DOI:10.7598/cst2017.1382
ISSN:2278-3458

Chemical Science Transactions
2017, **6(3),** 428-436

RESEARCH ARTICLE

One Pot Multicomponent Green Synthesis of Triaryl Imidazoles Catalysed by Nano Nickel Cobalt Ferrite

KORUPOLU RAGHU BABU, SRIVIDHYA MARIPI,
SURI BABU MADASU* and NOOKA RATNAM APPNA

Department of Engineering Chemistry, A. U. College of Engineering (A),
Andhra University, Visakhapatnam - 530 003, Andhra Pradesh, India
suribabumadasu@yahoo.co.in

Received 23 January 2017 / Accepted 24 February 2017

Abstract: 2,4,5-Triaryl imidazoles have been developed by the reaction of benzil, ammonium acetate and aldehydes in ethanol using magnetically reusable nano nickel cobalt ferrite ($NiCoFe_2O_4$). It is a simple, highly efficient, three component one-pot green synthetic method. The advantages of this procedure are high product yields, easy operational method, mild conditions, magnetically recoverable and re-usable catalyst. Most important to all, the catalyst can be recovered by simple filtration and reusable up to five cycles without losing its catalytic activity.

Keywords: Multi component reaction, Heterogeneous catalyst, Nickel cobalt ferrites, One pot synthesis, 2,4,5-Tri substituted imidazoles

Introduction

A multicomponent reaction (MCR) is a process in which three or more easily accessible components are combined together in a single reaction vessel to produce a final product displaying features of all inputs and thus offers greater possibilities for molecular diversity per step with a minimum of synthetic time and effort. In spite of the significant useful attributes of MCRs for modern organic chemistry and their suitability for building up large compound libraries these reactions were of limited interest in the past fifty years. However, in the last decade, with the introduction of high-throughput biological screening, the importance of MCRs for drug discovery has been recognized and considerable efforts from both academic and industrial researchers have been focussed especially on the design and development of multi-component procedures for the generation of libraries of heterocyclic compounds. The advantages of MCR's are atom economy, less time consuming, easy purification process and avoid protection-deprotection steps. Therefore, the design and development of efficient and green MCRs focussed on a target molecule is one of the most important challenges in organic synthesis.

Imidazoles are a class of heterocyclic compounds that contain nitrogen hetero atom, these exhibits a wide range of biological activities in both *in vitro* and *in vivo*. In recent years, chemical and biological usefulness of tri substituted imidazoles have great applications in the field of pharmacological studies. A wide variety of biological properties have been found for imidazole derivatives such as therapeutic agent[1], herbicidal[2], antibacterial[3], antitumor[4], glucagon receptors[5], fungicididal[6], antithrombotic activity[7]. These are also act as inhibitors of P^{38} MAP kinase[8], *B*-Raf kinase[9], transforming growth factor b1 (TGF-b1) type 1 activin receptor-like kinase (ALK5)[10], cyclooxygenese-2 (COX-2)[11] and biosynthesis of interleukin-1 (1L-1)[12]. These are also used as modulators of *p*-gycloprotein (p-gp), mediated multi drug resistance (MDR)[13] and pesticides[14].

Poly substituted imidazoles were first synthesised in 1882 by using an aldehyde with 1, 2- dicarbonyl compound and ammonia (Radziszewski 1882)[15]. Afterwards, various methods have been developed for the synthesis of 2,4,5-trisustituted imidazoles by using various catalysts such[16] as Al_2O_3-$ZrCl_4$, $NiCl_2.6H_2O$, Zeolite Hy[17,18], $InCl_3.3H_2O$, DABCO[19,20] and PEG-400[21], acetic acid[22], silica supported sulphuric acid[23], ceric ammonium nitrate[24], iodine[25], *L*-prolin[26] trichloro iso cyanuric acid[27,28,29] $Zr(acac)_4$ and KH_2PO_4. According to literature survey, various synthetic protocols have been reported for the synthesis of substituted imidazoles using various type of copper containing catalyst such as synthesis of imidazoles through the Cu_2O catalysed cross-cyclo addition between two different isocyanides, synthesis of multi substituted imidazoles via CuI–catalysed [3+2]cyclo additions[30].

