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ORIGINAL PAPER

Mesoporous nanocrystalline $MgAl_2O_4$: A new heterogeneous catalyst for the synthesis of 2,4,6-triarylpyridines under solvent-free conditions

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In this paper, one-pot synthesis of 2,4,6-triarylpyridine by condensation of subsisted acetophenone (*II*), aromatic aldehydes (*I*), and ammonium acetate (*III*) in the presence of nanocrystalline $MgAl_2O_4$ as a new heterogeneous catalyst under solvent-free conditions is reported. Advantages of this method are the use of spatially-hindered aldehydes such as 2-methoxy-, 2-fluoro-, and 2-chlorobenzaldehydes, a new nanocatalyst with high surface area, shorter reaction time, easier work-up, higher yield, and its environmental friendliness. The performance of this reaction under solvent free conditions using heterogeneous catalysts like $MgAl_2O_4$ could enhance its efficiency from an economic as well as green chemistry point of view.

Keywords: one-pot synthesis, multicomponent reactions, heterogeneous catalyst, nanocrystalline magnesium aluminate, solvent-free

Introduction

Multi-component reactions (MCR) are an attractive strategy in organic synthesis. In this strategy, complex molecules are formed without the isolation of intermediates after a cascade of bond-forming individual steps using three or more reactants. Attractive properties of MCRs are the reduction of the isolation and purification steps, simple operation, and low costs, energy, time, solvents, and waste production (Barluenga et al., 2008). MCRs are widely used for the synthesis of heterocyclic compounds such as pyridine derivatives. Pyridine derivatives have a unique position in the synthetic chemistry because they are present in natural products, pharmaceuticals such as vasodilators, anticonvulsants, antiepileptics, anaesthetics, and in agrochemicals such as pesticides and herbicides (Kim et al., 2004; Enyedy et al., 2003; Pillai et al., 2003; Klimešová et al., 1999; Constable et al., 2000).

Several methods for the synthesis of 2,4,6-triaryl pyridine are used solid-phase synthesis (Chiu et al., 1998), one-pot synthesis under microwave irradiation (Khosropour et al., 2011), solvent-free synthesis (Adib et al., 2006), and synthesis using heteropoly acids (Heravi et al., 2007), $HClO_4$–SiO_2 (Nagarapu et al., 2007), and ionic liquids (Davoodnia et al., 2010) as the catalyst. However, many of these protocols suffer from drawbacks such as expensive catalyst, undesired side products of the reaction with harsh reagents, long reaction time, cumbersome product isolation procedures, and environmental pollution (Montazeri & Mahjoob, 2012). Therefore, further development of versatile reaction conditions of the 2,4,6-triarylpyridine synthesis using an efficient, inexpensive, reusable, green, and eco-friendly catalyst with high selectivity is still necessary.

On the other hand, environmental concerns requiring the reduction of the amount of pollutants produced, including harmful organic solvents whose recovery is subject to ever more strict laws, have to be considered (Safari et al., 2010). Hence, the chal-

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