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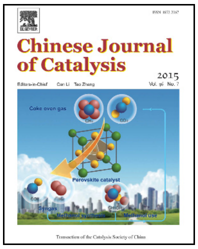

Article

KSF supported 10-molybdo-2-vanadophosphoric acid as an efficient and reusable catalyst for one-pot synthesis of 2,4,5-trisubstituted imidazole derivatives under solvent-free condition

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ABSTRACT

The one-pot three-component cyclocondensation has been developed involving the reaction of benzil with an aromatic aldehydes and ammonium acetate under thermal solvent-free conditions in the presence of a KSF supported 10-molybdo-2-vanadophosphoric acid catalyst. 10-Molybdo-2-vanadophosphoric acid was immobilized on KSF with a 20% loading, which showed the highest catalytic activity. The catalyst was fully characterized using FT-IR spectroscopy, thermal analysis, XRD and SEM analysis techniques. There are several distinct advantages to this protocol, including high yields, short reaction time, operational simplicity and a recyclable catalyst with a facile work-up procedure.

1. Introduction

The development of eco-friendly, efficient and economical methods for the synthesis of biologically interesting compounds remains one of the principle aims of synthetic chemistry [1]. The field of green chemistry has rapidly developed during the last 10 years following the recognition that environmentally friendly products and processes represent economically attractive long term synthetic strategies. The important areas of green chemistry include the development of novel chemical processes that show intrinsically high atom economies, pronounced energy savings, low levels of waste production and facile work-up procedures, as well as avoiding the use of hazardous chemicals [2].

Heteropoly acids (HPAs) have been widely used as homogeneous and heterogeneous acid catalysts for the oxidation of numerous substrates because of their unique physicochemical properties. To date, more than 100 HPAs of a variety of different compositions and structure have been reported in the literature [3]. Among them, only the Keggin-structure HPAs have been reported to play a significant role in catalysis because they are readily available and possess relatively high thermal stability [4]. Keggin type HPAs can be represented by the formula $H_{8-x}[XM_{12}O_{40}]$, where X is a hetero atom (usually P^{5+} or Si^{4+}), x is the oxidation state of the catalyst and M is its addenda atom (most frequently W^{6+} or Mo^{6+}). The M^{6+} ions can be substituted by many other metal ions, including V^{5+}, Co^{2+} and Zn^{2+} [5]. HPAs can be used either directly as a bulk material or in their supported form in both homogeneous and heterogeneous systems. HPAs are highly soluble in polar solvents, but insoluble in non-polar solvents. There are several advantages to HPAs, including their multifunctionality, structural mobility, non-corrosive nature, strong Brönsted acidity, reusability, non-toxicity and experimental simplicity [6]. However, there

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are also several disadvantages associated with the use of HPAs as catalysts, including their low surface areas and problems associated with their separation from the reaction mixture. It is envisaged that supported HPAs could be used to overcome some of the problems described above, and a number of porous supports with high surface areas such as silica, zirconia, clay, active carbon and zeolite have been reported as supports for HPAs [7].

2,4,5-Trisubstituted imidazoles has attracted considerable attention from synthetic chemists working in a variety of different field because of their wide range of biological properties, including their anti-allergic [8], antitumor [9], anti-inflammatory [10], antibacterial [11] and analgesic [12]. Compounds belonging to this structural class are also known as inhibitors of p38 MAP kinase [13], fungicides, herbicides, glucagon receptors [14], growth regulators and therapeutic agents. A variety of different procedures have been developed for the synthesis of 2,4,5-trisubstituted imidazoles, with the majority of these process involving nitriles and esters [15,16] as starting materials. The first synthesis of the 2,4,5-triphenyl imidazoles was reported by Japp and Radziszewski via the condensation of 1,2-dicarbonyl compounds with a variety of different aldehydes and ammonia [17,18]. Following on from this, numerous classical methods have been reported in the literature for the synthesis of 2,4,5-trisubstituted imidazoles [19,20]. However, there are limitations associated with some of these methods, including the requirement for expensive and toxic catalysts, long reaction time, harsh reaction conditions, low product yields and difficulties associated with the recovery and reusability of the catalysts. With this in mind, there is an urgent need for the development of clean processes utilizing eco-friendly and green catalysts, which can be readily recycled at the end of reactions.