The above reported methods are having its own importance and virtues, however most of these methods require harmful catalysts, longer reaction time, poor yield, difficult work up and effluent pollution. Thus they were not appropriate for synthesis of structurally diverse imidazoles. Development of clean and highly yielding and environmentally benign approaches is still desirable and much in demand.

In recent years, magnetic nano particles have emerged as a useful group of heterogeneous catalysts. Separation of magnetic nano particles is simple and an attractive alternative to filtration as it prevents loss of catalyst and enhances reusability. The use of low cost and readily available species as catalyst plays a significant role for economic feasibility of the chemical process. The greener generation of nanoparticles and their eco-friendly applications in catalysis via magnetically recoverable and recyclable nano-catalysts for a variety of oxidation, reduction and condensation reactions[31-34], has made an incredible impact on the development of sustainable pathways. Magnetically recyclable nano catalysts and their use in benign media is an ideal merge for the development of sustainable methodologies in organic synthesis.

Due to the effective activity of magnetically separable nickel cobalt ferrite nano particles (NiCoFeNPs) have the advantages of recyclability, easy work-up and clean reaction profiles apart from the lack of necessity ligands and in minimizing the organic waste generation when compared to the conventational catalytic systems. As a part of our ongoing research towards the synthesis of biologically active heterocyclic compounds using magnetically separable nano catalysts, keeping environmental friendly methods in mind, here we report an efficient and simple work-up method for the synthesis of tri substituted imidazoles using magnetic separable nano nickel cobalt ferrite as heterogeneous catalyst under reflux conditions. The synthesised imidazole derivatives were characterised by IR, H^1 NMR and Mass spectral data.

Experimental

All the chemicals were purchased from Sigma Aldrich with purity not less than 99.9%. Analytical Thin Layer Chromatography (TLC) was carried out by using silica gel 60 F254 pre-coated plates. Visualization was accomplished with UV lamp. All the products were characterized by their IR, H^1 NMR and Mass spectra. ^{1}H NMR was recorded on 300 MHz in CDCl3/DMSO and the chemical shifts were reported in parts per million (ppm, δ) downfield from the Tetramethyl silane (TMS).

General experimental procedure for synthesis of 2,4,5-tiarylimidazoles

Benzil (10 mmol), aromatic aldehyde (10 mmol), ammonium acetate (20 mmol) and nano nickel cobalt ferrite (20 mol %) were taken in a round bottomed flask and the contents are dissolved in 5 mL of ethanol. Then the reaction mixture was stirred for 20 min at reflux temperature. The progress of the reaction was monitored by TLC (*n*-hexane: ethyl acetate 4:1). After completion of the reaction the catalyst was separated from the reaction mixture by using an external magnet and then the reaction mixture was concentrated by evaporating solvent on a rotary evaporator. Then the dried product was recrystallized from hot ethanol for several times to get the corresponding pure product. The products were confirmed by IR, H^1 NMR and Mass spectras.

Spectral Data

2,4,5-Triphenyl -1-H-imidazole **(4a)**

White solid; M.P: 273-275 °C; ^{1}H NMR (300 MHz, $CDCl_3$): δ=7.95-7.36 (m, 15H), 9.31(brs, N-H), FTIR (KBr, cm^{-1}): 3453(N-H), 3060(C-H), 1657(C=C), 1576(C=N), ESI-MS (*m/z*): 297 (M^++1).

2-(4-Methyl phenyl)-4,5-diphenyl-1-H-imidazole **(4b)**

White solid; M.P: 184-186 °C. ^{1}H NMR (300 MHz, $CDCl_3$): δ = 2.32 (s, CH3), 7.12-7.63 (m, 10H), 7.75 (d, 2H, J=10 Hz), 7.32(d, 2H, J=10 Hz); FTIR (KBr, cm^{-1}): 3452 (N-H), 1605 (C=C), 1585 (C=N); ESI-MS (*m/z*): 311 (M^++1).

