

Short communication

# Catalytic performance of Preyssler heteropolyacid as a green and recyclable catalyst in oxidation of primary aromatic amines

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Received 12 February 2006; received in revised form 29 March 2006; accepted 29 March 2006

Available online 18 May 2006

## Abstract

Many substituted anilines were converted into the corresponding azoxy benzenes as major products, by hydrogen peroxide, using Preyssler catalyst.

The ability of various Preyssler catalysts as pure,  $H_{14}[NaP_5W_{30}O_{110}]$ , mixed addenda,  $H_{14}[NaP_5W_{29}MoO_{110}]$ , and cetylpyridinium acidic salt, was investigated. In all cases, the highest yield of azoxy compounds was observed, using  $H_{14}[NaP_5W_{30}O_{110}]$  as catalyst. The oxidations of anilines with 30%  $H_2O_2$  catalyzed by Preyssler catalysts in various solvents with different molar ratio of amine to oxidant, have been investigated and the results showed that the reaction yields were affected by changing the solvent as well as molar ratio of amine:oxidant.

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**Keywords:** Heteropolyacid; Catalyst; Preyssler; Primary aromatic amine; Azoxy compounds

## 1. Introduction

The oxidation of amines is a fundamental reaction for the synthesis of *o*-containing amine derivatives and both industry and academia have paid considerable attention to them. Therefore, a variety of oxidation and catalytic methods have been investigated [1–11]. In the most reactions mixed *o*-containing amines have been obtained, so it is very difficult to control the selectivity in such reactions. It has been reported the product composition can be controlled by the oxidant, catalyst, and reaction conditions, employed [7,10–13].

Among the various reported methods, for the conversion of amines into oxygen containing compounds formation of, nitro and nitroso compounds are common and has been most extensively studied while, a limited number of methods have been reported for the preparation of azoxy compounds.

The azoxy compounds are of interest because of their physiological activity, uses in liquid crystal displays and therapeutic medicines [6,14].

Sonawane et al. reported the use of Ts-1 catalyst for the selective oxidation of aniline to azoxy benzene and Sandler and co-workers have investigated the preparation of mixed azoxy compounds [15,16]. Melnikov et al. reported the oxidation of many primary aromatic amines to azoxy compounds, catalyzed by sodium tungstate in the presence of  $H_3PO_4$  [17].

Literature survey shows that the most of the reported methodologies involves, using metal catalysts. The majority of such metal catalysts are usually, expensive, of limited environmental compatibility, and even toxic. The current trend towards a green and sustainable chemistry has given life to an increasing number of new strategies for green catalysts, with the properties being easily recovered, re-used and being disposed at low costs [18].

The heteropolyacids (HPAs) could be a materials of choice for these oxidation reactions in view of their specific basic properties as underlined and their potential usefulness as catalysts [19,20]. There are several large-scale industrial processes utilizing HPA catalysts as oxidant and acid catalysts [21–24].

Until now, most of the research concerning catalytic properties of HPAs has been carried out using, Keggin structure and its derivatives as defect, mixed addenda, supported, etc. In the recent years, the interest in other HPAs, has been growing in the literature.

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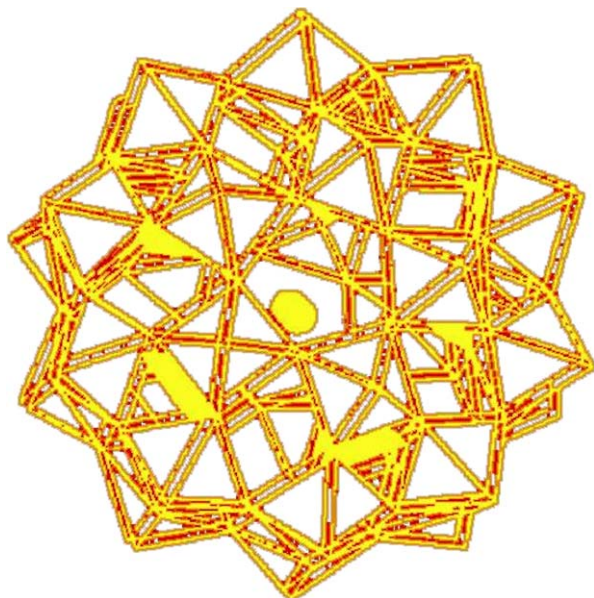


Fig. 1. Structure of Preyssler catalyst.

