

## Polyethylene glycol in water: A simple, efficient and green protocol for the synthesis of quinoxalines

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**Abstract.** A variety of biologically important quinoxaline derivatives has been efficiently synthesized in excellent yields under extremely mild conditions using PEG-600 and water. This inexpensive, non-toxic, eco-friendly and readily available system efficiently condensed several aromatic as well as aliphatic 1,2-diketones with aromatic and aliphatic 1,2-diamines to afford the products in excellent yield. Polyethylene glycol (PEG) can be recovered and recycled.

**Keywords.** Polyethylene glycol (PEG); water; 1,2-dicarbonyls; 1,2-diamines; recyclability; quinoxalines.

### 1. Introduction

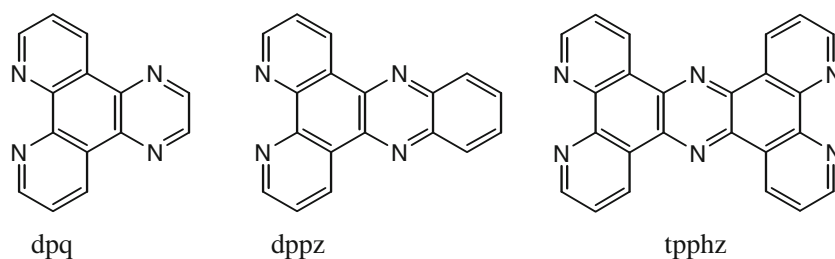
Quinoxaline derivatives are important class of nitrogen-containing heterocycles, having interesting therapeutic properties such as antiviral, antibacterial, antibiotic, antiinflammatory and kinase inhibition.<sup>1</sup> They have also been evaluated as anticancer, antimycobacterial, and anthelmintic agents.<sup>2</sup> Besides this, they are potential building blocks for the synthesis of anion receptors,<sup>3</sup> cavitands,<sup>4</sup> dehydroannulenes,<sup>5</sup> organic semiconductors,<sup>6</sup> and dyes.<sup>7</sup> Molecules like dipyrido[3,2-a:2',3'-c]phenazine (dppz),<sup>8</sup> dipyrido[3,2-f:2',3'-h]quinoxaline (dpq)<sup>9</sup> and tetrapyrido[3,2-a:2',3'-c:3'',2''-h:2''',3'''-j]phenazine (tpphz)<sup>10</sup> derivatives exhibit interesting properties, such as bidentate coordination ability, rigidity,  $\pi$ -accepting character and planar highly conjugated aromatic structure (figure 1). All these interesting properties and nitrogen sites enable them to act as therapeutic agents that are capable of binding and cleaving DNA under the physiological condition.<sup>11</sup> In addition, organometallic complexes of these ligands show interesting optical, electrochemical and electroluminescent properties.<sup>12</sup>

A number of methods have been developed for the synthesis of substituted quinoxaline derivatives and the most common method is the condensation of an aryl 1,2-diamine with 1,2-dicarbonyl compounds

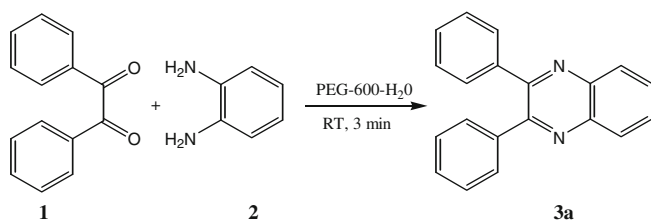
in refluxing ethanol or acetic acid.<sup>13</sup> Apart from this, other methods such as solid phase synthesis,<sup>14</sup> oxidative cyclisation of  $\alpha$ -hydroxy ketones with 1,2-diamines,<sup>15</sup> cyclisation-oxidation of phenacyl bromides with 1,2-diamines by  $\text{HClO}_4\text{-SiO}_2$ ,<sup>16</sup> cyclisation of *o*-phenylenediamine with propiophenone by using KOH,<sup>17</sup> and oxidative coupling of epoxides with ene-1,2-diamines<sup>18</sup> have also been reported. Nevertheless, most of the methods suffer from several drawbacks such as critical product isolation, expensive metal catalyst, harsh reaction conditions, and use of strong oxidizing agents, long reaction time and low yields. Therefore, the development of simple, more efficient and environmentally benign method is useful in generating organic compounds in drug discovery process.

