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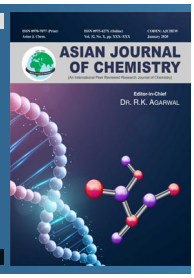

Grinding-Assisted Synthesis of Some Heterocyclic Compounds

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This article involves synthesis of some heterocyclic compounds from an amine derivative using eco-friendly approach (grinding). The first step involves synthesis of Schiff base from benzaldehyde derivatives (3-hydroxybenzaldehyde and 4-nitrobenzaldehyde) with 4-aminoantipyrine. These compounds were used as precursor for the synthesis of heterocyclic compounds and then synthesized tetrazole, oxazepine oxazepane derivatives from Schiff base with sodium azide, phthalic anhydride, maleic anhydride and succinic anhydride, respectively. The heterocyclic compounds were characterized by TLC, melting point, FTIR, ^{1}H NMR, ^{13}C NMR, GC-mass and HRMS analyses.

Keywords: Grinding, Heterocyclic compounds, Schiff base, Tetrazoles, 1,3-Oxazepine, 1,3-Oxazepane.

INTRODUCTION

Schiff base is derived by condensation reaction of aldehydes or ketones and primary amines, which contains -N=CHR group [1-5]. Antipyrine is very much used would equally be in medicine and believed that its amino derivative of much use in medicine possibly as intermediates in antipyretic and analgesic drugs [6-10]. Synthesis of Schiff base through classical condensation of aldehydes (or ketones) and amines were pursued, however, the yield of products were low [11,12].

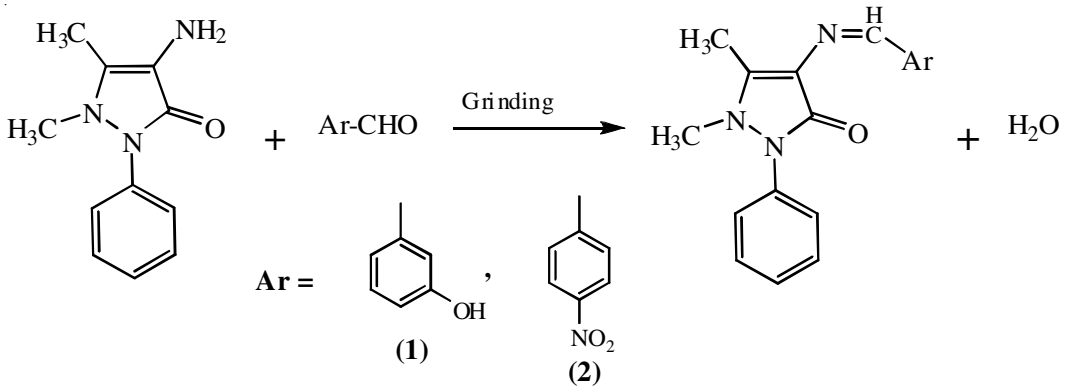
Synthesis of Schiff base is often carried out with acid catalyzed and generally by refluxing the mixture of aldehyde (or ketone) and amine in organic medium. However, with the assistance of grinding, it was found that condensation reaction of *p*-nitro or *m*-hydroxybenzaldehyde and 4-aminoantipyrine could proceed fast and efficiently without solvent [13,14]. The approach reported herein deals with the synthetic of some new derivatives of tetrazole, thiazolidinone and oxazepine starting Schiff base derived from reaction *p*-nitro or *m*-hydroxy benzaldehyde with 4-aminoantipyrine and then reacted with NaN_3, thioglycolic acid and (phthalic, succinic, maleic) anhydride.

Tetrazoles have found use in pharmaceuticals as lipophilic spacers and carboxylic acid surrogates [15], in specialty explosives, photography and information recording systems, not to mention as precursors to a variety of nitrogen containing heterocycles [16-18]. The most direct method to form tetrazoles is *via* the concerted and highly regioselective [2+3] cycloaddition between an inorganic azide (NaN_3) and a Schiff base this cycloaddition is too slow to be synthetically useful except when potent electron-withdrawing groups activate the nitrile component [19-21].

