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Liquid Phase Oxidation of Cyclohexene Over Selenite Doped MCM-41

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Abstract Liquid phase oxidation of cyclohexene was carried out under mild reaction conditions over selenite doped MCM-41 using aqueous hydrogen peroxide as oxidant and acetic acid as solvent. The major products of the reaction were 2-acetoxycyclohexanol and a small amount of 1,2-cyclohexanediol. The catalyst exhibited very high substrate conversion ($\sim 100\%$) and good product (2-acetoxycyclohexanol) selectivity (81%) and it can be reused with almost the same activity. The catalyst was characterized by a combination of various physicochemical techniques, such as N_2 physisorption, TEM, X-ray diffraction and FT-IR.

Keywords Oxidation of cyclohexene · Selenite doped MCM-41 · 2-Acetoxycyclohexanol

1 Introduction

Cyclohexene (CHE) oxidation is an important commercial reaction and the products of oxidation of CHE are important intermediates for the synthesis of medication, pesticide, and insect pheromone etc. [1, 2]. 2-Acetoxycyclohexanol (2-ACHOL) plays significant roles as versatile intermediates of complex natural products and biologically active compounds [3–5]. The classical synthesis methods such as the use of high oxidation state transition metals for CHE oxidation (e.g. chromium VI) [6] need critical conditions, especially, they are polluting and risky. Sodium bismuthate was also used in oxidation of CHE even though the yield of 2-ACHOL was only 25% [7]. These homogeneous media have several disadvantages owing to the typical problems of separation of products from the catalyst in addition to the very low yields. On the other hand, we have demonstrated that cobalt resinate was an efficient catalyst for the synthesis of 2-cyclohexene-1-ol (2-CHOL) and 2-cyclohexene-1-one (2-CHON) by allylic oxidation of CHE under atmospheric pressure of molecular oxygen in the absence of solvent [8]. Furthermore, Schiff base copper complex [9] and its immobilized analogue supported on organically modified silica [10], alumina-supported Mn(II) complexes [11], Nb-containing mesoporous and macroporous materials [12] and dendritic PAMAMSA-Mn(II) complexes [13] have also been used to oxidize CHE.

The well-ordered mesoporous materials, particularly, incorporated with metal ions, have a good use in catalysis for their unusual properties: monodispersed, single-pore distributed, huge pore volume and highly ordered mesoporous structures [14]. For example, Cr-MCM-41, Cr-MCM-48 [15], W-SBA-15 [16], Ti-AlSBA [17], TiO_2–SiO_2 [18] and RuO_2/TiO_2 [19] have been employed as catalysts for the allylic oxidation or epoxidation of CHE with *tert*-butylhydroperoxide or H_2O_2. The major products of the epoxidation of CHE are cyclohexene oxide (epoxide) and 1,2-cyclohexanediol (CHDL) [18–20]. In contrast to intensive studies of those metal ions doped mesoporous materials, so far there is no report on synthesis of selenite doped MCM-41, let alone the study of its catalytic oxidation of CHE that we can find even though electrochemical cycling behavior of selenium-doped $LiMn_2O_4$ spinel was reported [21].

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Here we report that when selenite doped MCM-41 (SeO_3^{2-}/MCM-41) was used as a catalyst, an excellent conversion rate (~100%) of CHE and high selectivity (~81%) of 2-ACHOL was obtained. This seems to be the main production of 2-ACHOL in this way. In particular, the major products were so different from those obtained from the oxidation of CHE by using those metal ions doped mesoporous materials as catalysts, such as Co-MCM-41, Co-SBA-15.

2 Experimental

2.1 Synthesis of SeO_3^{2-}/MCM-41

SeO_3^{2-}/MCM-41 with Si/Se ratio of 50, 40, 20, and 2 were synthesized by a modified process as described in the literatures [22, 23] as follows: 5.0 g cetyltrimethylammonium bromide (CTAB) was added to 66 mL solution containing 1.0 g sodium hydroxide. Then, 20 mL tetraethyl orthosilicate (TEOS) was added to it slowly until the solution became a clear gel. Subsequently the aqueous solution of sodium selenite hydrous was introduced dropwise. The final mixture was stirred for 2 h and then transferred into a Teflon bottle and treated under autogenous pressure without stirring at 363 K for 7 days, filtered, washed, dried and calcined at 823 K in air for 24 h. A detailed description of a process for synthesis of Co-MCM-41, Co/$MTiO_2$ and Co-SBA-15 is available elsewhere [23], [24] and [25], respectively.

2.2 Characterizations

Pore size distributions, BET surface areas and pore volumes were measured by nitrogen adsorption/desorption using a NOVA 2000e gas sorption analyzer (Quantachrome Corp.). Prior to the analysis, the samples were outgassed at 423 K for about 1 h.

High resolution transmission electron microscopy (HRTEM) micrographs were obtained on a JEM Fas-TEM-3010 electron microscope operated at 200 kV.

