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Tetralin oxidation over chromium-containing molecular sieve catalysts

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Abstract

A series of catalysts in which a transition metal (Fe, Co, and Cr) is incorporated into MCM-41, hexagonal mesoporous aluminophosphate (HMA), and $AlPO_4$-5 matrix was prepared and tested for liquid phase oxidation of tetralin using *tert*-butyl hydroperoxide as an oxidizing agent. In general, chromium-containing catalysts showed higher catalytic activity as well as higher α-tetralone selectivity than iron or cobalt-incorporated catalysts. In addition, HMA proved more effective as a support material than MCM-41. CrHMA was the most active (64.9% conversion) and selective (80% α-tetralone selectivity) catalyst, but developed metal leaching due to its amorphous pore walls. $CrAlPO_4$-5, having a crystalline structure, showed higher resistance to metal leaching than CrHMA.

Keywords: Tetralin; α-Tetralone; *tert*-Butyl hydroperoxide; Mesoporous aluminophosphate; $CrAlPO_4$-5

1. Introduction

Oxidation of benzylic heterocyclic compounds has been extensively employed in the synthesis of fine and specialty chemicals. Selective oxidation of tetralin produces α-tetralone, an important source of synthetic precursors and a reactive intermediate for a wide range of products including pharmaceuticals, dyes, and agrochemicals [1–3]. α-Tetralone is also used as an additive to enhance the cetane number in diesel fuels [4].

The majority of industrial oxidation processes employ catalysis by various metal complexes in homogeneous reaction mode [5,6]. Numerous studies of the reaction have been carried out either in solvent or in undiluted tetralin using metalloporphyrin catalysts [7] or in biphasic autoxidation [8,9]. A heterogeneously catalyzed reaction is obviously more desirable considering the separation problems. In this regard, liquid phase oxidation of tetralin catalyzed by several transition metal oxide catalysts, including Ti, Ni, Cu, and Cr has been investigated using various oxygen sources [10–13]. Recently, it has been shown that redox metal-containing molecular sieves are effective heterogeneous catalysts in oxidation reactions [14]. The transition metal ions in these catalysts can be either incorporated in true framework sites, anchored to the framework in defect sites, or present as isolated/clustered extra-framework cations.

We herein report on the synthesis of selected transition metal containing hexagonal mesoporous aluminophosphate (HMA), MCM-41, and $AlPO_4$-5 and compare their catalytic performances in liquid phase oxidation of tetralin.

2. Experimental

2.1. Catalyst preparation

Transition metal substituted HMA catalysts were hydrothermally synthesized following a procedure reported earlier [15]. Aluminum hydroxide, phosphoric acid (85 wt% in water), chromium, cobalt, and iron acetate reagents supplied by Aldrich were used as received. Cetyltrimethylammonium bromide (CTAB) was employed as a structure-directing template in the synthesis. In a typical synthesis, 3.46 g of aluminum hydroxide was added to 25 g of distilled water and 2.5 ml phosphoric acid was added under constant stirring for 1 h. The mixture was then mixed with 4.0 g of CTAB dissolved in 30 g water. The combined mixture was stirred for an

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additional 3 h and then an appropriate amount of metal salts dissolved in 15 g water was added into the gel mixture. pH was adjusted slowly to 9.5 by adding tetramethylammonium hydroxide (TMAOH) (25 ml, 25 wt% aqueous solution, Aldrich) and the solution was stirred for 72 h at room temperature. The metal content for the standard HMA catalyst in the substrate was controlled to 2 mol% and the gel composition of the synthesis mixture was 0.04 Cr_2O_3 (0.04 Fe_2O_3, 0.08 CoO): Al_2O_3: P_2O_5: 0:5 CTAB: 8.5 TMAOH: 400 H_2O. The resulting solid product was recovered by filtration, washed repeatedly with de-ionized water, and dried overnight in an oven at 80 °C. The as-synthesized materials were then calcined at 500 °C for 10 h. For CrHMA, catalysts with chromium content of 1, 3, and 5 mol% were also prepared.

