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# Microwave assisted alum[KAl $(SO_4)_2.12H_2O$] catalyzed multi-component synthesis of Bis-pyrazole derivatives

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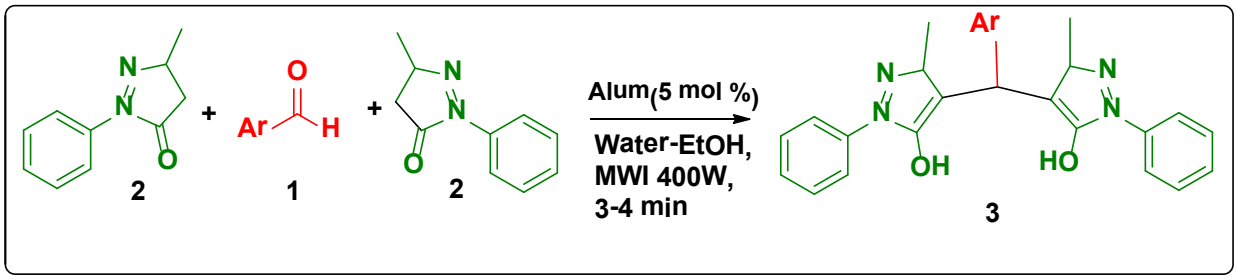
**Abstract:** A green approach multicomponent easy access to some known bis-pyrazole synthesis has been successfully achieved bycondensation between pyrazole-5-one and aromatic aldehydes in the presence of alum[KAl $(SO_4)_2.12H_2O$] as anenvironmentally benign, reusable catalyst in water-ethanol solvent under microwave irradiation method.This efficient and novel protocol has some advantages such as mild, high efficiency, high rate, a very less time of reaction and environmentally benign.

**Keywords:** Alum, Microwave irradiation, Knoevenagel condensation, Bis-pyrazole.

## Introduction

Nitrogen containing five member heterocycles, particularly pyrazole derivatives have become among the most extensively investigated biologically active compounds with various applications and uses in agriculture as well as medicinal chemistry [1-3]. They exhibit considerable antibacterial [4], antifungal [5], antitubercular [6], anti-inflammatory [7], and antitumor activities [8]. In various heterocycles[9-14].Previously reported pyrazole and its derivatives in the presence of several catalytic systems including cellulose sulfuric acid [15], sulfuric acid ([3-(3-silicapropyl)sulfanyl] propyl)ester[16],silica bonded s-sulfonic acid [17], 2-hydroxyethylammonium acetate [18], nano-$Fe_2O_3$ [19], PEG-400 [20], 1,3-disulfonic acid imidazoliumtetrachloroaluminate [21], electrolysis [22], sodium dodecyl sulfate [23], ceric ammonium nitrate(CAN) [24], PEG-$SO_3H$ [25], lithium hydroxide, Mohr's salt [26], and sonication method [27]. Among these, some suffer from shortcoming of these such as low product yield, timeconsuming reaction, use of extra tools and expensive catalysts, environmental pollution etc.

Alum [$KAl(SO_4)_2.12H_2O$], which is used in organic transformations, for example the




**Scheme 1.**Synthesis of Bis(pyrazolyl) methanes using Alum.

Beginelli, coumarins, benzylpyrazolyl coumarin and quinolinone, trisubstituted dimidazole,1*H*-spiro[isoindoline-1,2'-quinazoline]-3,4'(3*H*)-diones, 1,3,4-oxadiazoles, 1,5-benzodiazepines[28].Useofwater or water-ethanol as a reaction medium results in increase the rate and selectivity of many organic reactions[29]. In addition to using of green solvent, microwave irradiation reactions were performed and reduced the time compared to thermal reactions[30-38]. Herein, we report a facile, mild and effective methodology by using environmentally benign catalyst under microwave irradiation method for the synthesis of Bis-pyrazole.

