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Catalyst-free Preparation of 4*H*-1,3,4-Oxadiazines in H_2O

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This report provides a description of a novel and efficient procedure for the synthesis of substituted 4*H*-1,3,4-oxadiazines *via* reaction of various vinyl cyanides and acetohydrazide in H_2O under catalyst-free conditions. The remarkable advantages are the simplicity of the experimental procedures, high yields, and avoidance of organic solvents. Products were characterized by melting point, IR, ^1H and ^{13}C NMR spectra, and elemental analysis. The desired products were obtained in >85% yields.

Keywords: Acetohydrazide, Catalyst-free, Cyclization, Oxadiazines, Vinyl cyanides, Aqueous

INTRODUCTION

Cyclization has been widely used to construct carbocyclic or heterocyclic organic compounds [1-4]. Heterocyclic compounds containing five- or six-membered rings are successfully used as medicines [5-7]. Among heterocyclic compounds, nitrogen-containing heterocycles have attracted a great attention because of their unique chemical and physical properties [8]. Synthesis of azaheterocyclic compounds is of considerable interest owing to the wide-ranging biological activity of this series of compounds as medicinal and pesticidal agents. Moreover, most of them are extremely versatile building blocks for the manufacture of the bioactive compounds in pharmaceutical drug design and agrochemical industry [9-11]. Six-membered heterocyclic rings with three hetero atoms are not found broadly in nature, but they have superfine use in the most important fields. Thus, developing new heterocyclic compounds as medicinals are still an important region of interest in the life science [12]. Oxadiazines are an important category of heterocyclic compounds which they are considered in biological systems. They are oxa analogs of nucleosides-6-oxadihydro uracil and display important application in biological systems [13-15]. A diversity of biological effects is associated with oxadiazines bearing heteroatoms at 1,3,4 positions [16]. Given such intrinsic merits, various synthetic methods have been reported for the synthesis of oxadiazines such as condensation reaction of cyanoacetylhydrazine with ω-bromo(4-methyl) acetophenone [5], reaction of acetophenones with acid hydrazide in the presence of PhI(OH)OTs [17], condensation reaction of cyanoacetylhydrazine with ω-bromoacetophenone [18], [2+4] cycloaddition of allenoates with N-acyldiazenes [19], [3+3] cycloaddition of aza-oxyallylic cations with nitrile oxides [20], three-component reaction of cyclohexyl isocyanide, hydrazides and cyclic ketones in the presence of SBA-Pr-SO_3H [21], dehydrosulfurization of the 4-chloro-N-(2,2,2-trichloro-1-isothiocyanatoethyl)benzamide under the action of dicyclohexylcarbodiimide [22], and by the cyclocondensation of phenylhydrazones catalyzed by triethylamine [23]. Though these methods are quite satisfactory in many instances, most of them are associated with limitations such as harsh reaction conditions, application of toxic organic solvents as reaction media, and long reaction times. Accordingly, due to the biological importance of these kinds of heterocycles, an efficient, environmentally clean, commercially available, and cheap synthetic method is still a need. To the best of our knowledge, there have been no reports on the reaction of vinyl cyanides with acetohydrazide for the preparation of 4*H*-1,3,4-oxadiazine derivatives. In this work, we report a

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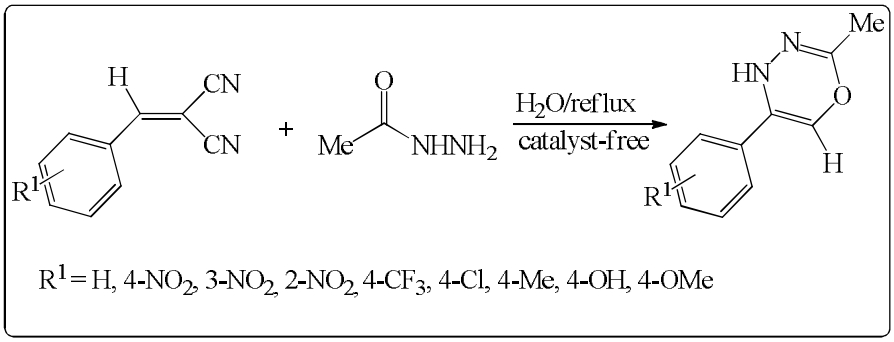


Scheme 1. Synthesis of 4*H*-1,3,4-oxadiazine derivatives

simple, efficient, cost-effective, and inexpensive method for the preparation of 2-methyl-5-aryl-4*H*-1,3,4-oxadiazine under catalyst-free conditions at the reflux temperature in H_2O (Scheme 1).