The metal (Cr, Co and Fe) substituted MCM-41 and microporous $AlPO_4$-5 were hydrothermal synthesized according to reported procedures [16,17] with 2 mol% metal content in the synthesis substrates.

2.2. Characterization

Structural characterization of the catalyst samples was performed by powder XRD (Rigaku, DMAX 2500) using Ni-filtered Cu Kα radiation. The metal content and morphology of the samples were analyzed using TEM/EDX (Philips, CM 200). Total BET surface area, pore volume, and average pore diameter were determined by nitrogen physisorption at −196 °C using a Micromeretics ASAP2000 automatic analyzer and the pore size distribution was calculated by the Barrett–Joyner–Halenda (BJH) method using the desorption branch of the isotherm. Prior to the measurement, samples were degassed at 100 °C for 1 h and at 350 °C for 4 h at a pressure less than 1.4 Pa. UV–vis diffuse reflectance spectroscopy was performed on a Varian CARY3E double beam spectrometer using MgO as a reference in the range of 200–800 nm in ambient conditions.

2.3. Tetralin oxidation

Catalytic reactions were carried out using a Chemistation PPS-2510 fitted with a condenser (Eyela). The oxidations were carried out in the liquid phase using 50 mg of the calcined catalyst, 5 ml of the chlorobenzene (solvent), and tetralin (8 mmol). In a typical reaction, a mixture containing the substrate, solvent, and catalyst were put into a Chemistation glass reactor and heated to 120 °C. The oxidant (TBHP, 16 mmol) was then added through a septum to the reactant mixture and stirred at 500 rpm. These reactions were carried out with an oxidation stoichiometeric molar ratio of substrate to oxidant of 1:2 and continued for 8–12 h. The product sample obtained after removing the solid phase using a HPLC syringe filter was analyzed by FID in a GC (Acme 6000 Series, Younglin instruments) employing a high performance HP-5 capillary column (60 m × 0.32 mm). Identification of reaction products was performed in a Varian 1200L single quadrupole GC/MS system using authentic standard compounds.

2.4. Recycling and chemical washing tests

Stability of the catalyst was tested by carrying out recycle runs of tetralin oxidation. Typically, catalyst powder was separated from the reaction mixture after a catalytic run, washed with acetone, and dried at 80 °C. Finally, the catalyst was calcined at 500 °C for 6 h in air to remove the adsorbed species. In another experiment, the original catalyst was treated with ammonium acetate in order to remove the extra-framework chromium species [16]. In a typical procedure, about 100 mg of the calcined catalyst was washed with 1 M ammonium acetate (30 ml) solution under constant stirring for 12 h at room temperature. The catalyst was then filtered and activated via the same procedure as that employed for the recycled catalyst, i.e., heat treatment at 500 °C in air for 6 h. Tetralin oxidation reaction was then carried out using the recycled or ammonium acetate-washed catalysts. Experiments using the filtrate of the reaction product were also carried out: after removing the catalyst from the reaction mixture by centrifuging at 15,000 rpm and passing it through a separation filter, 1 g filtrate solution was added to a fresh reaction mixture. The feed composition before and after the reaction was analyzed using a GC to test the possibility of the reaction being catalyzed by the dissolved Cr species.

