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CU(II) COMPLEX IMMOBILIZED ON PVA AS AN EFFICIENT CATALYST FOR PREPARATION OF BENZIMIDAZOLE DERIVATIVES

M. Kazemnejadi[1], A. Sardarian[2], H. Eslahi[3], M. Esmaeilpour[4]
Department of Chemistry, College of sciences, Shiraz University, Shiraz 71946 84795 Iran,[1] Milad Kazemnejadi@yahoo.com, [2]sardarian@susc.ac.ir,[3] Hassaneslahi7@gmail.com,[4] m1250m551085@yahoo.com

Abstract: An efficient and rapid one-pot method for the green synthesis of benzimidazole derivatives and catalysed by PVA@Cu(II) Schiff base complex is described. The catalyst demonstrates excellent performance, reusability and stability which can efficiently catalyst benzimidaoles in water medium. A number of benzimidazoles were prepared in up to high yields. The catalyst was recovered by simple filtration and reused for seven runs without any remarkable reactivity loss.

Keywords: Benzimidazole, PVA, Schiff base ligand, Cu catalyst, heterogeneous

1. INTRODUCTION

Benzimidazole derivatives have attracted a great deal of attention in recent years due to of their inevitable applications in medicinal. Benzimidazole derivatives have medicinal properties such as antiviral [1], antiulcer [2], antifungal [3], antihypertensive [4], anticancer [5], and antihistamine [6] compounds. Furthermore, they play a vital role as intermediates in different organic reactions [7]. Two general methods have been reported for the preparation of 2-substituted benzimidazoles include: (1) coupling of phenylenediamines with carboxylic acids [8]; (2) a two-step procedure that consists of the oxidative cyclodehydrogenation of aniline Schiff's bases, which are often generated in situ from the condensation of phenylenediamines with aldehydes [9]. The first method need harsh conditions and the latter method provide a vast utility because of the availability of a different types of aldehydes and oxidative reagents (i.e. nitrobenzene, 1,4-benzoquinone, air, heteropoly acids, MnO_2, $Pb(OAc)_4$, H_2O_2/HCl etc.). However, multistep processes, suffer from some impediments such drastic reaction conditions, tedious work-up procedures and low yields. The additional serious drawbacks relate to the use of homogeneous catalysts that are somewhat modified in the work-up procedure and cannot be recovered. In this point of view, the design of heterogeneous catalysts for a direct, simple and highly selective benzimidazole synthesis will be highly desirable. In the last decades, the use of heterogeneous catalysts has received considerable interest in the synthesis of various benzimidazole derivatives. Various oxide whether supported or unsupported catalysts including aluminosilicates [10], iron oxide [11], cobalt oxide [12], ZnO [13], CuO [14] and MoO_3 [15] have already been used. Recently, an efficient room temperature synthesis of benzimidazole has been developed using zeolite catalysts [16].
In the course of reaction, 2-nitroanilines was hydrogenated to o-phenylenediamine which subsequently underwent cyclization in presence of CO_2 and H_2. Most reported heterogeneously catalysed protocols nevertheless require high catalyst loadings, expensive and/ or sophisticated catalysts to be developed, low catalyst stability or high loadings of metals [11-20]. Therefore, cleaner, high yield and simpler approaches for these precious compounds is very desirable. Herein, we have developed a copper catalytic system for preparation of benzimidazole derivatives through environmentally benign conditions. This stable heterogeneous catalyst could be efficiently catalysed benzimidazole formation which can be simply recovered from the reaction mixture and reused for several times without significant reactivity loss.