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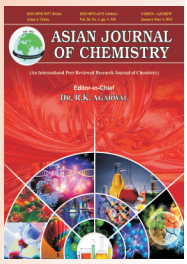

Multicomponent One-pot Green Synthesis of 1-Amidoalkyl-2-naphthols Promoted by *p*-Nitrobenzoic Acid Under Solvent-free Condition

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A series of sixteen 1-amidoalkyl-2-naphthols derivatives were synthesized by one-pot multicomponent condensation reaction of β-naphthol with various aromatic aldehydes and amides under thermal (hot plate and oil bath) and microwave irradiation techniques promoted by *p*-nitrobenzoic acid and the corresponding products were obtained in good to excellent yield (82-92 %) under solvent-free condition.

Keywords: 1-Amidoalkyl-2-naphthol, *p*-Nitrobenzoic acid, Multicomponent one-pot reaction, Microwave irradiation, Solvent-free.

INTRODUCTION

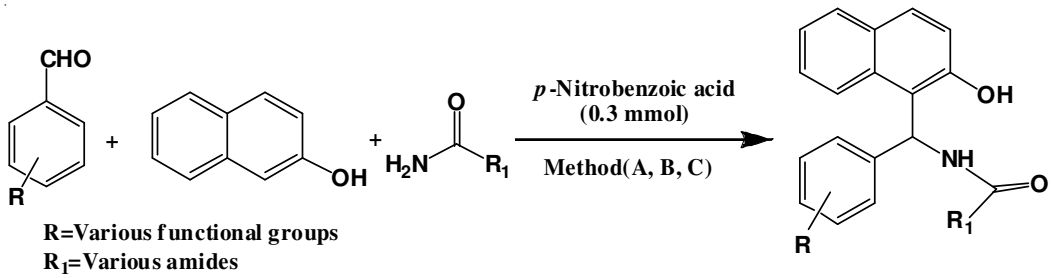
Since the last decade, there is a tremendous growth of research in the field of one-pot multicomponent reactions for the synthesis of wide variety of organic compounds. One of the multicomponent reactions of current interest is synthesis of 1-amidoalkyl-2-naphthols. The amidoalkyl-naphthols derivatives were paid much attention in synthetic organic chemistry for their various biological activities. It has been reported that 1-amidoalkyl-2-naphthols can be converted to biological active 1-aminoalkyl-2-naphthols derivatives by an amide hydrolysis reaction exhibiting depressor and bradycardic activities in humans[1-2]. In addition, 1-amidoalkyl-2-naphthols can also be converted to 1,3-oxazine derivatives[3-4] which has widespread area of useful biological activities[5-11]. It has been reported to be an essential synthons for drug design and discovery of potent drugs such as nucleoside antibiotics and HIV protease inhibitors such as ritonavir and lipinavir[12,13].

During last few years various methods have been reported for the synthesis of amidoalkyl-naphthols in the presence of variety of catalysts such as chlorosulphonic acid[14], *p*-toluene sulphonic acid[15], $NaHSO_4 \cdot H_2O_{16}$, $Fe(HSO_4)_3$[17], $Sr(OTf)_2$[18], iodine[19], hetropoly acid $K_5CoW_{12}O_{40} \cdot 3H_2O$[20], hetropoly acid catalysts like cation-exchange resins[21], silica supported perchloric acid[22,23], $FeCl_3 \cdot SiO_2$[24], montmorillonite K10 clay[25], silica sulfuric acid[26], sulfamic acid[27], *N*,*N*,*N*',*N*'-tetrabromobenzene-1,3-disulfonamide[28] and ionic liquids[29]. However, majority of them suffer from several limitations such as high temperature, long reaction time, the use of expensive reagents, low yields of products, high catalyst loading, corrosive reagents, strongly acidic conditions and further purification of products. Therefore development and introduction of convenient and efficient methods for the preparation of 1-amidoalkyl-2-naphthols is of practical importance is still in demand.

