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A highly efficient and reusable heterogeneous catalyst for the one-pot synthesis of tetrasubstituted imidazoles

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ABSTRACT

An efficient synthesis of 1,2,4,5-tetrasubstituted imidazoles is achieved by four component cyclocondensation of benzil or benzoin, aniline or benzyl amine, aldehyde and ammonium acetate by using novel polymeric catalyst [poly(AMPS-co-AA)] under solvent-free conditions. The key advantages of this process are high yields, shorter reaction times, easy work-up, purification of products by non-chromatographic method and the reusability of the catalyst.

1. Introduction

Multicomponent reactions (MCRs) have drawn great interest enjoying an outstanding status in modern organic synthesis and medicinal chemistry because they are one-pot processes bringing together three or more components and show high atom economy and high selectivity [1,2]. MCRs have great contribution in convergent synthesis of complex and important organic molecules from simple and readily available starting materials, and have emerged as powerful tools for drug discovery [3,4]. The imidazole nucleus is a fertile source of biologically important molecules. Compounds containing imidazole moiety have many pharmacological properties and play important roles in biochemical processes. They are well known as inhibitors of P38MAP kinase, fungicides, herbicides, anti-inflammatory agents, antithrombotic agents, plant growth regulators and therapeutic agents. In addition, they are used in photography as photosensitive compounds. Some substituted triarylimidazoles are selective antagonists of the glucagons receptor and inhibitors of IL-1 biosynthesis [5]. Radziszewski and Jaap [6,7] proposed the first synthesis of the imidazole core in 1882, starting from 1,2-dicarbonyl compounds, aldehydes and ammonia to obtain 2,4,5-triphenylimidazole. There are several methods for the synthesis of 2,4,5-trisubstituted and 1,2,4,5-tetrasubstituted midazoles using $ZrCl_4$ [8], zeolites HY/silica gel [9], $NaHSO_3$ [10], sulphanilic acid [11], iodine [12], ceric ammonium nitrate [13], oxalic acid [14], ionic liquids [15] and also by microwave irradiation using acetic acid [16]. Each of the above methods for this reaction has its own merits, while some of the methods are plagued by the limitations of poor yield, longer reaction time, difficult work-up and effluent pollution [5]. Therefore, the development of a new mild method to overcome these disadvantages still remains a challenge for organic chemists. One of the aims we have in mind is to introduce a new catalyst for synthesis of 1,2,4,5-tetrasubstituted imidazoles with cost effectiveness and mild condition in high yields. In 1963, Merrifield introduced a modified technique that overcame the problems associated with classical multistep synthesis. This technique has been used in the production of large amount of products. However, recently the chemistry of functional polymers has received great attention and became a practical method for the efficient preparation of novel chemical libraries [17]. Polymeric reagents have recently been developed for use in simple processes such as epoxidation, oxidation, acylation, halogenation and Wittig reactions. In all of these applications, advantage is taken of the selectivity, insolubility and reusable capacity of the

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