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MICROWAVE ASSISTED KNOEVENAGEL CONDENSATION: A REVIEW ARTICLE

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ABSTRACT

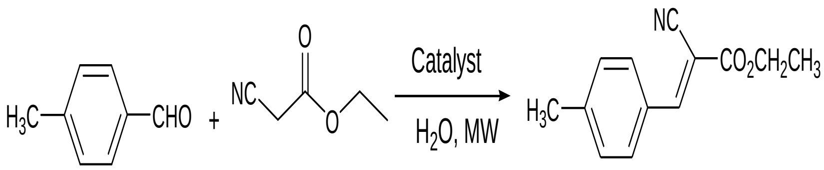
Chemistry has wide scope for Carbon-carbon bond forming reactions and growing from traditional towards modern synthetic methods. The preferred interesting topic in organic chemistry that may benefit from advantages of M.W. irradiation. The Microwave technique was intensively used to carry out many kinds of organic synthesis and has become a useful non-conventional means of performing organic synthesis. In these review I systematic plotted Knoevenagel condensation[1-2] and its applications according to recent years development in present global scenario.

KEYWORDS: The preferred interesting topic in organic chemistry that may benefit from advantages of M.W. irradiation.

INTRODUCTION

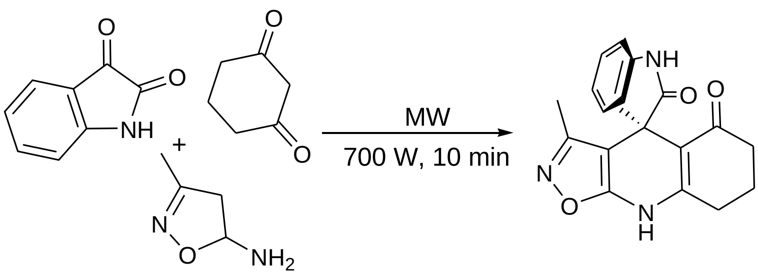
This article is aimed at giving insights into the new trends of MW-assisted reactions, placing the stress on the substantial areas of up-to-date synthetic organic chemistry by presenting a selection of the recent literature. Recently microwave assisted reactions[3-8] have attracted chemists due to their unique advantageous like rate acceleration, short reaction time and improved product yield. Therefore, this technique considered as a promising green approach in Heterocyclic[9-10], drug discovery and development processes. In most of chemists have been focused on developing clean and green protocols chemistry[11-12] for the synthesis of various organic compounds. Water has exclusive nature e.g. ready availability, non toxic nature, and safety in handling.

The Knoevenagel condensation[13] is one of the most important method to synthesis substituted alkenes. It is regarded as a key step in the synthesis of natural product, therapeutic drugs and pharmacological products.[14-15] Environmentally friendly Knoevenagel reaction, using green media, reusable heterogeneous catalysts, ultrasound irradiation and microwave heating technique has recently been paid great attention by organic chemists.[16-17] Amine functionalized mesoporous silica nanoparticles organocatalyst was synthesized by surfactant directed co-concentration, which exhibited comparable efficiencies with the propylamine homogeneous catalyst in water-medium under microwave. The catalytic activities of the NH_2-MSNS catalyst were investigated in water-medium Knoevenagel condensation. Under microwave irradiation, the NH_2-MSNS catalyst showed higher efficiency than the common amine-functionalized mesoporous silicas (NH_2-MCM-41) and silica gel/MW[18] with long channels synthesized by traditional method. Amine-functionalized MSNS has been found to be effective catalyst in the water-medium Knoevenagel condensation under microwave irradiation.[19]



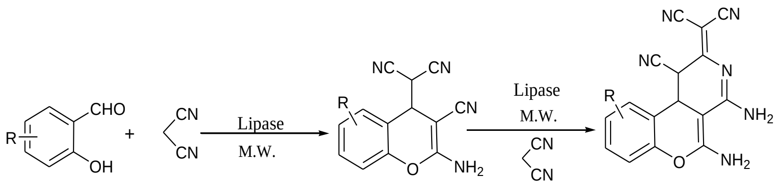
Scheme-1: The Knoevenagel condensation of aromatic aldehydes with active methylene compounds under Microwave Irradiation.