As a part of a research project to develop environmentally friendly catalysts, we have recently applied the Preyssler HPA catalyst to various reactions [25–27]. However, the capability of this catalyst still has been largely overlooked and there is still rooms to manoeuvre [28].

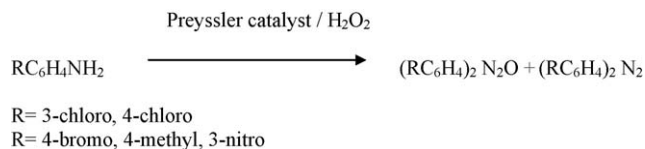
In our opinion, advantages such as: retention of structure in the presence of hydrogen peroxide, high hydrolytic stability (pH 0–12), high thermal stability, and having a large number of acidic protons (14) along with exclusive structure for Preyssler's anion are outstanding and make it a good candidate for further studying. This polyanion consists of a cyclic assembly of five  $PW_6O_{22}$  units; each derived from the Keggin anion,  $[PW_{12}O_{40}]^{3-}$ , by the removal of two sets of three corner shared  $WO_6$  octahedra [29]. The structure of this polyanion is shown in Fig. 1.

In our previous paper, we reported the oxidation of tertiary amines with aqueous hydrogen peroxide under the influence of Preyssler catalyst [30].

Due to importance of azoxy compounds in industry and pharmacy, and because of the limited number of methods on the synthesis of azoxy compounds and with the aim of extending of applications of Preyssler catalyst, in this paper we wish to report our results on the oxidation of amines for the synthesis of azoxy compounds as a major product in the presence of Preyssler catalyst. We have found that in these oxidations, the Preyssler catalyst in an organic solvent render effective oxidation of primary aromatic amines to azoxy compounds, in the presence of hydrogen peroxide in good yields.

## 2. Results and discussion

The oxidation of 4-chloro, 3-chloro, 3-nitro, 4-bromo and 4-methyl aniline were carried out with 30% hydrogen peroxide as the oxidant and Preyssler catalyst, as pure,  $H_{14}-P_5$ , mixed addenda,  $H_{14}-P_5Mo$  and cetylpyridinium salt,  $CPW-P_5$ . The azoxy compounds were major and azo compounds were minor products. Our study has been focused on azoxy compounds as



Scheme 1.

the major product. Surprisingly, aniline did not give any satisfactory results (Scheme 1).

The effects of various parameters such as solvent, mole number of oxidant, catalyst type, temperature and time of reactions were studied.

In the first stage, many oxidation reactions in various solvents with low boiling points such as ethanol, acetonitrile and methanol, and high boiling points solvents such as dimethylformamide have been performed.

We have found that the right chose of the solvent has an important role on the yields of azoxy products. Dimethylformamide was found to be the solvent of choice for the oxidation of 4-chloro, 3-chloro and 3-nitro aniline and for 4-bromo and 4-methyl aniline the best solvent was ethanol and acetonitrile, respectively.

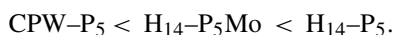
The influence of mole ratio of oxidant to substrate on the yields in the best solvent with Preyssler catalysts was investigated. The molar ratio of substrate:oxidant was varied as 1:1 to 1:8. The best molar ratio was found to be 1:7.

On the basis of these results, amines were oxidized with 30%  $H_2O_2$  along with molar ratio of 1:7 in the solvent of choice. The results are summarized in Table 1.

