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**ONE-POT AND EFFICIENT SYNTHESIS OF  
5-AMINOPYRAZOLE-4-CARBONITRILES CATALYZED BY  
POTASSIUM PHTHALIMIDE**

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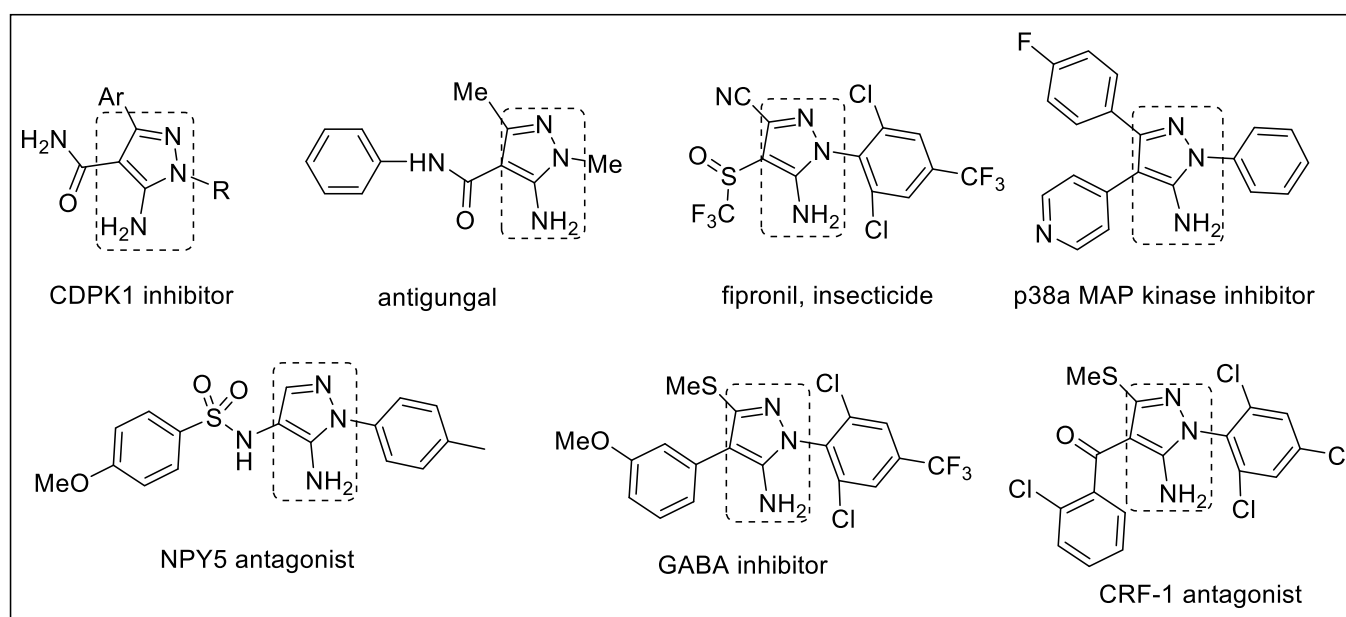
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**Abstract** – The potassium phthalimide (PPI) has been found to be an efficient, recyclable, inexpensive, commercially available, and easy to handle catalyst for the synthesis of 5-aminopyrazole-4-carbonitriles through the one-pot, three-component reaction of easily available starting reactants involving aldehydes, phenylhydrazine, and malononitrile. This reaction was performed in a mixture of EtOH:H<sub>2</sub>O as a green reaction media at 50 °C. The catalyst could be recovered easily from the filtrate via evaporation of solvent and reused many times. The present eco-friendly three-component cyclocondensation offers main benefits including, time-saving, mild conditions, minimize the amount of waste, high atom efficiency, avoiding the hazardous organic solvents or catalysts, and the ease of the work-up.