Panneerselvam Yuvaraj was studied the microwave-mediated, chemoselective and environmentally friendly synthesis of a variety of bioactive oxazolo[5,4- b]quinoline- fused spirooxindole derivatives via three-component tandem Knoevenagel/Michalel addtion reaction of 5-amino-3- methylisoxazole, diketones and isatin[20] shown in scheme-2.



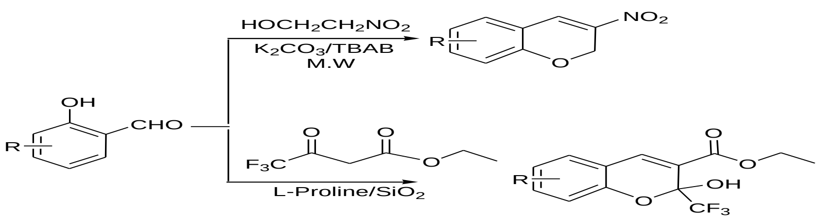
Scheme-2: Synthesis of oxazolo[5,4-b]quinoline – fused spirooxindole

To improve the catalytic performance of enzyme, microwave irradiation was adopted on the enzymatic condensation. Fengjuan Yang worked on the condensation with several substituted salicylaldehyde and malononitrile under microwave irradiation[22] lipas catalyzed.[24], this method is a more convenient and efficient method as shown in scheme-3.



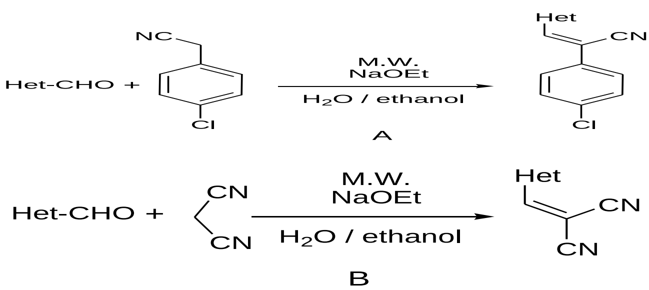
Scheme-3: Lipase-catalyzed condensation of substituted salicylaldehyde and malononitrile under microwave irradiation

Patil gave the Knoevenagel condensation of salicylaldehydes with ethyl trifluoroacetoacetate followed by intramolecular cyclization in the presence of silica-immobilized l-proline catalyst gave the desired 2-hydroxy-2- (trifluoromethyl)-2H-chromenes and other research groups have also developed green reactions to prepare 2-aminochromenes[25] shown in scheme-4.



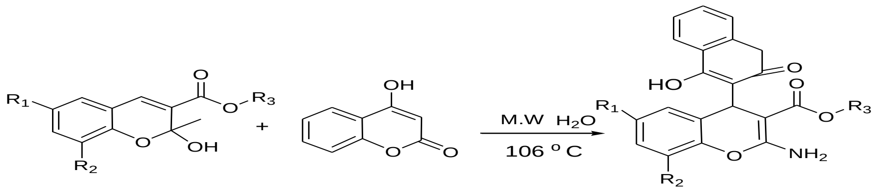
Scheme-4: The Knoevenagel condensation of salicylaldehydes with ethyl trifluoroacetoacetate and l-proline catalyzed 2-hydroxy-2 (trifluoromethyl)-2H-chromenes.

Shetake P. and Dhongade S. was modified and studied new fast and experimentally efficient solvent free synthesis of 3-(heteroaryl)-2-(aryl) prop- 2-enenitriles and 2-(heteroaryl)-2-yl methylenemalononitrile derivatives by condensation of heteroaryl aldehyde with substituted benzyl cyanide/ malononitrile[26] as shown in Scheme-5(A,B)



Scheme-5: Synthesis of 3-(heteroaryl)-2-(aryl) prop- 2-enenitriles(A) and 2-(heteroaryl)-2-yl methylenemalononitrile(B).

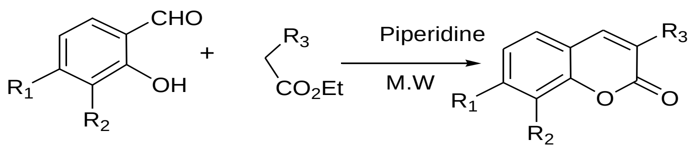
H. M. Meshram developed synthesis of 4-hydroxy coumarin substituted 4H-chromenes without using catalyst under microwave irradiation in aqueous medium. They claimed to synthesized novel coumarin substituted *4H*-chromenes[27] shown in scheme-6.