2-(4-Methoxy phenyl)-4,5-diphenyl-1-H-imidazole **(4c)**

white solid; M.P: 222-224 °C; ^{1}H NMR (300 MHz, $CDCl_3$): δ=3.84 (S, 3H), 6.93-6.92 (d, J=8.8Hz, 2H), 7.52-7.23 (m, 10H), 7.82-7.80 (d, J=8.8Hz, 2H); FTIR (KBr, cm^{-1}): 3450 (N-H), 1612 (C=C), 1579 (C=N), 1385 (C-O); ESI-MS (*m/z*): 327 (M^++1).

2-(2-Methoxy phenyl)-4,5-diphenyl-1-H-imidazole **(4d)**

White solid; M.P: 210-212 °C; ^{1}H NMR (300 MHz , $CDCl_3$): δ=11.81(s, 1H), 8.03 (d, J=7.5Hz, 1H), 7.50-7.01 (m, 13H), 3.92 (s, 3H); FTIR (KBr, cm^{-1}): 3430 (N-H), 1615 (C=C), 1580 (C=N), 1385 (C-O); ESI-MS(*m/z*): 327 (M^++1).

2-(4-Acetamido phenyl)-4,5-diphenyl-1-H-imidazole **(4e)**

White solid; M.P: 220-222 °C ; ^{1}H NMR (300 MHz, $CDCl_3$): δ=3.5 (s, 1H), 2,13 (s, 3H), 7.13 – 7.53 (m, 10H), 7.65 (d, 2H, J=10Hz), 7.22 (d, 2H, J=10 Hz); FTIR (KBr, cm^{-1}): 3450 (N-H), 1612 (C=C), 1579 (C=N), 1690 (C=O), 1610 (N-H def), 2920 (C-H), ESI-MS(*m/z*): 354 (M^++1).

2-(2-Hydroxy phenyl)-4,5-diphenyl-1-H-imidazole **(4f)**

White solid; M.P: 117-119 °C ; ^{1}H NMR (300 MHz, $CDCl_3$): δ=12.62 (s, 1H), 8.03 (dd, 2H, J=8.7Hz), 7.59 (d, 2H, J=8Hz), 7.49 (s, 1H), 7.27-7.34 (m, 10H); FTIR (KBr, cm^{-1}): 3431 (N-H), 3300 (O-H), 3062 (=C-H), 1615 (C=C), 1580 (C=N), 1200 (C-O); ESI-MS(*m/z*): 313 (M^++1).

2-(4-Hydroxy, 3-methoxy phenyl)-4,5-diphenyl-1-H-imidazole **(4g)**

White solid; M.P: 215-217 °C; ^{1}H NMR (300 MHz, $CDCl_3$): δ=12.50 (s, 1H), 9.11 (s, 1H), 7.46-7.54 (m, 6H), 7.42 (t, 2H), 7.35 (t, 1H), 7.25 (t, 2H), 7.21 (t, 1H), 7.01 (d, 1H), 3.80 (s, 3H); FTIR (KBr, cm^{-1}): 3408 (N-H), 3260 (O-H), 1595 (C=C), 1510 (C=N); ESI-MS(*m/z*): 343 (M^++1).

2-(4-Nitro phenyl)-4,5-diphenyl-1-H-imidazole **(4h)**

Yellow solid; M.P: 199-201 °C ; ^{1}H NMR (300 MHz, $CDCl_3$): δ=12.81 (s, 1H), 8.01-7.42 (m, 14H); FTIR (KBr, cm^{-1}): 3402 (N-H), 2928 (=C-H), 1598 (C=C), 1519 (C=N), 1346 (NO_2), 856 (C-N); ESI-MS(*m/z*): 342 (M^++1).

