Journal of Molecular Liquids 209 (2015) 224–232

Contents lists available at ScienceDirect

Journal of Molecular Liquids

journal homepage: www.elsevier.com/locate/molliq

Preparation, characterization and application of novel ionic liquid as an efficient and reusable catalyst for the solvent-free synthesis of hexahydroquinolines

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ARTICLE INFO

Article history:
Received 4 May 2015
Received in revised form 30 May 2015
Accepted 3 June 2015
Available online xxxx

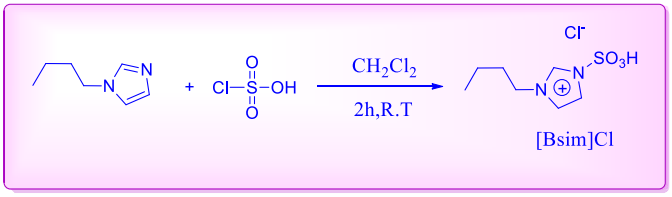
Keywords:
Brønsted acidic ionic liquid
1-Butyl-3-sulfonic acid imidazolium chloride
[Bsim]Cl
Solvent-free
Catalyst

ABSTRACT

A novel Brønsted acidic ionic liquid 1-butyl-3-sulfonic acid imidazolium chloride, [Bsim]Cl, was synthesized. Its structure was investigated using FT-IR, ^{1}H NMR, ^{13}C NMR, mass, UV, TGA and DTA spectra. This ionic liquid, with one acidic functional group, is utilized as a highly efficient and homogeneous catalyst for the promotion of hexahydroquinolines via one-pot multi-component condensation of aromatic aldehydes, dimedone, ethyl acetoacetate, and ammonium acetate under solvent-free conditions. This new method consistently has the advantages of excellent yields and short reaction times. Further, the catalyst could be reused and recovered at least four times without appreciable loss of activity.

1. Introduction

Ionic liquids, IL, (based imidazolium or other organic cations) have received considerable interest as eco-friendly solvents, catalysts and reagents in green synthesis because of their unique properties, such as low volatility, nonflammability, high thermal stability, negligible vapor pressure and ability to dissolve a wide range of materials [1–4]. Recently ionic liquids have been successfully employed as dual reagents (solvents + catalysts) for a variety of the reactions, but their use as catalyst under solvent-free conditions needs to be given more attention [5]. Among them, Brønsted acidic ionic liquids have been designed in order to replace solid acids and traditional mineral liquid acids like sulfuric acid and hydrochloric acid in chemical procedures [6]. A one-pot process is a promising plan of the novel organic synthesis in which a sequence of reactions without performing isolating intermediatesis. Thus proceeding with studies on the synthesis of the compounds by the one-pot multi-component reactions (MCRs) have been of ongoing interest, since MCRs preferably are facile, fast, and efficient with a minimal workup [7–9]. IL-MCR causes the rapid synthesis of highly functionalized heterocyclic molecules with high potential applications in medicinal chemistry [10]. In 1882, Arthur Hantzsch reported first synthesis of symmetrically substituted 1,4-dihydropyridines by the one-pot, four component condensation of two molecules of ethyl acetoacetate, aromatic aldehyde and ammonia [11]. Hantzsch 1,4-dihydropyridines (1,4-DHPs) form a class of heterocyclic compounds which represent interesting pharmacological and biological properties [12]. To realize the importance of polyhydroquinoline derivatives in the synthesis of various drug sources, many characteristic methods were reported. They include conventional heating [13,14], L-proline [15], various catalysts such as ammonium nitrate (CAN) [16], silica perchloric acid ($HClO_4$–SiO_2) [17], trimethylsilyl chloride [18], nickel nanoparticle [19], FeF_3 [20], K7[PW11CoO40] [21], p-TSA [22], solar heat [23], hafnium (IV) [24], SBA-Pr-SO_3H [25], Bakers' yeast [26], and



Scheme 1. The synthesis of 1-butyl-3-sulfonic acid imidazolium chloride, [Bsim]Cl.

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http://dx.doi.org/10.1016/j.molliq.2015.06.011
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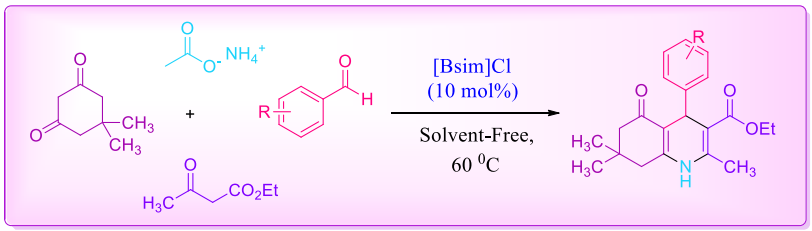

Scheme 2. The one-pot multi-component preparation of ethyl 4-(aryl)-2,7,7-trimethyl-5-oxo-1,4,5,6,7,8-hexahydroquinoline-3-carboxylate derivatives catalyzed by [Bsim]Cl.

