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SYNTHESIS OF ERLENMEYER-PLÖCHL AZLACTONES PROMOTED BY 5-SULFOSALICYLIC ACID

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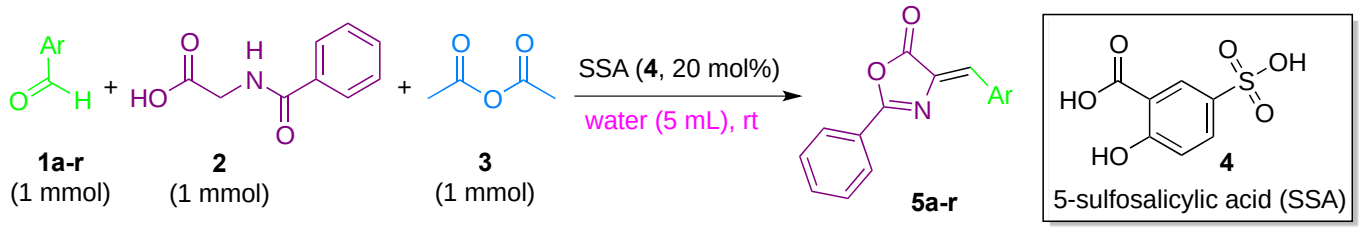
Abstract – 5-Sulfosalicylic acid was found as an efficient catalyst in the synthesis of 4-arylidene-2-phenyl-5(4*H*)-oxazolones by condensation and cyclodehydration of aromatic aldehydes with hippuric acid and acetic anhydride at room temperature. The catalyst was easily recycled from the filtrate via evaporation of solvent and reused. This process is green, simple to handle, commercially available starting materials and catalyst, mild conditions, as well as it does not require to hazardous organic solvents, heating, and microwave and ultrasound irradiations.

Nitrogen- and oxygen-containing five-membered heterocycles are important compounds that can be found in a wide range of drug-likes and biologically relevant molecules. Some examples of these five-membered heterocycles are 4-arylidene-2-phenyl-5(4*H*)-oxazolones (Erlenmeyer-Plöchl azlactones or oxazol-5(4*H*)-ones), which synthesis of them was started in 1893 by Erlenmeyer for the first time.[1] Some azlactones and their derivatives exhibited biological activities including, antimicrobial,[2] antiproliferative,[3] tyrosinase inhibitory,[4] antitubercular,[5] antioxidant,[6] anti-inflammatory,[7] anti-HIV,[8] anticonvulsant,[9] and monoacylglycerol lipase (MAGL) inhibitory.[10] These important heterocyclic compounds have been found to be generally useful intermediates for the synthesis imidazolones,[11] *N*-benzoylamino-3-arylacrylates,[12] oxazole-4-carboxamides,[13] α-amino acids,[14-15] optically active spiro-fused cyclohexanone/5-oxazolone derivatives,[16] peptides,[17] and some heterocyclic compounds.[18] Thus, the synthesis of this heterocyclic unit is of particular interest. Erlenmeyer-Plöchl azlactones can be synthesized through two steps from readily available *N*-substituted glycines and aldehydes in acetic anhydride, using anhydrous sodium acetate as a basic catalyst.[19] Various reagents and reaction conditions have been previously used for the cyclization of aldehydes and hippuric acid in presence of acetic anhydride to form azlactones such as $POCl_3$,[20] Al_2O_3,[21] $Bi(OAc)_3$,[22] $Bi(OTf)_3$,[23] silica-supported heteropolyacids,[24] $Yb(OTf)_3$,[25] $Ca(OAc)_2$,[26] supported KF,[27] anhydrous $ZnCl_2$,[28] Fe_2O_3,[29] ZnO,[30]

Al_2O_3-H_3BO_3,[31] 2,4,6-trichloro-1,3,5-triazine, catalytic triphenylphosphine, and sodium carbonate,[32] MgO/Al_2O_3,[33] cellulose supported ionic liquid phase,[34] 2-chloro-4,6-dimethoxy-1,3,5-triazine/*N*-methylmorpholine,[35] ionic liquid [Et_3NH][HSO_4],[36] 2-aminopyridine, supported on nano-sphere SiO_2,[37] Hünig's base,[38] nano-TiO_2,[39] triphenylphosphine (PPh_3),[40] zeolite NaY,[41] and *N,N*-dimethylacetamide (DMAC)/*N,N*-dicyclohexylcarbodiimide (DCC) under microwave irradiation.[42] The organic transformations for synthesis of isoxazol-5(4*H*)-ones,[43] pyran annulated heterocycles,[44] and aminopyrazoles[45] are of particular interest to our laboratory and this report describes synthesis of some derivatives of oxazol-5(4*H*)-ones in presence of 5-sulfosalicylic acid (SSA, **4**) under green conditions (Scheme 1).