2-(2-Nitro phenyl)-4,5-diphenyl-1-H-imidazole **(4i)**

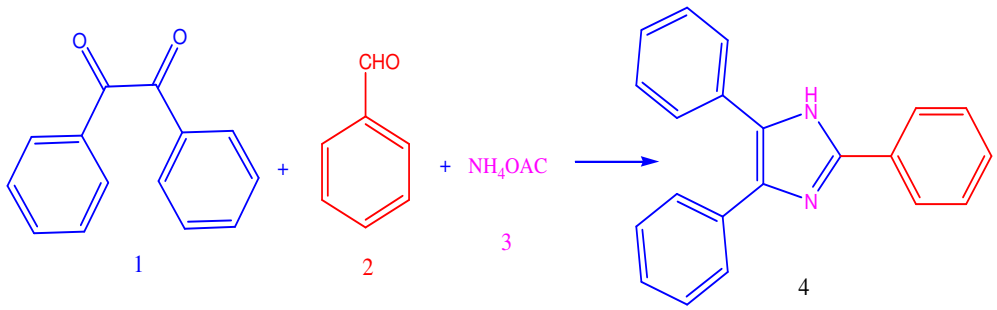
Yellow solid; M.P: 298-300 °C; ^{1}H NMR (300 MHz, $CDCl_3$): δ=13.10 (s, 1H), 8.95 (s, 1H), 8.53 (d, 1H, J=7.5 Hz), 8.23 (d, 1H, J=7.8 Hz), 7.81 (d, 1H, J=7.8 Hz), 7.54-7.33 (m, 10H); FTIR (KBr, cm^{-1}): 3448 (N-H), 3068 (=C-H), 1526 (=C-N), 1352 (NO_2); ESI-MS(*m/z*): 342 (M^++1).

2-(4-Chloro phenyl)-4,5-diphenyl-1-H-imidazole **(4j)**

White solid; M.P: 248-250 °C ; ^{1}HNMR (300 MHz, $CDCl_3$): δ=12.76 (s, 1H), 7.22-7.55 (m, 12H), 8.11 (d, 2H, J=8.8 Hz); FTIR (KBr, cm^{-1}): 3445 (N-H), 2924 (=C-H), 1646 (C=C) and 1255 (C-N); ESI-MS(*m/z*): 331 (M^++1).

Results and Discussion

Initially a model reaction was conducted by using different solvents and different mol% of catalyst for synthesis of 2,4,5-triaryl imidazoles to investigate the feasibility of the reaction. Benzil, benzaldehyde and ammonium acetate were taken in different solvents (DMSO, acetonitrile, dichloromethane, chloroform and ethanol) in the presence of catalyst $NiCoFe_2O_4$ NPs and results are summarized in Table 1. It was clearly observed that low yield of product is obtained with DMSO, acetonitrile, dichloromethane, chloroform (20%, 24%, 27% and 29%, Table 1, entry 1-4) respectively even after 2 h stirring. It was clearly observed that the reaction was carried out with ethanol as a solvent at 75-78 °C provided 60% yield with in 2 h. After finding the suitable solvent, the model reaction is performed with different mol% of $NiCoFe_2O_4$ catalyst and observed that 20 mol% suitable to obtained maximum yield at neat condition (Table 2, entry 4). No change was observed on further enhancing the catalyst mol%.



Scheme 1. Synthesis of 2,4,5-triaryl imidazoles

Table 1. Screening of solvent for synthesis of 2,4,5-trisubstituted imidazoles

Entry	Solvent	Catalyst mol %	Temperature °C	Time	Yield[a], %, w/w
1	DMSO	5	95-100	2 h	20
2	Acetonitrile	5	65-70	2 h	24
3	Dichloro methane	5	45-50	2 h	27
4	Chloroform	5	45-50	2 h	29
5	Ethanol	5	75-78	2 h	60

