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REGULAR ARTICLE

H_3PO_4 catalyzed one-pot synthesis of 1,3-diphenyl-1H-pyrazole-4-carbaldehyde to novel 1,3-diphenyl-1H-pyrazole-4-carbonitrile

TUKARAM S CHOUDHARE[a], DEVENDRA S WAGARE[b], SAGAR E SHIRSATH[c] and PRASHANT D NETANKAR[a,*]

[a]Department of Chemistry, Maulana Azad College, Aurangabad 431 004, India
[b]Department of Chemistry, Vivekanand Arts, Sardar Dalipsingh Commerce and Science College, Auranagbad 43002, India
[c]School of Materials Science and Engineering, University of New South Wales, Sydney, New South Wales 2052, Australia
E-mail: pdnchemi@gmail.com

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Abstract. One-pot condensation of pyrazole-4-aldehydes and hydroxylamine hydrochloride to form the corresponding oxime using formic acid as a medium and further dehydration of oxime using a catalytic amount of orthophosphoric acid to afford novel pyrazole-4-carbonitrile. This protocol serves as an ortho-phosphoric acid-catalyzed one-pot conversion of aldehyde to nitrile. Most remarkable features of this method are metal-free, cost-effective, atom efficiency with excellent yield (98–99%). This process will serve as a robust and scalable tool for the synthesis of valuable and versatile precursor (nitriles). This precursor will pave the way for the synthesis of various medicinally important valuable compounds.

Keywords. Pyrazole-4-carbaldehyde; pyrazole-4-carbonitrile; H_3PO_4; formic acid.

1. Introduction

The compounds bearing nitrile functional found to be active pharmacophores. Recently, pharmaceutical has trended in recognizing the roles of nitrile in medical agents has increased. The increasing number of structural advances of nitrile-containing agents offer insight into the binding of small molecule inhibitors.[1,2] Nitrile group serve as a versatile precursor for the synthesis of various functional groups such as carbonyl compounds (carboxylic acids, aldehydes, ketones, amides, etc.), alcohols, amines and important heterocycles (purines, pyrimidines, pyrazines imidazoles, thiazoles, biphenylene, etc. Moreover, it could be extensively utilized in the synthesis of agrochemicals, active pharmaceutical ingredient's, dyes, synthetic rubbers, herbicides and polymers.[3–5] Synthesis of cyanides reported by well-celebrated methods such as sandmayer reactions, Rosenmund−von Braun reaction, cyanide-halide exchange reactions, transoximination, cleavage of cyanide anion, aerobic oxidation of aldehyde using Nitroxyl/NOx catalyst system, used inorganic reagents such as $NH_2OH/Na_2CO_3/SO_2F_2$ in DMSO, etc. Despite the advantages of the above-reported methods; most of them suffered from serious limitations such as involvement of highly toxic reagents (e.g., NaCN, KCN, $Zn(CN)_2$, or CuCN), use of poisonous metal and catalyst, the release of halide by-products, harsh condition and tedious and laborious work-up procedure which violate the green chemistry protocol.[6–11]

Recently, a variety of one-pot cascade processes described the direct conversion of aldehydes to nitriles using hydroxylamine but in most of them used $CuCl_2/NaOMe/O_2$,[12] $Pb(OAc)_4$,[13] oxone,[14] H_2O_2,[15] I_2,[16] NBS,[17] IBX[18] and $NaICl_2$,[19] O-Benzoyl Hydroxylamine (BHA),[20] often suffered from one or more disadvantages such as non-selective, low yields, a multistep synthesis which increases the expenditure of time and efforts. These methods include the use of expensive, hazardous catalyst and volatile solvents. Some methods used NH_2-OSO_3H and O-(4-CF_3-

*For correspondence

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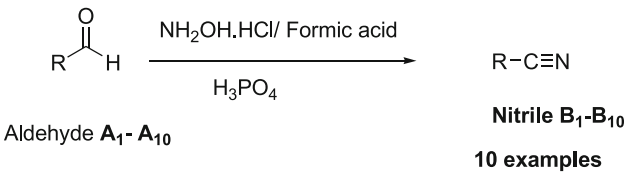
benzoyl)-hydroxylamine (CF_3-BHA) as the nitrogen source and acetic acid but it requires more time, low yield and expensive than the present method.[21-22]