## INTRODUCTION

Pyrazole and their derivatives are a class of nitrogen containing heterocyclic compounds that have much more attention owing to their a broad spectrum of biological properties such as anticancer, antimicrobial, anti-inflammatory, analgesic, neuroprotective, cytotoxic, antihypertensive, antiviral, anti-obesity, antibacterial, antimycobacterial, antitubercular, antifungal, anticonvulsant, cardiovascular, antiglaucoma, antihyperglycemic, antiparkinson, antioxidant, herbicidal, antipyretic, insecticide, fungicide, and protein kinase inhibitor activities<sup>1</sup> and have been used in the fields of pharmaceuticals and agrochemicals. Polysubstituted pyrazoles have been employed as cosmetic colorings, UV stabilizers, ligands in coordination chemistry, optical brighteners, photoinduced electron transfer systems, and in some transition-metal cross-coupling reactions.<sup>2</sup> They have also been used as an important building blocks and

structural motifs in the synthesis of natural products, biologically active molecules, and some heterocyclic compounds.<sup>3</sup> Furthermore, several reports showed that pyrazole-containing compounds had antimalarial and antileishmanial activity.<sup>4</sup> Aminopyrazoles, on the other hand, are increasingly being exploited as multipurpose building blocks in synthetic organic chemistry for the synthesis of bioactive promising organic molecules and drug candidates.<sup>5</sup> Structures of some reported biologically active highly functionalized pyrazole derivatives have shown in Figure 1.

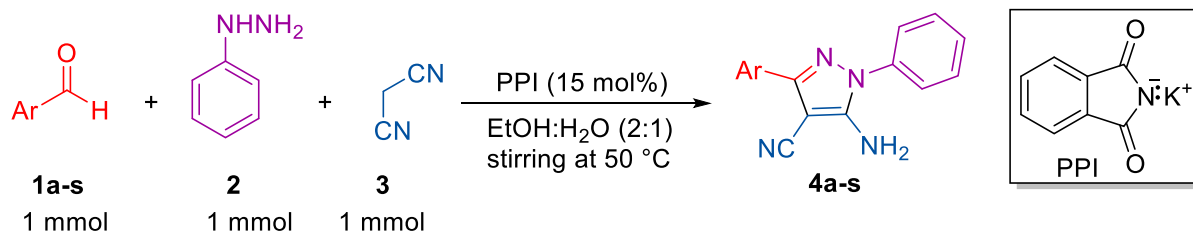


**Figure 1.** Structures of some biologically active polysubstituted pyrazoles containing aminopyrazole units

There are several synthetic methods for the synthesis of polysubstituted pyrazoles under different conditions and diverse catalysts, such as ionic liquids,<sup>6</sup> molecular iodine,<sup>7,8</sup> piperidinium acetate,<sup>9</sup> Cu(OAc)<sub>2</sub>,<sup>10</sup> rhodium,<sup>11</sup> ZnBr<sub>2</sub>,<sup>12</sup> FeCl<sub>3</sub>,<sup>12</sup> graphene oxide-TiO<sub>2</sub> composite (GO-TiO<sub>2</sub>),<sup>13</sup> Oxone,<sup>14</sup> CuO/ZrO<sub>2</sub>,<sup>15</sup> piperidine,<sup>16</sup> eosin Y under atmospheric oxygen,<sup>17</sup> 1-methylimidazolium trinitromethanide {[HMIM]C(NO<sub>2</sub>)<sub>3</sub>},<sup>18</sup> cerium(IV) ammonium nitrate (CAN) in PEG-400,<sup>19</sup> silica chloride,<sup>20</sup> potassium aluminum sulfate (Alum),<sup>21</sup> ytterbium perfluorooctanoate [Yb(PFO)<sub>3</sub>],<sup>22</sup> nano-TiO<sub>2</sub>,<sup>23</sup> palladium and copper,<sup>24</sup> and CuBr-bpy.<sup>25</sup> Furthermore, the use of ultrasound in PEG-400 and water medium is another synthetic technique en route for smooth access to these important heterocyclic core units.<sup>26</sup> The biological and medicinal importance of these heterocycles encourages us to search for an efficient and commercially available catalyst for their synthesis.

