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ULTRASONIC ASSISTED SYNTHESIS OF 2-IMINOCHROMENES CATALYZED BY $H_3PW_{12}O_{40}$ AS AN EFFICIENTAND REUSABLE CATALYST

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Abstract: Efficient and convenient synthesis of *N*-alkyl-2-imino-2*H*-chromene-3-carboxamides by reaction of *N*-alkyl-2-cyanoacetamides with salicylaldehydes using12-tungstophosphoric acid ($H_3PW_{12}O_{40}$) as a green and reusable catalyst under ultrasonic irradiation is described. The catalyst is inexpensive and readily available and could be efficiently used at least five times without substantial reduction in its catalytic activity. High activity of the catalyst, excellent yields, short reaction times, and simple procedure with an easy work-up are other advantages of the present methodology.

Keywords: 2-Iminochromenes, $H_3PW_{12}O_{40}$,Cyanoacetamides,Ultrasonicirradiation

Introduction

The chromenes have been the subject of intense research due to their wide range of pharmaceuticaland biological properties, including antivascular[i], antihypertensive[ii], antifungal[iii], antitumor[iv], analgesic[v], antibacterial[vi], anticonvulsant[vii], and some others. They have also been widely employed as cosmetics[viii], pigments[viii], and potent biodegradable agrochemicals[ix]. A number of compounds with chromene moiety are known as potential inhibitors of acetylcholinesterase[x], butyrylcholinesterase[x], TNF-α[xi],hMAO[xii], human rhinovirus capsid-binding[xiii],aromatase[xiv], aldose reductase[xv], notum pectinacetylesterase[xvi], and dihydrofolatereductase[xvii].Naturally occurring chromenes are used as valuable leads for the design and synthesis of new active analogs with potential therapeutic applications[xviii-xxii].

Because of the aforementioned properties of chromenes, the synthesis of these compounds by new methodologiesis of much importance to organic chemists. Several methods have been reported for the synthesis of 2-aminochromenes including one-pot reaction of salicylaldehydes, malononitrile,and nitroalkanes[xxiii], stepwise condensation of salicylaldehydes with 3 equiv of malononitrile[xxiv],reaction of malononitrile with *in situ* generated sensitive *ortho*-quinonemethides from 2-(aryl-sulfonyl)alkyl phenols[xxv],and one-pot three-component reaction of resorcinol, an aromatic aldehyde, and malononitrile or ethyl cyanoacetate in the presence of various catalysts[xxvi-xxx].However, there are a few reports on

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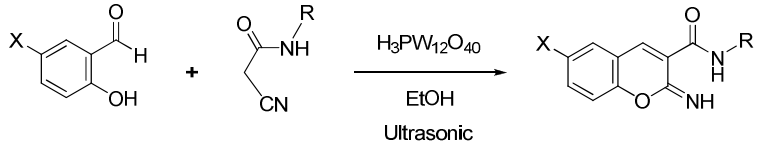
the synthesis of 2-iminochromenes. The classic method for the synthesis of these compounds is the reaction of salicylaldehydes with active methylene compounds initiated by a few catalysts such as piperidine in the presence[xxxi] or absence[xxxii] of microwave irradiation, potassium phthalimide[xxxiii], Na_2CO_3 or $NaHCO_3$[xxxiv], and polyethylene polyamine functionalized polyacrylonitrilefiber[xxxv].Although each of these individual methods has its own merits, many suffer from limitations such as long reaction times, unsatisfactory yields, and the use of relatively expensive catalysts. Thus, the exploration of novel methodologies using new efficient and reusable catalysts is still ongoing.

In recent years, Keggin-type heteropolyacids (HPAs) have attracted rising interest as eco-friendly catalysts in organic transformations due to their advantageous properties, such as environmental compatibility, reusability, non-corrosiveness,and relative lack of disposal problems[xxxvi-xxxviii]. Being stronger acids, HPAs, especially12-tungstophosphoric acid ($H_3PW_{12}O_{40}$), generally exhibit higher catalytic activities than conventional catalysts, such as mineral acids, ion-exchange resins,zeolites, etc.[xxxix]. On the other hand, the $H_3PW_{12}O_{40}$ is highly soluble in water and polar organic solvents, such as lower alcohols and carboxylic acids, and insoluble in hydrocarbons.

