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Polystyrene-Supported $AlCl_3$ as a Highly Active and Reusable Heterogeneous Lewis Acid Catalyst for the One-Pot Synthesis of Quinoxalines

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ABSTRACT: *A new and environmentally benign protocol for the synthesis of quinoxaline derivatives through the condensation reactions of 1,2-diketones and 1,2-phenylenediamines using cross-linked polystyrene-supported aluminum chloride (PS/$AlCl_3$) as a highly active and reusable heterogeneous Lewis acid catalyst is described. This polymeric catalyst is stable and can be easily recovered and reused without appreciable change in its efficiency.* Heteroatom Chem 00:1–6, 2012; View this article online at wileyonlinelibrary.com. DOI 10.1002/hc.21039

INTRODUCTION

Functionalized quinoxalines are an important class of nitrogen-containing heterocycles, and they constitute useful intermediates in organic synthesis. Quinoxaline derivatives have broad biological activities and have been used as anticancer [1], antiviral [2], anti-inflammatory [3], antibacterial agents [4], and kinase inhibition agents [5]. In addition to the medicinal applications, quinoxalines have been used as dyes [6] and key intermediates in the synthesis of organic semiconductors [7]. Over the years, a number of synthetic methods for the preparation of quinoxaline derivatives have been reported in the literature [8]. Among them, the condensation of 1,2-aryldiamine with 1,2-diketone in refluxing ethanol or acetic acid for 2–12 h is a general approach [9]. In addition to common homogeneous Lewis acids, many other catalysts including I_2/DMSO and CH_3CN [10], sulfamic acid/CH_3OH [11], $CuSO_4·5H_2O$ [12], polyaniline-salt/dichloroethane [13], $MnCl_2$[14], Zn[(l)proline]/HOAc [15], CAN [16], montmorillonite K-10 [17], $H_6P_2W_{18}O_{62}·24H_2O$ [18], *p*-TSA [19], $Bi(OTf)_3$[20], and $NbCl_5$[21] have been used to promote these condensations. Oxidative couplings of epoxides and ene-1,2-diamines [22] catalyzed by Bi(0), $Pd(OAc)_2$, $RuCl_2$-$(PPh_3)_3$-TEMPO, and MnO_2 have been reported [23]. The condensation has also been accomplished under catalyst-free conditions, but needs microwave and ultrasound irradiations [24]. In addition, the above cyclocondensation process could proceed in ionic liquid [25]. However, some of the reported procedures suffer from one or more drawbacks, such as the use of harmful organic solvents (pollution), harsh reaction conditions, unsatisfactory product yields, tedious work-up processes (especially in polar solvents), long reaction times, the use of toxic and detrimental metal precursors as catalysts, and relatively expensive reagents, requirement for special apparatus. The main disadvantage of almost all existing methods is that the catalysts are destroyed in the work-up procedure, and their recovery and reuse is often impossible, which limit their use under environmentally benign processes. Owing to the

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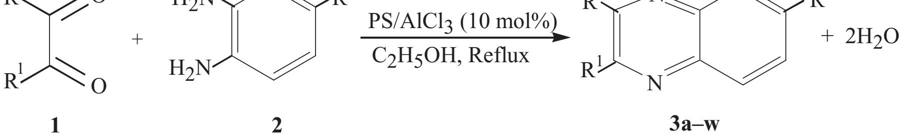
importance of quinoxalines from pharmaceutical, industrial, and synthetic points of view, introduction of a simple, efficient, versatile, and environmentally friendly method for the preparation of these compounds is still in demand.

In the past few decades, there have been significant developments in the application of effective and safe heterogeneous catalysts for the production of organic chemicals [26]. Heterogeneous catalysts, which are widely used in organic transformations, have good thermal stability, can be easily separated from the reaction mixture, and can be often regenerated and reused. Therefore, heterogenizing of a homogeneous metal species by supporting it on an insoluble support has attracted a lot of interest as a suitable method for solving many practical problems including recovery of the catalyst from a reaction mixture and recycling. The use of polymer-supported catalysts in organic transformations and the design of functionalized polymers carrying metal species have gained importance in organic synthesis since they offer economic advantage in developing reusable catalysts and environmental friendly processes [27]. Polystyrene-supported aluminum chloride, PS/$AlCl_3$, which is a tightly bound complex between anhydrous $AlCl_3$ and PS-divinylbenzene copolymer, has been described for the first time by Neckers and coworkers [28a–28d]. In recent years, a PS–$AlCl_3$complex has been reported to be an efficient catalyst for some organic transformations [28]. As part of our continuing interest in heterogeneously catalyzed organic reactions [29,30], we report herein an efficient and ecofriendly procedure for the synthesis of quinoxalines by the one-pot condensation reaction of 1,2-phenylenediamines with 1,2-diketones catalyzed by PS-supported $AlCl_3$ as a reusable heterogeneous Lewis acid catalyst (Scheme 1).