X-ray power diffraction (XRD) experiments were conducted on a D/max-3B spectrometer with Cu Kα radiation, and scans were made in the 2θ range 0–7° with a scan rate of 0.5°/min (low angle diffraction).

FI-IR measurements were performed on a Thermo Nicolet AVATAR FT-IR 360 instrument. Potassium bromide pellets containing 0.5% of the catalysts were accumulated for each spectrum in transmission at a spectral resolution of 4 cm^{-1}. The spectrum of dry KBr was taken for background subtraction.

XPS measurement was performed on a PHI5500ESCA analyzer. The main parameters were as follows: Mg Kα, 200 W, vacuum ~10^{-7} Pa.

Inductively coupled plasma–atomic emission spectrometry (ICP–AES; Labtam Plasma Lab 8440) analysis was used to determine the percentage of Se of the catalyst in the filtrate obtained from fast hot filtration.

2.3 Oxidation of CHE

The oxidation reactions were carried out at the atmospheri pressure as follows: the catalyst (100 mg), CHE (CP, 2 mL) and 30 mL of solvent (acetic acid, acetonitrile, acetone, methanol, chloroform, *N,N*-dimethylformamide (DMF)) were used as received without further purification and added successively into a temperature controlled, round bottom, three-necked-flask having a reflux condenser. The aqueous H_2O_2 (30%, approximate 5 mL) dissolved in acetic acid was added dropwise after the reaction mixture heated to the set temperature. Reaction mixture was filtered under reduced pressure after the set time (11 h) and the residue was extracted with chloroform. Anhydrous $MgSO_4$ (AR) was used to remove more water from the extracted organic phase. The obtained products were analyzed by gas chromatography using a Thermon 3000 column (10% on Celite545, 1 m, thermal conductivity detector) and GC-MS (Fínnigan GC800 TP/MS Voyager) using DB-5MS capillary column. Reference substances were used for the identification of the products.

3 Results and Discussion

Using above preparing process, SeO_3^{2-}/MCM-41 was white in color with BET surface area of 1,422 m^2/g, pore volume of 0.65 mL/g and pore diameter of 23.1 Å. The N_2 adsorption/desorption isotherm of SeO_3^{2-}/MCM-41 shown in Fig. 1 is a type IV adsorption isotherm, typical of mesoporous materials. The BET surface value (1,422 m^2/g) is much bigger than that of pure MCM-41 silica. Such a difference must be attributed to the presence of microspores. The BJH pore size distribution curve of SeO_3^{2-}/MCM-41 (inset of Fig. 1) shows two primary regions, one is in the microporous region; the other is in the mesoporous region which is majority part of the overall pores. The presence of microspores is not usual for MCM-41 samples.

The HRTEM images of SeO_3^{2-}/MCM-41 in Fig. 2 illustrate the regular hexagonal array of mesoporous channels which is similar to that of MCM-41 prepared under microwave irradiation [26].

The low angle X-ray diffractogram patterns of SeO_3^{2-}/MCM-41 are shown in Fig. 3. The presence of four Bragg angles can be distinguished in hexagonal lattice symmetry, characteristic of MCM-41 structure [27]. The pattern corresponds to regular hexagonal channels of the MCM-41 structure with a strong (100) peak followed by two (110)

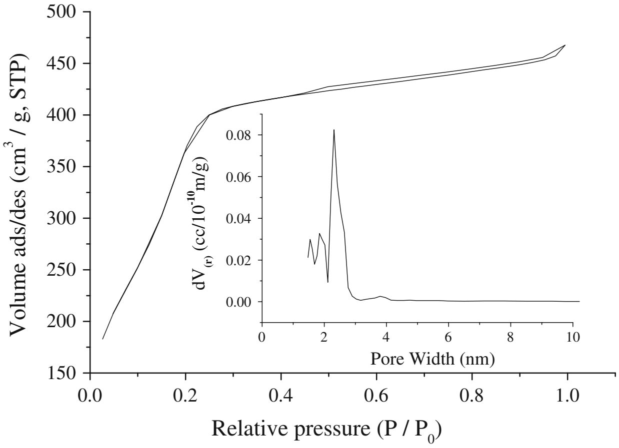


Fig. 1 N_2 adsorption/desorption isotherm and the pore size distribution of SeO_3^{2-}/MCM-41

and (200) peaks in the 2θ ranges of 2–7°, which indicated that the obtained mesoporous sample has a highly ordered pore system with a high porosity [27]. This is supported by the high BET surface areas (1,422 m^2/g) which is also significantly higher than that of Co-MCM-41 (1,041 m^2/g) [23]. This indicates that our selenite was well incorporated into the MCM-41 framework.

In order to investigate the nature of selenium phase of SeO_3^{2-}/MCM-41, the sample was characterized with XPS (not shown). XPS of Se3d indicates the presence of Se^{4+}. The atomic ratio of Se:Si calculated by using the integrated peak area and sensitivity factors, is about 1:55.8 which is close to the Se:Si ratio of 1:50 for the preparing SeO_3^{2-}/MCM-41.