Herein, we describe the preparation of a series of KSF-supported 10-molybdo-2-vanadophosphoric acid catalysts and their characterization by FT-IR spectroscopy, thermal analysis, XRD and SEM analysis techniques. These catalysts were subsequently used for one-pot synthesis of 2,4,5-trisubstituted imidazoles by the reaction of benzyl with a series of aromatic aldehydes and ammonium acetate under thermal, solvent-free conditions.

2. Experimental

2.1. Catalyst preparation

2.1.1. Preparation of 10-molybdo-2-vanadophosphoric acid $H_5[PMo_{10}V_2O_{40}]\cdot 30H_2O$

Sodium metavanadate (24.4 g, 0.20 mol) was dissolved in boiling water (100 mL), and the resulting solution was mixed with a solution of Na_2HPO_4 (7.1 g, 0.050 mol) in water (100 mL). The mixture was then cooled to room temperature and treated with concentrated sulfuric acid (5 mL), which caused the solution to become red in color. The mixture was then treated with a solution of $Na_2MoO_4\cdot 2H_2O$ (121 g, 0.50 mol) in water (200 mL), followed by concentrated sulfuric acid (85 mL), which was added slowly with vigorous stirring, and the resulting hot solution was then allowed to cool to room temperature. The 10-molybdo-2-vanadophosphoric acid was then extracted with diethyl ether (500 mL), and air was passed through the heteropoly etherate to remove the diethyl ether. The resulting solid was dissolved in water, and the aqueous solution was concentrated under vacuum in a desiccator over concentrated sulfuric acid until the first crystals appeared, and then allowed to crystallize further. The resulting large red crystals were collected by filtration, washed with water and air dried (yield, 30% based on molybdate) [21].

2.1.2. Preparation of KSF-supported 10-molybdo-2-vanadophosphoric acid $H_5[PMo_{10}V_2O_{40}]$ $30H_2O$

For preparation of the KSF-supported 10-molybdo-2-vandophosphoric acid catalysts, KSF montmorillonite was dried in an oven at 120 °C for 2 h prior to being used as a support. To prepare the catalyst loaded with 10, 20, 30 and 40 wt% of 10-molybdo-2-vandophosphoric acid on to the KSF supports, the appropriate amounts of 10-molybdo-2-vandophosphoric acid were dissolved in 6 mL of dry methanol. The resulting solutions were added to pre-dried KSF in a dropwise manner under constant stirring with a glass rod. For the initial addition of the 10-molybdo-2-vandophosphoric acid solutions, the clay was in its powdered form, but quickly turned into a paste following the addition of the 10-molybdo-2-vandophosphoric acid solution. However, after being stirred for 10 min, the paste turned into a free flowing powder. Following the impregnation process, all of the catalysts were dried at room temperature for 24 h. The catalysts were ground and sieved before being calcined at 200 °C for 3 h to give some uniformity to their particle size.

2.2. Characterization techniques

FT-IR spectra were obtained on a Bruker Model 3000 hyperion microscope equipped with a vertex 80 FT-IR system (Bruker, Germany). FEG-SEM images were recorded on a JSM-7600F microscope (Jeol, USA), which was operated at 30 kV. XRD patterns were obtained on a Philips X'pert MPD System (Philips, Netherlands) using Cu K_α radiation. The TG-DTA measurements for the samples were recorded on a Diamond TG-DTA Thermal Analyzer (PerkinElmer) with about 10 mg of sample in a platinum crucible at a heating rate of 10 °C/min in air. ^1H NMR spectra were recorded on a Bruker Avance 400 spectrometer (Bruker), and ^{13}C NMR spectra were recorded on a Bruker DRX-300 instrument (Bruker) using TMS as an internal reference standard. Mass spectra were recorded on a Waters UPLC-TQD mass spectrometer using electrospray ionization. The uncorrected melting points of the compounds were recorded in an open capillary tube in a paraffin bath.