In recent years, green and sustainable chemistry has become a subject of intensive research and the studies in this area have led the development of cleaner and relatively benign chemical processes. Among them, much effort has been devoted towards multicomponent and microwave assisted organic synthesis (MAOS)[30].

In continuation of our earlier work in the development of novel catalysts for the synthesis of various heterocyclic and synthetic intermediates[31], we now describe a simple, general and efficient protocol for the synthesis of 1-amidoalkyl-2-naphthols *via* one-pot condensation reaction using catalytic amount of *p*-nitrobenzoic acid using thermal and microwave techniques by **Method A, B** and **C**. The synthetic approach is outlined in (**Scheme-I**).

The *p*-nitrobenzoic acid is readily available, inexpensive reagents and environmentally benign light yellow crystals which is stable and can conveniently be handled and removed from the reaction mixture. Thus, the remarkable catalytic activities together with their operational simplicity make them the most suitable catalyst for the synthesis of 1-amidoalkyl-2-naphthols. The *p*-nitrobenzoic acid has earlier been used for the efficient synthesis of 1,5-benzodiazepines[32].



R=Various functional groups
R_1=Various amides

Method A: Microwave irradiation
Method B: Oil bath
Method C: Hot plate

Scheme-I: Synthesis of 1-amidoalkyl-2-naphthols

EXPERIMENTAL

All the chemicals were purchased from commercial suppliers. The melting points were determined on Veego-programmable melting point apparatus (microprocessor based) and are uncorrected. ^{1}H NMR spectra were obtained using Brucker AC-400 F, 400 MHZ spectrometer. IR spectra were obtained on Perkin Elmer 882 Spectrum and RXI, FT-IR. Elemental analyses for C, H and N were performed on Thermo-flash EA-1112 CHNS-O Analyzer. Reactions were monitored and the homogeneity of the products was checked by TLC. All chemicals were dried and freshly prepared prior to use according to standard procedure.

General procedure for synthesis of 1-amidoalkyl-2-naphthols

Method A: A mixture of substituted aromatic aldehydes (1 mmol), 2-naphthol (1 mmol) and substituted amides (1.2 mmol) in the presence of effective amount of *p*-nitrobenzoic acid (30 mol %, 0.3 mmol) were taken in 20 mL conical flask and was subjected to microwave irradiation in microwave oven (LG model MS1927C) at 450 W and each pulse was of 30 s for the appropriate time to avoid overheating. The progress of reactions was monitored by TLC (ethyl acetate/*n*-hexane = 2:8). After completion of the reaction, a solid was obtained. It was washed with water and filtered. Then the solid residue was recrystallized from ethanol to get purified product.

Method B: A mixture of substituted aromatic aldehydes (1 mmol), 2-naphthol (1 mmol) and substituted amides (1.2 mmol) in the presence of effective amount of *p*-nitrobenzoic acid (30 mol %, 0.3 mmol) were taken in 100 mL conical flask and heated in an oil bath at 110-120 °C for the appropriate time with occasionally stirring. The progress of the reaction was monitored by TLC. After completion of reaction, mass was cooled to room temperature, washed with water and filtered. The solid residue was purified by recrystallization from EtOH to get purified product.

Method C: A mixture of substituted aromatic aldehydes (1 mmol), 2-naphthol (1 mmol) and substituted amides (1.2 mmol) in the presence of effective amount of *p*-nitrobenzoic acid (30 mol %, 0.3 mmol) were crushed in mortar and pestle to a fine powder and transferred into a china dish and heated on hot plate at 110-120 °C for appropriate time with occasional stirring. After completion of the reaction, water (10 mL) was added, filtered and then residue recrystallized from ethyl alcohol to get pure product.

All the products were identified by their ^{1}H NMR, IR and CHN data and compared with literature reports.