3. Results and discussion

The XRD patterns of the calcined Cr-, Co-, and Fe-substituted molecular sieve catalysts are shown in Fig. 1. All the Cr-, Co-, and Fe-substituted HMA samples maintain an intense

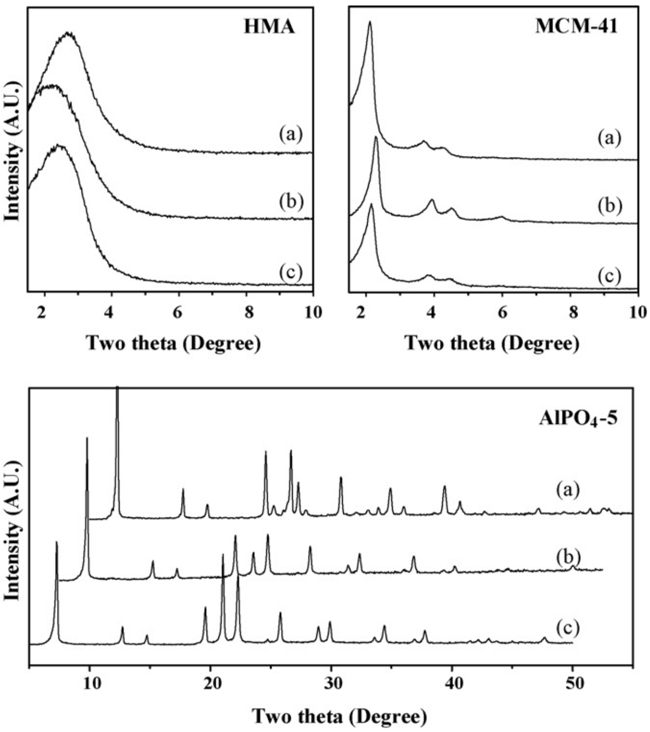

Fig. 1. XRD patterns of (a) Fe, (b) Co, and (c) Cr-substituted molecular sieve catalysts.

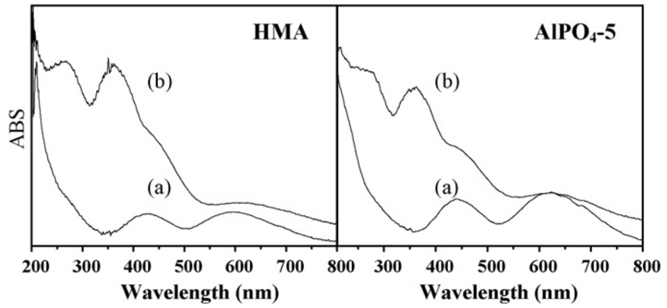

Fig. 2. DRS-UV–vis spectra of Cr-containing molecular sieve catalysts: (a) as-synthesized and (b) calcined.

peak due to (1 0 0) reflection in the low angle region, characteristic of hexagonal mesoporous aluminophosphates [18], whilst the X-ray diffractograms of the Cr-, Co-, Fe-substituted MCM-41 samples display three well resolved reflections corresponding to (1 0 0), (1 1 0), and (2 0 0) planes in a hexagonal lattice structure. The XRD patterns of the transition metal containing AlPO$_4$-5 series also confirm the typical characteristics of microporous aluminophosphates (AlPO$_4$-5), as reported in the literature [19]. The nitrogen adsorption–desorption isotherms of 1–3 mol% CrHMA samples were confirmed to have type IV isotherms, as reported by Selvam and Mohapatra [20]; as the relative pressure increases ($P/P_0 > 0.3$), all the isotherms exhibited an inflection characteristic of capillary condensation due to uniform mesopores (not shown). The corresponding textural properties in BET surface areas between 477 and 680 m^2/g and pore volumes in a range of 0.23–0.31 cm^3/g were obtained. Two percent CrHMA showed the highest surface area and pore volume.

