4TH National Conference of Iran Chmistry, Chemical Enginereeng And Nano

KF/HNTS CATALYZED ONE-POT THREE-COMPONENT BIGINELLI REACTIONS: AN EFFICIENT SYNTHESIS OF 3,4-DIHYDROPYRIMIDIN-2-(1H)-ONE

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Abstract: Potassium fluoride/Holloysite nonotube (KF/HNTs) acts an active hetrogeneos base catalyst for Biginelli reaction. Here we have describe An efficient one-pot synthesis of 3,4- dihydropyrimidine-2(1H)-one via a three-component reaction of aldehyde, urea, and β-ketoester in the presence of a catalytic amount of (KF/HNTs). The experimental results showed that the KF/HNTs had high catalytic activity and it can be recycled without significant loss of activity. This protocols exhibit advantages such as excellent yield, easy work-up and eco-friendly conditions.

Keywords: Biginelli reaction; KF/HNTs; 3,4-dihydropyrimidin-2(1H)-ones; multicomponent reaction; solid base catalyst.

1. INTRODUCTION

The Biginelli reaction [1,2] involving a multicomponent condensation of aldehyde, urea, and β- ketoester, is the most efficient method for the synthesis of 3,4-dihydropyrmidin-2-(1H)-one. Much effort was directed toward developing highly efficient Biginelli reaction owing to the exhibition of a wide range of biological activities in dihydropyrimidinones such as antitumor [3,4], antibacterial [3-5], such as calcium-channel blocker [4], alpha-la-antagonism [3], antihypertensive agents [6,7] and neuropeptide Y antagonism [8]. Traditional homogeneous systems suffer from problems such as low catalyst recovery, complicated product separation and harsh reaction conditions in most cases. To overcome these drawbacks, several methods have been developed to prepare recoverable heterogeneous catalysts, using various supports, for the Biginelli reaction [9-13]. Recently, Biginelli reaction has been conducted under basic conditions. This involves the use of PPh_3, under solvent free conditions [14], t-BuOK at 70°C [15], chiral primary amines [16], ammonium carbonate in water [17] and nano ZnO as a structure base catalyst [18]. We suggest a new protocol for the synthesis of DHPs by use of supported reagents. The major advantage of supported reagent is the reusability of the catalyst that makes the process inexpensive [19]. Furthermore, it also contributes towards the area of "Green Chemistry". Our interest in organic reactions carried out under heterogeneousconditions prompted us to investigate the utility of solid bases in carbon–carbon bond-forming reactions [20-22]. Among the salts with potentially basic properties potassium fluoride (KF) is more general owing to its cheapness and availability. A lot of supports have been introduced for increasing the basicity of KF such as KF/ZnO, KF/Ca–Mg–Al, KF/CaO–Fe_3O_4, KF/Cp, KF/celite, KF/zeolite, and KF/LDH mainly applied for organic production. Although, a majority of the abovementioned solid bases are effective in organic transformations, they suffer from some drawbacks such as low basicity, use of expensive solid supports and tedious preparation steps [23-29]. Zeolites are particularly interesting in this context because of their high capacity for cation exchange [30]. The main reason for the interest in natural zeolites [31] is the increasing demand for low-cost ion-exchange materials, catalysts, and adsorbent materials in fields such as waste-water treatment [32], animal nutrition, and aquacultural farms [32]. Holloysite clay is an aluminasilicate nanotube formed by rolling flat sheets of kaolinite clay [34,35]. They have a 15 nm lumen, 50-70 nm external diameter, length of 0.5-1 µm. Due to these nonoscale properties, they are used for loading, storage and controlled release of active chemical agent.