Potassium phthalimide (PPI) is readily available and cheap compound with weak basicity (pK<sub>a</sub> = 8.3 in H<sub>2</sub>O) and is often used in the Gabriel synthesis reaction. It has also been found that PPI showed an extremely high capacity for CO<sub>2</sub> capture.<sup>27</sup> We have recently discovered efficient approaches for the

PPI-promoted some organic transformations.<sup>28</sup> To date, there has been no article published in literature that describes the use of PPI for the synthesis of polysubstituted pyrazoles. In this contribution, PPI has been used for the green one-pot, three-component reaction (3CR) of various aldehydes (**1a-s**), phenylhydrazine (**2**), and malononitrile (**3**) to synthesis of highly functionalized 5-aminopyrazole-4-carbonitriles (**4a-s**) (Scheme 1).



Ar: C<sub>6</sub>H<sub>5</sub> (**1a**), 4-Me-C<sub>6</sub>H<sub>4</sub> (**1b**), 4-MeO-C<sub>6</sub>H<sub>4</sub> (**1c**), 4-HO-C<sub>6</sub>H<sub>4</sub> (**1d**), 4-Me<sub>2</sub>N-C<sub>6</sub>H<sub>4</sub> (**1e**), 4-NO<sub>2</sub>-C<sub>6</sub>H<sub>4</sub> (**1f**), 4-Cl-C<sub>6</sub>H<sub>4</sub> (**1g**), 4-Br-C<sub>6</sub>H<sub>4</sub> (**1h**), 4-F-C<sub>6</sub>H<sub>4</sub> (**1i**), 4-HO-3-MeO-C<sub>6</sub>H<sub>3</sub> (**1j**), 3,4-(MeO)<sub>2</sub>-C<sub>6</sub>H<sub>3</sub> (**1k**), 4-HO-3-EtO-C<sub>6</sub>H<sub>3</sub> (**1l**), 3-NO<sub>2</sub>-C<sub>6</sub>H<sub>3</sub> (**1m**), 2-Cl-C<sub>6</sub>H<sub>3</sub> (**1n**), 2-HO-C<sub>6</sub>H<sub>3</sub> (**1o**), 2-MeO-C<sub>6</sub>H<sub>3</sub> (**1p**), 2-NO<sub>2</sub>-C<sub>6</sub>H<sub>3</sub> (**1q**), 2-thienyl (**1r**), 4-pyridyl (**1s**)

**Scheme 1.** 3CR of aldehydes (**1a-s**) with phenylhydrazine (**2**), and malononitrile (**3**) to synthesis highly functionalized 5-aminopyrazole-4-carbonitriles (**4a-s**) catalyzed by PPI

## RESULTS AND DISCUSSION

Initial experiments were performed with benzaldehyde (**1a**), phenylhydrazine (**2**), and malononitrile (**2a**) in order to optimize a variety of reaction parameters, such as temperature, solvent, and catalyst loading (Table 1).

**Table 1.** Optimization of the reaction conditions for the synthesis of 5-amino-1,3-diphenyl-1*H*-pyrazole-4-carbonitrile (**4a**) via 3CR of benzaldehyde (**1a**), phenylhydrazine (**2**), and malononitrile (**3**)

Entry	Catalyst loading/mol%	Solvent	Temp./°C	Time/min.	Isolated yields/%
1	5	EtOH:H <sub>2</sub> O (2:1)	25	90	15
2	5	EtOH:H <sub>2</sub> O (2:1)	30	50	30
3	5	EtOH:H <sub>2</sub> O (2:1)	50	25	45
4	10	EtOH:H <sub>2</sub> O (2:1)	50	10	60
5 <sup>a</sup>	<b>15</b>	<b>EtOH:H<sub>2</sub>O (2:1)</b>	<b>50</b>	<b>5</b>	<b>95</b>
6	15	EtOH:H <sub>2</sub> O (2:1)	25	85	85
7	15	EtOH:H <sub>2</sub> O (2:1)	30	80	86
8	15	EtOH:H <sub>2</sub> O (2:1)	40	70	88

