Catal Lett (2008) 123:129–134
DOI 10.1007/s10562-008-9405-9

One-step Preparation of *N*-tosylimines Using Zeolite Catalysts

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Received: 6 December 2007 / Accepted: 13 January 2008 / Published online: 13 February 2008
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Abstract *N*-tosylimines have been successfully synthesized for the first time by condensations of *N*-tosylamine with aldehydes catalyzed by HY3, HY6, HY9 and HZSM-5 zeolites. HY9 was the best catalyst, leading to the highest yield of *N*-tosylimines, indicating that the reaction efficiency depends upon the concentration of the Bronsted acid sites and its hydrophobicity.

Keywords Zeolites · *N*-tosylimines · *N*-tosylamine · Aldehydes · Condensation

1 Introduction

N-Tosylimines are versatile synthetic intermediates in various organic reactions [1–3]. They are excellent substrates in aza-Diels-Alder reactions [4–8], nucleophilic additions [9–19], reduction [20–22], and radical [23, 24] or aza-Morita-Baylis-Hillman [25–29] reactions. They were used in the synthesis of aziridine derivatives [30–33]. The sulfonamides formed can be converted to amines or amino acids [16, 19].

Traditionally, *N*-tosylimines were prepared by two-step reactions. Several useful methods had been reported in the literature [34–44]. These included palladium catalyzed isomerization of *N*-tosyl aziridines [34]; oxidation of chloramine-T in the presence of tellurium metal [35]; in situ rearrangement of oxime *O*-sulfinates [36]; the in situ generation of *N*-tosylimines by the reaction of aldehydes and *N*-sulfinyl *p*-toluenesulfonamide [37]; reaction of *N*-trimethylsilylaldimine with sulfonyl chlorides [38]; condensations of racemic sulfinylamides with aldehydes or ketones and oxidation with 3-Chloroperoxybenzoic acid [39] and the synthesis of *N*-tosylimines using titanium tetrachloride in the presence of triethylamine [40]. Chemla and his coworkers [41] reported a two-step synthesis of *N*-sulfonylimines by the reaction of aldehyde, sulfonamide and sodium *p*-toluenesulfinate in aqueous formic acid followed by the treatment of generated sulfonamide sulfone intermediate with sodium bicarbonate.

Recently, several methods have been developed for the synthesis of *N*-tosylimines, such as dehydration by the use of molecular sieves and Amberlyst under Dean-Stark conditions [42], or the use of tetraethyl orthosilicate [43] or trifluoroacetic anhydride as dehydrating agents [44]. However, all these methods involved expensive reagents, long reaction time and harmful catalysts.

The application of solid acids and bases (natural and modified clay minerals, montmorillonites, zeolites, mixed oxides and layered double hydroxides) as efficient catalysts in organic synthesis has been widely studied [45]. For the environmental protection, this method is important because the zeolites produce less waste. Zeolites also have excellent activity and selectivity even in industrial scales. Moreover, in most cases these substances can be recovered from reaction mixtures and reused without significant loss of activity.

Earlier we have found that Friedal-Crafts acylation of anisole with substituted aromatic acids and alkanoic acids can be successfully performed over zeolite catalysts [46]. The acid zeolite can regioselectively catalyze the acylation of anisole with carboxylic acid [47]. Furthermore, acid

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Electronic supplementary material The online version of this article (doi:10.1007/s10562-008-9405-9) contains supplementary material, which is available to authorized users.

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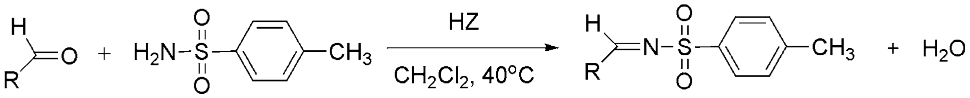


Scheme 1 The condensation reaction of *N*-tosylamine with various aldehydes over acid zeolite gave *N*-tosylimine as the major product

zeolite can be used as excellent catalyst with high yield for esterification such as the preparation of acetates, benzoates, and phthalates [48]. The Knoevenagel condensation of substituted benzaldehydes with active methylene compounds can be performed efficiently by using USY (Ultra Stable Y) zeolite as heterogeneous catalyst [49].