Spectral data for 1-amidoalkyl-2-naphthols

N-[(2-Hydroxynaphthalen-1-yl)-phenyl-methyl)]-acetamide (4a): IR(KBr, ν_{max}, cm^{-1}): 3441 (O-H, Ar, str), 3177 (N-H 2 *sec*-amide, str), 3057 (C-H, Ar, str), 1694 (>C=O, amide, str), 1555-1462 (C=C, Ar, str), 1243-1095 (C-N/C-O, str), 770 (C-H, Ar, out of plane , bend), 746 (N-H, out of plane, bend); ^{1}H NMR (400 MHz, DMSO-d_6): δ 9.85 (s, 1H, -CONH), 8.27 (d, *J* = 12 Hz, Ar-OH) 7.97 (t, *J* = 8 Hz, 1H, Ar-H), 7.75 (d, *J* = 8 Hz, 1H, Ar-H), 7.69 (d, *J* = 8 Hz, 1H, Ar-H), 7.40-7.12 (m, 9H, Ar-H), 2.04 (s, 3H, -$COCH_3$); Anal. calcd. (%) for $C_{19}H_{17}NO_2$: C 78.33, H 5.87, N 4.81; Found (%): C 77.47, H 5.79, N 4.76.

N-[(4-Chlorophenyl)-(2-hydroxynaphthalen-1-yl)-methyl)]urea (4d): IR (KBr, ν_{max}, cm^{-1}): 3320 (O-H, Ar, str), 3179 (N-H 2 *sec*-amide, str), 3022 (C-H, Ar, str), 1694 (>C=O, amide, str), 1592-1431 (C=C, Ar, str), 1402-1107 (C-N/C-O, str), 803 (C-H, Ar, out of plane, bend), 830 (C-Cl, str), 744 (N-H, out of plane, bend); ^{1}H NMR (400 MHz, DMSO-d_6): δ 8.75 (s,1H, -CONH), 7.88-7.83 (m, 4H, Ar-H), 7.40-7.27 (m, 7H, Ar-H), 7.59 (d, *J* = 8 Hz, Ar-OH), 6.09 (bs, 2H, -NH_2); Anal. calcd. (%) for $C_{18}H_{15}N_2O_2Cl$: C 66.16, H 4.63, N 8.57; Found (%): C 66.20, H 4.57, N 8.63.

N-[(4-Dimethylaminophenyl)-(2-hydroxynaphthalen-1-yl)-methyl)]acetamide (4h): IR (KBr, ν_{max}, cm^{-1}): 3372 (O-H, Ar, str), 3248 (N-H 2 *sec*-amide, str), 3084 (C-H, Ar, str), 2924 (C-H, alkane, str), 1652 (>C=O, amide, str), 1581-1397 (C=C, Ar, str), 1372-1031 (C-N/C-O, str), 827 (C-H, Ar, out of plane, bend), 698 (N-H, out of plane, bend); ^{1}H NMR (400 MHz, DMSO-d_6): δ 9.68 (s, 1H, -CONH), 7.90 (d, *J* = 8Hz, Ar-OH), 7.89-7.06 (m, 10H, Ar-H), 6.73 (s, 1H, -CHNH), 3.10 (s, 6H, -$N(CH_3)_2$), 2.13 (s, 3H, -$COCH_3$); Anal. calcd. (%) for $C_{21}H_{22}N_2O_2$: C 75.44, H 6.58, N 8.38; Found (%): C 75.76, H 6.84, N 8.36.

N-[(4-Methyphenyl)-(2-hydroxy-naphthalen-1-yl)-methyl)]benzamide (4k): IR (KBr, ν_{max}, cm^{-1}): 3414 (N-H 2 *sec*-amide, str), 3008 (O-H, Ar, str overlapping with C-H, Ar, str), 2824 (C-H, alkane, str), 1630 (C=O, amide, str), 1529-1483 (C=C, Ar, str), 1346-1248 (C-N/C-O, str), 817 (C-H, Ar, out of plane, bend), 711 (N-H, out of plane, bend); ^{1}H NMR (400 MHz, DMSO-d_6): δ 10.20 (s, 1H, -CONH), 8.93 (br d, *J* = 8Hz,1H Ar-OH), 7.84-7.70 (m, 4H, Ar-H), 7.51-7.16 (m, 9H, Ar-H), 7.02 (d, *J* = 8Hz, 2H, Ar-H), 2.23 (s, 3H, -CH_3);

Anal. calcd. (%) for $C_{25}H_{21}NO_2$: C 81.72, H 5.76, N 3.81; Found (%): C 81.63, H 5.68, N 3.88.