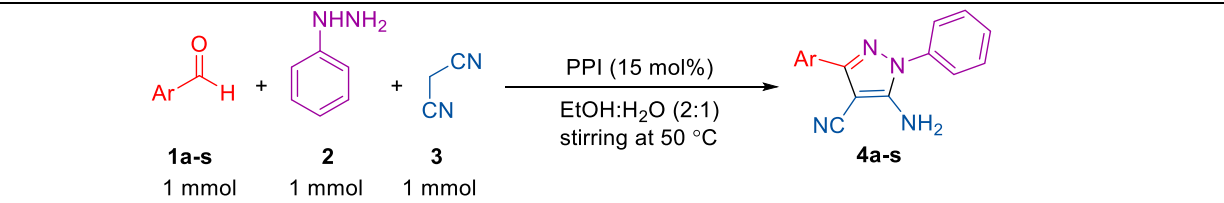
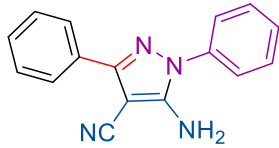
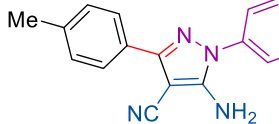
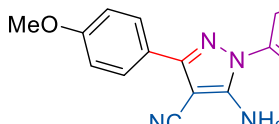
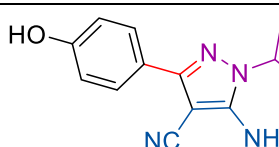
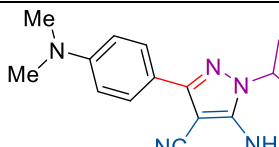
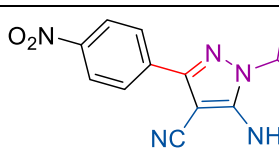
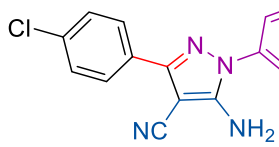
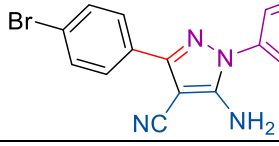
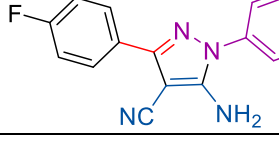
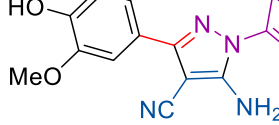
9	15	EtOH:H <sub>2</sub> O (2:1)	60	4	94
10	15	EtOH:H <sub>2</sub> O (2:1)	70	2	90
11	20	EtOH:H <sub>2</sub> O (2:1)	50	5	98
12	15	H <sub>2</sub> O	50	5	74
13	15	EtOH	50	5	91
14	15	DCM	50	5	83
15	15	DMF	50	5	50
16	15	EtOAc	50	5	85
17	15	THF	50	5	75
18	15	-	50	15	10
19	-	EtOH:H <sub>2</sub> O (2:1)	50	240	15

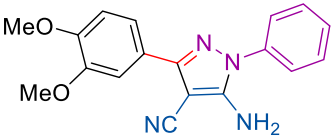
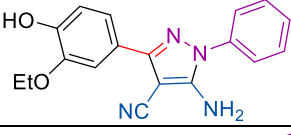
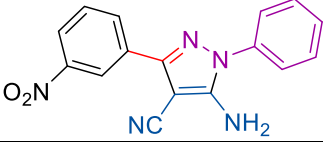
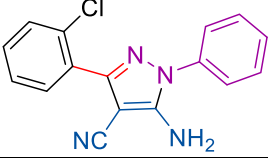
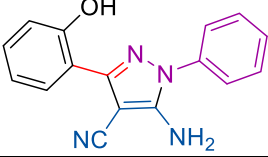
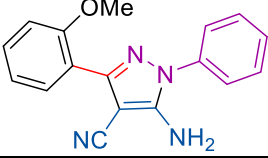
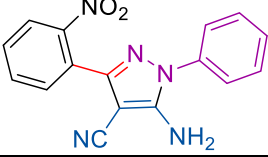
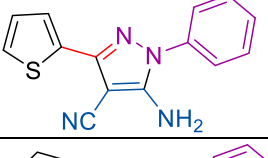
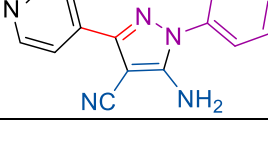
<sup>a</sup> Optimized reaction conditions shown in bold.