In continuation of our ongoing interest, we herein report the first example of the condensations of *N*-tosylamine with aldehydes using a zeolite catalyst (Scheme 1). This method provided an easy access to the *N*-tosylimines, using an ecofriendly zeolite catalyst, and is better than the methods reported earlier in the literature [34–44]. The advantages of this method over the existing synthetic routes will be discussed in this article.

2 Experimental

All chemical reagents and solvents were analytical grade and used without further purification. Melting points were measured by optical microscopic instrument of melting point and are uncorrected. ^{1}HNMR spectra were recorded on BRUKER AVANCE 300 MHz or BRUKER AVANCE 400 MHz spectrometer using tetramethylsilane as an internal standard ($CDCl_3$ solution).

2.1 Catalyst Preparation and Characterization

Samples of Y zeolites were prepared from a NaY commercial zeolite (Si/AI = 2.5, Oil Corporation of Qi Lu, China) through NH_4^+ exchange followed by calcination at 550 °C for 3 h. The samples were exchanged again and then ‘self-steamed’ for 3 h at 550, 760 and 810 °C, respectively. The framework Si/A1 ratio of the obtained samples, HY3, HY6 and HY9, determined from their unit-cell parameters by using Breck’s equation [50], were found to be 2.9, 5.7 and 9.2, while their bulk Si/A1 ratios were the same as the NaY zeolite (2.5). According to the Mobil procedure [51], HZSM-5 was prepared by treating four times the starting material NaZSM-5 (synthesized in our laboratory, Si/AI = 30) with a 1 M solution of nitric acid four times, using a liquid to solid weight ratio of 10:1 at 90 °C for 1 h, followed by drying at 110 °C for 5 h and ulterior calcinations at 520 °C for 3 h. But the final calcinations were carefully performed in absence of water in order to avoid framework dealumination. All of the catalysts were conserved in a vacuum desiccator for use.

Pyridine adsorption spectra were recorded in a Nicolet FT-IR 5DX spectrophotometer equipped with a high temperature vacuum chamber. The zeolite was taken in the sample holder and dehydrated at 400 °C for 10 h under vacuum (10^{-5} mbar). The spectrum was recorded after cooling the sample to room temperature. Then, pyridine vapor was passed into the sample holder and adsorbed onto the zeolite for a period of 30 min. The physically adsorbed pyridine was removed by heating the sample at 150 °C under vacuum (10^{-5} mbar) for 1 h. The material was cooled to room temperature, and the infrared spectrum was recorded. The concentration of the Bronsted and Lewis acid sites was calculated from the integrated area of the PyH^+ and PyL bands (at 1,540 and 1,450 cm^{-1}, respectively), using the values of the molar extinction coefficients of these bands (1.13 and 1.28 cm $mmol^{-1}$, respectively) [52, 53].

2.2 Typical Procedure for the Synthesis of *N*-tosylimine

A mixture of *N*-tosylamine (3.5 mmol), 4-chlorobenzaldehyde (3.8 mmol) and HY9 catalyst (180 mg) in dichloromethane (3 mL) was stirred and heated to reflux (40 °C) for 18 h (Dean-Stark traps filled with 4 Å molecular sieve). At the end of the reaction as indicated by thin layer chromatography, the mixture was filtered to remove the catalyst, washed three times with dichloromethane, and the filtrate was dried using a rotary evaporator, then crystallized from dichloromethane/hexane. The desired *N*-(4-Chlorobenzylidene)-*p*-tolylsulfonamide was obtained in 98% yield as a white solid, m.p. 166–168 °C; ^{1}HNMR: δ (300 MHz, $CDCl_3$), 2.46 (3H, s, $\mathbf{CH_3}C_6H_4$), 7.34–7.37 (2H, d, $CH_3C_6\mathbf{H_4}$), 7.46–7.48 (2H, d, ClC_6H_4), 7.85–7.90 (4H, m, $ClC_6\mathbf{H_4}$, $CH_3C_6\mathbf{H_4}$), 8.98 (1H, s, **H**C=N).

