Catalysis Today 157 (2010) 114–118

Contents lists available at ScienceDirect

Catalysis Today

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

Selective oxidation of pyridine to pyridine-*N*-oxide with hydrogen peroxide over Ti-MWW catalyst

Wei Xie, Yuting Zheng, Song Zhao, Junxia Yang, Yueming Liu*, Peng Wu*

Shanghai Key Laboratory of Green Chemistry and Chemical Processes, Department of Chemistry, East China Normal University, North Zhongshan Rd. 3663, Shanghai 200062, China

ARTICLE INFO

Article history:
Available online 15 March 2010

Keywords:
Oxidation
Pyridine
Pyridine-*N*-oxide
Ti-MWW
Hydrogen peroxide

ABSTRACT

The oxidation of pyridine to pyridine-*N*-oxide (PNO) with hydrogen peroxide has been investigated on various titanosilicate catalysts. Superior to other titanosilicates like TS-1, Ti-Beta and Ti-MOR, Ti-MWW showed a higher catalytic activity and product selectivity. The reaction parameters such as solvent, temperature, hydrogen peroxide/substrate ratio, the amount of catalyst and Ti content were optimized to maximize the PNO yield. Ti-MWW was capable of giving pyridine conversion and PNO selectivity both over 99% under optimum reaction conditions. Ti-MWW was a highly active, selective, and reusable catalyst for the synthesis of PNO. The mechanism for the oxidation of pyridine over titanosilicates has also been considered. Ti-MWW was further structurally modified to posses open reaction spaces by interlayer expansion, which reduced the diffusion limitations in the oxidation of pyridine derivatives with bulkier molecular dimensions.

© 2010 Elsevier B.V. All rights reserved.

1. Introduction

Pyridine-*N*-oxide (PNO) is an important fine chemical with a constantly increasing world market owing to its usefulness as synthetic intermediates and biological importance. Heterocyclic PNO is also used as protecting groups, auxiliary agents, oxidants, ligands in metal complexes and catalysts [1]. In addition, PNO is used as a functional chemical building block for synthesizing agrochemicals and pharmaceuticals.

The conventional processes for manufacturing PNO are of multisteps involving sulfuric acid-catalyzed oxidation of pyridine with H_2O_2 as an oxidant, acetic acid as the solvent, and the separation of PNO by neutralizing sulfuric acid with sodium hydroxide [2]. These processes are encountering serious disadvantages, such as using excessive solvent like acetic acid and corrosive sulfuric acid, producing onslaughts of valueless sodium sulfate by-product together with toxic waste solvent and bringing about environmental problems. Therefore, it is urgent to develop costless and environmentally benign methods of zero-emission for PNO synthesis.

The discovery of TS-1 titanosilicate has opened up new possibilities of developing zeolite catalyst-based green processes, as the material is capable of catalyzing a variety of organic compounds with H_2O_2 and with water as almost the sole by-product [3]. Recently, TS-1 is reported to be useful for the oxidation of primary, secondary amines and pyridine derivatives [4]. However, TS-1 is still facing some drawbacks. Its manufacture cost is generally high due to the use of organic silicon source and tetrapropylammonium hydroxide as structure-directing agent. Moreover, the MFI topology of TS-1 is composed of medium-sized pores of 10-membered ring (MR), which limits the application of TS-1 to the oxidations involving the substrates and oxidants both with relatively small molecular sizes.

Many other titanosilicates with various crystalline topologies have been developed after TS-1. Up to date, only Ti-MWW with the MWW structure seems to be intrinsically more active than TS-1 [5]. The advantage of Ti-MWW is firmly related to its unique pore system consisting of two independent 10-MR channels, 12-MR supercages and external side cups as well. Since the MWW zeolite has structural diversity, it is possible to transform Ti-MWW into the titanosilicates with open porosity, such as Del-Ti-MWW [6] and IEZ-Ti-MWW [7] through phase delamination and interlayer expanding, respectively. Ti-MWW has been shown to be an efficient for the epoxidation of functional alkenes and ammoximation of cyclohexanone. These successes encourage us to investigate its application in the synthesis of valuable oxygenated chemicals containing other functional groups.