1a-r (1 mmol) **2** (1 mmol) **3** (1 mmol) **5a-r** **4**

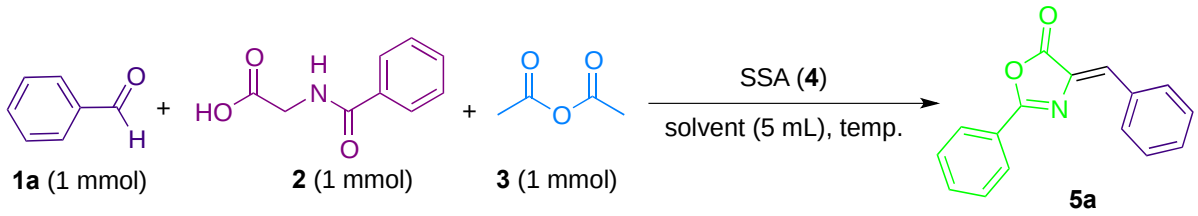
Ar: C_6H_5 (**1a**), 4-Me-C_6H_4 (**1b**), 4-HO-C_6H_4 (**1c**), 3-HO-C_6H_4 (**1d**), 2-HO-C_6H_4 (**1e**), 4-MeO-C_6H_4 (**1f**), 3-MeO-C_6H_4 (**1g**), 4-Me_2N-C_6H_4 (**1h**), 4-HO-3-MeO-C_6H_3 (**1i**), 2,4-$(MeO)_2$-C_6H_3 (**1j**), 2-Cl-C_6H_4 (**1k**), 4-Cl-C_6H_4 (**1l**), 2,4-diCl-C_6H_4 (**1m**), 4-NO_2-C_6H_4 (**1n**), 3-NO_2-C_6H_4 (**1o**), 2-NO_2-C_6H_4 (**1p**), 2-thienyl (**1q**), 2-furyl (**1r**)

Scheme 1. SSA-catalyzed synthesis of 4-arylidene-2-phenyl-5(4*H*)-oxazolones (**5a-r**)

Initially, to find the best reaction conditions for the synthesis of azlactones, the reaction of benzaldehyde (**1a**), hippuric acid (**2**) and acetic anhydride (**3**) was chosen as a model. The results are shown in Table 1. When the reaction was carried out without any catalyst, the formation of the product was not observed. Considering the fact that we had already used the SSA (**4**) in the synthesis of 1-amidoalkyl-2-naphthols and 3,4-disubstituted isoxazol-5(4*H*)-ones,[46] we decided to use it as a catalyst in the synthesis of azlactones. In the direction of optimize the catalyst loading, the model reaction was conducted in 5 mol%, 10 mol%, 15 mol%, and 20 mol% catalyst loadings under aqueous media at rt (Table 1, entries 2-5). The reaction proceeded sluggishly with SSA (**4**) at 5 mol% loading but, at 20 mol% loading, furnished 90% yield of corresponding product (**5a**) for 30 min (Table 1, entry 5). 25 mol% Catalyst loading furnished 91% yield of **5a** product. Subsequently 20 mol% of catalyst was selected as the optimum loading. Several other solvents such as EtOH, EtOAc, $CHCl_3$, MeOH, Ph-Me, and a mixed of EtOH-H_2O was screened and gave trace to good yields of product (**5a**) (Table 1, entries 7-12). The reaction was also conducted under solvent-free conditions and gave 40% isolated yield for 120 min (Table 1, entry 13). Furthermore, the reaction temperature was investigated. The implementation of the model using the optimal amount of catalyst (20 mol%) at temperatures higher than rt, the product was formed in 84-90% isolated yields (Table 1, entries 14-16). Implementation of model reaction using 5 mol% and 10 mol% SSA at 60 °C leads to 70 and 79%

isolated yields, respectively (Table 1, entries 17, 18). Finally, 20 mol% of SSA as the catalyst, water as solvent, and rt as reaction temperature were selected as optimized conditions (Table 1, entry 5).

