Chemistry & Biology Interface, 2018, 8, 2, 128-134

RESEARCH PAPER ISSN: 2249 –4820

CHEMISTRY & BIOLOGY INTERFACE

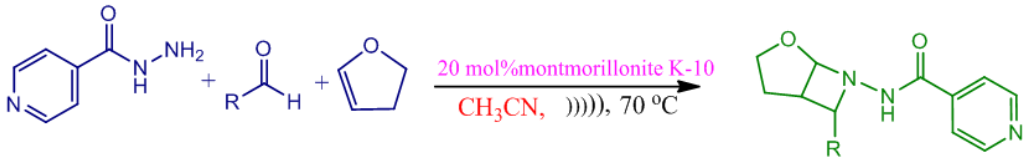
An official Journal of ISCB, Journal homepage; www.cbijournal.com

Synthesis of isonicotinamide derivatives viaone pot three-component Cycloaddition Reaction

Pravin N. Chavan[1]*, Shivaji L. Jadhav[2], D.D. Kayande[3], Megha J. Rai[2]

[1]*DoshiVakilCollege, Goregaon, District - Raigad,-402103. (MH) India.*
[2]*Dr. Rafiq Zakaria College for Women, Aurangabad - 431001. (MH) India.*
[3]*S.B.E.S College, Aurangabad - 431001. (MH) India.*
Email: chemistryp141286@gmail.com, drmegha08@gmail.com
Received 5 March 2018; Accepted 14 April 2018

Abstract: Herein we report a synthesisof N-(7-R)-2-oxa-6-azabicyclo[3.2.0]heptan-6-yl)isonicotinamide derivatives **4**(**a-n**)from isoniazide (**1**), aromatic aldehyde**2**(**a-n**) and 2,3-dihydrofuran (**3**), by novel methodology in which, one pot three-component [2π+2π] cycloaddition reaction were carried out in the presence of Montmorillonite K-10 (green) catalyst under ultrasonic irradiation and targeted compounds were isolated in good to excellent yields.



R= Aromatic aldehydes

Keywords: Cycloaddition, Substituted azitidine, Aromatic Aldehydes, Montmorillonite K-10 and Ultra-sound irradiation.

Introduction:

Now-a-days cycloaddition reactions are a center of heterocyclic synthesis due to its unique properties of cyclic ring formation in single step **[1-2].** Azetidineis one of the most important heterocycles exhibits wide spectrum of biological and pharmaceutical properties such as Antitubercular **[3]** Antimicrobial **[4-6],** Cardiovascular **[7],** Anti-inflammatory**[8],** and Anticonvulsant**[9].** Cycloaddition reactions like [4π+2π] and [2π+2π] using green catalysts have been extensively studied in the last decades as the most straight forward approach for the

preparation of quinoline, isoquinoline, azitidine, β-lactum ring derivatives (natural products) etc**. [10-11]. It** is carried out with high atom economy and low waste product**.** These reactions have beentransported by high pressure, heat, light, sonication, Lewisacids, and bases and recently by green catalyst. Aza-diels Alder reaction, Povarov reactions are mostlyfacilitated through metal catalysts (Lewis acids and Lewis bases) **[12-19].**

Recently most of the researchers have their interest to developed synthetic route for heterocyclic compounds by involving ultrasound irradiation **[20-22].** A large number of convenient one pot multicomponent reactions have been carried out to excellent yields, short reaction timeunder ultrasonic irradiations **[23].**A literature surveyrevealed that, cycloaddition reactions are not fully explored through green catalysts and Ultrasonication approach**[24].**

Nevertheless, the drawbacks associated with most of these methods are the exercise of expensive catalysts, lack of reusability for the catalyst, use of excess catalyst, low product yield, longer reaction, hazardous and tedious workup. Consequently simple, efficient, safer and greener catalytic system is highly demanded. Thus there is a prime need to develop alternative option for conventional methods. Herein, we report Montmorillonite K-10 as a solid acid catalyst for one pot three component synthesis of azetidine derivatives. Montmorillonite K-10 has been used as efficient solid acid catalysts for a number of organic and liquid phase reactions and offer several advantages over classic acids.