The exploration of mild, rapid, one-pot inexpensive protocol for expanding the conversion of aldehyde to nitriles is thus highly desirable. Herein, the reported protocol meets all the requirements of green chemistry such as a one-pot method to increase atom and step efficiency, metal-free and the use of easily available and cost-effective catalyst. Herein, we have described the transformation of numerous aliphatic, alicyclic and aromatic aldehydes to corresponding nitriles using NH_2OH as a nitrogen source in formic acid as a medium followed by dehydration by 1 mol% H_3PO_4 (Schemes 1, 2).

2. Experimental

All the chemicals were purchased from Sigma Aldrich and used as received without further purification. All compounds were matched with and confirmed by literature data for Melting point, IR, ^{1}H-NMR, ^{13}C-NMR and Mass Spectrometry. The melting points were determined on Labstar melting point apparatus and are uncorrected. The IR spectra were taken on a Perkin-Elmer FTIR-1600 spectrophotometer and the data expressed in cm^{-1} (KBr). ^{1}H and ^{13}C NMR spectra were recorded on Bruker Avance (300 MHz) spectrometer in $CDCl_3$ using TMS as the internal standard. Mass spectra were recorded on an Agilent spectrometer.



Scheme 1. H_3PO_4 catalyzed synthesis of nitrile derivatives.

2.1 *Procedure for the synthesis of 3-Aryl-1-phenyl-1H-pyrazole-4-carbaldehydes (C_1–C_{10})*

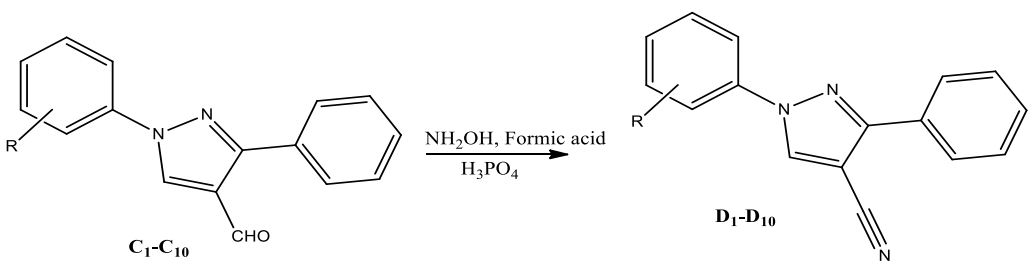
0.006 m (9.2 gm) $PoCl_3$ was added dropwise to a cold solution of hydrazone (2) in 8 mL DMF solution with continuous stirring at 0–5 °C. Afterwards, reaction mass was irradiated under microwave for about 4 min at regular time intervals of 15 s each. Reaction mixture poured on crushed ice to an obtained crude product which was recrystallized by DMF.

2.2 *General procedure for the preparation of nitrile derivatives (B_1–B_{10})*

A mixture of aldehyde (1 mmol), formic acid (10Vol) was cooled to 5–10 °C and hydroxylamine hydrochloride (1 mmol) was added. After 1 min orthophosphoric acid (1 mmol) was added and the reaction mixture was heated up to 100 °C. The reaction was monitored by TLC in MDC: Methanol (9:1) as a mobile phase. The reaction mixture was cooled and poured in 10 mL ice-water and the precipitated solid was filtered out to give desired crude product. The crude product was recrystallized with ethanol to get pure nitrile. The products were characterized by IR, ^{1}H NMR, ^{13}C NMR spectral techniques.

2.3 *General procedure for the preparation of nitrile derivatives (D_1–$_{10}$)*

A mixture of pyrazole-4-carbaldehyde (1 mmol), formic acid (2.5 mL) was cooled to 5–10 °C and hydroxylamine hydrochloride (1 mmol) was added. After 1 min, orthophosphoric acid (1 mol%) was added to the reaction mixture and heat about 100 °C till the completion of the reaction, monitored by TLC in DCM: Methanol (9:1) as a mobile phase.



