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ONE-POT, MILD AND EFFICIENT MULTICOMPONENT SYNTHESIS OF NOVEL VARIOUS *SPIRO*-NITROGEN HETEROCYCLE COMPOUNDS

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ABSTRACT. One-pot, mild and efficient synthesis of various *spiro*-nitrogen heterocycle compounds, based on the reaction of ninhydrin and 1,2-diamino-benzene, *(indenoquinoxalin)*, with *N*-heterocycle compounds and dialkylacetylenedicarboxylates is described. Using this approach, various *spiro*-nitrogen heterocycle compounds at a temperature of 50-60 °C in acetonitrile solvent, can be obtained very high yields.

KEY WORDS: Ninhydrin, 1,2-Diaminobenzene, *spiro*-Nitrogen heterocycle compounds, *Indenoquinoxalin*, *N*-heterocycle compounds, Dialkylacetylenedicarboxylates

INTRODUCTION

Heterocyclic chemistry is one of the most complex branches of chemistry and heterocyclic compounds, the largest and most diverse family of chemical compounds. The heterocyclic compounds have a stable structure that cannot be easily hydrolyzed or polymerized [1-3]. The heterocyclic compounds are a broad category of ring compounds that contain one or more non-carbon atoms, including nitrogen, oxygen, sulfur, or phosphorus in their structure [4-6].

Heterocyclic compounds play a vital role in biological processes and are widely found in natural compounds [7-11]. The main source of these compounds is plants. Heterocyclic compounds are used in the pharmaceutical industry. Some vitamins, proteins, and hormones have a heterocyclic structure. Multi-component reactions have always been a beneficial way of synthesizing heterocyclic compounds [12-17]. The quinoline, isoquinoline and *indenoquinoxalin* skeleton compounds are often used for the design of many synthetic compounds with diverse pharmacological properties such as antimicrobial, cytotoxic, HIV protease inhibitor, anti-inflammatory, anti-cancer, antitumor, antimalarial and anti-viral activities [18-20]. Yavari and co-workers had reported a huge number studies, utilizing quinoline, isoquinoline, *indenoquinoxalin* and acetylendicarboxylate esters [21-27].

Meanwhile, *indenoquinoxalin* is one of the most important compounds for the synthesis of many heterocyclic compounds using multiple reactions [28-34]. The extent and dynamics of this part of organic chemistry has made these compounds a special place. Hence, researchers are using new and modern techniques to produce heterocyclic compounds [35-44].

According to the introduction, in this paper, using a gentle, effective and one-pot method, we reported an effective synthesis of *spiro*-heterocyclic nitrogen compounds based on the reaction between ninhydrin and 1,2-diaminobenzen, (*indenoquinoxalin*), with heterocyclic nitrogen compounds and dialkylacetylenedicarboxylate. Based on this, various *spiro*-nitrogen heterocycle compounds at a temperature of 50-60 °C in acetonitrile solvent, can be obtained very good yields.

RESULTS AND DISCUSSION

Initially, for the sample the reaction of ninhydrin **1** with 1,2-diaminobenzene **2** with isoquinoline **3** and dimethylacetylenedicarboxylate **4** was selected. In this case, solvent optimization and

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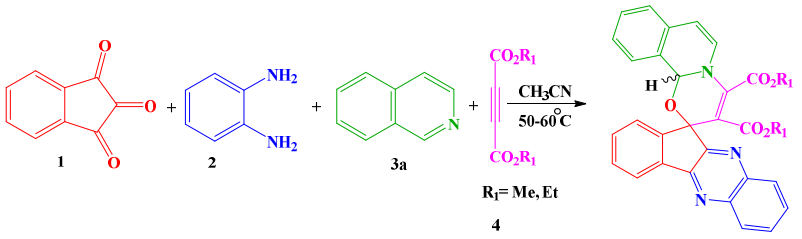
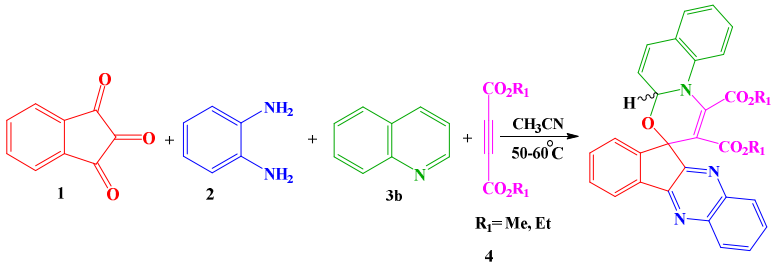
reaction conditions were performed. Acetonitrile was found to be a suitable solvent and the best yield of **5a** was obtained at a temperature of 50-60 °C in acetonitrile. In the following, the reaction of ninhydrin **1** and 1,2-diaminobenzene **2** with nitrogen heterocycle compounds **3** and dialkylacetylenedicarboxylate **4** in an acetonitrile solvent is complete at 50-60 °C for 12 hours and *spiro*-heterocyclic nitrogen-containing compounds **5a-f** produce very good yields (Scheme 1-3).