The FT-IR spectra of SeO_3^{2-}/MCM-41 was recorded between 400 and 4,000 cm^{-1} in transmission mode and shown in Fig. 4. It has been reported that vibration band at ca. 1,090 cm^{-1} was assigned to the symmetric stretching of Si–O–Si vibration of the MCM-41 [28]. In the hydroxyl region (3,000–4,000 cm^{-1}), the broad band is observed at ca. 3,420 cm^{-1} for SeO_3^{2-}/MCM-41. The band shift from 3,444 to 3,420 cm^{-1} for SeO_3^{2-}/MCM-41 compared with MCM-41 is probably attributed to the presence of more defect sites (SiOH group). This band was assigned to

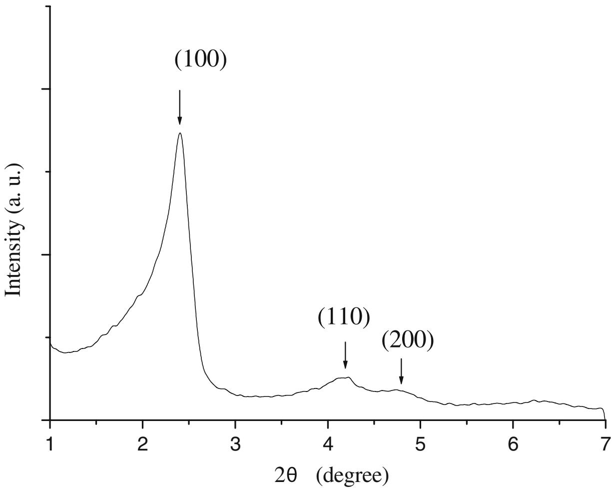


Fig. 3 XRD patterns of SeO_3^{2-}/MCM-41

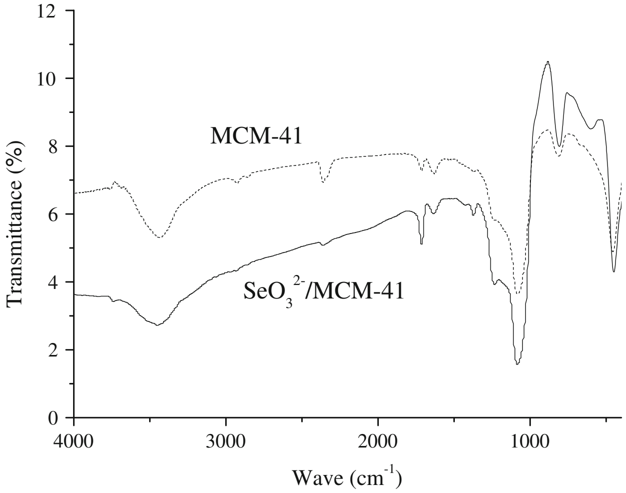
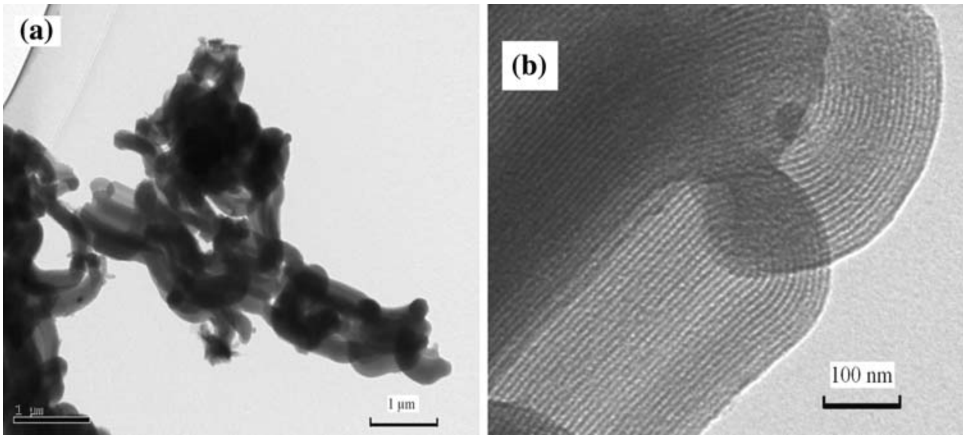


Fig. 4 FT-IR spectra of SeO_3^{2-}/MCM-41

Fig. 2 HRTEM images of SeO_3^{2-}/MCM-41



silanol group vibrations situated inside the channels of MCM-41, resulting from silanol groups interacting via hydrogen bonding [28].