Ultrasonic irradiation has increasingly been used as a very significant nontraditional technique for accelerating organic reactions[xl,xli]. Compared with traditional methods, the salient features and benefits of ultrasonic irradiation technique includes reduced reaction times, reduced energy consumption, enhanced selectivity, and improved yields[xlii,xliii]. Often, the reactions under ultrasound irradiation are commonly easier to work up than those in conventional methods[xliv-xlvi].

Considering the above facts and also in extension of our previous studies on the development of new environmental friendly methodologies in the synthesis of organic compounds using reusable catalysts[xlvii-xlxvii], herein we would like to report an efficient procedure for preparation of *N*-alkyl-2-imino-2*H*-chromene-3-carboxamides by Knoevenagel condensation of salicylaldehydes**1** with*N*-alkyl-2-cyanoacetamides**2** followed by cyclization reaction in the presence of $H_3PW_{12}O_{40}$ as reusable catalyst under ultrasonic irradiation (Scheme 1).



1 **2** **3a-g**

Scheme 1.Synthesis of *N*-alkyl-2-imino-2*H*-chromene-3-carboxamides catalyzedby $H_3PW_{12}O_{40}$

Experimental

All chemicals were purchased from Merck and Aldrich and used without purification. Melting points were measured on a Stuart SMP3 melting point apparatus. The ^1H spectra were measured on a Bruker 300 FT spectrometer using TMS as the internal standard.Elemental analysis was performed on a Thermo Finnigan Flash EA microanalyzer.

General procedure for the synthesis of *N*-alkyl-2-imino-2*H*-chromene-3-carboxamides 3a-g catalyzed by $H_3PW_{12}O_{40}$. A mixture of salicylaldehydes**1** (1 mmol), *N*-alkyl-2-cyanoacetamides**2**(1 mmol) and $H_3PW_{12}O_{40}$(10 mol%) in ethanol (5 mL) was sonicated at 60°C for 3-7 min.The reaction was monitored using TLC plates eluting with *n*-hexane/ethyl acetate (volume ratio, 3:2). Upon completion of the transformation, the reaction mixture was cooled to room temperature. This resulted in the precipitation of the product,

which was collected by filtration, washed repeatedly with cold water and recrystallized from ethanol to give products **3a-g**in high yields.

N-Benzyl-2-imino-2*H*-chromene-3-carboxamide (**3a**): ^{1}H NMR (300 MHz, $CDCl_3$): δ4.68 (d, 2H, J = 5.7 Hz, CH_2), 7.14 (d, 1H, J = 8.1 Hz, arom-H), 7.22 (t, 1H, J = 7.5 Hz, arom-H), 7.27-7.55 (m, 7H, arom-H), 7.58 (s br., 1H, NH), 8.53 (s, 1H, CH in pyran ring),10.74 (s br., 1H, NH).Anal.Calcd for $C_{17}H_{14}N_2O_2$: C 73.37, H 5.07, N 10.07, Found: C 73.11, H 5.26,N 10.29.

2-Imino-*N*-phenethyl-2*H*-chromene-3-carboxamide (**3b**): ^{1}H NMR (300 MHz, $CDCl_3$): δ2.96 (t, 2H, J = 7.2 Hz, CH_2), 3.72 (q, 2H, J = 7.2 Hz, CH_2), 7.13 (d, 1H, J = 8.4 Hz, arom-H),7.17-7.52 (m, 8H, arom-H), 7.53 (s br., 1H, NH), 8.48 (s, 1H, CH in pyran ring), 10.40 (s br., 1H, NH).Anal.Calcd for $C_{18}H_{16}N_2O_2$: C 73.95, H 5.52, N 9.58, Found: C 74.21, H 5.69,N 9.37.

2-Imino-*N*-methyl-2*H*-chromene-3-carboxamide(**3c**): ^{1}H NMR (300 MHz, $CDCl_3$): δ3.05 (d, 3H, J = 4.8 Hz, CH_3),7.38-7.46 (m, 2H, arom-H and NH), 7.66-7.76 (m, 3H, arom-H), 8.80 (s br., 1H, NH), 8.95 (s, 1H, CH in pyran ring).Anal. Calcd for $C_{11}H_{10}N_2O_2$: C 65.34, H 4.98, N 13.85, Found: C 65.58, H 4.79,N 13.64.

