

## CATALYTIC APPLICATION OF MESOPOROUS SILICAS IN SYNTHESIS OF SUBSTITUTED IMIDAZOLES UNDER MICROWAVE IRRADIATION AND SOLVENT-FREE CONDITIONS

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Mesoporous compounds, founded in 1992, are of great interest because of their remarkable properties, such as their large surface area and pore volume, narrow pore size distribution, and the ease with which their surface can be functionalized. There are many applications for these compounds namely hard template for nanopowder synthesis [1], molecular sieves [2], catalysis [3] and catalyst support [4]. Microwave-assisted rapid organic reactions, on the other hand, constitute an emerging technology, that make organic syntheses more effective and more eco-friendly than conventional reactions. Microwave-assisted synthesis of heterocyclic compounds is also being attended in combinatorial chemistry synthesis of fine chemicals and pharmaceuticals.

This report is focused on application of this technique for environmentally friendly synthesis of substituted imidazoles using SBA-15 mesoporous silica as an efficient catalyst. Some of tri and tetra substituted imidazoles have been known for several years in agrochemicals as herbicide, fungicide [5-6], and also in photography as photosensitive compound [7]. Moreover, compounds with the imidazole ring system have many pharmacological properties and play important roles in biochemical processes. In addition imidazole is the basic skeletal of imidazolium salts, well known as ionic liquids. The use of ionic liquids, composed entirely of ions with a melting point below 100 °C, has become one of the most prolific areas of research, due to their unique properties, including low volatility, high polarity, and good stability over a wide temperature range, as well as selective dissolving capacity with proper selection of cation and anion.

Mesoporous silica was briefly synthesized using a block copolymer as structure directing surfactant in aqueous solution of 2M HCl and Tetraethyl orthosilicate (TEOS) as silica source in hydrothermal condition. The catalytic reactions were done using a domestic microwave oven and a home made Teflon pot as reaction vessel. The reaction progress has been tracked by thin layer chromatography (TLC) and products were extracted by solvent following by crystallization in ethanol. Materials have been characterized using Fourier Transform Infrared (FT-IR), and Nuclear Magnetic Resonance (NMR) spectroscopies and BET surface area measurement methods.

All of the chemicals used were from MERCK, but block copolymers being an industrial co-emulsifier from Clariant and have been used without further purification. General procedure for imidazole synthesis started with 1 mM benzyl and equimolar amount of aldehyde in the presence of 4 times of stoichiometric amount of ammonium acetate as ammonia source. These substances were completely mixed together with 250 mg of the catalyst in a Teflon vessel and the mixture was irradiated by 2 minute microwave energy pulses up to elimination of aldehyde as limiting reagent. The total reaction time was between 6-10 min for different aldehydes.

From our results, we can conclude that SBA-15 mesoporous silica is an efficient catalyst for synthesis of imidazoles. The surface area measured by BET nitrogen absorption method was in the range of 690-730 m<sup>2</sup>/g which is greater than that of silica gel used in our previous work as

catalyst for this reaction. Its pore volume is also larger than HY zeolite, we applied for the same reaction [8]. The melting point and TLC of products confirm the catalytic effect of mesoporous silica. Moreover, NMR and IR spectroscopy results obtained in this work is in good agreement with our previous experiments and further study on application of this catalyst is under investigation in our laboratory.

### References:

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### Figures:

