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RECENT ADVANCES IN KNOEVENAGEL CONDENSATION USING SONOCHEMISTRY: A REVIEW

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ABSTRACT

The use of ultrasound to promote chemical reactions is called sonochemistry. The effects of ultrasound observed during organic reactions are due to cavitation, a physical process that creates, enlarges, and implodes gaseous and vaporous cavities in an irradiated liquid. Recently, the sustainability of chemical reactions has gained strength in scientific and political discussions. In this context, sonochemistry is discussed as being a matchingmethod for promote chemical reactions. Such (often called "green") techniques can help to reduce the total of undesired dangerous chemicals and solvents, decrease energy utilization and boost the selectivity towards the given product(s). In the field of organic synthesis, green chemistry rhymes with relevant choice of starting materials, atom economy, methodologies that minimize the number of chemical steps, suitable use of benign solvents and reagents, efficient strategies for product isolation and refinement and energy minimization. In that context, alternative methods and especially ultrasound can be a fine addition towards achieving these green needs. certainly, sonochemistry is known as being one of the most capable green chemical methods.[1] This review is devoted to the most striking results obtained in green organic sonochemistry.

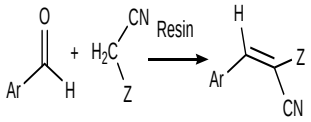
KEYWORDS:- The use of ultrasound sonochemistry.

INTRODUCTION

The ultrasound irradiation has been largely used as synthetic tool for organic synthesis.[2-4] A large number of organic reactions have been carried out in higher yield, shorter reaction time and milder condition under ultrasound irradiation.[5-7] When these methods compared with conventionaland ultrasound has been shown to have desirable effects on both homogeneous and heterogeneous reactions.[8] viable and environmentally-benign.[10] The chemical effect of ultrasonic comes from local hotspots produced by cavitation. Moreover, ultrasound is a mechanical acoustic wave with the frequency.[9-10] The Knoevenagel condensation is all time recognised useful and widely employed methods for the formation of C-C bonds.[11]

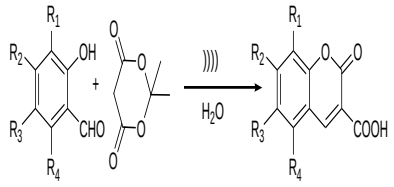
Knoevenagel condensation reaction is the mainlyancient route for the synthesis of α-β-unsaturated carbonyl compounds using aldehydes or ketones with active methylene compounds.[12] B. List introduced Knoevenagel in 1896.[13] it has attract attentionof chemist particularly in the preparation wide range of substituted alkenes, bioactive molecules and natural products, therapeutic drugs also on pharmacological products.[14] Overall, the Knoevenagel condensation is carried out homogenously using nitrogenous molecules. Ultrasonic energy promotes some chemical reactions and is nowadays presented as an alternative tool to prepare fine chemicals under mild conditions.[15-16] The sonochemical effect results from the interaction between a suitable field of acoustic waves and a reacting chemical system, interaction being given by the intermediate phenomena of acoustic cavitation.

The present research work HafedhBelhadj gave the Knoevenagel condensation of aromatic aldehydes with active methylene groups under ultrasound irradiation in the presence of anion-exchange resins.[17] as shown in scheme1.



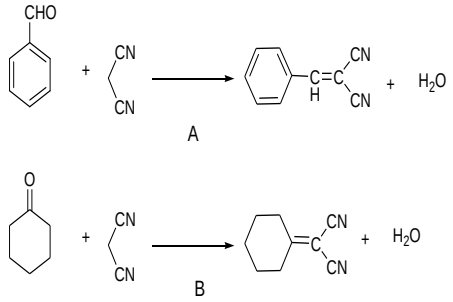
Scheme:1-Knoevenagel condensation of aromatic aldehydes with active methylene

Jian-Long DUhave found a practical and green synthesis procedure for preparing 3- carboxycoumarins by condensation of substituted salicylaldehydes and Meldrum's acid in water under ultrasound.[18] as shown in scheme 2.