N-Cyclohexyl-2-imino-2*H*-chromene-3-carboxamide (**3e**): ^{1}H NMR (300 MHz, $CDCl_3$): δ1.25-2.00 (m, 10H, cyclohexyl), 3.90-4.05 (m, 1H, CH in cyclohexyl), 7.10 (d, 1H, J = 8.1 Hz, arom-H), 7.18 (td, 1H, J = 7.5, 0.6 Hz, arom-H),7.40-7.50 (m, 2H, arom-H), 7.57 (s br., 1H, NH), 8.45 (s, 1H, CH in pyran ring), 10.30 (d, 1H,J = 6.6 Hz, NH). Anal.Calcd for $C_{16}H_{18}N_2O_2$: C 71.09, H 6.71, N 10.36, Found: C 70.88, H 6.52, N 10.60.

N-Benzyl-6-bromo-2-imino-2*H*-chromene-3-carboxamide (**3f**): ^{1}H NMR (300 MHz, $CDCl_3$): δ4.67 (d, 2H, J = 5.7 Hz, CH_2), 7.00-7.65 (m, 8H, arom-H), 7.66 (s br., 1H, NH), 8.45 (s, 1H, CH in pyran ring), 10.65 (s br., 1H, NH). Anal.Calcd for $C_{17}H_{13}BrN_2O_2$: C 57.16, H 3.67, N 7.84, Found: C 57.38, H 3.81,N 7.61.

6-Bromo-*N*-cyclohexyl-2-imino-2*H*-chromene-3-carboxamide (**3g**): ^{1}H NMR (300 MHz, $CDCl_3$): δ1.25-2.02 (m, 10H, cyclohexyl), 3.90-4.05 (m, 1H, CH in cyclohexyl), 7.02 (d, 1H, J = 8.7 Hz, arom-H), 7.54 (dd, 1H, J = 8.7, 2.4 Hz, arom-H), 7.63 (d, 1H, J = 2.4 Hz, arom-H),7.66 (s br., 1H, NH), 8.39 (s, 1H, CH in pyran ring), 10.21 (d, 1H, J = 6.9S Hz, NH). Anal.Calcd for $C_{16}H_{17}BrN_2O_2$: C 55.03, H 4.91, N 8.02, Found: C 54.79, H 5.08,N 8.28.

Results and discussion

To search for the optimal conditions, we examined the reaction of salicylaldehyde (1 mmol) and *N*-benzyl-2-cyanoacetamide (1 mmol) as test reaction for the synthesis of *N*-benzyl-2-imino-2*H*-chromene-3-carboxamide **3a** in the absence and presence of $H_3PW_{12}O_{40}$ as catalyst under ultrasonic irradiation.Various parameters such as the amount of catalyst, the effect of solvent, and the influence of temperature were studied and the results are summarized in Table 1.First, a non-catalyzed reaction was tested in H_2O or EtOH at 60 °C, but low yield of the product was formed (entries 1 and 2). On the contrary, we were pleased to see that the reaction was efficiently catalyzed by $H_3PW_{12}O_{40}$. Ethanol proved to be a much better solvent in terms of yield as well as reaction time than all the others.The excellent yield of the product was obtained when the reaction was conducted in EtOHat 60 °C in the presence of 10 mol% of the $H_3PW_{12}O_{40}$ catalyst (entry 14). No significant improvement in yields or reaction times was observed using a higher amount of the catalyst. All subsequent reactions were carried out in these optimized conditions.

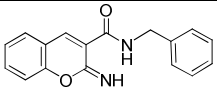
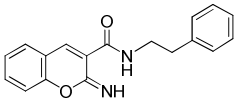
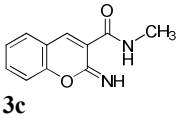
Table 1
Optimization of reaction conditions for synthesis of compound **3a** catalyzed by $H_3PW_{12}O_{40}$[a]

