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A convenient [hydroxy(tosyloxy)iodo]benzene-mediated one-pot synthesis of 2-arylimidazo[2,1-*b*]benzothiazoles

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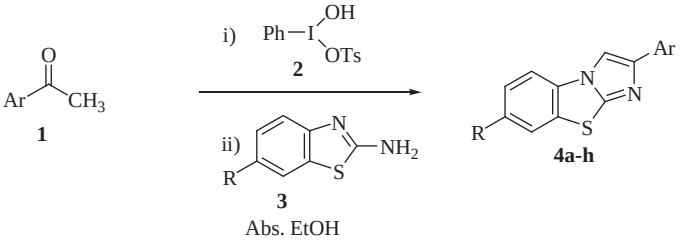
Several 2-arylimidazo[2,1-*b*]benzothiazoles (**4**) have been conveniently synthesized in one-pot reactions *via* α-tosyloxylation of enolizable ketones (**1**) using [hydroxy(tosyloxy)iodo]benzene **2** in acetonitrile, followed by treatment with 2-amino-6-(substituted)benzothiazoles (**3**). The present protocol offers several advantages towards general access of 2-arylimidazo[2,1-*b*]benzothiazoles, including an intriguing alternative to the literature protocols, a readily available nontoxic reagent, operational simplicity and an environmentally benign procedure.



Keywords: α-tosyloxyketones; [hydroxy(tosyloxy)iodo]benzene; imidazo[2,1-*b*]benzothiazoles; hypervalent iodine; 2-amino-6-(substituted)benzothiazoles

1. Introduction

The imidazo[2,1-*b*]benzothiazole structural motif may be found in a large number of pharmaceutical agents with a diverse range of physiological activities, namely, antitumor,[1] antibacterial,[2,3] antifungal [4] and immunosuppressive.[5] A series of substituted imidazo[2,1-*b*]benzothiazoles possess inverse-agonist profiles toward the central benzodiazepine [6] and 5-hydroxytryptamine$_3$ receptors.[7] As shown in Figure 1, imidazo[2,1-*b*]benzothiazole derivative (AC220) **I** was identified as one of the most potent and selective FMS-like tyrosine kinase-3 inhibitor.[8] Furthermore, imidazo[2,1-*b*]benzothiazoles can be synthetically manipulated easily

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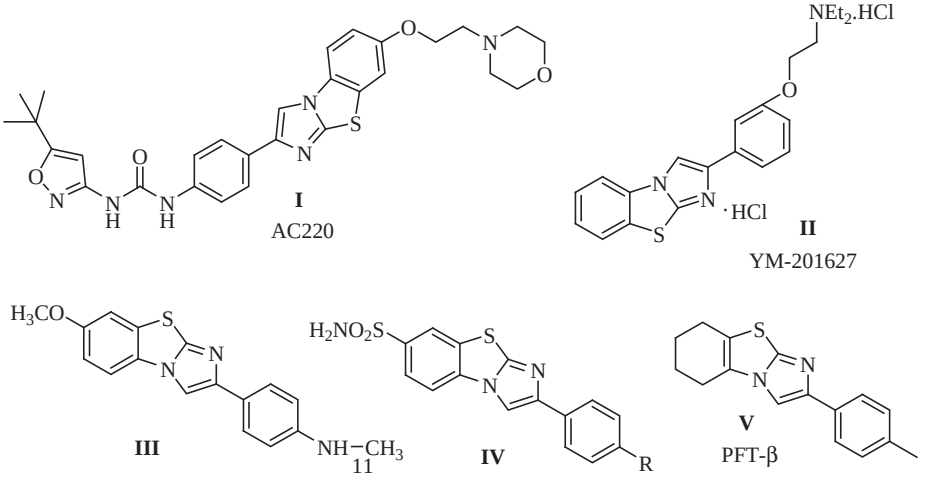