## 3. Effect of the catalyst type

In our studies, we investigated the activity of various Preyssler forms as pure,  $H_{14}-P_5$ , mixed addenda,  $H_{14}-P_5Mo$ , and cetylpyridinium salt,  $CPW-P_5$ . Representative results in Table 1 are shown. The primary aromatic amines were oxidized to the azoxy compounds in satisfactory yields along with a small amount of azo compounds.

The results indicate that the nature of the catalyst plays an important role on their catalytic activities. The highest yield of products has been achieved in the presence of  $H_{14}-P_5$  as catalyst, and  $CPW-P_5$  gave lowest yields. The yield of obtained products increased in the following order:



Due to the complicated nature of the reaction, obtaining the variety of products, and two-phase conditions used for the reactions, it seemed rather difficult to make an exact assessment of the catalyst role. However, we may make some assumptions that agree with the experimental data and literature.

Comparison between  $H_{14}-P_5$  and  $CPW-P_5$  shows that the number of protons as well as steric hindrance are important factors. In an aqueous solution, HPAs are strong fully dissociated acids [31].

Additionally, the nature of counter cation in HPA salts is critical with respect to their acidity. Salts with rather small cations resemble the parent HPAs; they are readily soluble in water, and

Table 1  
Oxidation of primary aromatic amines with H<sub>2</sub>O<sub>2</sub> catalyzed by Preyssler catalyst in solvent of choice<sup>a</sup>

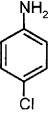
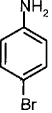
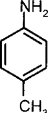
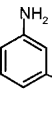
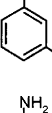
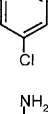
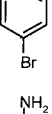
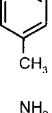
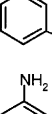
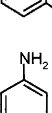
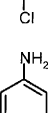
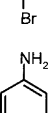



Run	Substrate	Solvent	Catalyst	% Azoxy	% Azo
1		<i>N,N</i> -Dimethylformamide	H <sub>14</sub> -P <sub>5</sub>	52.16	31.582
2		Ethanol	H <sub>14</sub> -P <sub>5</sub>	68.205	28.321
3		Acetonitrile	H <sub>14</sub> -P <sub>5</sub>	61.859	15.203
4		<i>N,N</i> -Dimethylformamide	H <sub>14</sub> -P <sub>5</sub>	82.273	16.826
5		<i>N,N</i> -Dimethylformamide	H <sub>14</sub> -P <sub>5</sub>	87.305	12.695
6		<i>N,N</i> -Dimethylformamide	H <sub>14</sub> -P <sub>5</sub> Mo	41.9	25.37
7		Ethanol	H <sub>14</sub> -P <sub>5</sub> Mo	42	17.4
8		Acetonitrile	H <sub>14</sub> -P <sub>5</sub> Mo	50.8	12.48
9		<i>N,N</i> -Dimethylformamide	H <sub>14</sub> -P <sub>5</sub> Mo	43.66	8.93
10		<i>N,N</i> -Dimethylformamide	H <sub>14</sub> -P <sub>5</sub> Mo	63	9.16
11		<i>N,N</i> -Dimethylformamide	CPW-P <sub>5</sub>	40.91	24.77
12		Ethanol	CPW-P <sub>5</sub>	31.1	12.9
13		Acetonitrile	CPW-P <sub>5</sub>	49.6	12.2

Table 1 (Continued)

Run	Substrate	Solvent	Catalyst	% Azoxy	% Azo
14		<i>N,N</i> -Dimethylformamide	CPW-P <sub>5</sub>	20.57	4.21
15		<i>N,N</i> -Dimethylformamide	CPW-P <sub>5</sub>	48.3	7.02

<sup>a</sup> Reaction conditions: solvent (5 ml), amine: H<sub>2</sub>O<sub>2</sub>, 1:7, catalyst (1.6 × 10<sup>-3</sup> mmol), reflux 4 h. Yields were determined by GC-mass.

non porous. In contrast, water insoluble salts with large monovalent cations, such as CPW, have a rigid microporous/mesoporous structure. Even if these HPAs are prepared by precipitation from aqueous solutions to be stoichiometric, residual quantities of protons still remain, which are apparently responsible for the catalytic activities of these salts [32].

