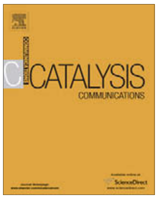
Catalysis Communications 11 (2010) 792–796

Contents lists available at ScienceDirect

Catalysis Communications

journal homepage: www.elsevier.com/locate/catcom



Electronic effects of substituents on the oxidation potentials of vanadyl complexes with tetradentate Schiff base ligands derived from 1,2-propylenediamine

Saeed Rayati [a,*], Akbar Ghaemi [b], Nasrin Sadeghzadeh [a]

[a] *Department of Chemistry, K.N. Toosi University of Technology, P.O. Box 16315-1618, Tehran, Iran*
[b] *Department of Chemistry, Islamic Azad University of Saveh Branch, P.O. Box 39187-366, Saveh, Iran*

ARTICLE INFO

Article history:
Received 24 December 2009
Received in revised form 12 February 2010
Accepted 15 February 2010
Available online 19 February 2010

Keywords:
Schiff base
Oxovanadium
Catalyst
Olefins
tert-Butylhydroperoxide

ABSTRACT

Two tetradentate Schiff base ligands (H_2L^{1-2}) (H_2L^1 = bis(2-hydroxy-3-methoxy-benzaldehyde)-1,2-propandiimine, H_2L^2 = bis(2-hydroxy-4-methoxy-acetophenone)-1,2-propandiimine) were prepared by reaction of 1,2-propylenediamine and *o*-hydroxycarbonyls compounds containing a methoxy group and characterized by elemental analysis, FT-IR, ^1H and ^{13}C NMR. The vanadyl complexes were synthesized and characterized. The catalytic potential of these complexes was tested for the oxidation of cyclooctene and styrene using *tert*-butylhydroperoxide (TBHP) as oxidant. It has been shown that the presence of electron-donating substituents on the aromatic ring as well as the imine bond can effectively improve the catalytic activity and the product selectivity of catalysts.

1. Introduction

The oxidation of hydrocarbons using transition metals Schiff base complexes has attracted both academic and industrial interest [1]. They are efficient catalysts in homogeneous and heterogeneous conditions. The catalytic activity of these complexes depends on the nature of substituents as well as the metal centre [1–4]. The interest in vanadium coordination chemistry promoted by the presence of this element in biological systems [5], and by catalytic [4], inhibitory [6], medicinal [7,8] and structural [9–11] properties of its compounds. The potential catalytic abilities of vanadium compounds have lead to an increasing interest in vanadium coordination chemistry in recent years [2]. Homogeneous catalysts of oxovanadium(IV) complexes have been shown to induce organic reactions such as the oxidation of sulfides to sulfoxides and sulfones [12–14], the epoxidation of alkenes [15–18], the hydroxylation of hydrocarbons [19], and the oxidation of alcohols to aldehydes and ketones [20,21]. These studies are indicative that oxovanadium(IV) complexes are potential catalysts to influence the yield and selectivity in chemical transformation.

In continuation of our efforts to develop new oxovanadium(IV)-based homogeneous oxidation catalysts, we report herein the synthesis of two new oxovanadium(IV) Schiff base complexes with electron donor substituents that exhibited higher potential catalytic activity and product selectivity for the oxidation of cyclooctene and styrene with *tert*-butylhydroperoxide (TBHP).

2. Experimental

2.1. Instruments and reagents

All chemicals were supplied by either Merck or Fluka. Solvents (ethanol and acetonitrile) were dried and distilled by standard methods before use [22]. Chemicals were used as received. Infrared spectra were recorded as KBr pellets using a Unicam Matson 1000 FT-IR. Elemental analyses (C, H, N) were performed using a Heraeus Elemental Analyzer CHN-O-Rapid (Elemental-Analyze system KBr pellets, Gmbh, West Germany). ^1H and ^{13}C NMR spectra were obtained in $CDCl_3$ solution on a Bruker FT-NMR 250 (250 MHz) spectrometer. A varian (AA220) flame atomic absorption spectrometer was used for vanadium determination. A Metrohm 757 VA Computrace was employed to evaluate electrochemical measurements at room temperature (25 °C) under nitrogen with 0.1 M tetrabutylammonium hexafluorophosphate (TBAH) as the supporting electrode. The reaction products of oxidation analyzed by HP Agilent 6890 gas chromatograph equipped with a HP-5 capillary column (phenyl methyl siloxane 30 m × 320 μm × 0.25 μm) and flame-ionization detector.

