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One-Pot Efficient Pseudo Five Components Synthesis of 4,4'-(Arylmethylene)bis(3-methyl-1-phenyl-1*H*-pyrazol-5-ols) at Room Temperature Assisted by K_2CO_3

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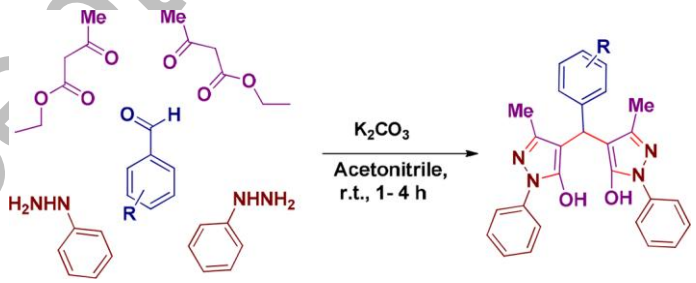
Abstract

A convenient approach for the synthesis of 4, 4'-(arylmethylene)-bis (3-methyl-1-phenyl-1*H*-pyrazol-5-ols) by an efficient one-pot reaction of aromatic/aliphatic aldehydes, phenylhydrazine and ethyl acetoacetate by using sub-stoichiometric amount of K_2CO_3 in acetonitrile solvent at room temperature is described. This protocol of one-pot synthesis produced a library of bis-pyrazolylmethane derivatives, a potential bioactive compound in very good to excellent yields.

Graphical Abstract



KEYWORDS: Phenyl hydrazine; *β*-keto ester; Aromatic aldehyde; Bis-pyrazolylmethane; Multicomponent reaction; K_2CO_3-catalyst

INTRODUCTION

Pyrazoles and their derivatives are an important class of biologically active drug targets in the pharmaceutical industry, because they are found as the core structures of numerous biologically active compounds.[1, 2] For example, they exhibit antipyretic, ant-anxiety, anti-inflammatory and analgesic properties.[3-4] Moreover, 2,4-dihydro-3*H*-pyrazol-3-one derivatives including 4, 4′-(arylmethylene) bis(3-methyl-1-phenyl-1*H*-pyrazol-5-ols) have a broad spectrum of approved biological activity, being used as gastric secretion stimulatory,[5] antipyretic,[6] anti-inflammatory,[7] antidepressant,[8] antibacterial,[9] and anti filarial agents.[10] 4,4′-(arylmethylene) bis(1*H*-pyrazol-5-ol)s are also useful as insecticides,[11] pesticides,[12] fungicides,[13] and dyestuffs.[14] Furthermore, pyrazoles are not only of interest as intermediates in chemical synthesis but also as chelating and extracting reagents for different metal ions.[15] The studies on bis-pyrazolone-based complexes reveal that they have strong fluorescence properties[16-18] and some of them have antitumor activities in vitro and high herbicidal activities. The conventional chemical approach for the synthesis of 4, 4′-(arylmethylene) bis(3-methyl-1-phenyl-1*H*-pyrazol-5-ols) involves successive Knoevenagel synthesis to the corresponding arylidenepyrazolones followed by base-promoted Michael reaction and also one-pot tandem Knoevenagel-Michael reaction of arylaldehydes with two equivalents of 5-methyl-2-phenyl-2,4-dihydro-3*H*-pyrazol-3-one performed under a variety of reaction conditions.[19]

There are quite a good number of catalysts have been used for this transformation. These include acetic acid or piperidine in ethanolic solution,[20] sodium dodecyl sulfate

(SDS),[21] silica-bonded S-sulfonic acid,[22a] 3-aminopropylated silica gel (AP-SiO_2),[22b] silica-sulfuric acid (SSA),[22c] PEG-SO_3H,[23] PEG-400 at 110 °C,[24] lithium hydroxide monohydrate ($LiOH.H_2O$),[25a] SBPPSA catalyst,[25b] ([Sipmim]HSO_4),[25c] CAN,[26] and an electrocatalytic tandem Knoevenagel-Michael reaction,[27] $Ce(SO_4)_2.4H_2O$,[28] ammonium acetate[29] etc. Recently, Hasaninejed *et al.* [30] reported the pseudo five component methods for the synthesis of title compound under ultrasound irradiation. Under solvent free conditions pseudo five component reaction in the presence of 2-hydroxy ethylammonium propionate[31a] as a recyclable catalyst and acidic ionic liquid [Et_3NH][HSO_4] [31b] as reusable catalyst was reported by Zhang *et al.* Very recently, acetic acid functionalized ionic liquid, pyridinium salt (1-carboxymethyl)pyridinium chloride [cmpy]Cl)[32] has been reported for the synthesis of bis-pyrazolylmethane derivatives as homogeneous organic catalyst by Moosavi-Zare and his coworkers.