According to the earlier studies for thermogravimetric analysis, potentiometric and conduction titrations, Preyssler's anion with large counter cations such as tetrahexyl ammonium bromide and tetraheptyl ammonium bromide, can not replace more than six or seven protons [33].

In this study for CPW-P<sub>5</sub>, the results showed that about six protons can be replaced. The large anion of Preyssler provided many "sites" on the oval-shaped molecule that are likely render the catalyst effectiveness.

We suggest, the rigidity, steric hindrance and lower number of protons in CPW-P<sub>5</sub> is tentatively responsible for lower activity of CPW-P<sub>5</sub>. The larger number of protons may lower the activation barrier to the oxidation reaction.

Between H<sub>14</sub>-P<sub>5</sub> and H<sub>14</sub>-P<sub>5</sub>Mo, the lower yield is observed for H<sub>14</sub>-P<sub>5</sub>Mo. As mentioned earlier, it seems, by replacing tungsten with molybdenum in Preyssler catalyst, because of decrease in symmetry and distortion, acidity is decreased [25a]. In addition, usually tungsten HPAs are the catalysts with stronger acidity [31]. Generally, if the reaction rate is controlled by the catalyst acid strength, tungsten HPAs show the highest catalytic activity.

With respect to the higher oxidation potential of molybdenum HPAs compared to tungsten HPAs, and observed the highest yields for H<sub>14</sub>-P<sub>5</sub>, it seems reaction yields are controlled and determined by acidity of catalyst.

The acid strength and catalytic activity of Bronsted acids are best quantified in terms of their some characteristics, namely the hardness of the acid or the softness of the corresponding base. These are the key parameters of the hard and soft acid–base theory related to the polarizabilities of the acids and bases can be generally applied to Lewis acids and bases [34].

The primary aromatic amines such as substituted anilines are moderate bases and can be reacted with hard and soft acids. In this study, an acid–base interaction between catalyst and substrate is suggested and the catalyst structure assumed to play an important role in stabilizing organic intermediates.

The radical products from the decomposition of hydrogen peroxide (HO• and HO<sub>2</sub>•) act as the amine oxidants in the presence of Preyssler's anion at 70 °C [35–37].

#### 4. Effects of the mole numbers of oxidant

Various oxygen-donors such as *t*-BuOOH and H<sub>2</sub>O<sub>2</sub> were examined for oxidation of primary aromatic amines under various reaction conditions. Higher activity was observed with hydrogen peroxide. Hydrogen peroxide is also preferable because of safety and ease of handling. The oxidation of amines was investigated with various molar ratios of substrate:oxidant (1:1 to 1:8) under optimum conditions. The results are shown in Fig. 2 for the catalyst of choice, H<sub>14</sub>-P<sub>5</sub>. Our studies show that, the reaction yield was affected by changing the molar ratio of amine:oxidant. The yield increases with that of the molar ratio up to 1:7 and any further increasing of the molar ratio does not have any appreciable effect on the yield.

In the absence of Preyssler catalyst, H<sub>2</sub>O<sub>2</sub> has a very poor ability to oxidize primary aromatic amines.

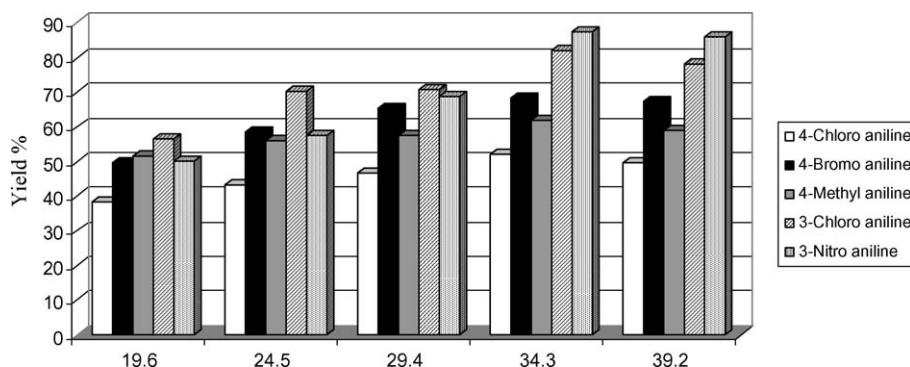


Fig. 2. Dependence of azoxy yields with mole numbers of hydrogen peroxide.

