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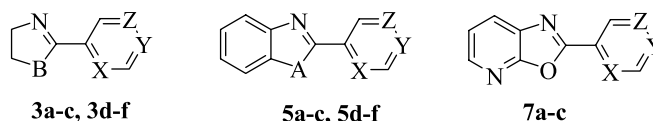
Microwave-assisted conversion of aromatic heterocyclic nitriles to various heterocyclic molecules

Sham M. Sondhi*, Surbhi Arya and Reshma Rani

Department of Chemistry, Indian Institute of Technology-Roorkee, Roorkee 247667, Uttarakhand, India

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Dihydroimidazole (**3a-c**), dihydrooxazole (**3d-f**), benzoxazole (**5a-c**), benzothiazole (**5d-f**) and oxazolopyridine (**7a-c**) derivatives have been synthesized by condensation of various heterocyclic aromatic nitriles with diamine, aminoalcohol, aminophenol, aminothiophenol, and 3-aminopyridine-2-ol, respectively, under microwave irradiation and under solvent free conditions. This catalyst free and solvent free approach provided heterocyclic compounds in quantitative yields. Time taken for the condensation to occur is < 20 min.



3a-c, 3d-f

5a-c, 5d-f

7a-c

Keywords: microwave; imidazole; oxazole; benzoxazole; benzothiazole

Introduction

Heterocyclic compounds bearing imidazole, oxazole, benzoxazole, benzothiazole, and oxazolopyridine moieties are of current research interest (1–6) due to their pharmacological applications (7). These types of heterocyclic compounds exhibit antitumor (8, 9) anti-inflammatory (10, 11), antibacterial (12), antiviral (13), and antimicrobial (14) activities. Since this investigation deals with a new approach to synthesize these medicinally important heterocyclic compounds, it is essential to review the procedures reported in literature. These procedures involve condensation *via* using (i) heterogeneous catalyst such as silphox with dimethylformamide (15), heteropolyacid supported on silica gel [Cu_{3/2}PMo₁₂O₄₀/SiO₂] (16), silica sulfuric acid (17), (ii) metal catalyst that is Pd(OAc)₂PPh₃ (18), H₃PW₁₂O₄₀ (19), ZrOCl₂·8H₂O (20), CuCl & Cu(OTf)₂ (21), (iii) solid support reagents (22), polyphosphoric acid (23), and *N*-bromosuccinimide (24). These heterocyclic molecules also have been synthesized by microwave irradiation (25–27), ultrasound irradiation by using catalyst (27), and by using ceric ammonium nitrate in presence of hydrogen peroxide under solvent free condition (28). Nitrile have also been converted to heterocyclic molecules by using catalyst and/or solvent under microwave

irradiation (29–32). To the best of our knowledge no method is reported in literature involving condensation of heterocyclic aromatic nitriles with aliphatic diamino, aliphatic aminoalcohol, aromatic phenol, aromatic amino thiophenol, and hetroaromatic amino phenol using solvent free, catalyst free and microwave assisted conditions.

In related development microwave chemistry has a profound impact on the synthesis of heterocyclic compounds under solvent free condition, *via* using microwave irradiation for acceleration of organic reactions (33–36). Microwave irradiation technique reduces reaction time drastically and thus less chances of getting side products, which ultimately lead to high yields of required products and easy workup procedure.

In continuation of our efforts (37–40) in search of biologically important heterocyclic compounds which can be synthesized easily, we have synthesized dihydro-1*H*-imidazole, dihydrooxazole, benzoxazole, benzothiazole, and oxazolopyridine derivatives by reaction of heterocyclic aromatic nitriles with diamine, aminoalcohol, aminophenol, aminothiophenol, and 3-aminopyridine-2-ol, respectively, using microwave irradiation technique under solvent free condition, which we wish to report in this article.