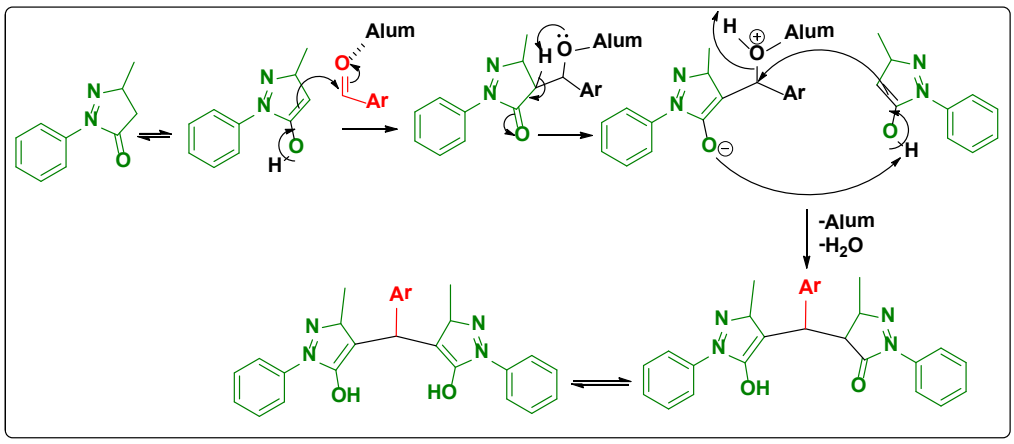
In continuation of our research interest to search an environmentally benign method and protocol [39-44]. Herein, we wish to report a new access for the synthesis of bis-pyrazole **3** by using alum an efficientcatalyst as alum in under microwave irradiation method.(Scheme **1**).

## Results and discussion

In our literature survey there is no any report synthesis of Bis-pyrazole using alum as catalyst under the microwave irradiation method.

Initially, we optimized reaction conditions for the reaction of 3-methyl-1-phenyl-5-pyrazolone with benzaldehyde was chosen as a model (compound **3a**) in various solvent at different time interval, series of solvents DCM, EtOH, MeOH, $CH_3CN$,THF and $CHCl_3$were screenedwith selected alum catalyst (5 mol%) and we found that the use of protic solvents such as EtOH and MeOH forcefully reduces the reaction time with improved yield of product at room temperature, especially with water-ethanol(1:1) (Table **1**).

## Plausible mechanism for the synthesis of bis-pyrazole



The required amount of the catalyst evaluated for this transformation of same compound. Without catalyst, the reaction did not carried out even for 10 min. Further,increase amount catalystby 1-6,8 and 10mol%, the reaction showed an significant progress and completed in about 3 min with excellent yield (98%) There is no considerable improvement on the yield of product and reaction time was observed by further increasing amount catalyst. We also the reaction was established at room temperature in the presence of alum catalyst (5mol%) as catalyst, but the reaction rate decreased and did not complete even after 08 min. In our observation for model reaction an excellent yield was obtained in presence of alum in water-ethanol, thus all examples were tested in 5mol% of alum in 12mL of water-ethanol (1:1)reasonably good to excellent yield.(Table **2**).The product **3a-3m**were purified by simple filtration and recrystallization from hot EtOH. Reactions of aromatic aldehydes bearing electron-withdrawing/donating group with phenyl pyrazolone gave the corresponding products in excellent yields under the same reaction conditions. The yield and structure of the compounds **3** were confirmed by CHN analyses, $^{1}H$, and $^{13}C$ NMR spectroscopy and

**Table 1.Solvent effect in the synthesis of model compound 3a.**

| Entry | Solvent | Time (min) | Yield (%) |
|---|---|---|---|
| 1 | DCM | 6 | 48 |
| 2 | THF | 6 | 55 |
| 3 | MeOH | 4 | 89 |
| 4 | EtOH | 3 | 93 |
| 5 | $H_2O$-Ethanol | **3** | **98** |
| 6 | $CH_3CN$ | 5 | 78 |
| 7 | $CHCl_3$ | 6 | 58 |

Catalyst(alum) selected as 5 mol% for each entry of solvent.