The UV–vis spectra of CrHMA samples are shown in Fig. 2. The spectra of as-synthesized CrHMA and CrAlPO$_4$-5 show two intense bands at ca. 440 and 610 nm due to the spin-allowed d–d transitions $^4A_{2g} \rightarrow {}^4T_{1g}$ and $^4A_{2g} \rightarrow {}^4T_{2g}$, respectively. However, in the AlPO$_4$-5 sample, $^4A_{2g}(F) \rightarrow {}^4T_{2g}(F)$ transition was observed at 630 nm. The spectra of the calcined CrHMA and CrAlPO$_4$-5 sample, on the other hand, show two strong bands at 260 and 362 nm due to charge transfer (O → Cr) in chromate species, with a weak shoulder at 445 nm due to polychromate species [18,21]. It is clearly demonstrated that chromium species are mostly well isolated in the CrHMA matrix but a small amount of polychromates species is also present. The as-synthesized CrHMA samples were green in color and changed to yellow upon calcination. The former coloration is due to the presence of trivalent chromium ions and the latter is due to the presence of higher-valent chromium ions, i.e., chromate, dichromate and/or polychromates. The intensity of the color from light green to dark green increases proportionally to the metal loading, and these change are observed in both mesoporous and microporous materials.

A catalyst screening test for the liquid phase tetralin oxidation reaction using TBHP as an oxidant was conducted to compare catalytic performances among the Cr-, Co-, and Fe-containing molecular sieve catalysts. The tetralin conversion and product distribution data are summarized in Table 1. α-Tetralone was the major product followed by tetralol, naphthalene, and α-napthol. Trace amounts of 1,4-dihyroxytetralin, 4-hydroxy-1-tetralone, and 1,4-napthaquinone were also detected. Blank reactions with TBHP but in the absence of a catalyst under the same experimental conditions yielded no oxygenated products, thus confirming that the formation of oxygenated products is promoted by the transition metal species in the micro/mesoporous matrix.

HMA group catalysts exhibit superior catalytic activity and α-tetralone selectivity compared to siliceous MCM-41 and microporous AlPO$_4$-5 catalysts. Among the different transition metals (Cr, Co, Fe) in the HMA matrix, CrHMA exhibited higher conversion of tetralin accompanied by higher selectivity to α-tetralone. CrHMA showed ca. 65% conversion of tetralin with ca. 80% selectivity to α-tetralone. The observed high conversion and high selectivity of the catalyst are believed to be a consequence of the catalytically active hexavalent chromium incorporated in HMA with uniform mesopores. A small amount of oxidatively dehydrogenated product, α-naphthol, was also formed. The catalytic performances of CoHMA and FeHMA were similar: 30–31% tetralin conversion and 67–68% selectivity to α-tetralone and moderate selectivity to tetralol.

Among the MCM-41 group catalysts, CrMCM-41 again performed the best in terms of tetralin conversion (40.7%) and selectivity to α-tetralone (82.1%). FeMCM-41 and CoMCM-41 behaved similarly with low catalytic activity while significantly higher selectivity to naphthalene than in aluminophosphate-

Table 1
Oxidation of tetralin over transition metal incorporated molecular sieves

Catalysts	Tetralin conversion (%)	Product selectivity (%)			
		Naphthalene	α-Tetralone	Tetralol	α-Naphthol
CrMCM-41	40.7	3.8	82.1	11.7	2.4
CoMCM-41	13.7	16.3	71.7	8.5	3.5
FeMCM-41	11.8	18.6	72.6	4.9	3.9
CrAlPO$_4$-5	61.4	2.8	78.0	4.7	14.5
CoAlPO$_4$-5	33.0	5.9	46.2	15.9	32.0
FeAlPO$_4$-5	21.6	11.5	32.0	10.2	46.3
CrHMA	64.9	2.0	80.0	3.5	14.5
CoHMA	30.7	2.4	67.5	30.1	0.0
FeHMA	30.0	6.2	67.6	24.8	1.4

Reaction conditions: 5 ml chlorobenzene, 8 mmol tetralin, 16 mmol TBHP, and 50 mg catalyst, 120 °C, 12 h. Metal content in the catalyst was 2 mol%.

based catalysts was observed. Among the $AlPO_4$-5 catalyst series, $CrAlPO_4$-5 performed well with high selectivity (78%) to α-tetralone accompanied by 61% conversion. Fe- and Co-containing $AlPO_4$-5 exhibited relatively poor selectivity to ketone and a significant amount of α-naphthol was formed.