Seven membered heterocyclic ring system oxazepine has already been reported in the literature. Many of oxazepines and their oxides show interesting sedatives, muscle relaxant, and anticonvulsant properties in animals. The discovery of central nervous system activity (CNS) of 1,4-benzodiazepine several clinically useful drugs have been found which contain a heterocyclic moiety fused onto the 7-membered ring oxazepin-5-one derivatives [22-24]. In this work, an attempt has been made for the synthesis of some heterocyclic compounds using grinding method. This method given a good products and high yields. The synthesized compounds were ascertained from spectral and physio-chemical analysis. All of the periods of the completed reactions were checked by TLC.

EXPERIMENTAL

All chemicals and solvents were purchased from Sigma-Aldrich company. The melting point was determined by Stuat



Scheme-I: Preparation of Schiff base (**1, 2**)

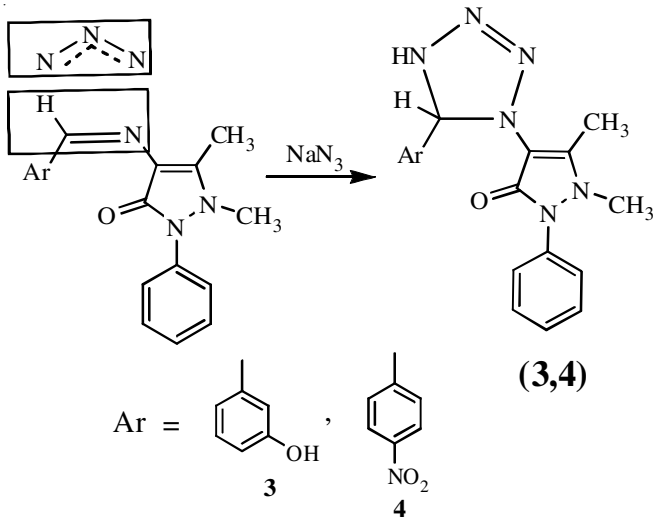
melting apparatus SM30. Ultrasound irradiation was recorded using Unisonics PTY (LTD type fxp12). Infrared spectra were recorded by a Bruker FT-IR Spectrophotometer (Tensor 27 Germany) and a biotech Engineering FT-IR-600 using KBr discs, Alpha Bruker/ATR Diamond. Ultra-Violet spectra were recorded by UV-Visible spectrophotometer (Shimadzu UV-1650 pc, Japan) using chloroform as a solvent. ^{1}H & ^{13}C NMR spectra were recorded by Bruker AC 250, Bruker ARX 300 and Bruker ARX 500. The ^{1}H NMR spectra presented in this work were collected in $CDCl_3$ or in DMSO-d_6 solution. Mass spectrometry has recorded by AMD MS40, varian MAT CH7, MAT 731 (EI, 7 0 eV), intecta AMD 402 (EI, 70 eV and CI), finnigan MAT 95 (CI, 200ev). High resolution mass spectrometry (HRMS) has recorded using Varian MAT 311, Intecta AMD 402. Elemental analysis has recorded by Thermoquest Flash EA 1112 (Leco CHNS-932) to ensure the purity of synthesized compounds. Thin layer chromatography (TLC) was carried out to ensure the completion of the reactions.

Synthesis of Schiff bases: 4-(3-hydroxybenzylideneamino)antipyrine (1) and 4-(4-nitrobenzylideneamino)antipyrine (2): A mixture of 4-aminoantipyrine (0.01 mol) and substituted benzaldehyde (0.01 mol) was grinded in a mortar with a pestle made of porcelain for 5-10 min. The mixture turns pastry after few minutes of grinding with the colour change during grinding process. A 15 mL of diethyl ether was added and then filtered the solid product, dried and recrystallized from absolute ethanol [25] (**Scheme-I**).

Synthesis of 4-(5-(*m*-hydroxyphenyl)-2,5-dihydro-1-tetrazolyl)antipyrine (3) and 4-(5-(*p*-nitrophenyl)-2,5-dihydro-1-tetrazolyl)antipyrine (4): The same procedure in the synthesis of compounds **1** and **2** was followed for the synthesis of compounds **3** and **4**. A mixture of Schiff bases (**1** and **2**) (0.01 mol) and sodium azide (0.01 mol) was taken for the synthesis of compounds **3** and **4** (**Scheme-II**).

Synthesis of 4-(4-antipyrinyl)-3-(*m*-hydroxyphenyl)-3,4- dihydrobenzo[*e*][1,3]oxazepine-1,5-dione (5) and 4-(4-antipyrinyl)-3-(*p*-nitrophenyl)-3,4-dihydrobenzo[*e*][1,3]-oxazepine-1,5-dione (6): In this case, mixture of Schiff base (0.001 mol) and phthalic anhydride (0.001 mol) was grinded for 10-15 min. The solid product filtered, dried and recrystallized from dichloromethane (DCM) (**Scheme-III**).