N-[(3-Nitrophenyl)-(2-hydroxynaphthalen-1-yl)-methyl)]urea (4l): IR (KBr, ν_{max}, cm^{-1}): 3636 (O-H, Ar, str), 3399-3316 (N-H 2 *sec*-amide, str), 3183-3076 (C-H, Ar, str), 1640 (>C=O, amide, str), 1481-1436 (C=C, Ar, str), 1346-1068 (C-N/C-O, str), 1528 (N-O, str), 810 (C-H, Ar, out of plane, bend), 744 (N-H, out of plane, bend); ^{1}H NMR (400 MHz, DMSO-d_6): δ 8.93 (s, 1H, -CONH), 8.64 (d, J = 8Hz, Ar-OH), 8.11-7.60 (m, 7H, Ar-H), 7.59-7.23 (m, 3H, Ar-H), 6.88 (s, 2H, -NH_2), 6.36 (s, 1H, -CHNH); Anal. calcd. (%) for $C_{18}H_{15}N_3O_4$: C 64.09, H 4.48, N 12.46; Found (%): C 64.16, H 4.56, N 12.38.

N-[(2,5-Dimethoxyphenyl)-(2-hydroxynaphthalen-1-yl)-methyl)]acetamide (4o): IR (KBr, ν_{max}, cm^{-1}): 3367 (O-H, Ar, str overlapping with N-H 2 *sec*-amide, str), 3056 (C-H, Ar, str), 2927-2832 (C-H, alkane, str), 1640 (>C=O, amide, str), 1595-1408 (C=C, Ar, str), 1235-1043 (C-N/C-O, str), 814 (C-H, Ar, out of plane, bend), 748 (N-H, out of plane, bend); ^{1}H NMR (400 MHz, DMSO-d_6): δ 9.68 (s, 1H, -CONH), 8.45 (d, *J* = 8Hz, 1H), -7.88-7.04 (m, 9H, Ar-H), 6.45 (s, 1H, -CHNH), 3.62 (s, 3H, -OCH_3), 3.45 (s, 3H, -OCH3), 1.87 (s, 3H, -$COCH_3$); Anal. calcd. (%) for $C_{21}H_{21}NO_4$: C 71.78, H 6.02, N 3.99; Found (%): C 71.50, H 6.50, N 3.90.

N-[(4-Chloro-phenyl)-(2-hydroxynaphthalen-1-yl)-methyl)]nicotinamide (4p): IR (KBr, ν_{max}, cm^{-1}): 3340 (N-H, 2 *sec*-amide, str), 3200 (O-H, Ar, str), 3057-3021(C-H, Ar, str), 1654 (>C=O, amide, str), 1596-1464 (C=C, Ar, str), 1371-1032 (C-N/C-O, str), 883-856 (C-H, Ar, out of plane, bend), 738 (C-Cl, str), 716 (N-H, out of plane, bend); ^{1}H NMR (400 MHz, DMSO-d_6): δ 8.55 (d, *J* = 8Hz, Ar-OH), 7.89-7.85 (m, 4H, Ar-H), 7.62-7.13 (m, 10H, Ar-H), 6.66 (s, 1H, -CHNH); Anal. calcd. (%) for $C_{21}H_{21}NO_4$: C 71.78, H 4.37, N 7.20; Found (%): C 71.10, H 4.51, N 7.95.