In this study, we have carried out the oxidation of pyridine and its derivatives on Ti-MWW, TS-1, Ti-Beta and Ti-MOR with H_2O_2 or *tert*-butyl hydroperoxide (TBHP) with the purpose to synthesize *N*-oxides actively and selectively. The stability and reusability of Ti-MWW have been investigated in detail. In particular, IEZ-Ti-MWW with an open reaction space has been prepared to improve

---

* Corresponding authors at. Tel.: +86 21 6223 2292; fax: +86 21 6223 2292.
E-mail addresses: ymliu@chem.ecnu.edu.cn (Y. Liu), pwu@chem.ecnu.edu.cn (P. Wu).

0920-5861/$ – see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.cattod.2010.02.045

Table 1
The results of pyridine oxidation over different titanosilicate catalysts[a].

No.	Catalyst[b]	Oxidant	Pyridine conv. (%)	PNO sel. (%)	$X_{H_2O_2}$ (%)	$U_{H_2O_2}$ (%)	TOF[c] (h^{-1})
1	Ti-MWW (43)	H_2O_2	97.7	99.4	96.0	77.8	252
2	TS-1 (40)	H_2O_2	99.2	95.5	97.1	75.1	238
3	Ti-Beta (70)	H_2O_2	51.5	95.7	70.2	54.0	216
4	Ti-MOR (92)	H_2O_2	24.5	90.2	60.3	28.2	135
5	Silicalite-1	H_2O_2	0	0	–	–	0
6	none	H_2O_2	0	0	–	–	0
7	Ti-MWW (43)	TBHP	13.0	99.0	–	–	33
8	TS-1 (40)	TBHP	1.4	98.0	–	–	3
9	Ti-Beta (70)	TBHP	2.4	97.6	–	–	10

[a] Reaction conditions: cat., 0.15 g; pyridine, 30 mmol; H_2O_2 (30%), 39 mmol; temp., 348 K; time, 2 h; without solvent.
[b] The number in parentheses indicates the Si/Ti molar ratio.
[c] TOF: turnover frequency.

the catalytic activity in the oxidation of bulky amine substrates. This study may provide ideas for developing cleaner processes for the production of PNO and other oxides of pyridine derivatives.

2. Experimental

2.1. Catalyst preparation and characterization

Ti-containing MWW lamellar precursor was hydrothermally synthesized following the procedures reported previously [5]. The precursor was then refluxed with 2 M HNO_3 to result in the samples with Si/Ti ratios of 43–110 and Si/B ratios of 36–56. The acid-treated sample was washed with deionized water and subsequently dried at 373 K (Ti-MWW-dry) or further calcined at 823 K (Ti-MWW-cal.). Interlayer-expanded zeolite, IEZ-Ti-MWW, was postsynthesized by alkoxysilylating Ti-MWW-dry with diethoxydimethylsilane [7]. TS-1 with a Si/Ti molar ratio of 40 was hydrothermally synthesized with Enichem method [8]. Al-free Ti-Beta was hydrothermally synthesized by a seeding method [9], while Ti-MOR was postsynthesized by vapor-phase treatment of delaminated mordenite with $TiCl_4$ [10].

The samples were characterized by inductively coupled plasma (ICP) on a Thermo IRIS Intrepid II XSP atomic emission spectrometer, by X-ray diffraction (XRD) on a Bruker D8 Advance diffractometer (Cu Kα), by N_2 adsorption (Autosorb Quancachrome 02108-KR-1) and by UV–vis (Shimadzu UV-2400PC) and IR (Nicolet NEXUS 670) spectroscopies.

2.2. Catalytic reactions

The liquid-phase oxidation of pyridine was carried out under vigorous stirring in a 50 mL glass flask connected to a cooling condenser. In a typical run, a desirable amount of solvent, 30 mmol of pyridine, 39 mmol of H_2O_2 (30 wt% aqueous solution) or TBHP (70 wt% aqueous solution) and 0.15 g catalyst were mixed in the flask and heated at different temperature under agitation for 2 h. The reaction mixture was converted into a homogeneous phase by mixing with enough ethanol. After removal of the catalyst powder, the products formed were then determined using authentic chemicals on a GC–MS (Agilent GC–MS (6890-5973N)). And their amounts were quantified by GC (Shimadzu, FID detector and 30-m OV-1 column). The amount of unconverted H_2O_2 was quantified by iodometry.