RESULTS AND DISCUSSION

For our investigations, cross-linked PS-supported aluminum chloride (PS/$AlCl_3$) was prepared by addition of anhydrous aluminum chloride to PS (8% divinyl benzene, grain size 0.25–0.65 mm) in carbon disulfide under reflux conditions. The loading capacity of the polymeric catalyst obtained by using the gravimetric method and checked by an atomic absorption technique was 0.485 mmol $AlCl_3$/g of complex beads catalyst [31]. The data obtained by these two techniques showed, within experimental error, that the catalyzing species are in the form of $AlCl_3$ supported on the polymeric support. The UV spectrum of the solution of the PS–$AlCl_3$ complex in CS_2 showed a new strong band at 480 nm, which is due to the formation of a stable $\pi \rightarrow p$ type coordination complex between the benzene rings in the PS carrier with aluminum trichloride. The IR spectrum of PS/$AlCl_3$ showed new absorption peaks due to the C—C-stretching vibration and the C—H-bending vibration of the benzene ring at 1550–1650 and 400–800 cm^{-1}, by which complex formation was demonstrated. Although $AlCl_3$ is a water-sensitive, corrosive, and environmentally harmful compound, the PS–$AlCl_3$ complex is a stable and water-tolerant species. This polymeric catalyst is easy to prepare, stable in air for a long time without any change, easily recycled, and reused without appreciable loss of its activity.

At the onset of the research, the model reaction between 1,2-phenylenediamine (**2a**) and benzil (**1a**) was investigated to find the most appropriate reaction conditions and evaluate the catalytic efficiency of the PS/$AlCl_3$ catalyst. The results are summarized in Table 1. It was found that polar solvents such as CH_3CN and C_2H_5OH were better than nonpolar solvents. No reaction occurred when H_2O was used as a solvent (Table 1, entry 6); it may be because of the aggregation of the catalyst caused by its hydrophobic nature, leading to inadequate access of substrates to the active sites of the catalyst. We therefore selected ethanol as a solvent. Further studies showed that the reaction temperature has a great influence on the model reaction, and the best results were observed when the reaction temperature was 78°C (Table 1, entries 9,12,13). Consequently, we chose 78°C as the optimal temperature for the model



R^1 = Ph, 4-(Cl)-Ph,4-(Br)-Ph,4-(MeO)-Ph, 4-(Me)-Ph, Me, 2-Furil, H
R^2 = H, Me, NO_2, Cl, PhCO

SCHEME 1 Synthesis of quinoxalines catalyzed by PS/$AlCl_3$.

TABLE 1 PS/$AlCl_3$-Catalyzed Condensation Reaction of Benzil (**1a**, 1 mmol) with 1,2-Phenylenediamine (**2a**, 1 mmol) under Different Reaction Conditions

Entry	*Solvent*[a]	*Catalyst (mol%)*	*Time (min)*	*Yield (%)*[b]
1	Toluene	10	60	40
2	Cyclohexane	10	60	<5
3	CH_2Cl_2	10	60	15
4	CH_3Cl	10	60	15
5	CH_3CN	10	60	71
6	H_2O	10	60	NR[c]
7	C_2H_5OH	–	10	NR
8	C_2H_5OH	5	10	63
9[d]	C_2H_5OH	10	10	96, 93, 91, 90,88
10	C_2H_5OH	20	10	95
11[e]	C_2H_5OH	–	10	NR
12[f]	C_2H_5OH	10	10	54
13[g]	C_2H_5OH	10	10	80
14[h]	C_2H_5OH	10	10	NR

[a] All reactions were carried out under reflux conditions with 10 mol% of catalyst in 3 mL of solvent.
[b] Isolated yield.
[c] NR: No reaction.
[d] Catalyst was reused for five times.
[e] PS was used as a catalyst.
[f,g] Two reactions were carried out in ethanol at 45 and 60°C, respectively.
[h] Catalyst was removed by filtration after 10 min.

reaction. Next, the amount of the catalyst was examined and we found that the yields were obviously affected by different amounts of catalyst. No reaction was observed in the absence of catalyst, whereas 10 mol% of PS/$AlCl_3$ was sufficient to complete the reaction and excessive amount of catalyst did not increase the yields significantly (Table 1, entries 7–10).