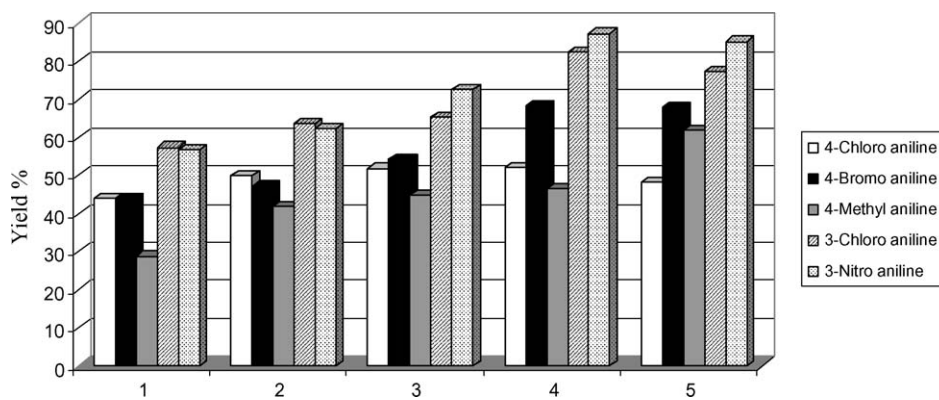


Fig. 3. Dependence of azoxy yields with time of reaction (h).

### 5. Effect of the reaction time

In order to obtain the best reaction time, the effect of reaction time on the % yield of major product, azoxy, was studied. The results (in optimum conditions) for H<sub>14</sub>–P<sub>5</sub> (best catalyst) are shown in Fig. 3. The results show that there is an increase in the percentage of produced azoxy with increase in reaction time. This behavior was quiet general for all of the used catalysts. The optimum reaction time has been found to be 4 h in reflux condition.

### 6. Effect of the reaction temperature

All of the reactions, in various solvents and with different moles of oxidant in the presence of Preyssler catalysts were carried out at two temperatures including room temperature (lowest temperature) and reflux temperature (highest temperature). The highest yield of azoxy compounds was obtained at reflux temperature and the yields are lower when the reactions were carried out at room temperature or between.

### 7. Recovery of the Preyssler catalyst

In order to compare the stability of Preyssler catalyst structure in the presence of H<sub>2</sub>O<sub>2</sub>, with respect to that of Keggin HPAs, separation of the Preyssler catalyst was made after the catalytic reaction. Interestingly, in contrary of Keggin HPAs, Preyssler's anion catalyzes oxidations of primary aromatic amines by H<sub>2</sub>O<sub>2</sub> without any degradation of structure.

It is well known that Keggin HPAs at 60–70 °C with the use of H<sub>2</sub>O<sub>2</sub> have degraded to peroxo polyoxometalates and the reaction takes place via the oxygen atom transfer from the peroxo polyoxometalate to the substrate [38,39]. Our studies have also shown that the Preyssler catalyst can catalyze oxidations of amines with retention of structure, which is very important in catalytic processes, specially, in industry.

This behavior is accordance to earlier works [25a,30]. Fig. 4 shows the IR spectrum of Preyssler's anion before (a) and after (b) catalytic reaction. Several times recovery has decreased the catalytic activity only 3–5%.

### 8. Conclusion

In conclusion, we have shown that the oxidation of anilines with H<sub>2</sub>O<sub>2</sub> catalyzed by Preyssler catalyst provides a simple, clean, and general procedure for the preparation of azoxy compounds as major products that are difficult to prepare by the conventional methods.