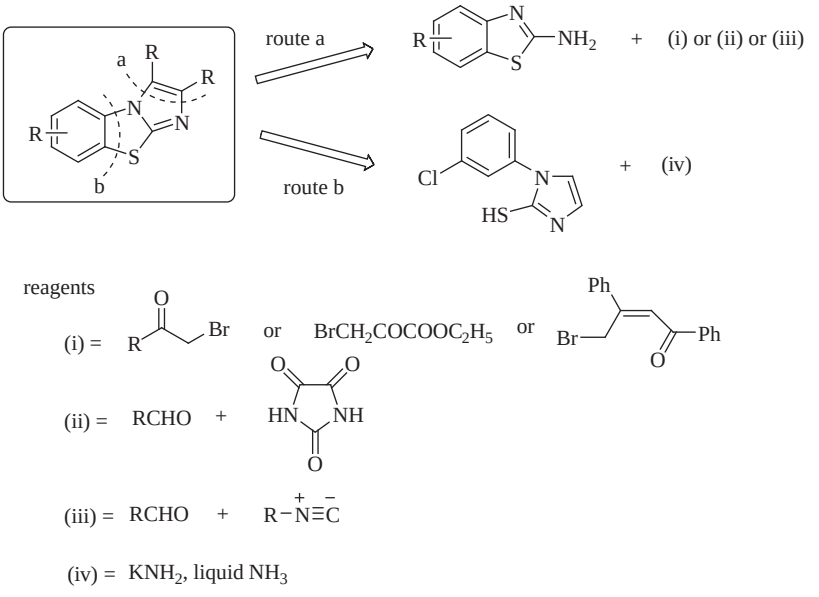
Figure 1. 2-Arylimidazo[2,1-*b*]benzothiazoles and related compounds with biological and medicinal activity.

by Mannich, substitution and addition reactions for preparing a wide range of compounds that have immense pharmaceutical value.[9–11]

In particular, 2-arylimidazo[2,1-*b*]benzothiazole derivative (YM-201627) **II** has proven to be a potent and orally active antitumor agent (Figure 1).[12] ^{11}C-labeled imidazo[2,1-*b*]benzothiazole **III** has been shown to be a superb fluoroprobe in positron emission tomography analysis of Alzheimer's disease.[13] Imidazobenzothiazoles **IV** are well-known inhibitors of apoptosis following the p-53 independent pathway in contrast to the well-known standard drug piffithrin-α (PFT-α or PFT-β **V**).[14] It has recently been reported that Mannich bases of 2-arylimidazo[2,1-*b*]benzothiazoles possess potential anti-cancer activities.[11]

Mainly two distinct routes have been employed in the literature for the synthesis of imidazo[2,1-*b*]benzothiazoles (Scheme 1). The first one involves the construction of a imidazole ring on the starting 2-aminobenzothiazole by condensation with 1,2-difunctionalized units, such as α-haloketones,[1,4,5,8,9,11,14–16] α-haloester,[6] and γ-bromodypnone [17]; a multicomponent reaction between 2-aminobenzothiazoles, benzaldehydes and imidazoline-2,4,5-trione under solvent-free conditions [18]; and Groebke–Blackburn multicomponent reaction of 2-aminobenzothiazoles with aldehydes and isocynides (**route a**).[2,19] The second approach involves appending the thiazole ring by reacting 1-(*m*-chlorophenyl)-2-mercaptoimidazole with potassium amide in liquid ammonia (**route b**).[20] Other synthetic routes for the preparation of these compounds involve annulation reactions of amidines with substituted bromomethyl compounds, acid chlorides and acrylic dienophiles [21] and Pd/Cu catalyzed Sonogashira coupling reaction of iodobenzenes with 2-imino-3-(2-propynyl)-1,3-benzothiazole.[22,23]

However, many of these procedures require sometimes multistep synthesis, harsh reaction conditions, cumbersome product isolation procedure, unsatisfactory yields, special care in handling and storing the toxic and expensive reagents. Therefore, a need still exists for further development of a versatile and convenient synthetic protocol for the synthesis of 2-arylimidazo[2,1-*b*]benzothiazoles. Our ongoing program on the development of greener protocols in heterocyclic synthesis coupled with significant biological importance,[24–28] prompted us to extend the versatility of [hydroxy(tosyloxy)iodo]benzene HTIB for the synthesis of 2-arylimidazo[2,1-*b*]benzothiazoles involving the intermediacy of α-tosyloxyketones (α-TK).