**Table 2.Synthesis of bispyrazoles 3 using Alum (5 mol%) under microwave irradiation method.**

| Entry | Aldehyde | Product | Time (min) | Yield[a] (%) [Lit.] |
|---|---|---|---|---|
| 1 | C6H5CHO | **3a** | 3 | 98[20] |
| 2 | 2,4-(MeO)2C6H3CHO | **3b** | 4 | 92 |
| 3 | 3-(EtO)-4-(HO)C6H3CHO | **3c** | 4 | 94 |
| 4 | 1-NaphthylCHO | **3d** | 4 | 90 |
| 5 | 4-(C6H5)C6H4CHO | **3e** | 3 | 96 |
| 6 | 3-IndolylCHO | **3f** | 4 | 92 |
| 7 | 2,4-Cl2C6H3CHO | **3g** | 3 | 97[15] |
| 8 | 3-O2NC6H4CHO | **3h** | 3 | 97 [20] |
| 9 | 4-O2NC6H4CHO | **3i** | 3 | 97 [20] |
| 10 | 4-MeC6H4CHO | **3j** | 4 | 96 [20] |
| 11 | 2-ClC6H4CHO | **3k** | 3 | 97 [20] |
| 12 | 3-BrC6H4CHO | **3l** | 3 | 96 [20] |
| 13 | 4-MeOC6H4CHO | **3m** | 3 | 95 [26] |

[a] Isolated yields. [b]Reaction condition: aromatic aldehyde **1** (1 mmol), the pyrazolone**2** (2 mmol) and Alum (5 mol %) in $H_2O$-EtOH 12 mL)

compared with those reported methods (Table **3**.) According to our observation, summarized data, the presence of alum catalyst on water-ethanol as solvent for the synthesis of compound **3** under microwave method can be considered as an efficient, environmentally benign method for the preparation of desired product in high yield, short reaction time and environmentally safe conditions using a environmentally benign catalyst in comparison with previously reported method.

**Table 3. Comparison of present work with other reported methods in the literature for synthesis of 3a.**

| Entry | Catalyst and Conditions | Time (min) | Yield[a] (%) [Lit.] |
|---|---|---|---|
| 1 | Alum K[$AlSO_4$. 10($H_2O$)], H2O-EtOH, MWI | **3** | **98**[b] |
| 2 | Mohr's salt (2 mol%), H2O/EtOH, Reflux | 20 | 90 [26[b]] |
| 3 | SBSSA[c] (0.1 g), EtOH, Reflux | 120 | 80 [15] |
| 4 | SASPSPE[d] (0.1 g), EtOH, Reflux | 180 | 90 [16] |
| 5 | Cellulose sulfuric acid (0.2 g), H2O/EtOH, Reflux | 120 | 74 [17] |
| 6 | 2-HEAA[e] (5 mol%), EtOH, r.t. | 15 | 90 [18] |
| 7 | NPS- -Fe2O3[f] (2 mol%), H2O, r.t. | 180 | 93 [19] |
| 8 | PEG-400[g], 110 °C | 120 | 92 [20] |
| 9 | [Dsim]AlCl4[h] (1 mol%), 50 °C | 60 | 86 [21] |
| 10 | Electrolysis, EtOH, NaBr (0.1 g), 20 °C | 33 | 82 [22] |
| 11 | Sodium dodecyl sulfate (5 mol%), H2O, Reflux | 60 | 87 [23] |

[a] Isolated yields. 98[b] Present work.[c]Alum K[AlSO4. 10(H2O)],. [d]H2O-EtOH, MWI

## Experimental

### General

The melting points of the compounds were determined in open head capillary and are uncorrected. Elemental analyses were performed on a PerkinElmer 2400 Series II analyzer. $^1H$ and $^{13}C$ NMR spectra were recorded on a Bruker spectrometer 300 MHz, DMSO- $d_6$ as a solvent. Chemical shifts are reported as $\delta_{ppm}$ units. All the compounds were checked for purity by thin layer chromatography.