According to the results, products have two diastereomeric forms (60:40). But it is noteworthy that the reaction between dimethylacetylenedicarboxylate with pyridine or isoquinoline or quinoline and *N*-heterocycle compounds generates only one diastereoisomer, while with diethylacetylenedicarboxylate produce two diastereo isomers. Unfortunately, we could not separate these diastereoisomers (Scheme 4).



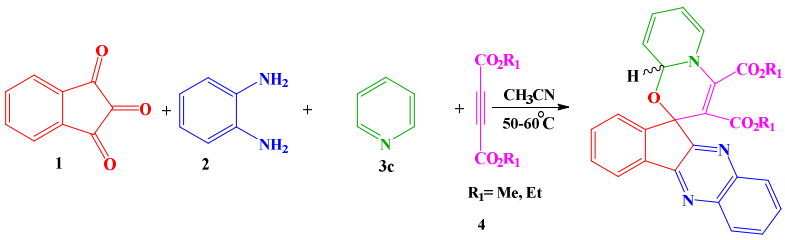
	R_1	Yield[%]
5a	Me	89
5b	Et	87

Scheme 1. Formation of *spiro*-nitrogen heterocycle compounds **5a–5b**.



	R_1	Yield[%]
5c	Me	85
5d	Et	77

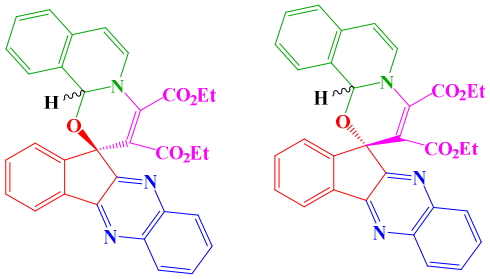
Scheme 2. Formation of *spiro*-nitrogen heterocycle compounds **5c–5d**.



	R_1	Yield[%]
5e	Me	86
5f	Et	79

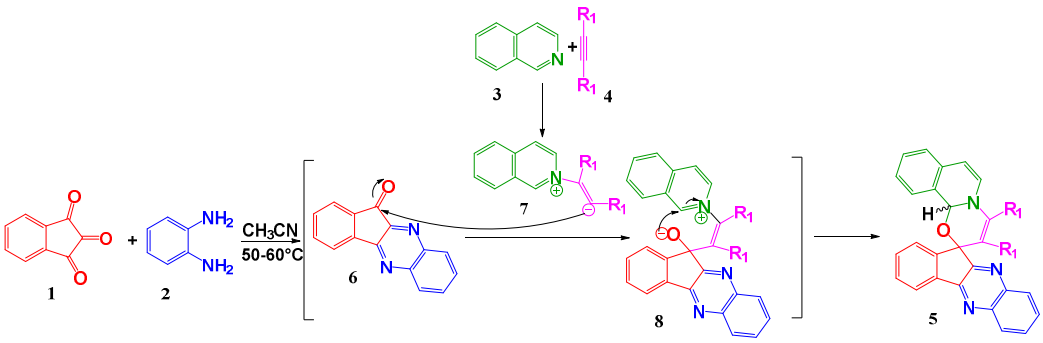
Scheme 3. Formation of *spiro*-nitrogen heterocycle compounds **5e–5f**.

The structure of the *spiro*-heterocyclic nitrogen compounds **5a-f** is deduced from the ^{1}H NMR, ^{13}C NMR and IR spectra and elemental analysis. For example, the spectrum of the ^{1}H NMR composition of **5a** in $CDCl_3$ showed a singlet at δ 3.23 and a singlet at δ 4.07 for the methoxyprotons (2OMe), a doublet at δ 5.91 and a doublet at δ 6.57 for the methine proton ($2=CH_{dihydroisoquinoline}$), a singlet at δ 6.88 for the ($CH_{diastereoisomer}$) proton, multiplets at δ 7-8.40 for aryl protons(12H). The spectrum of the ^{13}C NMR composition of **5a** in $CDCl_3$ showed 30 distinct resonances in agreement with the proposed structure.



Scheme 4. Two diastereoisomers of *spiro*-nitrogen heterocycle compounds **5b**, **5d** and **5f**.

The proposed mechanism for the synthesis of various *spiro*-nitrogen heterocycle compounds can be shown below (Scheme 5). Initially, in the reaction vessel, ninhydrin **1** reacted with 1,2-diaminobenzene **2** and produced an intermediate of the indenoquinoxalin **6**. At the same time, nitrogen isoquinoline **3** attacked the dialkylacetylenedicarboxylate **4** and produced an intermediate **7**. Then, the carbonyl group of intermediate **6** was attacked by carbon with a negative average of intermediate **7** to furnish intermediate **8**. In the following, by forming a ring, the final product **5** is produced.