To confirm the activity of PS-supported $AlCl_3$, a comparison experiment was carried out by using cross-linked PS alone under the same experimental conditions. No product was isolated when using PS as a catalyst, which indicated that PS itself did not promote the reaction (Table 1, entry 11). To demonstrate that the reaction catalyzed by the catalyst PS/$AlCl_3$ is really a heterogeneous process and to find out whether the reaction occurred in the solid matrix of the supported catalyst PS/$AlCl_3$ or whether aluminum chloride simply released into the ethanol was responsible for the reaction, PS/$AlCl_3$ was heated under reflux in ethanol for 10 min and then isolated by filtration. When reactants were added to the filtrate and heated under reflux for 10 min, no reaction took place (Table 1, entry 14). Therefore, we may conclude that any $AlCl_3$ that leached into the reaction mixture is not an active homogeneous catalyst and that the observed catalysis is truly heterogeneous in nature. However, one of the important points regarding the heterogeneous catalysts is their deactivation and reusability. To test this, a series of five consecutive runs were carried out with the same catalyst PS/$AlCl_3$ sample, without a noticeable decrease in the activity, furnishing the corresponding product with 96%, 93%, 91%, 90%, and 88% isolated yields (Table 1, entry 9). This reusability demonstrates the high stability and turnover of a polymeric Lewis acid catalyst under operating conditions. After using the catalyst PS/$AlCl_3$, the solid was simply filtered off, washed with ethanol and ether, and reused.

To evaluate the efficiency and generality of this methodology, we next investigated the scope and limitation of this condensation reaction under optimized conditions (C_2H_5OH, 10 mol% of PS/$AlCl_3$, reflux), and the results are summarized in Table 2. As can be seen from Table 2, a variety of structurally diverse 1,2-phenylenediamines and 1,2-diketones underwent the condensation reaction smoothly to afford the corresponding quinoxaline derivatives (**3**) in excellent yields. The electronic property of the substituents on the aromatic ring of the diamine part had an obvious effect on reaction time under the above optimal reaction conditions.

TABLE 2 Preparation of Quinoxaline Derivatives[a] Catalyzed by PS/$AlCl_3$ Using Different Aromatic 1,2-Phenylenediamines (**2**) and 1,2-Diketones(**1**)

Entry	*R^1*	*R^2*	*Product*	*Time (min)*	*Yield (%)*[b]	*Ref.*
1	Ph	H	**3a**	10	96	[16]
2	Ph	Me	**3b**	8	97	[16]
3	Ph	NO_2	**3c**	40	90	[16]
4	Ph	Cl	**3d**	20	91	[16]
5	4-(Cl)C_6H_4	H	**3e**	15	96	[15]
6	4-(Cl)C_6H_4	Me	**3f**	15	95	[15]
7	4-(Br)C_6H_4	H	**3g**	15	94	[21]
8	4-(Br)C_6H_4	Me	**3h**	10	93	[21]
9	4-(OMe)C_6H_4	H	**3i**	25	89	[15]
10	4-(OMe)C_6H_4	Me	**3j**	30	88	[15]
11	4-(OMe)C_6H_4	NO_2	**3k**	60	84	[15]
12	4-(OMe)C_6H_4	Cl	**3l**	35	87	[15]
13	4-(Me)C_6H_4	H	**3m**	25	92	[11]
14	4-(Me)C_6H_4	Me	**3n**	25	92	[11]
15	4-(Me)C_6H_4	NO_2	**3o**	55	85	[11]
16	4-(Me)C_6H_4	Cl	**3p**	35	91	[11]
17	Ph	PhCO	**3q**	55	91	[21]
18	Me	H	**3r**	20	92	[32]
19	Me	Me	**3s**	20	94	[32]
20	2-Furil	H	**3t**	10	95	[16]
21	2-Furil	Me	**3u**	10	96	[16]
22	H	H	**3v**	15	95	[33]
23	H	Me	**3w**	12	94	[33]

[a] *Reaction conditions*: diamine (1 mmol), 1,2-diketone (1 mmol), PS/$AlCl_3$(10 mol%), ethanol (3 mL), reflux.
[b] Isolated yield.

It was observed that electron-withdrawing groups (Table 2, entries 3,4,11,12,15–17) associated with 1,2-phenylenediamines decreased the reactivity of the substrate, and long reaction times were required. In contrast, electron-donating groups at the phenyl ring of 1,2-diamine favored the formation of product (Table 2, entry 2). On the other hand, the effect of electronic factors associated with aromatic 1,2-diketones is opposite. Since only symmetric 1,2-diketones were used for the condensation reactions, no regioisomers were generated as the products. To check the versatility of this method, other 1,2-dicarbonyls such as 2,3-butanedione (Table 2, entries 18,19), furil (Table 2, entries 20,21), and glyoxal (Table 2, entries 22,23) were subjected to a condensation reaction and the corresponding products were obtained with excellent yields.