3 Results and Discussion

3.1 Catalyst Characterization

The physical–chemical properties of the zeolites are given in Table 1. It is well known that steam treatment has a marked effect on both stability and the crystallinity of the HY zeolites. The unit cell size and crystallinity decrease with increase in steaming temperature due to the presence

Table 1 Characteristics of the zeolite catalysts

Catalyst	Crystallinity[a] (%)	Unit cell size (nm)	Bronsted acid sites (mmol g^{-1})	Lewis acid sites (mmol g^{-1})	Frame work Si/Al[c]
HY3	90	2.4618	0.74	0.33	2.9
HY6	82	2.4440	0.47	0.49	5.7
HY9	82	2.4354	0.22	0.51	9.2
HZSM−5	95[b]	–	0.45	0.08	30

[a] The crystallinity of the starting Na^+ form zeolite was considered as 100%; the crystallinity and the unit cell size of the Y samples were obtained from the XRD data by using the ASTM methods D3906-80 and D3942-80

[b] The crystallinity of HZSM-5 was obtained by adsorption of N_2

[c] The bulk Si/AI ratio of the Y zeolites was 2.5

of extra-framework aluminum in the pores and channels. Similarly, increase in the framework Si/Al ratio from 2.9 (for HY3) to 9.2 (for HY9) indicates increase dealumination with increase steaming severity.

The acidity of the zeolites was measured by the FT-IR spectra of pyridine adsorption. All zeolites showed bands at 1,540 and 1,450 cm^{-1} corresponding to concentration of Bronsted and Lewis acid. For the HY zeolites, the ratio of Lewis to Bronsted acid sites increased with the framework Si/Al ratio; however, the relative concentration of these acid sites decreased with decrease in framework aluminum content. The main acid sites in HZSM-5 were Bronsted, instead of Lewis acid sites. This study reveals that the acidity of the catalysts decreases in the order: HY3 > HY6 > HY9 > HZSM-5.

3.2 Catalytic Activity of Different Catalysts

Initial investigations of the effect of the catalyst type on the reaction yielded some interesting results (Table 2). All of the four zeolites were active catalysts for the reaction, generally led to *N*-tosylimine **1b** as the main product (Scheme 1). On the other hand, their activities were quite different (Table 2, entry 1–4). The yield of *N*-tosylimine **1b** during 8 h of reaction increases in the order: HY3 ≈ HY6 > HZSM-5 > HY9, which is similar to the order of the increase in Bronsted acid site densities of the catalysts (Table 1). Although the activity of HY6 was close to that of HY3, its Bronsted acid site density was quite different (the difference between HY3 and HY6 Bronsted acidity is 0.74 vs. 0.47 mmol/g, respectively). This fact can be explained by means of two possible assumptions: Firstly, the activity enhancement in dealuminated samples can be attributed to increase in strength of Bronsted acid sites; steam dealumination generates non-framework Al species, which could withdraw electron density from nearby Bronsted acid sites, causing an enhancement in the strength of the site [54, 55]. Therefore, it is widely accepted that the higher the framework Si/Al ratio of the zeolite, the higher the strength of its acid sites and lower their density; the above yield of **1b** implies that the activity per acid sites in HY6 is much greater than that of HY3 and that the condensation reactions require strong acid sites capable of protonating the carbonyl group of 4-chlorobenzaldehyde [56]. Secondly, HY6 is more hydrophobic than HY3 since the framework dealumination enhances the hydrophobicity of the zeolite, the 'micro-equilibrium' of the condensation taking place in the pore of the zeolite could naturally shift towards the formation of *N*-tosylimine and water, the latter would leave the hydrophobic surface of the zeolite as soon as it was formed. On the contrary, the amount of Bronsted acid sites for HZSM-5 and HY6 (0.45 and 0.47 mmol/g, respectively) is about the same, but the yield of **1b** for HZSM-5 was much lower than that for HY6. This difference is naturally due to the smaller pore of HZSM-5 in which the diffusion of the voluminous molecules is difficult. For the above reasons, this order of activity of the zeolites, being consistent with and opposite to the order of the Bronsted and Lewis acid sites, respectively, could imply that the condensation was preferentially catalyzed by Bronsted acid site, instead of Lewis acid site. The fact that the moderate yield of **1b** (58.1%) (Table 2, entry 3) with HZSM-5 confirms above hypothesis, since the concentration of the Lewis acid sites in HZSM-5 is only 0.08 mmol/g (Table 1, entry 4). However, when the reaction was allowed to run to 18 h, the yield of **1b** with HY9, HZSM-5, HY6 and HY3 catalysts was 98.0, 91.0, 89.1 and 87.5% (Table 2, entry 5–8), respectively. Apparently, with HY6 and HY3 catalysts, the condensation reactions were faster than that with HY9 and HYZSM-5, which attained the equilibrium in several hours. The higher hydrophobicity character of the HY9 in respect to the other samples, assigned to its higher extra-framework aluminum content [57], can indeed explain the higher yield observed over this zeolite. Due to its extra-framework aluminum content, the HY9 zeolite has the highest hydrophobicity, which can play an important role influencing the 'micro-equilibrium' of the condensation. Therefore, the higher hydrophobicity of HY9 is a key factor in its better performance.