RESULTS AND DISCUSSION

In order to optimize the quantity of *p*-nitrobenzoic acid, we carried out the synthesis of N-[4-chloro-phenyl-(2-hydroxynapthalene-1-yl)-methyl] acetamide by using 2-naphthol, 4-chloro benzaldehyde and acetamide in the ratio (1:1:1.2 mmol) with different quantities of *p*-nitrobenzoic acid at same temperature and time under solvent free conditions. It was found that the best result was obtained when the reaction was carried in the presence of 0.3 mmol (30 mol %) of *p*-nitrobenzoic acid (Table-1, Entry-3). The fewer amounts gave a low yield even after long reaction time and the more amounts could not cause the obvious increase for the yield of product. Therefore, 0.3 mmol of catalyst was chosen as the optimal quantity of *p*-nitrobenzoic acid.

Slight excess of amide was found to be advantageous. Hence, molar ratio of aromatic aldehyde to amide was kept to be (1:1.2 mmol). Encouraged by these results, a wide variety of aromatic aldehydes and amides were treated with β-naphthol using *p*-nitrobenzoic acid under the optimized conditions to afford the corresponding 1-amidoalkyl-2-naphthols (Table-2) in good to excellent yields at 110-120 °C without formation of any side products such as dibenzoxanthenes, which are normally observed under the influence of strong acids. It is important to note that the synthesis of 1-amidoalkyl-2-naphthols could not be achieved in the absence of catalyst.

TABLE-1
OPTIMIZATION STUDY OF *p*-NITROBENZOIC ACID (PNBA) FOR THE SYNTHESIS OF 1-AMIDOALKYL-2-NAPHTHOLS

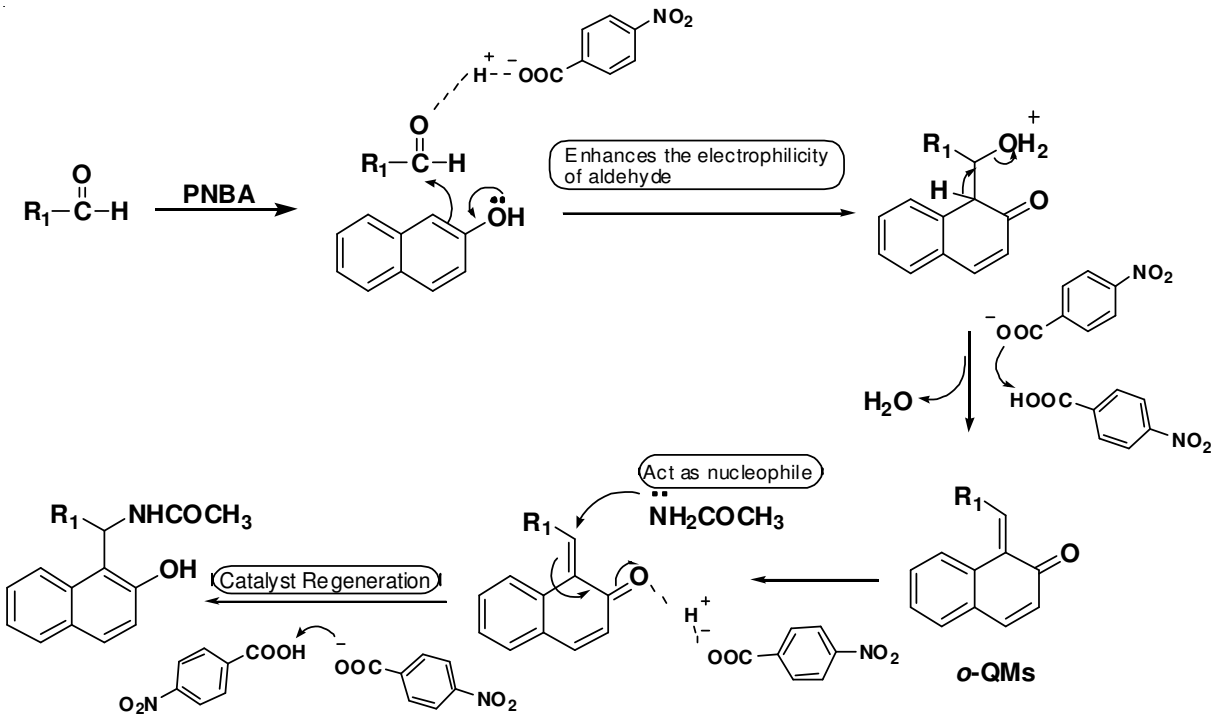
Entry	PNBA (mmol)	Method A Time (min)/ yield (%) (Microwave)	Method B Time (min)/ yield (%) (Oil-bath)	Method C time (min)/ yield (%) (Hot-plate)
1.	0.1	10/75	15/71	20/56
2.	0.2	10/82	15/80	20/62
3.	0.3	10/88	15/85	20/66
4.	0.4	10/88	15/86	20/66