Scheme-6: Microwave assisted synthesis of new 4H-chromenes

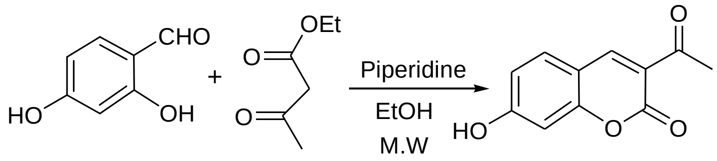
Lucrecia A. studied the Knoevenagel condensation and substituent effect of p-substituted benzaldehydes with malononitrile / ethyl cyanoacetate over potassium, calcium and lanthanum modified MgO was carried out. They carried experiment under microwave activation and the results revealed that the reaction is much faster under microwave activation as compared to conventional thermal activation.[28]

Coumarins are valuable group of organic compounds which was used by food industries and cosmetics utility purposes. Synthesis of Coumarins can be done by Claisen rearrangement, Perkin reaction and Pechmann reaction as well as Knoevenagel condensation.[29] Dariusz Bogdal study microwave irradiation in the Knoevenagel condensation show that the synthesis of a number of coumarins[30-31] He also reported the method having simple, fast. Condensation of salicylaldehyde or its derivatives with various derivatives of ethyl acetate carried out under solvent-free conditions in the presence of piperidine which leads to the synthesis of coumarins[32-33] shown in scheme-7.



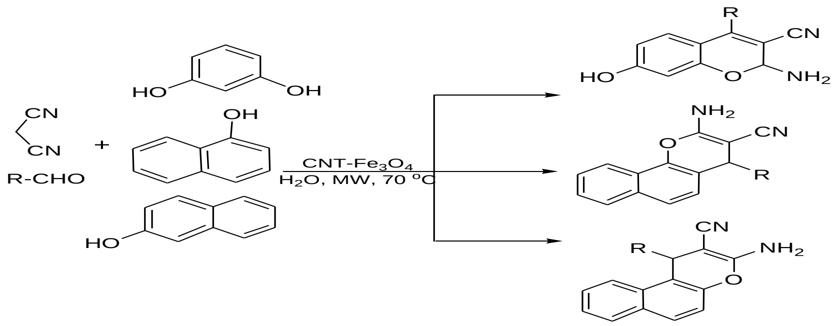
Scheme-7: Synthesis of coumarins by Knoevenagel condensation under microwave irradiation

Thiago Moreira Pereira et al gave synthesis of novel N-acylhydrazone and semicacarbazone-7-hidroxy-coumarins derivatives, starting from 3-acetyl-7-hydroxy-2H-chromen-2-one, under microwave irradiation.[34-35] Resulting molecules have properties of pH dependent luminescence and a strong batochromic shift (65 nm in a less polar medium (methanol) shown in scheme-8.



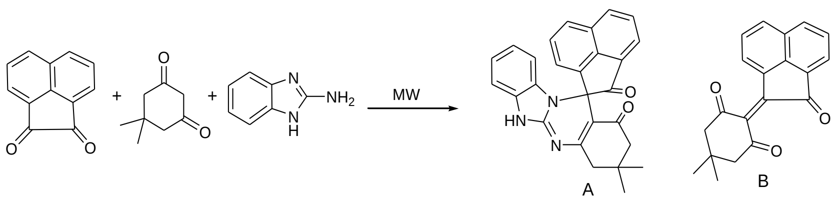
Scheme-8: Synthesis of 3-acetyl-7-hydroxy-2H-chromen-2-one

Zohre Zarnegar and Javad Safari gave synthesis of CNT–Fe_3O_4–IL, magnetite supported on CNTs (CNT–Fe_3O_4) and modified with 1-methyl-3- (3-trimethoxysilylpropyl)-1H-imidazol-3-ium chloride. The resulting nanocomposite was studied for organocatalyst to synthesise 2-amino-chromenes under microwave irradiation MCR's type reactions in aqueous media[36] shown in scheme-9.



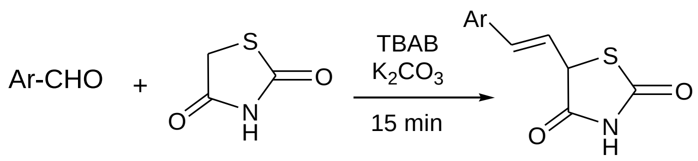
Scheme-9: Microwave-assisted synthesis of 2-amino-chromenes catalyzed by CNT–Fe_3O_4–IL in aqueous medium.