Synthesis of 2-(3-hydroxyphenyl)-3-(4-antipyrinyl)-2,3-dihydro-1,3-oxazepine-4,7-dione (7), 2-(4-nitrophenyl)-

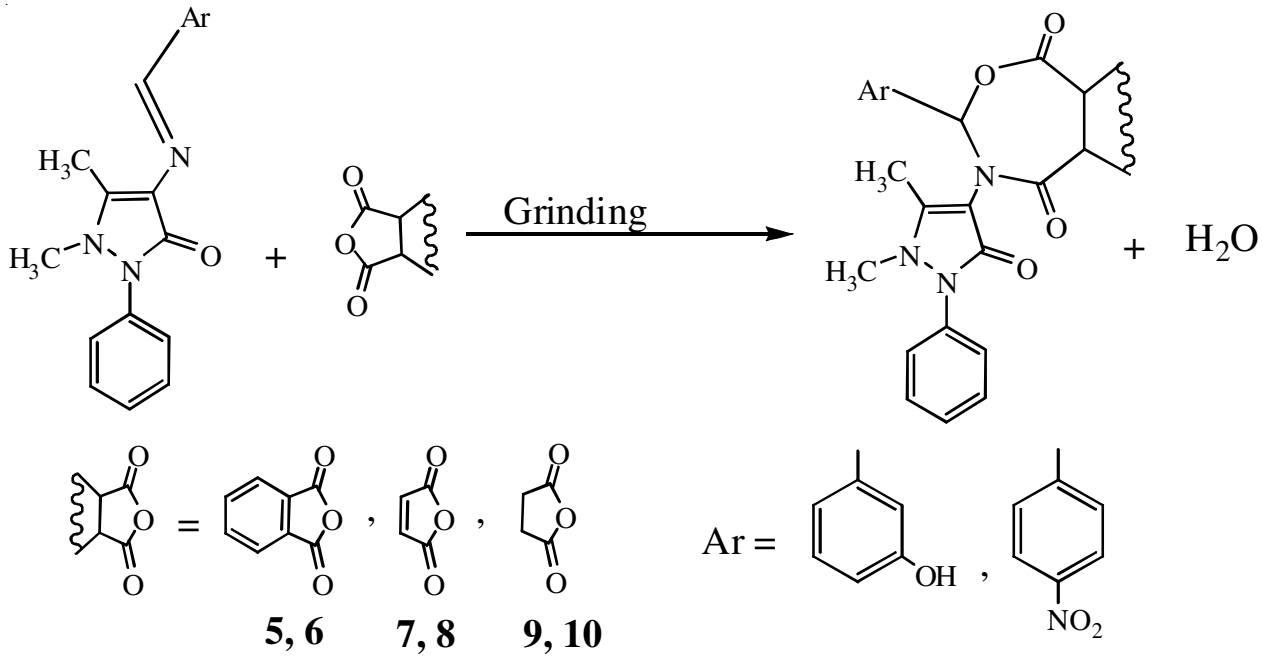


Scheme-II: A mechanism of tetrazole synthesis

3-(4-antipyrinyl)-2,3-dihydro-1,3-oxazepine-4,7-dione (8), 2-(3-hydroxyphenyl)-3-(4-antipyrinyl)-1,3-oxazepane-4,7-dione (9) and 2-(3-nitrophenyl)-3-(4-antipyrinyl)-1,3-oxazepane-4,7-dione (10): Follow the same procedure for preparation of compounds **5** and **6**. A mixture of Schiff base (0.001 mol) and maleic or succinic anhydride (0.001 mol) was grinded for 5 min. The solid product filtered, dried and recrystallized from THF (**Scheme-III**). The physico-chemical parameters of all the synthesized heterocyclic compounds are shown in Table-1.