TABLE-2
p-NITROBENZOIC ACID (PNBA) (0.3 mmol, 30 mol %) CATALYZED SYNTHESIS OF 1-AMIDOALKYL-2-NAPHTHOLS

Entry	R	R_1	Product[a]	Time (min)/Yield (%) Method A	Method B	Method C	m.p. (°C) (Lit.)
1	H	CH_3	**4a**	10/88	15/85	20/66	242-245 (245-246)[22]
2	H	NH_2	**4b**	11/85	16/86	19/65	171-172 (170-173)[29b]
3	H	C_6H_5	**4c**	9/83	16/85	19/67	234-237 (234-236)[15]
4	4-Cl	NH_2	**4d**	8/90	16/88	25/72	165-167 (166-168)[29b]
5	4-Cl	CH_3	**4e**	9/92	14/88	23/74	222-224 (223-225)[22]
6	4-Cl	C_6H_5	**4f**	8/91	13/87	21/74	173-175 (175-177)[29b]
7	4-$N(CH_3)_2$	NH_2	**4g**	11/87	20/85	25/70	205-207
8	4-$N(CH_3)_2$	CH_3	**4h**	14/84	26/82	28/64	122-124 (123-125)[24]
9	4-CH_3	NH_2	**4i**	12/82	25/80	32/62	120-122 (118-120)[17]
10	4-CH_3	CH_3	**4j**	14/83	22/82	30/61	222-224 (222-223)[24]
11	4-CH_3	C_6H_5	**4k**	12/82	24/79	23/60	1190-191 (190-192)[29d]
12	3-NO_2	NH_2	**4l**	8/92	13/89	20/72	192-194 (194-196)
13	3-NO_2	CH_3	**4m**	9/92	16/88	22/65	242-243 (241-242)[22]
14	3-NO_2	C_6H_5	**4n**	9/89	15/81	21/67	213-215 (214-216)[29b]
15	2,5-$(OCH_3)_2$	CH_3	**4o**	12/82	20/80	20/63	252-253 (251-253)[22]
16	4-Cl	CH_3	**4p**	8/92	12/90	13/68	209-211

[a]All known products have been reported previously in the literature and were characterized by comparison of IR and NMR spectra with authentic samples.

As reported in literatures, a mechanism for the synthesis of 1-amidoalkyl-2-naphthols is shown in **Scheme-II**. A role of *p*-nitrobenzoic acid has been proposed to activate the aldehyde by binding H^+ with the oxygen atom which ultimately enhances the electrophilicity of the aldehyde and leads to reduction in reaction time. The condensation of 2-naphthol with the activated aldehyde give *ortho*-quinone (*o*-QMs) as a highly reactive and ephemeral intermediate. The same *ortho*-quinone methides, generated *in situ*, have been reacted with amide to form 1-amidoalkyl-2-naphthol derivatives. Thus, the 2-naphthol acts as Michael acceptors and araldehydes as nucleophile resulting in Michael adduct under the influence



Scheme-II: Mechanism of action of *p*-nitrobenzoic acid promoted synthesis of 1-amidoalkyl-2-naphthols

of *p*-nitrobenzoic acid. The electron withdrawing groups (EWD) substituted on benzaldehyde in *o*-QM intermediate increase the rate of 1, 4-nucleophilic addition reaction because of alkene LUMO is at lower energy in the neighboring withdrawing groups than electron donating groups (EDG). Hence benzene ring with electron withdrawing groups gave better yield as compared to electron donating groups.