In comparison with other reported oxidative systems, and Keggin catalyzed reactions in the presence of hydrogen peroxide, performance of the Preyssler catalyst can be described as outstanding.

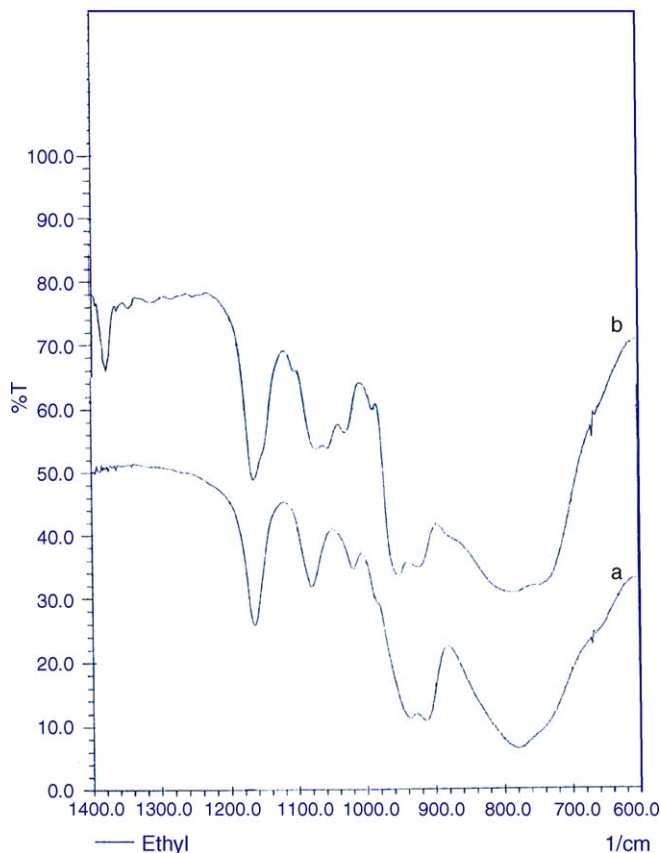


Fig. 4. IR spectrum of Preyssler catalyst before (a) and after (b) the catalytic reaction.

## 9. Experimental section

### 9.1. Materials

All amines, solvents and reagents were purchased from commercial sources. Hydrogen peroxide was obtained from Merck Company and was standardized by well-known methods (Hydrogen Peroxide Product Information Manual, Analytical Procedure). H<sub>14</sub>-P<sub>5</sub> and H<sub>14</sub>-P<sub>5</sub>Mo were prepared according to our earlier works [25].

For preparation of CPW-P<sub>5</sub>, to a solution of H<sub>14</sub>-P<sub>5</sub> was added dropwise CPC in H<sub>2</sub>O with stoichiometric ratio of 1:14, and the mixture was stirred at room temperature for 2 h. The resulting white precipitate was filtered and then washed several times with distilled water and dried in vacuo to give cetylpyridinium acidic salt of Preyssler.

### 9.2. Instruments

IR spectra were obtained with a Bruker 500 scientific spectrometer. GC-Mass analysis was performed on a GC-Mass model: 5973 network mass selective detector, GC 6890 Agilent. Mass spectra were obtained with a Massens POEKTROMETER CH-7A VARIN MAT BREMEN spectrometer. <sup>1</sup>H NMR spectra were recorded on a FT-NMR Bruker 100 MHz Aspect 3000 spectrometer.

### 9.3. Oxidation of primary aromatic amines: general procedure

To a stirred solution of catalyst ( $1.6 \times 10^{-3}$  mmol) in appropriate solvent (Table 1) was added the aromatic amine and hydrogen peroxide with molar ratio of 1:7 and the mixture was stirred at room or reflux temperature for 7 and 4 h, respectively. The solvent was evaporated to dryness under reduced pressure. The residue was extracted with dichloromethane, dried over sodium sulfate, and evaporated under reduced pressure. The product was purified by column chromatography on silica gel. The melting points and spectral data of each product were compared with those of authentic samples and the literature values. Yields were determined by GC-mass.

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