Scheme 5. A plausible mechanism for compound **5**.

EXPERIMENTAL

Chemicals and apparatus

Solvents, ninhydrin, 1,2-diaminobenzene, isoquinoline, quinoline, pyridine and dialkylacetylenedicarboxylate were obtained from Merck, Fluka, and Aldrich, and were used without further purification. Electrothermal-9100 was used to measure the melting temperature. Elemental analysis was performed with Heraeus CHN-O-Rapid. ^{1}H, ^{13}C NMR spectra were recorded with a Bruker DRX-500 Avance instrument using $CDCl_3$ as the deuterated solvent containing TMS as internal standard, at 300, 500.1, 125.8 MHz, respectively; δ in ppm, J in Hz. IR Spectra (ν/cm^{-1}) were recorded as KBr pellets with a Shimadzu IR-460 spectrometer.

*General procedure for the preparation of compounds **5a-f***

To a mixture of (1 mmol) ninhydrin and (1 mmol) of 1,2-diaminobenzene in 10 mL of acetonitrile solvent, (1 mmol) isoquinoline was added and the solution was stirred. Then (1 mmol) of dialkylacetylenedicarboxylate in 2 mL of acetonitrile was dropwise added to the solution for 15 min at room temperature. Then the reaction mixture was allowed to warm to 50-60 °C and was stirred for 12 hours. The product was filtered and the solvent was removed under reduced pressure. The remaining residue was recrystallized and poured from ethanol.

*Dimethyl 11b'H-spiro[indeno[1,2-b]quinoxaline-11,2'-[1,3]oxazino[2,3-a]isoquinoline]-3',4'-dicarboxylate (**5a**)*. Yellow powder; m.p. = 240; yield: 0.448 g (89%); IR (KBr) (ν_{max}/cm^{-1}) = 1700, 1736 (2C=O); ^{1}H NMR: δ 3.23 (3H, s, MeO), 4.07 (3H, s, MeO), 5.91 (1H, d, ^3J = 7.9 Hz, =CH), 6.57 (1H, d, ^3J = 7.9 Hz, =CH), 6.88 (1H, s, CH), 7-8.40 (12H, m, =CH); ^{13}C NMR: δ 51.45, 53.41 (2OMe), 80.82 (CH), 105.12 (C_{spiro}), 122.72, 123.43, 123.97, 125.18, 126.37, 127.06, 127.78, 128.81, 129.07, 129.45, 129.73, 129.85, 130.10, 130.41 (14=CH), 132.33, 137.80, 141.36, 142.06, 145.50, 147.49, 154.23 (10C), 162.33, 163.90 (2C=O). Anal. calcd. for $C_{30}H_{21}N_3O_5$ (503.15): C, 71.56; H, 4.20; N, 8.35; O, 15.89%. Found: C, 71.54; H, 4.23; N, 8.32; O, 15.91%.

*Diethyl 11b'H-spiro[indeno[1,2-b]quinoxaline-11,2'-[1,3]oxazino[2,3-a]isoquinoline]-3',4'-dicarboxylate (**5b**)*. Yellow powder; m.p. = 240; yield: 0.462 g (87%); IR (KBr)(ν_{max}/cm^{-1}) = 1701, 1736 (2C=O); NMR data for the major isomer (60%): ^{1}H NMR: δ 0.51(3H, t, ^3J = 7.1 Hz, Me), 1.49 (3H, t, ^3J = 7.1 Hz, Me), 3.60 (2H, m, OCH_2), 4.53 (2H, m, OCH_2), 5.89 (1H, d, ^3J = 7.8 Hz, =CH), 6.59 (1H, d, ^3J = 7.7 Hz, =CH), 6.90 (1H, s, CH), 7-8.30 (12H, m, =CH); ^{13}C NMR: δ 13.19 (Me), 13.96 (Me), 60.19 (OCH_2), 62.79 (OCH_2), 80.77 (CH), 104.79 (C_{spiro}), 122.33, 123.44, 124.09, 125.12, 126.45, 126.98, 127.82, 128.94, 129.08, 129.40, 129.84, 129.91, 130.28, 130.41 (14=CH), 131.81, 132.14, 137.12, 138.26, 141.32, 142.53, 145.75, 147.65, 149.01, 154.23 (10C), 163.19, 163.39 (2C=O); NMR data for the minor isomer (40%): ^{1}H NMR: δ 0.57 (3H, t, ^3J = 7.1 Hz, Me), 1.50 (3H, t, ^3J = 7.1 Hz, Me), 3.69 (2H, m, OCH_2), 4.56 (2H, m, OCH_2), 5.91 (1H, d, ^3J = 7.8 Hz, =CH), 6.59 (1H, d, ^3J= 7.7 Hz, =CH), 6.90 (1H, s, CH), 7-8.30 (12H, m, =CH); ^{13}C NMR: δ 13.11 (Me), 13.93 (Me), 60.24 (OCH_2), 62.79 (OCH_2), 80.23 (CH), 106.13 (C_{spiro}), 122.68, 123.91, 125.07, 125.21, 126.24, 126.96, 127.36, 128.79, 128.85, 129.46, 129.79, 129.86, 130.05, 130.12 (14=CH), 131.81, 132.14, 137.12, 138.26, 141.82, 142.38, 145.57, 147.65, 149.01, 154.23 (10C), 162.21, 162.42 (2C=O). Anal. calcd. for $C_{32}H_{25}N_3O_5$ (531.18): C, 72.31; H, 4.74; N, 7.91; O, 15.05%. Found: C, 72.28; H, 4.70; N, 7.93; O, 15.10%.