Scheme 1. Retrosynthetic routes for imidazo[2,1-*b*]benzothiazoles.

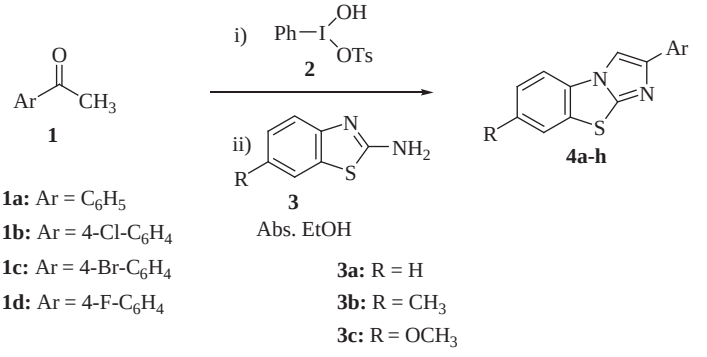
2. Results and discussion

α-TK, readily accessible through oxidation of enolizable ketones with HTIB, offer an attractive alternative due to their versatility in organic synthesis, as these are relatively stable chemical entities, have ready accessibility, site-selectivity in the reaction and provide a safe eco-friendly alternative route to conventional toxic and lachrymatory α-haloketones for clean synthesis.[29–32]

The starting material, 2-amino-6-(substituted)benzothiazoles (**3**), was synthesized by the reaction of appropriate aniline with potassium thiocyanate which resulted in the formation of substituted phenylthiourea. The ensuing reaction of substituted phenylthiourea in the presence of bromine in chloroform followed by neutralization with aqueous ammonia afforded **3**.[33]

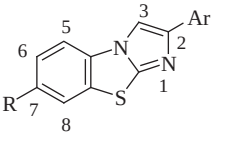
A one-pot HTIB-mediated sequential synthesis of title compounds is depicted in Scheme 2. Initially, acetophenone (**1a**) was oxidized with one equivalent of HTIB **2** in acetonitrile under reflux condition for 2 h and subsequently the α-TK so generated *in situ* was treated with 2-aminobenzothiazole (**3a**) in absolute ethanol. The progress of the reaction was monitored by thin layer chromatography (TLC) which indicated the formation of α-TK after 2 h and a single spot for **4a** after 5 h. Simple workup of the reaction resulted in the formation of 2-phenylimidazo[2,1-*b*]benzothiazole (**4a**) in 62% yield (Scheme 2). ^{1}H NMR spectrum of compound **4a** exhibited a characteristic singlet at δ 7.87 due to the C_3-H of the imidazole ring confirming the cyclization.

In order to study the scope of this approach, various enolizable ketones (**1b–d**) were subjected to α-tosyloxylation with HTIB **2** and ensuing reaction with 2-amino-6-(substituted)benzothiazoles (**3a–c**) under the above conditions in order to achieve the synthesis of **4b–h**. In all the cases, the corresponding 2-arylimidazo[2,1-*b*]benzothiazoles (**4b–h**) (Scheme 2) were obtained in excellent yields ranging from 60% to 72% (Table 1). The synthesized compounds **4** were characterized by comparing their mps [5,15] and spectral properties with those reported in the literature.[11] The versatility of the present protocol was further proved by the fact that when the reaction was scaled from milligram to gram level under the same conditions, it took



Scheme 2. Synthesis of 2-arylimidazo[2,1-*b*]benzothiazoles (**4**).

Table 1. Yield and characterization of 2-arylimidazo[2,1-*b*]benzothiazoles (**4**).



