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Tetrahedron Letters 45 (2004) 7641–7644
Tetrahedron Letters

Magnesium perchlorate as an efficient catalyst for the synthesis of imines and phenylhydrazones

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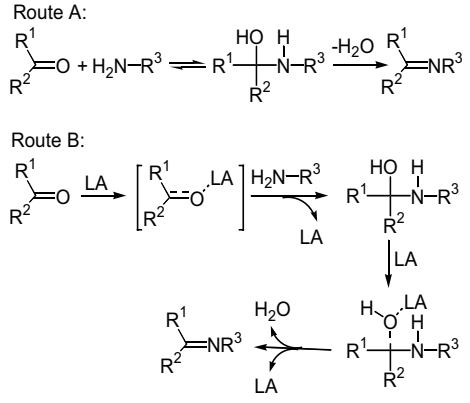
Received 10 July 2004; revised 12 August 2004; accepted 17 August 2004

Abstract—Magnesium perchlorate has been found to be an efficient catalyst for the synthesis of imines and phenylhydrazones by the reaction of carbonyl compounds with amines and phenylhydrazine in high yields at room temperatures and in short times. The condensation of less electrophilic carbonyl compounds with poorly nucleophilic amines affords the imines in excellent yields.

1. Introduction

The condensation of amines with carbonyl compounds is a venerable and useful organic transformation[1] as the resultant imines are used as versatile components in nucleophilic addition with organometallic reagents,[2] in cycloaddition reactions,[3] and have potential for therapeutic applications such as lipoxygenase inhibitors, anti-inflammatory agents[4] and anti-cancer agents.[5] Various synthetic routes for the synthesis of imines are depicted in Scheme 1. As nucleophilic attack by the amine at the carbonyl carbon in the firststep is reversible, the feasibility of imine formation largely depends on the rate of removal of water in the final step (route A). The classical synthesis of imines, originally reported by Schiff,[6] involves condensation of a carbonyl compound with an amine under azeotropic distillation[7] to separate the liberated water. Subsequently, removal of water was facilitated by the use of molecular sieves.[8] Recently an in situ dehydration strategy has been adopted by the use of dehydrating solvents such as tetramethyl orthosilicate[9] and trimethyl orthoformate.[10] In an alternative approach (route B), the condensation reaction has been carried out in the presence of $ZnCl_2$,[11] $TiCl_4$,[12] $MgSO_4$–PPTS,[13] alumina,[14] K-10 under microwave irradiation,[10b,15] $Ti(OR)_4$,[16] and $CuSO_4$,[16b] which act as Lewis acids to catalyze nucleophilic attack on the carbonyl group by the amine as well as serving as dehydrating agents to facilitate the removal of water in the final step.



Scheme 1. Synthetic routes for imine formation.

The methodologies reported have disadvantages such as the requirement for high reaction temperatures, prolonged reaction periods, an excess of costly dehydrating reagents/catalysts, moisture sensitive catalysts, and special apparatus. Moreover, the efficiency of the procedures reported is limited to the reaction of highly electrophilic carbonyl compounds and strongly nucleophilic amines.

2. Results and discussion

We thought that the condensation between a carbonyl compound and an amine leading to the formation of

Keywords: Magnesium perchlorate; Catalyst; Imines; Phenylhydrazones.
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0040-4039/$ - see front matter © 2004 Elsevier Ltd. All rights reserved.
doi:10.1016/j.tetlet.2004.08.097