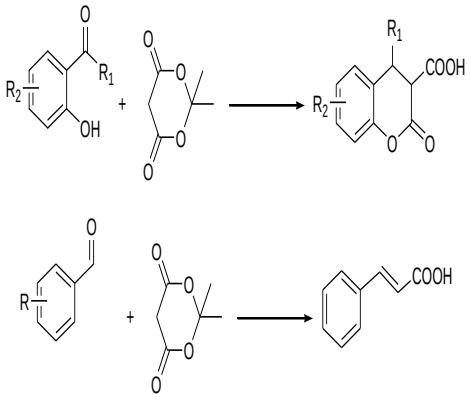
Scheme: 2- Synthesis of 3-Carboxycumarine

RM Martın-Aranda gave result of two basic saponites (Li+-SA and Cs+-SA) whichefficient catalysts for the Knoevenagel condensation of benzaldehyde or cyclohexanone with malononitrile (synthesis of α,β-unsaturated nitriles).[19] as shown in scheme 3.



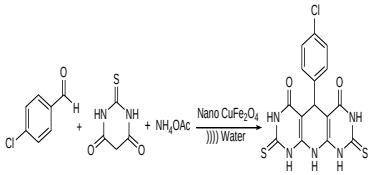
Scheme-3: Knoevenagel condensation of malononitrile with carbonylic compounds: (A) benzaldehyde, (B) cyclohexanone.

Serena Fiorito have demonstrated that non toxic, safe, non polluting, cheap, easy to obtain natural juices and waste waters deriving from agricultural practices and industrial processes are efficient and high yielding promoters for the Knoevenagel condensation of 2-hydroxybenzaldehydes, 2-hydroxyacetophenones, and benzaldehydes with Meldrum's acid leading to 3-carboxycoumarins and cinnamic acids respectively as shown in scheme-4.[20]



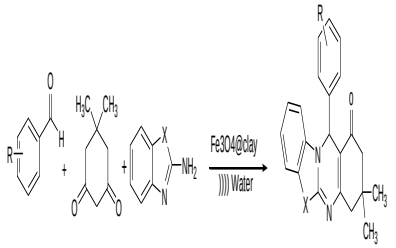
Scheme:4-Knoevenagel condensation routes to coumarin-3-carboxylic (R1 = H, CH_3)and cinnamic acids.

Hossein Naeimi have described the synthesis of pyrido[2,3-d:6,5- d]dipyrimidine derivatives using 2-thiobarbituric acid and ammonium acetate with substituted aromatic aldehydes. This reaction was performed in the presence of catalytic amount of copper ferrite nanoparticles as an efficient, completely magnetically recoverable, easy work-up catalyst in water ultrasonic irradiation make the present procedure eco-friendly and economically acceptableas shown in scheme-5.[21]



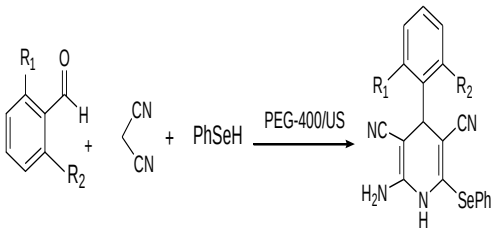
scheme-5: Model reaction for the synthesis of 5-(4-chloro)-2,8-dithioxo-2,3,7,8,9,10-hexahydropyrido[2,3-d:6,5-d]dipyrimidine-4,6(1H,5H)-dione

Ali Malekigave ultrasonic mediated Fe_3O_4@clay core/shell nanocomposite catalyzed green synthesis of imidazo(thiazolo) pyrimidine derivatives has been demonstrated via an efficient and environmentally benign one-pot multicomponent protocolas shown in scheme-6.[22]



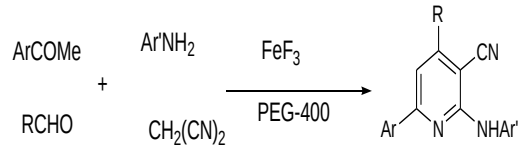
Scheme-6: Ultrasonic-assisted synthesis of imidazo(thiazolo)pyrimidines by using Fe_3O_4@clay nanocatalyst.