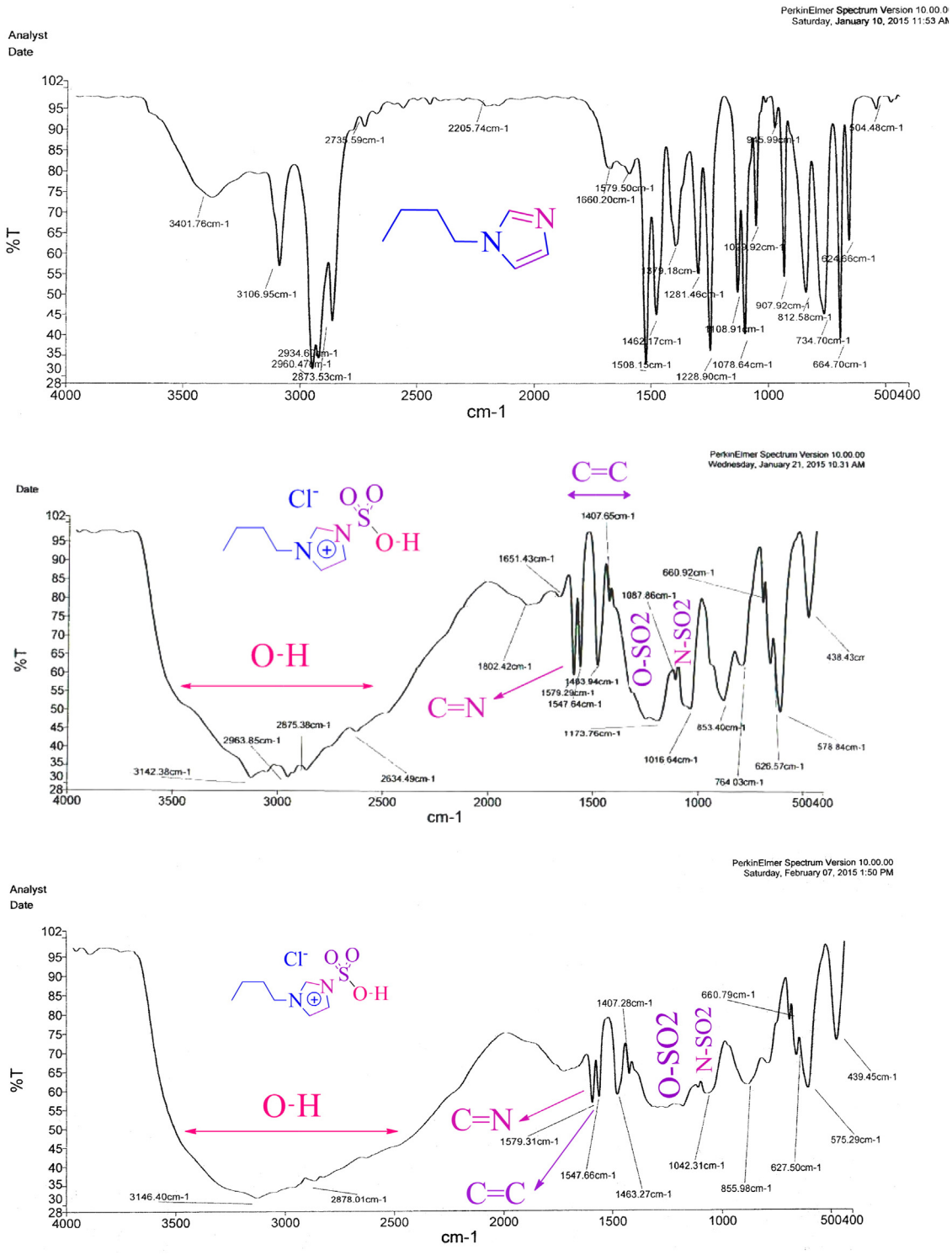

Fig. 1. FT-IR spectra of 1-butyl imidazole (top), 1-butyl-3-sulfonic acid imidazolium chloride, [Bsim]Cl, (middle) and recycle [Bsim]Cl, after four runs.