*Dimethyl 4a'H-spiro[indeno[1,2-b]quinoxaline-11,3'-[1,3]oxazino[3,2-a]quinoline]-1',2'-dicarboxylate (**5c**)*. Yellow powder; m.p. = 240; yield: 0.427 g (85%); IR (KBr) (ν_{max}/cm^{-1}) = 1707, 1713 (2C=O); ^{1}H NMR: δ 3.27 (3H, s, OMe), 3.93 (3H, s, OMe), 5.95 (1H, dd, ^3J = 9.8 Hz, =CH), 6.52 (1H, d, ^3J = 4.3 Hz, =CH), 6.83 (1H, d, ^3J = 9.8 Hz, CH), 7.05-8.16 (12H, m, =CH); ^{13}C NMR: δ 52.02, 53.27 (2OMe), 81.03 (CH), 114.31 (C_{spiro}), 118.58, 121.79, 122.42, 122.45, 123.75, 128.65, 128.93, 129.22, 129.24, 129.27, 129.79, 129.84, 129.93 (14=CH), 130.42, 132.13, 135.89, 138.29, 141.41, 142.89, 143.67, 146.46, 154.01, 161.24 (10C), 164.02, 164.45 (2C=O). Anal. calcd. for $C_{30}H_{21}N_3O_5$ (503.15): C, 71.56; H, 4.20; N, 8.35; O, 15.89%. Found: C, 71.53; H, 4.25; N, 8.33; O, 15.88%.

*Diethyl 4a'H-spiro[indeno[1,2-b]quinoxaline-11,3'-[1,3]oxazino[3,2-a]quinoline]-1',2'-dicarboxylate (**5d**)*. Yellow powder; m.p. = 240; yield: 0.409 g (77%); IR (KBr)(ν_{max}/cm^{-1}) = 1726 (2C=O), NMR data for the major isomer (60%): ^{1}H NMR: δ 0.60 (3H, t, ^3J = 7.1 Hz, Me), 1.31 (3H, t, ^3J = 7.1 Hz, Me), 3.72 (2H, m, OCH_2), 4.41 (2H, m, OCH_2), 5.95 (1H, dd, ^3J = 9.6 Hz, =CH), 6.01 (1H, d, ^3J = 4.7 Hz, =CH), 6.88 (1H, d, ^3J = 9.6 Hz, CH), 7.02-8.20 (12H, m, =CH); ^{13}C NMR: δ 13.18 (Me), 13.96 (Me), 60.20 (OCH_2), 62.78 (OCH_2), 81.03 (CH), 114.31 (C_{spiro}),

118.58, 121.79, 122.42, 122.45, 123.75, 128.65, 128.93, 129.22, 129.24, 129.27, 129.79, 129.84, 129.93 (14=CH), 130.42, 132.13, 135.89, 138.29, 141.41, 142.89, 143.67, 146.46, 154.01, 161.24 (10C), 164.02, 164.45 (2C=O); NMR data for the minor isomer (40%): ^{1}H NMR: δ 0.59 (3H, t, ^3J = 7.1 Hz, Me), 1.31(3H, t, ^3J = 7.1 Hz, Me), 3.60 (2H, m, OCH_2), 4.32 (2H, m, OCH_2), 5.95 (1H, dd, ^3J = 9.6 Hz, =CH), 6.01 (1H, d, ^3J = 4.7 Hz, =CH), 6.88 (1H, d, ^3J = 9.6 Hz, CH), 7.02-8.20 (12H, m, =CH); ^{13}C NMR: δ 13.18 (Me), 13.96 (Me), 60.20 (OCH_2), 62.78 (OCH_2), 79.93 (CH), 114.31(C_{spiro}), 118.58, 121.79, 122.42, 122.45, 123.75, 128.65, 128.93, 129.22, 129.24, 129.27, 129.79, 129.84, 129.93 (14=CH), 130.42, 132.13, 135.89, 138.29, 141.41, 142.89, 143.67, 146.46, 154.01, 161.24 (10C), 164.02, 164.45 (2C=O). Anal. calcd. for $C_{32}H_{25}N_3O_5$ (531.18): C, 72.31; H, 4.74; N, 7.91; O, 15.05%. Found: C, 72.35; H, 4.73; N, 7.89; O, 15.07%.