Table 1. Optimization of the reaction conditions for the synthesis of 4-benzylidene-2-phenyloxazol-5(4*H*)-one (**5a**)

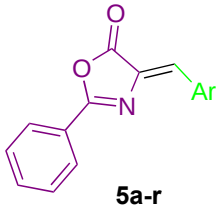


Entry	Catalyst loading/mol%	Solvent	Temp/°C	Time/min	Isolated yields/%
1	-	H_2O	rt	120	trace
2	5	H_2O	rt	60	65
3	10	H_2O	rt	50	68
4	15	H_2O	rt	40	75
5	**20**	**H_2O**	**rt**	**30**	**90**
6	25	H_2O	rt	27	91
7	20	EtOH	rt	120	trace
8	20	EtOAc	rt	120	78
9	20	$CHCl_3$	rt	120	85
10	20	MeOH	rt	120	trace
11	20	Ph-Me	rt	70	96
12	20	EtOH-H_2O (1:1)	rt	120	15
13	20	no solvent	rt	120	40
14	20	H_2O	40	60	84
15	20	H_2O	50	45	87
16	20	H_2O	60	30	90
17	5	H_2O	60	40	70
18	10	H_2O	60	40	79

Optimized conditions shown in bold.

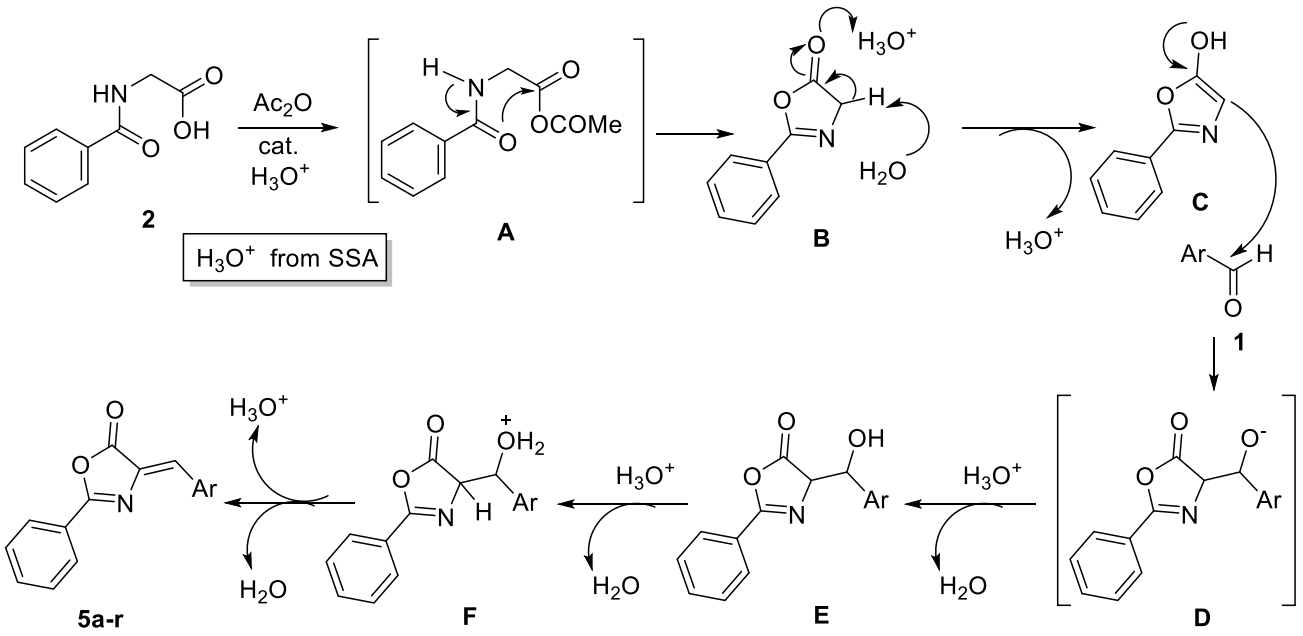
After optimization of conditions for synthesis of Erlenmeyer products, our study was focused on different substituted benzaldehydes containing both electron-withdrawing and electron-donating groups and heteroaromatic aldehydes such as thiophene-2-carboxaldehyde and furan-2-carboxaldehyde towards synthesis of diversity azlactones. The results showed that the corresponding products (**5a-r**) were synthesized within 82-98% isolated yields in 28-53 min. In this study, the catalyst was recovered and reused for synthesis of **5a**. After completion of the reaction, the solid product was filtered and washed with water. The catalyst was recovered by simple evaporation from the filtrate, dried in the air and reused. The remained SSA showed good catalytic activity as the product **5a** obtained in 90%, 88%, 84%, and 70% isolated yields in four successive runs, respectively.