3. Results and discussion

3.1. Summary of catalyst characterization

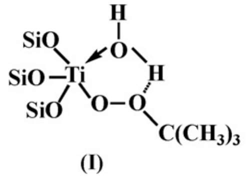
The XRD patterns confirmed that all titanosilicates had objective zeolite structures with a relatively high crystallinity (Fig. S1). No impurity phase was detected. The samples showed the characteristic adsorption bands around 220 nm in UV–vis spectra (Fig. S2) and at 960 cm^{-1} in IR spectra, respectively (not shown), which indicated that they contained the tetrahedral Ti species dispersed isolatedly in the zeolite framework [3]. The Langmuir specific surface areas determined from N_2 adsorption were in the range of 590–635 $m^2 g^{-1}$ for the Ti-MWW samples with the Si/Ti molar ratios of 43–110, 530 $m^2 g^{-1}$ for TS-1, 553 $m^2 g^{-1}$ for Ti-MOR and 653 $m^2 g^{-1}$ for Ti-Beta. Above characterizations verified that these titanosilicates were good in quality as liquid-phase oxidation catalysts.

3.2. A comparison of oxidation among various titanosilicates

Table 1 compares the results of pyridine oxidation over different titanosilicates using hydrogen peroxide or TBHP as an oxidant. Ti-MWW showed a pyridine conversion of 97.7% and PNO selectivity of 99.4% with H_2O_2 as an oxidant. The absolute amount of by-products other than PNO was extremely small (No. 1). Under the reaction conditions same as those adopted for Ti-MWW, TS-1 showed a comparably high pyridine conversion but a lower PNO selectivity than Ti-MWW (No. 2). The by-products were mainly 2-hydroxypyridine, 4-hydroxypyridine, oxaziranes, and azoxy compounds, which were consistent with those reported in previous studies [11]. The specific activity with respect to Ti species, i.e. turnover frequency (TOF), indicated that TS-1 was less active. The superiority of Ti-MWW to TS-1 has already been observed in the epoxidation of various kinds of alkenes [12], which is attributed to the unique pore system of MWW zeolite favoring the adsorption and access of substrate molecules to the Ti active sites. The efficiency of H_2O_2 utilization was 75–78% for Ti-MWW and TS-1. Nonproductive decomposition of H_2O_2 was mainly due to the basic media with the presence of pyridine.

Ti-MOR and Ti-Beta turned out to be much less active and selective than Ti-MWW and TS-1 (Nos. 3 and 4). Ti-Beta exhibited highly hydrophilic features due to stacking defaults, which made it to be an unsuitable catalyst for pyridine oxidation. Since Ti-MOR had larger crystal size and only one-dimensional channels, both of which hinder the diffusion and access of substrate molecules to the Ti sites, it was also an unsuitable catalyst for pyridine oxidation. In spite of the difference in optimum reaction conditions, the titanosilicates exhibited in pyridine oxidation a catalytic activity order of Ti-MWW > TS-1 ≫ Ti-Beta > Ti-MOR. In particular, no product was obtained either in the absence of any catalyst or in the presence of Ti-free Silicalite-1 (Nos. 5 and 6). The results clearly indicate that the isolated tetrahedral Ti^{4+} ions in titanosilicates are the active sites for pyridine oxidation.

In comparison with H_2O_2, the use of TBHP with a bulky molecular size made the reaction greatly retard (Nos. 7–9). In general, the titanosilicate-catalyzed reactions are considered to involve the 5-MR intermediates which are formed through the coordination



Scheme 1.