EXPERIMENTAL

General Procedure

A mixture of vinyl cyanides (1 mmol), and acetohydrazide (0.074 g, 1 mmol) was stirred in H_2O (2 ml) at reflux temperature for desired time (Table 2). The completion of the reaction was monitored by TLC. After cooling, the reaction mixture was poured onto crushed ice (10 g). The resulting precipitate was filtered under suction and then recrystallized from EtOH to afford the pure product.

2-Methyl-5-phenyl-4*H*-1,3,4-oxadiazine. Yield 160 mg (92%) as an withe crystal. m.p.: 139-142 °C; ^{1}H NMR spectrum (500 MHz, $CDCl_3$), δ, ppm: 2.50 (s, 3H, CH_3), 7.35-7.45 (m, 3H, Ar), 7.65-7.75 (m, 2H, Ar), 7.80 (s, 1H, H-6), 10.40 (s, 1H, NH). ^{13}C NMR spectrum (125 MHz, $CDCl_3$), δ, ppm: 20.40, 127.10, 128.70, 130.00, 133.90, 143.10, 166.10, 174.40. IR (neat), ῡ, cm^{-1}: 3188 (N-H), 3081 (=C-H), 2974 (C-H), 1672 (C=N), 1604 and 1465 (C=C), 1339 (C-N), 1134 (C-O). Anal. Calcd. Found, %: C, 68.6; H, 5.9; N, 16.3. $C_{10}H_{10}N_2O$. Calculated, %: C, 68.95; H, 5.79; N, 16.08.

2-Methyl-5-(4-nitrophenyl)-4*H*-1,3,4-oxadiazine. Yield 208 mg (95%) as an withe crystal. m.p.: 203-206 °C; ^{1}H NMR spectrum (500 MHz, $CDCl_3$), δ, ppm: 2.50 (s, 3H, CH_3), 7.90 (d, J = 8.8 Hz, 2H, Ar-H), 8.00 (s, 1H, H-6), 8.40 (d, J = 8.8 Hz, 2H, Ar-H), 10.00 (s, 1H, NH). ^{13}C NMR spectrum (125 MHz, DMSO-d_6), δ, ppm: 20.65, 124.44, 127.97, 140.60, 143.59, 148.03, 166.44, 172.76. IR (neat), ῡ, cm^{-1}: 3180 (N-H), 3080 (=C-H), 2954 (C-H), 1679 (C=N), 1581 (C=C), 1519 and 1387 (NO_2), 1333 (N-H), 1154 (C-O). Anal. Calcd. Found, %: C, 55.0; H, 3.9; N, 19.1. $C_{10}H_9N_3O_3$. Calculated, %: C, 54.79; H, 4.14; N, 19.17.

2-Methyl-5-(3-nitrophenyl)-4*H*-1,3,4-oxadiazine. Yield 208 mg (95%) as an yellow crystal. m.p. 240-244 °C; ^{1}H NMR spectrum (500 MHz, $CDCl_3$+co-solvent DMSO-d_6), δ, ppm: 2.70 (s, 3H, CH_3) 7.20-7.25 (m, 1H, Ar-H), 7.60-7.85 (m, 3H, Ar-H), 8.00 (s, 1H, H-6), 10.84 (s, 1H, NH). ^{13}C NMR spectrum (125 MHz, DMSO-d_6), δ, ppm: 20.66, 124.23, 130.77, 133.14, 136.63, 140.63, 143.64, 148.69, 166.38, 172.61. IR (neat), ῡ, cm^{-1}: 3183 (N-H), 3094 (=C-H), 2968 (C-H), 1681 (C=N), 1644 (C=C), 1522 and 1388 (NO_2), 1333 (C-N), 1136 (C-O). Anal. Calcd. Found, %: C, 54.8; H, 3.8; N, 19.4. $C_{10}H_9N_3O_3$. Calculated, %: C, 54.79; H, 4.14; N, 19.17.