Table 2. SSA-catalyzed synthesis of Erlenmeyer-Plöchl azlactones (**5a-r**) based on the optimized conditions according to the Scheme 1

Structure of Erlenmeyer-Plöchl azlactones  **5a-r** Ar	Time (min)	Isolated yields (%)	Mp (°C)	
			Found	Reported (Melting points are matched with data from the literature[33,34,39,42,47])
C_6H_5 (**5a**)	30	90	171-173	169-170
4-Me-C_6H_4 (**5b**)	53	95	143-144	144-145
4-HO-C_6H_4 (**5c**)	40	97	171-173	172-173
3-HO-C_6H_4 (**5d**)	35	94	145-156	144-145
2-HO-C_6H_4 (**5e**)	40	96	138-139	138-139
4-MeO-C_6H_4 (**5f**)	34	86	154-156	153-154
3-MeO-C_6H_4 (**5g**)	42	88	102-103	101-102
4-$(Me)_2N$-C_6H_4 (**5h**)	28	89	212-214	213-214
4-HO-3-MeO-C_6H_3 (**5i**)	40	90	159-191	157-158
2,4-diMeO-C_6H_3 (**5j**)	45	87	154-156	151-152
2-Cl-C_6H_4 (**5k**)	37	85	162-163	163-164
4-Cl-C_6H_4 (**5l**)	32	86	188-190	198
2,4-diCl-C_6H_3 (**5m**)	46	82	157-159	159-160
4-NO_2-C_6H_4 (**5n**)	42	94	240-241	238-240
3-NO_2-C_6H_4 (**5o**)	32	98	169-171	167-168
2-NO_2-C_6H_4 (**5p**)	45	94	164-165	158-159
thiophen-2-yl (**5q**)	40	90	172-174	173-175
furan-2-yl (**5r**)	45	86	165-167	169-170

A plausible reaction mechanism for this transformation is outlined in Scheme 2. The reaction occurs through initial formation of intermediate **A** by the reaction between the activated hippuric acid (**2**) using SSA catalyst and acetic anhydride (**3**) followed by cyclization to 2-phenyloxazol-5(4*H*)-one (**B**). Enol (**C**) is formed via protonation using acid catalyst and deprotonation by conjugate base of the SSA and this enol form is then condensed with aldehyde (**1**) to form intermediate **E**. In the last step, protonation and dehydration leads to the formation of the corresponding heterocyclic products (**5a-r**).

In conclusion, this work describes an efficient, mild and SSA-catalyzed condensation and cyclodehydration reaction of hippuric acid, a wide range of aldehydes, and acetic anhydride to form 4-arylidene-2-phenyl-5(4*H*)-oxazolone derivatives in good to excellent yields. This method is easy to implement and was successfully carried out with satisfactory results in a green medium.



Scheme 2. Proposed reaction mechanism

EXPERIMENTAL

All chemicals, unless otherwise specified, were purchased from commercial sources and were used without further purification, with the exception of liquid aldehydes, which were distilled before using. Melting points were measured on a Buchi 510 melting point apparatus and are uncorrected. ^{1}H NMR and ^{13}C NMR spectra were recorded on a Bruker AVANCE DRX spectrometer. FT-IR spectra were recorded on a Perkin Elmer RXI spectrometer. The development of reactions was monitored by thin layer chromatography (TLC) analysis on Merck pre-coated silica gel 60 F_{254} aluminum sheets, visualized by UV light. Elemental microanalyses were performed on Elementar Vario EL III analyzer at Damghan University, Damghan, Iran. All of the targeted products are reported in the literature and are characterized by comparison of their spectral and physical data on the basis of literature descriptions.