In considering the factor discussed above and as part of our ongoing research on the chemical synthesis and biological properties of azetidine derivatives, it is proposed to synthesize of azetidine derivatives by using Montmorillonite K-10 as a catalyst via one-pot three components Aza- Diels-Alder reactions under neat conditions which may helpful to society to get pharmacologically more active compounds.In the present study a series of azetidine derivatives were synthesized under ultra-sonication method and characterized by means of IR, ^{1}HNMR, ^{13}CNMR.

Experimental section

General procedure for (2π+2π) cycloaddition reaction of the N-(7-R)-2-oxa-6-azabicyclo[3.2.0]heptan-6-yl) isonicotinamidederivatives: A mixture of (montmorillonite k10) (20 mol %), isoniazide (1 mmol), aromatic aldehyde (1 mmol) and DHF (1 mmol) in Acetonitrile as solvent (5 mL) weretaken in 25ml round bottom flask and irradiated under ultra-sonication (frequency of 50 Hz and power of 250 V AC, 5.5 L) at 70^{o}C temperature for 2 hours. The reaction progress was assessed on TLC plate using ethyl acetate: hexane (3:7) as solvents. After completion of the reaction, usual work-up procedure was followed to obtain the crude products. The products eluted in ethyl acetate/hexane. At the onset of this work 2,5-dimethoxybenzaldehyde, isoniazid, and 3, 4- dihydropyran was chosen as model reaction to optimize the reaction parameters (Scheme 1).

The chemicals used were of laboratory grade. Melting points of all the synthesized compounds were determined in open capillary tubes and are uncorrected. The IR spectra (KBr disks) were recorded on Brukar FT-IR spectrometer. ^{1}HNMR spectra were recorded on a Brukar DRX-300 and 400 MHz NMR spectrometer and 13C NMR spectra were recorded on a Brukar DRX- 75 and 100 MHz NMR in CDCl3/ DMSO-d6 using (TMS) as internal standard and chemical shifts are inδ(ppm). The purity of each of the compound was checked by thin-layer chromatography (TLC) usingsilica-gel, 60F254 aluminiumsheets as an adsorbent, and visualization was accomplished by iodine/

ultravioletlight.

N-(7-(2,5-dimethoxyphenyl)-2-oxa-6-azabicyclo[3.2.0]heptan-6-yl) isonicotinamide(4a): Buttercream-white powder,m.p. 192-195°C. FTIR (KBr cm^{-1}): 3212, 2999, 2856, 1655, 1170; ^{1}H NMR 400 MHz, DMSO) δ 11.92 (s, 1H, D_2O exchangeable NH), 8.67-8.78 (m, 2H,Ar), 7.7-7.81 (m, 2H,Ar), 7.48-7.49 (m, 1H,Ar), 6.88 (s, 1H,Ar), 6.84-6.87, (m, 1H,Ar), 5.10 (m, 1H,), 4.10 (m, 1H,), 3.97-3.99 (dd, 1H,), 3.78-3.80 (dd,1H), 3.26 (m, 1H), 3.8 (s, 6H), 2.56-2.58 (dd, 1H), 1.80-1.82 (dd, 1H); ^{13}C NMR (100 MHz, DMSO): 162.12, 153.67, 152.82, 150.25, 149.00, 140.70, 132.00,123.65, 122.88, 118.43, 112.85, 110.07, 78.75, 56.28, 55.00, 40.00, 39.11.

N-(7-(3,4-dihydroxyphenyl)-2-oxa-6-azabicyclo[3.2.0]heptan-6-yl) isonicotinamide(4b):
Beige-white powder,m.p132-135°C. FTIR (KBr cm^{-1}): 3211, 3010, 2704, 1675, 1167; ^{1}H NMR 400 MHz, DMSO) δ 12.2 (s, 1H, D_2O exchangeable NH), 8.67-8.80 (m, 2H,Ar), 7.80-8.0(m, 2H,Ar), 7.4 (s, 1H Ar), 7.3 (m, 2H,Ar), 5.30 (s, 2H,), 5.10 (m, 1H,), 4.10 (m, 1H,), 3.97-3.99 (dd, 1H,), 3.78-3.80 (dd,1H), 3.26 (m, 1H), 2.56-2.58 (dd, 1H), 1.80-1.82 (dd, 1H); ^{13}C NMR (100 MHz, DMSO): 162.26, 150.43, 149.91, 148.9, 144.43, 140.64, 136.13, 130.61, 128.56, 123.71, 121.81, 113.06, 78.00, 77.10, 41.38, 40.32, 39.09.