RESULTS AND DISCUSSION

Carbonates of alkali metals are weak bases and have eco-friendly features such as water soluble, not contaminated with product and in principle can be recovered by evaporation of water. Both cesium carbonate and potassium carbonate (K_2CO_3) are extensively used in synthetic organic chemistry.[33-34] Potassium carbonate base has been employed in many common organic reactions such as Micheal addition,[35] reaction of carbonyl compounds with active methylene compounds[36] and C/N/O-alkylation[37] etc.

We wish to report here an efficient method for the synthesis of 4,4'-(arylmethylene)bis(3-methyl-1-phenyl-1*H*-pyrazol-5-ol)s *via* K_2CO_3 assisted one-pot pseudo-five-component

condensation- reaction. In our initial endeavour, to find an appropriate reaction condition for K_2CO_3-promoted synthesis of 4,4′-(arylmethylene)-bis-(3-methyl-1-phenyl-1*H*-pyrazol-5-ol), a one-pot-pseudo-five-component reaction between *m*-nitrobenzaldehyde (1 mmol), phenylhydrazine (2 mmol), and ethyl acetoacetate (2 mmol) was selected as a model reaction and was examined under different bases, solvents and reaction conditions. The results are summarized in Table 1**.**

From the results of Table 1, it clearly show that potassium carbonate is an effective catalyst for this pseudo five component reaction, and in the absence of K_2CO_3 the condensation reaction gave low yield (<25%) after 24 h at room temperature. Amongst the solvents tested $CHCl_3$, 1,4-dioxane, DCE and THF, acetonitrile was found to be the best in terms of yield of the reaction.

With the optimized condition in our hand we have investigated the applicability of this synthetic methodology for 4, 4′-(arylmethylene) bis(3-methyl-1- phenyl-1*H*-pyrazol-5-ols) derivative. Thus a series of reaction of *β*-ketoester (2 mmol), phenyl hydrazine (2 mmol), and a wide range of structurally diverse aromatic aldehydes (1 mmol) were treated in the presence of K_2CO_3 at room temperature. Almost in all cases the desired product was obtained in good to excellent yields. The results are summarized in Table 2.

It was found that aromatic aldehydes with electron withdrawing (Table 2, entries 1-12) substituent gave the desired product 4,4′-(arylmethylene) bis(3-methyl-1-phenyl-1*H*-pyrazol-5-ol)s in high yield and within short reaction times. However, in case of aromatic

aldehydes with moderately strong electron donating groups gave the desired products with lower yield (Table 2, entries 13-17). Interestingly, the condensation of *β*-ketoester (2 mmol), phenyl hydrazine (2 mmol) with terephthalaldehyde (1 mmol) in the presence of K_2CO_3 under acetonitrile solvent, afforded 4,4′-(arylmethylene)-bis (3-methyl-1-phenyl-1*H*-pyrazol-5-ol) in 78% yield, as the only isolable product. This observation is in contrast with that reported by Khazaei *et al.* But, we did not get di-4,4′-(arylmethylene)-bis(3-methyl-1-phenyl-1*H*-pyrazol-5-ol) which have been discussed by Khazaei, and Niknam group[38,39] (Table 2, entry 19). When the similar protocol was applied for the condensation of *β*-ketoester, phenyl hydrazine with 4-N,N-dimethylaminobenzaldehyde (Table 2, entry 20), did not respond to the reaction probably due to the +R effect of dimethylamino group which decreases the reactivity of the aldehyde group. To show, the generality and scope of this synthetic method further, substituted phenyl hydrazines (Table 2, entries 21-22) and ethyl benzoylacetate (Table 2, entries 23-24) was used to afford the corresponding products in good yield. Aliphatic aldehydes (Table 3, entries 1-4) also reacted with phenyl hydrazine and *β*-ketoester under similar condition to afford the title compounds in good to moderate yield.