*Dimethyl 9a'H-spiro[indeno[1,2-b]quinoxaline-11,2'-pyrido[2,1-b][1,3]oxazine]-3',4'-dicarboxylate (**5e**)*. Yellow powder; m.p. = 192 °C; yield: 0.390 g (86%); IR (KBr) (ν_{max}/cm^{-1}) = 1737 (2C=O), 1259; ^{1}H NMR: δ 1.15 (6H, s, 2MeO), 5.32 (1H, dd, ^3J= 6.7 Hz, =CH), 5.49(1H, dd, ^3J = 9.9 Hz, =CH), 6.20 (1H, dd, ^3J = 9.9 Hz, =CH), 6.45 (1H, d, ^3J = 7.5 Hz, =CH), 6.96 (1H, dd, ^3J = 3.2 Hz, CH), 7.42-8.17 (8H, m, =CH); ^{13}C NMR: δ 53.42, 57.99 (2OMe), 79.37 (CH), 101.69 (C_{spiro}), 107.28, 116.48, 122.42, 123.66, 124.84, 125.11, 128.93, 129.04, 129.73, 129.91, 130.34 (12=CH), 132.09, 138.28, 141.21, 142.58, 145.50, 147.28, 153.91, 161.70(8C), 163.54, 163.77 (2C=O). Anal. calcd. for $C_{26}H_{19}N_3O_5$ (453.45): C, 68.87; H, 4.22; N, 9.27; O, 17.64%. Found: C, 68.79; H, 4.23; N, 9.30; O, 17.66%.

*Diethyl 9a'H-spiro[indeno[1,2-b]quinoxaline-11,2'-pyrido[2,1-b][1,3]oxazine]-3',4'-dicarboxylate (**5f**)*. Yellow powder; m.p. = 198 °C; yield: 0.380 g (79%); IR (KBr) (ν_{max}/cm^{-1}) = 1695, 1734 (2C=O); NMR data for the major isomer (60%): ^{1}H NMR: δ 0.50 (3H, t, ^3J = 7.1 Hz, Me), 1.44(3H, t, ^3J = 7.1 Hz, Me), 3.59 (2H, m, OCH_2), 4.48 (2H, m, OCH_2), 5.35 (1H, dd, ^3J = 10 Hz, =CH), 5.54 (1H, dd, ^3J = 9.9 Hz, =CH), 6.24 (1H, dd, ^3J = 9.9 Hz, =CH), 6.51 (1H, d, ^3J = 7.6 Hz, =CH), 7.03 (1H, dd, ^3J = 3.2 Hz, CH), 7.50-8.16 (8H, m, =CH); ^{13}C NMR: δ 13.15 (Me), 13.96 (Me), 60.23 (OCH_2), 62.78 (OCH_2), 79.44 (CH), 101.38 (C_{spiro}), 107.19, 116.47, 122.27, 123.81, 124.84, 125.11, 128.84, 129.16, 129.78, 130.23 (12=CH), 132.04, 138.53, 141.24, 142.69, 145.76, 147.56, 154.12, 161.91 (8C), 163.08, 163.35 (2C=O); NMR data for the minor isomer (40%): ^{1}H NMR: δ 0.50 (3H, t, ^3J = 7.1 Hz, Me), 1.44 (3H, t, ^3J = 7.1 Hz, Me), 3.60 (2H, m, OCH_2), 4.49 (2H, m, OCH_2), 5.35 (1H, dd, ^3J = 10 Hz, =CH), 5.54 (1H, dd, ^3J = 9.9 Hz, =CH), 6.24 (1H, dd, ^3J = 9.9 Hz, =CH), 6.51 (1H, d, ^3J = 7.6 Hz, =CH), 7.03 (1H, dd, ^3J = 3.2 Hz, CH), 7.50-8.16 (8H, m, =CH); ^{13}C NMR: δ 13.15 (Me), 13.96 (Me), 60.23 (OCH_2), 62.78 (OCH_2), 79.99 (CH), 101.38 (C_{spiro}), 107.19, 116.47, 122.27, 123.81, 124.84, 125.11, 128.84, 129.16, 129.78, 130.23 (12=CH), 132.04, 138.53, 141.24, 142.69, 145.76, 147.56, 154.12, 161.91 (8C), 163.08, 163.35 (2C=O). Anal. calcd. for $C_{28}H_{23}N_3O_5$ (481.51): C, 69.84; H, 4.81; N, 8.73; O, 16.61%. Found: C, 69.80; H, 4.83; N, 8.75; O, 16.60%.