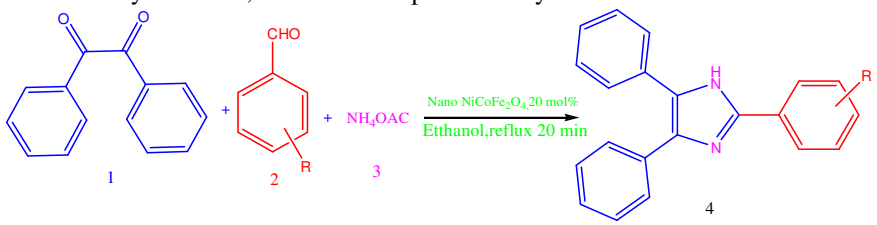
[a]*Isolated yields*

Table 2. Effect of $NiCoFe_2O_4$ catalyst percentage on synthesis of 2,4,5-trisubstituted imidazoles

Entry	Solvent	Catalyst, mol %	Temperature, °C	Time	Yield[a], %, w/w
1	Ethanol	5	75-78	2 h	60
2	Ethanol	10	75-78	1 h	72
3	Ethanol	15	75-78	40 min	85
4	Ethanol	20	75-78	20 min	95
5	Ethanol	25	75-78	20 min	95
6	Ethanol	30	75-78	20 min	95

[a]*Isolated yields*

With the optimised conditions in hand, the reaction was performed with different benzaldehydes (Scheme 2) to explore the scope and generality of the present protocol and the results of these observations are summarized in Table 3. From the results, it can be concluded that the aromatic aldehydes with having electron withdrawing groups (Table 3, entries 8 & 9) reacts at faster rate compared with aromatic aldehydes substituted with electron releasing groups (Table 3, entries 2-5). The structures of synthesized imidazoles were confirmed by H^1 NMR, IR and Mass spectral analysis.



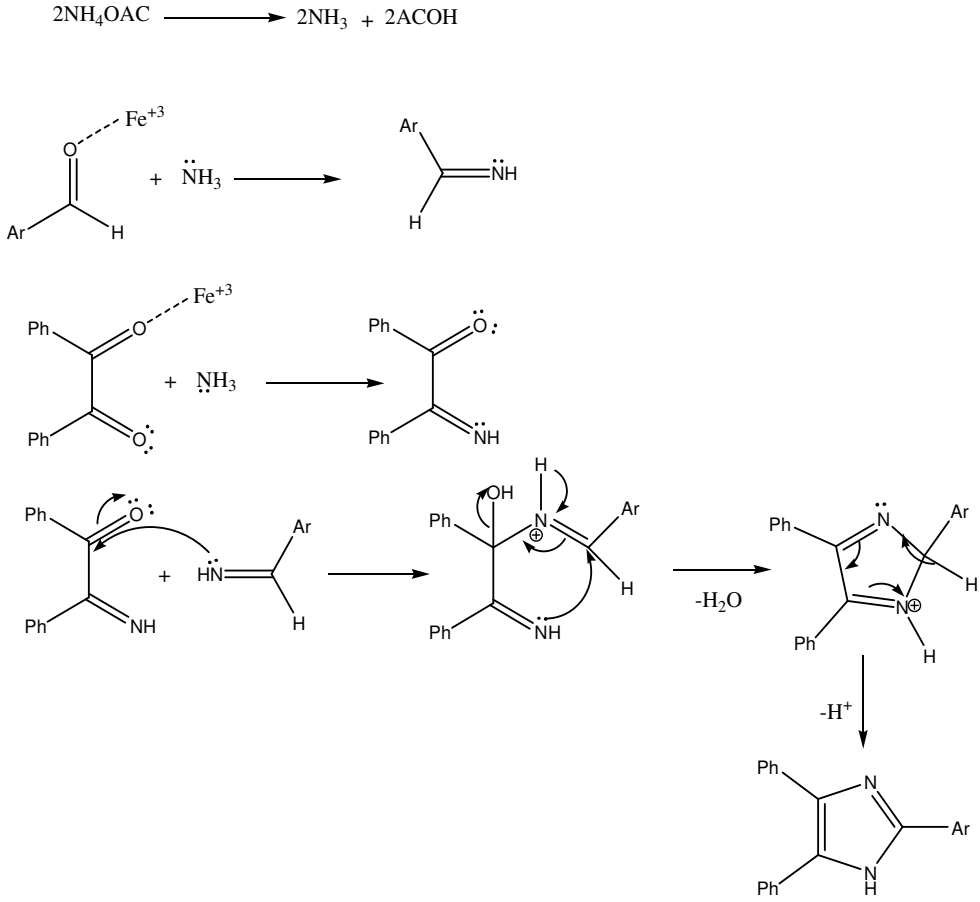
Scheme 2. $NiCoFe_2O_4$ catalysed synthesis of 2,4,5-trisustituted imidazoles derivatives