2.3. General procedure for synthesis of 2,4,5-trisubstituted imidazoles using KSF supported $H_5[PMo_{10}V_2O_{40}]\cdot 30H_2O$ as a catalyst

A mixture of benzil (1 mmol), an aromatic aldehyde (1 mmol), ammonium acetate (2 mmol) and 20% $H_5PMo_{10}V_2O_{40}$/

KSF (0.1 g) was heated in the oil bath at 110 °C for the appropriate time. The progress of reaction was monitored by thin layer chromatography (TLC). Upon completion of the reaction, the mixture was cooled to room temperature and then treated with hot ethanol to separate the catalyst, which was collected from residue. The filtrate was then poured into 50 mL of ice-water to give a precipitate, which was collected by filtration, washed with ice-water and recrystallized from ethanol to give compounds **4a–4n** in high yields. The recovered catalyst was washed with ethanol (10 mL) and calcined at 140 °C for 1 h before reusing.

2.3.1. Spectral data for selected 2,4,5-trisubstituted imidazoles

2-(3,4,5-Trimethoxyphenyl)-4,5-diphenyl-1H-imidazole **(4i)**: Off-white solid; ^{1}H NMR (CDCl$_3$, 400 MHz): δ = 3.96 (s, 6H), 3.91 (s, 3H), 7.32 (d, J = 6 Hz, 2H), 7.35–7.37 (m, 8H), 7.58 (d, 2H); ^{13}C NMR: δ = 128.6, 127.8, 102.8, 56.3; IR (KBr, cm^{-1}): 3448, 1649, 1588, 1243, 731; EI-MS: m/z = 387 [M+H]$^+$.

2-(2-Furyl)-4,5-diphenyl-1H-imidazole **(4l)**: Black solid; (CDCl$_3$, 400 MHz): δ = 3.75 (m, 1H), 6.55 (m,2H), 7.03 (d, J = 10 Hz, 1H), 7.41–7.54 (m, 4H), 7.84–8.19 (m, 6H); ^{13}C NMR: δ = 145.48, 142.2, 138.8, 128.6, 127.8, 127.1, 112.0, 107.5; IR (KBr, cm^{-1}): 3366, 3057, 1641, 1618, 1126, 731; EI-MS: m/z = 287 [M+H]$^+$.

2-(3-Pyridyl)-4,5-diphenyl-1H-imidazole **(4m)**: Off-white solid; ^{1}H NMR (CDCl$_3$, 400 MHz): δ = 3.71 (m, 1H), 7.29–7.36 (m, 4H), 7.38–7.43 (m, 4H), 7.48–7.51 (m, 2H), 7.55 (d, J = 6.8 Hz, 1H), 8.36 (d, J = 8 Hz, 1H), 8.4 (d, 1H), 9.1 (s, 1H); ^{13}C NMR: δ = 148.8, 145.7, 143.0, 133.5, 128.5, 128.2, 127.9, 127.6, 126.6, 123.9; IR (KBr, cm^{-1}): 3367, 3053, 1623, 1246, 765; EI-MS: m/z = 298 [M+H]$^+$.

3. Results and discussion

3.1. Characterization of the catalysts

3.1.1. FT-IR Analysis

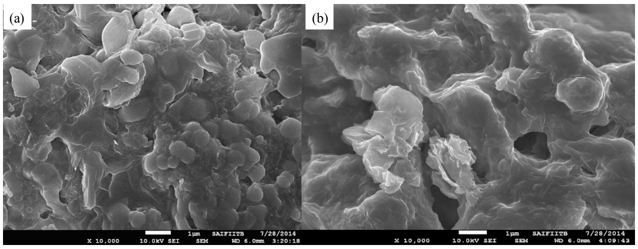


Fig. 2. FEG-SEM images of the KSF clay (a) and 20% $H_5PMo_{10}V_2O_{40}$/KSF (b).