An increase in regulatory restrictions on the use, manufacture and disposal of organic solvents has focused attention on the development of non-hazardous alternatives for the sustainable development of chemical enterprise. Liquid polymers have been used as green reaction media with unique properties such as thermal stability, commercial availability, non-volatility, immiscibility with organic solvents and recyclability. Recently, polyethylene glycol (PEG) and its aqueous solutions represent interesting solvent systems for solvent replacement, and may stand comparison to other currently favoured systems such as ionic liquids, supercritical carbon dioxide, and micellar systems.<sup>19</sup> PEGs are selected instead of other polymers because they are inexpensive, biodegradable, non-halogenated and having low toxicity.

\*For correspondence



**Figure 1.** Representatives of quinoxaline ring containing compounds.



**Scheme 1.** Synthesis of quinoxaline.

As a part of ongoing program on the development of novel methods in organic synthesis,<sup>20</sup> we report here a simple, efficient and green protocol for the synthesis of quinoxalines. In this protocol, 1,2-diketone **1** and 1,2-diamine **2** in PEG-H<sub>2</sub>O after stirring at room temperature for specified time resulted in the formation of product **3a** in 98% yield (scheme 1).

## 2. Experimental

### 2.1 General procedure for the synthesis of quinoxalines

A mixture containing 1,2-dicarbonyl compound **1** (1 mmol) and 1,2-diamine **2** (1 mmol) in PEG-600-H<sub>2</sub>O (1:1, 5 ml) was stirred at room temperature for the specified time. After completion of the reaction (TLC), water (10 ml) was added to the reaction mixture. Precipitate obtained was filtered, washed with water and dried to obtain quinoxaline **3** in almost pure form. All the products were characterized by IR, <sup>1</sup>H NMR, <sup>13</sup>C NMR, and Mass Spectroscopy.

### 2.2 Spectral data of some representative compounds

**2.2a 2,3-Diphenyl-quinoxaline (3a):** White solid; yield 98%; mp 126°C; IR (KBr): 3056, 1964, 1577, 1556, 1540, 1495 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, DMSO-d<sub>6</sub>): δ 7.35 (m, 6H), 7.50 (m, 4H), 7.90 (dd, *J* = 8, 2 Hz, 2H), 8.20 (d, *J* = 8, 2 Hz, 2H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 128.2, 128.8, 129.2, 129.8, 129.9, 139.1, 141.2, 153.5; MS (ESI): *m/z* 283 (M+H)<sup>+</sup>.

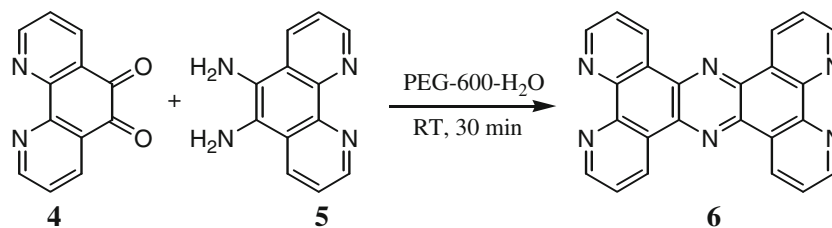
**2.2b 2,3-Bis-(4-fluoro-phenyl)-quinoxaline (3b):** Colourless solid; yield 96%; mp 104°C; IR (KBr): 3074, 3061, 1664, 1599, 1512 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, DMSO-d<sub>6</sub>): δ 7.24 (m, 4H), 7.52 (m, 2H), 7.90 (m, 2H), 8.05 (m, 2H), 8.20 (m, 2H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 128.4, 129.2, 130.1, 132.6, 133.4, 144.2, 155.1; MS (ESI): *m/z* 319 (M+H)<sup>+</sup>.