For comparison, the oxidation of CHE with the aqueous H_2O_2 over different catalysts is summarized in Table 1. The catalysts are compared under similar conditions and found that 2-ACHOL was obtained as the main products, a small amount of (CHDL) and 2-CHON were also detected. As expected, without adding any catalyst, no meaningful activity (conversion of CHE was ~5%) was observed. Undoped MCM-41 exhibited extremely low activity. On the other hand, since Co-MCM-41 [23], Co-SBA-15 [29] and Co/MTiO$_2$ [24] exhibited efficient catalytic activity in selective oxidation of diphenylmethane to benzophenone, 2-methyl pyridine to 2-pyridinecarboxylic acid and cyclohexane to cyclohexanol and cyclohexanone, respectively, they were compared with SeO_3^{2-}/MCM-41. It is apparent that SeO_3^{2-}/MCM-41 exhibits highest conversion and TON. This may suggest that BET surface areas of the catalysts would play a more important role because the BET surface areas of SeO_3^{2-}/MCM-41 (1,442 m^2/g) is much bigger than those of Co-MCM-41 (1,041 m^2/g) [23], Co-SBA-15 (690 m^2/g) [25] and Co/MTiO$_2$ (79.8 m^2/g) [24]. Moreover, the micro/mesoporous structure makes SeO_3^{2-}/MCM-41 a highly effective catalyst since the intradiffusion resistance is minimized and the efficiency of selective oxidation is enhanced. Other factors such as the difference between the dopant ions, number of active sites, difference between silica and titania, can also modify the efficiency. Therefore, the advantage of SeO_3^{2-}/MCM-41 over those metal ions doped mesoporous materials is obvious. Due to the good performance of SeO_3^{2-}/MCM-41, in the following we concentrate on the study of the influence of various parameters on the CHE conversion and selectivity of 2-ACHOL over SeO_3^{2-}/MCM-41.

The effects of various solvents on the reaction are summarized in Table 2. It shows that a significantly lower conversion was obtained in the case of methanol, acetone, acetonitrile, DMF and chloroform. This is due to possible partial decomposition of H_2O_2 because it was reported that the decomposition of H_2O_2 are faster in these solvents than in acetic acid [30]. The higher catalytic activity of SeO_3^{2-}/

Table 1 Comparison of catalytic activities of different catalysts for the oxidation of CHE

Catalyst	Conversion of CHE (wt%)	TON	Selectivity (wt%)			
			2-CHON	CHDL	2-ACHOL	By-products
Co/MTiO$_2$	66.4	557	10.0	5.7	38.6	45.7
Co-SBA-15	59.3	381	9.0	7.5	42.4	41.1
Co-MCM-41	80	514	6.7	7.7	46.6	39.0
SeO_3^{2-}/MCM-41	~100	646	–	7.7	81.1	11.2
MCM-41	13.2	2	20.3	1.1	1.6	77.0
First recycle	93.1	601	4.9	8.1	74.0	13.0
Second recycle	95.1	614	3.1	8.3	73.5	15.1

Reaction condition: 2 mL CHE; 100 mg catalyst; 30 mL acetic acid; 5 mL H_2O_2; 350 K for reaction temperature; 11 h reaction time

TON turn over number (millimole of oxidized products per millimole of cobalt or selenium in the catalysts)

By-products were cyclohexanol, 2-cyclohexene-1-ol, and products of the diol cleavage

Table 2 Effect of solvents on the CHE reaction

Solvents	Conversion of CHE (wt%)	TON	Selectivity (wt%)		
			2-ACHOL	CHDL	By-products
Acetic acid	100	646	81.1	7.7	11.2
Methanol	10.9	70	4.2	8.3	88.1
Acetonitrile	4.1	27	13.4	42.7	43.9
Acetone	7.8	50	–	4.8	95.2
Chloroform	18.8	121	22.7	59.6	17.7
DMF	17.3	112	35.2	24.8	40.0

Reaction condition: 2 mL CHE; 100 mg catalyst; 30 mL solvent; 5 mL H_2O_2; 350 K for reaction temperature (except for methanol at 340 K, acetone at 330 K);11 h reaction time

TON turn over number (millimole of oxidized products per millimole of selenium in the catalyst)

By-products were cyclohexanol, 2-cyclohexene-1-one, 2-cyclohexene-1-ol, and products of the diol cleavage

MCM-41 is due to that selenite ions were incorporated in the framework positions of MCM-41 during synthesis [31]. Moreover, it has been reported that acetic acid does not only act as a solvent, but also serves as a good oxidizing agent because of the formation of the framework titanium–peracetic acid complex when it reacts with H_2O_2 in the presence of titanium-containing zeolites (TS-1) [30, 32, 33]. Similarly, if in our system a complex with peracetic acid was possibly formed in the pores of SeO_3^{2-}/MCM-41 then it would be relatively more hydrophobic and stable, as compared to H_2O_2. In short, the synergistic effects among doped selenite, mesoporous framework of MCM-41, acetic acid and H_2O_2 make SeO_3^{2-}/MCM-41 an effective catalyst for the oxidation of CHE under mild conditions.