N-(7-(3-hydroxyphenyl)-2-oxa-6-azabicyclo[3.2.0]heptan-6-yl) isonicotinamide(4c):
Light orange powder,m.p 240-242°C. FTIR (KBr cm^{-1}):3231, 2924.84, 2854, 1655, 1178; ^{1}H NMR 400 MHz, DMSO) δ 11.8 (s, 1H, D_2O exchangeable NH), 8.8-8.78 (m, 2H,Ar), 8.32-7.82(m, 2H,Ar), 7.2 (s, 1H Ar), 6.72-6.97 (m, 3H,Ar), 6.2 (s, 1H, OH), 5.12 (m, 1H,), 4.12 (m, 1H,), 3.96-3.98 (dd, 1H,), 3.78-3.80 (dd,1H), 3.25 (m, 1H), 2.54-2.56 (dd, 1H), 1.81-1.83 (dd, 1H); ^{13}C NMR (100 MHz, DMSO): 166.05, 150.96, 148.59, 147.68, 146.74, 145.42, 132.45, 125.69, 121.33, 115.69, 113.65, 110.00, 78.00, 77.11, 41.18, 40.00, 39.12.

N-(7-(4-cyanophenyl)-2-oxa-6-azabicyclo[3.2.0]heptan-6-yl) isonicotinamide(4d):
Mustard-yellow powder,m.p. 210-212°C. FTIR (KBr cm^{-1}): 3234, 2924, 2842, 1250, 1655, 1178; ^{1}H NMR 400 MHz, DMSO) δ 11.9 (s, 1H, D_2O exchangeable NH), 8.34-8.68 (m, 2H,Ar), 7.9 (m, 2H,Ar), 7.64-7.86 (m, 2H,Ar), 6.59-6.93 (m, 2H,Ar), 5.14 (m, 1H,), 4.60 (m, 1H,), 3.97-3.99 (dd, 1H,), 3.78-3.80 (dd,1H), 3.25 (m, 1H), 2.54-2.56 (dd, 1H), 1.81-1.83 (dd, 1H); ^{13}C NMR (100 MHz, DMSO): 162.19, 157.94, 149.98, 140.89, 135.40, 129.79, 123.68, 121.82, 119.29, 118.0, 113.59, 78.90, 76.80, 45.02, 40.00, 38.83.

N-(7-(4-methoxyphenyl)-2-oxa-6-azabicyclo[3.2.0]heptan-6-yl) isonicotinamide(4h):
Light clear goldpowder, m.p. 200-204°C. FTIR (KBr cm^{-1}): 3213, 3001, 2843, 1655, 1134, 1105; ^{1}H NMR 400 MHz, DMSO) δ 11.8 (s, 1H, D_2O exchangeable NH), 8.34-8.78 (m, 2H,Ar), 8.0 (s, 1H,Ar), 7.79-7.81 (m, 2H,Ar), 7.76-7.83 (m, 2H,Ar), 5.14 (m, 1H,), 4.7 (m, 1H,), 3.97-3.99 (dd, 1H,), 3.82 (s,3H), 3.78-3.80 (dd,1H), 3.25 (m, 1H), 2.54-2.56 (dd, 1H), 1.81-1.83 (dd, 1H); ^{13}C NMR (100 MHz, DMSO): 162.56, 150.13, 149.12, 147.51, 132, 130.00, 128.10, 127.59, 121.95, 118.54, 113.06, 110.03, 78.88, 77.00, 56.00, 41.00, 40.00, 39.01.