EXPERIMENTAL

General Procedure For The Synthesis *Of 4,4-(Arylmethylene)-Bis(3-Methyl-1-Phenyl-1H-Pyrazol-5-Ol) (4).*

A magnetically stirred mixture of *β*-ketoester (2 mmol) and phenyl hydrazine (2 mmol) in acetonitrile (2 ml) was assisted by K_2CO_3 (50 mol %) at room temperature for 10 to 15 min., and then aldehyde (1 mmol) was added to the reaction mixture and continued for

the required time (Table 2). Progress of the reaction was monitored by TLC. After the completion of the reaction, the residue was filtered. The solvent was removed under reduced pressure and the crude residue was purified by column chromatography over silica gel (100-200 mesh), eluting with 25 % to 30 % ethyl acetate in petroleum ether to afford compound **4/6**. The structures of all products were confirmed from physical and spectroscopic data such as melting points, IR, ^{1}H NMR, ^{13}C NMR spectra and mass spectra.

SPECTRAL DATA OF PRODUCTS

4,4'-((2-Fluorophenyl)Methylene)Bis(3-Methyl-1-Phenyl-1*H*-Pyrazol-5-Ol) (4h):

A mixture of ethyl acetoacetate (260 mg, 2.0 mmol), phenyl hydrazine (216 mg 2.0 mmol) and K_2CO_3 (69 mg, 0.5 mmol) in acetonitrile (2 ml) was stirred at room temperature for 15 min., then 2-fluorobenzaldehyde (124 mg, 1.0 mmol) was added to the reaction mixture and stirring was continued for stipulated time. Following the general procedure as mentioned above, **4h** (375 mg, 83 %) was isolated as light brown solid after column chromatography by using 25% ethyl acetate in petroleum ether. Melting point: 170-171°C; IR (KBr): 3446, 3056, 2922, 1595, 1491, 1412, 1293, 1221, 1158, 744, 680 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.30 (s, 6H), 5.14 (s, 1H), 7.09-7.17 (m, 2H), 7.24-7.28 (m, 3H), 7.42-7.47 (m, 4H), 7.63-7.73 (m, 5H), 14.33 (s,1H); ^{13}C NMR (75 MHz, DMSO-d_6): δ 12.0, 27.9, 104.0, 115.5, 115.8, 121.2, 124.5, 124.5, 126.1, 128.6, 128.7, 129.0, 129.4, 129.8, 129.9, 130.0, 137.7, 146.4, 158.1, 158.6, 161.8; HRMS (ES^{+}): calcd for $[C_{27}H_{23}FN_4O_2]$ Na^{+}: 477.1697; found: 477.1701.

CONCLUSION

In conclusion, we have demonstrated that one-pot pseudo five component reaction between phenylhydrazine, *β*-keto esters and aromatic/aliphatic aldehyde to gave a library of 4,4'-(arylmethylene)bis(3-methyl-1-phenyl-1*H*-pyrazol-5-ols) in very good to excellent yields using anhydrous K_2CO_3 is an efficient and an inexpensive, mild, green and basic common laboratory chemicals. We believe that these protocols will be a good addition to the most recent environmentally benign methods for the synthesis of this class of compounds.

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SUPPLEMENTARY DATA

Full experimental detail, ^{1}H and ^{13}C NMR spectra etc, this material can be found *via* the "Supplementary Content" section of this article's webpage.'

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Table 1. Effect of catalysts and temperature on the Synthesis of 4, 4'-((3-nitrophenyl)methylene)bis(3-methyl-1-phenyl-1*H*-pyrazol-5-ol)[a].

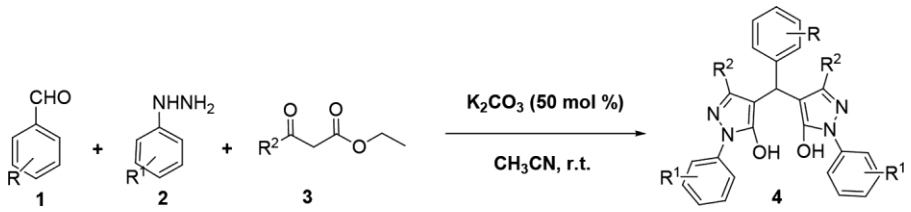
Entry	Catalysts	Mol% of catalyst	T(°C)	Time	Yield(%)[c]
1	K_2CO_3	25.0	rt	4 h	64
2	K_2CO_3	40.0	rt	2 h	72
3	K_2CO_3	50.0	rt	1 h	90
4	K_2CO_3	50.0	70	1 h	57
5	K_2CO_3	55.0	rt	1 h	87
6	K_2CO_3	60.0	rt	1 h	83
7	K_2CO_3	70.0	rt	1 h	75
8	-	-	rt	24 h	<25
9	CS2C03	50.0	rt	1 h	49
10	Ag_2C0_3	50.0	rt	1 h	31
11	Et_3N	50.0	rt	1 h	52
12	DIPA[b]	50.0	rt	1 h	46
13	K^{+t}BuO'	50.0	rt	1 h	35
14	Pyrollidine	50.0	rt	1 h	51
15	N32003	50.0	rt	1 h	65