In terms of α-tetralone yield, the following order was observed among the Cr-based catalysts: CrHMA > $CrAlPO_4$-5 > CrMCM-41. The better catalytic performance in HMA can be attributed to the hydrophilic cavities, which offer high surface area and high accommodation capacity for transition metal ions in the aluminophosphate matrix. HMAs may not be suitable for oxidation in aqueous media, i.e., with aqueous H_2O_2, but they appear to compatible with TBHP. In addition, mesopores in HMA are apparently more advantageous for enhanced diffusion than are micropores in $AlPO_4$-5, given the sizes of the organic molecules involved in the reaction. On the other hand, $AlPO_4$-5, with a crystalline wall structure, may provide higher stability against metal leaching than HMA, which may develop a stability problem due to its amorphous pore walls. The hydrophobic nature of the silica matrix, as well as a lower capacity to accommodate transition metals in MCM-41, could at least in part account for the lower tetralin oxidation activity of the transition metal substituted MCM-41 catalysts [6].

A general reaction path suggested for tetralin oxidation is given in Fig. 3. A catalyst is proposed to interact with TBHP to form a peroxo-complex, which subsequently interacts with a substrate molecule to give the intermediate product of 1-*tert*-butylperoxytetralin [22,23]. This intermediate product was, however, not detected in the product analysis during runs due to its short lifetime. The relatively slow reaction step of tetralol to α-tetralone has also been proposed [24].

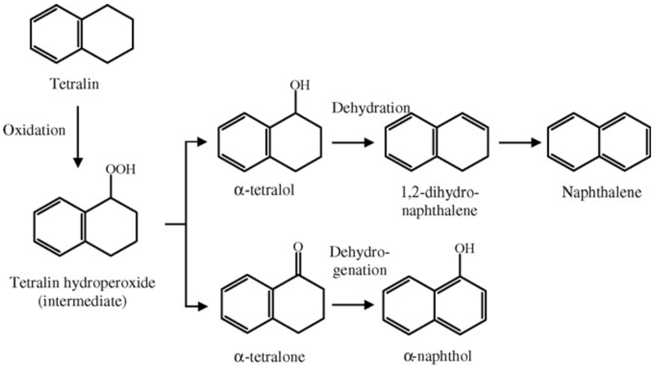

Fig. 3. Mechanism and reaction pathway of tetraline oxidation.

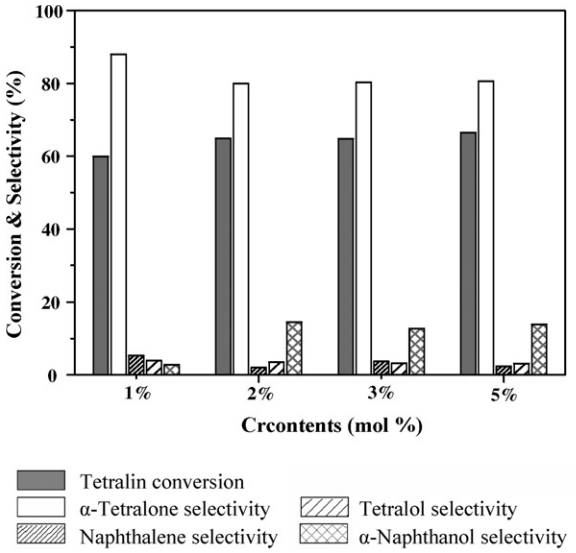

Fig. 4. . Catalytic performance of CrHMA with different Cr contents; reaction conditions: 5 ml chlorobenzene, 8 mmol tetralin, 16 mmol TBHP, and 50 mg catalyst, 120 °C, 12 h.