N-(7-(2,4-dichlorophenyl)-2-oxa-6-azabicyclo[3.2.0]heptan-6-yl) isonicotinamide(4k):
Yellowpowder,m.p. 210-213°C. FTIR (KBr cm^{-1}): 3205, 2998, 2822, 1658, 1151; ^{1}H NMR 400 MHz, DMSO) δ 11.95 (s, 1H, D_2O exchangeable NH), 8.3-8.71 (m, 2H,Ar), 8.28-8.62 (s, 2H,Ar), 7.78-7.84 (s, 1H,Ar), 7.62-7.77

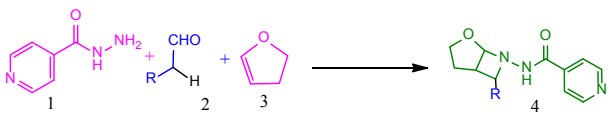
(m, 2H,Ar), 5.14 (m, 1H,), 4.13 (m, 1H,), 3.97-3.99 (dd, 1H,), 3.78-3.80 (dd,1H), 3.25 (m, 1H), 2.53-2.55 (dd, 1H), 1.80-1.82 (dd, 1H); ^{13}C NMR (100 MHz, DMSO): 162.52, 150.45, 148.22, 146.57, 138.21, 131.42, 128.20, 125.82, 122.05, 118.54, 112.66, 111.00, 78.75, 77.00, 41.10, 40.03, 38.71.

N-(7-(4-nitrophenyl)-2-oxa-6-azabicyclo[3.2.0]heptan-6-yl) isonicotinamide(4l):

Yellow powder,m.p. 130-133°C. FTIR (KBr cm^{-1}): 3210, 3049, 3001, 2837, 1652, 1168; ^{1}H NMR 400 MHz, DMSO) δ 11.79 (s, 1H, D_2O exchangeable NH), 8.59-8.67 (m, 2H,Ar), 8.52 (m, 2H,Ar), 7.2-7.68 (m, 2H,Ar), 6.67-7.00 (m, 2H,Ar), 5.14 (m, 1H,), 4.13 (m, 1H,), 3.97-3.99 (dd, 1H,), 3.78-3.80 (dd,1H), 3.25 (m, 1H), 2.54-2.56 (dd, 1H), 1.81-1.83 (dd, 1H); ^{13}C NMR (100 MHz, DMSO): 162.20, 150.33, 148.71, 145.94, 141.23, 130.00, 128.15, 125.00, 121.73, 120.55, 112.32, 111.05, 78.12, 77.40, 41.00, 40.10, 39.19

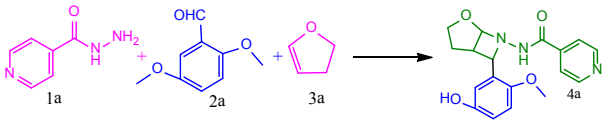
Results and Discussion

The Lewis acid and base catalysts has been extensively used in one pot multi- component [4π+2π] and [2π+2π] cycloaddition reaction but green catalysts are very rarely used for synthesis of substituted azetidine derivatives.In present work, using catalytic amount of Montmorillonite K10 we had carried out the synthesis of N-(7-*R*)-2-oxa-6-azabicyclo[3.2.0]heptan-6-yl) isonicotinamide derivatives through [2π+2π] cycloaddition reaction with good to excellent yields (Table **1,2** and **3**).Montmorillonite K-10 clay as in acidic nature catalyst.



Scheme 1. (General synthetic route)

In our initial studies synthesis of **N-(7-(2,5-dimethoxyphenyl)-2-oxa-6-azabicyclo[3.2.0] heptan-6-yl)isonicotinamide (4a)** through one-pot multi component hetero Diels–Alder reaction of 2,5-dimethoxybenzaldehyde, isoniazid, and 3, 4- dihydropyran was chosen as model reaction to optimize the reaction parameters by using Montmorillonite K10 catalyst.



Scheme 2. (Reaction model)

Initially, we had carried out the model reaction (i.e. isoniazid, 2,5-dimethoxybenzaldehyde, 2,3-dihydrofuran and Montmorillonite k-10 catalyst) at a room temperature by using different solvents such as ACN, DCE, EtOH and THF,we obtained the poor yield (10 to 36%)shown in table 1. Due to the disappointing results at room temperature we forwarded the reaction at reflux temperature with different solvents such as ACN, DCE, EtOH and THF, we observed the yield improvement with respect to room temperature but it was not as per our expectation (19 to 55%) shown in table 1. So we further proceed the same reaction model under ultrasonic irradiation, we observed the improvement in product yield (37-84%) and reduction of reaction time at 70°C in different solvents such as THF, EtOH, CH_3CN and DCE shown in (**Table 1,** entries 9-12).