### General Procedure to the Synthesis of reported Bispyrazols Using Alum catalyst

A solution of the aromatic aldehyde **1** (1 mmol), the pyrazolone**2** (2 mmol) and Alum (5 mol %) in $H_2O$-EtOH (1:1)12 mL) were subjected to a microwave oven for appropriate time programmed at 400 w. The progress of the reaction was monitored by TLC. After completion, the reaction mixture was cooled at room temperature cold water was added precipitate formed filtered and recrystallized from ethyl alcohol to pure product yield (90-98%).

### Spectral characterization data:

***4,4'-(Phenylmethylene)bis(3-methyl-1-phenyl-1H-pyrazol-5-ol) (3a)***

Light yellow; mp 165-167 °C (lit.[20] 168-170 °C);[1]H NMR (300 MHz,DMSO-$d_6$ ) (ppm): 13.93 (s, 1H, OH), 12.37 (s, 1H, OH), 7.73 (d, *J*= 8.4 Hz, 4H, aromatic CH), 7.43 (t, *J* = 7.2 Hz, 4H, aromatic CH), 7.32-7.26 (m, 6H, aromatic CH), 7.19-7.21 (m, 1H, aromatic CH), 5.03 (s, 1H, CH), 2.31 (s, 6H, 2CH3); [13]C NMR (100.62 MHz, DMSO -*d6*) (ppm): 156.6, 145.4, 140.6, 136.8, 128.9, 128.5, 127.3, 126.5, 126.3, 121.5, 105.8, 33.7, 11.6., Elementalanal.for $C_{27}H_{24}N_4O_2$: M/Z; 436.18

***4,4'-((2,4-Dimethoxyphenyl)methylene)bis(3-methyl-1-phenyl-1H-pyrazol-5-ol) (3b)***

Yellow ; mp 189-190 °C;[1]H NMR (300 MHz, DMSO-$d_6$) (ppm): 14.33 (s, 1H, OH), 12.36 (s, 1H, OH), 7.67 (d, *J*= 7.8 Hz, 4H, aromatic CH), 7.37-7.53 (m, 5H, aromatic CH), 7.19 (t, *J*= 6.9 Hz, 2H, aromatic CH), 6.47 (t, *J*= 9.6 Hz, 2H, aromatic CH), 5.07 (s, 1H, CH), 3.78 (s, 3H, OCH 3), 3.68 (s, 3H, OCH3), 2.23 (s, 6H, 2CH 3); [13]C NMR (76.46 MHz, DMSO -$d_6$) (ppm): 158.7, 156.5, 146.2, 137.5, 137.5, 137.5, 137.2, 136.8, 133.7, 131.7, 128.7, 125.5, 122.8, 120.5, 104.3, 98.3, 55.3, 55.1, 26.8, 11.5 ;Elemental anal. For $C_{29}H_{28}N_4O_4$ M/Z; 496.21

***4,4'-((3-Ethoxy-4 hydroxyphenyl)methylene) bis(3-methyl-1-phenyl-1H-pyrazol-5-ol) (3c)***

Grey, mp 214-215 °C; [1]H NMR (300 MHz, DMSO- $d_6$) (ppm): 13.99 (s, 1H, OH), 12.34 (s, 1H, OH), 8.68 (s, 1H, OH), 7.58 (d, *J*= 8.1 Hz, 4H, aromatic CH), 7.43 (t, *J*= 7.8 Hz, 4H, aromatic CH), 7.20 (t, *J*= 7.2 Hz, 2H, aromatic CH), 6.80 (s, 1H, CH), 6.63 (s, 2H, CH) 4.80 (s, 1H, CH), 3.91 (q, *J* = 6.7 Hz, 2H, CH2), 2.26 (s, 6H, 2CH3), 1.23 (t, *J* = 6.7 Hz, 3H, CH3). [13]C NMR (76.46 MHz, DMSO-$d_6$) (ppm): 146.2, 145.3, 142.5, 137.4, 137.2, 133.3, 131.5, 128.8, 125.6, 120.6, 119.9, 115.3, 113.5, 63.8, 32.8, 14.8, 11.3;Elemental anal. For $C_{29}H_{28}N_4O_4$ M/Z; 496.21