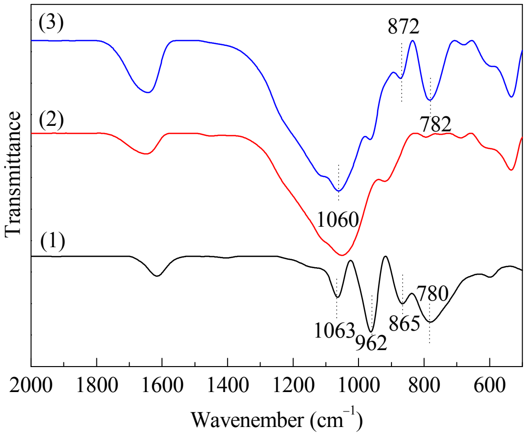


Fig. 1. FT-IR spectra of the unsupported $H_5PMo_{10}V_2O_{40}$ (1), KSF clay (2) and 20% $H_5PMo_{10}V_2O_{40}$/KSF (3).

Figure 1 shows the FT-IR spectra of the unsupported $H_5PMo_{10}V_2O_{40}$, KSF clay and 20% $H_5PMo_{10}V_2O_{40}$/KSF. The primary structure of $H_5PMo_{10}V_2O_{40}$ was identified based on the four characteristic IR bands in its FT-IR spectra in the range of 700–1200 cm^{-1}. The four characteristic IR bands of unsupported $H_5PMo_{10}V_2O_{40}$ appeared at 1063 (P–O stretching), 962 (M=O stretching), 865 (inter-octahedral M–O–M stretching) and 780 cm^{-1} (intra-octahedral M–O–M stretching). After being supported on KSF clay, some of the characteristic bands of the $H_5PMo_{10}V_2O_{40}$/KSF were observed at 1060 (P–O stretching), 872 (inter-octahedral M–O–M stretching) and 782 cm^{-1} (intra-octahedral M–O–M stretching), whereas its remaining bands merged with those of the KSF clay. This result indicated that $H_5PMo_{10}V_2O_{40}$ had been successfully immobilized on the KSF support via the formation of strong chemical interaction between the two components.

3.1.2. SEM Analysis

Figure 2 shows the surface morphologies of the synthesized catalysts. Figure 2(a) shows the surface morphology of the KSF clay, and Figure 2(b) shows the surface morphology of the 20% $H_5PMo_{10}V_2O_{40}$/KSF catalyst. These images clearly show that the surface morphology of the KSF clay was altered as a consequence of the deposition and insertion of the catalyst onto its surface.

3.1.3. Thermal analysis

Figure 3 shows the results for the thermal analysis of unsupported $H_5PMo_{10}V_2O_{40}$, KSF clay and 20% $H_5PMo_{10}V_2O_{40}$/KSF. The TGA results for $H_5PMo_{10}V_2O_{40}$ revealed a mass loss of about 7% up to a temperature of 130 °C, which was attributed to the loss of free and adsorbed water. The gradual mass loss of

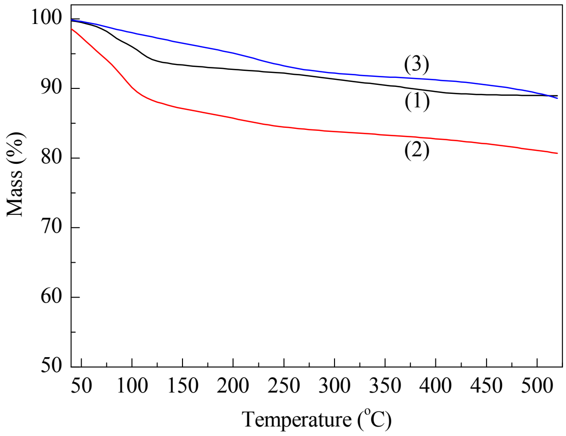


Fig. 3. Thermal analyses of unsupported $H_5PMo_{10}V_2O_{40}$ (1), KSF clay (2) and 20% $H_5PMo_{10}V_2O_{40}$/KSF (3).