Table-1. Screening of solvents at different temperature and time.

Entry	Solvent	method	Temp (°C)	Time (h)	Yield (%)
1	CH_3CN	Stir	Rt	12	36
2	DCE	Stir	Rt	12	14
3	EtOH	Stir	Rt	12	22
4	THF	Stir	Rt	12	10
5	THF	Reflux	110°C	08	19
6	EtOH	Reflux	110°C	08	32
7	DCE	Reflux	110°C	08	29
8	CH_3CN	Reflux	110°C	08	55
9	THF	Ultrasound	70°C	02	37

10	EtOH	Ultrasound	70°C	1.30	67
11	CH_3CN	Ultrasound	70°C	1.26	84
12	DCE	Ultrasound	70°C	02	55

(**Reaction conditions**- The reaction was carried out by the addition of isoniazide (1 mmol), 2,5-dimethoxybenzaldehyde (1 mmol), 2,3-dihydrofuran (1 mmol), montmorillonite k10 (20 mol%) catalyst and each solvent 5 mL).

From the data presented in table 1, we had observed that % yield in ACN and EtOH is slightly more as compared to DCE, THF in all three different reaction conditions. It reflects that the yield of products depends on the polarity of solvents. (ACN have more polarity than other solvents).

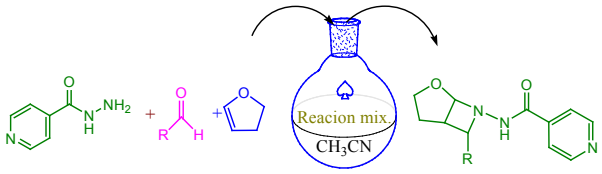


Figure1. One pot three components**,**

Figure 2. Reaction set up

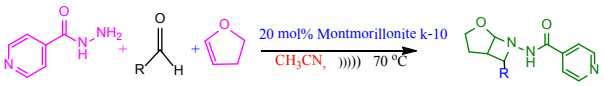
In order to study the quantitative effect of Montmorillonite k10 varied from 10 to 25 mol% the resultant yield observed is 35-84% shown in table 2.Montmorillonite K10 was found to be the suitable catalyst which shows excellent catalytic activity without any byproduct formation. Using this catalyst under ultrasonic irradiation the model reaction furnished 84% of the desired product within 1.26 h at the expense of catalytic amount of Montmorillonite K10 (Scheme 3). The optimum amount of the catalyst in this one-pot three component reaction, was found to be 20 mol % by lowering the catalyst amount, the desired product was obtained in lower yield, while as increased catalyst amount above has no significant effect on reaction rate and isolated yield of product.

Table 2. Screening of the catalyst (Montmorillonite K10) in [2π+2π] cycloaddition reaction.

Entry	(mol %)	Time(h)	Yield (%)
1	10	2	76
2	15	2	81
3	5	2	35
4	20	1.26	84
5	25	2	82

(Reactionconditions- The reaction was carried out by the addition of isoniazide (1 mmol), 2,5-dimethoxybenzaldehyde (1 mmol) and 2,3-dihydrofuran (1 mmol) in Acetonitrile solvent under Ultrasound irradiation at 70°C temperature).

From the observed data, it is foundthat 20 mol % amount of catalyst is suitable for preparation of series of azetidine derivatives (Table **2**), accordingly reaction scheme was planned and well executed for other derivatives.



Scheme 3.

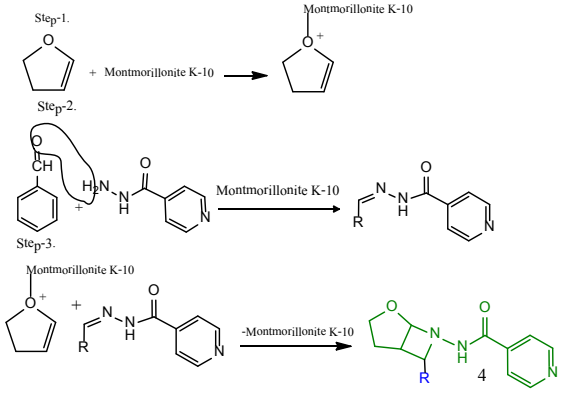
Table3.Synthesis of N-(7-R)-2-oxa-6-azabicyclo[3.2.0] heptan-6-yl) isonicotinamide derivatives 4(a-n) via [2π+2π] cycloaddition reaction.

