

An efficient solvent-free synthesis of imidazolines and benzimidazoles using $K_4[Fe(CN)_6]$ catalysis

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Abstract: Imidazolines and Benzimidazoles have been efficiently synthesized in high yields by treatment of 1,2-diamine with aldehydes using the metal co-ordinate complex $K_4[Fe(CN)_6]$ as a catalysis. The method was carried out under solvent free condition via oxidation of carbon-nitrogen bond. The process is green, mild and inexpensive.

Keywords: Aldehydes; $K_4[Fe(CN)_6]$; imidazolines; benzimidazoles; solvent free

1. Introduction

The development of simple, efficient and general synthetic method for biological active compounds from readily available catalyst is one of the major challenges in organic synthesis. The importance of imidazolines and benzimidazoles units arises, because they are found in many biologically active compounds.¹⁻² Imidazolines are biologically active pharmacophore and synthetic intermediates in medicinal chemistry.³⁻⁵ They are also used as chiral catalysts,⁶ chiral auxiliaries⁷ and ligands for asymmetric catalysis.⁸⁻⁹ As a continuation of our interest in the synthesis of imidazolines due to its broad spectrum of biological activities including antihyperglycemic,¹⁰⁻¹¹ antiinflammatory,¹²⁻¹³ antihypertensive,¹⁴⁻¹⁵ anticancer¹⁶ and antihypercholesterolemic¹⁷ agents. In addition, the benzimidazol moiety shown excellent biological activity like antiulcers, antihypertensives, antivirals, antifungals, anticancers, antihistaminics, antibacterial, antitubercular, antiasthmatic, anti-diabetic and antiprotozoal.¹⁸⁻²⁶

Recently, several methods have been developed, for the synthesis of benzimidazoles in presence of various catalyst such as sulfur/ultrasonic,²⁷ homogeneous Lewis acids,²⁸ $I_2/KI/K_2CO_3/H_2O$,²⁹ pyridinium-p-toluenesulfonate,³⁰ ionic liquids,³¹ polyaniline-sulfate,³² (bromodimethyl)sulfonium bromide³³ and Zeolite.³⁴ However, all of the synthetic protocols reported so far suffer from disadvantages such as, use of organic solvents,^{28,30,32} harsh reaction conditions,^{29,33} excess temperature,²⁹ prolonged reaction times,^{30,32} use of expensive reagents.^{28,31} To overcome all this disadvantages we report a practical, inexpensive and green method for the synthesis of imidazolines and benzimidazoles by using potassium ferro-cyanide as a catalyst under solvent free condition.

In recent years, potassium ferro-cyanide has gained special attention as a catalyst in organic synthesis like synthesis of anti-Alzheimer drug(-) Galanthamine³⁵ due to its high stability, oxidizing power selectivity and a nontoxic by product Fe(III). It promoted oxidative cyclization of 5-S

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