Table 3. $NiCoFe_2O_4$ catalysed synthesis of 2,4,5-tri substituted imidazole derivatives

Entry	Product	R	Time, min	Yield[a], %, w/w
1	**4a**	H	20	94%
2	**4b**	4-CH_3	20	92%
3	**4c**	4-OCH_3	20	90%
4	**4d**	2-OCH_3	20	89%
5	**4e**	4-$NHCOCH_3$	20	91%
6	**4f**	2-OH	20	90%
7	**4g**	4-OH, 3-OCH_3	20	92%
8	**4h**	4-NO_2	15	98%
9	**4i**	2-NO_2	15	97%
10	**4j**	4-Chloro	20	92%

[a]*Isolated yields*

The plausible mechanism for the formation 2,4,5-trisubstituted imidazoles by using Ni CoFeNPs is shown in Scheme 3.



Scheme 3. Plausible mechanism for the formation of 2,4,5-tri substituted Imidazoles

Table 4. Comparison of various catalysts with nickel cobalt ferrite in the synthesis

Entry	Catalyst	Solvent	Temperature, °C	Time, min	Yield[a], %, w/w	Reference
1	$InCl_3.3H_2O$	Methanol	25-30	492	76	19
2	$NiCl_2.6H_2O/Al_2O_3$	Ethanol	75-80	90	89	17
3	L-proline	Methanol	60-64	540	87	26
4	Zeolite-Hy	-	120	90	56	18
5	KH_2PO_4	Methanol	25-30	60	89	29
6	$Zr(acac)_4$	Ethanol	75-80	120	95	28
7	DABCO	t-Butanol	60-65	720	92	20
8	CAN	Methanol		600	75	24
9	$NiCoFe_2O_4$	Ethanol	75-80	20	95	Present work

[a]*Isolated yields*

The reusability of $NiCoFe_2O_4$ NPs is one of the most important advantages of this protocol that makes it useful for practical commercial applications. We have examined the recyclability of $NiCoFe_2O_4$ NPs catalyst for the model reaction. Interestingly, the recovered catalyst could be reused for up to five cycles under optimized reaction conditions without leaching of the Co, Ni and Fe metals which is evident from Table 5. The catalyst was separated by using a magnet after completion of the reaction, washed with water followed by chloroform, dried in oven and reused for the next cycle.

Table 5. Productivity with re-cycle catalyst

Entry	Catalyst re-use	Yield[a], %, w/w
1	1st cycle	94
2	2nd cycle	92
3	3rd cycle	90
4	4th cycle	89
5	5th cycle	87

[a]*Isolated yields*

Conclusion

We have described a novel, efficient, multi-component one pot green synthetic method using nano nickel cobalt ferrite catalyst and ethanol as a solvent. The novelty and synthetic utility of this method is demonstrated in the efficient synthesis of 2,4,5-triphenyl imidazole derivatives. The advantages of this method include its simplicity of operation, cleaner reaction, and good to excellent yields. Further, the purification of the product is simple involving filtration. The catalyst is easily separated by using external magnet and is reusable up to five cycles.

Acknowledgement

The corresponding author is grateful to CSIR, New Delhi for supporting through fellowship (JRF&SRF) & Department of Engineering Chemistry, AUCE (A), Andhra University and Visakhapatnam for providing general lab facilities. The corresponding author is also grateful to Prof. K. Raghu Babu and Dr. M. Suri Babu for their valuable and constant support.