Product	Ar	R	M.p. (°C)	Lit. M.p. (°C)	Yield[a] (%) one pot	Lit. yield[b] (%)
4a		H	102	100 [5,15]	62	30 [5], N.R.[c] [15]
4b	4-Cl	H	160	159 [5,15]	65	44 [5], 28.1 [15]
4c	4-Br	H	164	161[5,15]	60	16 [5], 36.47 [15]
4d	4-F	H	151	148 [15]	72	26.11 [15]
4e		CH_3	165	165 [15]	63	41.4 [15]
4f	4-Cl	CH_3	212	215 [15]	66	43.95 [15]
4g		OCH_3	164	164 [15]	67	N.R. [15]
4h	4-Cl	OCH_3	217	217 [15]	70	57.2 [15]

[a] Yield of isolated product.
[b] Yield of isolated pure product based on amount of appropriate 2-aminobenzothiazole used.
[c] N.R.: not reported.

place successfully to give the 2-arylimidazo[2,1-*b*]benzothiazoles **4**. The process is environmentally benign and the reaction conditions and workup procedure are mild, simple and convenient. The results summarized in Table 1 clearly indicate that the new one-pot synthesis affords much better overall yields (60–72%) as compared with the literature procedure (16–57%) involving the lachrymatory α-haloketones.[5,15]

3. Conclusions

In a nutshell, the noteworthy features of present study are:

(i) HTIB-mediated one-pot sequential protocol offers general applicability for synthesis of 2-arylimidazo[2,1-*b*]benzothiazoles, which have diverse synthetic applications.
(ii) α-TK, generated *in situ* as intermediate, provide a safe eco-friendly alternative approach which avoids the use of highly toxic, unstable and lachrymatory α-haloketones.

4. Experimental

Melting points were determined in open capillaries in an electrical apparatus and are uncorrected. The homogeneity of the compounds was checked by TLC using silica gel-G (Acme). IR spectra were recorded on a Buck Scientific IR M-500 spectrophotometer in KBr discs (ν_{max} in cm^{-1}), ^1H NMR spectra in $CDCl_3$ on a Bruker (300 and 400 MHz) spectrophotometer; chemical shifts are expressed in δ-scale downfield from TMS as an internal standard.

4.1. *General procedure for one-pot synthesis of 2-arylimidazo[2,1-b]benzothiazoles (4a–h) from ketones 1*

A mixture of appropriate methyl ketone **1** (5 mmol) and HTIB **2** (1.95 g, 5 mmol) in acetonitrile (20 mL) was heated under reflux for 2 h. Excess of acetonitrile was distilled off. The formation of α-TK was confirmed by TLC monitoring of the reaction mixture. Then a solution of 2-amino-6-(substituted)benzothiazole **3** (5 mmol) in absolute ethanol (10 mL) was added dropwise to the reaction mixture. The resultant reaction mixture was heated under reflux for 4–5 h. The progress of the reaction was accomplished by TLC using pet ether:ethyl acetate (8:2) as the solvent system. After completion of the reaction, the excess of solvent was distilled *in vacuo*, and a solid product separated out on cooling. The solid thus separated was filtered, neutralized with aq. sodium bicarbonate solution, washed with water and purified by recrystallization from ethanol to afford **4**. The spectroscopic data were consistent with literature.

4.1.1. *2-Phenylimidazo[2,1-b]benzothiazole (**4a**)*

^1H NMR ($CDCl_3$, 300 MHz): δ 7.03 (t, 1H, ArH), 7.32–7.37 (m, 4H, ArH), 7.47 (m, 1H, ArH), 7.49–7.59 (m, 2H, ArH), 7.80–7.83 (m, 1H, ArH), 7.87 (s, 1H, Imidazole-H).

4.1.2. *2-(4-Chlorophenyl)imidazo[2,1-b]benzothiazole (**4b**)*

Synthesis of this compound was accomplished on gram scale following the same procedure, employing appropriate methyl ketone **1** (0.01 mol), HTIB **2** (3.9 g, 0.01 mol) and compound **3a** (0.01 mol) to obtain pure product **4b** as a white solid.