CONCLUSIONS

In this work, one-pot, mild and efficient synthesis of various *spiro*-nitrogen heterocycle compounds based on the reaction of ninhydrin and 1,2-diaminobenzene with isoquinoline or quinoline or pyridine and dialkylacetylenedicarboxylate in CH_3CN at 50-60 °C is described. Using this approach, all *spiro*-nitrogen heterocycle compounds are obtained in high yields. The method offers several advantages including high diversity via various functional groups, operational simplicity and high yields.

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REFERENCES

1. Nasri, S.; Bayat, M.; Vasheghani Farahani, H.;Karami, S. Synthesis of new functionalized thiazolo pyridine-fused and thiazolopyridopyrimidine-fused spirooxindoles *via* one-pot reactions. *Heliyon* **2020**, 6, e03687.
2. Dömling, A.; Ugi, I. Multicomponent reactions with isocyanides. *Angew. Chem. Int. Ed*. **2000**, 39, 3168–3210.
3. Poor Heravi, M.R. An efficient synthesis of bis-indolylindane-1,3-diones, indan-1,3-diones, and indene-1,3(2H)-denies using [Hbim]BF_4 ionic medium. *ISRN Org. Chem.* **2013**, 2013, Article ID 528329, 8 pages. http://dx.doi.org/10.1155/2013/ 528329.
4. Campos, J.F.; Berteina-Raboin, S. Greener synthesis of nitrogen-containing heterocycles in water, PEG, and bio-based solvents. *Catalysts* **2020**, 10, 429.
5. Singh, R.; Bhardwaj, D.; Ram Saini, M. Recent advancement in the synthesis of diversespiro-indeno[1,2-*b*]quinoxalines: a review. *RSC Adv*. **2021**, 11, 4760.
6. Zangouei, M.; Esmaeili, A.A. One-pot, catalyst-free synthesis of novelspiro[indole-3,4'-pyrano[2',3':4,5]pyrimido[2,1-*b*][1,3]benzothiazole] derivatives. *J. Chem. Res*. **2020**, 44, 646–652.
7. Dömling, A. Recent developments in isocyanide based multicomponent reactions in applied chemistry. *Chem. Rev*. **2006**, 106, 17–89.
8. Naskar, S.; Paira, P.; Paira, R.; Mondal, S.; Maity, A.; Hazra, A.; Sahu, K.B.; Saha, P.; Banerjee, S.; Luger, P.; Webe, M.; Mondal, N.B. Montmorillonite K-10 clay catalyzed solvent-free synthesis of bis-indolylindane-1,3-dione, 2-(1',3'-dihydro-1*H*-[2,3']biindolyl-2'-ylidene)-indan-1,3-dioneand bisindolylindeno[1,2-*b*]quinoxaline under microwave irradiation. *Tetrahedron* **2010**, 66, 5196–5203.
9. Suresh Babu, A.R.; Raghunathan, R. TiO_2-mediated, one-pot, four-component1,3-dipolar cycloaddition reaction: A facile synthesis of dispiro-pyrrolidine ring systems. *Synth. Commun*. **2009**, 39, 347–354.
10. Nahid, S.; Mahdavinia, G.H.; Notash, B. A synthesis of spirofuran-indenoquinoxalines via isocyanid-based one-pot four-component reaction. *Chin. Chem. Lett*. **2016**, 27, 1040–1043.
11. Selvarangam, E.K.; Neelakandan Vidhya, L.; Babu Rasidha, B.; Paramasivan, T.P. A facile strategy for the one pot multicomponent synthesis of spirodihydropyridines from amines and activated alkynes. *Tetrahedron Lett*. **2011**, 52, 6508–6511.
12. Azizian, J.; Karimi, A.R.; Dastkhan, R.; Mohammadi, A.A.; Mohammadizadeh, M.R. One-pot highly diastereoselective synthesis of some novel spiropyrrolizidines via 1,3-dipolar cycloaddition reaction of azomethineylide. *J. Chem. Res*. **2004**, 5, 347–349.
13. Cheng, C.; Jiang, B.; Tu, S-J.; Li, G. [4+2+1] Domino cyclization in water for chemo- and regioselective synthesis of spiro-substituted benzo[b]furo[3,4-e][1,4]diazepine derivatives. *Green Chem*. **2011**, 13, 2107–2115.
14. Kucukguzel, S.G.; Oruc, E.F.; Rollas, S.; Shahin, F.; Ozbek, A. Synthesis, characterisation and biological activity of novel 4-thiazolidinones, 1,3,4-oxadiazoles and some related compounds. *Eur. J. Med. Chem.* **2002**, 37, 197–206.
15. Kathiravan, S.; Raghunathan, R. Expedient synthesis of novel ferrocenyl spiropyrrolidines through 1,3-dipolar cycloaddition reaction. *J. Heterocycl. Chem*. **2014**, 51, 906–910.
16. Rao, V.R.; Kumar, R.A.; Rao, T.V.P. Synthesis of 11H-indeno[1,2-*b*] quinoxaline-11-one azines containing 4-substitutedphenyl-3-thiazolin-2-one moiety. *Indian J. Chem. Sect. B* **1984**, 6, 563–564.