2-Methyl-5-(2-nitrophenyl)-4*H*-1,3,4-oxadiazine. Yield 190 mg (87%) as an yellow crystal. m.p.: 240-244 °C; ^{1}H NMR spectrum (500 MHz, $CDCl_3$+co-solvent DMSO-d_6), δ, ppm: 2.75 (s, 3H, CH_3) 7.25-7.70 (m, 4H, Ar-H), 8.20 (s, 1H, H-6), 10.95 (s, 1H, NH). ^{13}C NMR spectrum (125 MHz, DMSO-d_6), δ, ppm: 20.64, 124.89, 128.54, 130.68, 133.82, 138.19, 141.43, 148.45, 166.36, 172.67. IR (neat), ῡ, cm^{-1}: 3183 (N-H), 3094 (=C-H), 2968 (C-H), 1681 (C=N), 1644 (C=C), 1522 and 1388 (NO_2), 1333 (N-H), 1136 (C-O). Anal. Calcd. Found, %: C, 54.6; H, 3.9; N, 19.0. $C_{10}H_9N_3O_3$. Calculated, %: C, 54.79; H, 4.14; N,

19.17.

2-Methyl-5-(4-(trifluoromethyl)phenyl)-4*H*-1,3,4-oxadiazine. Yield 230 mg (95%) as an withe crystal. m.p.: 143-146 °C; ^{1}H NMR spectrum (500 MHz, $CDCl_3$), δ, ppm: 2.44 (s, 3H, CH_3), 7.65 (d, *J* = 8.0 Hz, 2H, Ar-H), 7.80 (d, *J* = 8.0 Hz, 2H, Ar-H), 8.00 (s, 1H, H-6), 10.80 (s, 1H, NH). ^{13}C NMR spectrum (125 MHz, DMSO-*d6*), δ, ppm: 20.65, (121.07, 123.46, 125.63, 127.75), 126.07, 127.61, 127.93, 138.70, 141.23, 166.31, 172.63 ppm; IR (neat), ῡ, cm^{-1}: 3192 (N-H), 3080 (=C-H), 2961 (C-H), 1681 (C=N), 1604 (C=C), 1519 and 1394 (NO_2),1323 C-N), 1286 (C-O). Anal. Calcd. Found, %: C, 54.8; H, 3.9; N, 11.5. $C_{11}H_9F_3N_2O$. Calculated, %: C, 54.55; H, 3.75; N, 11.57.

5-(4-Chlorophenyl)-2-methyl-4*H*-1,3,4-oxadiazine. Yield 188 mg (90%) as an withe crystal. m.p.: 153-154 °C; ^{1}H NMR spectrum (500 MHz, $CDCl_3$), δ, ppm: 2.44 (s, 3H, CH_3), 7.36 (d, *J* = 8.0 Hz, 2H, Ar-H), 7.60 (d, *J* = 8.0 Hz, 2H, Ar-H), 7.82 (s, 1H, H-6), 10.60 (s, 1H, NH); ^{13}C NMR spectrum (125 MHz, DMSO-d_6), δ, ppm: 20.65, 128.66, 133.68, 134.47, 141.61, 144.70, 166.10, 172.43. IR (neat): ῡ, cm^{-1}: 3192 (N-H), 3080 (=C-H), 2961 (=C-H), 1681 (C=N), 1604 and 1394 (C=C), 1323 (C-N), 1134 (C-O). Anal. Calcd. Found, %: C, 57.2; H, 4.6; N, 13.7. $C_{10}H_9ClN_2O$. Calculated, %: C, 57.57; H, 4.35; N, 13.43.