Scheme 2. Transformation of pyrazole carbaldehydes to novel pyrazole nitriles.

Table 1. Solvent optimization for one-pot synthesis of nitrile[a].

Entry	H_3PO_4 Catalyst mole %	Solvent	Condition	Time (hrs)	Yield (%)[b]
1	1%	Acetic acid	100 °C	7	90
2	1%	Acetic acid : Formic acid (1:1)	100 °C	9	85
3	1%	Acetic acid : Formic acid (2:1)	100 °C	7	88
4	1%	Formic acid : Water (1:1)	100 °C	12	88
5	**1%**	**Formic acid**	**100 °C**	**5**	**99**

[a]Experimental conditions: Benzaldehyde (1 mmol), NH_2OH. HCl (1 mmol), H_3PO_4 (1 mmol).

[b]Isolated yield.

[c]Bold signifies the optimized and suitable catalyst and solvent for the synthesis of nitriles.

The reaction mixture was cooled and poured in 10 mL ice-cold water and the precipitated solid was recrystallized with ethanol to get pure nitrile. The products were analyzed by IR, ^1H, ^{13}C NMR and Mass spectral techniques.

2.4 *Spectroscopic data*

2.4a 4-(dimethyl amino) benzonitrile: (B_3): Scale (151.3 mg, 1 mmol); ethyl acetate: petroleum ether = 1:20; 93% yield (135.8 mg); off-white crystalline powder; ^1H NMR (500 MHz, $CDCl_3$) δ 7.47 (d, $J = 9.0$ Hz, 2H), 6.64 (d, $J = 9.0$ Hz, 2H), 3.04 (s, 6H); ^{13}C NMR (125 MHz, $CDCl_3$) δ 152.52, 133.32, 120.60, 111.43, 97.44, 39.85; HRMS (ESI) m/z calcd. For $C_9H_{11}N_2$ [M+H]+: 147.0922, found: 147.0920.

2.4b Methoxybenzonitrile: (B_5): Purification by chromatography on SiO_2 (10% MTBE/hexanes) provided 7 (400 mg, 78% yield) as a colorless oil.2 1H NMR (400 MHz, $CDCl_3$) 7.51 (m, 2H) 6.89 (m, 2H), 3.79 (s, 3H); ^{13}C NMR (100 MHz, $CDCl_3$) 162.8, 133.9, 119.2, 114.7, 103.7, 55.5.

2.4c Tetradecanenitrile: (B10): 214.2 mg, 1 mmol); dichloromethane: petroleum ether = 1:10; 90% yield (188.2 mg); colourless oil; 1H NMR (400 MHz, CDCl3) δ 2.33 (t, $J = 7.2$ Hz, 2H), 1.68– 1.57 (m, 2H), 1.44 (s, 2H), 1.26–1.20 (m, 18H), 0.88 (t, $J = 6.9$ Hz, 3H); ^{13}C NMR (100 MHz, CDCl3) δ 119.94, 32.03, 29.74, 29.70, 29.61, 29.45, 29.42, 28.88, 28.78, 25.50, 22.80, 17.23, 14.21; MS (70 eV): m/z (%) 209.2 (M+, 100)

2.4d (4-fluoro-phenyl)-1-phenyl-1H-pyrazole-4-carbonitrile (D_1): White solid, M.p. 170–173 °C; IR (KBr) ν: cm^{-1}; 2217, 1677,1226, 836 (cm^{-1}); ^1H NMR (400 MHz, DMSO-d6) δ 7.4–7.60 (m, 5H), 7.92–8.01 (d, $J = 6.5$ Hz, 2H), 8.01–8.03 (d, $J = 6.8$, 2H), 9.45 (s, 1H); MS (m/z) 265.25.