[a]Reaction condition: 3-nitrobenzaldehyde (1.0 mmol), phenylhydrazine (2.0 mmol), ethylacetoacetate (2.0 mmol), catalyst, in 2 mL acetonitrile.

[b] diisopropylamine,

[c]Pure, isolated yield after column chromatography.

Table 2. One-pot synthesis of 4,4'-(arylmethylene) bis(3-methyl-1-phenyl-1*H*-pyrazol-5-ols) derivatives using K_2CO_3.[a]



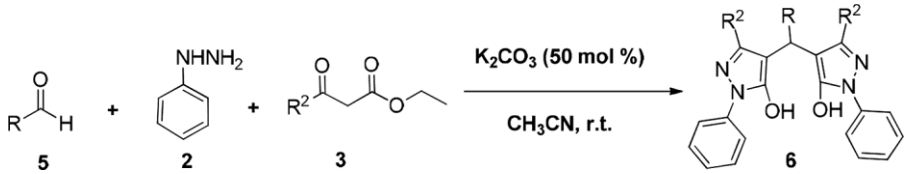
Entr	R	R^1	R^2	Time(h)	Products	Yield(
1	3-Nitro	H	Me	1	4a	90
2	4-Nitro	H	Me	1	4b	93
3	2-Nitro	H	Me	1	4c	89
4	4-Chloro	H	Me	1.5	4d	87
5	4-Bromo	H	Me	1.5	4e	88
6	4-Fluoro	H	Me	1.5	4f	91
7	3-Bromo	H	Me	2	4g	89
8	2-Fluoro	H	Me	1.5	4h	83
9	2-Chloro	H	Me	1.5	4i	82
10	4-Bromo-2-	H	Me	2	4j	86
11	2,6- Dichloro	H	Me	4	4k	71
12	4-CN	H	Me	1	4l	84
13	4-Methyl	H	Me	2	4m	76
14	4-Methoxy	H	Me	2	4n	74
15	2-Methoxy	H	Me	2	4o	68
16	3,4-Dimethoxy	H	Me	2.5	4p	73
17	3,4,5-	H	Me	2	4q	70
18	H	H	Me	1	4r	88
19	4-CHO	H	Me	1.5	4s	78
20[c]	4-N,N-	H	Me	24	4t	-
21	H	4-	Me	2	4u	83
22	3-Nitro	4-	Me	3	4v	77
23	4-Chloro	H	Ph	4	4w	56
24	4-CN	H	Ph	4.5	4x	58

[a]Reaction condition: Aromatic aldehyde (1.0 mmol), phenylhydrazine (2.0 mmol), *β*-keto ester (2.0 mmol), potassium carbonate (50 mol%), in 2 mL solvent.

[b]Pure, isolated yield after column chromatography.

[c]No product was isolated only starting materials.

Table 3. One-pot synthesis of bis-pyrazolylmethane derivatives using aliphatic aldehydes.[a]



Entr	Aldehyde (5)	R^2	Time(h)	Products	Yield(
1	cyclohexanecarbaldehyd	Me	2.5	6a	79
2	octanal	Me	4	6b	85
3	*i*sobutyraldehyde	Me	3	6c	88
4	*i*sobutyraldehyde	Ph	4	6d	69

[a]Reaction condition: Aliphatic aldehyde (1.0 mmol), phenylhydrazine (2.0 mmol), ethylacetoacetate or ethyl benzoylacetate (2.0 mmol), potassium carbonate (50 mol%), in 2 mL acetonitrile.

[b] Pure, isolated yield after column chromatography.

Scheme 1. K_2CO_3-assisted one-pot pseudo-five-component synthesis of 4,4'-(arylmethylene) -bis (3-methyl-1- phenyl-1*H*-pyrazol-5-ol).