References

1. Kidwai M, Saxena S, Ruby and Rastogi S, *Bull Korean Chem Soc.,* 2005, **26(12),** 2051-2053; DOI:10.5012/bkcs.2005.26.12.2051
2. Zhang C, Sarshar S, Moran E J, Krane S, Rodarte J C, Benbatoul K D, Dixon R and Mjalli A M M, *Bioorg Med Chem Lett.,* 2000, **10(23),** 2603-2605; DOI:10.1016/S0960-894X(00)00521-7
3. Antolini M, Bozzoli A, Ghiron C, Kennedy G, Rossi T and Ursini A, *Bioorg Med Chem Lett.,* 1999, **9(7),** 1023-1028; DOI:10.1016/S0960-894X(99)00112-2
4. Wang L, Woods K W, Li Q, Barr K J, McCroskey R W, Hannick S M, Gherke L, Credo R B, Hui Y H, Marsh K, Warner R, Lee J Y, Zielinski M N, Frost D, Rosenberg S H and Sham H L, *J Med Chem.,* 2002, **45(8),** 1697-1711; DOI:10.1021/jm010523x
5. De Laszlo S E, Hacker C, Li B, Kim D, MacCoss M, Mantlo N, Pivnichny J V, Colwell L, Koch G E, Cascieri M A and Hagmann W K, *Bioorg Med Chem Lett*., 1999, **9(5),** 641-646; DOI:10.1016/S0960-894X(99)00081-5

6. a) Ucucu U, Karaburun N G and Isikdag I, *Farmaco*., 2001, **56(4),** 285-290; DOI:10.1016/S0014-827X(01)01076-X b) Wolkenberg S E, Wisnoski D D, Leister W H, Wang Y, Zhao Z and Lindsley C W, *Org Lett*., 2004, **6(9),** 1453-1456; DOI:10.1021/ol049682b
7. Lombardino J G and Wiseman E H, *J Med Chem*., 1974, **17(11),** 1182-1188; DOI:10.1021/jm00257a011
8. Lee J C, Laydon J T, McDonnell P C, Gallagher T F, Kumar S, Green D, McNulty D, Blumenthal M J, Keys J R, Vatter S W L, Strickler J E, McLaughlin M M, Siemens I R, Fisher S M, Livi G P, White J R, Adams J L and Young P R, *Nature,* 1994, **372,** 739-746; DOI:10.1038/372739a0
9. Takle A K, Brown M J, Davies S, Dean D K, Francis G, Gaiba A, Hird A W, King F D, Lovell P J, Naylor A, Reith A D, Steadman J G and Wilson D M, *Bioorg Med Chem Lett*., 2006, **16(2),** 378-381; DOI:10.1016/j.bmcl.2005.09.072
10. Khanna I K, Weier R M, Yu Y, Xu X D, Koszyk F J, Collins P W, Koboldt C M, Veenhuizen A W, Perkins W E, Casler J J, Masferrer J L, Zhang Y Y, Gregory S A, Seibert K and Isakson P C, *J Med Chem*., 1997, **40(11),** 1634-1647; DOI:10.1021/jm9700225
11. Lange J H, Van S H H, Coolen H K, Adolfs T J, McCreary A C, Keizer H G, Wals H C, Veerman W, Borst A J, De Looff W, Verveer P C and Kruse C G, *J Med Chem*., 2005, **48(6),** 1823-1838; DOI:10.1021/jm040843r
12. Gallagher T H, Fier-Thompson S M, Garigipati R S, Sorenson M E, Smietana J M, Lee D, Bender P E, Lee J C, Laydon J T and Griswold D E, *Bioorg Med Chem Lett.,* 1995, **5(11),** 1171-1176; DOI:10.1016/0960-894X(95)00189-Z
13. Newman M J, Rodarte J C, Benbatoul K D, Romano S J, Zhang C, Krane S, Moran E J, Uyeda R T, Dixon R, Guns E S and Mayer L D, *Cancer Res*., 2000, **60(11),** 2964-2972.
14. Wang L, Woods K W, Li Q, Barr K J, McCroskey R W, Hannick S M, Gherke L, Credo R B, Hui Y H, Marsh K, Warner R, Lee J Y, Zielinsky- Mozng N, Frost D, Rosenberg S H and Sham H L, *J Med Chem*., 2002, **45(8),** 1697-1711; DOI:10.1021/jm010523x
15. Esfahani M N, Montazerozohori M and Tooba A, *Chemical Papers,* 2015, **69(11),** 1491-1499; DOI:10.1515/chempap-2015-0156
16. Sharma G V M, Jyothi Y and Lakshmi P S, *Synthetic Commun.,* 2006, **36(20),** 2991-3000; DOI:10.1080/00397910600773825
17. Heravi M M, Bakhtiari K, Oskooie H A and Taheri S, *J Mole Cataly A: Chem.,* 2007, **263(1-2),** 279-281; DOI:10.1016/j.molcata.2006.08.070
18. Balalaei S and Arabanian A, *Green Chem.*, 2000, **2,** 274-276; DOI:10.1039/B006201O
19. Sharma S D, Hazarika P and Konwar D, *Tetrahedron Lett.,* 2008, **49,** 2216-2220; DOI:10.1016/j.tetlet.2008.02.053
20. Murthy S N, Madhav B and Nageswar Y V D, *Tetrahedron Lett.,* 2010, **51(40),** 5252-5257; DOI:10.1016/j.tetlet.2010.07.128
21. Wang X C, Gong H P, Quan Z J, Li L and Ye H L, *Chin Chem Lett.,* 2009, **20(1),** 44-47; DOI:10.1016/j.cclet.2008.10.005
22. Sarshar S, Siev D and Mjalli A M M, *Tetrahedron Lett,* 1996, **37(5),** 835-838; DOI:10.1016/0040-4039(95)02334-8
23. Shaabani A and Rahmati A, *J Mole Cataly A: Chem.,* 2006, **249(1-2),** 246-248; DOI:10.1016/j.molcata.2006.01.006