The actual mechanism of the reaction is unclear. However, based on the experimental results and by referring to the literature [14,16], the mechanism of the quinoxaline formation proceeds by the usual pathway proposed using Lewis acids, involves the complexation of aluminum with diketone by acting as a Lewis acid and also plays a role in promoting the dehydration steps.

Finally, the efficacy of PS/$AlCl_3$ was compared with that of other catalysts reported earlier. The synthesis of **3a** was considered as a representative example (Table 3). As indicated in Table 3, in addition to having the general advantages attributed to polymeric-supported catalysts, PS/$AlCl_3$ is an equally or more efficient catalyst for this condensation reaction in terms of the yield and reaction rate.

In conclusion, PS/$AlCl_3$ as a noncorrosive, environmentally benign, and stable heterogeneous polymer-supported Lewis acid has proved to be an efficient catalyst for the efficient synthesis of quinoxaline derivatives. The short reaction times, simple experimental procedure and product isolation, wide applicability, high yields, mild reaction conditions, low cost, and easy preparation and safe handling of the catalyst are important features of this new methodology. In addition, the use of PS/$AlCl_3$ has resulted in a reduction in the unwanted and hazardous waste that is produced during conventional homogeneous processes. Finally, this polymeric Lewis acid catalyst can be recovered and reused at least five times with negligible loss in its activity.

EXPERIMENTAL

Materials and Methods

All chemical used were obtained from commercial suppliers and used without further purification. Melting points were measured on an Electrothermal 535 apparatus and were uncorrected. ^1H and ^{13}C NMR spectra were recorded on a Bruker DPX-250 Avance spectrometer (Karlsruhe, Germany) at 250.13 MHz. IR spectra were recorded with a Unicam Matteson 1000 spectrophotometer. Reaction monitoring and purity determination of the products were accomplished by thin layer chromatography (TLC) on silica gel polygram SILG/UV 254 plates. The capacity of the catalyst was determined by the Mohr titration method and atomic absorption technique using a Philips atomic absorption instrument. All yields refer to isolated products. Elemental analyses were performed using a Heraeus CHN-O-Rapid analyzer (Hanau, Germany) at Research Institute of

TABLE 3 Comparison of some previously reported procedures with the present method for the synthesis of **3a**

Entry	*Catalyst (Loading)*	*Reaction Conditions*	*Time (min)*	*Yield (%)*[a]	*Ref.*
1	Montmorillonite K-10 (10% w/w)	H_2O, rt	2.5 h	100	[17]
2	Polyaniline sulfate (5% w/w)	CH_2ClCH_2Cl, rt	20	95	[13]
3	Zn[(l)proline] (10 mol%)	HOAc, rt	10	94	[15]
4	Amberlyst-15 (24 mol%)	H_2O, 70°C	19	98	[35]
5	$MnCl_2$(10 mol%)	C_2H_5OH, rt	17	94	[14]
6	I_2(10 mol%)	CH_3CN, rt	30	90	[10]
7	*p*-TSA (10 mol%)	Solvent free, rt	5	90	[19]
8	I_2(10 mol%)	DMSO, rt	40	90	[10]
9	ZrO_2/Ga_2O_3/MCM-41	CH_3CN, rt	40	90	[34]
10	$FeCl_3 \cdot 6H_2O$ (10 mol%)	Solvent free, rt	5	35	[19]
11	$CoCl_2 \cdot 6H_2O$ (10 mol%)	Solvent free, rt	5	24	[19]
12	$AlCl_3$(10 mol%)	Solvent free, rt	6	34	[19]
13	$SnCl_4 \cdot 5H_2O$ (10 mol%)	Solvent free, rt	7	32	[19]
14	HCl (10 mol%)	Solvent free, rt	6	66	[19]
15	H_2SO_4(10 mol%)	Solvent free, rt	5	57	[19]
16	PS/$AlCl_3$(10 mol%)	C_2H_5OH, reflux	10	96	Table 2

[a] Isolated yields.

Petroleum Industry (Tehran, Iran), and the results agreed favorably with calculated values.