RESULTS AND DISCUSSION

Schiff base compounds: Schiff base compounds were synthesized using condensation reaction of 4-aminoantipyrine with aldehyde derivatives (*viz*. 3-hydroxybenzaldehyde and 4-nitrobenzaldehyde). FTIR analysis of Schiff bases showed four major peaks based on the substitution groups in the compound and their association with their corresponding azomethine groups [26]. Of the groups -N=CH-, =CHAr, C=O Ap and C=C, -N=CH- and =CHAr have four different peaks appeared at 1605-1596 and 3095-3051 cm^{-1}, respectively; C=O Ap and C=C peaked at 1641-1622 and at 1580-1572 cm^{-1}, respectively. Furthermore, -N=CH- was observed at 3165 cm^{-1} for C-OH group (Table-2).



Scheme-III: Preparation of compounds (**5-10**)

TABLE-1
PHYSICAL AND ELEMENTAL ANALYSIS DATA OF SYNTHESIZED HETEROCYCLIC COMPOUNDS (**1-10**)

Compd. No.	m.p. (°C)	Colour	Solvent	R_f	Time (min)	m.f.	m.w.	Yield (%)	Elemental analysis (%): Calcd. (Found)		
									C	H	N
1	280-282	White crystals	Abs. EtOH	0.60	10	$C_{18}H_{17}N_3O_2$	307	96	70.13 (70.232)	5.58 (5.682)	13.67 (13.875)
2	260-261	Orange	Abs. EtOH	0.70	5	$C_{18}H_{16}N_4O_3$	336	98	64.28 (64.473)	4.79 (4.811)	16.66 (16.753)
3	138-140	White	Dioxane	0.83	15	$C_{18}H_{18}N_6O_2$	350	93	–	–	–
4	240-242	Red	Dioxane	0.85	12	$C_{18}H_{17}N_7O_3$	379	95	56.99 (57.011)	4.52 (4.734)	25.84 (24.628)
5	181-183	Yellow	DCM	0.77	10	$C_{26}H_{21}N_3O_5$	455	90	–	–	–
6	195-197	Deep yellow	DCM	0.72	15	$C_{26}H_{20}N_4O_6$	484	93	64.46 (64.571)	4.16 (4.371)	11.56 (11.746)
7	148-150	Yellow	THF	0.55	5	$C_{22}H_{19}N_3O_5$	405	88	–	–	–
8	214-216	Deep red	THF	0.53	5	$C_{22}H_{18}N_4O_6$	434	92	60.83 (60.937)	4.18 (4.293)	12.90 (12.714)
9	198-200	Pale yellow	THF	0.58	5	$C_{22}H_{21}N_3O_5$	407	83	–	–	–
10	204-206	Yellow	THF	0.67	5	$C_{22}H_{20}N_4O_6$	436	87	60.55 (60.844)	4.62 (4.717)	12.84 (12.953)

TABLE-2
KEY IR SPECTRAL BANDS (cm^{-1}) OF COMPOUNDS (**1-10**)

Compd. No.	C=O lacton	C=O lactam & Ap	C=N, C=C	N=N, C-S	N-H, O-H	Azide N=N-N	UV ($CHCl_3$) λ_{max} (nm)
1	–	–, 1622	1605, 1580	–	–, 3156	–	342
2	–	–, 1641	1596, 1572	–	–	–	398
3	–	–, 1613	–, 1589	1499, –	3137, 3156	2075-2175	326
4	–	–, 1637	–, 1595	1503, –	3175, –	2122-2179	332
5	1743	1635	–, 1583	–	–, 3156	–	288
6	1763	1642	–, 1596	–	–	–	292
7	1720	1640	–, 1590	–	–, 3156	–	338
8	1715	1641	–, 1589	–	–	–	316
9	1688	1637	–, 1587	–	–, 3156	–	294
10	1696	1643	–, 1596	–	–	–	302

^{1}H NMR analysis revealed that the peaks of CH for-N=CH(s), CHAr(m), OH(b), C-CH_3(s) and N-CH_3(s) were at δ 9.48- 9.52, δ 7.13-8.43, δ 9.46, δ 2.12-2.43 and δ 3.13-3.15 ppm, respectively (Table-3). In all the compounds, C-CH_3 and N-C-CH_3 peaks were almost constant. ^{13}C NMR analysis showed a major peaks of C for -N=C, Ar-C and C-OH at δ 157.7-159.8, δ 116-152 and 153.21 ppm, respectively. For solvent DMSO, the peaks were observedf at δ 38-41 ppm (Table-4).

GC-MS, EI and HRMS (Table-5) showed exactly the same molecular weights of these molecules, and also an elemental analysis (CHN) provided acceptable results.