General procedure for the synthesis of 4-arylidene-2-phenyl-5(4*H*)-oxazolones (5a-r) catalyzed by SSA. The appropriate aldehyde (**1**, 1 mmol), hippuric acid (**2**, 1 mmol), acetic anhydride (**3**, 1 mmol), water (5 mL), and SSA as a catalyst (20 mol%) was stirred at rt for the appropriate time (Table 2). After completion of the reaction as indicated by TLC (hexane:EtOAc; 70:30) analysis, the reaction mixture allowed to stand for 5 h at rt. The resulting precipitated products were filtered off, washed with ice water and dried at room temperature. The crude products were purified by recrystallization from absolute EtOH, if required. The filtrate containing the catalyst was used as such for exploring the reusability of the catalyst. Spectral data for **5a** and **5n** were as follows:

4-Benzylidene-2-phenyloxazol-5(4*H*)-one (5a). ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.27 (s, 1H), 7.43-7.61 (m, 6H), 8.18-8.24 (m, 4H) ppm; ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 113.2, 125.7, 128.2, 128.5, 128.7,

129.2, 129.8, 131.2, 131.7, 135.5, 160.9, 166.0; Anal. Calcd for $C_{16}H_{11}NO_2$: C, 77.10; H, 4.45; N, 5.62. Found: C, 77.07; H, 4.41; N, 5.66.

4-(4-Nitrobenzylidene)-2-phenyloxazol-5(4*H*)-one (5n). ^{1}H NMR ($CDCl_3$, 400 MHz): δ = 7.28 (s, 1H), 7.61 (dd, J = 4.7, 10.9 Hz, 2H), 7.71-7.73 (m, 1H), 8.24 (dd, J = 1.2, 8.6 Hz, 2H), 8.34-8.37 (m, 2H), 8.29-8.32 (m, 2H); ^{13}C NMR ($CDCl_3$, 100 MHz): δ = 112.8, 122.2, 127.5, 128.9, 129.3, 129.8, 131.2, 131.3, 142.0, 146.7, 160.5, 166.3; Anal. Calcd for $C_{16}H_{10}N_2O_4$: C, 65.31; H, 3.43; N, 9.52. Found: C, 65.27; H, 3.39; N, 9.48.

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REFERENCES

1. E. Erlenmeyer, *Annalen*, 1893, **275**, 1.
2. C. Voosala, P. S. Kilarua, and U. K. Dasaric, *J. Chin. Chem. Soc.*, 2016, **63**, 909; N. S. Thombare, N. Aggarwal, R. Kumar, and M. Gopal, *J. Environ. Sci. Health, Part B*, 2012, **47**, 326.
3. C. Granchi, F. Rizzolio, V. Bordoni, I. Caligiuri, C. Manera, M. Macchia, F. Minutolo, A. Martinelli, A. Giordano, and T. Tuccinardi, *J. Enzyme Inhib. Med. Chem.*, 2016, **31**, 137.
4. H. Hamidian, R. Tagizadeh, S. Fozooni, V. Abbasalipour, A. Taheri, and M. Namjou, *Bioorg. Med. Chem.*, 2013, **21**, 2088; H. Hamidian and S. Azizi, *Bioorg. Med. Chem.*, 2015, **23**, 7089; K. M. Khan, U. R. Mughal, M. T. H. Khan, Zia-Ullah, S. Perveen, and M. I. Choudhary, *Bioorg. Med. Chem.*, 2006, **14**, 6027.
5. K. P. Suhasini, P. K. Chintakindi, K. Chaguruswamya, and Y. L. N. Murthy, *J. Chin. Chem. Soc*., 2015, **62**, 455.
6. M. Parveen, A. Ali, S. Ahmed, A. M. Malla, M. Alam, P. S. P. Silva, M. R. Silva, and D. U. Lee, *Spectrochim. Acta A*, 2013, **104**, 538.
7. U. S. Goksen, N. G. Kelekci, O. Goktas, Y. Koysal, E. Kilic, S. Isik, G. Aktay, and M. Ozalp, *Bioorg. Med. Chem.*, 2007, **15**, 5738.
8. M. Witvrouw, C. Pannecouque, E. D. Clercq, E. F. Alvarez, and J. L. Marco, *Arch. Pharm. Med. Chem.*, 1999, **352**, 163; I. L. Pinto, A. West, C. M. Debouckm, A. G. DiLella, J. G. Gorniak, K. C. O'Donnell, D. J. O'Shannessy, A. Patel, and R. L. Jarvest, *Bioorg. Med. Chem. Lett.*, 1996, **6**, 2467.
9. H. M. F. Madkour, *Chem. Pap*., 2002, **56**, 313.
10. C. Granchi, F. Rizzolio, V. Bordoni, I. Caligiuri, C. Manera, M. Macchia, F. Minutolo, A. Martinelli, A. Giordano, and T. Tuccinard, *J. Enzyme Inhibit. Med. Chem.*, 2016, **31**, 137.
11. V. K. Ahluwalia, M. K. Sharma, and R. Sharma, *Org. Prep. Proced. Int.*, 1992, **24**, 698; N. S. H. N.