Preeti Maloo et al gave novel multi-component synthesis of spiro-benzimidazoquinazolinones which have been developed under microwave irradiation. It involves a one-pot three-component reaction of acenaphthoquinone or isatin, 1,3-diketone and 2-aminobenzimidazole in ethanol at 180 W and 160°C.[37] shown in scheme-10.



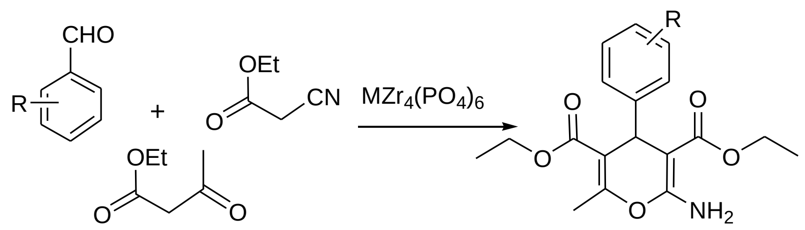
Scheme-10: One pot microwave synthesis of novel spiro-benzimidazoloquinazolinones from acenaphthoquinone or isatin 5, 1,3-diones 2-aminobenzimidazole

T. Durai Ananda Kumara has devloped Phase-transfer catalyzed, energy-efficient and facile synthesis of 5-arylidene-1,3-thiazolidine-2,4-diones. The Knoevenagel condensation of the active methylene group of thiazolidinone (1,3- thiazolidine-2,4-dione/rhodanine) and aryl aldehydes generates 5-arylidene-thiazolidinones.[38] shown in scheme-11.



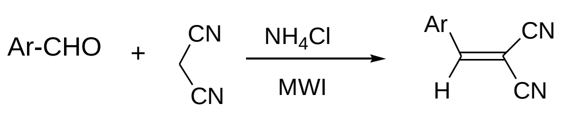
Scheme-11: TBAB catalyzed synthesis of 5-arylidene-1,3-thiazolidine-2,4-diones

Javad Safaei-Ghomi reported synthesis of nanocrystalline $MIIZr_4(PO_4)_6$ ceramics (M: Mn, Ni, Fe, Co). The catalyst utilised for Synthesis of 2-amino-4H-pyran-3,5-dicarboxylate derivatives has property of reusable and robust under microwave irradiation.[39] shown in scheme-12.



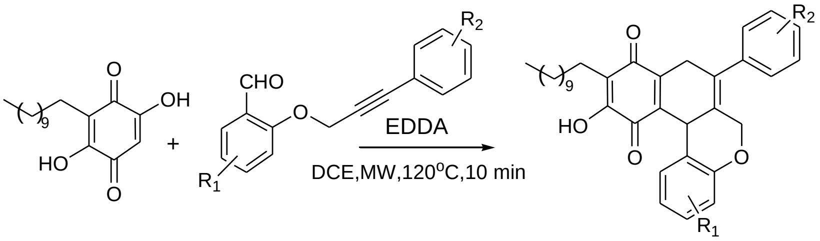
Scheme-12: The Synthesis of 2-amino-3,5-dicarboxylate 4H-pyran derivatives

Bhuiyan et al gave procedure for the synthesis of ylidene malononitrile by microwave irradiation using NH_4OAc carried out under solvent free condition. The main outcome from this method was excellent yields, short reaction time, cheap, simple experimental and as isolation procedures. Finally it is in agreement with the green chemistry protocols[40] shown in scheme-13.



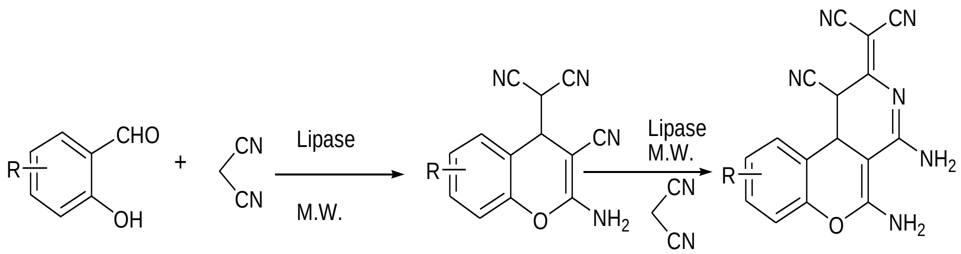
Scheme-13: Knoevenagel condensation reaction of malononitrile with corresponding aromatic aldehydes in presence of ammonium acetate (NH_4OAc) under MWI.