an imine should be a facile reaction due to the good electrophilic and nucleophilic properties of the carbonyl and amine groups, respectively, and may not require any catalytic assistance in the absence of electronic/steric factors that might decrease the electrophilicity/nucleophilicity of the carbonyl/amine groups. Thus, the condensation of aniline with benzaldehyde, 4-methoxybenzaldehyde, 2-hydroxybenzaldehyde, and 4-hydroxybenzaldehyde afforded 85–100% yields, in 5 min–1 h, under neat conditions at room temperature without the requirement of any additional reagent/catalyst although the reported procedures[14,15] highlight the need to use a catalyst for these reactions. The treatment of aniline with 4-nitrobenzaldehyde and 4-dimethylaminobenzaldehyde afforded the corresponding imines in 75% and 97% yields in 1 and 6 h, respectively, and a quantitative yield was obtained during the reaction of 4-methoxyaniline with 4-nitrobenzaldehyde in 1 h in DCE at room temperature in the absence of catalyst. Similarly, the reaction of benzaldehyde, 4-methoxybenzaldehyde, 4-dimethylaminobenzaldehyde, and cinnamaldehyde with (*R*)-(+)-α-methylbenzylamine resulted in imine formation in >90% yields in DCE or under neat conditions in the absence of catalyst in 0.25–0.5 h although the literature reports[14] the use of catalyst for the reaction of benzaldehyde with α-methylbenzylamine and the reaction of cinnamaldehyde with the less sterically hindered benzylamine. However, no significant imine formation was observed (GC–MS) when 4-methoxybenzaldehyde was treated with 4-nitroaniline either under neat conditions or in DCE for 70 h at room temperature or under reflux in EtOH or PhMe for 24 h.

The presence of the methoxy group in 4-methoxybenzaldehyde reduces the electrophilicity of the carbonyl carbon through resonance and the strong electron withdrawing property of the nitro group in 4-nitroaniline decreases the nucleophilicity of the amine group. Thus, the combination of these substrates constitutes a model reaction for evaluating the efficiency of a catalyst. Hence, 4-methoxybenzaldehyde was treated with 4-nitroaniline under various conditions (Table 1). While planning to select a suitable Lewis acid for catalyzing the reaction, we thought that a catalyst known for activation of an electrophile should serve the purpose. Logically, we focused our attention toward acylation catalysts. Recently, we reported that $Mg(ClO_4)_2$ efficiently catalyzes the acylation of various phenols, alcohols, thiols, and amines.[17] To our satisfaction we found that the use of 5 mol% of $Mg(ClO_4)_2$ resulted in quantitative formation of the corresponding imine in DCE at room temperature within 8 h (entry 1). Increase in the reaction temperature led to imine formation in comparable yield, in 1 h. Inferior yields were obtained when carrying out the condensation reaction in other solvents such as PhMe, THF, MeCN, and EtOH (entries 2–5). The decrease in product yield in PhMe was due to the poor solubility of the reactant 4-nitroaniline. However, in case of THF, MeCN, and EtOH the inferior results were due to the decreased catalytic efficiency of $Mg(ClO_4)_2$ because of competitive coordination of Mg^{2+} with the solvent. The distinct role of $Mg(ClO_4)_2$ in catalyzing the condensation reaction was demonstrated by the lack of imine formation when carrying out the reaction in DCE for 70 h at room temperature, and in PhMe or EtOH under reflux for 24 h, and when heating under microwave irradiation for 5 min in the absence of $Mg(ClO_4)_2$ (entries 6–9). Next we explored the catalytic efficiency of other magnesium derivatives such as $MgSO_4$, $MgBr_2$, and $Mg(OTf)_2$. However, $MgSO_4$ did not exhibit any significant catalytic activity[13] and only a 60% yield was obtained in the presence of $MgBr_2$ and $Mg(OTf)_2$ establishing that amongst the various magnesium salts used, $Mg(ClO_4)_2$ was the most effective in catalyzing imine formation. This was further substantiated by the observation that a 95% yield was obtained when carrying out the reaction under reflux for 1 h in the presence of $Mg(ClO_4)_2$, whereas the corresponding reaction carried out in the presence of $Mg(OTf)_2$ required 3 h

Table 1. Condensation of 4-methoxybenzaldehyde with 4-nitroaniline in the presence of various catalysts[a]

