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A facile [hydroxy(tosyloxy)iodo]benzene mediated synthesis of 2-arylimidazo[1,2-*a*]pyrimidines and their conversion into 3-bromo-2-arylimidazo[1,2-*a*]pyrimidines

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α-Tosyloxyketone **2**, obtained through hypervalent iodine oxidation of enolizable ketones using [hydroxy(tosyloxy)iodo]benzene (HTIB) in acetonitrile, on treatment with 2-aminopyrimidine **3** generates regioselectively 2-arylimidazo[1,2-*a*]pyrimidine **6** which upon subsequent treatment with bromine undergoes electrophilic substitution at position-3 to yield 3-bromo-2-arylimidazo[1,2-*a*]pyrimidine **8**.

Keywords: α-Tosyloxyketones, [hydroxy(tosyloxy)iodo]benzene, 2-arylimidazo[1,2-*a*]pyrimidines, electrophilic substitution, 3-bromo-2-arylimidazo[1,2-*a*]pyrimidines

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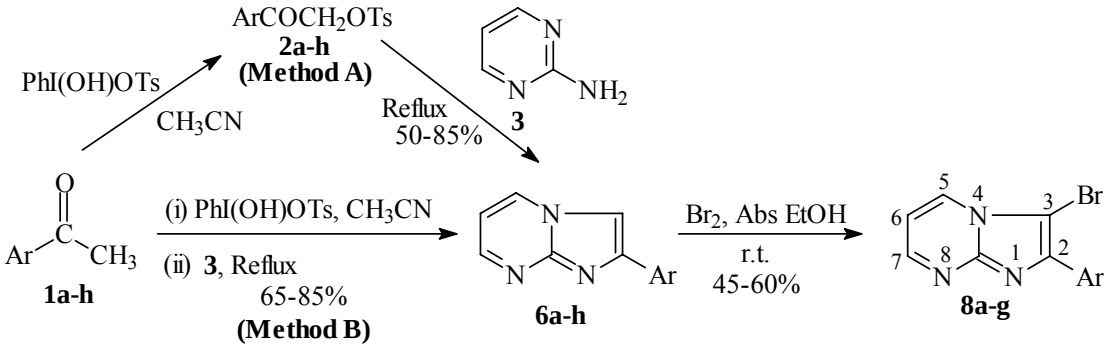
[Hydroxy(tosyloxy)iodo]benzene (HTIB or Koser's reagent) is an extremely useful reagent in organic synthesis[1]. An important application of HTIB, α-tosyloxylation of enolizable carbonyl compounds, was first reported in 1982 by Koser *et al*[2]. Recent work has shown that α-tosyloxyketones can offer a superior alternative to the existing synthesis of a variety of heterocycles involving highly lachrymatory α-haloketones[3-5]. During the literature survey, imidazo[1,2-*a*]pyrimidines attracted the authors' attention as these are a class of active compounds currently employed in the field of medicinal chemistry for their remarkable anthelmintic, bronchiodilatory[6], antithrombotic, cardiovascular[7], antifungal[8], antibacterial and antiprotozoal[9] properties, and also as potent GnRH receptor antagonists[10], benzodiazepine[11] and $GABA_A$ receptor agonists[12].

The synthesis of imidazo[1,2-*a*]pyrimidines has been widely investigated[13-21] and one of the most common routes for synthesis of imidazo[1,2-*a*]pyrimidines involves the condensation of 2-aminopyrimidine with α-halocarbonyl compounds[13–16]. However, the generality of this reaction is somewhat limited by the difficulty in preparation, purification, toxicity and lachrymatory property of α-halocarbonyl compounds. Keeping this in mind and as a part of the ongoing comprehensive programme to explore the utility of HTIB[22-24] for the synthesis of various types of heterocyclic compounds associated with a wide range of biological activity, it is now desired to report a new, facile and efficient method for the synthesis of 2-arylimidazo[1,2-*a*]pyrimidines **6** involving the intermediacy of α-tosyloxyketones **2**.