*Corresponding author. Email: sondifcy@iitr.ernet.in

Results and discussion

Equimolar ratio of 2-cyanopyridine **1a** (Scheme 1) and ethane 1,2 diamine **2a** (Scheme 1) were mixed together to make a homogeneous mixture and then this reaction mixture was subjected to microwave irradiation for 2 min. Thin layer chromatography (TLC) of reaction mixture showed formation of a new spot. This mixture was further subjected to microwave irradiation for 2×3 min. TLC of reaction mixture indicated absence of starting materials. This crude product was recrystallized from ethyl acetate to give pure product **3a** in quantitative yield. IR, ^1H NMR, GC-MS spectral and analytical data indicate that compound **3a** is 2-(4,5-dihydro-1H-imidazol-2-yl)pyridine which is formed by addition of diamine on nitrile functional group and then elimination of one molecule of NH_3 . By following one step process heterocyclic molecules **3b-f** were also synthesized in quantitative yield. Percentage yield and melting point (MP) of **3a-f** are mentioned in Table 1.

To further diversify this synthetic procedure we studied the reaction of 2-aminophenol **4a** (Scheme 1) and 2-cyanopyridine **1a** (Scheme 1) by irradiating their mixture in equimolar ratio for 1×5 min. A new product **5a** was formed which was identified to be 2-(pyridin-2-yl)benzo[d]oxazole, which is formed by cyclization and loss of one molecule of NH_3 . By following one step process heterocyclic molecules **5b-f** were also synthesized in quantitative yield. Percentage yield and MP of **5a-f** are mentioned in Table 1.

To verify the versatility of our synthetic procedure we condensed 3-aminopyridin-2-ol (**6**; Scheme 1) with 2-cyanopyridine, 4-cyanopyridine, and 2-cyanopyrazine (**1a-c**; Scheme 1) under microwave irradiation and got heterocyclic cyclized product **7a-c** in quantitative yield. Percentage yield and MP of **7a-c** are mentioned in Table 1.

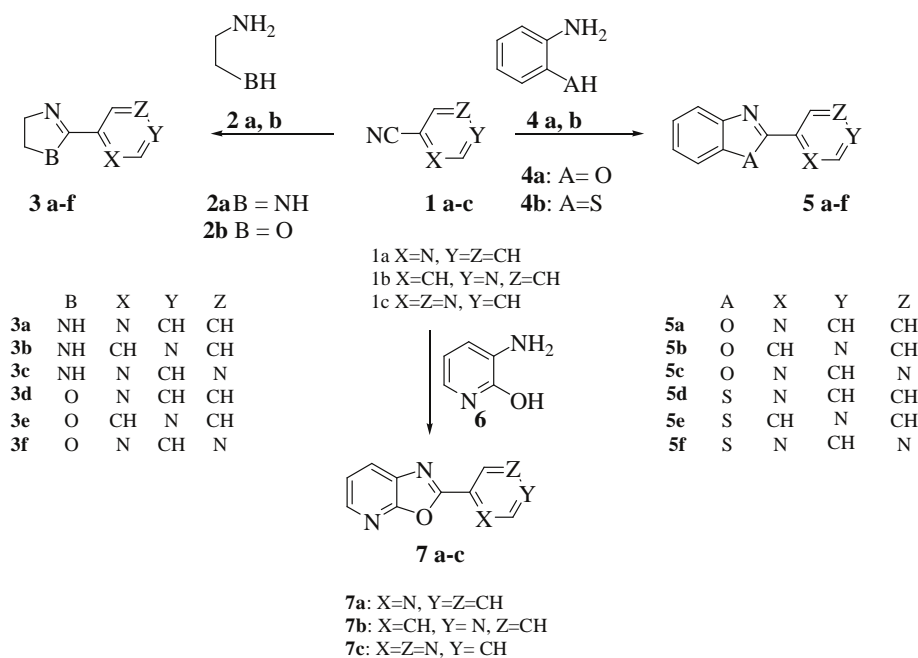
Experimental section

Microwave reactor model CEM Discover model No. 908010 were used for microwave irradiation. MP were determined on a JSGW apparatus and are uncorrected. IR spectra were recorded using a Perkin Elmer 1600 FT spectrometer. ^1H NMR spectra were recorded on a Bruker WH-500 MHz NMR spectrometer at a ca 5–15% (w/v) solution in $\text{DMSO-}d_6$ (TMS as internal standard). GC-MS was recorded on Perkin Elmer Clarus 500 gas chromatograph where built in MS detector was used. Elemental analysis was carried out on a Vario EL III elementor. TLC was performed on silica gel G for TLC (Merck) and spots were visualized by iodine vapor or by irradiation with ultraviolet light (254 nm).