Md. Nasim Khan have developed a simple and efficient multicomponent reaction using PEG-400 as a reusable green solvent assisted by ultrasonication for the easy access to a series of selenopyridine derivatives as shown in scheme-7.[23]



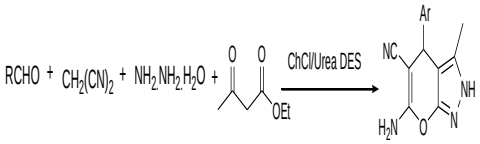
Scheme-7: N-substituted 2-aminopyridines.

Dinne Naresh Kumar Reddy et al have catalyzed FeF_3 four component reaction under ultrasound irradiation was explored for the first time to prepare N-substituted 2-aminopyridines. The methodology involved the use of cheap starting materials and PEG-400 under mild reaction conditions in the presence of air as.[24] shown in scheme-8.



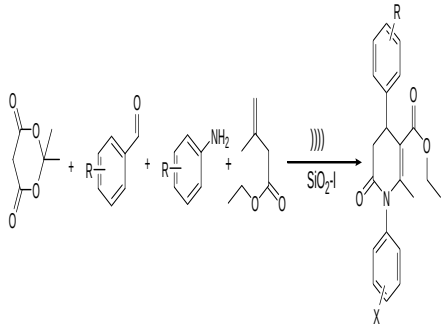
Scheme-8:FeF_3have catalyzed four component reaction under ultrasound irradiation

Adeleh Moshtaghi Zonouz & DavoodMoghani have developed a simple and efficient multi-component protocol for the synthesis of dihydropyrano[2,3-c]pyrazole derivatives using ChCl–Urea deep eutectic solvent as a dual catalyst and environmentally benign reaction mediumas shown in scheme-9.[25]



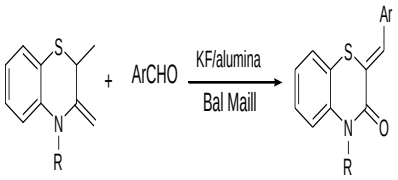
Scheme-9: Synthesis of dihydropyrano[2,3-c]pyrazolescatalysed byChCl–Urea DES medium

Santhosh Govinda raju et al have described a facile, versatile, environmentally benign, rapid and economical protocol for the synthesis of novel 1,4,5,6-tetrahydropyridine-3-carboxylate derivatives under ultrasonic irradiation. The effect of SiO2–I as a catalyst along with the use of benevolent processes like ultrasound is significant for the preparation of 1,4,5,6-tetrahydropyridine-3-carboxylates in excellent yield under the aspect of green chemistry as.[26] shown in scheme-10.



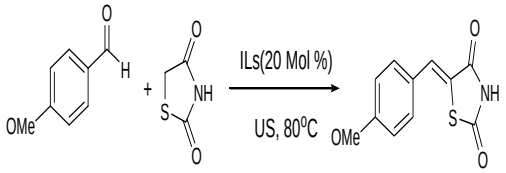
Scheme-10: Synthesis of 1,4,5,6-tetrahydropyridine-3-carboxylate derivatives

Ali Sharifi have developed which under solvent-free conditions leads to high yields of 2-arylidene-benzothiazinones. The attractive features of the procedure are themild reaction conditions, the use of acid-free reagents and to overall very low waste generation.[27] as shown in scheme-11.