2.4e 1,3-diphenyl-1H-pyrazole-4-carbonitrile (D_2): White solid, M.p. 120–123 °C; IR (KBr) ν: cm^{-1}; 2213, 1589,1214, 819 (cm^{-1}); ^1H NMR (400 MHz, DMSO-d6) δ 7.36–7.56 (m, 5H), 7.60–7.94 (m, 5H), 9.42 (s, 1H) ; HRMS (ESI) m/z calcd. For $C_{16}H_{11}N_3$ [M+H]+: 246.22, found: 246.30.

2.4f 3-(4-chloro-phenyl)-1-phenyl-1H-pyrazole-4-carbonitrile (D_3): White solid, M.p. 158–161 °C; IR (KBr) ν: cm^{-1}; 2221, 1594,1229, 834 (cm^{-1}); ^1H NMR (400 MHz, DMSO-d6) δ 7.37–7.52 (m, 5H), 7.54–7.57 (d, $J = 8.2$ Hz, 2H), 7.59–7.99 (d, $J = 8.0$ Hz, 2H), 9.45 (s, 1H); MS (m/z) 280.25 [M+H]+, 282.30 [M+2].

3. Result and Discussion

Initially, benzaldehyde treated with hydroxylamine in the presence of acetic acid followed by dehydration with orthophosphoric acid to obtained benzonitrile (**B**) with **87%** yield within 7 min (Table 1). The same reaction was performed using formic acid as a medium. Surprisingly, we obtained an excellent yield (99%) of benzonitrile (**B**) within a short reaction time. Hence, formic acid is a better choice of medium for subsequent reactions**.**

Similarly, a mixture of pyrazole carbaldehyde (**C_1**) and formic acid was cooled to 5–10 °C and subsequently, hydroxylamine hydrochloride was added to form oxime, which on dehydration with H_3PO_4 at 100 °C till TLC complies to form pyrazole nitrile (**D_1**). The product obtained was isolated by simple quenching reaction mass into ice-cold water (Table 1, Entry

Table 2. Catalyst optimization for one-pot synthesis of nitrile[a].

Entry	H_3PO_4 Catalyst mole %	Time (h)	Yield (%)[b]
1	0.5%	9	85
2	**1.0%**	**5**	**99**
3	1.5%	3.5	95
4	2.0%	2.5	95
5	2.5%	2.0	95

[a]Experimental conditions: Benzaldehyde (1 mmol), NH_2-OH. HCl (1 mmol), H_3PO_4 (1 mmol) at 100 °C.

[b]Isolated yield.

[c]Bold signifies the optimized and suitable catalyst and solvent for the synthesis of nitriles.

5) to obtain corresponding nitrile derivatives. The product was obtained with a good yield (99%).

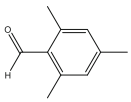
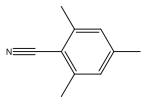
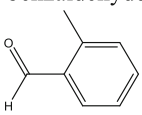
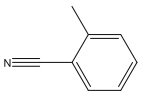
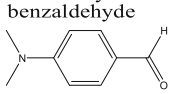
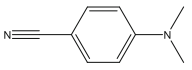
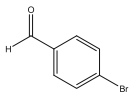
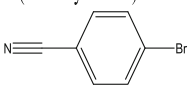
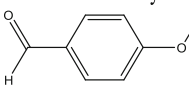
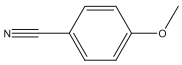
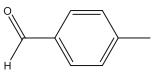
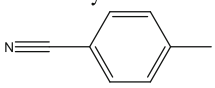
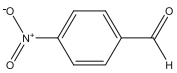
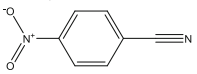
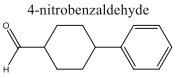
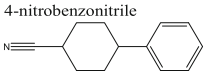
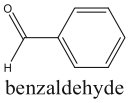
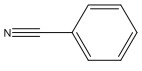
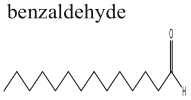
Investigations of solvent optimization were carried out and are summarized in Table 1 (Table 1, Entry 5, 99%).

Similarly, the study of optimization of catalyst was also carried out and is summarized in Table 2. The concentration of catalyst increased from 0.5 mol%, to 1.0 mol% both yield and rate of the reaction was increased (Table 2, Entries 1, 2, 3). However, further increment of catalyst amount (1.5 mol%, 2.5 mol%) did not appreciably affect the yield and the rate of the reaction (Table 2, Entries 4, 5).