***4,4'-(Naphthalen-1-ylmethylene)bis(3-methyl-1-phenyl-1H-pyrazol-5-ol) (3d)***

White , mp 228-229 °C; [1]H NMR (300 MHz, DMSO-$d_6$ ) (ppm): 13.19 (s, 1H, OH), 12.13 (s, 1H, OH), 8.8-8.05 (m, 1H, aromatic CH), 7.91 (d, *J* = 6.3 Hz, 1H, aromatic CH), 7.78 (d, *J*= 7.2 Hz, 1H, aromatic CH), 7.60-7.31 (m, 5H, aromatic CH), 7.32-7.59 (m, 7H, aromatic CH), 7.11-7.23 (m, 2H, aromatic CH), 5.57 (s, 1H, CH), 2.23 (s, 6H, $2CH_3$). [13]C NMR (76.46 MHz, DMSO- $d_6$) (ppm): 146.2, 144.3, 140.7, 137.5, 136.8, 133.7, 130.9, 128.9, 128.8, 127.2, 125.7, 125.8, 125.6, 125.5, 123.4, 119.9, 105.5, 30.8, 11.6, 11.7 ;Elemental anal. For $C_{31}H_{26}N_4O_2$ M/Z; 486.20

***4,4'-([1,1'-Biphenyl]-4-ylmethylene)bis(3-methyl-1-phenyl-1H-pyrazol-5-ol) (3e)***

White ; mp 217-219 °C; [1]H NMR (300 MHz, DMSO-$d_6$) (ppm): 14.01 (s, 1H, OH), 12.53 (s, 1H, OH), 7.73 (d, *J*= 7.8 Hz, 4H, aromatic CH), 7.52-7.67 (m, 4H, aromatic CH), 7.45-7.39 (m, 9H, aromatic CH), 7.20 (t, *J* = 7.6 Hz, 2H, aromatic CH), 4.98 (s, 1H, CH), 2.33 (s, 6H, $2CH_3$); [13]C NMR (76.46 MHz, DMSO -$d_6$) (ppm): 146.5, 141.3, 140.1, 137.8, 137.5, 137.5, 128.7, 128.9, 127.7, 127.3, 126.7, 125.6, 120.6, 104.7, 104.5, 32., 11.5 ;Elemental anal. For $C_{33}H_{28}N_4O_2$ M/Z; 512.22

***4,4'-((1H-Indol-3-yl)methylene)bis(3-methyl-1-phenyl-1H-pyrazol-5-ol) (3f)***

Yellow ; mp 242-243 °C;[1]H NMR (300 MHz, DMSO-$d_6$ ) (ppm): 12.63 (s, 2H, OH), 9.83 (s, 1H, NH), 8.15-8.13 (m, 2H, aromatic CH), 8.07 (s, 1H, aromatic CH), 8.01- 8.05 (m, 2H, aromatic CH), 7.58-7.63 (m, 1H, aromatic CH), 7.40-7.43 (m, 4H, aromatic CH), 7.32-

7.28 (m, 4H, aromatic CH), 7.15-7.15 (m, 1H, aromatic CH), 7.11-7.31 (m, 2H, aromatic CH), 3.47 (s, 1H, CH), 2.43 (s, 6H, $2CH_3$); $^{13}C$ NMR (100.62 MHz, DMSO- $d_6$) (ppm): 162.5, 150.7, 138.7, 138.3, 136.7, 136.3, 128.5, 128.2, 123.7, 123.3, 122.1, 118.3, 118.1, 112.7, 112.3, 18.6, 12.7;Elemental anal. For $C_{29}H_{25}N_5O_2$ M/Z; 475.20