2-Methyl-5-(p-tolyl)-4*H*-1,3,4-oxadiazine. Yield 173 mg (92%) as an withe crystal. m.p.: 125-129 °C; ^{1}H NMR spectrum (500 MHz, $CDCl_3$), δ, ppm: 2.38 (s, 3H, CH_3), 2.39 (s, 3H, CH_3), 7.20 (d, *J* = 8.0 Hz, 2H, Ar-H), 7.57 (d, *J* = 7.5 Hz, 2H, Ar-H), 7.84 (s, 1H, H-6), 10.50 (s, 1H, NH); ^{13}C NMR spectrum (125 MHz, DMSO- d_6), δ, ppm: 20.67, 21.41, 127, 129.85, 132.04, 139.79, 143.01,165.88, 172.24. IR (neat), ῡ, cm^{-1}: 3184 (N-H), 3087 (=C-H), 2973 (C-H), 1667 (C=N), 1600 and 1506 (C=C), 1393 (C-N), 1133 (C-O) Anal. Calcd. Found, %: C, 69.9; H, 6.7; N, 14.9. $C_{11}H_{12}N_2O$. Calculated, %: C, 70.19; H, 6.43; N, 14.88.

4-(2-Methyl-4*H*-1,3,4-oxadiazin-5-yl)phenol. Yield 171 mg (90%) as an withe crystal. m.p.: 249-252 °C; ^{1}H NMR spectrum (500 MHz, $CDCl_3$+co-solvent DMSO-d_6), δ, ppm: 2.68 (s, 3H, CH_3), 6.21 (broad, 2H, Ar-H), 6.89 (d, *J* = 7.5 Hz 2H, Ar-H), 8.87 (s, 1H, H-6), 10.20 (s, 1H, NH). ^{13}C NMR spectrum (125 MHz, DMSO-d_6), δ, ppm: 20.67, 116.10, 126.80, 128.72, 143.26, 145.90, 159.45, 172.98. IR (neat), ῡ, cm^{-1}: 3183 (N-H), 3087 (=C-H), 2968 (C-H), 1670 (C=N), 1606 and 1508 (C=C), 1348 (C-N), 1169(C-O). Anal. Calcd. Found, %: C, 63.4; H, 5.6; N, 14.6. $C_{10}H_{10}N_2O_2$. Calculated, %: C, 63.15; H, 5.30; N, 14.73.

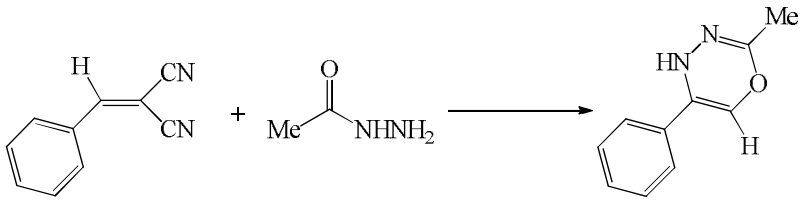
5-(4-Methoxyphenyl)-2-methyl-4*H*-1,3,4-oxadiazine. Yield 188 mg (92%) as an light yellow crystal. m.p.: 128-131 °C; ^{1}H NMR spectrum (500 MHz, $CDCl_3$), δ, ppm: 2.35 (s, 3H, CH_3), 3.82 (s, 3H, CH_3), 6.91 (d, *J* = 8.0 Hz, 2H, Ar-H), 7.61 (d, *J* = 7.4, Hz 2H, Ar-H), 7.79 (s, 1H, H-6), 10.38 (s, 1H, NH); ^{13}C NMR spectrum (125 MHz, DMSO-d_6), δ, ppm: 20.69, 55.67, 114.69, 127.33, 128.59, 142.80, 160.89, 165.76, 172.13 ppm; IR (neat), ῡ, cm^{-1}: 3183 (N-H), 3087 (=C-H), 2968 (C-H), 1670 (C=N), 1606 and 1508 (C=C), 1390 (C-N), 1169 (C-O). Anal. Calcd. Found, %: C, 64.6; H, 5.7; N, 13.8. $C_{11}H_{12}N_2O_2$. Calculated, %: C, 64.69; H, 5.92; N 13.90.

RESULTS AND DISCUSSIONS

The 4*H*-1,3,4-oxadiazines were synthesized by the reaction of acetohydrazide and various vinyl cyanides under catalyst-free conditions. For optimizing the experimental conditions, the reaction between acetohydrazide and 2-benzylidenemalononitrile was considered as a model reaction. Initially, the effect of solvent on the reaction was studied (Table 1, entries 1-6) and water was found to be the best one. Next, the influence of the reaction time on the yield was investigated (Table 1, entries 7, 8). It was clear that the highest yield was produced when the reaction time was 25 min, although the yield did not improve to any greater extent when the reaction time was increased from 25 min to 30 min. We therefore chose 25 min as the reaction time. Finally, to find the optimum temperature, the reaction was conducted at 20, 60 °C, and at reflux temperature, which resulted in the isolation of the product in a trace amount, 50 and 92% yields, respectively (Table 1, entries 2, 9,10). Hence, the best condition employs the 1:1 mol ratio of acetohydrazide and 2-benzylidenemalononitrile at the reflux temperature over 25 min using water as a solvent.