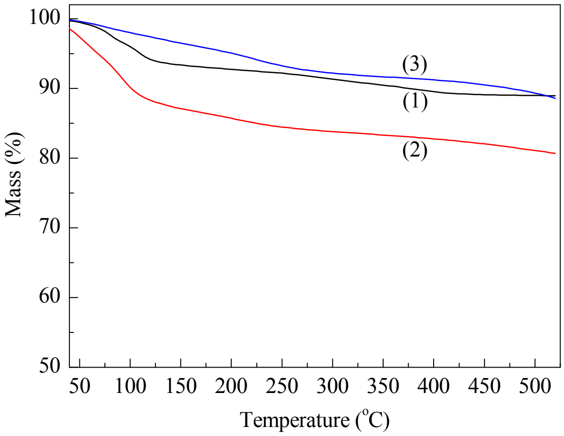


Fig. 3. Thermal analyses of unsupported $H_5PMo_{10}V_2O_{40}$ (1), KSF clay (2) and 20% $H_5PMo_{10}V_2O_{40}$/KSF (3).

about 4% up to 500 °C was attributed to the release of more hydrated or structural water. The TGA results for the KSF clay showed a steady mass loss of about 19% up to 500 °C, which was attributed to the loss of physisorbed and interlayer water, as well as dehydroxylation reactions resulting from the breaking of the structural –OH groups on the support. The TGA results for 20% $H_5PMo_{10}V_2O_{40}$/KSF showed a mass loss of about 3% up to a temperature of 140 °C, which was attributed to the loss of free and adsorbed water. A gradual mass loss of about 7% was observed up to 500 °C, which was caused by the release of more hydrated or structural water. Taken together, these results indicated that the thermal stability of $H_5PMo_{10}V_2O_{40}$ increased following its immobilization on the KSF support. This increase in the thermal stability could be attributed to the formation of intermolecular bonding interactions between KSF and $H_5PMo_{10}V_2O_{40}$.

3.1.4. XRD measurements

Figure 4 shows the XRD patterns of unsupported $H_5PMo_{10}V_2O_{40}$, KSF clay and 20% $H_5PMo_{10}V_2O_{40}$/KSF. The XRD pattern of $H_5PMo_{10}V_2O_{40}$ showed two intense peaks with 2θ values of 8.90° and 27.98°, which corresponded to basal *d*-spacing (d_{001}) values of about 0.991 and 0.318 nm. Several other peaks were also observed in the diffractogram of $H_5PMo_{10}V_2O_{40}$, which indicated that $H_5PMo_{10}V_2O_{40}$ was crystalline in nature. The XRD pattern of the KSF clay contained two intense peaks with 2θ values of 19.97° and 26.76°, which corresponded to basal *d*-spacing (d_{001}) values of about 0.444 and 0.333 nm. These data confirmed that the KSF clay was crystalline in nature. The peaks in the XRD pattern of 20% $H_5PMo_{10}V_2O_{40}$/KSF were less intense than those observed in the individual diffractograms, which indicated that the impregnation of the $H_5PMo_{10}V_2O_{40}$ on KSF was resulting in a decrease in its crystallinity.

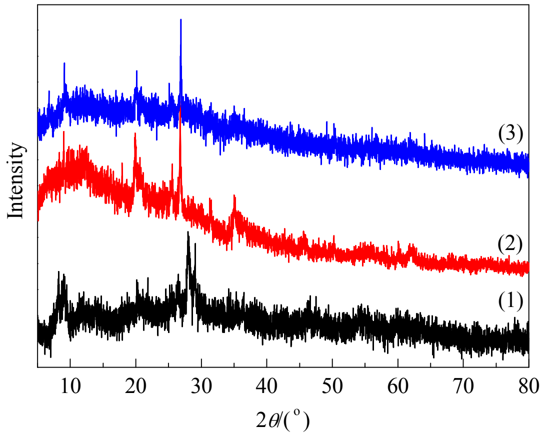


Fig. 4. XRD patterns of unsupported $H_5PMo_{10}V_2O_{40}$ (1), KSF clay (2) and 20% $H_5PMo_{10}V_2O_{40}$/KSF (3).