Various amounts of PPI were tested in a mixture of EtOH:H<sub>2</sub>O (2:1) medium to promote the synthesis of 5-amino-1,3-diphenyl-1*H*-pyrazole-4-carbonitrile (**4a**) at different temperatures (Table 1, entries 1-11). Among the catalytic quantities loaded and temperatures studied, 15 mol% PPI loading and 50 °C were found to be the most effective (Table 1, entry 5). Reactions were next monitored in H<sub>2</sub>O, EtOH and other different organic solvents such as dichloromethane (DCM), dimethylformamide (DMF), ethyl acetate (EtOAc), and tetrahydrofuran (THF) in order to explore the best solvent (Table 1, entries 12-17). The use of any of these solvents, did not obtain better results than EtOH:H<sub>2</sub>O solvent. When the reaction was conducted in the presence of optimum catalyst loading under solvent-free condition, only 10% of the product was isolated (Table 1, entry 18). It was found that the low yield of the corresponding product (**4a**) was obtained in the absence of catalyst (Table 1, entry 19). The model reaction was also investigated using other bases, such as KOH, and K<sub>2</sub>CO<sub>3</sub> and the results were not satisfactory.

After optimization of conditions for 5-aminopyrazole-4-carbonitriles synthesis and to examine the generality of this method, a variety of aromatic aldehydes bearing electron-withdrawing and electron-donating groups and heterocyclic aldehydes were reacted with phenylhydrazine (**2**) and malononitrile (**3**) in presence of PPI under aqueous ethanol conditions at 50 °C (Table 2). The results showed that the corresponding products (**4a-s**) were synthesized in 85-99% isolated yields in 2-10 min. Moreover, the recyclability of the catalyst was explored for synthesis of **4a**. After completion of the reaction, the PPI catalyst was separated from the filtrate by removing of solvent, washed with small amounts of ethanol and dried in the air. The remained catalyst was reused for four times and the corresponding product was formed in 92%, 88%, 80%, and 70% isolated yields, respectively.

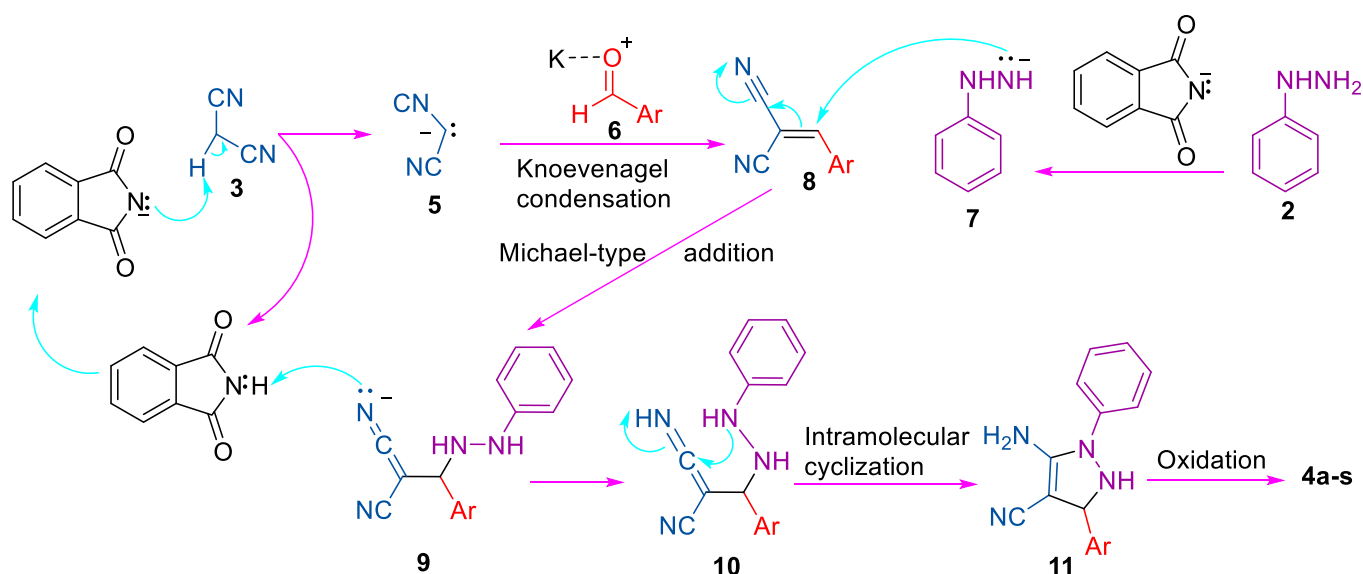
**Table 2.** Potassium phthalimide (PPI) promoted 3C synthesis of highly functionalized pyrazoles