17. Li, M.; Gong, F.-M.; Wen, L.-R.; Li, Z.-R. Expeditious construction of spiro-pyrrolidines by an autocatalytic one-pot, five-component, 1,3-dipolar cycloaddition of in situ generated azomethine ylides and olefinic dipolarophiles. *Eur. J. Org. Chem*. **2011**, 19, 3482–3490.
18. Valluri, K.K.; Kasi Viswanath, I.V.; Naga Raju, P.V.V.S.; Rajasekher, B.; Rama Dasu, B. Synthesis and in-vitro studies of some new quinoline 1,3,4-thiadiazolo pyrimidin derivatives. *Bull. Chem. Soc. Ethiop.* **2017**, 31*,* 337–344.
19. Liao, L.G.; Song, M.M.; Feng, J.F.; Tan, M.; Liu, F.; Qiu, Z.J.; Zhang, S.; Li, B.J. Green synthesis of indeno[1,2-b]quinoxalines using β-cyclodextrin as catalyst. *Molecules* **2022**, 27, 580.
20. Borah, B.; Bora, J.; Ramesh, P.; Chowhan, L.R. Sonochemistry in an organocatalytic domino reaction: An expedient multicomponent access to structurally functionalized dihydropyrano[3,2-b] pyrans, spiro-pyrano[3,2-b]pyrans, and spiroindenoquinoxaline-pyranopyrans under ambient conditions. *RSC Adv*. **2022**, 12, 12843–12857.
21. Yavari, I.; Solgi, R.; Khajeh-Khezri, A.; Askarian-Amiri, M.; Halvagar, M.R. Synthesis of spiroindolo[2,1-*b*]quinazolines from Huisgen zwitterions and tryptanthrin-malononitrile adducts. *J. Heterocycl. Chem*. **2019**, 1–7.
22. Yavari, I.; Safaei, M.; Alireza Abbasi, A. Ph_3P-mediated synthesis of fused 1,2-dihydropyridines. *Monatsh. Chem.* **2020**, 151, 107–112.
23. Yavari, I.; Sheikhi, S.; Sheykhahmadi, J.; Taheri, Z.; Halvagar, M.R. Ultrasound-promoted synthesis of spirocyclopropanes from switchable starting materials *via* azomethine ylide [3+2]-cycloaddition. *Synthesis* **2021**, 53, 2057–2066.
24. Azad, L.; Ghazvini, M.; Sanaeishoar, H.; Yavari, I. Synthesis of functionalized 1,2-dihydro-isoquinolines via one-pot reactions of isoquinoline, alkyl propiolate, and thiazolidin-2,4-dione. *J. Chem. Res.* **2019**, 43, 457–460.
25. Yavari, I.; Askarian-Amiri, M. A synthesis of spiroindolo[2,1-*b*]quinazoline-6,2'-pyrido [2,1-*b*][1,3]oxazines from tryptanthrins and Huisgen zwitterions. *Synth. Commun*. **2021**, 51, 1602–1608.
26. Yavari, I.; Hojati, M.; Azad, L.; Halvagar, M.R. A synthesis of spirocyclic oxazinoisoquinolines and oxazinoquinolines bearing thiazolopyrimidine moieties. *Synlett.* **2018**, 29, 1024–1027.
27. Yavari, I.; Piltan, M.; Moradi, L. Synthesis of pyrrolo[2,1-a]isoquinolines from activated acetylenes, benzoylnitromethanes, and isoquinoline. *Tetrahedron* **2009**, 65, 2067–2071.
28. Kamal, A.; Reddy, B.V.S.; Sridevi, B.; Ravikumar, A.; Venkateswarlu, A.; Sravanthi, G.; Sridevi, J.P.; Yogeeswari, P.; Sriram, D. Synthesis and biological evaluation of phaitanthrin congeners as anti-mycobacterial agents. *Bioorg. Med. Chem. Lett*. **2015**, 25, 3867–3872.
29. Azizian, J.; Karimi, A.R.; Kazemizadeh, Z.; Mohammadi, A.A.; Mohammadizadeh, M.R.A Novel one-pot synthesis of some new interesting pyrrole derivatives. *J. Org. Chem*. **2005**, 70, 1471–1473.
30. Nishtala, V.B.; Nanubolu, J.B.; Basavoju, S. Ultrasound-assisted rapid and efficient one-pot synthesis of furanyl spirooxindolo and spiroquinoxalinopyrrolizidines by 1,3-dipolar cycloaddition: A green protocol. *Res. Chem. Intermed*. **2017**, 43, 1365–1381.
31. Rashid, Z.; Naeimi, H.; Ghahremanzadeh, R. Highly efficient one-pot four-component Kabachnik-Fields synthesis of novel α-amino phosphonates under solvent-free and catalyst-free conditions. *RSC Adv*. **2015**, 5, 99148–99152.
32. Bagley, M.C.; Dale, J.W.; Bower, J. A new one-pot three-component condensation reaction for the synthesis of 2,3,4,6-tetrasubstituted pyridines. *Chem. Commun*. **2002**, 16, 1682–1683.
33. Velikorodov, A.V.; Stepkina, N.N.; Shustova, E.A.; Ionova, V.A. Synthesis of new spirocompounds proceeding from 11*H*-indeno[1,2-*b*]quinoxalin-2-one. *Russ. J. Org. Chem.* **2015**, 51, 674–679; *Zh. Org. Khim*. **2015**, 51, 693–697.
34. Moemeni, M.; Arvinnezhad, H.; Samadi, S.; Tajbakhsh, M.; Jadidi, K.; Khavasi, H. R. An efficient multicomponent and stereoselective synthesis of new spiro[indeno[1,2-