Entry	R	Prod.	Time (h)	Yield (%)
1	2,5- $(MeO)_2C_6H_3$	4a	1.26	84
2	3,4-$(HO)_2C_6H_3$	4b	1.43	81
3	3- HOC_6H_4	4c	1.35	78
4	4- CNC_6H_4	4d	1.19	79
5	4-HO-3-$MeOC_6H_3$	4e	1.28	78
6	Ph	4f	1.32	81
7	3-BrC_6H_4	4g	1.37	76
8	4-$MeOC_6H_4$	4h	1.30	80
9	4- HOC_6H_4	4i	1.18	78
10	4- ClC_6H_4	4j	1.24	80

11	2,4- $(Cl)_2C_6H_3$	4k	1.32	78
12	4- $NO_2C_6H_4$	4l	1.31	76
13	4- FC_6H_4	4m	1.25	77
14	2,3- $(HO)_2C_6H_3$	4n	1.30	81

(Reaction condition- isoniazide (1 mmol), Aromatic aldehydes (1 mmol), 2,3-dihydrofuran (1 mmol) and (20 mol%) montmorillonite k10 catalyst in Acetonitrile solvent (5 mL) at temperature 70°C under Ultrasonic irradiation).

The feasibility and versatility of this novel methodology was demonstrated by investigations on aza-diels alder cycloaddition reaction of a range of substrates with modified variants of substituted aldehydes (Table**3**). Isoniazid on reaction with substituted aldehydes provided 76-84% yield of corresponding products.All the above reactions were performed at 70°C. Interestingly, the yield of the reaction was decreased when the reaction was performed at room temperature with similar mole ratio; no desired product formation was observed. Encouraging result was obtained (84% yield) by addition of 20 mol% of catalyst. Subsequently, the temperature increased further to 70°C.The reaction was completed in 1.19–1.43 h. Time required for product formation under ultrasonication, % of product yield with respect to substituted aldehyde of corresponding product (**4a-4n)**are summarized in Table- **3**.



Scheme 4. Reaction mechanism of Schiff's base andDHF using Montmorillonite K-10catalyst underultrasounic irradiation.

Synthesis of N-(7-R)-2-oxa-6-azabicyclo[3.2.0] heptan-6-yl)isonicotinamide derivatives using (20 mol%) montmorillonite K 10 catalyst at 70°C in ACN under ultrasonic method evolved an efficient procedure in terms of high yields and less reaction time.The reaction of isoniazid (**1**), substituted aromatic aldehyde **2**(**a-n**) and 2,3-dihydrofuran (**3**) using montmorillonite K-10 as green catalyst lead to desired product (**4a-4n**) in modrate to excellent yields in short reaction times, It proves the feasibility and suitability of ultrasonication method over conventional methods.

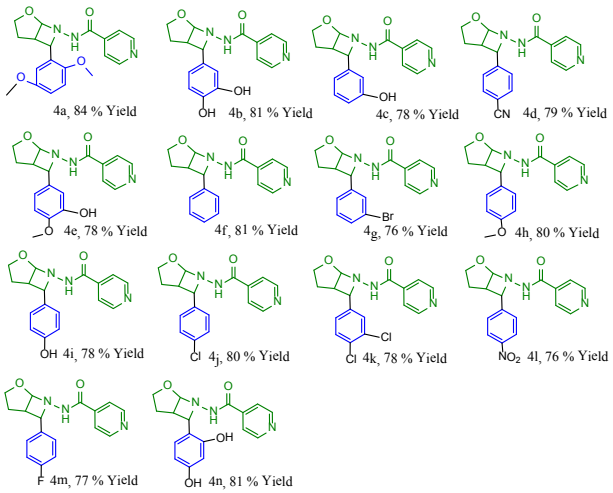


Figure 4.Synthesis of Azetidine derivatives with % yield.