Table 2 Effect of catalyst on the yield of *N*-(4-Chlorobenzylidene)-*p*-tolylsulfonamide **1b**

Entry[a]	Catalyst	Catalyst weight (g)	Time (h)	Yield (%)[c]
1	HY3	0.18	8	85
2	HY6	0.18	8	84.9
3	HZSM-5	0.18	8	58.1
4	HY9	0.18	8	49.9
5	HY3	0.18	18	87.5
6	HY6	0.18	18	89.1
7	HZSM-5	0.18	18	91.0
8	HY9	0.18	18	98.0
9	–	0	18	0
10	HY9	0.09	19	90.2
11	HY9	0.36	14[b]	86.6
12	HY9	0.54	11[b]	76.4

[a] The reaction was carried out according to typical experimental procedure

[b] 100% conversion at this time

[c] Isolated yield

The catalyst plays a crucial role in the product formation. *N*-tosylimine **1b** was not formed when the reaction was carried out in the absence of any catalyst (Table 2, entry 9). It is worthy to mention that the HY9 catalyst is readily recyclable and can be reused without significant loss of activity. Variations of the amount of catalyst were explored for the condensations in the presence of HY9 (Table 2). With 0.09 g HY9, *N*-tosylimine **1b** was obtained in 90.2% yield after 19 h (Table 2, entry 10). The rate was faster when more HY9 was used; however, the reaction yield decreased from around 86–76% on increasing the weight of HY9 from 0.36 to 0.54 g (Table 2, entry 11 and 12), since the side products increased and more product molecules remained adsorbed inside the pores. The optimal amount of HY9 was found to be 0.18 g, which gave an excellent yield (98.0%).

3.3 Influence of Different Solvents

The condensation of *N*-tosylamine with 4-chlorobenzaldehyde was chosen as a model reaction for the investigations of the effect of solvent. Various organic solvents such as tetrahydrofuran, benzene, dichloroethane, dichloromethane and ethanol were selected for the study (Table 3). Among the solvents examined, dichloromethane was found to be the most effective solvent. The reaction performed equally well in dichloroethane and benzene whereas in ethanol the reaction proceeded slowly.

Table 3 Effect of solvents on the yields of *N*-(4-Chlorobenzylidene)-*p*-tolylsulfonamide **1b**

Entry[a]	Solvent	Time (h)	Yield (%)[b]
1	CH_2Cl_2	18	98.0
2	$ClCH_2CH_2Cl$	18	96.9
3	Benzene	14[c]	91.3
4	THF	14	53.9
5	EtOH	14	19.0

[a] The reaction was carried out according to typical experimental procedure

[b] Isolated yields

[c] 100% conversion at this time

3.4 Influence of Different Substrates

Under the optimized reaction condition (with 0.18 g HY9 zeolite in CH_2Cl_2 at reflux), we examined the scope of the HY9 zeolite mediated condensations of *N*-tosylamine with various aldehydes. The results are shown in Table 4. In general, condensation of aromatic aldehydes afforded the corresponding *N*-tosylimine in excellent yield (above 90%) (Table 4, entries 2–5, 9). Both electron rich and deficient substrates worked well in this reaction conditions. Nevertheless, when the conjugated aldehyde was subjected to this reaction (Table 4, entries 7), the yield dropped to 65.2%. In contrast to the aromatic aldehydes, the less active aliphatic aldehydes such as butyraldehyde proceeded slower with lower yield (59.5%). Low yields were also observed with all bulky substrates (Table 4, entries 10–12). For 4-tert-butylbenzaldehyde, being lower diffusion coefficient of the reaction product, the yield of **1k** decreased to 53% in a longer reaction time (37 h). The conversion (<6%) of 4-formyl[2,2]paracyclophane was drastically suppressed in dichloromethane solvent at 40 °C after 48 h, due to the bulkier size of the condensation product did not have easy access under the typical reaction condition. However, the lower yield (26%) of *N*-tosylimine **1l** was finally obtained using 1,2-dichloroethane as solvent at elevated temperature (83 °C) over the 66 h time period. The above results showed that the *N*-tosylimine **1l** seems to be too bulky to diffuse in microporous structure, but higher temperature can help the product run out of the inner surface of HY9 zeolite (That is, in higher temperature large molecules can still penetrate the relative small channels with higher vibration energy). It has been suggested that for the esterification of aromatic carboxylic acids the active sites are available inside as well as outside the zeolite pores. However more of these sites have been found to be available inside the pore than on the outside or near the pore mouth [58]. It was suggested that the external surface area of the zeolite has little effect on yield of the reaction, thus indicating that it occurs mainly inside the pores.