### *4,4'-((2,4-Dichlorophenyl)methylene)bis(3-methyl-1-phenyl-1H-pyrazol-5-ol) (3g)*

Bisque, mp 228- 229 °C (lit.[15] 227–229 °C); $^1H$ NMR (300 MHz, DMSO-$d_6$) (ppm): 13.98 (s, 1H, OH), 12.65 (s, 1H, OH), 7.60-7.73 (m, 5H, aromatic CH), 7.50 (d, $J$ = 2.6Hz, 1H, aromatic CH), 7.39- 7.53 (m, 5H, aromatic CH), 7.30 (t, $J$ = 7.5 Hz, 2H, aromatic CH), 5.09 (s, 1H, CH), 2.29 (s, 6H, 2CH3 ); $^{13}C$ NMR (62.89 MHz, DMSO -*d6*) (ppm): 148.5, 147.2, 146.1, 137.5, 134.2, 128.7, 125.5, 120.4, 119.3, 111.7, 111.7, 104.7, 104.4, 31.4, 11.5 ;Elemental anal. For $C_{27}H_{22}Cl_2N_4O_2$ M/Z; 504.11

### *4,4'-((3-Nitrophenyl)methylene)bis(3-methyl-1-phenyl-1H-pyrazol-5-ol) (3h)*

Yellow, mp 154-155 °C (lit.[20,16] 151-153 °C);

$^1H$ NMR (300)MHz, DMSO -$d_6$) (ppm): 13.98 (s, 2H, OH), 8.01-8.17 (m, 2H, aromatic CH), 7.56-7.79 (m, 5H, aromatic CH), 7.62 (t, $J$ = 8.2 Hz, 1H, aromatic CH), 7.39 (t, $J$ = 7.8 Hz, 4H, aro-matic CH), 7.23 (t, $J$ = 7.6 Hz, 2H, aromatic CH), 5.18 (s, 1H, CH), 2.32 (s, 6H, 2CH3). $^{13}$ C NMR (100.62 MHz, DM-SO-$d_6$) (ppm): 147.9, 146.5, 144.6, 137.5, 134.5, 129.6, 128.7, 125.3, 125.6, 121.6, 121.3, 120.7, 32.7, 11.4 ;Elemental anal. For $C_{27}H_{33}N_5O_4$ M/Z; 481.17

### *4,4'-((4-Nitrophenyl)methylene)bis(3-methyl-1-phenyl-1H-pyrazol-5-ol) (3i)*

Yellow, mp 229- 231 °C (lit.[20] 229-231 °C); $^1H$ NMR (300 MHz, DMSO-*d6*) (ppm): 13.93 (s, 1H, OH), 12.53 (s, 1H, OH), 8.23 (d, $J$ = 8.8 Hz, 2H, aromatic CH), 7.70 (d, $J$ = 8.2Hz, 4H, aro-matic CH), 7.58 (d, $J$ = 8.6 Hz, 2H, aromatic CH), 7.39 (t, $J$ = 8.2 Hz, 4H, aromatic CH), 7.20 (t, $J$ = 8.2 Hz, 2H, aromatic CH), 5.16 (s, 1H, CH), 2.39 (s, 6H, 2CH3); $^{13}C$ NMR (100.62 MHz, DMSO-$d_6$) (ppm): 150.5, 146.3, 145.6, 137.3, 129.3, 128.8, 128.7, 125.4, 123.5, 120.4, 104.2, 33.2, 11.3 ;Elemental anal. For $C_{27}H_{23}N_5O_4$ M/Z; 481.17