3.2. Catalytic activity

Various amounts of $H_5PMo_{10}V_2O_{40}$/KSF were used to study the effect of the composition of the catalyst on the conversion, and the results are summarized in Table 1. Pure KSF clay showed only moderate catalytic activity in terms of the reaction time and the yield of the desired product (Table 1, entry 2). Bulk $H_5PMo_{10}V_2O_{40}$ gave a good yield of the product but only after an extended reaction time (Table 1). In terms of optimizing the amount of catalyst immobilized on the KSF support, it was observed that 20% $H_5PMo_{10}V_2O_{40}$/KSF gave an excellent yield of the desired product over a short reaction time (Table 1, entry 4). However, increasing the loading of the $H_5PMo_{10}V_2O_{40}$ catalyst on the KSF support from 10% to 20% led to a significant increase in the yield (Table 1, entries 3 and 4). Further increases in the loading, however, resulted in a decrease in the yield of the reaction because of the leaching of the catalyst from the support. The increase in activity of the catalyst following its immobilization on the KSF support was attributed to the high dispersion of the $H_5PMo_{10}V_2O_{40}$ catalyst on the support, which would lead to an increase in its surface area and the number of active sites compared with the bulk $H_5PMo_{10}V_2O_{40}$ catalyst.

Considering the above observations we next carried out a series of reactions using benzil (1 mmol), aromatic aldehydes (1 mmol) and ammonium acetate (2 mmol) in presence of 20% $H_5PMo_{10}V_2O_{40}$/KSF (0.1 g) at 110 °C under solvent free conditions. Most importantly, aromatic aldehydes with substituent's bearing either electron-donating or electron-withdrawing groups reacted successfully in the presence of 20% $H_5PMo_{10}V_2O_{40}$/KSF as a catalyst. In all cases the expected

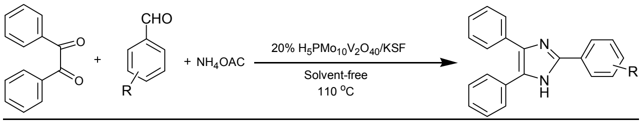
Table 1
Effect of various $H_5[PMo_{10}V_2O_{40}]$ loadings of the efficiency of the catalyst.

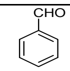
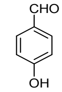
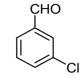
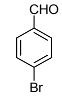
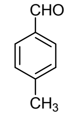
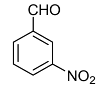
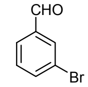
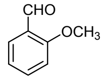
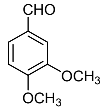
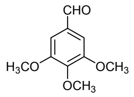
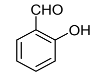
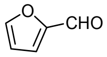
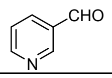
Entry	Catalyst	T/(°C)	Time (min)	Isolated yield (%)
1	$H_5[PMo_{10}V_2O_{40}]$ $30H_2O$	110	45	85
2	KSF	110	75	71
3	10% $H_5PMo_{10}V_2O_{40}$ / KSF	110	55	78
4	20% $H_5PMo_{10}V_2O_{40}$ / KSF	110	25	92
5	30% $H_5PMo_{10}V_2O_{40}$ / KSF	110	30	90
6	40% $H_5PMo_{10}V_2O_{40}$ / KSF	110	32	89

Reaction conditions: benzil (1 mmol), benzaldehyde (1mmol), ammonium acetate (2 mmol) and KSF supported $H_5[PMo_{10}V_2O_{40}]\cdot 30H_2O$ (0.1 g) under solvent free conditions.