Pedro Martín-Acosta gave Knoevenagel condensation /intramolecular hetero Diels- Alder reaction using O-(arylpropynyloxy)-salicylaldehydes is a type of polycyclic embelin derivatives formed through a domino in the presence of ethylenediamine diacetate (EDDA)[41] shown in scheme-14.



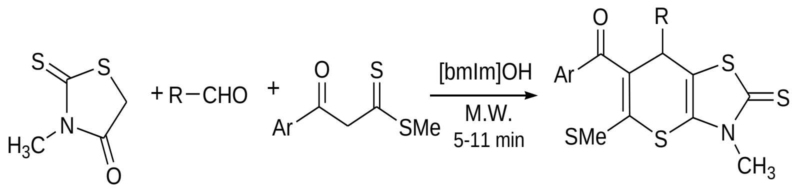
Scheme-14: Synthesis of novel tetracyclic embelin adducts

Enzymes have attracted chemists when they realised their potential use as catalyst in many chemical transformation.[42] This was totally different to the physiological similarities which called as catalytic promiscuity. Fengjuan Yang have done research on enzyme and reported a facile and efficient method for the condensation of substituted salicylaldehyde and malononitrile catalyzed by lipase under microwave irradiation[43] shown in scheme-15.



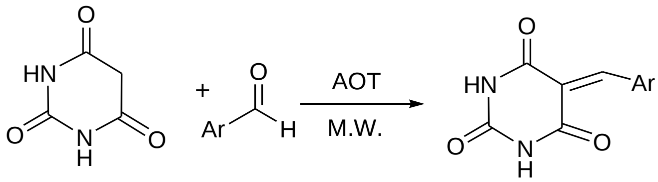
Scheme-15: Lipase-catalyzed condensation of substituted

The basic ionic liquid [bmIm]OH catalysed synthesis of thiopyrano[2,3-d]thiazole-2-thiones has been developed by I. R. Siddiqui et al. This reaction carried out under microwave irradiation and products are formed with high yields in less reaction time (5–11 min). The ionic liquid possesses a dual solvent-catalyst role[44] shown in scheme-16.



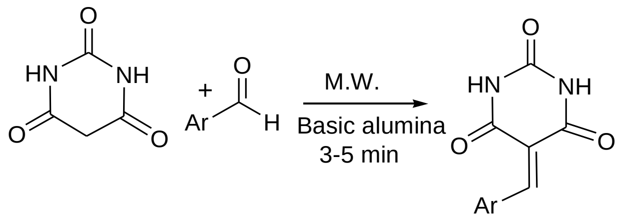
Scheme-16: Solvent screening for the synthesis of compound under microwave irradiation

According to biological point of view heterocyclic compounds gain attention of chemists and encourage them for the Synthesis of heteroaromatic compounds. Mithu Saha gave AOT mediated microwave-irradiation method which is efficiently using for synthesis Knoevenagel product. The reaction of dimedone or 4-hydroxy coumarin with aryl aldehydes, First time synthesized Knoevenagel products underwent Michael addition to give corresponding products. Various aryl aldehydes with barbituric acid[45] shown in scheme-17.



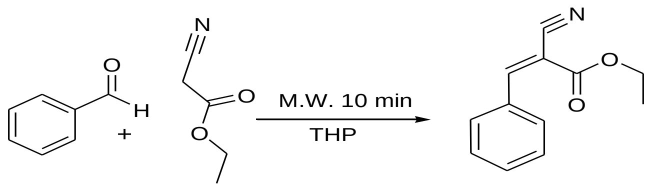
Scheme-17: Synthesis of Knoevenagel condensation Product under Microwave Irradiation Conditions in Presence of AOT

M.H. Moemeni 2-(Pyridin-4-ylmethylene) malononitrile[46] and Khalafi-Nezhad studied the Knoevenagel condensation reaction between barbituric acid and various aromatic aldehydes in presence of basic alumina without using organic solvents under microwave irradiation.[47-48] shown in scheme-18.