of a solvent molecule such as alcohol or H_2O to a Ti peroxo species [13]. Species I and II shown in Scheme 1 are thus presumed to be the intermediates for the oxidations with TBHP and H_2O_2, respectively, since both oxidants contained water. The species I is much larger than the species II owing to a larger molecular dimension of *tert*-butyl groups. An actual reaction occurs only when the molecules can reach the above intermediates. The medium pores of TS-1 imposed a serious steric restriction for the intermediate of TBHP, which led to a very low activity. Similarly, the oxidation of pyridine with TBHP was restricted seriously on Ti-Beta. Although the catalytic performance of Ti-MWW was also affected by the molecular sizes of the oxidant, it showed a higher pyridine conversion than TS-1 and Ti-Beta in the case of TBHP. This is because in addition to two-dimensional 10-MR sinusoidal channels running throughout the structure parallel to the *ab*-plane, the MWW structure contains also an independent channel system which is comprised of large supercages (0.7 nm × 0.7 nm × 1.8 nm), as well as the pocket or cup moieties (0.7 nm × 0.7 nm) of the supercages on the crystal exterior. These open reaction spaces make Ti-MWW more active than TS-1 and Ti-Beta for the pyridine oxidation with TBHP.

3.3. Effects of reaction parameters on the oxidation of pyridine over Ti-MWW

3.3.1. Effect of H_2O_2/pyridine ratio

Fig. 1 shows the effect of H_2O_2/pyridine ratio on the pyridine oxidation over Ti-MWW. The pyridine oxidation stoichiometrically needs equivalent moles of H_2O_2 and pyridine, but the pyridine conversion was only 75% at H_2O_2/pyridine ratio of 1.0. With increasing H_2O_2/pyridine ratio, the pyridine conversion increased simultaneously. Due to unproductive decomposition of H_2O_2, the reaction needed more H_2O_2 than pyridine to proceed to a high level. A high PNO selectivity (>97%) was retained at different H_2O_2/pyridine ratios. The pyridine conversion reached nearly 100% when the H_2O_2/pyridine ratio was above 1.3, and showed no change at higher ratios.

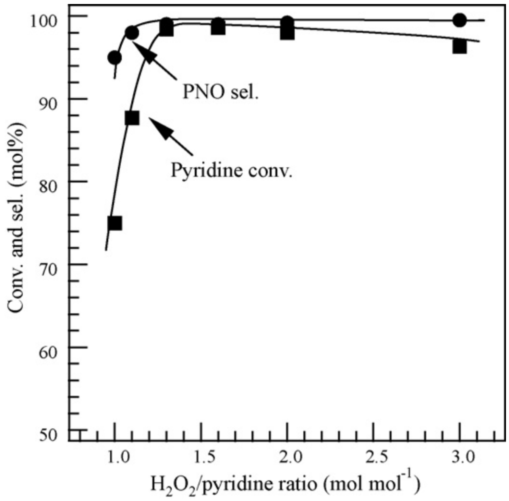


Fig. 1. Effect of H_2O_2/pyridine ratio on the pyridine oxidation over Ti-MWW (43). Oxidation conditions: see Table 1.

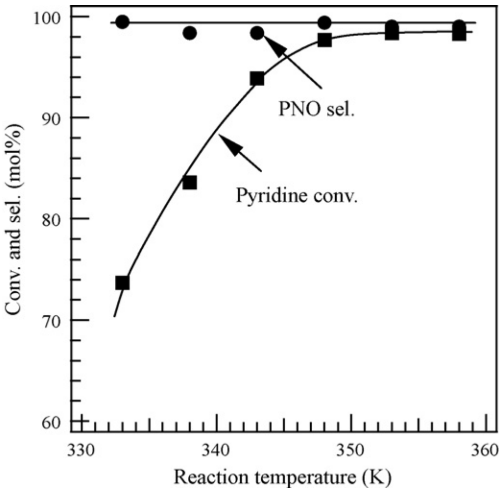


Fig. 2. Effect of reaction temperature on the pyridine oxidation over Ti-MWW (43). Oxidation conditions: see Table 1.

3.3.2. Effect of reaction temperature

The reaction temperature had a great effect on the oxidation of pyridine over Ti-MWW. As shown in Fig. 2, the conversion of pyridine increased with increasing temperature and reached the maximum at 353 K, whereas the selectivity to PNO remained over 98.0% at the temperatures investigated. Further increasing the temperature to 358 K, no further changes were observed for either the pyridine conversion or the PNO selectivity.