					
Comp. no	Structure of pyrazole derivatives	Time (min)	Isolated yields (%)	Mp (°C)	
				Found	Reported <sup>ref.</sup>
4a		5	95	161-162	159-160 <sup>7</sup>
4b		2	99	117-119	118-120 <sup>7</sup>
4c		2	98	107-109	106-108 <sup>7</sup>
4d		2	94	209-211	210-211 <sup>15</sup>
4e		3	85	104-106	105-107 <sup>7</sup>
4f		4	90	167-168	164-166 <sup>7</sup>
4g		3	98	129-131	128-130 <sup>7</sup>
4h		4	97	164-166	163-165 <sup>15</sup>
4i		4	94	Oil	Oil <sup>7</sup>
4j		4	95	119-120	120-121 <sup>26</sup>

<b>4k</b>		9	90	120-121	120-123 <sup>7</sup>
<b>4l</b>		10	90	220-222	221-223 <sup>18</sup>
<b>4m</b>		4	88	127-129	128-130 <sup>7</sup>
<b>4n</b>		5	98	Semi solid	Semi solid <sup>7</sup>
<b>4o</b>		9	89	161-163	160-161 <sup>26</sup>
<b>4p</b>		8	91	129-131	130-132 <sup>7</sup>
<b>4q</b>		4	96	160-162	159-160 <sup>26</sup>
<b>4r</b>		7	95	137-138	138-140 <sup>26</sup>
<b>4s</b>		10	92	223-225	224-226 <sup>18</sup>

The probable reaction mechanism for this transformation is summarized in Scheme 2. The reaction occurs through initial formation of nitrile intermediate **8** by the Knoevenagel condensation between the activated aldehyde **6** and intermediate anion **5** from malononitrile (**3**) as a C-H acid followed by loss of water molecules. This Knoevenagel reaction of the aldehydes and malononitrile has been reported to occur in the presence of base.<sup>28, 29</sup> Subsequently, Michael-type addition of the hydrazide anion **7** to the Michael acceptor **8**, leads to intermediate **9** and then **10** that cyclizes to heterocyclic intermediate **11** *via* the intramolecular

cyclization. A final dehydrogenation reaction, presumably by the oxygen in the air, leads to the corresponding heterocyclic products (**4a-s**).



**Scheme 2.** Proposed reaction mechanism of the 3C synthesis 5-aminopyrazole-4-carbonitriles (**4a-s**)

In conclusion, this work describes an efficient, mild and PPI-catalyzed one-pot 3CR to rapidly access diverse 5-aminopyrazole-4-carbonitriles using simple aldehydes, phenylhydrazine, and malononitrile as starting materials. All reactions proceeded with high efficiency under simple conditions and gave excellent yields avoiding time-consuming, costly synthesis, and tedious work-up, and purification of products. This approach is easy to implement and was successfully carried out with satisfactory results in a green solvent (EtOH:H<sub>2</sub>O).