Entry	Catalyst	Solvent	Yield (%)[b]
1	$Mg(ClO_4)_2$	DCE	100[c]
2	$Mg(ClO_4)_2$	PhMe	60[d]
3	$Mg(ClO_4)_2$	THF	50
4	$Mg(ClO_4)_2$	MeCN	60
5	$Mg(ClO_4)_2$	EtOH	60
6	None	DCE	Nil[e]
7	None	PhMe	Nil[f]
8	None	EtOH	Nil[f]
9	None	Neat	Nil[g]
10	$Mg(SO_4)_2$	DCE	9
11	$MgBr_2$	DCE	60
12	$Mg(OTf)_2$	DCE	60[h]
13	$LiClO_4$	DCE	16
14	$Sc(OTf)_3$	DCE	Nil
15	$Yb(OTf)_3$	DCE	66
16	$InCl_3$	DCE	14
17	Zeolite (ZSM-5)	DCE	Nil[i]
18	Zeolite (K/L)	DCE	Nil[i]
19	Montmorillonite KSF	DCE	Nil[i]
20	Montmorillonite K 10	DCE	23
21	Montmorillonite K 10	Neat	Nil[f,i]
22	$La_2(SO_4)_3$	DCE	35
23	$LaCl_3{\cdot}7H_2O$	DCE	61
24	$ZrCl_4$	DCE	40
25	$ZrOCl_3{\cdot}8H_2O$	DCE	27
26	$ZnCl_2$	DCE	46
27	$TiCl_4$	DCE	35
28	$Ti(OPr^i)_4$	DCE	70
29	$CuSO_4{\cdot}H_2O$	DCE	65
30	Neutral Al_2O_3	DCE	Nil[i]

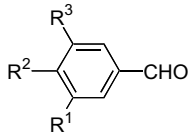
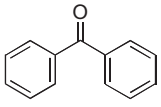
[a] An equimolar amount of the substrates were reacted in the presence of 5 mol% (except for entries 6–9, 17–21, and 30) of the catalyst at room temperature (except for entries 7–9, 20) for 8 h.
[b] Determined by ^1H NMR and GC–MS.
[c] A 95% yield was obtained when carrying out the reaction under reflux for 1 h.
[d] An 80% yield was obtained when carrying out the reaction under reflux for 2.5 h.
[e] The reaction was carried out for 70 h at room temperature.
[f] The reaction was carried out for 24 h under reflux.
[g] The reaction mixture was heated under microwave irradiation (BPL, Model BMC 900T, 1.35 KW, micromode, full power) for 5 min.
[h] A 90% yield was obtained when carrying out the reaction under reflux for 3 h.
[i] The catalyst was used in 10% w/w with respect to the aldehydes.

to afford a 90% yield (Table 1, compare footnotes c and h). Being encouraged by the catalytic effect of $Mg(ClO_4)_2$, we planned to evaluate the catalytic efficiency in imine formation of other reported acylation catalysts such as $ZrCl_4$,[18] $InCl_3$,[19] $LiClO_4$,[20] zeolite,[21] $Yb(OTf)_3$,[22] montmorillonite clays,[23] and $Sc(OTf)_3$.[24] None of these were found to be effective except for $Yb(OTf)_3$, which resulted in the formation of the imine in 66% yield (entry 15). The use of other Lewis acids such as $La_2(SO_4)_3$, $ZrOCl_2{\cdot}8H_2O$, $ZnCl_2$,[11] $TiCl_4$,[12] neutral alumina,[14] $Ti(OPr^i)_4$,[16] and $CuSO_4$[16b] afforded lower yields (0–46%) and a 61% yield was obtained in the presence of $LaCl_3{\cdot}7H_2O$.

To establish the scope and limitations of $Mg(ClO_4)_2$ as a catalyst for imine formation, structurally diverse carbonyl compounds were treated with various amines such as aniline (**1**), 4-methoxyaniline (**2**), 4-hydroxyaniline (**3**), 4-methylaniline (**4**), 4-chloroaniline (**5**), 4-nitroaniline (**6**), and benzylamine (**7**) under the catalytic influence of $Mg(ClO_4)_2$ and the results are summarized in Table 2. Excellent results were obtained in most cases.