General procedure for synthesis of heterocyclic molecules

Reaction procedure for synthesis of 3a

Ethane-1,2-diamine (0.240 g, 4 mmol) and 2-cyanopyridine (0.416 g, 4 mmol) were mixed together



Scheme 1. Synthesis of various heterocyclic compounds.

Table 1. Physical constants of various heterocyclic compounds synthesized* *i.e.* **3a-f**, **5a-f** & **7a-c**.

Compound no.	Solvent of crystallization	% Yield	MP (°C)
3a	Ethyl acetate	80	98–100 [lit(41) 101–102]
3b	Ethyl acetate	82	135–137 [lit(41) 136–137]
3c	Acetone	80	117–118
3d	Washing with chilled diethyl ether	75	BP. 87 [lit(43) 90]
3e	EtOH	82	98–100 [lit(44) 97–99]
3f	EtOH	88	88–90
5a	Ethyl acetate	68	105 [lit(45) 106–107]
5b	Ethyl acetate	70	115–117 [lit(23) 116–119]
5c	Ethyl acetate	72	115
5d	EtOH	62	132–135 [lit(46) 132–133]
5e	Ethyl acetate	60	134–136 [lit(46) 135–136]
5f	EtOH	63	171–173 [lit(47) 170–172]
7a	Ethyl acetate	71	218–220
7b	Ethyl acetate	72	138–140
7c	Ethyl acetate	75	164–165

*Using microwave reactor CEM Discover model No. 908010, reaction temperature for **3a-f** & **5d-f** was 70 °C and for **5a-c** & **7a-c** 75 °C.

thoroughly to form homogeneous reaction mixture. This reaction mixture was subjected to microwave irradiation for 2 × 4 min at 70 °C. Completion of reaction was checked by TLC. Crude reaction product was crystallized from ethyl acetate to give pure product **3a**. Similarly, compounds **3b-f**, **5a-f**, and **7a-c** were prepared under microwaves. These reactions were also performed using household microwave oven (model M197DL, Samsung) and found comparable in terms of yield and reaction times.

2-(4,5-Dihydro-1H-imidazol-2-yl)-pyridine (3a)(41)

Irradiation time 6 min. IR (KBr) ν_{\max} : 1639 (–C=N–), 1532 & 1461 (Ar) cm^{-1} . ^1H NMR (CDCl_3) δ : 3.571 (bs, 2H, aliphatic), 4.09 (bs, 2H, aliphatic), 5.99 (bs, 1H, NH), 7.37–7.39 (m, 1H, py), 7.77–7.80 (m, 1H, py), 8.154–8.172 (dd, $J_1=1$ Hz, $J_2=8$ Hz, 1H, py), 8.592–8.60 (m, 1H, py). GC-MS m/z 147 (M^+ , 44.74%); Anal. Calcd. for $\text{C}_8\text{H}_9\text{N}_3$ C: 65.30; H: 6.12; N: 28.57; Found C: 65.25; H: 6.08; N: 28.53%.

4-(4,5-Dihydro-1H-imidazol-2-yl)-pyridine (3b)(41)

Irradiation time 5 min. IR (KBr) ν_{\max} : 1630 (–C=N–), 1592 & 1545 (Ar) cm^{-1} . ^1H NMR (CDCl_3) δ : 3.827–3.88 (bs, 4H, aliphatic), 4.851 (bs, 1H, NH), 7.646–7.658 (m, 2H, py), 8.713–8.725 (d, $J_1=6$ Hz, 2H, py). GC-MS m/z 147 (M^+ , 55.91%); Anal. Calcd. For $\text{C}_8\text{H}_9\text{N}_3$ C: 65.30; H: 6.12; N: 28.57; Found C: 65.33; H: 6.10; N: 28.55%.