Tetrazole compounds (3 and 4): Tetrazole compounds (**3** and **4**) were synthesized by reacting Schiff base (**1** and **2**) with NaN_3 using the grinding method. FTIR analysis showed that -N=CH- group disappeared at 1605-1596 cm^{-1} and reappeared at 1499-1503 cm^{-1} with a new peak relative to the N=N group and another peak appeared at 3137-3175 cm^{-1} due to the NH group [27] (Table-2). Moreover, C=O Ap, C=C, azide (N=N-N) and OH groups also observed at 1637-1613, 1595-1589, 2179-2075 and 3156 cm^{-1}, respectively.

^{1}H NMR analysis showed peaks at δ 9.68-10.16 ppm (s) for CH, δ 7.15-8.43 ppm for CHAr, δ 3.86-4.00 ppm for NH and δ 9.46 ppm for OH (Table-3). ^{13}C NMR analysis showed that C for CH, C=O Ap, Ar-C, C-OH and solvent $CDCl_3$ peaked at δ 76.73-80.19, δ 161.13-161.23, δ 116-151, δ 153.33 and δ 75-78 ppm, respectively (Table-4). Similarly, GC-MS, EI and HRMS data (Table-5) were in good agreement with the IR and NMR data.

TABLE-3
^{1}H NMR ANALYSIS DATA OF COMPOUNDS (**1-10**)

Comp. No	C-CH_3	N-CH_3	CH=N-	CH_2-CH_2	CH=CH olefinic	CH_2	$NH_{tz.}$	OH	C-H	Ar-H
1	s, 2.12	s, 3.13	s, 9.48					s.br, 9.46		7.13-7.51 (m, 9H)
2	s, 2.43	s, 3.15	s, 9.52							7.21-8.43 (m, 10H)
3	s, 2.10	s, 3.15					s.br, 3.86	s.br, 9.46	s, 9.68	7.15-7.55 (m, 9H)
4	s, 2.45	s, 3.17					s.br, 4.00		s, 10.16	7.35-8.43 (m, 9H)
5	s, 2.11	s, 3.10						s.br, 9.46	s, 9.63	7.15-7.55 (m, 9H)
6	s, 2.45	s, 3.16							s, 9.73	7.20-8.19 (m, 13H)
7	s, 2.10	s, 3.08			6.43-6.96 (d, 2H)			s.br, 9.46	s, 9.71	7.15-7.55 (m, 9H)
8	s, 2.46	s, 3.17			5.94-6.79 (d, 2H)				s, 9.70	7.19-8.34 (m, 9H)
9	s, 2.10	s, 3.08		1.21-1.93 (t, 4H)				s.br, 9.46	s, 9.67	7.15-7.55 (m, 9H)
10	s, 2.45	s, 3.16		1.18-1.83 (t, 4H)					s, 9.71	7.21-8.19 (m, 9H)

TABLE-4
^{13}C NMR ANALYSIS DATA OF COMPOUNDS (**1-10**)

Compd. No.	C-$^*CH_3$	N-$^*CH_3$	CH_3-$^*C$=$^*C$-N	CH_2, C-OH	$^*CH$, CH=N	Ar-C	CH=CH olefinic	CH_2-CH_2	C=O Ap	C=O Lactam	C=O lactone
1	9.97	35.33	105.23-136.73	–, 153.21	–, 157.7	116-147			160.33		
2	10.12	34.87	109.83-135.97		–, 159.8	122-152			161.47		
3	9.82	35.53	112.77-135.13	153.33	76.73	116-145			161.13		
4	9.85	34.91	115.83-135.92		80.19	123-151			161.23		
5	9.81	35.23	117.81-135.35	154.11	105.8	120-151			161.22	163.32	186.87
6	10.02	35.39	117.84-134.32		107.7	123-154			160.20	163.33	187.54
7	9.93	35.00	116.56-134.37	154.17	100.1	118-154	151.35-158.37		162.20	167.32	187.31
8	9.00	34.19	116.65-135.22		101.2	116-151	152.52-158.93		161.87	166.31	189.25
9	9.95	34.97		153.31	101.2	122-153		23.57-29.66	161.25	164.15	188.43
10	10.01	35.38	113.88-134.32		102.3	117-154		26.37-30.87	160.19	163.61	185.33

TABLE-5
GC-MASS, EI AND HRMS SPECTRUM OF COMPOUNDS (**1-10**)