The IR, ^{1}HNMR and ^{13}CNMR data of synthesized compound confirm the formation of substituted N-(7-R)-2-oxa-6-azabicyclo[3.2.0] heptan-6-yl)isonicotinamide derivatives and Mass spectra confirmed the molecular massof the synthesized derivatives.

Conclusion

Substituted N-(7-R)-2-oxa-6-azabicyclo[3.2.0] heptan-6-yl)isonicotinamide derivatives**4**(**a-n**) has been successfully and congenital prepared by using Montmorillonite K10 as green catalyst via one pot multi-component [2π+2π] hetero Diels–Alder reaction under ultra-sonication, which is the important class of heterocyclic

compounds with a diverse pharmacological activity.

Acknowledgements

We are thanks to Management ofDoshiVakil Arts, G.C.U.B. Science and Commerce College, *Goregaon,* for providing the necessary facility during our research.

References

1. W. Carruthers, Cycloadditions Reactions in Organic Synthesis, Pergamon, Oxford, **1990**.
2. S. Kobayashi, K. A. Jorgensen, Cycloadditions Reactions in Organic Synthesis, Wiley-VCH, Weinheim, Germany, **2002**.
3. R. B.Patel, P. S. Desai & et al., Indian J. Chem, **2006**, 45B, 773-778.
4. S. K.Srivastava, R. Dua, S. D. Srivastava, Proc. Nat. Acad. Sci. India, Sec. A: Phys. Sci. **2010**, 80,117-121.
5. P. B.Trivedi, Undavia, Indian J. Chem., **1993**, 32B (7), 760-765.
6. H.Panwar, & et al., Indian J. Chem., **2006**, 45B, 2099-2104.
7. T.Akihisa, S.Mafune& et al., J. Nat. Prod. **2004**, 67(3), 479-480.
8. F.Couty, E.Gwilherm, Syn. LetT.,**2009** (19), 3053-3064.
9. N.Siddiqui, Arana& et al., ActaChim. Slov. **2009**, 56, 462–469.
10. L.Ghosez, J Marchand-Brynaert, Eds.Pergamon, Oxford, **1991**,5, 85.
11. A.de Meijere, Methods of Organic Chemistry, Ed., Thieme, Stuttgart, **1997**, 17e/f.
12. M.Rai, S. Jadhav, M. Farooqui, Journal of Medicinal Chemistry and Drug Discovery, **2015**,ACTRA special issue 1, 2347-9027.
13. P.Emel, TZuhal, Ultrasonic Sonochemistry, **2014,**21, 1600-1607.
14. G.Sundarajan, N. Prabagaran, B. Varghese, Org. Letts. **2001**, 3(13) 1973-1976.
15. J Zhang, C.J.Li,J. Org. Chem. **2002**, 67, 3969.
16. R. B.Borel, L. C.Barbosa, C. R. A. Maltha, S. A. Fernades,Tetrahedron lett.,**2015**,56, 662-665.
17. M. S.Sokamisa, Y. M. Nyondlo, H. Kinfe, ARKIVOV,**2016**, (iii), 313-324.
18. G.Bergonzini, L.Gramigna, A. Mazzanti, M. Fochi, L. Bernardi, A. Ricci, Chem.Commun.,**2010**, 46, 327-329.
19. P.Chavan, A. Ghoti, S. Jadhav, M. Rai, IJCPS, **2018**, 7, 53-57.
20. P.Chavan, S. Jadhav, D. D. Kayande, M. Rai,Heterocyclic Lett.,**2017**,7, 1027-1034.
21. P.Chavan, S. Jadhav, M. Rai, IAJPS,**2018**, (Suppl.) 05 (01), S17-S22.
22. P.Chavan, S. Jadhav, D. Pansare, M. Farooqui, M. Rai, WJPR.,**2018**. 7(5) 1171-1178.
23. R.Cell, H. A.Stefani,Tetrahedron**2009**, 65, 2619-2641.
24. G.Cravotto, P.Cintas, Angew. Chem. Int. Ed.,**2007**, 46, 5476-5478.
25. Y.Hu, Y.Zou, H. Wu, D. Q. Shi, Ultrason.Sonochem. **2012**, 19,264-269.