2-(4,5-Dihydro-1H-imidazol-2-yl)-pyrazine (3c)(42)

Irradiation time 6 min. IR (KBr) ν_{\max} : 1601 (–C=N–), 1498 & 1461 (Ar), cm^{-1} . ^1H NMR

(CDCl_3) δ : 3.579 (bs, 2H, aliphatic), 4.040 (bs, 2H, aliphatic), 5.783 (s, 1H, NH), 8.466–8.474 (m, 1H, pyrazine), 8.573–8.578 (d, $J_1=2.5$ Hz, 1H, pyrazine), 9.309–9.311 (d, $J_1=1$ Hz, 1H, pyrazine). GC-MS m/z 148 (M^+ , 80.74%); Anal. Calcd. For $\text{C}_7\text{H}_8\text{N}_4$ C: 56.75; H: 5.40; N: 37.83; Found C: 56.70; H: 5.42; N: 37.78%.

2-(4,5-Dihydro-oxazol-2-yl)-pyridine (3d)(43)

Irradiation time 3 min. IR (KBr) ν_{\max} : 1658 (–C=N–), 1533 & 1469 (Ar), cm^{-1} . ^1H NMR (CDCl_3) δ : 4.43–4.182 (t, $J_1=9.5$ Hz, 2H, aliphatic), 4.540–4.578 (t, $J_1=9.5$ Hz, 2H, aliphatic), 7.428–7.437 (m, 1H, py), 7.795–7.830 (m, 1H, py), 8.067–8.083 (d, $J_1=8$ Hz 1H, py), 8.730–8.739 (d, $J_1=4.5$ Hz 1H, py). GC-MS m/z 148 (M^+ , 26.75%); Anal. Calcd. For $\text{C}_8\text{H}_8\text{N}_2\text{O}$ C: 64.86; H: 5.40; N: 18.91; Found C: 64.83; H: 5.37; N: 18.90%.

4-(4,5-Dihydro-oxazol-2-yl)-pyridine (3e)(44)

Irradiation time 4 min. IR (KBr) ν_{\max} : 1652 (–C=N–), 1547 & 1414 (Ar), cm^{-1} . ^1H NMR (CDCl_3) δ : 4.116–4.155 (t, $J_1=9.5$ Hz, 2H, aliphatic), 4.489–4.528 (t, $J_1=9.5$ Hz, 2H, aliphatic), 7.803–7.815 (m, 2H, py), 8.737–8.749 (m, 2H, py). GC-MS m/z 148 (M^+ , 50.31%); Anal. Calcd. For $\text{C}_8\text{H}_8\text{N}_2\text{O}$ C: 64.86; H: 5.40; N: 18.91; Found C: 64.86; H: 5.35; N: 18.93%.

2-(4,5-Dihydro-oxazol-2-yl)-Pyrazine (3f)

Irradiation time 2 min. IR (KBr) ν_{\max} : 1653 (–C=N–), 1573 & 1469 (Ar), cm^{-1} . ^1H NMR (CDCl_3) δ : 4.168–4.207 (t, $J_1=9.5$ Hz, 2H, aliphatic), 4.547–4.585 (t, $J_1=9.5$ Hz, 2H, aliphatic),

8.682–8.692 (d, $J_1=5$ Hz, 2H, pyrazine), 9.277 (s, 1H, pyrazine). GC-MS m/z 149 (M^+ , 72.19%); Anal. Calcd. For $C_7H_7N_3O$ C: 56.37; H: 4.69; N: 28.18; Found C: 56.35; H: 4.65; N: 28.13%.