Table 2

Synthesis of 2,4,5-trisubstituted imidazoles using 20% $H_5PMo_{10}V_2O_{40}$/KSF as a catalyst



Entry	Aldehyde	Product	Time (min)	Isolated yield (%)	Melting point (°C) Found	Melting point (°C) Reported [Ref.]
1		**4a**	25	92	273–275	274–276 [22]
2		**4b**	35	95	241–243	240–242 [23]
3		**4c**	30	78	283–285	285–287 [30]
4		**4d**	25	90	250–252	252–254 [24]
5		**4e**	40	94	232–234	230–232 [25]
6		**4f**	35	96	302–304	>300 [25]
7		**4g**	40	83	298–300	301–303 [26]
8		**4h**	50	89	203–205	204–206 [28]
9		**4i**	40	95	219–221	220–221 [27]
10		**4j**	45	92	260–262	—
11		**4k**	25	84	210–212	209–211 [28]
12		**4l**	33	88	202–204	199–201 [27]
13		**4m**	35	94	230–232	231–233 [29]

Reaction conditions: benzil (1 mmol), aromatic aldehyde **4a**–**4m** (1 mmol), ammonium acetate (2 mmol) and 20% $H_5PMo_{10}V_2O_{40}$/KSF (0.1 g) at 110 °C under solvent-free conditions.

products were obtained in good yields. The results are shown in Table 2.

3.3. *Recyclability of the catalyst*

The catalyst prepared in the current study could be readily recovered from the reaction mixture upon completion of the reaction by a simple filtration process. The recovered catalyst could then be washed several times with ethanol before being calcined at 140 °C and reused for a fresh reaction mixture. The results of a recyclability study revealed that the catalyst could be reused at least three times with only a slight reduction in its original activity (e.g., 90% yield for the first run, 87% yield for the second run and 83% yield for the third run). This decrease in catalytic activity was attributed to the leaching of the catalyst from the support.

4. Conclusions

A simple and convenient procedure has been developed for the one-pot three-component cyclocondensation of benzyl with a variety of aromatic aldehydes and ammonium acetate in the presence of KSF-supported 10-molybdo-2-vanadophosphoric acid catalyst under thermal, solvent-free conditions. These reactions were carried out under solvent-free conditions over short reaction time and gave the corresponding products in good to excellent yields. The 20% $H_5PMo_{10}V_2O_{40}$ supported on KSF clay exhibited higher catalytic activity than the bulk $H_5PMo_{10}V_2O_{40}$ catalyst, as well as the (10% and 30%) $H_5PMo_{10}V_2O_{40}$ catalysts supported on KSF clay. The observed increase in the catalytic activity of the KSF-supported catalysts was attributed to the high surface area and large number of Brönsted acidic sites on the catalyst. Moreover, this newly developed catalyst was thermally stable, green and inexpensive to prepare, as well as being easy to separate from the reaction mixture and reused several times.

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References

[1] Sheldon R A. *Green Chem*, 2007, 9: 1273

[2] Tang S Y, Bourne R A, Smith R L, Poliakoff M. *Green Chem*, 2008, 10: 268

[3] Kozhevnikov I V. *Russ Chem Rev*, 1993, 62: 473

[4] Dias J A, Osegovic J P, Drago R S. *J Catal*, 1999, 183: 83

[5] Kozhevnikov I V. Catalysis for Fine Chemical Synthesis, Vol. 2. Chichester: Wiley, 2002

[6] Kozhevnikov I V. *Appl Catal A*, 2003, 256: 3

[7] Chimienti M E, Pizzio L R, Caceres C V, Blanco M N. *Appl Catal A*, 2001, 208: 7

[8] Shelke K F, Sapkal S B, Sonar S S, Madje B R, Shingte B B, Shingare M S. *Bull Korean Chem Soc*, 2009, 30: 1057

[9] Wang L, Woods K W, Li Q, Barr K J, McCroskey R W, Hannick S M, Gherke L, Credo R B, Hui Y H, Marsh K, Warner R, Lee J Y, Zielinsky-Mozng N, Frost D, Rosenberg S H, Sham H L. *J Med Chem*, 2002, 45: 1697