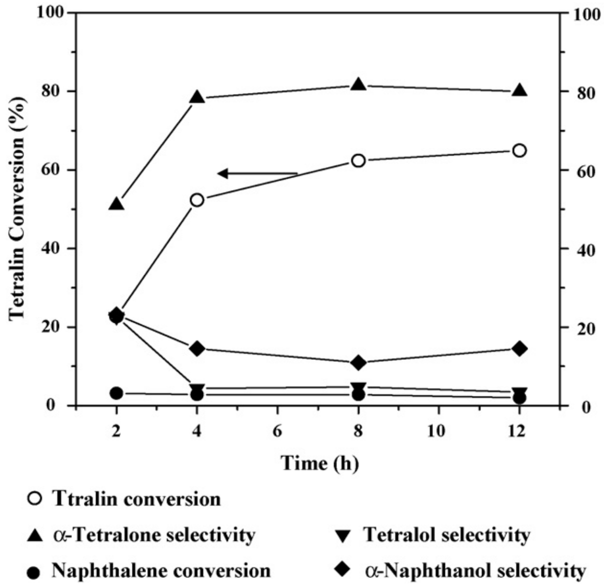

Fig. 5. Conversion and selectivity profiles of tetralin oxidation on 2% CrHMA catalyst; reaction conditions: 5 ml chlorobenzene, 8 mmol tetralin, 16 mmol TBHP, and 50 mg catalyst, 120 °C.

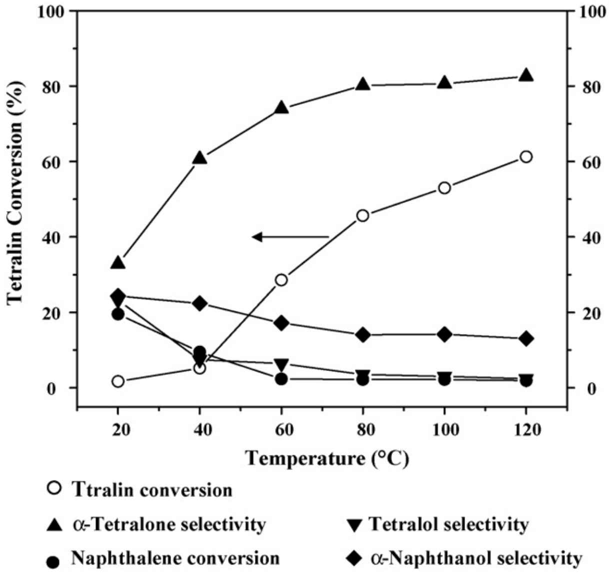

Fig. 6. Effect of temperature on tetralin oxidation using 2% CrHMA catalyst; reaction conditions: 5 ml chlorobenzene, 8 mmol tetralin, 16 mmol TBHP, and 50 mg catalyst, 8 h.

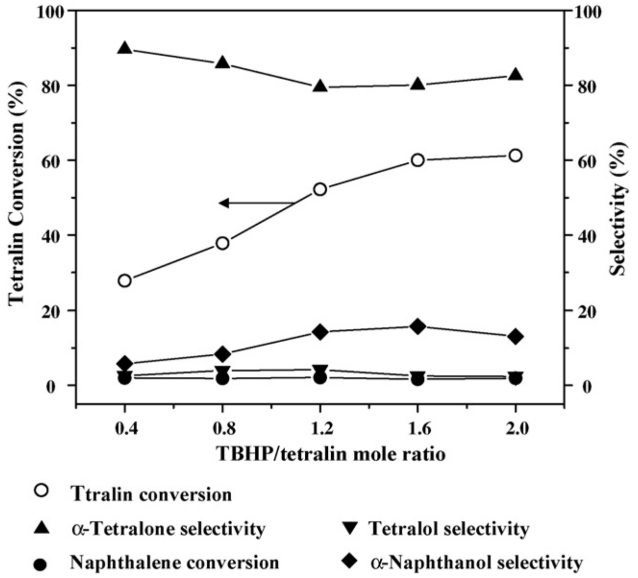

Fig. 7. Influence of TBHP/tetralin mole ratio on tetralin oxidation using 2% CrHMA; reaction conditions: 5 ml chlorobenzene, 8 mmol tetralin, 16 mmol TBHP, and 50 mg catalyst, 120 °C, 8 h.