Conclusion

In conclusion, we have found *p*-nitrobenzoic acid as the best promoter for the synthesis of 1-amidoalkyl-2-naphthols under solvent-free conditions. It is noted that the solvent-free microwave irradiation can promote reactive pathways in very convenient way from green chemical point of view compared to the direct hot plate heating method. These advantages makes this protocol an attractive and user friendly alternative for the synthesis of 1-amidoalkyl-2-naphthols.

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REFERENCES

1. A.Y. Shen, C.T. Tsai and C.L. Chen, *Eur. J. Med. Chem.*, **34**, 877 (1999).
2. I. Szatmari and F. Fulop, *Curr Org Synth*, **1**, 155 (2004).
3. M. Damodiran, N. Panneer Selvam and P.T. Perumal, *Tetrahedron Lett.*, **50**, 5474 (2009).
4. A.H. Kategaonkar, S.S. Sonar, K.F. Shelke, B.B. Shingate and M.S. Shingare, *Org. Commun.*, **3**, 1 (2010).
5. Y. Kusakabe, J. Nagatsu, M. Shibuya, O. Kawaguchi, C. Hirose and S. Shirato, *J. Antibiot.*, **25**, 44 (1972).
6. J.B. Chylinska, T. Urbanski and M. Mordarski, *J. Med. Chem.*, **6**, 484 (1963).
7. J.L. Peglion, J. Vian, B. Goument, N. Despaux, V. Audinot and M.J. Millan, *Bioorg. Med. Chem. Lett.*, **7**, 881 (1997).
8. H. Ren, S. Grady, D. Gamenara, H. Heinzen, P. Moyna, S. Croft, H. Kendrick, V. Yardley and G. Moyna, *Bioorg. Med. Chem. Lett.*, **11**, 1851 (2001).
9. F. Benedini, G. Bertolini, R. Cereda, G. Doná, G. Gromo, S. Levi, J. Mizrahi and A. Sala, *J. Med. Chem.*, **38**, 130 (1995).
10. R.D. Clark, J.M. Caroon, A.F. Kluge, D.B. Repke, A.P. Roszkowski, A.M. Strosberg, S. Baker, S.M. Bitter and M.D. Okada, *J. Med. Chem.*, **26**, 657 (1983).
11. J.B. Chylinska, M. Janowiec and T. Urbanski, *Br. J. Pharmacol.*, **43**, 649 (1971).
12. S. Knapp, *Chem. Rev.*, **95**, 1859 (1995).
13. S.M. Vrouenraets, W.F. Wit, J. Tongeren and J.M. Lange, *Expert Opin. Pharmacother.*, **8**, 851 (2007).
14. M. Anary-Abbasinejad, A. Hassanabadi, M. Kamali-Gharamaleki, A. Saidipoor, H. Anaraki-Ardakani, *J. Chem. Res.*, 644 (2007).
15. H.R. Shaterian and H. Yarahmadi, *Synlett*, 916 (2006).
16. H.R. Shaterian and H. Yarahmadi, *ARKIVOC*, 105 (2007).
17. H.R. Shaterian, H. Yarahmadi and M. Ghashang, *Bioorg. Med. Chem. Lett.*, **18**, 788 (2008).
18. W.K. Su, W.Y. Tang, J.J. Li, *J. Chem. Res.*, 123 (2008).
19. B. Das, K. Laxminarayana, B. Ravikanth and R. Rao, *J. Mol. Catal. Chem.*, **261**, 180 (2007).
20. L. Nagarapu, M. Baseeruddin, S. Apuri and S. Kantevari, *Catal. Commun.*, **8**, 1729 (2007).
21. S.B. Patil, P.R. Singh, M.P. Surpur and S.D. Samant, *Synth. Commun.*, **37**, 1659 (2007).
22. H.R. Shaterian, H. Yarahmadi and M. Ghashang, *Tetrahedron*, **64**, 1263 (2008).
23. B. Das, D.N. Kumar, K. Laxminarayana and B. Ravikanth, *Helv. Chim. Acta*, **90**, 1330 (2007).
24. H.R. Shaterian and H. Yarahmadi, *Tetrahedron Lett.*, **49**, 1297 (2008).
25. S. Kantevari, S.V.N. Vuppalapati and L. Nagarapu, *Catal. Commun.*, **8**, 1857 (2007).
26. G. Srihari, M. Nagaraju and M.M. Murthy, *Helv. Chim. Acta*, **90**, 1497 (2007).