2.2. Synthesis of ligands

2.2.1. Bis(2-hydroxy-3-methoxy benzaldehyde)-1,2-propandiimine (H_2L^1)

H_2L^1 was prepared according to the described procedure [23]. To a stirred ethanolic solution (20 ml) of 1,2-propylenediamine

---

[*] Corresponding author. Tel.: +98 21 22850266; fax: +98 21 22853650.
E-mail addresses: srayati@yahoo.com, rayati@kntu.ac.ir (S. Rayati).

1566-7367/$ - see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.catcom.2010.02.017

(0.074 g, 1 mmol), 2-hydroxy-3-methoxybenzaldehyde (0.308 g, 2 mmol) was added. The bright yellow solution was stirred and heated to reflux for 1 h. A yellow precipitate was obtained that was filtered off and washed with diethyl ether. Yield (95%), melting point 125 °C. Analysis calculated for $C_{19}H_{22}N_2O_4$ (342.2): C, 68.68; H, 6.42; N, 8.18. Found: C, 68.93; H, 6.01; N, 8.72%. Selected FT-IR data, υ (cm^{-1}): 3423 (O–H), 2923–3015 (C–H), 1630 (C=N), 1561 (C=C), 1031 (C–O). ^{1}H NMR (δ): 1.39, 1.41 (d, 3H, $NCH_2CH(CH_3)N$), 3.58, 3.65 (d, 2H, $NCH_2CH(CH_3)N$), 3.65–3.93 (m, 1H, $NCH_2CH(CH_3)N$), 4.03, 4.16 (s, 6H, OMe), 6.44–6.90 (m, 6H, Ar*H*), 8.27, 8.31 (s, 2H, *H*C=N), 13.70, 13.78 (s, 2H, OH). ^{13}C{^{1}H}NMR (δ): 20.4 ($NCH_2CH(CH_3)N$), 55.9 ($NCH_2CH(CH_3)N$), 56.0 ($NCH_2CH(CH_3)N$), 64.7, 65.5 (OCH_3), 113.9, 151.3 (aromatic C), 164.5, 166.4 (C=N).

2.2.2. Bis(2-hydroxy-4-methoxy acetophenone)-1,2-propandiimine (H_2L^2)

To a stirred ethanolic solution (20 ml) of 1,2-propylenediamine (0.074 g, 1 mmol), 2-hydroxy-4-methoxyacetophenone (0.304 g, 2 mmol) was added. The mixture was stirred and heated to reflux for 1 h. A yellow precipitate was obtained that was filtered off and washed with diethyl ether. Yield (90%), melting point 130 °C. Analysis calculated for $C_{21}H_{26}N_2O_4$ (370.2): C, 68.12; H, 7.02; N, 7.56. Found: C, 68.36; H, 6.82; N, 7.83%. Selected FT-IR data, υ (cm^{-1}): 3400 (O–H), 2916–3000 (C–H), 1623 (C=N), 1560 (C=C), 1160 (C–O). ^{1}H NMR (δ): 1.39, 1.50 (d, 3H, $NCH_2CH(CH_3)N$), 2.50, 2.60 (d, 2H, $NCH_2CH(CH_3)N$), 3.40–3.89 (m, 1H, $NCH_2CH(CH_3)N$), 3.76, 3.81 (s, 6H, O7.02–6.82Me), 6.24–7.70 (m, 6H, Ar*H*), 2.25, 2.35 (s, 6H, $(CH_3)C$=N), 12.75, 12.81 (s, 2H, OH). ^{13}C{^{1}H}NMR (δ): 14.24, 14.43 ($NCH_2CH(CH_3)N$), 54.6 ($NCH_2CH(CH_3)N$), 53.7 ($NCH_2CH(CH_3)N$), 55.2 (OCH_3), 101.9, 168.8 (aromatic C), 170.5, 172.0 (*C*=N), 20.0 ($(CH_3)C$=N).