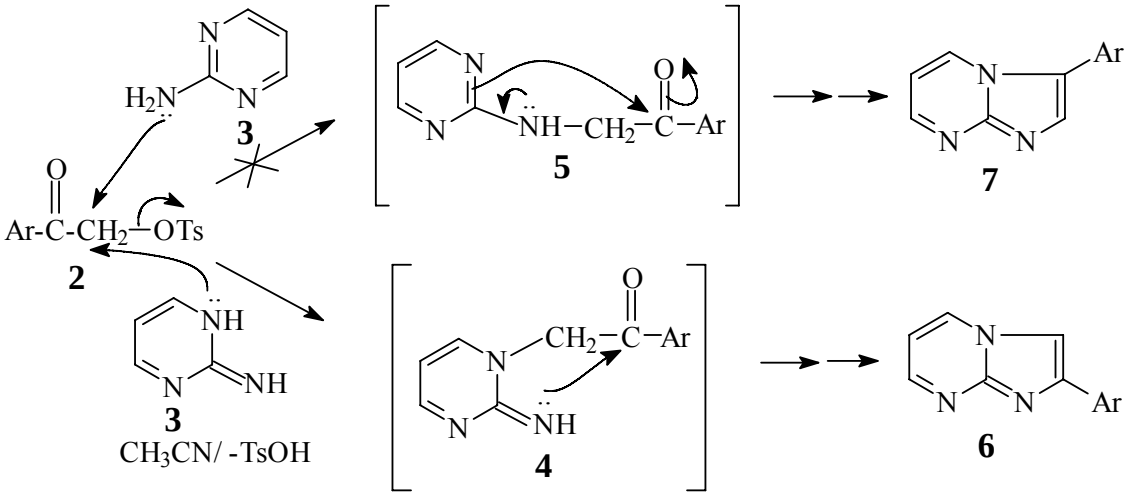
Results and Discussion

The synthesis of 2-arylimidazo[1,2-*a*]pyrimidines **6** can be achieved by two alternative methods as outlined in **Scheme I**. The synthesis was first attempted in a stepwise manner **(Method A)** involving the isolation of intermediate α-tosyloxyketones **2**, which are stable and crystalline solids. Initially, enolizable ketones **1** were made to react with HTIB to yield α-tosyloxyketones **2a-h** according to the literature procedure[25,26]. **2** on treatment with 2-aminopyrimidine **3** in acetonitrile resulted in the formation of a single isomer, which may be either 2-arylimidazo[1,2-*a*]pyrimidines **6** or 3-arylimidazo[1,2-*a*]pyrimidines **7**. The formation of structural isomers **6** and **7** could be expected *via* the intermediates **4** and **5**, which may be obtained in principle by the initial nucleophilic attack of the ring nitrogen or amino group on the α-tosyloxyketones, respectively **(Scheme II)**. Though comparison of the melting points of the products obtained with those reported in the literature indicated the formation of



where Ar, **a**= C_6H_5
b= 4-ClC_6H_4
c= 4-BrC_6H_4
d= 4-FC_6H_4
e= 3-$NO_2C_6H_4$
f= 4-$OCH_3C_6H_4$
g= 4-$CH_3C_6H_4$
h= 4-$NO_2C_6H_4$

Scheme I



Scheme II

isomer **6**, but there was no spectral or chemical evidence in the literature to differentiate isomer **6** from **7**. In order to distinguish between the isomers **6** and **7**, ^{1}H NMR spectra of the products were recorded. ^{1}H NMR spectra of the fused heterocyclic compounds displayed three double doublets at δ 6.7-6.9, 8.3-8.5, 8.3-8.8 in ABX pattern, due to H-6, H-5, H-7 (pyrimidine ring), respectively along with their coupling constant values $J_{5,6}$ = 6.6, $J_{6,7}$ = 4.2, $J_{5,7}$ = 2.1 Hz and a characteristic methine proton (=CH) resonance signal as a sharp singlet for one proton at δ 7.64-7.79 for 2-H or 3-H (imidazole ring) of

imidazo[1,2-*a*] pyrimidine system. However, ^{1}H NMR proved to be of little help in assigning the structures **6** or **7** to the products obtained as the value of imidazole-H singlet (δ 7.64-7.79) is very close to the value δ 7.77 (2-H) and 7.66 (H-3) reported for the parent imidazo[1,2-*a*]pyrimidine ring[27].

To overcome this problem, it was planned to undertake electrophilic substitution reaction such as bromination on the aryl-substituted imidazo[1,2-*a*]pyrimidines. Imidazo[1,2-*a*]pyrimidines are condensed aromatic system with ten delocalised π-electrons and are highly susceptible to electrophilic substitution at position-3 as predicted by HMO frontier-electron density calculations[27]. Bromination of the aryl-substituted imidazo[1,2-*a*]pyrimidines was thus carried out with 1.2 moles of Br_2 in absolute ethanol at RT and a monobrominated product **8** was obtained. ^{1}H NMR spectra of **8** exhibited the disappearance of the sharp singlet for imidazole-H at δ 7.74, while the splitting pattern of the pyrimidine ring remained unaltered. Disappearance of the singlet at δ 7.74 confirmed that bromination has occurred at position-3 and thus aryl group was present at position-2 of imidazopyrimidine ring. It can be claimed with certainity that the structural isomer formed during the present reaction was 2-arylimidazo[1,2-*a*]pyrimidines **6**. Had it been the 3-aryl isomer **7**, the bromination would not have taken place so easily as the frontier electron density at C-2 is 0.078, which is much less as compared to C-3 (0.543) of the parent imidazo[1,2-*a*]pyrimidine[27].

Encouraged by the results of the stepwise procedure, the one pot synthesis was further attempted **(Method B)** to obtain 2-arylimidazo[1,2-*a*]pyrimidines **6** starting from enolizable ketones. Thus, in this procedure, ketones **1** were oxidized with HTIB in CH_3CN, the resulting α-tosyloxyketones (*in situ*) on treatment with 2-aminopyrimidine generated **6** in excellent yields.

Conclusion

Finally, the present work is significant since the synthesis of several biologically active imidazo[1,2-*a*]pyrimidines can be achieved through an eco-friendly method directly from ketones without using the toxic and lachrymatory α-haloketones. Electrophilic substitution on the imidazo[1,2-*a*]pyrimidine ring provides a useful tool to differentiate between 2- or 3-aryl-substituted isomers. Lastly, the method extends the versatility of HTIB as an oxidizing agent in the synthesis of bridgehead nitrogen heterocycles.

Experimental Section

Melting points were determined in open capillaries using a melting point apparatus and are uncorrected. RT and m.p. represent room temperature and melting point, respectively. The IR spectra of the compounds were recorded on a Buck Scientific IR M-500 spectrometer using KBr pellets, ^{1}H and ^{13}C NMR spectra in $CDCl_3$ on a Bruker instrument at 300 MHz and 75 MHz, respectively. Chemical shifts are expressed in δ-scale downfield from TMS as an internal standard. Elemental microanalysis were performed at RSIC, CDRI, Lucknow. All the new compounds **8a-g** gave satisfactory elemental analyses.

Preparation of 2-arylimidazo[1,2-*a*]pyrimidines, 6a-h.

Method A: Step 1: Synthesis of α-tosyloxyketones, ***2a-h.***

To a solution of acetophenone **1a** (0.6 g, 5 mmol) in acetonitrile (20 mL) was added HTIB (1.95 g, 5 mmol) and the reaction mixture was refluxed for 3 h. The excess of solvent was distilled off and the residual mass was purified by recrystallization from ethanol. The solid was filtered and again washed with cold ethanol to give **2a**, yield (1.05 g, 72%), m.p. 90°C (lit.[26] m.p. 90°C).

Thus, all α-tosyloxyketones were prepared according to the literature procedure[2, 26].

Step 2: Synthesis of 2-arylimidazo[1,2-a]pyrimidines, ***6a-h;*** *General procedure:*

Typically, to a solution of 2-aminopyrimidine **3** (0.65 g, 6.8 mmol) in acetonitrile (30 mL) was added (1.71 mmol) of appropriate α-tosyloxyketones **2**. The heterogenous mixture was heated to reflux for 6 h. Progressive TLC monitoring of the reaction mixture showed the formation of a single isomer. 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 give **6**.

Method B: One-pot synthesis of 6a-h from ketones 1.

To a mixture of **1a** (0.6 g, 5 mmol) and acetonitrile (20 mL) was added HTIB (1.95 g, 5 mmol) and the resulting reaction mixture was refluxed for 2 h. The formation of α-tosyloxyketone **2** was confirmed by TLC monitoring of the reaction mixture. Then was added dropwise a solution of **3** (2.0 g, 20 mmol) in

acetonitrile (10 mL). The resultant reaction mixture was heated under reflux for 6 h. The solid obtained after cooling was filtered, neutralized with aq. sodium bicarbonate solution and washed with water to give **6a**, yield (0.7 g, 72%), m.p. 200°C (lit.[15] m.p. 202°C).

Adopting the same procedure, all other derivatives of title compounds **6** were synthesized. The isolated yields obtained by **Method B** are given in parenthesis.

6a: yield 50% [72%]; m.p. 200°C (lit.[15] m.p. 202°C).

6b: yield 67% [75%]; m.p. 272°C (lit.[15] m.p. 274°C).

6c: yield 65% [78%]; m.p. 278°C (lit.[15] m.p. 279°C).

6d: yield 59% [66%]; m.p. 235°C (lit.[15] m.p. 238°C).

6e: yield 72% [72%]; m.p. 240°C; IR (KBr): 3075 (aromatic C-H), 1580 (NO_2), 1511 (C-N), 1342 cm^{-1} (NO_2); ^1H NMR ($CDCl_3$): δ 6.85-6.89 (dd, 1H, *J*=6.6 Hz, *J*=4.2 Hz, H-6), 7.57-7.60 (m, 1H, Ph-5'-H), 7.90 (s, 1H, H-3), 8.20-8.27 (m, 1H, Ph-6'-H), 8.37-8.43 (m, 2H, Ph-2', 4'-H), 8.53-8.55 (dd, 1H, *J*=6.6 Hz, *J*=2.1 Hz, H-5), 8.73-8.80 (dd, 1H, *J*=3.9 Hz, *J*=2.1 Hz, H-7); ^{13}C NMR ($CDCl_3$): δ 107.12 (C-3), 109.35 (C-6), 120.88, 123.10 (2C, Ph-2', 4'), 129.55, 129.85 (2C, Ph-1', 5'), 132.19 (C-2), 133.32 (Ph-6'), 146.10 (C-5), 147.82 (C-7), 148.83 (C-8a), 150.81 (Ph-3').

6f: yield 52% [65%]; m.p. 194°C; IR (KBr): 3134 (aromatic C-H), 1615 (C=C and C=N), 1515 (C-N), 1244 cm^{-1} (C-O of OCH_3); ^1H NMR ($CDCl_3$): δ 3.78 (s, 3H, OCH_3), 6.73-6.76 (dd, 1H, *J*=6.6 Hz, *J*=4.2 Hz, H-6), 6.89-6.92 (d, 2H, *J*=8.7 Hz, Ph-3', 5'-H), 7.65 (s, 1H, H-3), 7.86-7.89 (d, 2H, *J*=8.7 Hz, Ph-2', 6'-H), 8.31-8.34 (dd, 1H, *J*=6.6 Hz, *J*=1.8 Hz, H-5), 8.40-8.42 (dd, 1H, *J*=3.9 Hz, *J*=1.8 Hz, H-7); ^{13}C NMR ($CDCl_3$): δ 55.33 (OCH_3), 105.19 (C-3), 108.57 (C-6), 114.18 (2C, Ph-3', 5'), 125.77 (Ph-1'), 127.59 (2C, Ph-2', 6'), 132.72 (C-2), 147.36 (C-5), 148.68 (C-7), 149.40 (C-8a), 160.08 (Ph-4').

6g: yield 73% [75%]; m.p. 222-224°C; IR (KBr): 3129 (aromatic C-H), 1612 (C=C and C=N), 1513 cm^{-1} (C-N); ^1H NMR ($CDCl_3$): δ 2.31 (s, 3H, CH_3), 6.70-6.74 (dd, 1H, *J*=6.9 Hz, *J*=4.2 Hz, H-6), 7.15-7.18 (d, 2H, *J*=8.1 Hz, Ph-3', 5'-H), 7.68 (s, 1H, H-3), 7.81-7.84 (d, 2H, *J*=8.1 Hz, Ph-2', 6'-H), 8.31-8.33 (dd, 1H, *J*=6.8 Hz, *J*=2.1 Hz, H-5), 8.39-8.41 (dd, 1H, *J*=4.2 Hz, *J*=2.1 Hz, H-7); ^{13}C NMR ($CDCl_3$): δ 19.91 (CH_3), 104.35 (C-3), 107.17 (C-6), 124.71 (2C, Ph-2', 6'), 128.02 (2C, Ph-3', 5'), 128.84 (Ph-1'), 131.43 (C-2), 137.09 (Ph-4'), 146.0 (C-5), 147.21 (C-7), 148.10 (C-8a).

6h: yield 85% [85%]; m.p. >370°C (lit.[13,14] m.p. >370°C).

Synthesis of 3-bromo-2-arylimidazo[1,2-*a*]pyrimidines, 8a-g.

To a stirred solution of compound **6a** (0.19 g, 1 mmole) in absolute EtOH (5 mL) was added dropwise a solution of Br_2 (0.19 g, 1.2 mmole) in absolute EtOH (6 mL). When the bromine color no longer faded, addition was stopped, and the mixture was stirred for additional 5 min. The colourless precipitated solid, **8a**-HBr, was filtered and rinsed with EtOH. Concentration of the filtrate gave a second crop. The free base was obtained by treating an aqueous solution of compound **8a**-HBr with saturated aq. solution of $NaHCO_3$. (Yields, m.p.'s, analyses and ^{13}C NMR spectral data of **8a-g** are given in **Table I** and **Table II** and ^1H NMR spectral data are given below).