24. Sangshetti J N, Kokare N D, Kotharkara S A and Shinde D B, *J Chem Sci.,* 2008, **120(5),** 463-467; DOI:10.1007/s12039-008-0072-6
25. Kidwai M, Mothsra P, Bansal V, Somvanshi R K, Ethayathulla A S, Dey S and Singh T P, *J Molecular Cataly A*, 2007, **265(1-2),** 177-182; DOI:10.1016/j.molcata.2006.10.009
26. Samai S, Nandi G C, Singh P and Singh M S, *Tetrahedron,* 2009, **65(49),** 10155-10161; DOI:10.1016/j.tet.2009.10.019
27. Hojati S F, Nezhadhoseiny S A and Beykzadeh Z, *Monatshefte für Chemie*, 2013, **144,** 387-390; DOI:10.1007/s00706-012-0830-5
28. Khosropour A R, *Ultrasonics Sonochem.,* 2008, **15(5),** 659-664; DOI:10.1016/j.ultsonch.2007.12.005
29. Joshi R S, Mandhane P G, Shaikh M U, Kale R P and Gill C H, *Chine Chem Lett.,* 2010, **21(4),** 429-432; DOI:10.1016/j.cclet.2009.11.012.
30. Tang D, Wu P, Liu X, Chen Y X, Guo S B, Chen W L, Li J G and Chen B H, *J Org Chem.*, 2013, **78(6),** 2746-2750; DOI:10.1021/jo302555z
31. Polshettiwar V, Baruwati B and Varma R S, *Chem Commun.,* 2009, **14,** 1837-1839; DOI:10.1039/B900784A
32. Baig R B N and Varma R S, *Green Chem.,* 2013, **15,** 398-417; DOI:10.1039/-C2GC36455G
33. Polshettiwar V, Baruwati B and Varma R S, *Green Chem.,* 2009, **11,** 12-31.
34. Polshettiwar V and Varma R S, *Tetrahedron* 2010, *66*, 1091-1097.