Finally, among all the experimental variations, 1.0 mol% H_3PO_4 condition at 100 °C temperature gave the best results with 99% yield (Table 2, Entry 2). To check the generality and the scope of the optimized reaction, the methodology was evaluated by employing different aliphatic, alicyclic, aromatic aldehydes or ketones. The resultant nitriles (**B_1-B_{10}**) were obtained in good to excellent yields.

An identical method was applied for the synthesis of novel pyrazole-4-nitriles **(D_1–D_{10})** from pyrazole-4-

Table 3. Synthesis of nitrile (B_1–B_{10}) from aldehyde[a].

Compound	Pyrazole aldehyde	Product	Yield[b] (%)	Observed melting point °C
B_1	 2,4,6-trimethyl benzaldehyde	 2,4,6-Trimethylbenzonitrile	98	50–55
B_2	 2-methyl benzaldehyde	 2-methylbenzonitrile	99	Boiling point 205
B_3	 4-(dimethylamino)benzaldehyde	 4-(dimethylamino)benzonitrile	98	**72–75**
B_4	 4-bromobenzaldehyde	 1-bromo-4-ethynylbenzene	99	235–237
B_5	 4-methoxybenzaldehyde	 4-methoxybenzonitrile	99	**256–257**
B_6	 4-methylbenzaldehyde	 4-Methylbenzonitrile	97	217.611
B_7	 4-nitrobenzaldehyde	 4-nitrobenzonitrile	98	**144–147**
B_8	 4-phenylcyclohexanecarbaldehyde	 4-Phenylcyclohexane-1-carbonitrile	97	
B_9	 benzaldehyde	 Benzonitrile	99	188–191
B_{10}	 tetradecanal	 Tetradecanenitrile	98	

[a]Reaction conditions: aldehyde (1 mmol), NH_2OH. HCl (1 mmol), H_3PO_4 (1.0 mol%) solvent formic acid at 100 °C.
[b]Isolated yield.
[c]Bold signifies the optimized and suitable catalyst and solvent for the synthesis of nitriles.

Table 4. Synthesis of pyrazole nitrile (D_1–D_{20}) from pyrazole aldehyde[a].