Compd. No.	*m/z*	%	*m/z*	%	*m/z*	%	*m/z*	%	*m/z*	%	HRMS	
											Calc.	Meas.
1	308	16	307	75	199	23	188	25	171	19	307.13228	308.14353
	121	33	89	15	77	17	56	100				
2	337	6	336	34	188	20	121	33	91	22	336.12172	337.12952
	78	14	44	13								
4	380	12	379	86	336	90	281	15	244	25	379.14739	380.14664
	216	21	188	35	121	50	56	100				
6	485	15	484	65	431	17	336	70	306	30	484.13764	485.14571
	244	30	188	40	121	50	56	100				
8	435	17	434	72	410	42	336	50	242	25	434.12216	435.12991
	188	45	121	35	77	25	56	100				
10	437	13	436	60	405	15	336	82	244	15	436.13536	437.14552
	188	31	121	43	77	13	56	100				

*$[M+H]^+$ = Calculated molecular ion mass or measured molecular ion mass for some compounds.

Oxazipene compounds (5-10): Oxazipene compounds (**5-10**) were synthesized by reacting Schiff base (**1** and **2**) with phthalic anhydride (**5** and **6**)/maleic anhydride (**7** and **8**)/succinic anhydride (**9** and **10**) using the grinding method in absence of any solvent [28].

FTIR analysis showed that -N=CH- group disappeared at 1605-1596 cm^{-1} and reappeared with a new peak at 1763-1743 cm^{-1} (phthalic moiety), 1696-1688 cm^{-1} (maleic moiety) and 1720-1715 cm^{-1} (succinic moiety) relative to the C=O of lactone group, and another peak appeared at 1642-1635 cm^{-1} (phthalic moiety), 1641-1640 cm^{-1} (maleic moiety) and 1643-1637 cm^{-1} (succinic moiety) attributed due to the C=O lactam and Ap. In compounds **5**, **7** and **9**, C=C and OH groups appeared at 1596-1583, 1596-1587, 1590-1589 cm^{-1} and 3156 cm^{-1}, respectively (Table-2).

^{1}H NMR analysis showed peaks at δ 9.63-9.73 ppm (phthalic moiety), δ 9.71-9.70 (maleic moiety), δ 9.67-9.71 ppm (succinic moiety) for CH, δ 7.15-8.19 ppm (phthalic moiety), δ 7.15-8.34 ppm (maleic moiety) for ArH and δ 9.46 ppm for OH. Moreover, at peak at 5.94-6.96 ppm for CH=CH olefinic was also observed (maleic moiety), similarly, peaks at δ 1.21-1.18 and δ 1.93-1.83 ppm were observed for CH_2CON and CH_2COO, (succinic moiety), respectively (Table-3).

^{1}H NMR analysis showed that CH (s), CH_2 (q), Ar-H (m), and OH (s) peaked at δ 6.12-6.03, δ 2.76-4.08, δ 6.85-8.51, and δ 9.46 ppm, respectively (Table-3). ^{13}C NMR analysis showed that C for CH, C=O of lactam, C=O Ap, Ar-C, CH_2, and solvent DMSO observed at δ 61.56-64.17, δ 170.22-170.17, δ 167.45-161.35, δ 115-151, δ 33.89-31.65 and δ 38-41 ppm, respectively (Table-4). Similarly, GC-MS, EI and HRMS data (Table-5) are in good agreement with the structure of synthesized compounds.

Conclusion

High yields of several heterocyclic compounds (tetrazole, oxazepine, oxazepane and thiazolidinone derivatives) were synthesized through the grinding method. The characterization data (FTIR, ^{1}H & ^{13}C NMR, GC-MS, EI, HRMS and CHNS) is fully agreed with the synthesized compounds. The merits of this method are low energy requirement, short processing time, cheap, eco-friendly and no side products.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