The synthesized oxadiazine was characterized by FT-IR, ^{1}H, and ^{13}C NMR spectra, m.p. and elemental analyses. The FT-IR spectrum of 2-methyl-5-phenyl-4*H*-1,3,4-oxadiazine showed a band for stretching vibration of N-H at 3188 cm^{-1}. The appearance signals at 3081, 2974 and1672 cm^{-1} were assigned respectively to the stretching vibration of aromatic

Table 1. Optimization of the Reaction Conditions for the Synthesis of 2-Methyl-5-phenyl-4*H*-1,3,4-oxadiazine[a]



Entry	Solvent	Time (min)	Temp (°C)	Yield (%)
1	Solvent-free	25	Crushing	85
2	H_2O	25	Ref.	92
3	EtOH	25	Ref.	92
4	H_2O:EtOH (1:1)	25	Ref.	60
5	MeCN	25	Ref.	55
6	AcOEt	25	Ref.	40
7	H_2O	20	Ref.	90
8	H_2O	30	Ref.	92
9	H_2O	25	20	Trace
10	H_2O	25	60	50

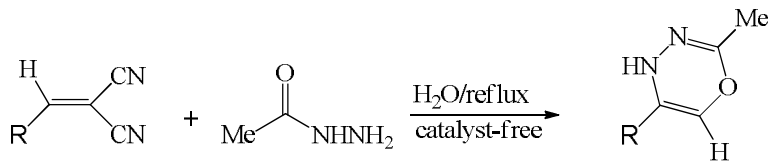
[a]2-Benzylidenemalononitrile (1 mmol), acetohydrazide (1 mmol).

C-H groups, methyl C-H groups, and the C=N bond of oxadiazine ring. The peaks at 1604, and 1465 cm^{-1} were attributed to vibrational modes of C=C stretching vibration of the phenyl ring. The signals at 1339 and 1134 cm^{-1} were also assigned to the stretching vibration of C-N and C-O groups, respectively.

The ^{1}H NMR spectrum of 2-methyl-5-phenyl-4*H*-1,3,4-oxadiazine exhibited a sharp singlet at δ = 2.45 ppm for CH_3 group on the oxadiazine ring and a multiplet signal at δ = 7.43-7.69 ppm for the 5 H atoms of the phenyl ring. The peaks at δ = 10.36 and δ = 7.88 ppm were assigned to the N-H and C-H groups of the oxadiazine ring, respectively. The ^{13}C NMR spectrum of 2-methyl-5-phenyl-4*H*-1,3,4-oxadiazine showed a signal at 20.0 ppm for the CH_3 group. The signal at 144.0 ppm attributed to the carbon atom of the oxadiazine ring. The carbon atom of the N=C-O group was recorded at 174.5 ppm, and finally, the peaks at 126.0-134.4 ppm were also assigned to the carbon atoms of the phenyl ring. The elemental analysis of 2-methyl-5-phenyl-4*H*-1,3,4-oxadiazine showed the existing of carbon (68.6%), hydrogen (5.9%), and nitrogen (16.3%), confirming the synthesis of the product. The melting point of 2-methyl-5-phenyl-4*H*-1,3,4-oxadiazine was also recorded (Table 2, entry 1).