2.3. Preparation of vanadyl complexes (VOL^x (x = 1–2))

The complexes were prepared by a general procedure: the ligand, H_2L^1 (0.34 g, 1 mmol) or H_2L^2 (0.40 g, 1 mmol) was dissolved in 30 ml of ethanol. An ethanolic solution of oxobis(pentane-2,4-dionato)vanadium(IV) (0.265 g, 1 mmol) was added and the reaction mixture was refluxed for 2 h. The colored solution was concentrated to yield colored powders. The products washed with warm ethanol. General structure of oxovanadium(IV) complexes have been shown in Fig. 1. The melting points, yields, and elemental analyses for the complexes are given in Table 1.

2.4. General oxidation procedure

Catalytic experiments were carried out in a 50 ml round bottom flask fitted with a water condenser. In a typical procedure, 0.032 mmol vanadyl complex was dissolved in 10 ml solvent (chloroform, acetonitrile or dichloromethane). Then 10 mmol alkene (cyclooctene or styrene) was added to the reaction mixture and 30 mmol TBHP was added. The reaction mixture was refluxed for 6 h. The oxidation products were identified by comparison with authentic samples (retention times in GC).

3. Results and discussion

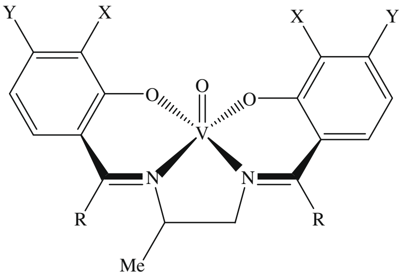
3.1. Characterization of the ligands and oxovanadium(IV) complexes

3.1.1. IR spectral studies

A practical list of IR spectral data is presented in Table 2. Comparison of the spectra of the complexes with the ligands provides evidence for the coordination mode of ligand in catalysts. The Schiff base ligands exhibit a broad band around 2969–3430 cm^{-1} region due to the presence of extensive hydrogen bonding between phenolic hydrogen and azomethine nitrogen atoms. Absence of this band in the spectra of complexes indicates the breaking of hydrogen bonding followed by coordination of phenolic oxygen to the metal ion after deprotonation. A sharp band appearing at 1623–1630 cm^{-1} due to υ(C=N) (azomethine), shifts to lower wave number by 15–16 cm^{-1} and appears at 1607–1623 cm^{-1}. This indicates the involvement of azomethine nitrogen in coordination. IR spectra of VOL^1 shows υ(V=O) at 969 cm^{-1} and for VOL^2 at 984 cm^{-1}. Tetradentate Schiff base oxovanadium(IV) complexes with υ(V=O) around 970 cm^{-1} are in monomeric form. Thus, both of VOL^1 and VOL^2 are assigned to have monomeric structure [24].

3.1.2. Electrochemical studies

The electrochemical properties of the VO–Schiff base complexes (VOL^1 and VOL^2) were investigated in CH_3CN, using TBAH (0.1 M) as the supporting electrolyte by cyclic voltammetry. Fig. 2 shows the cyclic voltammograms of 0.01 mmol of VOL^1 complex in acetonitrile at various scan rates. Both vanadyl complexes exhibit a



Complex	R	X, Y
VOL^1	H	**X**: OMe, **Y**: H
VOL^2	Me	**X**: H, **Y**: OMe

Fig. 1. Oxovanadium(IV) complexes used in this study.

Table 1
Physical and analytical data of the complexes.

	Compound formula	Formula weight	Yield (%)	Color	Found (calculated)			
					%C	%H	%N	%V
VOL^1	$VC_{19}H_{20}N_2O_5$	406.9	90	Green	56.39 (56.08)	4.61 (4.91)	6.93 (6.88)	12.46 (12.51)
VOL^2	$VC_{21}H_{24}N_2O_5$	432.7	85	Green	58.03 (58.34)	5.14 (5.55)	6.79 (6.47)	11.89 (11.77)

Table 2
IR spectral data of ligands and the vanadium complexes.

Compound	Selective IR bands (cm^{-1})	
	V=O	C=N
H_2L^1	–	1630
H_2L^2	–	1623
VOL^1	969	1615
VOL^2	984	1607

reversible electrochemical behavior as shown in the cyclic voltammograms of Fig. 2. Redox potentials for two complexes are given in Table 3. Introduction of electron-donating substituents instead of electron-withdrawing ones [14] decreases the oxidation potential of V(IV).