2-Pyridin-2-yl-benzooxazole (5a) (45)

Irradiation time 5 min. IR (KBr) ν_{\max} : 1621 ($-C=N-$), 1591 & 1454 (Ar), cm^{-1} . 1H NMR (DMSO- d_6) δ : 7.456–7.527 (m, 2H, Ar), 7.641–7.666 (m, 1H, py), 7.858–7.901 (m, 2H, Ar), 8.064–8.099 (dt, 1H, py), 8.351–8.371 (dt, 1H, py), 8.807–8.819 (m, 1H, py). GC-MS m/z 196 (M^+ , 100%); Anal. Calcd. For $C_{12}H_8N_2O$ C: 73.46; H: 4.08; N: 14.28; Found C: 73.40; H: 4.00; N: 14.25%.

2-Pyridin-4-yl-benzooxazole (5b) (23)

Irradiation time 12 min. IR (KBr) ν_{\max} : 1633 ($-C=N-$), 1456 (Ar), cm^{-1} . 1H NMR (DMSO- d_6) δ : 7.470–7.551 (m, 2H, Ar), 7.866–7.918 (m, 2H, Ar), 8.115–8.127 (dd, $J_1=1.5$ Hz, $J_2=4.5$ Hz, 2H, py), 8.855–8.868 (d, $J_1=2$ Hz, $J_2=4.5$ Hz, 2H, py). Anal. Calcd. For $C_{12}H_8N_2O$ C: 73.46; H: 4.08; N: 14.28; Found C: 73.42; H: 4.02; N: 14.20%.

2-Pyrazin-2-yl-benzooxazole (5c)

Irradiation time 5 min. IR (KBr) ν_{\max} : 1684 ($-C=N-$), 1596 & 1446 (Ar), cm^{-1} . 1H NMR (DMSO- d_6) δ : 7.494–7.572 (m, 2H, Ar), 7.901–7.938 (m, 2H, Ar), 8.891–8.906 (d + s, 2H, pyrazine), 9.525–9.527 (d, $J_1=1$ Hz, 1H, pyrazine). GC-MS m/z 197 (M^+ , 100%); Anal. Calcd. For $C_{11}H_7N_3O$ C: 67.00; H: 3.55; N: 21.31; Found C: 66.94; H: 3.53; N: 21.35%.

2-Pyridin-2-yl-benzothiazole (5d) (46)

Irradiation time 1 min. IR (KBr) ν_{\max} : 1643 ($-C=N-$), 1569 & 1506 (Ar), cm^{-1} . 1H NMR (DMSO- d_6) δ : 7.495–7.528 (m, 1H, Ar), 7.565–7.623 (m, 2H, Ar), 8.038–8.073 (m, 1H, Ar), 8.109–8.125 (d, $J_1=8$ Hz, 1H, py), 8.175–8.192 (m, 1H, py), 8.335–8.355 (m, 1H, py), 8.736–8.751 (m, 1H, py). GC-MS m/z 212 (M^+ , 100%); Anal. Calcd. For $C_{12}H_8N_2S$ C: 67.92; H: 3.77; N: 13.20; S: 15.09; found C: 67.90; H: 3.70; N: 13.24; S: 15.05%.

2-Pyridin-4-yl-benzothiazole (5e) (46)

Irradiation time 2 min. IR (KBr) ν_{\max} : 1594 & 1469 (Ar), cm^{-1} . 1H NMR (DMSO- d_6) δ : 7.560–7.642 (m, 2H, Ar), 8.044–8.056 (d, $J_1=6$ Hz, 2H, py), 8.241–8.257 (d, $J_1=8$ Hz, 2H, Ar), 8.803–8.815 (d, $J_1=6$ Hz, 2H, py). GC-MS m/z 212 (M^+ , 100%); Anal. Calcd. For $C_{12}H_8N_2S$ C: 67.92; H: 3.77; N:

13.20; S: 15.09; Found C: 67.89; H: 3.75; N: 13.15; S: 15.02%.