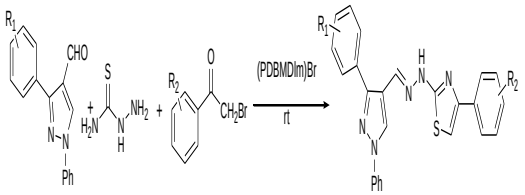
Scheme-11: solvent-free synthesis of 2-arylidene-benzothiazinones

Suresh and Jagir Singh Sandhu gave a clean and efficient method for the synthesis of pharmacologically important thiazolidine derivatives via Knoevenagel condensation of aldehydes with 2,4-thiazolidinedione/rhodanine.[28] as shown in scheme-12.



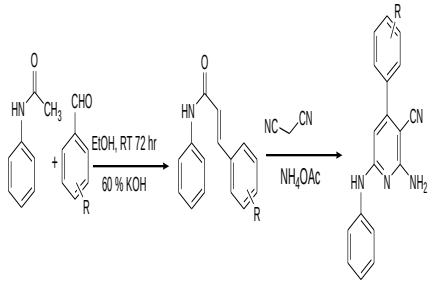
Scheme-12: Synthesis of (Z)-5-(4-methoxybenzylidene)thiazolidine-2,4-dione via Knoevenagel condensation.

Mohammad Nikpass developed a simple, convenient and efficient procedure for the synthesis of 2-hydrazonyl-4-phenylthiazoles using catalyst [PDBMDIm]Br as shown in scheme-13.[29]



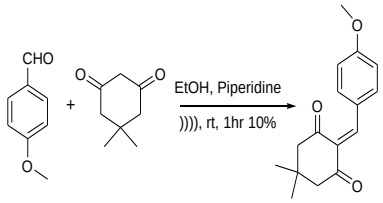
Scheme-13: Synthesis of 2-hydrazonyl-4-phenylthiazoles

M. Purushothaman gave the synthesis of 2-amino-3-cyano-4-phenyl-6-(phenylamino)pyridine and its analogues synthesized by condensation of N-phenylacetamide, substituted benzaldehydes, malononitrile and ammonium acetate via Michael addition followed by Knoevenagel reactionas shown in scheme-14.[30-31]



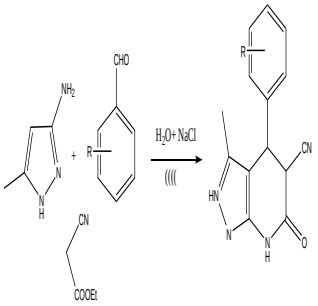
Scheme-14: synthesis of 2-amino-3-cyano-4-phenyl-6-(phenylamino)pyridine

Li-Hsun Chen gave a novel three-component coupling reaction mechanism of substituted 2-aminobenzimidazoles, aromatic aldehydes and 1,3-cyclohexadiones under ultrasonic acceleration to synthesize benzimidazoquinazolinones as shown in scheme-15.[32]



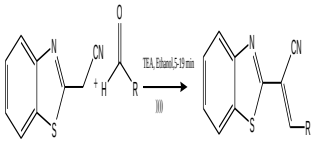
Scheme- 15: Ultrasound-Assisted, One-Pot Synthesis of Functionalized Benzimidazo[2,1‑b]quinazolin-1(1H)‑ones

Anshu Dandia concluded in his ultrasound accelerated study, when three-component reaction of 3-amino-5-methylpyrazole, ethyl cyanoacetate and aromatic aldehydes condensed in presence of Sodium Chloride(NaCl). She achieved chemoselective synthesis of pyrazolo[3,4-b]pyridine advantage of this method for economical and environmental aspects as shown in scheme-16.[33]



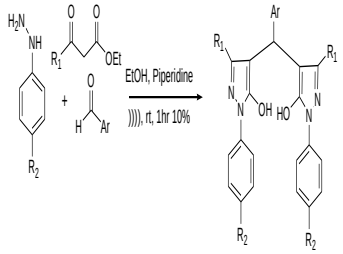
Scheme:16-Chemoselective synthesis of pyrazolo[3,4-b]pyridine derivatives.

