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RESEARCH ARTICLE

SYNTHESIS AND EFFECT AMOUNT OF COBALT (II) ACETYLACETONECATALYST ON SYNTHESIS OF BENZIMIDAZOLE DERIVATIVES

***Anil B. Chidrawar**

Research Centre of Chemistry, Degloor College, Degloor. Dist: Nanded - 431717

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ABSTRACT

It Synthesis of 2-phenyl benzimidazole derivatives by oxidative condensation reaction with o-phenylenediamine (1.05 mmol), substituted benzaldehyde (1 mmol), catalyst cobalt (II) acetylacetoneand solvent CH_3OH stirring at room temperature for 4 hours. Solubility experiments showed that the cobalt (II) acetylacetone is miscible with methanol and relatively readily soluble in polar solvents such as ethanol, and they are partially immiscible with no-polar solvents such as ethyl acetate, and tetrahydrofuran. The efficiency of the reaction was mainly affected by the amount of the catalyst. As can be seen from Table 1, the optimal amount of cobalt (II) acetylacetone was 0.05 mmol (entry 2). The isolated yield of 2-phenylbenzimidazole decreases with the increase of cobalt (II) acetylacetone from 0.05 mmol to 0.5 mmol.

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INTRODUCTION

Among these potential heterocyclic drugs, benzimidazole scaffolds are considerably prevalent. Due to their isostructural pharmacophore of naturally occurring active biomolecules, benzimidazole derivatives have significant importance as chemotherapeutic agents in diverse clinical conditions. Synthetically produced heterocycles designed by organic chemists are used as pharmaceuticals, dyestuff, agrochemicals and are of increasing importance in many other areas including adhesives, molecular engineering, polymers etc. In biological processes naturally occurring heterocyclic moieties played a vital role. They are broadly found in naturally in plant alkaloids, nucleic acids, and anthocyanins and flavones as well as in chlorophyll. Additionally, several proteins, hormones, vitamin's contain aromatic heterocyclic ring system (Xiang et al., 2016; Ouattara, 2011 and Shim, 2011). Heterocycles act as drugs because they have specific chemical reactivity and they provide convenient building blocks to which pharmacologically active substituent can be attached. A recent analysis of the nitrogen heterocyclic composition of U.S. FDA (Food and Drug Administration) approved drugs has revealed the relative frequency by which various nitrogen heterocyclic compounds have been incorporated into approved drugs architecture (Chari, 2011). Benzimidazole is an aromatic N-heterocyclic formed by the fusion of benzene and imidazole ring. Nitrogen containing heterocycles, are present in vitamins, proteins and nucleic acids. Benzimidazole is a heterocyclic aromatic organic compound. The most prominent compound available in the nature containing benzimidazole skeleton is Nribosyl dimethyl benzimidazole, which serves as axial ligands for cobalt in vitamin B_{12} (Sun et al., 2011).

****Corresponding author:*** **Anil B. Chidrawar,**
Research Centre of Chemistry, Degloor College, Degloor. Dist: Nanded - 431717

Benzimidazole moieties are very important class of heterocyclic compounds that have many applications in pharmaceutical industry. Benzimidazole derivatives have attracted a significant attention in recent years because of their medicinal applications as antiviral, antifungal (Wright, 1951), antihypertensive (Preston, 1974), and anticancer (Grimmett, 1997), compounds. Apart from therapeutic applications, benzimidazoles formed as intermediates in different organic reactions. In addition to this benzimidazoles are used as fluorescent whitening agent, dyes, organic ligands and functional materials (Chakrabarty, 2006; Gogoi, 2006 and Lee, 2001). Pharmacologically active molecules such as albendazole/ mebendazole/ thiabendazole (antihelmentic), omeprazole (antiulcer), astimizole (antihistaminic) etc. contains the substituted benzimidazoles and display a broad spectrum of potential pharmacological activities. Benzimidazole derivatives also show cytotoxic activity. Substituted benzimidazole derivatives is evaluated by their ability to inhibit gastric H^+/K^+ ATPase and by blocking the gastric acid secretion (Lin, 2005). Some of the benzimidazole derivatives such as Albendazole, Mebendazole were widely used in treatment of parasitic worm infestations. It has been reported that many molecules containing benzimidazole moiety exhibit a wide range of different biological activities as a result of changing the groups on the core structure, as shown in Fig. 2. These biological activities include anti-cancer (1) (Kurakata, 2001), bactericidal (2), (Carcanague, 2002), fungicidal (3) (Lezcano, 2002) and (Aghatabay, 2007), analgesic (4) (Demirayak, 2005) and anti-viral properties (5) (Tewari, 2006). Some have cardiovascular applications (6) (Austel, 1989) while some derivatives have been synthesized and evaluated for inhibition of HIV-1 infectivity.