b]quinoxaline-11,2'-pyrrolidine] derivatives. *J. Heterocycl. Chem*. **2012**, 49, 190–194.
35. Middleton, R.J.; Mellor, S.L.; Chhabra, S.R.; Bycroft, B.W.; Chan, W.C. Expedient synthesis of a novel class of pseudoaromatic amino acids: Tetrahydroindazol-3-yl- and tetrahydrobenzisoxazol-3-ylalanine derivatives. *Tetrahedron Lett*. **2004**, 45, 1237–1242.
36. Tenorio, R.P.; Carvalho, C.S.; Pessanha, C.S.; de Lima, J.G.; de Faria, A.R.; Alves, A.J.; de Melo, E.J.T.; Goes, A.J.S. Synthesis of thiosemicarbazone and 4-thiazolidinone derivatives and their in vitro anti-*toxoplasmagondii* activity. *Bioorg. Med. Chem. Lett*. **2005**, 15, 2575–2578.
37. Carpino, P.A.; Griffith, D.A.; Sakya, S.; Dow, R.L.; Black, S.C.; Hadcock, J.R.; Iredale, P.A.; Scott, D.O.; Fichtner, M.W.; Rose, C.R.; Day, R.; Dibrino, J.; Butler, M.; DeBartolo, D.B.; Dutcher, D.; Gautreau, D.; Lizano, J.S.; O'Connor, R.E.; Sands, M.A.; Kelly-Sullivan, D.; Ward, K.M. New bicyclic cannabinoid receptor-1 (CB_1-R) antagonists. *Bioorg. Med. Chem. Lett*. **2006**, 16, 731–736.
38. Azizian, J.; Shaabanzadeh, M.; Hatamjafari, F.; Mohammadizadeh, M.R. One-pot rapid and efficient synthesis of new spiro derivatives of 11*H*-indeno[1,2-*b*]quinoxalin-11-one, 6*H*-indeno[1,2-*b*]pyrido[3,2-*e*]pyrazin-6-one and isatin-based 2-pyrazolines. *Arkivoc.* **2006**, 11, 47-58.
39. Maghsoodlou, M.T.; Habibi-Khorassani, S.M.; Moradi, A.; Hazeri, N.; Amir Davodi, A.; Sajadikhah, S.S. One-pot three-component synthesis of functionalized spirolactones by meansof reaction between aromatic ketones, dimethyl acetylenedicarboxylate, and *N*-heterocycles. *Tetrahedron* **2011**, 67, 8492–8495.
40. Pfeiffer, P.; Milz, E. Kondensationsprodukte des Indanons-(1) mitnitrosokörpern und aldehyden. *Chem. Ber.* **1938**, 71, 272–279.
41. Li, Y.X.; Wang, S.H.; Li, Z.M.; Su, N.; Zhao, W.G. Synthesis of novel 2-phenylsulfonyl-hydrazono-3-(2',3',4',6'-tetra-*O*-acetyl-β-D-glucopyranosyl)thiazolidine-4-ones from thio-semicarbazide precursors. *Carbohydr. Res*. **2006**, 341, 2867–2870.
42. Pearson, B.D.; Mitsch, R.A.; Cromwell, N.H. Indenoquinolines. III. Derivatives of 11*H*-indeno[1,2-*b*]quinoxaline and related indenoquinolines. *J. Org. Chem*. **1962**, 27, 1674–1678.
43. Kathiravan, S.; Raghunathan, R.; Suresh, G.; Siva, G.V. Synthesis and antimicrobial activities of novel ferrocenyldispiropyrrolidines and pyrrolizidines*. Med. Chem. Res.* **2012**, 21, 3170–3176.
44. Ameri Rad, J.; Jarrahpour, A.; Ersanlı, C.C.; Atioğlu, Z.; Akkurt, M.; Turos, E. Synthesis of some novel indeno[1,2-*b*]quinoxalinspiro-*β*-lactam conjugates. *Tetrahedron* **2017**, 73, 1135–1142.