Since, phenylhydrazones are important intermediates for the synthesis of 1,3-diphenylpyrazoles with potential antiparasitic activities,[25] we next planned to extend the application of $Mg(ClO_4)_2$ as a catalyst for hydrazone formation. However, treatment of phenylhydrazine with 4-methoxybenzaldehyde, 4-hydroxybenzaldehyde, 4-nitrobenzaldehyde, furfural, thiophene-2-carboxaldehyde, and pyridine-3-carboxaldehyde either under neat conditions or in DCE at room temperature for 0.5–2 h afforded the corresponding phenylhydrazones in 75–95% yields without any catalytic assistance. Although in each of these reactions quantitative formation of the phenylhydrazones took place in 2–30 min under the catalytic influence of $Mg(ClO_4)_2$ (5 mol%) in DCE at room temperature, we decided to evaluate the catalytic efficiency of $Mg(ClO_4)_2$ for the condensation of phenylhydrazine with less electrophilic aldehydes/ketones such as 3,4-dimethoxybenzaldehyde, 3,4,5-trimethoxybenzaldehyde, and acetophenone as no significant phenylhydrazone formation was observed when these substrates were treated with phenylhydrazine in the absence of catalyst. The use of 5 mol% of $Mg(ClO_4)_2$ afforded 98%, 90%, and 95% yields of the corresponding phenylhydrazones in DCE at room temperature in 0.25–2 h from 3,4-dimethoxybenzaldehyde, 3,4,5-trimethoxybenzaldehyde, and acetophenone, respectively.

Table 2. $Mg(ClO_4)_2$-catalyzed reaction of carbonyl compounds with amines[a]

Entry	Substrate	Amine	Time (h)	Yield (%)[b,c]
				
1	$R^1 = R^3 = H$; $R^2 = OMe$	**6**	8	95
2	$R^1 = R^3 = H$; $R^2 = NMe_2$	**1**	0.25	95
3	$R^1 = R^2 = OMe$; $R^3 = H$	**1**	4	94
4	$R^1 = R^2 = OMe$; $R^3 = H$	**5**	8	90
5	$R^1 = R^2 = OMe$; $R^3 = H$	**3**	8	95
6	$R^1 = OMe$; $R^2 = OH$; $R^3 = H$	**1**	4	85
7	$R^1 = OMe$; $R^2 = OH$; $R^3 = H$	**2**	5	92
8	$R^1 = OMe$; $R^2 = OH$; $R^3 = H$	**3**	8	91
9	$R^1 = OMe$; $R^2 = OH$; $R^3 = H$	**5**	4	90
10	$R^1 = R^2 = OH$; $R^3 = H$	**5**	8	80
11	$R^1 = R^2 = R^3 = OMe$	**2**	6	96
12	$R^1 = R^2 = R^3 = OMe$	**3**	4	95
13	$R^1 = R^2 = R^3 = OMe$	**4**	4	90
14	$R^1 = R^2 = R^3 = OMe$	**5**	8	93
15	$R^1 = R^2 = R^3 = OMe$	**6**	1	90[d]
16		**7**	8	90[e]

[a] The substrate (1 equiv) was treated with amine (1 equiv) in the presence of $Mg(ClO_4)_2$ (5 mol%) in DCE at room temperature (except for entry 15).
[b] Isolated yield of the corresponding imine.
[c] The compounds were characterized by IR, NMR, and MS.
[d] The reaction was carried out under reflux.
[e] The corresponding reaction, catalyzed by alumina, afforded 82% yield at 120 °C in 6 h.[14]

3. Conclusion

In conclusion, $Mg(ClO_4)_2$ is a new and highly efficient catalyst for the synthesis of imines and phenylhydrazones. The advantages include high yields, mild reaction conditions and use of catalytic amounts of $Mg(ClO_4)_2$.