To explore the generality and scope of this method, a wide variety of vinyl cianides and acetohydrazide were reacted under the optimized experimental conditions to afford the corresponding 4*H*-1,3,4-oxadiazines in excellent

Table 2. Reaction of Vinyl Cyanides with Acetohydrazide under Catalyst-free Conditions[a]



Entry	R	Time (min)	Yield (%)	M.p.
1	C_6H_5	25	92	139-142
2	4-$NO_2C_6H_4$	15	95	203-206
3	3-$NO_2C_6H_4$	20	95	240-244
4	2-$NO_2C_6H_4$	25	87	210-213
5	4-CF_3 C_6H_4	15	95	143-146
6	4-Cl C_6H_4	25	90	153-154
7	4-Me C_6H_4	100	92	125-129
8	4-OH C_6H_4	120	90	249-252
9	4-OMe C_6H_4	115	92	128-131

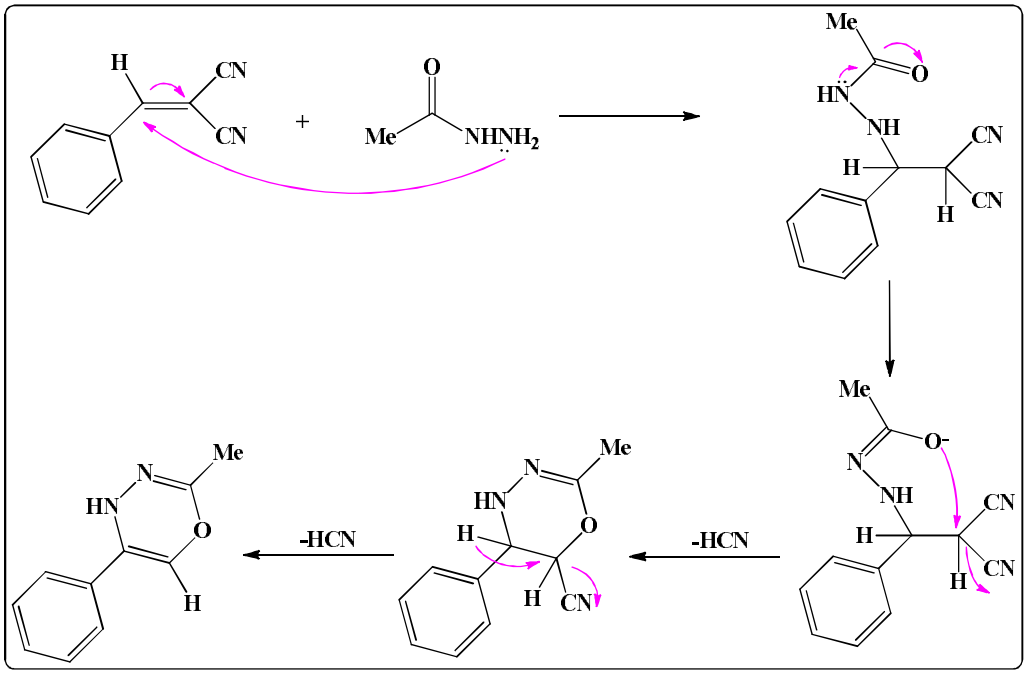
[a]Reaction conditions: vinyl cyanide (1 mmol), acetohydrazide (1 mmol), H_2O (2 ml), reflux temperature.

yields (Table 2). The results revealed that the reaction for the (arylmethylidene)malononitriles bearing electron-withdrawing substituent such as NO_2 and CF_3 increased the rate of reaction (Table 2, entries 2-5). Aldehydes such as 4-methybenzaldeyde, 4-hydroxybenzaldeyde, and 4-methoxybenzaldeyde are well tolerated and afforded excellent product yields (Table 2, entries 7-9). All the products were characterized by m.p. elemental analysis, FT-IR, ^1H and ^{13}C NMR spectra. No reaction occurred between 2-cyano-3-phenylacrylamide, ethyl 3-phenyl-2-cyanoacrylate or methyl 3-phenyl-2-cyanoacrylate, and acetohydrazide under optimized conditions (Table 2, entries 10-12).

Although the mechanism of the reaction has not been established experimentally, the formation of the product can be rationalized as outlined in Scheme 2. In order to assess the efficiency and generality of this methodology, we compared the results of this method with some reported results in the literature. By comparing our results with the results depicted in Table 3, it was found that 4*H*-1,3,4-oxadiazine can be synthesized in a shorter time in aqueous media.