Moorthy, V. Saxena, C. Karthikeyan, and P. Trivedi, *J. Enzyme Inhibit. Med. Chem.*, 2012, **27**, 201; K. M. Khan, U. R. Mughal, N. Ambreen1, Samreen, S. Perveen, and M. I. Choudhary, *J. Enzyme Inhibit. Med. Chem.*, 2010, **25**, 29; M. Muselli, L. Colombeau, J. Hedouin, C. Hoarau, and L. Bischoff, *Synlett*, 2016, **27**, 2819; F. Chavez, C. Pavy, T. Williamson, and T. Cleary, *Synth. Commun.*, 2012, **42**, 3321; A. K. Dhingra, B. Chopra, R. Dass, and S. K. Mittal, *Chin. Chem. Lett.*, 2016, **27**, 707; H. Deng and X. Zhu, *Mater. Chem. Front.*, 2017, DOI: 10.1039/C6QM00148C.

12. G. D. Roiban, T. Soler, M. Contel, I. Grosu, C. Cativiela, and E. P. Urriolabeitia, *Synth. Commun.*, 2012, **42**, 195.
13. V. M. Prokopenko, S. G. Pil'o, V. S. Brovarets, A. N. Vasilenko, and B. S. Drach, *Russ. J. Gen. Chem.*, 2010, **80**, 121.
14. R. B. Mosey, J. S. Fisk, and J. J. Tepe, *Tetrahedron: Asymmetry*, 2008, **19**, 2755.
15. A. R. Genady and H. Nakamura, *Org. Biomol. Chem.*, 2011, **9**, 7180.
16. M. Q. Zhou, J. Zuo, B. D. Cui, J. Q. Zhao, Y. You, M. Bai, Y. Z. Chen, X. M. Zhang, and W. C. Yuan, *Tetrahedron*, 2014, **70**, 5787.
17. M. Palcut, *J. Chem. Res.*, 2005, **2005**, 766; S. C. Khadse and V. A. Chatpalliwar, *Arab. J. Chem.*, 2017, **10**, S859.
18. H. M. F. Madkour, *Heterocycl. Commun.*, 2002, **8**, 501.
19. P. P. de Castro, A. G. Carpanez, and G. W. Amarante, *Chem. Eur. J.*, 2016, **22**, 10294.
20. A. R. Khosropour, M. M. Khodaei, and S. J. Hoseini Jomor, *J. Heterocycl. Chem.*, 2008, **45**, 683.
21. P. A. Conway, K. Devine, and F. Paradisi, *Tetrahedron*, 2009, **65**, 2935; S. Chandrasekhar and P. Karri, *Tetrahedron Lett.*, 2007, **48**, 758.
22. K. A. Monk, D. Sarapa, and R. S. Mohan, *Synth. Commun.*, 2000, **30**, 3167.
23. M. M. Khodaei, A. R. Khosropour, and S. J. Hoseini Jomor, *J. Chem. Res. Synop.*, 2003, **10**, 638.
24. G. Romanelli, J. C. Autino, P. Vazquez, L. Pizzio, M. Blanco, and C. Caceres, *Appl. Catal., A*, 2009, **352**, 208.
25. Y. Chuanming, Z. Baocheng, S. Weike, and X. Zhenyuan, *Synth. Commun.*, 2006, **36**, 3447.
26. S. Paul, P. Nanda, R. Gupta, and A. Loupys, *Tetrahedron Lett.*, 2004, **45**, 425.
27. F. M. Bautista, J. M. Campelo, A. Garcia, D. Luna, J. M. Marinas, and A. Romero, *J. Chem. Soc., Perkin Trans. 2*, 2002, 227.
28. P. S. Rao and R. V. Venkataratnam, *Indian J. Chem.*, 1994, **33B**, 984.
29. S. J. Ahmadi, S. Sadjadi, and M. Hosseinpour, *Ultrason. Sonochem.*, 2013, **20**, 408.
30. M. A. Pasha, V. P. Jayashankara, K. N. Venugopala, and G. K. Rao, *J. Pharmacol. Toxicol.*, 2007, **2**, 264.
31. J. Kashyap, A. B. Chetry, and P. J. Das, *Synth. Commun.*, 1998, **28**, 4187.