[10] Mohammadi A, Keshvari H, Sandaroos R, Rouhi H, Sepehr Z. *J Chem Sci*, 2012, 124: 717

[11] Joshi R S, Mandhane P G, Shaikh M U, Kale R P, Gill C H. *Chin Chem*

Graphical Abstract

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KSF supported 10-molybdo-2-vanadophosphoric acid as an efficient and reusable catalyst for the one-pot synthesis of 2,4,5-trisubstituted imidazole derivatives under solvent-free condition

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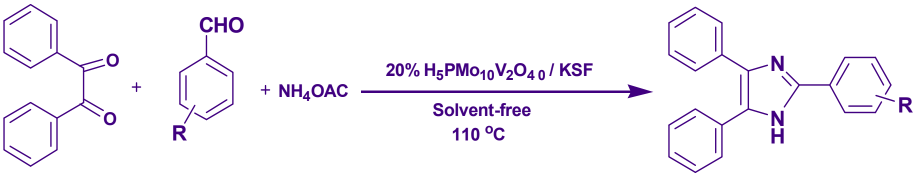

Twenty percent 10-molybdo-2-vanadophosphoric acid supported on KSF clay acts as an efficient, green and reusable solid acid catalyst for one-pot synthesis of 2,4,5-trisubstituted imidazole derivatives.

Lett, 2010, 21: 429
[12] Ucucu U, Karaburun N G, Isikdag I. *Farmaco*, 2001, 56: 285
[13] Ashrafi M, Davoodnia A, Tavakoli-Hoseini N. *Bull Korean Chem Soc*, 2013, 34: 1508
[14] De Laszlo S E, Hacker C, Li B, Kim D, MacCoss M, Mantlo N, Pivnichny J V, Colwell L, Koch G E, Cascieri M A, Hagmann W K. *Bioorg Med Chem Lett*, 1999, 9: 641
[15] Gadekar L S, Mane S R, Katkar S S, Arbad B R, Lande M K. *Cent Eur J Chem*, 2009, 7: 550
[16] Balalaie S, Arabanian A, Hashtroudi M S. *Monatsh Fur Chem*, 2000, 131: 945
[17] Radziszewski B. *Chem Ber*, 1882, 15: 1493
[18] Japp F, Robinson H. *Chem Ber*, 1882, 15: 1268
[19] Sharma G V M, Jyothi Y, Lakshmi P S. *Synth Commun*, 2006, 36: 2991
[20] Ahmad S, Abbas R, Elham F, Zahra B. *Catal Commun*, 2007, 8: 1149
[21] Shojaei A F, Rezvani M A, Baei M S. *World Appl Sci J*, 2010, 11: 727
[22] Karimi A R, Alimohammadi Z, Amini M M. *Mol Divers*, 2010, 14: 635
[23] Wang L M, Wang Y H, Tian H, Yao Y F, Shao J H, Liu B. *J Fluorine Chem*, 2006, 127: 1570
[24] Mohammadi A A, Mivechi M, Kefayati H. *Monatsh Chem*, 2008, 139: 935
[25] Samai S, Nandi G C, Singh P, Singh M S. *Tetrahedron*, 2009, 65: 10155
[26] Chuanming Y, Min L, Weike S, Yuan Y X. *Synth Commun*, 2007, 37: 3304
[27] Sangshetti J N, Kokare N D, Kotharkar S A, Shinde D B. *Monatsh Chem*, 2008, 139: 125
[28] Samai S, Nandi G C, Singh P, Singh M S. *Tetrahedron Lett*, 2009, 65: 10155
[29] Sangshetti J N, Kokare N D, Kotharkara S A, Shinde D B. *J Chem Sci*, 2008, 120: 463
[30] Shaabani A, Rahmati A, Farhangi E, Badri Z. *Catal Commun*, 2007, 8: 1149