CUNCLUSIONS

In summary, we have developed an improved and convenient procedure for the synthesis of 4*H*-1,3,4-oxadiazine through the reaction of (arylmethylidene)



Scheme 2. The proposed mechanism for the synthesis of 4*H*-1,3,4-oxadiazines

Table 3. Comparing Different Methods for the Synthesis of 1,3,4-Oxadiazines

Entry	Catalyst/solvent/temp (°C)	Time	Yield (%)	Ref.
1	PPA/AcOH/140	6-8 h	60	1
2	HBr/AcOH/r. t.	18 h	95	2
3	PhI(OH)OTs/MeCN/reflux	6 h	74	17
4	-/1,4-dioxan/reflux	4 h	81	18
5	SBA-Pr-SO_3H/solvent-free/r.t.	2 h	90	21
6	catalyst-free/H_2O/reflux	25 min	92	This work

malononitrile and acetohydrazide under catalyst-free conditions.

The remarkable advantages of this method are simple experimental procedure, appropriate reaction times, high yields, and the ease of product isolation.

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REFERENCES

[1] K. Suhasini, T.V.P. Rao, V. Thirupathaiah, Curr. Sci. 52 (1983) 1133.

[2] D.L. Trepanier, V. Spracmanis, D.S. Tharpe, P.E. Krieger, J. Heterocyclic Chem. 2 (1965) 403.

[3] H. Emtiazi, M.A. Amrollahi, Helv. Chim. Acta 96 (2013) 2196.

[4] A.R. Kartirizky, C.W. Rees, Comprehensive Heterocyclic Chemistry, Pergamon Press, New York, 1984.

[5] R.M. Mohareb, A.A. Mohamed, Molecules 15 (2010) 3602.

[6] R.P. Robinson, K.M. Donahue, J. Heterocycl. Chem. 31 (1994) 1541.

[7] T. Naito, Chem. Pharm. Bull. 56 (2008) 1367.

[8] N.R. Candeias, L.C. Branco, P.M. Gois, C.A.M. Afonso, A.F. Trindade, Chem. Rev. 109 (2009) 2703.

[9] M.J. Kornet, J. Heterocycl. Chem. 33 (1996) 2047.

[10] P.T. Berkowitz, R.A. Long, P.R.K. Dea Robins, T.R. Mathews, J. Med. Chem. 20 (1977) 134.

[11] K.M. Khan, S. Rahat, M.I. Choudhary, A. Rahman, U. Ghani, S. Perveen, S. Khatoon, A. Dar, A. Malik, Helv. Chim. Acta. 85 (2002) 559.

[12] A.R. Shaikh, M. Farooqui, R.H. Satpute, S. Abed, J. Drug Deliv. 8 (2018) 424.

[13] B. Monica, K. Sandra, G. Magdalena, Z. Branca, Acta Pharm. 53 (2003) 175.

[14] F.M. Eissa, J. Chin. Chem. Soc. 56 (2009) 843.

[15] S. Ke, X. Cao, Y. Liang, K. Wang, Z. Yang, Mini-Rev. Med Chem. 11 (2011) 642.

[16] M.H. Abdellattif, O.A. Ali, M.H. Arief, M.A. Hussien, Curr. Org. Synth. 23 (2017) 369.

[17] N.K. Nandkishore, J.M. Kondre, S.V. Gamapawar, S.V. Shinde, Synth. Commun. 39 (2009) 1.

[18] R.M. Mohareba, Schatz, J. Bioorg. Med. Chem. 19 (2011) 2707.

[19] Q. Zhang, L.G. Meng, J. Zhang, L. Wang, Org. Lett. 17 (2015) 3272.

[20] G. Wang, R. Chen, S. Zhao, L. Yang, H. Guo, S. Sun, J. Wang, J. Domena, Y. Xing, Tetrahedron Lett. 59 (2018).

[21] V. Fathi Vavsari, G. Mohammadi Ziarani, S. Balalaie, A. Badiei, F. Golmohammadi, S. Ramezanpour, F. Rominger, Chem. Select 2 (2017) 3496.

[22] P.V. Zadorozhnii, I.O. Pokotylo, V.V. Kiselev, A.V. Kharchenko, O.V. Okhtina, Res. J. Pharm. Biol. Chem. Sci. 10 (2019) 1508.

[23] P. Gurunanjappa, A.K. Kariyappa, Asian J. Chem. 29 (2017) 1687.