Entry	Catalyst (mol%)	Solvent	T (°C)	Time (min)	Isolated Yield (%)
1	-----	H_2O	60	90	32
2	-----	EtOH	60	90	37
3	5	H_2O	r.t.	45	68
4	5	H_2O	60	25	75
5	5	EtOH	r.t.	35	75
6	5	EtOH	60	15	89
7	5	MeOH	r.t.	42	70
8	5	MeOH	60	28	78
9	5	$CHCl_3$	r.t.	65	63
10	5	$CHCl_3$	60	45	75
11	10	H_2O	r.t.	38	70
12	10	H_2O	60	18	80
13	10	EtOH	r.t.	20	84
14	10	EtOH	60	5	98
15	10	MeOH	r.t.	35	73
16	10	MeOH	60	20	82
17	10	$CHCl_3$	r.t.	52	68
18	10	$CHCl_3$	60	38	79
19	15	H_2O	60	17	80
20	15	EtOH	60	5	97
21	15	MeOH	60	18	84
22	15	$CHCl_3$	60	30	80

[a]Reaction conditions: salicylaldehyde (1 mmol) and *N*-benzyl-2-cyanoacetamide (1 mmol) under ultrasonic irradiation.
[b]Isolated yields.

Thereafter, applicability of the method was evaluated for the synthesis of other *N*-alkyl-2-imino-2*H*-chromene-3-carboxamides using a variety of salicylaldehydes and *N*-alkyl-2-cyanoacetamides.The obtained results are summarized in Table 2.As shown, all reactions proceed very cleanly to give the corresponding *N*-alkyl-2-imino-2*H*-chromene-3-carboxamides**3a-g**in high yields over short reaction times. Melting points, TLC and the ^{1}H NMR spectroscopic data were used to establish that only one product is formed in all cases with no undesirable side-products being present after purification.

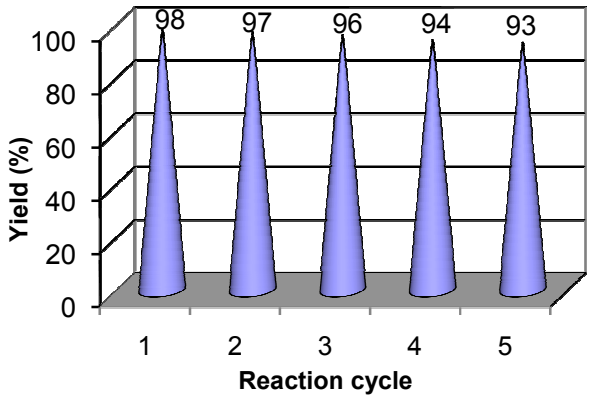
Table 2
Synthesis of *N*-alkyl-2-imino-2*H*-chromene-3-carboxamides **3a-g** using $H_3PW_{12}O_{40}$as catalyst[a]

Entry	X	R	Product	Time (min)	Isolated Yield (%)
1	H	$PhCH_2$	 **3a**	5	98
2	H	$PhCH_2CH_2$	 **3b**	5	95
3	H	CH_3	 **3c**	3	93

4	H	$CH_3(CH_2)_2CH_2$		7	92
			3d		
5	H	Cyclohexyl		5	95
			3e		
6	Br	$PhCH_2$		3	95
			3f		
7	Br	Cyclohexyl		3	90
			3g		

[a]Reaction conditions: salicylaldehydes**1** (1 mmol), *N*-alkyl-2-cyanoacetamides**2**(1 mmol), $H_3PW_{12}O_{40}$ (10 mol%), EtOH, ultrasonic irradiation,60 °C.

Because of importance of recyclability and reusability of catalysts for commercial applications, the recovery and catalytic activity of recycled $H_3PW_{12}O_{40}$was explored in the synthesis of compound **3a**. After completion of the reaction, the reaction mixture was cooled to room temperature, the product was collected by filtration, and washed repeatedly with cold water. The combined filtrate was evaporated to dryness under reduced pressure. The solid catalyst was collected, dried at 60 °C under vacuum for 2 h and reused for the same experiment. The catalyst could be used at least five times with little loss of activity (Fig. 1). This reusability demonstrates the high stability and turnover of $H_3PW_{12}O_{40}$under the employed conditions.



Conclusion

In summary, we have found that $H_3PW_{12}O_{40}$ can be used as a new, reusable and efficient catalyst for the preparation of a variety of *N*-alkyl-2-imino-2*H*-chromene-3-carboxamides by reaction of salicylaldehydes and *N*-alkyl-2-cyanoacetamides.The reaction occur in ethanol at 60 °C and furnishes the expected products in high yields over short reaction times. Other attractive features of this protocol are environmentally friendly method, simple procedure, easy work-up, high catalytic activity and recyclability and reusability of the catalyst.

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