The presence of two methoxy substituents at *ortho* positions of VOL^1 phenyl groups enhances the electron densities on the ligating atoms of the ligand through π-donation to the phenyl groups. It should be noted that the methoxy substituent enhances the electron density at the *ortho* and *para* positions with respect to the substituent. Therefore, the "O" atoms of VOL^1 are expected to be more affected by the π-donation of methoxy groups whereas in the case of VOL^2 the "N" atoms are the more affected one. Furthermore, in VOL^2, there are two methyl groups at imine moieties of the ligand as well as the two methoxy residues at the phenyl rings. In addition to the π-donation from –OMe to the ligand, methyl group may also be considered as a π-donor due the hyperconjugation effect of this group [25]. Accordingly, the metal centre of VOL^2 is expected to be more electron-rich than that of VOL^1 (vide infra) and the redox potentials of V(V)/V(IV) shifts in the cathodic direction and the complex become easier to oxidize.

3.2. Catalytic activity studies

3.2.1. Oxidation of cyclooctene

The oxidation of cyclooctene, catalyzed by VOL^1 and VOL^2, was carried out using *tert*-butylhydroperoxide as an oxidant to give cyclooctene oxide as the sole product. To achieve the maximum epoxide yield, several parameters such as, the effect of oxidant concentration (moles of TBHP per moles of cyclooctene), solvent and temperature of the reaction have been optimized. Different TBHP/cyclooctene molar ratios (0.5:1, 1:1, 2:1 and 3:1) were considered while keeping the fixed amount of cyclooctene (1.102 g, 10 mmol) and catalyst VOL^1 (0.013 g, 0.032 mmol) in 10 ml of $CHCl_3$ at 60 °C. Increasing the TBHP/cyclooctene ratio from 0.5:1 to 3:1 the conversion increases from 32% to 94%, probably due to fast formation of active intermediate in the presence of higher amount of oxidant.

Table 3
Electrochemical data for the oxidation of oxovanadium(IV) complexes in CH_3CN.

Complex	E_p^a (V)	E_p^c (V)	$E_{1/2}$ (V)	ΔE_p (V)
VOL^1	0.614	0.504	0.559	0.110
VOL^2	0.595	0.490	0.545	0.105

The same result was obtained using VOL^2 and in the 3:1 M ratio (TBHP/cyclooctene), the maximum conversion (99.8%) was achieved. Another important feature of this catalytic system with respect to the similar systems [26] is the excellent product selectivity (100%) for epoxide that may be a result of using electron rich vanadyl Schiff base complexes.

The influence of the solvent nature in the catalytic epoxidation of cyclooctene by VOL^1 and VOL^2 has been investigated. Chloroform, acetonitrile and dichloromethane were used as solvent and the highest conversion (98% in VOL^1 and 98.8% in VOL^2) was obtained in chloroform. It was observed that the catalytic activity of the vanadyl complexes decreases in order chloroform > acetonitrile > dichloromethane. Hundred percentage selectivity for epoxide formation was obtained for all solvents. More efficient oxidation of cyclooctene in chloroform relative to dichloromethane at reflux condition seems to be due to the higher boiling point of chloroform. The decreased conversion of cyclooctene in acetonitrile with respect to that of chloroform, in spite of the higher b.p of acetonitrile may be due to the donor ability of acetonitrile which can compete with the oxidant in coordination to the metal centre (see the proposed mechanism, Fig. 3).

In order to get the best reaction temperature, reaction mixture was stirred at 40 °C, 50 °C and 60 °C and for both catalysts, a maximum of conversion of cyclooctene were obtained at 60 °C (93.6% for VOL^1 and 98.8% in VOL^2).