3.3.3. Effect of solvent

Titanosilicate-catalyzed reactions are well known to depend greatly on the nature of solvent. The oxidation of pyridine over Ti-MWW has been carried out in the solvents such as methanol, ethanol, isopropanol, *t*-BuOH, water, acetonitrile, acetone, ether, or free of any additional solvent (Fig. 3). The PNO selectivity was over 99.4% in all cases except for acetonitrile and acetone which showed a selectivity of 96–98.0%. With methanol and water as effective solvents for the oxidation of pyridine, the PNO yield decreased in the order of methanol ≈ H_2O > non-solvent > ethanol > acetonitrile > isopropanol > *t*-BuOH ≫ acetone > ether. The effect of solvent is one of the most complicated issues in the catalytic sys-

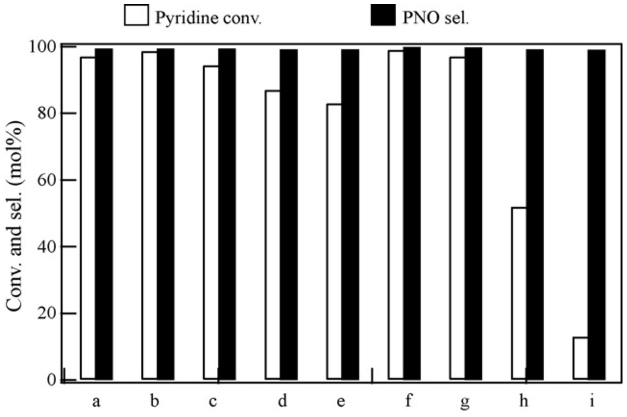


Fig. 3. The results of pyridine oxidation on Ti-MWW free of any additional solvent (a), and in the presence of methanol (b), ethanol (c), isopropanol (d), *t*-BuOH (e), water (f), acetonitrile (g), acetone (h), and ether (i). Oxidation conditions: the amount of solvent, 0.19 mol; others see Table 1.

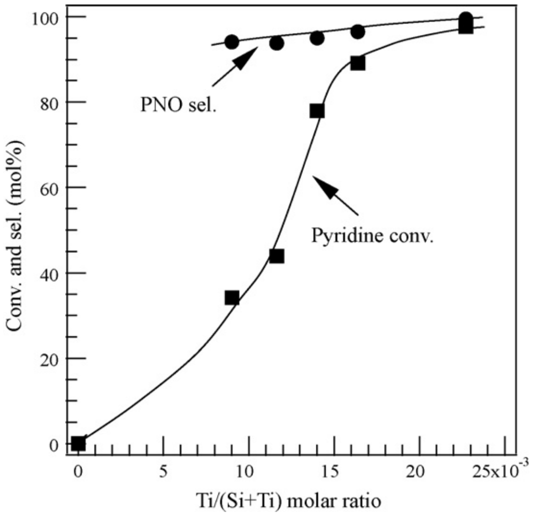


Fig. 4. Dependence of the pyridine oxidation on the Ti content of Ti-MWW. Reaction conditions: see Table 1.

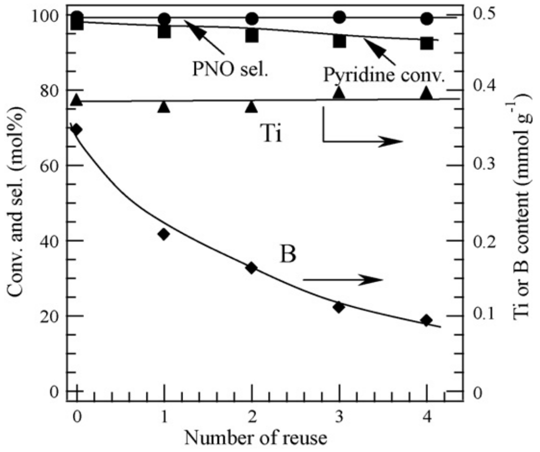


Fig. 5. The reuse of Ti-MWW in pyridine oxidation. The used catalyst was regenerated by water washing and further calcination in air at 823 K for 6 h. Reaction conditions: see Table 1.

tem of titanosilicate/H_2O_2. Although MeCN has been reported to be a suitable solvent for the epoxidation of alkenes on Ti-MWW, it only led to a moderate activity (96.4% pyridine conversion). In contrast, when the system was free of solvent except for the water originally coming from the aqueous H_2O_2 solution, the conversion increased to 97.7% at 348 K. In comparison to other solvent, isopropanol or *t*-BuOH with a large molecular dimension made the oxidation retard greatly. The pyridine conversion decreased significantly with non-polar solvents. The conversion turned to be 50.6% and 15.1% in acetone and ether, respectively. Above results suggest the reaction rate is closely related to the polarity and molecular size of the solvents, and also the solubility of the substrates, products, etc.