**2.2c Dipyrido[3,2-*a*:2',3'-*c*]phenazine (dppz) (3c):** Light brown; yield 95%; mp 246°C; IR (KBr): 3079, 3052, 3025, 1590, 1492, 1081 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, DMSO-d<sub>6</sub>): δ = 9.68 (dd, *J* = 8.2, 1.3 Hz, 2H); 7.95 (dd, *J* = 8.2, 5.4 Hz, 2H); 8.30 (dd, *J* = 5.4, 1.3 Hz, 2H); 8.42 (d, *J* = 8.0 Hz; 2H); 8.14 (d, *J* = 8.0 Hz, 2H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 125.8, 129.6, 130.2, 130.8, 131.1, 138.4, 144.6, 147.8, 153.2; MS (ESI): *m/z* 283 (M+H)<sup>+</sup>.

**2.2d Dibenz[*a,c*]phenazine (3d):** Yellow solid; yield 97%; mp 218°C; IR (KBr): 3059, 3032, 1604, 1500 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, DMSO-d<sub>6</sub>): δ = 7.82 (m, 2H), 7.90 (m, 2H), 8.00 (m, 2H), 8.35 (m, 2H), 8.80 (d, *J* = 8 Hz, 2H), 9.28 (d, *J* = 8 Hz, 2H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 126.2, 127.1, 127.6, 128.1, 129.8, 130.7, 132.4, 135.9, 142.6, 151.2; MS (ESI): *m/z* 281 (M+H)<sup>+</sup>.

**2.2e 2,3-Difuran-2-yl-quinoxaline (3e):** Pale brown solid; yield 97%; mp 130°C; IR (KBr): 3107, 3061, 1572, 1529, 1400 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, DMSO-d<sub>6</sub>): δ = 6.72 (s, 4H), 7.90 (m, 4H), 8.10 (m, 2H); <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>): δ 120.1, 129.8, 132.2, 144.6, 146.5, 148.4, 156.9; MS (ESI): *m/z* 263 (M+H)<sup>+</sup>.

**2.2f 2,3-Diphenylpyrido[2,3-*b*]pyrazine (3g):** Yellow solid, yield 94%; mp 257°C; IR (KBr): 1588, 1545, 1430, 1382, 1328 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, DMSO-d<sub>6</sub>): δ = 7.31–7.41 (m, 6H), 7.56 (d, *J* = 7.8 Hz, 2H), 7.64 (d, *J* = 7.8 Hz, 2H), 7.74–7.76 (dd, *J* = 8.4, 4.1 Hz, 1H), 8.55–8.57 (d, *J* = 8.2 Hz, 1H), 9.19 (d, *J* = 4.2 Hz,



**Scheme 2.** Synthesis of tetrapyrido[3,2-*a*:2',3'-*c*:3'',2''-*h*:2''',3'''-*j*]phenazine (tpphz).

1H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  125.2, 128.2, 128.5, 129.4, 129.6, 129.8, 130.3, 136.2, 138.0, 138.5, 149.4, 153.6, 155.0, 156.6; MS (ESI):  $m/z$  283 ( $\text{M}^+$ ).

**2.2g** 5,6-Diphenyl-2,3-dihydro-pyrazine (**3l**): White solid; yield 94%; mp 156°C; IR (KBr): 3081, 2943, 2832, 1964, 1896, 1610, 1572  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR ( $\text{DMSO}-d_6$ ):  $\delta$  = 3.60 (s, 4H), 7.30 (m, 10H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  52.4, 128.2, 129.4, 130.8, 136.8, 161.6; MS (ESI):  $m/z$  235 ( $\text{M}+\text{H}$ ) $^+$ .