3.2.2. Oxidation of styrene

The oxidation of styrene with VOL^1 and VOL^2 gave styrene oxide as dominant product and benzaldehyde as by product. Reaction temperature, solvent and oxidant/styrene molar ratio have been optimized. In order to obtain the best oxidant/styrene molar ratio, three different TBHP/styrene molar ratios (0.5:1, 1:1, 2:1 and 3:1) were considered while keeping the fixed amount of styrene (1.04 g, 10 mmol) and catalyst (0.032 mmol) in 10 ml of CH_3CN for the reaction at 60 °C. A maximum of 48% conversion for VOL^1 (with 87.5% selectivity for epoxide formation) and 88% conversion for VOL^2 (with 99% selectivity for epoxide formation) was obtained at 3:1 M ratio. Further increment in oxidant concentration hardly improved the conversion. Table 4 shows product selectivity and percent conversion of styrene with both catalysts. Oxidation of styrene has been influenced by the nature of solvent. Three different

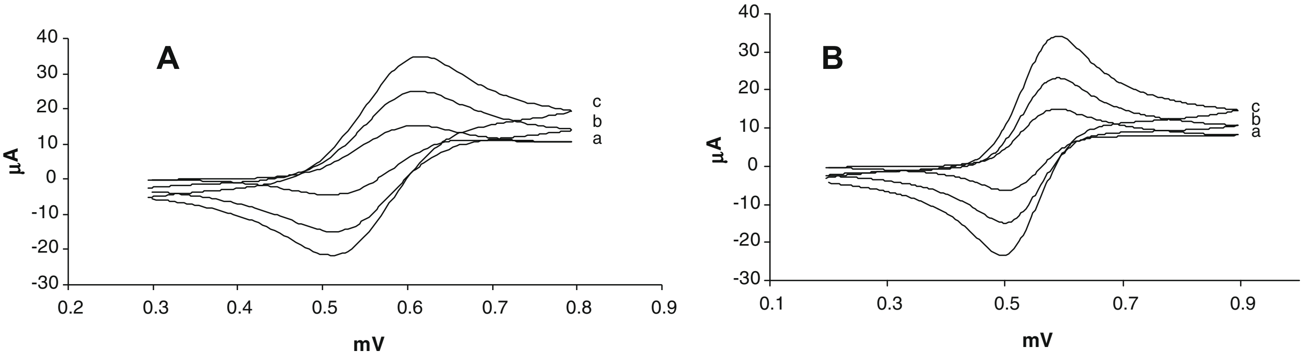


Fig. 2. Cyclic voltammograms of 0.01 mmol VOL^1 (A) and VOL^2 (B) in acetonitrile containing 0.1 M TBAH as supporting electrolyte. Scan rates (mV/s) are (a) 20, (b) 50 and (c) 100, respectively.

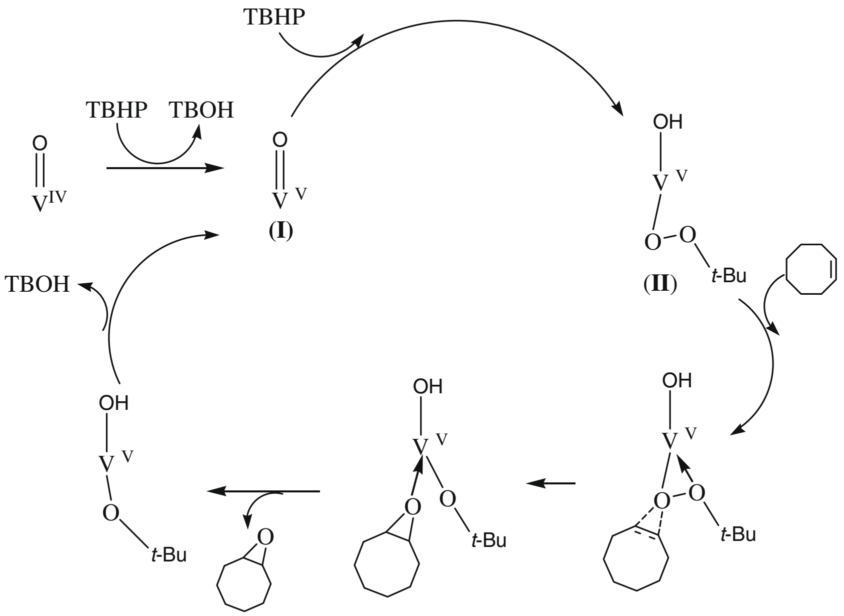


Fig. 3. Proposed catalytic cycle for the oxidation of cyclooctene (or styrene) by VOL^1 or VOL^2.