3.3.4. *Effect of Ti content*

A series of Ti-MWW catalysts with Si/Ti ratios of 43–110 were prepared and used in the oxidation of pyridine (Fig. 4). Reasonably, the yield of PNO increased with an increasing amount of the Ti active sites in Ti-MWW. It should be noted that the oxidation did not proceed in the presence of Ti-free MWW zeolite.

3.4. *The stability and regeneration of Ti-MWW*

In addition to the catalytic activity and PNO selectivity, the reusability of Ti-MWW is also important issue as it determines the real applicability. The stability and reusability of Ti-MWW in the pyridine oxidation have thus been checked. The experiments were initiated with an enlarged reaction scale using 0.3 g of Ti-MWW, and the used catalyst was subjected to ICP analyses and repeated reactions under the same conditions. After calcination in air at 823 K for 6 h, the color of the used catalyst changed from light brown to snow white, indicating a total removal of heavy organic species. This kind of regeneration actually prevented the catalyst from deactivating. As shown in Fig. 5, no obvious Ti leaching occurred after the repeated reaction while the content of boron was gradually decreased as the boron species moved out of the framework easily due to a small ionic radius.

To further verify the heterogeneous nature of catalysis and assess the absence of soluble active species, we have carried out the hot filtration test following the suggestion in the literature [14]. The experiments involved performing two parallel reactions in the presence of the solid catalyst (Fig. S4). When the conversion of 62% was reached at 20 min, the solid catalyst was removed by filtration for one reaction. The reaction was terminated since the pyridine conversion was almost unchanged with prolonging the reaction time. On contrast, the pyridine conversion for the reaction without hot filtration increase with reaction time and reached about 98% at 120 min. This result indicates that the liquid-phase oxidation of pyridine actually takes place in a heterogeneous manner, and also rules out the leaching of Ti species during the reaction.

The used catalyst showed similar XRD pattern and UV–vis. spectrum to the fresh one (Figs. S5 and S6), indicating no obvious changes occurred in the structure and Ti coordination state. The catalyst remained to be highly active and selective to the pyridine oxidation after reused for four times. A slight decrease in conversion is probably due to a partial dissolution of framework silicon in basic reaction media. Very similar results have already been obtained with Ti-MWW in the epoxidation of various alkenes [12].

3.5. *Oxidation of pyridine derivatives*

The oxidation of pyridine derivatives has been carried out on different titanosilicate catalysts (Table 2). The larger the substrate

Table 2
Oxidation of pyridine derivatives over different titanosilicates[a].

No.	Catalyst	3-Picoline oxidation			4-Picoline oxidation		
		Conv. (%)	3-PNO sel.[b] (%)	TOF[c] (h^{-1})	Conv. (%)	4-PNO sel.[d] (%)	TOF[c] (h^{-1})
1	TS-1 (40)	29.9	97.0	72	30.4	96.5	73
2	Ti-MWW-cal. (43)	34.0	98.9	87	38.8	97.6	100
3	Ti-MWW-dry (43)	57.0	99.5	147	45.3	97.2	117
4	IEZ-Ti-MWW (40)	71.9	99.0	177	71.3	98.9	176

[a] Reaction conditions: cat., 0.15 g; 3-picoline or 4-picoline, 30 mmol; H_2O_2 (30%), 39 mmol; temp., 348 K; time, 2 h; without solvent.
[b] 3-PNO: 3-picoline-*N*-oxide.
[c] TOF: turnover frequency.
[d] 4-PNO: 4-picoline-*N*-oxide.