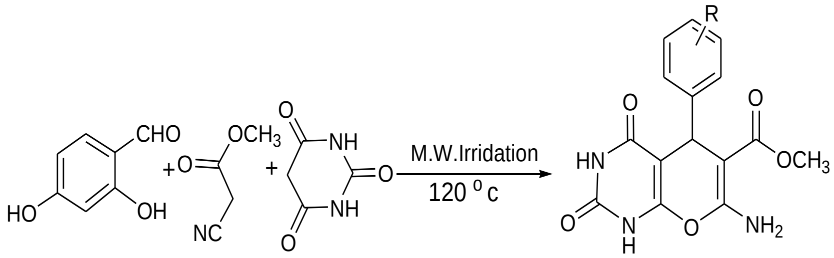
Scheme-18: 5-Synthesis of Substituted benzylidenepyrimidine-2,4,6(1H,3H,5H)-trione

Mohd Bismillah Ansari was developed microwave-mediated Knoevenagel condensation using metal free heterogeneous catalyst which can be operates under water as a green solvent media. The TMP catalyst also shows a wide scope for substrate study[49] shown in scheme-19.



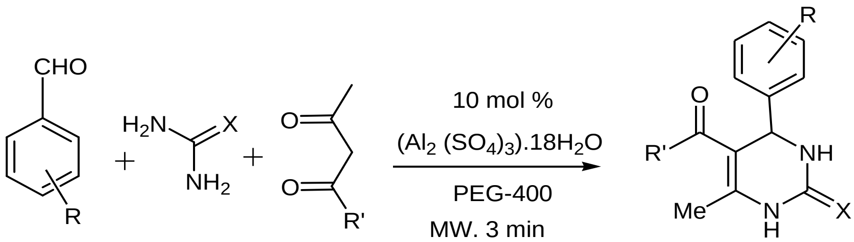
Scheme-19: TMP-catalyzed Knoevenagel condensation of benzaldehyde with ethylcyanoacetate in different solvents

Ajmal R. Bhat gave Microwave-assisted methodology catalyst which is catalyst free, simple and green pathway useful for synthesis of methyl 7 amino-4- oxo-5-phenyl-2-thioxo-2, 3, 4, 5-tetrahydro-1H-Pyrano [2, 3-d] pyrimidine-6-carboxylate derivatives.[50] shown in scheme-20.



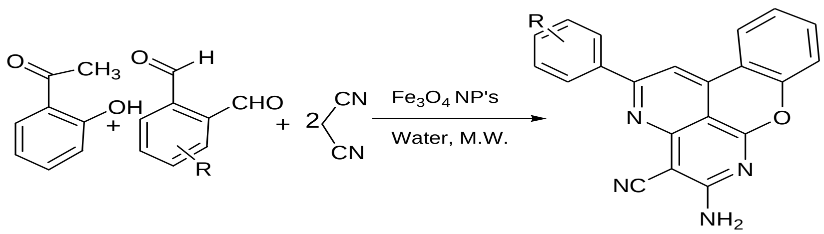
Scheme-20: Microwave synthesis of methyl 7-amino-4-oxo-5-phenyl-2-thioxo-2,3,4,5-tetrahydro-1H-pyrano[2,3-d]pyrimidine-6-carboxylate derivatives

B. R. Chaudhari and its coworkers studied Microwave promoted route for synthesis of 3,4-dihydropyrimidin-2(1H)-one/thione (DHPMs) in presence of Aluminium sulphate as a catalyst for aromatic aldehydes, 1,3-dicarbonyl compounds & (thio)urea in PEG as a reaction solvent[51] shown in scheme-21.



Scheme-21: Synthesis of DHPMs by using PEG-400 under Microwave.

The present work illustrates the advantages of microwave-assisted nano-catalysis in aqueous medium. Fe_3O_4 NPs have been synthesized, characterized and probed as a catalyst for the synthesis of chromeno[1,6]naphthyridines in water. Under microwave catalytic activity of Fe_3O_4 NPs.[52] shown in scheme-22.



Scheme-22: Synthesis of chromeno[1,6]naphthyridines.

CONCLUSION

Microwave now bring a convenient way to achieve target of green and sustainable development in organic chemistry and is strongly recommended to use in organic synthesis. This chemistry is interesting, impressive and provides a good opportunity to chemists to project their vision as tool in synthesis under microwave assisted methods. The outcome from this method is excelled yield, short reaction time, cheap, simple experimental procedure and ecofriendly.

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