**2.2h** 1,2,3,4-Tetrahydro-phenazine (**3m**): white solid, yield 90%, mp 178°C, IR (KBr): 1459, 1423, 1384, 1330, 1291, 1238  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ )  $\delta$  = 2.04 (m, 4H), 3.16 (m, 4H), 7.65 (m, 2H), 7.97 (m, 2H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  22.8, 33.2, 128.3, 128.9, 141.2, 154.1; MS (ESI):  $m/z$  184 ( $\text{M}^+$ ).

**2.2i** 2,3,2',3'-Tetraphenyl-[6,6'] biquinoxalinyll (**3q**): Yellow solid; yield 89%; mp > 295°C; IR (KBr): 3084, 3055, 3032, 1612, 1599  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR ( $\text{DMSO}-d_6$ ):  $\delta$  = 7.35 (m, 12H), 7.55 (d,  $J$  = 8.1 Hz, 8H), 8.23 (d,  $J$  = 8.5 Hz, 2H), 8.32 (d,  $J$  = 8.5 Hz, 2H), 8.60 (s, 2H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  155.8, 154.2, 142.1, 141.7, 141.3, 140.0, 131.1, 130.8, 130.5, 130.1, 129.0, 128.3; MS (ESI):  $m/z$  563 ( $\text{M}+\text{H}$ ) $^+$ .

**Table 1.** Effect of the PEG-600 to  $\text{H}_2\text{O}$  volume ratio on quinoxaline synthesis.

Entry	PEG (ml)	$\text{H}_2\text{O}$ (ml)	Time (min/hr)	Yield (%) <sup>a</sup>
1	5	0	20 min	83
2	4	1	15 min	89
3	2.5	2.5	3 min	98
4	1	4	1.5 hr	82
5	0	5	6 hr	59

Reaction conditions: Benzil (1 mmol), *o*-phenylenediamine (1 mmol), PEG- $\text{H}_2\text{O}$ , RT

<sup>a</sup>Isolated yields after purification

**2.2j** Tetrapyrido[3,2-*a*:2',3'-*c*:3'',2''-*h*:2''',3'''-*j*]phenazine (tpphz) (scheme 2, compound **6**): Yellow solid; Yield 73%; mp > 295°C; IR (KBr): 3082, 3061, 3026, 1595, 1496, 1082  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  = 7.92 (dd,  $J$  = 8.4, 5.2 Hz, 4H); 9.40 (dd,  $J$  = 5.2, 1.7 Hz, 4H); 9.87 (dd,  $J$  = 8.4, 1.7 Hz, 4H);  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  125.2, 130.6, 131.1, 138.5, 146.8, 155.4; MS (ESI):  $m/z$  385 ( $\text{M}+\text{H}$ ) $^+$