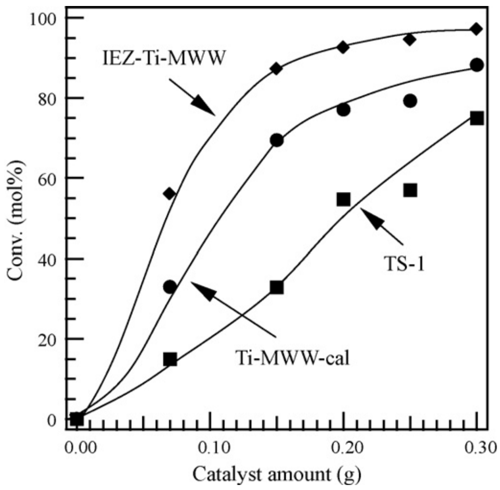


Fig. 6. Effect of catalyst amount on the 4-picoline oxidation. Oxidation conditions: solvent was methanol; others see Table 2.

is the more open reaction space is needed. The conversion of 3-picoline and 4-picoline was only 29.9% and 30.4% over medium pore TS-1 (No. 1). Ti-MWW in the calcined form was more active than TS-1 by showing the conversion of 34.0% and 38.8% for 3-picoline and 4-picoline, respectively (No. 2). This is mainly because it has 12-MR supercages as well as external half cups. Nevertheless, the catalytic activity of Ti-MWW for these methyl-substituted derivatives was obviously lower than for pyridine as shown in Table 1.

We reported that the sample without calcination, that is Ti-MWW-dry, may still contain layered structure and then have open pore system [15]. It is thus superior to the calcined Ti-MWW in the oxidation of cyclic alkenes. The same phenomenon was also observed in the oxidation of pyridine derivatives. The conversion of 3-picoline and 4-picoline over Ti-MWW-dry increased to 57.0% and 45.3%, respectively (Table 2, No. 3). However, a subsequent calcination would cause the dehydroxylation between the layer sheets to narrow the interlayer pore entrance. Ti-MWW-dry suffers a disadvantage that its structure and catalytic activity is not totally maintained when subjected to regeneration by calcination. It is desirable to develop the titanosilicate which is thermally and hydrothermally stable and contains the basic MWW structure and larger pores as well.

Very recently, a methodology has been established to make the interlayer structure expanded for the MWW, FER, CDO, and MCM-47 topologies by the silylation of corresponding precursors [7]. The interlayer expanded zeolites thus obtained are composed of the fundamental building units identical to directly calcined precursors but exhibit a larger porosity. Interlayer expanded MWW-type titanosilicate, i.e. IEZ-Ti-MWW, was then prepared and applied to the oxidation of picolines. IEZ-Ti-MWW showed a much higher conversion than other catalysts (Table 2, No. 4). The conversion of 3-picoline and 4-picoline reached high 71.9% and 71.3%, respectively. With the interlayer spacing expanded by silylation by 0.25 nm [7], IEZ-Ti-MWW possessed larger pore windows which were accessible to the substrates with large molecular dimensions and were also benefit for product desorption. As a result, the 3-PNO or 4-PNO yield decreased in the order of IEZ-Ti-MWW > Ti-MWW-dry > Ti-MWW-cal. > TS-1.

Fig. 6 shows the effect of catalyst amount on the 4-picoline oxidation over different titanosilicates. The reaction was carried out by using the same amount of substrate, while varying the amount of catalysts in the range of 0–0.3 g. The 4-PNO selectivity was always over 97%. The 4-picoline conversion increased with increasing amount of catalyst. It is noteworthy that the conversion of 4-picoline reached 97.1% when the amount of the IEZ-Ti-MWW catalyst was 0.3 g (corresponding to ca. 11 wt% of 4-picoline). However, the conversion of 4-picoline was only 75% and 88.3% over TS-1 and Ti-MWW-cal., respectively, when the same amount of catalyst was used. The interlayer pore expansion of the MWW structure is thus an effective way to enhance the activity for the oxidation of pyridine derivatives.

4. Conclusion

Ti-MWW is capable of catalyzing the liquid-phase oxidation of pyridine to pyridine-*N*-oxide at a conversion and selectivity >99% in the presence of water or methanol, and proves to be a reusable catalyst for pyridine-*N*-oxide synthesis. The catalytic behavior of Ti-MWW depends greatly on the amount of Ti active sites. Ti-MWW is more effective than TS-1, Ti-Beta and Ti-MOR. The catalytic performance of Ti-MWW for bulky substrates pyridine derivatives is greatly improved by interlayer silylation of Ti-MWW precursor. Thus, MWW structure-based titanosilicates are promising catalysts for the clean synthesis of PNO and other oxides of pyridine derivatives.