Table 4 Results for the condensation reaction of *N*-tosylamine (3.5 mmol) with aldehyde (3.8 mmol) over HY9 (0.18 g) in dichloromethane at reflux (40 °C)

Entry	Product	R	Yield (%)[b]	Melting point (°C)	Time (h)
1	**1a**	Ph[a]	87.4	111–112	8
2	**1b**	4-ClPh	98.0	166–168	18
3	**1c**	4-MeOPh	92.5	124–125	31
4	**1d**	4-NO_2Ph	92.0	196–197	31
5	**1e**	1-Naph	90.6	196–197	12
6	**1f**	2-Furyl[a]	93.2	100–101	13
7	**1g**	Ph–CH=CH	65.2	117–118	16
8	**1f**	Pr	59.5	116–118	25
9	**1h**	2-Cl-6-FPh	90.3	118–120	16
10	**1i**	3,5-Me_2-4-MeOPh	67.2	99–101	19
11	**1j**	4-Me_3CPh	53.0	116–117	37
12	**1k**	PCP[c]	26.0	166–168	66[d]

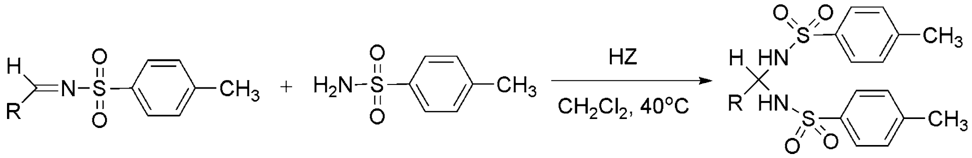
[a] The product was shown as Scheme 2

[b] Isolated yield

[c] PCP: [2,2]paracyclophane

[d] The reaction was carried out in 1,2-dichloroethane at reflux (83 °C)

Scheme 2 Arylidenebis(*p*-tolylsulfonamide) would be formed through attack of *N*-tosylamine at the produced *N*-tosylimine in the presence of zeolites



In general, the condensations of *N*-tosylamine with various aldehydes over HY9 zeolite gave *N*-tosylimine as the major product (Scheme 1) and a trace of *N*,*N*′-arylidenebis(*p*-tolylsulfonamide) as the byproduct (Scheme 2). The byproduct would be formed through attack of *N*-tosylamine at the produced *N*-tosylimine in the presence of HY9 zeolite. In contrast to all the other aldehydes, *N*,*N*′-benzylidenebis(*p*-tolylsulfonamide) and *N*,*N*′-furfurylidenebis(*p*-tolylsulfonamide) were obtained in high yields with benzaldehyde and furaldehyde in the condensations (Table 4, entry 1, 6), in spite of the shape-selectivity of the zeolite towards. These results suggested that the addition of *N*-tosylamine to *N*-tosylimines from benzaldehyde or furaldehyde could take place inside the pores of HY9 zeolite, since the molecular dimensions of benzaldehyde and furaldehyde are smaller than that of other aromatic aldehydes.

4 Conclusion

We have described a highly efficient method for the preparation of *N*-tosylimines using acidic zeolites as catalysts. The condensation is preferentially catalyzed by Bronsted acid sites and occurs mainly inside the pores of zeolites. HY9 zeolite is applicable to the condensations of a variety of aldehydes, including electron deficient and electron rich aromatic aldehydes, and aliphatic aldehydes. The catalyst is mild, affordable and commercially available. This new method offers several advantages, including mild reaction conditions, high yields and readily recyclable catalysts. Therefore, zeolites could be a viable, ecofriendly and recyclable solid acid catalyst for the synthesis of *N*-tosylimines.

Acknowledgments Financial support from the National Natural Science Foundation of China (Grant No. 20441004, 20671059) and Department of Science and Technology of Shandong Province is gratefully acknowledged.

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