Typical Procedure for Preparation of PS/$AlCl_3$

Anhydrous $AlCl_3$ (4.5 g) was added to PS (8% divinylbenzene, grain size 0.25–0.68 mm, 3.5 g) in carbon disulfide (25 mL) as the reaction medium. The mixture was stirred using a magnetic stirrer under reflux condition for 1 h, cooled, and then water (50 mL) was cautiously added to hydrolyze the excess $AlCl_3$. The mixture was stirred until the deep orange color disappeared, and the polymer became light yellow. The polymer beads were collected by filtration and washed with water (300 mL) and then with ether (30 mL) and acetone (30 mL). The catalyst was dried in a vacuum oven overnight at 50°C. Complex formation was demonstrated by a new band at 1630 cm^{-1}in the IR spectrum of polymer. The chlorine content of PS/$AlCl_3$ was 5.16% analyzed by the Mohr titration method [31], and the loading capacity of $AlCl_3$ on the polymeric catalyst was calculated to be 0.485 mmol/g. One gram of solid catalyst (PS/$AlCl_3$) was decomposed by burning with Na metal, extracted with 10 mL of water and filtered. The chlorine content of the filtrate was determined by the Mohr titration method.

General Synthetic Procedure for the Preparation of Quinoxaline Derivatives

In a round-bottomed flask (25 mL) equipped with a condenser and a magnetic stirrer, a mixture of 1,2-phenylenediamine (**2,** 1 mmol), 1,2-diketone (**1**, 1 mmol), and PS/$AlCl_3$ (0.206 g, 0.1 mmol of $AlCl_3$) in ethanol (3 mL) was heated under reflux for an appropriate time as indicated by TLC. After completion of the reaction, hot ethanol was added to the mixture and the catalyst was collected by filtration and then washed with ether (2×5 mL). The filtrate was concentrated on a rotary evaporator under reduced pressure, and the solid product was washed with water (2×10 mL) and recrystallized from ethanol to afford the pure products **3a–w**. The products were characterized by IR, NMR spectroscopies data, and their melting points, and they are also compared with respect to authentic compounds reported in the literature, which agreed with reported values [11,15,16,21,32,33].

Spectral and Physical Data for Selected Products

*6-Chloro-2,3-bis(4-methoxyphenyl)quinoxaline (***3l***, $C_{22}H_{17}ClN_2O_2$)*. Pale yellow solid; mp 150–151°C; IR (KBr): 2935, 2835, 1605, 1510, 1345, 1292, 1245, 1175, 832 cm^{-1}; ^1H NMR (250 MHz, $CDCl_3$): δ_H 8.10 (d, J = 2.30 Hz, 1H), 8.05 (d, J = 8.90 Hz, 1H), 7.64 (dd, J = 2.20 Hz, 1H), 7.50 (m, 4H), 6.88 (d, J = 8.70 Hz, 4H), 3.80 (s, 6H, OCH_3) ppm; ^{13}C NMR (62.50 MHz, $CDCl_3$): δ_C 160.45, 160.35, 153.82, 141.32, 139.54, 135.15, 131.38, 131.33, 131.25, 130.46, 130.22, 127.88, 113.85, 55.35 ppm.

*6-Methyl-2,3-di-p-tolylquinoxaline (***3n***, $C_{23}H_{20}N_2$)*. Pale yellow solid; mp 136–137°C; IR (KBr): 3430, 2912, 1611, 1450, 1336, 820 cm^{-1}; ^1H NMR (250 MHz, $CDCl_3$): δ_H 8.06 (d, J = 8.50 Hz, 1H), 7.93 (s, 1H), 7.55 (dd, J = 8.50 Hz, 1H), 7.45 (d, J = 7.95 Hz, 4H), 7.14 (d, J = 7.90 Hz, 4H), 2.59 (s, 3H), 2.36 (s, 6H) ppm; ^{13}C NMR (62.50 MHz, $CDCl_3$): δ_C 153.34, 152.60, 141.26, 140.11, 139.68, 138.60, 138.52, 136.63, 132.02, 129.85, 129.81, 128.98, 128.70, 128.02, 21.90, 21.40 ppm.

*6-Methyl-2,3-dimethylquinoxaline (***3s***, $C_{11}H_{12}N_2$)*. Pale yellow oil; IR (KBr): 2970, 1622, 1565, 1452, 820 cm^{-1}; ^1H NMR (250 MHz, $CDCl_3$): δ_H 7.90 (d, J = 8.50, 1H), 7.79 (s, 1H), 7.53 (d, J = 8.50 Hz, 1H), 2.75 (s, 6H), 2.60 (s, 3H) ppm; ^{13}C NMR (62.50 MHz, $CDCl_3$): δ_C 153.70, 152.80, 141.60, 139.90, 139.50, 131.40, 128.20, 127.70, 23.60, 23.50, 22.10 ppm.

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