Acknowledgments

We gratefully acknowledge NSFC of China (20925310, 20873043), Science and Technology Commission of Shanghai Municipality (08JC1408700, 09XD1401500), 973 Program (2006CB202508), 863 Program (2007AA03Z34, 2008AA030801), and Shanghai Leading Academic Discipline Project (B409).

Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.cattod.2010.02.045.

References

[1] (a) A. Albini, Synthesis (1993) 263;
(b) A.V. Malkov, M. Bell, F. Castelluzzo, P. Kocovsky, Org. Lett. 7 (2005) 3219;
(c) R. Sarma, A. Karmakar, J.B. Baruah, Inorg. Chim. Acta 361 (2008) 2081.
[2] V. Bockelhe, W.J. Linn, J. Am. Chem. Soc. 76 (1954) 1286.
[3] G. Bellussi, M.S. Rigguto, Stud. Surf. Sci. Catal. 137 (2001) 911.
[4] (a) J.S. Reddy, P.A. Jacobs, J. Chem. Soc., Perkin Trans. 22 (1993) 2665;
(b) J.S. Reddy, P.A. Jacobs, Catal. Lett. 37 (1996) 213;
(c) M.R. Prasad, G. Kamalakar, G. Madhavi, S.J. Kulkarni, K.V. Raghavan, Chem. Commun. 17 (2000) 1577;
(d) D.J. Robinson, P. McMorn, D. Bethell, P.C. Bulman-Page, C. Slyd, F. King, F.E. Hancock, G.J. Hutchings, Catal. Lett. 72 (2001) 233.
[5] P. Wu, T. Tatsumi, T. Komatsu, T. Yashima, J. Phys. Chem. B 105 (2001) 2897.
[6] P. Wu, D. Nuntasri, J.F. Ruan, Y.M. Liu, M.Y. He, W.B. Fan, O. Terasaki, T. Tatsumi, J. Phys. Chem. B 108 (2004) 19126.
[7] (a) P. Wu, J.F. Ruan, L.L. Wang, L.L. Wu, Y. Wang, Y.M. Liu, W.B. Fan, M.Y. He, O. Terasaki, T. Tatsumi, J. Am. Chem. Soc. 130 (2008) 8178;
(b) L.L. Wang, Y. Wang, Y.M. Liu, H.H. Wu, X.H. Li, M.Y. He, P. Wu, J. Mater. Chem. 19 (2009) 8594.
[8] T. Taramasso, G. Perego, B. Notari, U.S. Patent 4,410,501, 1983.
[9] T. Blasco, M.A. Camblor, A. Corma, P. Esteve, J.M. Guil, A. Martínez, J.A. Perdigón-Melón, S. Valencia, J. Phys. Chem. B 102 (1998) 75–88.
[10] P. Wu, T. Komatsu, T. Yashima, J. Catal. 168 (1997) 400.
[11] (a) F.E. Clslak, Ind. Eng. Chem. 47 (1955) 800;
(b) R. Kluge, M. Schulz, S. Liebsch, Tetrahedron 52 (1996) 5773;
(c) F. Cardona, M. Bonanni, G. Soldaini, A. Goti, Chem. Sus. Chem. 1 (2008) 327.
[12] (a) P. Wu, T. Tatsumi, J. Catal. 214 (2003) 317;
(b) P. Wu, Y.M. Liu, M.Y. He, T. Tatsumi, J. Catal. 228 (2004) 183.
[13] (a) G. Bellussi, A. Carati, M.G. Clerici, G. Maddinelli, R. Millini, J. Catal. 133 (1992) 220;
(b) M.G. Clerici, P. Ingallina, J. Catal. 140 (1993) 71.
[14] R.A. Sheldon, M. Wallau, I. Arends, U. Schuchardt, Acc. Chem. Res. 31 (1998) 485.
[15] P. Wu, D. Nuntasri, Y.M. Liu, H.H. Wu, Y.W. Jiang, W.B. Fan, M.Y. He, T. Tatsumi, Catal. Today 117 (2006) 199.