The effect of reaction temperature on the reaction over SeO_3^{2-}/MCM-41 is shown in Fig. 5. It is clear that the conversion of CHE was found to increase with increase in reaction temperature and passed through a maximum at 330 K, but the highest selectivity was obtained at 340 K. A further increase in the reaction temperature resulted in slight decrease in the conversion but significant decrease in the selectivity of CHDL, probably owing to a quicker decomposition of H_2O_2 at higher temperature and the formation of cyclohexyl acetate by interaction of cyclohexyl and acetoxy radicals and/or due to the reaction between CHDL with excess acetic acid. Considering the effect of temperature on both conversion of CHE and selectivity of 2-ACHOL, 350 K was chosen as the suitable temperature for the oxidation of CHE.

The effect of reaction time on CHE reaction over SeO_3^{2-}/MCM-41 was also investigated and depicted in Fig. 6. It is seen that the conversion of the CHE increased with time up to 11 h while the selectivity of 2-ACHOL was 81%; meanwhile selectivity of CHDL was only 1%. Therefore, the optimum conversion and selectivity could be achieved at about 11 h.

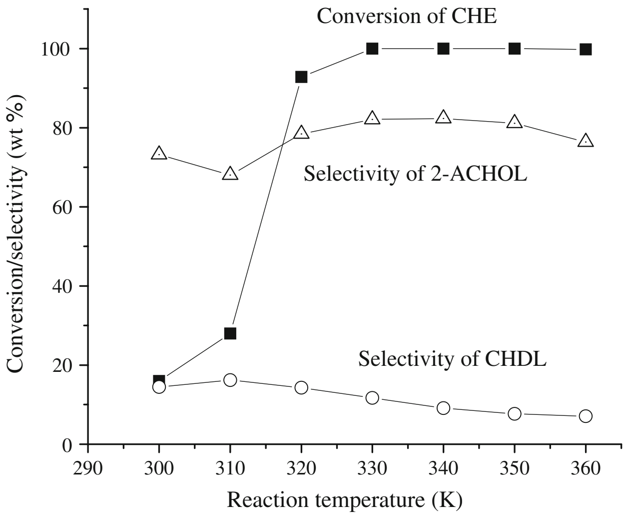


Fig. 5 Effect of temperature on the conversion and selectivities over SeO_3^{2-}/MCM-41 (Reaction conditions: 2 mL CHE; 30 mL acetic acid; 5 mL H_2O_2 (30%); reaction time, 11 h; catalyst, 100 mg)

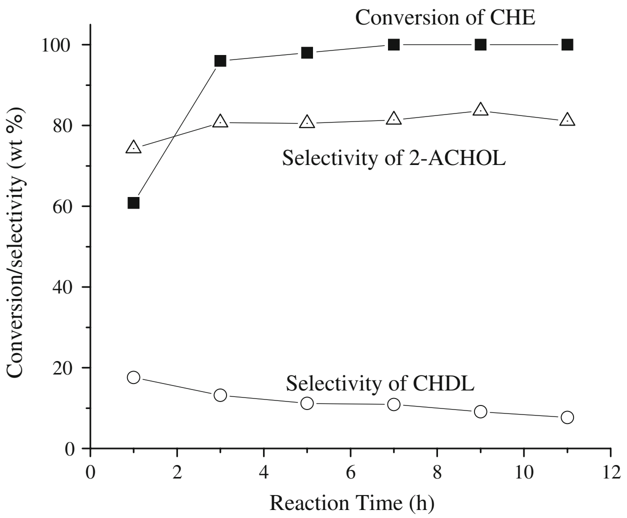


Fig. 6 Effect of reaction time on the conversion and selectivities over SeO_3^{2-}/MCM-41 (Reaction conditions: 2 mL CHE; 30 mL acetic acid; 5 mL H_2O_2 (30%); reaction temperature, 350 K; catalyst, 100 mg)

Figure 7 presents the effect of catalyst concentration on the reaction over SeO_3^{2-}/MCM-41 at 11 h by using acetic acid as solvent. An initial steep increase in the conversion of CHE and a slightly increase in selectivity of 2-ACHOL were observed when the amount of the catalyst was increased up to 100 mg. Beyond this amount, the conversion of CHE and selectivity of 2-ACHOL only increased a little. Therefore, 100 mg was selected as the suitable amount of the catalyst for the selectively producing

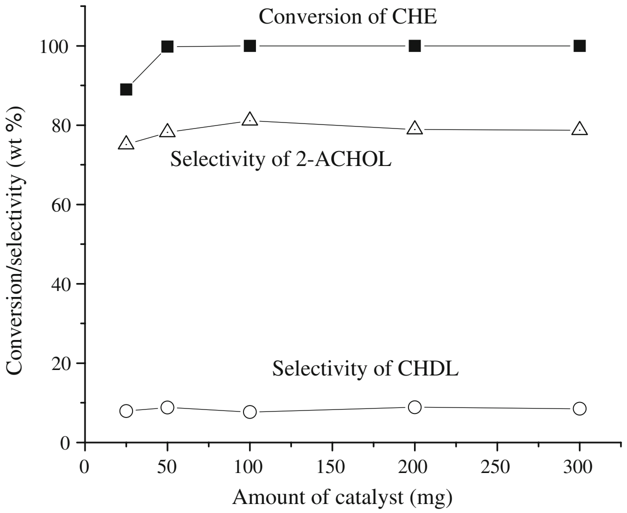


Fig. 7 Effect of the amount of catalyst on the conversion and selectivities over SeO_3^{2-}/MCM-41 (Reaction conditions: 2 mL CHE; 30 mL acetic acid; 5 mL H_2O_2 (30%); reaction temperature, 350 K; reaction time, 11 h)