## EXPERIMENTAL

All chemicals, unless otherwise specified, were purchased from commercial sources and were used without further purification, with the exception of liquid aldehydes, which were distilled before using. The products were characterized by comparison of their physical data with those of known samples or by their spectral data. Melting points were measured on a Buchi 510 melting point apparatus and are uncorrected. <sup>1</sup>H NMR and <sup>13</sup>C NMR spectra were recorded on a Bruker AVANCE DRX spectrometer. FT-IR spectra were recorded on a Perkin Elmer RXI spectrometer. The development of reactions was monitored by thin layer chromatography (TLC) analysis on Merck pre-coated silica gel 60 F<sub>254</sub> aluminum sheets, visualized by UV light. Elemental microanalyses were performed on Elementar Vario EL III analyzer at Damghan University, Damghan, Iran. All of the targeted products are reported in the literature and are characterized by comparison of their spectral and physical data on the basis of literature descriptions.

**General Procedure for the synthesis of 5-aminopyrazole-4-carbonitrile derivatives (4a-s) catalyzed by PPI.** The appropriate aldehyde (1 mmol), phenylhydrazine (**2**, 1 mmol), malononitrile (**3**, 1 mmol), and PPI as a catalyst (15 mol%) was mixed in a mixture of EtOH:H<sub>2</sub>O (2:1). Then, the reaction mixture was heated at 50 °C for the appropriate time (Table 2). After completion of the reaction as indicated by TLC analysis, the reaction mixture was cooled to room temperature and poured into water. The precipitated products were filtered off, washed with ice water and dried at room temperature. The crude products (except **4i** and **4n**) were purified by recrystallization from EtOH. The filtrate containing the catalyst was used as such for exploring the reusability of the catalyst. The catalyst is soluble in water while the products are insoluble in water. Spectral data for **4a** and **4j** were as follows:

**5-Amino-1,3-diphenyl-1H-pyrazole-4-carbonitrile (4a).** <sup>1</sup>H NMR (CDCl<sub>3</sub>, 500 MHz): 6.82 (t, *J* = 7.3 Hz, 1H), 7.12 (d, *J* = 7.6 Hz, 2H), 7.28-7.36 (m, 3H), 7.42 (t, *J* = 7.7 Hz, 2H), 7.63 (s, 1H), 7.71 (d, *J* = 7.2 Hz, 2H), 7.73 (s, 1H); <sup>13</sup>C NMR (CDCl<sub>3</sub>, 125 MHz): 112.9, 113.3, 120.7, 127.0, 128.9, 129.2, 129.8, 135.7, 137.8, 146.1, 150.4, 156.6; Anal. Calcd for C<sub>16</sub>H<sub>12</sub>N<sub>4</sub>: C, 73.83; H, 4.65; N, 21.52%. Found: C, 73.48; H, 4.86; N, 21.72%.

**5-Amino-3-(4-hydroxy-3-methoxyphenyl)-1-phenyl-1H-pyrazole-4-carbonitrile (4j).** <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>): δ 3.93 (s, 3H), 6.86-6.94 (m, 2H), 7.06 (d, *J* = 7.1 Hz, 1H), 7.13 (d, *J* = 8.2 Hz, 2H), 7.30 (d, *J* = 7.4 Hz, 2H), 7.40 (s, 1H), 7.63 (s, 1H), 9.90 (s, 1H), 10.83 (s, 1H); <sup>13</sup>C NMR (100 MHz, DMSO-*d*<sub>6</sub>): δ 56.2, 113.6, 114.7, 118.8, 119.0, 119.5, 120.4, 131.7, 131.9, 132.9, 133.2, 138.6, 140.4, 150.7, 157.5; Anal. Calcd for C<sub>17</sub>H<sub>14</sub>N<sub>4</sub>O<sub>2</sub>: C, 66.66; H, 4.61; N, 18.29; Found: C, 66.61; H, 4.65; N, 18.26.

## ACKNOWLEDGEMENT

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