### 3. Results and discussion

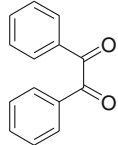
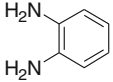
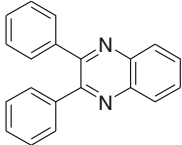
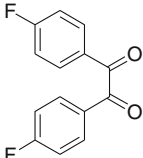
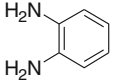
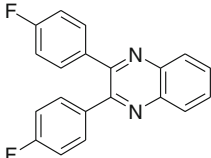
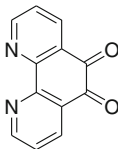
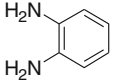
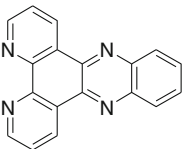
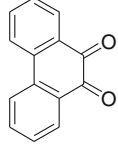
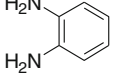
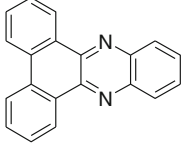
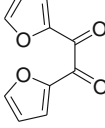
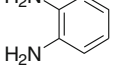
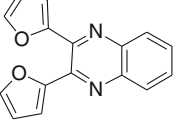
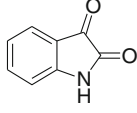
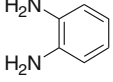
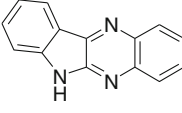
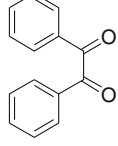
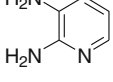
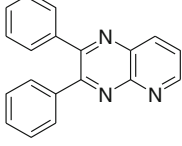
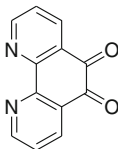
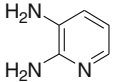
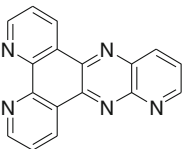
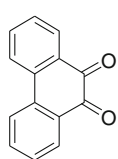
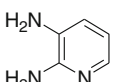
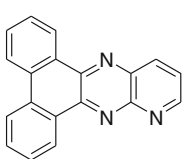
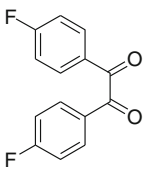
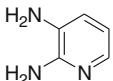
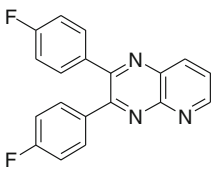
In the beginning, a reaction of benzil and *o*-phenylenediamine in water was carried out at room temperature, which resulted in the formation of a condensation product **3a** after 6 hours (59%). With similar substrates, reaction was carried out in PEG-600 afforded the title compound **3a** in 20 min (83% yield). Inspired by these results, we introduced PEG- $\text{H}_2\text{O}$  as a solvent system and found that the reaction rate was significantly improved. By changing proportion of PEG- $\text{H}_2\text{O}$ , dramatic effect on the conversion rate of quinoxaline has been observed (table 1). As shown in table 1, when pure PEG was used as a solvent, the reaction was completed in 20 min. However, addition of water to PEG increased the rate of the reaction as well as yield of the product (table 1, entries 2, 3). When the proportion of PEG- $\text{H}_2\text{O}$  was 1:1, the highest reaction rate was observed (table 1, entry 3). On the other hand,

**Table 2.** Optimisation of reaction conditions using different PEG- $\text{H}_2\text{O}$  systems.

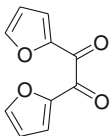
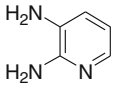
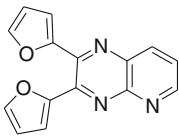
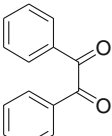

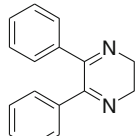

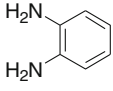
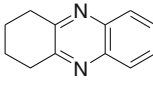

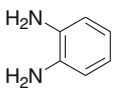
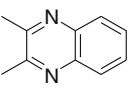

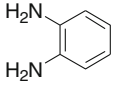
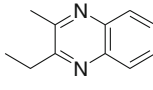

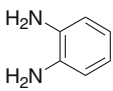
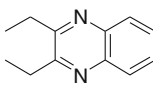
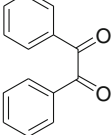
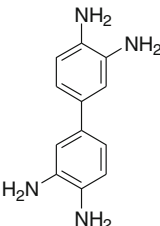
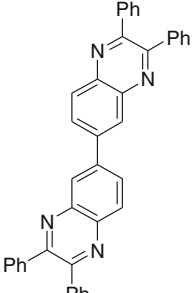
Entry	Solvent <sup>a</sup>	Time (min)	Yield (%) <sup>b</sup>
1	PEG-200- $\text{H}_2\text{O}$	12	87
2	PEG-300- $\text{H}_2\text{O}$	8	94
3	PEG-400- $\text{H}_2\text{O}$	5	97
4	PEG-600- $\text{H}_2\text{O}$	3	98
5	PEG-800- $\text{H}_2\text{O}$	5	96

<sup>a</sup>PEG- $\text{H}_2\text{O}$  volume ratio: (1:1). <sup>b</sup>Isolated yields after purification