Pedro De-la-Torre gave efficient US-PTC method which is most useful for three different methods promoting Knoevenagel condensation reaction for the synthesis of acrylonitrile derivatives. (E)-2-(benzo[d]thiazole-2-yl)-3-heteroaryl- acrylonitriles were obtained in High yield, less reaction times and under eco-friendly conditions as shown in scheme-17.[34]



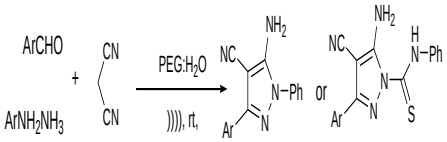
Scheme:17- Synthesis of heteroaryl-acrylonitriles.

Hasaninejad, A gave One-pot pseudo five-component synthesis bis(3-methyl-1-phenyl-1H-pyrazol-5-ols) under ultrasonic irradiation as shown in scheme- 18.[35]



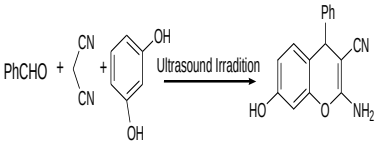
Scheme:18- Synthesis bis(3-methyl-1-phenyl-1H-pyrazol-5-ols) under ultrasonic irradiation

Firouzeh Nemati reported herein, an efficient method for the synthesis of highly functionalized pyrazole derivatives using ultrasound as an energy source in aqueous PEG medium without the use of any catalyst. This system can cause rapid synthesis of highly functionalized pyrazole derivatives to show the versatility of this method as shown in scheme- 19.[36]



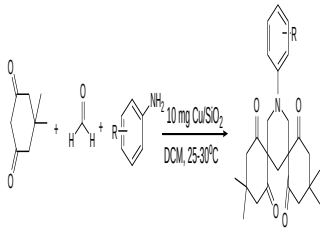
Scheme-19: Synthesis of highly functionalized pyrazole derivatives using ultrasound

The synthesis of 2-amino-4H-chromenes, different reaction conditions have been examined in the reaction of benzaldehyde, resorcinol and malononitrile as a model reaction. J. safari investigated the effect of various solvents such as H_2O, EtOH, DMSO, DMF, CH_3CN, and CH_3Cl on a model reaction under ultrasound irradiation (power intensity: 80%) at 50^oC as shown in scheme-20.[37]



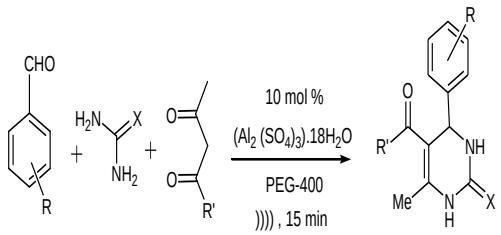
Scheme-20: Synthesis of 2-amino-4H-chromene derivatives

Suman Ray gave the surface acidity of Cu/SiO2 in the synthesis of 3,5-dispirosubstituted piperidines using silica-supported copper as catalyst under ultrasound irradiation as shown in scheme- 21.[38]



Scheme-21: synthesis of 3,5-dispirosubstituted piperidines

Bhata R. Chaudhari et al have developed an efficient aluminium sulphate mediated, green method for the synthesis of DHPMs by using PEG-400 as the solvent medium under ultrasound irradiation.[39-42]



Scheme-22: Synthesis of 3,4-dihydropyrimidin-2(1H)-one/thione by using Ultrasound in PEG.

CONCLUSION

Sonochemistry is green and sustainable organic chemistry and is largely suggested to use in organic synthesis. Ultrasound synthesis is attractive, extraordinary and provides tool in synthetic chemistry with a good outcome from the method is excelled yield, short reaction time, cheap, simple experimental procedure and also environmentally efficient.

ACKNOWLEDGEMENTS

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