After it was established that CrHMA performs better than the other catalysts, CrHMA with different Cr loadings (1, 2, 3, and 5 mol%) were prepared and their catalytic activities were evaluated. As shown in Fig. 4, changes in Cr loading under the given reaction conditions generated little variation in catalytic performance; tetralin conversion (62–65%) and selectivity to α-tetralone (80–85%) remained virtually the same within the range of experimental error. It appears that introduction of an excess amount of Cr under the given reaction conditions offers no advantage.

The conversion and product distribution as a function of reaction time are shown in Fig. 5. As the reaction time increases, conversion and α-tetralone selectivity increases continuously, but the reaction reaches a steady state in 8 h and further increase of time does not improve either conversion or selectivity. Therefore, further reactions parameters were studied with 8 h reaction time.

Effect of reaction temperature on tetralin oxidation over 2% CrHMA catalyst is shown in Fig. 6. Virtually no reaction took place below 40 °C; tetralin conversion then increased rapidly above 40 °C to 120 °C. α-Tetralone selectivity also increased concurrently and remained steady after 80 °C. Tetralol selectivity, on the other hand, decreased steadily with reaction temperature, indicating that further oxidation to α-tetralone occurs at higher temperatures. Naphthalene decreased as the tetralol concentration decreased with temperature. α-Naphthol remained relatively constant and did not appear to follow a consecutive reaction path from α-tetralone.

The effect of TBHP over tetralin mole ratio on the reaction product distribution was investigated and the results are shown in Fig. 7. Tetralin conversion increased with the amount of TBHP introduced, but leveled off above 80% of the stoichiometric amount. α-Tetralone selectivity decreased somewhat with TBHP but also leveled off above 60% of the stoichiometric amount. Conversion of α-tetralone to α-naphthol appears to be promoted with TBHP.

Leaching is a result of solvolysis of the metal–oxygen bonds, and is a problem generally associated with the use of heterogeneous metal catalysts in liquid phase oxidation. Catalyst recycling tests were thus performed over 2% CrHMA catalyst to verify its stability, and the catalytic reaction results are summarized in Table 2. As shown in Table 2, tetralin conversion dropped continuously in the 1st and 2nd recycle runs, reflecting metal leaching of Cr species from CrHMA. The same reaction conducted using the filtrate solution after a run confirmed the occurrence of metal leaching. Subsequently, we conducted catalyst pre-washing with ammonium acetate solution at room temperature; after drying and calcination, the reaction was carried out again in order to evaluate the problem of active metal ion leaching and the stability of the chromium ions in the CrHMA matrix [16]. A significant decrease in conversion after pre-washing was observed, but the decline in activity leveled off significantly in the next recycle run. As such, ammonium acetate pre-washing appears to be an effective tool to remove the less stable Cr species from the HMA support material.

It is commonly recognized that the metal–oxygen bonds in crystalline molecular sieves are more stable than those formed on silica or alumina supports. Microporous $AlPO_4$-5 has a crystalline three-dimensional network, and the framework Al atoms can be replaced by various transition metal ions, $CrAlPO_4$-5 being the most prominent material in oxidation

Table 2
Oxidation of tetralin over fresh, recycled, and ammonium acetate-washed CrHMA and $CrAlPO_4$-5 catalysts

Catalysts	Tetralin conversion (%)	Product selectivity (%)			
		Naphthalene	α-Tetralone	Tetralol	α-Naphthol
CrHMA	61.3	1.9	82.6	2.4	13.1
CrHMA (recycle 1)	53.0	1.8	79.8	6.0	12.4
CrHMA (recycle 2)	42.9	2.5	78.5	3.5	15.5
CrHMA[a] (AA-washed)	37.9	2.1	78.3	5.2	14.4
CrHMA[a] (AA-washed and recycled)	35.8	2.4	77.2	5.4	15.0
$CrAlPO_4$-5[b]	61.4	2.8	78.0	4.7	14.5
$CrAlPO_4$-5[a,b] (AA-washed)	55.4	3.7	78.0	7.3	11

Reaction conditions: 5 ml chlorobenzene, 8 mmol tetralin, 16 mmol TBHP, and 50 mg catalyst, 120 °C, 8 h. Metal content in the catalyst was 2 mol%.

[a] AA designates ammonium acetate.

[b] Reaction was carried out at 120 °C for 12 h.

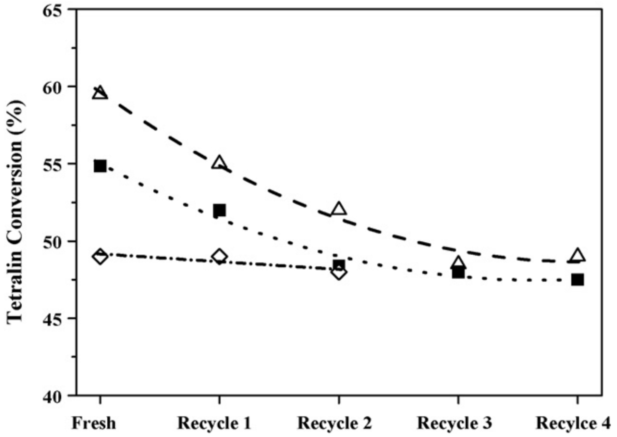

Fig. 8. Recycle test of tetralin oxidation using $CrAlPO_4$-5: 5 ml chlorobenzene, 8 mmol tetralin, 16 mmol TBHP, and 50 mg catalyst, 100 °C, 8 h; (△) $CrAlPO_4$-5 without ammonium acetate washing; (■) $CrAlPO_4$-5 washed with 1 M ammonium acetate solution; (◇) $CrAlPO_4$-5 washed with 3 M ammonium acetate solution.

[17]. The $CrAlPO_4$-5 sample prepared in this work was also washed with an ammonium acetate solution, and no color developed in the filtrate during the washing. Thus, it seems very little trace of the extra-framework Cr species was present in $CrAlPO_4$-5. Tetralin oxidation was performed using the washed $CrAlPO_4$-5 sample. As shown in Table 2, both the fresh $CrAlPO_4$-5 and ammonium acetate-washed $CrAlPO_4$-5 catalysts produced similar catalytic activity and α-tetralone selectivity, and clearly demonstrated superior Cr ions stability compared with CrHMA in amorphous walls. To investigate the washing effect of ammonium acetate on $CrAlPO_4$-5 more closely, we had conducted a catalyst recycle test at 100 °C for $CrAlPO_4$-5 catalysts washed with different concentrations of ammonium acetate. As shown in Fig. 8, washing with 3 M ammonium acetate solution produced a sufficiently stable catalytic operation for $CrAlPO_4$-5. Detailed investigation of tetralin oxidation using $CrAlPO_4$-5 to optimize reaction conditions is underway.

4. Conclusions

We reported on the preparation and characterization of Cr-, Fe-, and Co-substituted HMA, MCM-41, and $AlPO_4$-5 catalysts for the liquid phase oxidation of tetralin using TBHP as oxidant in chlorobenzene and conducted comparative catalytic studies. The chromium-substituted HMA catalyst showed the best catalytic performance among them with 65% conversion accompanied by selectivity to α-teralone greater than 80%. The catalyst suffered leaching, however, although it could be reused after some activity loss. Microporous $CrAlPO_4$-5 demonstrated somewhat lower catalytic activity than CrHMA but showed little extra-framework Cr species. It showed good reproducible catalytic performance when washed with ammonium acetate. The chromium ions are believed to be more strongly bound in a crystalline microporous structure than in mesoporous materials.

Acknowledgement

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