°TOF = TON / time (in hours).					

Table S13: Data analysis for oxidation of 9,10-dihydroanthracene <sup>a</sup>					
Complex	% Conversion	TON <sup>b</sup>	℃TOF (h <sup>-1</sup> )		
Co <sub>2</sub> (L <sup>1</sup> )(H <sub>2</sub> O) <sub>2</sub> Cl <sub>4</sub> ( <b>1</b> )	59	14	1.3		
$Co_2(L^2)(H_2O)_2Cl_4(2)$	52	13	1		
<sup>a</sup> All reactions were carried out in CH <sub>3</sub> CN at 60 °C. 9,10-dihydroanthracene (1 mmol), <b>Complex</b> (0.04 mmol), and TBHP (8 mmol).					

% Conversion = mmole of product/ mmole of reactant

<sup>b</sup>TON = mmol of product/mmol of catalyst loading

<sup>c</sup>TOF = TON / time (in hours).

Table S14: Data analysis for oxidation of adamantane <sup>a</sup>						
Complex	% Conversion		TON <sup>b</sup>	℃TOF (h <sup>-1</sup> )		
	Α	B + C	-			
Co <sub>2</sub> (L <sup>1</sup> )(H <sub>2</sub> O) <sub>2</sub> Cl <sub>4</sub>	36	15	13	1		
$Co_2(L^2)(H_2O)_2Cl_4$	28	13	10	0.8		
<sup>a</sup> All reactions were carried out in CH <sub>3</sub> CN at 60 °C. adamantane (1 mmol), <b>Complex</b> (0.04 mmol), and TBHP (8 mmol).						
% Conversion = mmole of p	product/ mmole o	f reactant				
~ TON = mmor of product/mmor of catalyst loading						

<sup>c</sup>TOF = TON / time (in hours).

















Fig. S42: Packing diagram of complex 1 (projection at c\*-axis).

Table S15: Comparative study of literature for the oxidation of different alkanes					
Catalyst	Substrate	Product	% conversion or	Reference	
			% Yield		
[Co(bis(imino)pyridine) (OTf) <sub>2</sub> ]	Cyclohexane	Cyclohexanol	33	13	
[Co(bis(amino)pyridine) (OTf) <sub>2</sub> ]		Cyclohexanone			
[Cp*Co(bipy)Cl]PF <sub>6</sub>	Cyclohexane	Cyclohexanol	24	15	
[Cp*Co(bipy)I]PF <sub>6</sub>					
[Cp*Co(phen)I]PF <sub>6</sub>					
[Co([PhB(MeIm)₃])(acac)(MeImH)] <sup>+</sup>	Cyclohexane	Cyclohexanol	29	16	
[Co([PhB(MeIm) <sub>3</sub> ]) <sub>2</sub> ] <sup>+</sup>		Cyclohexanone			
Complex 1	Cyclohexane	Cyclohexanol	35	This work	
		Cyclohexanone			
Complex 2	Cyclohexane	Cyclohexanol	39	This work	
		Cyclohexanone			
$[Co{C6H4C(NH2)NC(ONCMe2)2]2]2+$	Adamantane	1-adamantanol	66		
		2-adamantanol			
		2-adamantanone			
Complex 1	Adamantane	1-adamantanol	51	This work	
		2-adamantanol			
		2-adamantanone			

Complex 2	Adamantane	1-adamantanol	41	This work
		2-adamantanol		
		2-adamantanone		
Cobalt complex supported with MCM	Diphenyl	Benzophenone	29	10
Nano particle.	methane			
[Co(tptz)Cl <sub>2</sub> ].2H <sub>2</sub> O	Diphenyl	Benzophenone	64	9
	methane			
Complex 1	Diphenyl	Benzophenone	72	This work
	methane			
Complex 2	Diphenyl	Benzophenone	35	This work
	methane			
[Co(tptz)Cl <sub>2</sub> ].2H <sub>2</sub> O	Fluorene	Fluorenone	99	9
Complex 1	Fluorene	Fluorenone	94	This work
Complex 2	Fluorene	Fluorenone	91	This work
	1,2,3,4-tetra	3,4-dihydro	52	14
	hydronaphthale	naphthalen-1(2 <i>H</i> )-		
	ne	one		
Complex 1	1,2,3,4-tetra	3,4-dihydro	80	This work
	hydronaphthale	naphthalen-1(2 <i>H</i> )-		
	ne	one		
Complex 2	1,2,3,4-tetra	3,4-dihydro	78	This work
	hydronaphthale	naphthalen-1(2 <i>H</i> )-		
	ne	one		
Silica supported cobalt complexes	Ethylbenzene	Acetophenone	69	12
Complex 1	Ethylbenzene	Acetophenone	79	This work
Complex <b>2</b>	Ethylbenzene	Acetophenone	75	This work
Silica supported cobalt complexes	1-bromo-4-	1-(4-bromo	75	12
	ethylbenzene	phenyl)ethan-1-one		
Complex 1	1-bromo-4-	1-(4-bromo	82	This work
	ethylbenzene	phenyl)ethan-1-one		
Complex 2	1-bromo-4-	1-(4-bromo	78	This work
	ethylbenzene	phenyl)ethan-1-one		
Silica supported cobalt complexes	1-chloro-4-	1-(4-chloro	76	12
	ethylbenzene	phenyl)ethan-1-one		
Complex 1	1-chloro-4-	1-(4-chloro	79	This work
	ethylbenzene	phenyl)ethan-1-one		
Complex 2	1-chloro-4-	1-(4-chloro	73	This work
	ethylbenzene	phenyl)ethan-1-one		

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