### *4,4'-((4-Methylphenyl)methylene)bis(3-methyl-1-phenyl-1H-pyrazol-5-ol) (3j)*

White, mp 205-206 °C (lit.[20] 201-203 °C); $^1H$ NMR (300 MHz, DMSO-$d_6$) (ppm): 13.90 (s, 1H, OH), 12.36 (s, 1H, OH), 7.03–7.73 (m, 14H, aromatic CH), 4.93 (s, 1H, CH), 2.33 (s, 6H, 2CH3), 2.23 (s, 3H, CH3). $^{13}$ C NMR (100.62 MHz, DMSO-$d_6$) (ppm): 147.2, 140.2, 135.5, 129.7, 129.5, 127.7, 126.3, 121.5, 40.8, 33.6, 21.3, 12.3 ;Elemental anal. For $C_{28}H_{26}N_4O_2$ M/Z; 450.20

### *4,4'-((2-Chlorophenyl)methylene)bis(3-methyl-1-phenyl-1H-pyrazol-5-ol) (3k)*

White, mp 239-241 °C (lit.[20,16] 236-237 °C); $^1H$ NMR (400.13 MHz, DMSO-$d_6$) (ppm): 13.92 (br, 2H, 2OH), 7.70-7.79 (m, 4H, aromatic CH), 7.39-7.47 (m, 8H, aromatic CH), 7.27- 7.37 (m, 2H, aromatic CH), 5.02 (s, 1H, CH), 2.29 (s, 6H, $2CH_3$). $^{13}C$ NMR (100.62 MHz, DMSO -$d_6$) (ppm): 141.3, 140.5, 137.4, 135.7, 130.5, 129.3, 128.7, 128.2, 126.8, 123.7, 120.5, 32.3, 11.5 ;Elemental anal. For $C_{27}H_{23}ClN_4O_2$M/Z; 470.15

### *4,4'-((3-Bromophenyl)methylene)bis(3-methyl-1-phenyl-1H-pyrazol-5-ol) (3l)*

White, mp 175-176 °C (lit.[20] 172-175 °C); $^1H$ NMR (300 MHz, DMSO-*d6)* (ppm): 13.90 (s, 1H, OH), 12.38 (s, 1H, OH), 6.78-7.86 (m, 14H, aromatic CH), 4.86 (s, 1H, CH), 2.35 (s, 6H, 2CH3). $^{13}C$ NMR (125 MHz, DMSO-$d_6$) (ppm): 154.5, 145.7, 140.5, 133.5, 132.5, 129.6, 128.5,

128.2, 127.6, 127.3, 125.3, 123.5, 117.3, 14.2, 10.1 ;Elemental anal. For $C_{27}H_{23}BrN_4O_2$ M/Z; 514.10

### *4,4'-((4-Methoxyphenyl)methylene)bis(3-methyl-1-phenyl-1H-pyrazol-5-ol) (3m)*

White, mp 165-166°C (lit.[26] 162-163 °C); $^1$H NMR (300 MHz, DMSO-$d_6$) (ppm): 13.89 (br, 2H, 2OH), 7.56 (d, $J$ = 8.2 Hz, 4H, aromatic CH), 7.43 (d, $J$ = 7.2 Hz, 4H, aromatic CH), 7.23 (t, $J$ = 7.45 Hz, 2H, aromatic CH), 7.15 (d, $J$ = 8.4 Hz, 2H, aromatic CH), 7.3 (d, $J$ = 8.2 Hz, 2H, aromatic CH), 4.83 (s, 1H, CH), 3.89 (s, 3H, OCH3), 2.49 (s, 6H, 2CH3). $^{13}$C NMR (125 MHz, DMSO-$d_6$) (ppm): 157.3, 146.3, 137.4, 134.5, 129.3, 128.5, 128.2, 125.1, 120.3, 113.7, 105.1, 54.3, 32.4, 11.4 ;Elemental anal. For $C_{28}H_{26}N_4O_3$ M/Z; 466.20

## Conclusion:

In conclusion, the present work describedan efficient facile, novel strategy for the synthesis of reported Bis-pyrazole compound in easily available, reusable catalyst under the microwave irradiation method. The notable advantages forthisprotocol short reaction times, simplicity, excellent yields and environmental impact.

## Conflict of interest:

The authors confirm that this article content has no con-flict of interest.

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