Table 4
Oxidation of styrene and cyclooctene with TBHP in the presence of VOL^1 and VOL^2.[a]

Catalyst	Olefin	Conversion (%)	TON[b] (Total)	Product selectivity (%)	
				Epoxide	Benzaldehyde
None	Styrene	10	3.1	45	55
VOL^1	Styrene	48	150	87.5	12.5
VOL^2	Styrene	88	275	99	1
None	Cyclooctene	4	1.2	100 (cyclooctene oxide)	
VOL^1	Cyclooctene	94	293	100 (cyclooctene oxide)	
VOL^2	Cyclooctene	99.8	312	100 (cyclooctene oxide)	

[a] Solvent: 10 ml; catalyst: 0.032 mmol; styrene and/or cyclooctene: (10 mmol); TBHP: 30 mmol.

[b] TON: (total turnover number) the ratio of the number of moles of product to the number of moles of catalyst.

solvents (chloroform, dichloromethane and acetonitrile) were used. It was observed that the catalytic activity of the vanadyl complexes decreases in order acetonitrile > chloroform > dichloromethane. Interestingly, in the case of styrene, acetonitrile is better than chloroform which may be partly due to the higher b.p of acetonitrile (82 °C) and also dielectric constant ($\varepsilon/\varepsilon_0 = 35.94$) and dipole moment ($\mu = 3.90$ D) which are the highest of all the solvents used. Also, it is proposed that the involvement of an intermediate with some carbocation character in the catalytic cycle of styrene cause the observed trend. This assumption is according to the stability of carbocation of styrene due to the conjugation with the phenyl ring in organic reaction. The presence of solvent such as acetonitrile which can stabilize the cationic centres increases the rate of reactions bearing intermediates with more cationic character.

In order to get the best reaction temperature, the reaction mixture was stirred at various temperatures (40, 50, 60, 70 and 80 °C) and the obtained results show that the maximum conversion was obtained at 80 °C.

The reusability of the catalyst was examined. The catalyst can be reused four times without a detectable catalyst leaching or a significant loss of its activity.

3.2.3. Proposed mechanism

In the proposed reaction mechanism oxovanadium(IV) oxidized to oxovanadium(V) (Fig. 3, **I**) in the presence of TBHP [27] and another TBHP coordinate to the V(V)═O species to form *tert*-butylOOV^V–OH intermediate (Fig. 3, **II**) that is probably the active oxidizing species, which can react with olefin to produce epoxide.

Titration of oxovanadium(IV) complexes in chloroform solution with TBHP has been done and the reaction was monitored by UV–Vis spectroscopy. Upon the addition of TBHP, the weak broad band due to d–d transition (574 nm for VOL^1 and 554 nm for VOL^2), slowly become weaker and finally disappears. This observation suggests the formation of peroxovanadium(V) species [28] which is in accord with the previously reported studies by Maurya et al. [19,29].

The higher conversion and selectivity obtained in the presence of the title electron rich vanadyl complexes in comparison with other reports [17,26] suggests that the catalytic activity significantly increases with a decrease in the V^{5+}/V^{4+} potential. Therefore, it seems that the oxidation of V^{IV}═O to V^V═O occur in rate limiting step.

In oxidation of styrene, the benzaldehyde formation is possibly due to over-oxidation of styrene oxide by nucleophilic attack of hydroperoxide [30].

4. Conclusions

VOL^1 and VOL^2 complexes were prepared and characterized. Oxidation of cyclooctene and styrene with *tert*-butylhydroperoxide (TBHP) in the presence of the title complexes shows that the introduction of electron-donating substituents on the aromatic rings as well as the imine bonds of salene type Schiff bases effectively improve their catalytic activity. The catalytic performance of vanadyl complexes depends on the oxidant-to-olefin molar ratio, the type of solvent and the reaction temperature. The conversion of cyclooctene to cylcooctene oxide was 98% for VOL^1 and 98.8% for VOL^2 and a selectivity of 100% was attained. In the case of styrene, 48–88% conversion to epoxide and benzaldehyde has been observed.

Acknowledgements

We gratefully acknowledge practical support of this study by K.N. Toosi University of Technology and Islamic Azad University of Saveh Branch.