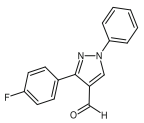
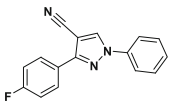
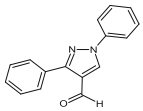
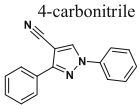
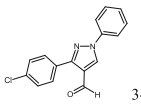
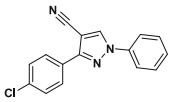
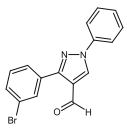
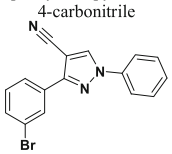
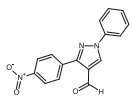
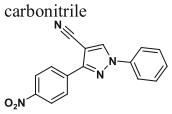
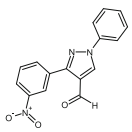
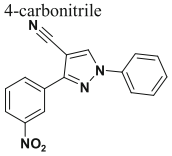
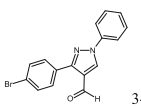
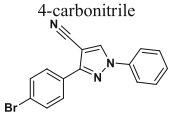
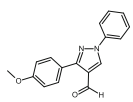
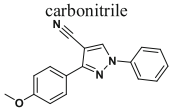
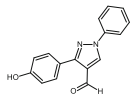
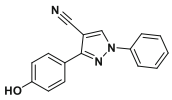
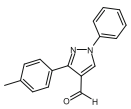
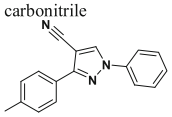
Sl. No.	Pyrazole aldehyde	Product	Yield[b] (%)	Observed Melting point °C
D_1	 3-(4-fluoro-phenyl)-1-phenyl-1*H*-pyrazole-4-carbaldehyde	 3-(4-fluoro-phenyl)-1-phenyl-1*H*-pyrazole-4-carbonitrile	97	170–173
D_2	 1,3-diphenyl-1*H*-pyrazole-4-carbaldehyde	 1,3-diphenyl-1*H*-pyrazole-4-carbonitrile	94	120–123
D_3	 3-(4-chloro-phenyl)-1-phenyl-1*H*-pyrazole-4-carbaldehyde	 3-(4-chloro-phenyl)-1-phenyl-1*H*-pyrazole-4-carbonitrile	99	158–161
D_4	 3-(3-bromophenyl)-1-phenyl-1*H*-pyrazole-4-carbaldehyde	 3-(3-Bromo-phenyl)-1-phenyl-1*H*-pyrazole-4-carbonitrile	95	160–162
D_5	 3-(4-nitrophenyl)-1-phenyl-1*H*-pyrazole-4-carbaldehyde	 3-(4-nitro-phenyl)-1-phenyl-1*H*-pyrazole-4-carbonitrile	98	198–200
D_6	 3-(3-nitrophenyl)-1-phenyl-1*H*-pyrazole-4-carbaldehyde	 3-(4-nitro-phenyl)-1-phenyl-1*H*-pyrazole-4-carbonitrile	97	195–197
D_7	 3-(4-bromophenyl)-1-phenyl-1*H*-pyrazole-4-carbaldehyde	 3-(4-Bromo-phenyl)-1-phenyl-1*H*-pyrazole-4-carbonitrile	92	164–166
D_8	 3-(4-methoxyphenyl)-1-phenyl-1*H*-pyrazole-4-carbaldehyde	 3-(4-Bromo-phenyl)-1-phenyl-1*H*-pyrazole-4-carbonitrile	91	170–172

Table 4. *(contd)*

Sl. No.	Pyrazole aldehyde	Product	Yield[b] (%)	Observed Melting point °C
D_9	 3-(4-hydroxyphenyl)-1-phenyl-1*H*-pyrazole-4-carbaldehyde	 3-(4-hydroxy-phenyl)-1-phenyl-1*H*-pyrazole-4-carbonitrile	94	182–183
D_{10}	 3-(4-methylphenyl)-1-phenyl-1*H*-pyrazole-4-carbaldehyde	 3-(4-methyl-phenyl)-1-phenyl-1*H*-pyrazole-4-carbonitrile	99	165–167

[a]Reaction conditions: Pyrazole aldehyde (1 mmol), NH2OH.HCl (1 mmol), H_3PO_4 (1.0 mol%) solvent formic acid at 100 °C.
[b]Isolated yield.

carbaldehydes. The resultant nitriles were obtained in excellent yield.

All the novel compounds were characterized by IR spectral analysis, the appearance of peak near 2220 cm^{-1} clearly indicates the formation of nitrile. In compound **D_1**, peak observed at 2217 cm^{-1} for **CN** stretching frequency and disappearance of strong peak near 1720 cm^{-1} confirmed the conversion of aldehyde into nitrile. Structure elucidation of all the compounds were confirmed by ^1H NMR spectral analysis, chemical shift values and the number of NMR signals confirmed the structure of synthesized compounds. In addition to this, all the novel compounds were characterized by mass spectrometrically, and the molar masses of all these compounds consistent with theoretical data (Tables 3, 4).

4. Conclusions

A novel, cost-effective, eco-friendly, metal-free and mild protocol has been developed for the synthesis of nitrile derivatives using H_3PO_4 as a catalyst. This protocol offers several significant advantages, including operational simplicity, superior atom-economy, short reaction time and good to excellent yields. Moreover, we have successfully attempted the synthesis of valuable novel pyrazole nitriles (**D_1–D_{10}**) and their structures were confirmed by IR, ^1H, ^{13}C NMR and mass spectral techniques.

Supplementary Information (SI)

Tables S1–S3, NMR spectra, IR spectra, Mass spectra are available at www.ias.ac.in/chemsci.

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Declarations

Conflict of interest There is no conflict of interest related to this article.

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