Experimental Section: All melting points were determined in open capillary tube and were uncorrected. IR spectra were recorded with potassium bromide pellets technique, ^1H NMR spectra were recorded on AVANCE 300 MHz Spectrometer in DMSO using TMS as

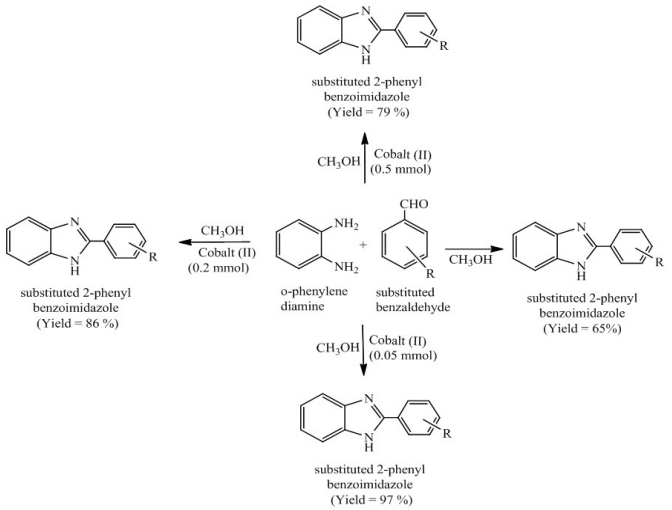
internal standard. Mass spectra were recorded on a FT VG-7070 H Mass Spectrometer using EI technique at 70 eV. All the reactions were monitored by thin layer chromatography.

MATERIAL AND METHODS

In the initial catalytic activity experiments, different solvents were screened for the reaction. Herein the reaction of benzaldehyde and o-phenylenediamine was selected as the model reaction.

Effect of amount of cobalt (II) acetylacetone on synthesis of benzimidazoles:

a) Synthesis of 2-phenyl benzimidazole derivatives by oxidative condensation reaction with o-phenylenediamine (1.05 mmol), substituted benzaldehyde (1 mmol) and solvent CH_3OH stirring at room temperature for 4 hours.
b) Synthesis of 2-phenyl benzimidazole derivatives by oxidative condensation reaction with o-phenylenediamine (1.05 mmol), substituted benzaldehyde (1 mmol), catalyst cobalt (II) acetylacetone (0.05 mmol) and solvent CH_3OH stirring at room temperature for 4 hours.
c) Synthesis of 2-phenyl benzimidazole derivatives by oxidative condensation reaction with o-phenylenediamine (1.05 mmol), substituted benzaldehyde (1 mmol), catalyst cobalt (II) acetylacetone (0.2 mmol) and solvent CH_3OH stirring at room temperature for 4 hours.
d) Synthesis of 2-phenyl benzimidazole derivatives by oxidative condensation reaction with o-phenylenediamine (1.05 mmol), substituted benzaldehyde (1 mmol), catalyst cobalt (II) acetylacetone (0.5 mmol) and solvent CH_3OH stirring at room temperature for 4 hours.



RESULTS AND DISCUSSION

Solubility experiments showed that the cobalt (II) acetylacetone is miscible with methanol and relatively readily soluble in polar solvents such as ethanol, and they are partially immiscible with no-polar solvents such as ethyl acetate, and tetrahydrofuran. In the initial catalytic activity experiments, different solvents were screened for the reaction. Herein the reaction of benzaldehyde and o-phenylenediamine was selected as the model reaction. The efficiency of the reaction was mainly affected by the amount of the catalyst. As can be seen from Table 1, the optimal amount of cobalt (II) acetylacetone was 0.05 mmol (entry 2).

The isolated yield of substituted 2-phenylbenzimidazole decreases with the increase of cobalt (II) acetylacetone from 0.05 mmol to 0.5 mmol. In general, the reaction proceeded smoothly at room temperature to give the corresponding products in reasonable to good yields ranged from 80% to 97%. Compared with the other methods and catalysts, the separation procedure of products and catalyst from the reactor was easier.

Table 1. Effect of amount of cobalt (II) acetylacetone on synthesis of benzimidazoles

Entry	Catalyst (mmol)	Isolated Yield (%)
1	0	65
2	0.05	97
3	0.2	86
4	0.5	79

CONCLUSION

In conclusion we have developed a simple methodology for the preparation of substituted benzimidazoles derivatives. The advantage of this method are extremely mild reaction conditions, short reaction time, high yield, simple experimental technique and compliance with green chemistry protocols.

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