^1H NMR ($CDCl_3$, 300 MHz): δ 7.23–7.26 (t, 1H, C_7-H), 7.27–7.30 (d, 2H, $J_o = 8.4$ Hz, Ph-2′, 6′-H), 7.33–7.39 (t, 1H, $J = 7.8$ Hz, C_6-H), 7.48–7.51 (d, 1H, $J = 7.8$ Hz, C_8-H), 7.59–7.62 (d, 1H, $J = 7.8$ Hz, C_5-H), 7.69–7.72 (d, 2H, $J_o = 8.4$ Hz, Ph-3′, 5′-H), 7.84 (s, 1H, Imidazole-H).

4.1.3. *2-(4-Bromophenyl)imidazo[2,1-b]benzothiazole (**4c**)*

^{1}H NMR ($CDCl_3$, 400 MHz): δ 7.37 (dt, 1H, $J = 7.96$ Hz, $J = 1.0$ Hz, C_7-H), 7.47 (dt, 1H, $J = 7.82$ Hz, $J = 1.04$ Hz, C_6-H), 7.54 (m, 2H, $J = 8.52$ Hz, $J = 1.72$ Hz, Ph-2′, 6′-H), 7.61 (d, 1H, $J = 7.68$ Hz, C_8-H), 7.72 (d, 1H, $J = 7.96$ Hz, C_5-H), 7.75 (m, 2H, $J = 8.56$ Hz, $J = 1.8$ Hz, Ph-3′, 5′-H), 7.96 (s, 1H, Imidazole-H).

4.1.4. *2-(4-Fluorophenyl)imidazo[2,1-b]benzothiazole (**4d**)*

^{1}H NMR ($CDCl_3$, 400 MHz): δ 7.12 (t, 2H, $J = 8.72$ Hz, Ph-3′, 5′-H), 7.35 (dt, 1H, $J = 7.8$ Hz, $J = 0.88$ Hz, C_7-H), 7.46 (dt, 1H, $J = 7.72$ Hz, $J = 0.88$ Hz, C_6-H), 7.60 (d, 1H, $J = 7.92$ Hz, C_8-H), 7.70 (d, 1H, $J = 7.92$ Hz, C_5-H), 7.84 (m, 2H, $J = 7.72$ Hz, $J = 5.4$ Hz, Ph-2′, 6′-H), 7.90 (s, 1H, Imidazole-H).

4.1.5. *7-Methyl-2-phenylimidazo[2,1-b]benzothiazole (**4e**)*

^{1}H NMR ($CDCl_3$, 300 MHz): δ 2.47 (s, 3H, CH_3), 7.24 (d, 1H, $J = 7.6$ Hz, C_6-H), 7.29 (t, 1H, $J = 7.16$ Hz, Ph-4′-H), 7.39 (t, 2H, Ph-3′, 5′-H), 7.4–7.52 (m, 2H , C_8, C_5-H), 7.72–7.74 (m, 2H, phenyl-2′, 6′-H), 7.88 (s, 1H, Imidazole-H).

4.1.6. *2-(4-Chlorophenyl)-7-methylimidazo[2,1-b]benzothiazole (**4f**)*

^{1}H NMR ($CDCl_3$, 400 MHz): δ 2.47 (s, 3H, CH_3), 7.25 (d, 1H, $J = 7.6$ Hz, C_6-H), 7.38 (m, 2H, $J_o = 8.52$ Hz, $J_m = 2.48$ Hz, $J_p = 1.8$ Hz, Ph-2′, 6′-H), 7.46 (m, 1H, C_8-H), 7.49 (m, 1H, C_5-H), 7.79 (m, 2H, $J_o = 8.52$ Hz, $J_m = 2.48$ Hz, $J_p = 1.8$ Hz, Ph-3′, 5′-H), 7.90 (s, 1H, Imidazole-H).