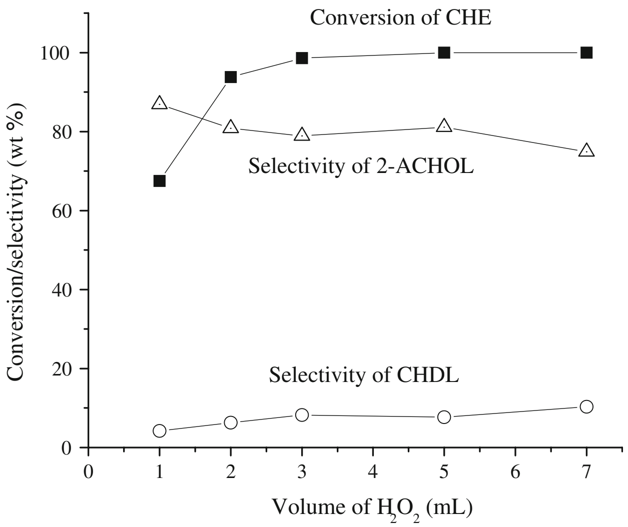


Fig. 8 Effect of the volume of H_2O_2 on the conversion and selectivities over SeO_3^{2-}/MCM-41 (Reaction conditions: 2 mL CHE; 30 mL acetic acid; reaction temperature, 350 K; reaction time, 11 h; catalyst, 100 mg)

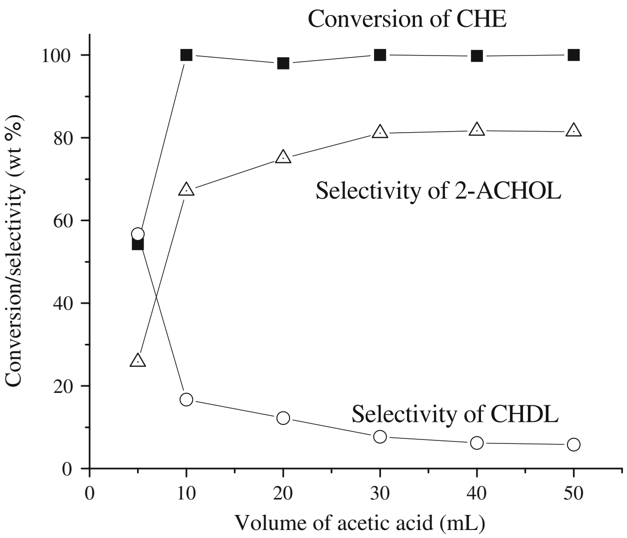


Fig. 9 Effect of volume of acetic acid on the conversion and selectivities over SeO_3^{2-}/MCM-41 (Reaction conditions: 2 mL CHE; 5 mL H_2O_2 (30%); reaction temperature, 350 K; reaction time, 11 h; catalyst, 100 mg)

2-ACHOL. These results indicate that in the system catalyzed by SeO_3^{2-}/MCM-41 only small amount of catalyst is active in the oxidation of CHE at higher amount of catalyst.

The effects of the volume of H_2O_2 and acetic aced on CHE reaction over SeO_3^{2-}/MCM-41 at 11 h were shown in Figs. 8 and 9, respectively. In Fig. 8, it is clear that the conversion of CHE was found to increase with increase in volume of H_2O_2 and passed through a maximum at 5 mL while the selectivity of 2-ACHOL decreased with increase in volume of H_2O_2. Differently, the selectivity of CHDL increased. In contrast, Fig. 9 shows that the conversion of the CHE and the selectivity of 2-ACHOL increased with the volume of acetic acid while the selectivity of CHDL decreased. Considering these effects, 5 mL H_2O_2 and 30 mL acetic acid were chosen as suitable volumes in this study, respectively. Meanwhile, the influence of the Se loading on the catalytic activity is shown in Table 3. It indicates that the suitable ratio of Si/Se is 50.

In order to prove whether the catalyst is a heterogeneous one, experiments with fast hot catalyst filtration and studying the reactivity of the filtrate had been done by a modified process as [34]: 100 mg catalyst, 2 mL CHE, 30 mL HAc and 2 mL H_2O_2 was stirred at the temperature of 350 K for about 30 min (conversion of CHE was 26%), then quickly filtrate the catalyst. Let the filtration to react further with by adding 3 mL H_2O_2. We found that after hot filtration the filtrate reacted little further comparing that observed when the catalyst was not filtered. This observation was also well supported by ICP–AES analysis of the filtrates obtained from the fast hot catalyst filtration where negligible amount of leaching of active Se ($\leq$0.001 wt%) was detected. These indicated that the active component (selenite) did not leach to the solution and the SeO_3^{2-}/MCM-41 did act as a heterogeneous catalyst.