27. (a) S.B. Patil, P.R. Singh, M.P. Surpur and S.D. Samant, *Ultrason. Sonochem*., **14**, 515 (2007); (b) R.R. Nagawade and D.B. Shinde, *Chin. J. Chem*., **25**, 1710 (2007).
28. R. Ghorbani-Vaghei and S.M. Malaekehpour, *Cent. Eur. J. Chem*., **8**, 1086 (2010).
29. (a) A. Kumar, M.S. Rao, I. Ahmad and B. Khungar, *Can. J. Chem*., **87**, 714 (2009); (b) H. Khabazzadeh, K. Saidi and N. Seyedi, *J. Chem. Sci*., **121**, 429 (2009); (c) A.R. Hajipour, F. Ghayeb, N. Sheikhan and A.E. Ruoho, *Tetrahedron Lett*., **50**, 5649 (2009); (d) S.B. Sapkal, K.F. Shelke, B.R. Madje, B.B. Shingate and M.S. Shingare, *Bull. Korean Chem. Soc*., **30**, 2887 (2009); (e) T.T. She, Z.L. Liu and K. Gong, *Chinese J. Appl. Chem*., **27**, 778 (2010).
30. (a) P.T. Anastas and J.C. Warner, Green Chemistry: Theory and Practice; Oxford University Press, Oxford, UK (1998); (b) P.T. Anastas and T. Williamson, Green Chemistry, Frontiers in Benign Chemical Synthesis and Process; Oxford University Press, Oxford, UK (1998).
31. (a) R. Duvedi and R.K. Singh, *Asian J. Chem*., **24**, 5665 (2012); (b) S. Sharma, D.N. Prasad and R.K. Singh, *J. Chem. Pharm. Res*, **3**, 382 (2011); (c) A. Sandhar, D.N. Prasad and R.K. Singh, *Indian J. Heterocycl. Chem*., **21**, 369 (2012); (c) A. Sandhar, D.N. Prasad, A. Kapoor and R.K. Singh, *Curr. Res. Chem.*, **4**, 68 (2012); (d) S. Malik, S. Sharma and R.K. Singh, *Asian J. Chem*., **24**, 5669 (2012); (e) P. Kaur, H. Sharma, R. Rana, D.N. Prasad and R.K. Singh, *Asian J. Chem*, **24**, 5649 (2012); (f) A. Sandhar and R.K. Singh, *Asian J. Chem.*, **24**, 5643 (2012); (g) A. Sandhar and R.K. Singh, *Chem. Sci. Trans*., **2(S1)**, 176 (2012); (h) R.K. Singh, B. Singh, R. Duvedi and S. Kumar, *Res. Chem. Intermed*., (2014); doi:10.1007/s11164-013-1513-5.
32. R. Varala, R. Enugala and S.R. Adapa, *J. Braz. Chem. Soc*., **18**, 291 (2007).