4.1.7. *7-Methoxy-2-phenyl imidazo[2,1-b]benzothiazole (**4g**)*

^{1}H NMR ($CDCl_3$, 400 MHz): δ 3.88 (s, 3H, OCH_3), 7.01 (dd, 1H, $J = 8.78$ Hz, $J = 2.44$ Hz, C_6-H), 7.21 (d, 1H, $J = 2.44$ Hz, C_8-H), 7.32 (t, 1H, $J = 7.40$ Hz, Ph-4′-H), 7.43 (t, 2H, $J = 7.48$ Hz, Ph-3′, 5′-H), 7.50 (d, 1H, $J = 8.76$ Hz, C_5-H), 7.88 (d, 2H, $J = 7.24$ Hz, Ph-2′, 6′-H), 7.91 (s, 1H, Imidazole-H).

4.1.8. *2-(4-Chlorophenyl)-7-methoxyimidazo[2,1-b]benzothiazole (**4h**)*

^{1}H NMR ($CDCl_3$, 400 MHz): δ 3.90 (s, 3H, OCH_3), 7.03 (dd, 1H, $J = 8.84$ Hz, $J = 2.44$ Hz, C_6-H), 7.23 (d, 1H, $J = 2.44$ Hz, C_8-H), 7.39 (m, 2H, $J_o = 8.56$ Hz, $J_m = 2.44$ Hz, $J_p = 1.76$ Hz, Ph-2′, 6′-H), 7.52 (d, 1H, $J = 8.8$ Hz, C_5-H), 7.80 (m, 2H, $J_o = 8.56$ Hz, $J_m = 2.48$ Hz, $J_p = 1.76$ Hz, Ph-3′, 5′-H), 7.90 (s, 1H, Imidazole-H).

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Disclosure statement

No potential conflict of interest was reported by the authors.

Supplemental material

Supplemental data for this article can be accessed at doi:10.1080/17415993.2014.996221.

References

[1] Furlan A, Colombo F, Kover A, Issaly N, Tintori C, Angeli L, Leroux V, Letard S, Amat M, Asses Y, Maigret B, Dubreuil P, Botta M, Dono R, Bosch J, Piccolo O, Passarella D, Maina F. Identification of new aminoacid amides containing the imidazo[2,1-*b*]benzothiazol-2-ylphenyl moiety as inhibitors of tumorigenesis by oncogenic Met signaling. Eur J Med Chem. 2012;47:239–254.

[2] Al-Tel TH, Al-Qawasmeh RA, Zaarour R. Design, synthesis and *in vitro* antimicrobial evaluation of novel imidazo[1,2-*a*]pyridine and imidazo[2,1-*b*][1,3]benzothiazole motifs. Eur J Med Chem. 2011;46:1874–1881.

[3] Palkar M, Noolvi M, Sankangoud R, Maddi V, Gadad A, Nargund LVG. Synthesis and antibacterial activity of a novel series of 2,3-diaryl-substituted-imidazo(2,1-*b*)-benzothiazole derivatives. Archiv der Pharmazie. 2010;343:353–359.

[4] Malik JK, Soni H, Singhai AK. Synthesis, characterization and evaluation for antifungal activity of substituted diaryl imidazo [2,1-*b*]-benzothiazole. J Pharm Res. 2013;7:39–46.

[5] Mase T, Arima H, Tomioka K, Yamada T, Murase K. Imidazo[2,1-*b*]benzothiazoles. 2. New immunosuppressive agents. J Med Chem. 1986:29;386–394.

[6] Trapani G, Franco M, Latrofa A, Carotti A, Genchi G, Serra M, Biggio G, Liso G. Synthesis and benzodiazepine receptor binding of some imidazo- and pyrimido[2,1-*b*] benzothiazoles. Eur J Med Chem. 1996;31:575–587.

[7] Mahesh R, Venkatesha Perumal R, Pandi PV. Microwave-assisted solvent-free synthesis of 3-[(4-substituted piperazin-1-yl)alkyl] imidazo[2,1-*b*][1,3]benzothiazol-2(3*H*)-ones as serotonin$_3$ (5-HT$_3$) receptor antagonists. Pharmaize. 2005;60:411–414.