2-Pyrazin-2-yl-benzothiazole (5f) (47)

Irradiation time 2 min. IR (KBr) ν_{\max} : 1630 ($-C=N-$), 1508 & 1447 (Ar), cm^{-1} . 1H NMR (DMSO- d_6) δ : 7.533–7.632 (m, 2H, Ar), 8.161–8.177 (d, $J_1=8$ Hz, 1H, Ar), 8.217–8.233 (d, $J_1=8$ Hz, 1H, Ar), 8.820–8.828 (m, 1H, pyrazine), 8.849–8.854 (d, $J_1=2.5$ Hz, 1H, pyrazine), 9.509 (s, 1H, pyrazine). GC-MS m/z 213 (M^+ , 100%); Anal. Calcd. For $C_{11}H_7N_3S$ C: 61.97; H: 3.28; N: 19.71; S: 15.02; Found C: 61.94; H: 3.25; N: 19.67; S: 15.00%.

2-Pyridine-2-yl-oxazolo[4,5-b]pyridine (7a) (48)

Irradiation time 4 min. IR (KBr) ν_{\max} : 1614 ($-C=N-$), 1584 & 1549 (Ar), cm^{-1} . 1H NMR (DMSO- d_6) δ : 7.532–7.558 (m, 1H, Ar), 7.688–7.715 (m, 1H, Ar), 7.715–8.100 (m, 1H, Ar), 8.325–8.344 (dd, $J_1=1.5$ Hz, $J_2=8$ Hz, 1H, py), 8.413–8.432 (m, 1H, py), 8.617–8.629 (dd, $J_1=1$ Hz, $J_2=4.5$ Hz, 1H, py), 8.840–8.849 (m, 1H, py). GC-MS m/z 197 (M^+ , 67.44%); Anal. Calcd. For $C_{11}H_7N_3O$ C: 67.00; H: 3.55; N: 21.31; Found C: 66.96; H: 3.49; N: 21.30%.

2-Pyridine-4-yl-oxazolo[4,5-b]pyridine (7b) (48)

Irradiation time 20 min. IR (KBr) ν_{\max} : 1632 ($-C=N-$), 1586 & 1500 (Ar), cm^{-1} . 1H NMR (DMSO- d_6) δ : 7.562–7.588 (q, 1H, Ar), 8.166–8.178 (dd, $J_1=2$ Hz, $J_2=4.5$ Hz, 2H, py), 8.350–8.369 (dd, $J_1=1$ Hz, $J_2=8$ Hz, 1H, Ar), 8.639–8.651 (dd, $J_1=1$ Hz, $J_2=4.5$ Hz, 1H, Ar), 8.900–8.912 (dd, $J_1=1.5$ Hz, $J_2=4.5$ Hz, 2H, py). GC-MS m/z 197 (M^+ , 65.51%); Anal. Calcd. For $C_{11}H_7N_3O$ C: 67.00; H: 3.55; N: 21.31; Found C: 63.25; H: 3.50; N: 21.25%.

2-Pyrazine-2-yl-oxazolo[4,5-b]pyridine (7c)

Irradiation time 8 min. IR (KBr) ν_{\max} : 1612 ($-C=N-$), 1551 & 1458 (Ar), cm^{-1} . 1H NMR (DMSO- d_6) δ : 7.579–7.605 (q, 1H, Ar), 8.379–8.398 (dd, $J_1=1$ Hz, $J_2=8$ Hz, 1H, Ar), 8.662–8.674 (dd, $J_1=1$ Hz, $J_2=4.5$ Hz, 1H, Ar), 8.944 (s, 2H, pyrazine), 9.580–9.582 (d, $J_1=1$ Hz, 1H, pyrazine). GC-MS m/z 198 (M^+ , 68%); Anal. Calcd. For $C_{10}H_6N_4O$ C: 60.60; H: 3.03; N: 14.14; Found C: 57.25; H: 2.50; N: 14.12%.

Conclusion

In conclusion, we have developed a very simple, high yielding, easy to workup one step process for

conversion of heterocyclic aromatic nitriles to various heterocyclic compounds. Further efforts to synthesize more complex heterocyclic compounds are in progress.

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