REFERENCES

1. M.A. Malik, O.A. Dar, P. Gull, M.Y. Wani and A.A. Hashmi, *Med. Chem. Commun.*, **9**, 409 (2018); https://doi.org/10.1039/C7MD00526A
2. S. Kumar, D.N. Dhar and P.N. Saxena, *J. Sci. Ind. Res. India*, **68**, 181 (2009).
3. J.H. Tomma, M.S. Khazaal and A.H. Al-Dujaili, *Arab. J. Chem.*, **7**, 157 (2014); https://doi.org/10.1016/j.arabjc.2013.08.024
4. W. Qin, S. Long, M. Panunzio and S. Biondi, *Molecules*, **18**, 12264 (2013); https://doi.org/10.3390/molecules181012264
5. C.J. Dhanaraj, J. Johnson, J. Joseph and R.S. Joseyphus, *J. Coord. Chem.*, **66**, 1416 (2013); https://doi.org/10.1080/00958972.2013.782008
6. Y. Teng, F. Ji, C. Li, Z. Yu and R. Liu, *J. Lumin.*, **131**, 2661 (2011); https://doi.org/10.1016/j.jlumin.2011.07.005
7. A. Lang, C. Hatscher, C. Wiegert and P. Kuhl, *Amino Acids*, **36**, 333 (2009); https://doi.org/10.1007/s00726-008-0074-1
8. C.A. Lang, P.K. Hatscher and P. Kuhl, *Tetrahedron Lett.*, **48**, 3371 (2007); https://doi.org/10.1016/j.tetlet.2007.03.067
9. R. Teran, R. Guevara, J. Mora, L. Dobronski, O. Barreiro-Costa, T. Beske, J. Pérez-Barrera, R. Araya-Maturana, P. Rojas-Silva, A. Poveda and J. Heredia-Moya, *Molecules*, **24**, 2696 (2019); https://doi.org/10.3390/molecules24152696
10. M.S. Alam, D.-U. Lee and L. Bari, *J. Korean Soc. Appl. Biol. Chem.*, **57**, 613 (2014); https://doi.org/10.1007/s13765-014-4201-2
11. R. Hassan, H. Arida, M. Montasser and N.A. Latif, *J. Chem.*, **2013**, 240568 (2013); https://doi.org/10.1155/2013/240568
12. J. Xu, Y. Liu and S.-H. Hsu, *Molecules*, **24**, 3005 (2019); https://doi.org/10.3390/molecules24163005
13. R.T. Haiwal, *J. Kerbala Univ.*, **6**, 216 (2008).
14. Z.H. Abood, *J. Kerbala Univ.*, **7**, 297 (2009).
15. C.G. Neochoritis, T. Zhao and A. Domling, *Chem. Rev.*, **119**, 1970 (2019); https://doi.org/10.1021/acs.chemrev.8b00564
16. Y. Zou, L. Liu, J. Liu and G. Liu, *Future Med. Chem.*, **12**, 91 (2020); https://doi.org/10.4155/fmc-2019-0288
17. N. Kaur, *Synth. Commun.*, **49**, 1633 (2019); https://doi.org/10.1080/00397911.2018.1542497

18. J.F. Campos and S. Berteina-Raboin, *Catalysts*, **10**, 631 (2020); https://doi.org/10.3390/catal10060631
19. Y. Chen, G. Nie, Q. Zhang, S. Ma, H. Li and Q. Hu, *Org. Lett*., **17**, 1118 (2015); https://doi.org/10.1021/ol503687w
20. M.S. Singh, S. Chowdhury and S. Koley, *Tetrahedron*, **72**, 5257 (2016); https://doi.org/10.1016/j.tet.2016.07.044
21. S. Kamijo, T. Jin, Z. Huo and Y. Yamamoto, *J. Am. Chem. Soc*., **125**, 7786 (2003); https://doi.org/10.1021/ja034191s
22. N. Kaur, *Synth. Commun*., **49**, 987 (2019); https://doi.org/10.1080/00397911.2019.1574351
23. J.-Y. Wang, X.-F. Guo, D.-X. Wang, Z.-T. Huang and M.-X. Wang, *J. Org. Chem.*, **73**, 1979 (2008); https://doi.org/10.1021/jo7024306
24. M. Driowya, A. Saber, H. Marzag, L. Demange, K. Bougrin and R. Benhida, *Molecules*, **21**, 1032 (2016); https://doi.org/10.3390/molecules21081032
25. A. Naqvi, M. Shahnawaaz, A.V. Rao, D. Seth and N.K. Sharma, *J. Chem.*, **6**, 75 (2009).
26. K. Patel, J. Modha and H. Mehta, *Int. J. ChemTech Res.*, **7**, 2108 (2015).
27. C.K. Mathews, K.E. Hold and K.G. Ahern, *National J. Chem.*, **41**, 77 (2011).
28. M.S. Al-Rawi, J.H. Tomma and D.F. Hussein, *Iraqi National J. Chem.*, **55**, 264 (2014).