[8] Chao Q, Sprankle KG, Grotzfeld RM, Lai AG, Carter TA, Velasco AM, Gunawardane RN, Cramer MD, Gardner MF, James J, Zarrinkar PP, Patel HK, Bhagwat SS. Identification of *N*-(5-*tert*-butyl-isoxazol-3-yl)-N′-{4-[7-(2-morpholin-4-yl-ethoxy)imidazo-[2,1-*b*][1,3]benzothiazol-2-yl]phenyl}urea dihydrochloride (AC220), a uniquely potent, selective, and efficacious FMS-like tyrosine kinase-3 (FLT3) inhibitor. J Med Chem. 2009;52:7808–7816.

[9] El-Shorbagi A-N, Sakai S-I, El-Gendy MA, Omar N, Farag HH. Imidazo[2,1-*b*]benzothiazoles. I. Chem Pharm Bull. 1988;36:4760–4768.

[10] Pandey AK, Sharma R, Singh A, Shukla S, Srivastava K, Puri SK, Kumar B, Chauhan PMS. Synthesis of biologically active pyridoimidazole/imidazobenzothiazole annulated polyheterocycles using cyanuric chloride in water. RSC Adv. 2014;4:26757–26770.

[11] Kumbhare RM, Kumar KV, Ramaiah MJ, Dadmal T, Pushpavalli SNCVL, Mukhopadhyay D, Divya B, Devi TA, Kosurkar U, Pal-Bhadra M. Synthesis and biological evaluation of novel Mannich bases of 2-arylimidazo[2,1-*b*]benzothiazoles as potential anti-cancer agents. Eur J Med Chem. 2011;46:4258–4266.

[12] Amino N, Ideyama Y, Yamano M, Kuromitsu S, Tajinda K, Samizu K, Matsuhisa A, Kudoh M, Shibasaki M. YM-201627: an orally active antitumor agent with selective inhibition of vascular endothelial cell proliferation. Cancer Lett. 2006;238:119–127.

[13] Yousefi BH, Manook A, Drzezga A, Reutern BV, Schwaiger M, Wester H-J, Henriksen G. Synthesis and evaluation of ^{11}C-labeled imidazo[2,1-*b*]benzothiazoles (IBTs) as PET tracers for imaging β-amyloid plaques in Alzheimer's disease. J Med Chem. 2011;54:949–956.

[14] Chandak N, Bhardwaj JK, Sharma RK, Sharma PK. Inhibitors of apoptosis in testicular germ cells: synthesis and biological evaluation of some novel IBTs bearing sulfonamide moiety. Eur J Med Chem. 2013;59:203–208.

[15] Buu-Hoi NP, Xuong ND, Cuc TT. Reaction of ω-bromoacetophenones with 2-aminothiazole and 2-aminobenzothiazoles. Bull Soc Chim France. 1966;4:1277–1279; Chem. Abstr. 1966, 65, 7164a.

[16] Rani R, Makrandi JK. Microwave assisted synthesis and antimicrobial activity of some 2-(benzo-furan-2-yl)-7-(substituted)imidazo[2,1-*b*]benzothiazoles. Indian J Chem. 2009;48B:1614–1617.

[17] Kovtunenko V, Potikha L, Turov A. Synthesis of fused imidazoles based on γ-bromodypnone. Synthetic Commun. 2004;34:3609–3613.

[18] Adib M, Sheibani E, Zhu L-G, Bijanzadeh HR. Efficient synthesis of imidazo[2,1-*b*][1,3]benzothiazoles and 9*H*-imidazo-[1,2-*a*][1,3]benzimidazoles under solvent-free conditions. Synlett. 2008(19):2941–2944.

[19] Tsirulnikov S, Kysil V, Ivachtchenko A, Krasavin M. Trimethylsilyl chloride as a promoter for the Groebke-Blackburn reaction: preparation of imidazo[2,1-*b*][1,3]benzothiazoles. Synth Commun. 2009;40:111–119.