References

[1] K.C. Gupta, A.K. Sutar, Coord. Chem. Rev. 252 (2008) 1420–1450.
[2] K.C. Gupta, A.K. Sutar, C.C. Lin, Coord. Chem. Rev. 253 (2009) 1926–1946.
[3] K.C. Gupta, A.K. Sutar, J. Mol. Catal. A: Chem. 272 (2007) 64–74.
[4] A.G.J. Ligtenbarg, R. Hage, B.L. Feringa, Coord. Chem. Rev. 237 (2003) 89–101.
[5] D. Rehder, Coord. Chem. Rev. 182 (1999) 297–322.
[6] H. Sakurai, Y. Kojima, Y. Yoshikawa, K. Kawabe, H. Yasui, Coord. Chem. Rev. 226 (2002) 187–198.
[7] D. Rehder, G. Santoni, G.M. Licini, C. Schulzke, B. Meier, Coord. Chem. Rev. 237 (2003) 53–63.
[8] K.H. Thompson, C. Orvig, Coord. Chem. Rev. 219–221 (2001) 1033–1053.
[9] E. Tsuchida, K. Oyaizu, Coord. Chem. Rev. 237 (2003) 213–228.
[10] M. Kojima, H. Taguchi, M. Tsuchimoto, K. Nakajima, Coord. Chem. Rev. 237 (2003) 183–196.
[11] M.R. Maurya, Coord. Chem. Rev. 237 (2003) 163–181.
[12] M.R. Maurya, A.K. Chandrakar, S. Chand, J. Mol. Catal. A: Chem. 274 (2007) 192–201.
[13] R. Ando, S. Mori, M. Hayashi, T. Yagyu, M. Maeda, Inorg. Chim. Acta 357 (2004) 1177–1184.
[14] R. Ando, H. Ono, T. Yagyu, M. Maeda, Inorg. Chim. Acta 357 (2004) 817–823.
[15] S. Rayati, N. Sadeghzadeh, H.R. Khavasi, Inorg. Chem. Commun. 10 (2007) 1545–1548.
[16] S. Rayati, N. Torabi, M. Koliaei, F. Ashouri, S. Mohebbi, A. Wojtczak, A. Kozakiewicz, Appl. Catal. A: Gen. 346 (2008) 65–71.
[17] S. Rayati, N. Torabi, A. Ghaemi, S. Mohebbi, A. Wojtczak, A. Kozakiewicz, Inorg. Chim. Acta 361 (2008) 1239–1248.
[18] S. Mohebbi, D.M. Boghaei, A.H. Sarvestani, A. Salimi, Appl. Catal. A: Gen. 278 (2005) 263–267.
[19] M.R. Maurya, A.K. Chandrakar, S. Chand, J. Mol. Catal. A: Chem. 270 (2007) 225–235.
[20] M.R. Maurya, S. Sikarwar, P. Manikandan, Appl. Catal. A: Gen. 315 (2006) 74–82.
[21] M.R. Maurya, A. Kumar, P. Manikandan, S. Chand, Appl. Catal. A: Gen. 277 (2004) 45–53.
[22] D.D. Perrin, W.L.F. Armarego, D.R. Perrin, Purification of Laboratory Chemicals, second ed., Pergamon Press, Elmsford, NY, 1980.
[23] S.A. Fairhurst, D.L. Hughes, U. Kleinkes, G.J. Leigh, J.R. Sanders, J. Weisner, J. Chem. Soc. Dalton Trans. (1995) 321–326.
[24] N.F. Choudhary, N.G. Connelly, P.B. Hitchcock, G.J. Leigh, J. Chem. Soc. Dalton Trans. (1999) 4437–4446.
[25] F.A. Carey, R. Sundberg, Advanced Organic Chemistry, Part A: Structure and Mechanisms, fifth ed., Springer, 2007 (Chapters 1, 6 and 12).
[26] M. Salavati-Niasari, M.R. Elzami, M.R. Mansournia, S. Heidarzadeh, J. Mol. Catal. A: Chem. 221 (2004) 169–175.
[27] C.J. Chang, J.A. Labinger, H.B. Gray, Inorg. Chem. 36 (1997) 5927–5930.
[28] T. Joseph, D. Srinivas, C.S. Gopinath, S.B. Halligudi, Catal. Lett. 83 (2002) 209–214.
[29] M.R. Maurya, A. Kumar, J. Mol. Catal. A: Chem. 250 (2006) 190–198.
[30] V. Hulea, E. Dumitriu, Appl. Catal. A: Gen. 277 (2004) 99–106.