32. M. Pattarawarapan, S. Jaita, and W. Phakhodee, *Tetrahedron Lett.*, 2016, **57**, 3171.
33. N. Rostamizadeh, A. Khajeh-Amiri, and H. Moghanian, *Synth. React. Inorg., Met.-Org., Nano-Met. Chem.*, 2016, **46**, 631.
34. R. Kurane, S. Khanapure, D. Kale, R. Salunkhe, and G. Rashinkar, *RSC Adv.*, 2016, **6**, 44135.
35. V. Siddaiah, G. Mahaboob Basha, D. Sudhakar, R. Srinuvasarao, and Y. S. Kumar, *Synth. Commun.*, 2013, **43**, 2191.
36. M. Parveen, F. Ahmad, A. M. Malla, S. Azaz, M. R. Silva, and P. S. P. Silva, *RSC Adv.*, 2015, **5**, 52330.
37. A. Mobinikhaledi, H. Moghanian, and S. Pakdel, *Chin. Chem. Lett.*, 2015, **26**, 557.
38. T. Cleary, T. Rawalpally, N. Kennedy, and F. Chavez, *Tetrahedron Lett.*, 2010, **51**, 1533.
39. P. Anandgaonker, G. Kulkarni, S. Gaikwad, and A. Rajbhoj, *Chin. J. Catal.*, 2014, **35**, 196.
40. A. M. L. Punna Rao, A. S. Rao, M. S. Babu, and M. K. Rao, *J. Heterocycl. Chem.*, 2017, **54**, 429.
41. M. A. Bodaghifard, H. Moghanian, A. Mobinikhaledi, and F. Esmaeilzadeh, *Synth. React. Inorg., Met.-Org., Nano-Met. Chem.*, 2017, **47**, 845.
42. M. Kidwai and R. Kumar, *Org. Prep. Proced. Int.*, 1998, **30**, 451.
43. H. Kiyani and F. Ghorbani, *Heterocycl. Lett.*, 2013, **3**, 359; H. Kiyani and F. Ghorbani, *Res. Chem. Intermed.*, 2015, **41**, 2653; H. Kiyani and F. Ghorbani, *Elixir Org. Chem.*, 2013, **58A**, 14948; H. Kiyani and F. Ghorbani, *J. Saudi Chem. Soc.*, 2017, **21**, S112; H. Kiyani and F. Ghorbani, *Org. Chem. Indian J.*, 2013, **13**, 97; H. Kiyani and F. Ghorbani, *Open J. Org. Chem.*, 2013, **1**, 5; H. Kiyani and F. Ghorbani, *Heterocycl. Lett.*, 2013, **3**, 145; H. Kiyani, M. Jabbari, and A. Mosallanezhad, *Jordan J. Chem.*, 2014, **9**, 279; H. Kiyani, A. Kanaani, D. Ajloo, F. Ghorbani, and M. Vakili, *Res. Chem. Intermed.*, 2015, **41**, 7739; H. Kiyani and F. Ghorbani, *Res. Chem. Intermed.*, 2015, **41**, 7847; H. Kiyani and F. Ghorbani, *Res. Chem. Intermed.*, 2016, **42**, 7559.
44. H. Kiyani and F. Ghorbani, *Res. Chem. Intermed.*, 2015, **41**, 4031; H. Kiyani and F. Ghorbani *J. Saudi Chem. Soc.*, 2014, **18**, 689; H. Kiyani and F. Ghorbani, *Chem. Pap.*, 2014, **68**, 1104; H. Kiyani and F. Ghorbani, *Jordan J. Chem.*, 2014, **9**, 1; H. Kiyani and M. S. Jalali, *Heterocycles*, 2016, **92**, 75; H. Kiyani and M. S. Jalali, *Comb. Chem. High Throughput Screen.*, 2016, **19**, 275; H. Kiyani and M. Bamdad, *Rev. Roum. Chim.*, 2017, **62**, 221.
45. H. Kiyani and M. Bamdad, *Heterocycles*, 2017, **94**, 276.
46. H. Kiyani, H. Darbandi, A. Mosallanezhad, and F. Ghorbani, *Res. Chem. Intermed.*, 2015, **41**, 7561.
47. L. R. Jat, R. Mishra, and D. Pathak, *Int. J. Pharm. Pharm. Sci.*, 2012, **4**, 378.