[20] Ogura H, Itoh T. Derivatives of imidazo[2,1-*b*]benzothiazole (studies on heterocyclic compounds. VII). Chem Pharm Bull. 1970;18:1981–1986.

[21] Landreau C, Deniaud D, Evain M, Reliquet A, Meslin J-C. Efficient regioselective synthesis of triheterocyclic compounds: imidazo[2,1-*b*]benzothiazoles, pyrimido[2,1-*b*]benzothiazolones and pyrimido[2,1-*b*]benzothiazoles. J Chem Soc, Perkin Trans. 2002;1:741–745.

[22] Kamali TA, Habibi D, Nasrollahzadeh M. Synthesis of 6-substituted imidazo[2,1-*b*][1,3]thiazoles and 2-substituted imidazo[2,1-*b*][1,3]benzothiazoles *via* Pd/Cu-mediated Sonogashira coupling. Synlett. 2009(16):2601–2604.
[23] Bakherad M, Keivanloo A, Hashemi M. One-pot synthesis of 2-substituted imidazo[2,1-*b*][1,3]benzothiazoles *via* coupling-cyclization under Pd-Cu catalysis in water. Lett in Org Chem. 2009;6:165–170.
[24] Aggarwal R, Sumran G, Singh SP, Saini A. Hypervalent iodine oxidation of benzil-α-arylimino oximes: An efficient synthesis of 2,3-diphenylquinoxaline-1-oxides. Tetrahedron Lett. 2006;47:4969–4971.
[25] Aggarwal R, Kumar R, Kumar V. A facile and rapid one-pot synthesis of 1,4-diaryl-2-mercaptoimidazoles under solvent-free conditions. J Sulfur Chem. 2007;28:617–623.
[26] Aggarwal R, Sumran G, Garg N, Aggarwal A. A regioselective synthesis of some new pyrazol-1'-ylpyrazolo[1,5-*a*]pyrimidines in aqueous medium and their evaluation as antimicrobial agents. Eur J Med Chem. 2011;46:3038–3046.
[27] Aggarwal R, Kumar V, Bansal A, Sanz D, Claramunt RM. Multi-component solvent-free versus stepwise solvent mediated reactions: regiospecific formation of 6-trifluoromethyl and 4-trifluoromethyl-1*H*-pyrazolo[3,4-*b*]pyridines. J Fluorine Chem. 2012;140:31–37.
[28] Aggarwal R, Rani C, Sumran G. Efficient synthesis of some new 2,3-dimethylquinoxaline-1-oxides using bis(acetoxy)phenyl-λ^3-iodane as an oxidant. Synth Commun. 2014;44:923–928.
[29] Aggarwal R, Rani C, Sadana R, Busari K, Sharma C, Aneja KR. Hypervalent iodine mediated synthesis of some new *N*-(4,6-dimethylpyrimidin-2-yl)-*N*′-(4-arylthiazol-2-yl)hydrazines and their evaluation as antibacterial and cytotoxic agents. Chem Biol Interface. 2013;3(6):389–397.
[30] Aggarwal R, Kumar R. Novel and expedient regioselective synthesis of 2-imino-3-methyl-2,3-dihydrothiazoles. Synth Commun. 2008;38:2096–2102.
[31] Prakash O, Wadhwa D, Hussain K, Kumar R. [Hydroxy(tosyloxy)iodo]benzene in organic synthesis: a facile synthesis of furo[3,2-*c*]coumarins using α-tosyloxyketones. Synth Commun. 2012;42:2947–2951.
[32] Nicolaou KC, Montagnon T, Ulven T, Baran PS, Zhong YL, Sarabia F. Novel chemistry of α-tosyloxy ketones: applications to the solution- and solid-phase synthesis of privileged heterocycle and enediyne libraries. J Am Chem Soc. 2002;124:5718–5728.
[33] Stuckawisch CG. Derivatives of 2-amino-6-methoxybenzothiazole. J Am Chem Soc. 1949;71:3417–3417.