**Table 3.** Synthesis of quinoxaline using different aromatic, heterocyclic and aliphatic 1,2-diketones.

Entry	Dicarbonyls	Diamines	Product <sup>a</sup>	Time (min)	Yield (%) <sup>b</sup>
3a				3	98 <sup>c</sup>
3b				2	96
3c				3	95
3d				5	97
3e				5	97
3f				5	92
3g				5	94
3h				5	90
3i				5	94
3j				3	95

**Table 3.** (continued).

Entry	Dicarbonyls	Diamines	Product	Time (min)	Yield (%) <sup>b</sup>
<b>3k</b>				5	95
<b>3l</b>				2	94
<b>3m</b>				5	90
<b>3n</b>				2	92
<b>3o</b>				5	89
<b>3p</b>				5	85
<b>3q</b>				5	89

Reaction conditions: PEG-600-H<sub>2</sub>O. <sup>a</sup>All products were characterised by IR, <sup>1</sup>H NMR, <sup>13</sup>C NMR and MS.

<sup>b</sup>Isolated yields after purification. <sup>c</sup>PEG was recovered and reused for three consecutive runs

further addition of water resulted in decrease reaction rate due to decreased solubility of substrates in PEG-H<sub>2</sub>O. In summary, to obtain high reactivity within a short period of time, an optimum volume ratio of PEG to H<sub>2</sub>O was necessary, for example, 1:1 during quinoxaline synthesis.

We have also studied the effect of PEG with different molecular weights on the rate of quinoxaline formation. The rate of conversion increased continuously with the increase in the molecular weight of PEG

from PEG-200 to PEG-600 and decreased when the molecular weight was over 600. This could be attributed to the lower viscosity and better hydrophilic character but the low solubility of the substrates in PEG with low molecular weight. When PEGs with molecular weight over 600 were used, decreased rate of reaction was observed due to higher viscosity of the reaction medium. As a result of the above studied parameters, PEG-600-H<sub>2</sub>O (1:1) system, found to be effective for the synthesis of quinoxalines at room

temperature with excellent yield within a short reaction time (table 2). Furthermore, PEG-600 may be recovered in almost pure form by removal of water under diminished pressure and recycled.<sup>16</sup> There was no decrease in the yield even after three subsequent experiments with recovered and reused PEG-600.

In an optimized reaction condition, 1,2-diketone (1 mmol) and 1,2-diamine (1 mmol) in PEG-600–H<sub>2</sub>O (5 mL) were mixed and stirred at room temperature for 2–5 min. After completion of the reaction (TLC), a simple work-up afforded the product in excellent yield. Present methodology is very simple, efficient, clean and without any side-products. In order to demonstrate the versatility of the PEG-600–H<sub>2</sub>O promoted synthesis of quinoxalines, a series of 1,2-diketones and 1,2-diamines were subjected to condensation (table 3). All these reactions showed rapid formation of quinoxalines at room temperature with high efficiency. However, the variations in the yields were very small and both aromatic and aliphatic 1,2-diketones gave the condensed products in excellent yields with different 1,2-diamines. The products were characterized by <sup>1</sup>H NMR, IR and Mass spectroscopic data. In addition, this methodology is also useful for the synthesis of sterically hindered quinoxaline such as tetrapyrido[3,2-a:2',3'-c:3'',2''-h:2''',3'''-j]phenazine (tpphz) **6** (73% yield) by condensing [1,10]phenanthroline-5,6-dione **4** with [1,10]phenanthroline-5,6-diamine **5** in PEG-600–H<sub>2</sub>O (scheme 2). This new molecule may have biological potential, synthetic and technological importance and they may also provide new prototypes for DNA interaction.

#### 4. Conclusion

In conclusion, we have developed a novel, efficient and eco-friendly method for the synthesis of quinoxalines from various 1,2-diketones and 1,2-diamines using PEG–H<sub>2</sub>O as a new reaction medium. This medium rendered this procedure attractive and environmentally benign. In addition to its efficiency and simplicity, this method provided high yields of biologically potent quinoxalines in short reaction time.

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