Caution: It should be mentioned that care must be taken in handling perchlorates as they are potentially explosive, when used in the presence of combustible substances at high temperature.[26] However, some perchlorate salts have high thermal stability,[27] [$LiClO_4$] is thermally stable at or above its melting point of 247 °C and can be dried under vacuum at 160 °C for a prolonged period (~50 h).[28] Exposure to nominal levels of perchlorate does not adversely affect health and safety.[27] Thus, in the present work the requirement of catalytic quantities of $Mg(ClO_4)_2$ and the mild reaction conditions (room temperature) should circumvent the problems of the hazards associated with the use of perchlorates.

4. Experimental

Typical procedure for imine formation: 4-Methoxybenzaldehyde (1.36 g, 10 mmol) was treated with 4-nitroaniline (1.38 g, 10 mmol) in DCE (10 mL) at rt for 8 h (GC–MS) with magnetic stirring in the presence of $Mg(ClO_4)_2$ (0.1 g, 0.5 mmol, 5 mol%). The reaction mixture was diluted with DCE (10 mL), filtered through a bed of Na_2SO_4, and concentrated to afford the desired product (2.43 g, 95%) IR (KBr): ν = 3482, 3362, 1679, 1627, 1599, 1508, 1335, 1165, 1109, 1028, 886, 837 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$): δ = 3.9 (s, 3 H), 7.00 (d, J = 8.5 Hz, 2H), 7.22 (d, J = 8.73 Hz, 2H), 7.86 (d, J = 8.49 Hz, 2H), 8.25 (d, J = 8.68 Hz, 2H), 8.35 (s, 1H); EIMS m/z 256 (M^+), identical with an authentic sample of 4-nitro-*N*-(4-methoxybenzylidene)aniline.[29] This general procedure was followed for

the preparation of other imines. In most cases, the isolated products gave satisfactory physical data (mp, IR, NMR, and MS), which compared well with those of the literature without any further purification. Wherever applicable, purification was performed through crystallization (petroleum ether–EtOAc or petroleum ether–EtOH). *N*-(4-Dimethylaminobenzylidene)aniline (Table 2, entry 2),[30] *N*-(4-hydroxy-3-methoxybenzylidine)aniline (Table 2, entry 6),[31] 4-methoxy-*N*-(4-hydroxy-3-methoxybenzylidine)aniline (Table 2, entry 7),[31a] 4-hydroxy-*N*-(4-hydroxy-3-methoxybenzylidine)aniline (Table 2, entry 8),[31a] 4-chloro-*N*-(4-hydroxy-3-methoxybenzylidine)aniline (Table 2, entry 9),[31a] 4-hydroxy-*N*-(3,4,5-trimethoxybenzylidine)aniline (Table 2, entry 12),[32] and *N*-methylphenylbenzhydrylimine (Table 2, entry 16),[14] were identified by comparison of their spectroscopic data (IR, NMR, and MS) with those reported. *N*-(3,4-Dimethoxybenzylidene)aniline (Table 2, entry 3),[33] 4-chloro-*N*-(3,4-dimethoxybenzylidene)aniline (Table 2, entry 4),[34] 4-hydroxy-*N*-(3,4-dimethoxybenzylidene)aniline (Table 2, entry 5),[35] 4-chloro-*N*-(3,4-dihydroxybenzylidene)aniline (Table 2, entry 10),[36] 4-methoxy-*N*-(3,4,5-trimethoxybenzylidene)aniline (Table 2, entry 11),[5b] 4-methyl-*N*-(3,4,5-trimethoxybenzylidene)aniline (Table 2, entry 13),[5b] 4-chloro-*N*-(3,4,5-trimethoxybenzylidene)aniline (Table 2, entry 14),[31] 4-nitro-*N*-(3,4,5-trimethoxybenzylidene)aniline (Table 2, entry 15),[37] *N*-(3,4-dimethoxybenzylidene)phenylhydrazine,[38] *N*-(3,4,5-trimethoxybenzylidene)phenylhydrazine,[39] *N*-(1-phenylethylidene)phenylhydrazine[25] were identified by comparison of the spectroscopic (IR, NMR, and MS) data with those of authentic samples.

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