To study the stability and recycling ability of SeO_3^{2-}/MCM-41 under reaction conditions, recycling experiments were carried out. The reactions were also under the optimum conditions with those used for SeO_3^{2-}/MCM-41. The typical recycling procedure was as follows: after the initial reaction, the catalyst was separated from the reaction mixture and washed with acetone and dried at 363 K, followed by the activation at 723 K for 4 h. The catalyst SeO_3^{2-}/MCM-41 showed excellent reusability in the oxidation reactions. The results are also summarized in Table 1. The major product was also 2-ACHOL, and the conversion of CHE and the selectivity of 2-ACHOL were stable after first recycle.

Based on the above experimental results and the mechanisms proposed for the oxidation of CHE in [15, 35], a possible reaction pathway for the oxidation of CHE over SeO_3^{2-}/MCM-41 catalyst is depicted in Scheme 1.

At first, acetic acid interacts with H_2O_2 leading to the formation of peracetic acid 1a, which further reacts with Se species 1b to produce selenium peroxospecies 2a. The latter than interacts with CHE to produce a complex of the type 2b and leads to the formation of epoxide, 2c (7-oxabicyclo [4, 1, 0] heptane). Obviously, ring-opening of epoxide is the major side reactions in oxidation of CHE when using SeO_3^{2-}/MCM-41 as a catalyst which resulted in the formation of its corresponding diol (CHDL) and ester (2-ACHOL) in acetic acid.

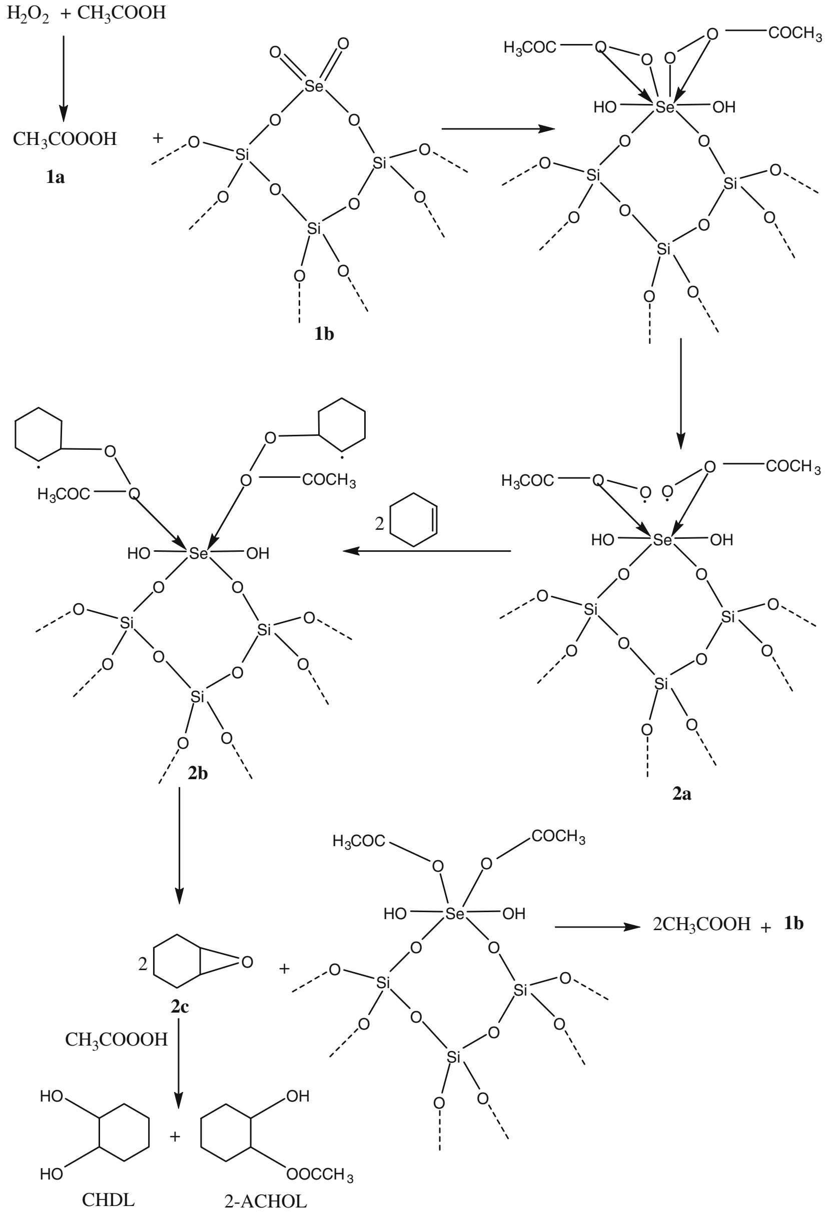
Table 3 Influence of the Se loading on the catalytic activity

Ratio of Si/Se	Conversion of CHE (wt%)	TON	Selectivity (wt%)		
			CHDL	2-ACHOL	By-products
2	63.4	25	3.4	56.8	39.8
20	71.9	181	4.7	61.2	34.1
40	82.1	401	6.5	69.4	24.1
50	~100	646	7.7	81.1	11.2

Reaction condition: 2 mL CHE; 100 mg catalyst; 30 mL acetic acid; 5 mL H_2O_2; 350 K for reaction temperature; 11 h reaction time

TON turn over number (millimole of oxidized products per millimole of selenium in the catalysts)

By-products were 2-cyclohexene-1-one, cyclohexanol, 2-cyclohexene-1-ol, and products of the diol cleavage



Scheme 1 A possible reaction pathway for the oxidation of CHE over SeO_3^{2-}/MCM-41 catalyst

4 Conclusions

In conclusion, SeO_3^{2-}/MCM-41 was an efficient and high substrate conversion catalyst for the oxidation of CHE under relatively mild reaction conditions without adding any initiator. Recycling of the catalyst indicates that the catalyst can be reused with almost the same activity.

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References

1. Ingold KU (1989) Aldri Chim Acta 22:69
2. Madhavareddy M, Purmiyamurthy T, Cobalt JI (1995) Tetrahedron Lett 36:159
3. Prior MJ, Whitham GH (1986) J Chem Soc Perkin Trans 1:683
4. Hudson LF, Parker TM, Whittaker D (1993) J Chem Soc Chem Commun 1427
5. Gupton JT, Duranceau SJ, Miller JF, Kosiba ML (1988) Synth Commun 18:937
6. Roček J, Drozd JC (1970) J Am Chem Soc 92:6668
7. Truesdale LK, Reuman ME (1980) J Org Chem 45:726
8. Yin C, Yang Z, Li B, Zhang F, Wang J, Ou E (Accepted) Catal Lett
9. Mukherjee S, Samanta S, Roy BC, Bhaumik A (2006) Appl Catal A: Gen 301:79
10. Wang R, Hao C, He Y, Xia C, Wang J, Wang Y (2000) J Appl Polym Sci 75:1138
11. Salavati Niasari M, Farzaneh F, Ghandi M (2002) J Mol Catal A: Chem 186:101
12. Nowak I, Kilos B, Ziolek M, Lewandowska A (2003) Catal Today 78:487
13. Yang Z, Kang Q, Ma H, Li C, Lei Z (2004) J Mol Catal A: Chem 213:169
14. Corma A (1997) Chem Rev 97:2373
15. Sakthivel A, Dapurkar SE, Selvam P (2003) Appl Catal A: Gen 246:283
16. Cheng CY, Lin KJ, Prasad MR, Fu SJ, Chang SY, Shyu SG, Sheu HS, Chen CH, Chuang CH, Lin MT (2007) Catal Commun 8:1060
17. Lapisardi G, Chiker F, Launay F, Nogier JP, Bonardet JL (2004) Catal Commun 5:277
18. Okia AR, Xu Q, Shpeizer B, Clearfield A, Qiu X, Kirumakki S, Tichy S (2007) Catal Commun 8:950
19. Sreethawong T, Yamada Y, Kobayashi T, Yoshikawa S (2006) J Mol Catal A: Chem 248:226
20. Rinaldi R, Sepúlveda J, Schuchardt U (2004) Adv Synth Catal 346:281
21. Yoon CS, Kim CK, Sun Y-K (2002) J Power Source 109:234
22. Yao W, Chen Y, Min L, Fang H, Yan Z, Wang H, Wang J (2006) J Mol Catal A: Chem 246:162
23. Chang F, Li W, Xia F, Yan Z, Xiong J, Wang J (2005) Chem Lett 34:1540
24. Yao W, Fang H, Ou E, Wang J, Yan Z (2006) Catal Commun 7:387
25. Xia F, Ou E, Wang L, Wang J (2008) Dyes Pigments 76:76
26. Wu C, Gao Q, Hu J, Chen Z, Shi W (2009) Micropor Mesopor Mater 117:165
27. Carvalho WA, Varaldo PB, Wallau M, Schuchardt U (1997) Zeolites 18:408
28. Laha SC, Mukherjee P, Sainkar SR, Kumar R (2002) J Catal 207:213
29. Li B, Li L, Sun L, Xia F, Wang S, Wang J (2008) Catal Commun 9:2287
30. Sooknoi T, Limtrakul J (2002) Appl Catal A: Gen 233:227
31. Kadgaonkar MD, Laha SC, Pandey RP, Kumar P, Mirajkar SP, Kumar R (2004) Catal Today 97:225
32. Velusamy S, Punniyamurthy T (2003) Tetrahedron Lett 44:8955
33. Sato T, Dakka J, Sheldon RA (1994) J Chem Soc Chem Commun 16:1887
34. Sheldon R, Wallau M, Arends I, Schuchardt U (1998) Acc Chem Res 31:485
35. Muzart J (1992) Chem Rev 92:113