8a: ^1H NMR ($CDCl_3$): δ 6.91-6.94 (dd, 1H, *J*=6.6 Hz, *J*=4.2 Hz, H-6), 7.31-7.44 (m, 3H, Ph-4', 3', 5'-H), 8.13-8.16 (d, 2H, *J*=8.1 Hz, Ph-2', 6'-H), 8.37-8.40 (dd, 1H, *J*=6.6 Hz, *J*=2.1 Hz, H-5), 8.51-8.53 (dd, 1H, *J*=4.2 Hz, *J*=2.1 Hz, H-7).

8b: ^1H NMR ($CDCl_3$): δ 6.92-6.96 (dd, 1H, *J*=6.9 Hz, *J*=4.2 Hz, H-6), 7.36-7.39 (d, 2H, *J*=8.4 Hz, Ph-2', 6'-H), 8.08-8.11 (d, 2H, *J*=8.7 Hz, Ph-3', 5'-H), 8.36-8.39 (dd, 1H, *J*=6.9 Hz, *J*=2.1 Hz, H-5), 8.50-8.52 (dd, 1H, *J*=4.2 Hz, *J*=1.8 Hz, H-7).

8c: ^1H NMR ($CDCl_3$): δ 6.94-6.97 (dd, 1H, *J*=6.9 Hz, *J*=4.2 Hz, H-6), 7.53-7.56 (d, 2H, *J*=8.7 Hz, *J*=1.8 Hz, Ph-2', 6'-H), 8.03-8.06 (d, 2H, *J*=8.7 Hz, *J*=1.8 Hz, Ph-3', 5'-H), 8.38-8.41 (dd, 1H, *J*=6.9 Hz, *J*=1.8 Hz, H-5), 8.52-8.54 (dd, 1H, *J*=4.2 Hz, *J*=1.8 Hz, H-7).

8d: ^1H NMR ($CDCl_3$): δ 6.92-6.96 (dd, 1H, *J*=6.9 Hz, *J*=4.2 Hz, H-6), 7.07-7.13 (t, 2H, *J*=8.7 Hz, Ph-3', 5'-H), 8.11-8.16 (dd, 2H, *J*=9.0 Hz, *J*=5.4 Hz, Ph-2', 6'-H), 8.37-8.40 (dd, 1H, *J*=6.9 Hz, *J*=1.8 Hz, H-5), 8.50-8.52 (dd, 1H, *J*=3.9 Hz, *J*=2.1 Hz, H-7).

8e: ^1H NMR ($CDCl_3$): δ 7.00-7.04 (dd, 1H, *J*=6.6 Hz, *J*=4.2 Hz, H-6), 7.58-7.64 (m, 1H, Ph-5'-H), 8.18-8.21 (m, 1H, Ph-6'-H), 8.44-8.52 (m, 2H, Ph-2', 4'-H), 8.59-8.60 (dd, 1H, *J*=6.6 Hz, *J*=2.1 Hz, H-5), 9.04-9.05 (dd, 1H, *J*=3.9 Hz, *J*=2.1 Hz, H-7).

8f: ^1H NMR ($CDCl_3$): δ 3.80 (s, 3H, OCH_3), 6.90-6.92 (dd, 1H, *J*=6.6 Hz, *J*=4.2 Hz, H-6), 6.94-6.97 (d, 2H, *J*=8.7 Hz, Ph-3', 5'-H), 8.10-8.13 (d, 2H, *J*=8.7 Hz, Ph-2', 6'-H), 8.36-8.39 (dd, 1H, *J*=6.6 Hz, *J*=2.1 Hz, H-5), 8.49-8.51 (dd, 1H, *J*=4.2 Hz, *J*=2.1 Hz, H-7).

Table I — Characterization data of 3-bromo-2-arylimidazo[1,2-*a*]pyrimidines, **8a-g**

Compd	m.p. (°C)	Yield (%)	Mol. formula	Found (Calcd)%		
				C	H	N
8a	142-44	52%	$C_{12}H_8BrN_3$	52.72 (52.55	3.12 2.91	15.21 15.32)
8b	208-10	45%	$C_{12}H_7BrClN_3$	46.63 (46.67	2.33 2.26	13.72 13.61)
8c	210-12	48%	$C_{12}H_7Br_2N_3$	40.69 (40.79	1.78 1.98	11.71 11.89)
8d	186-88	45%	$C_{12}H_7BrFN_3$	49.42 (49.31	2.21 2.39	14.21 14.38)
8e	208	47%	$C_{12}H_7BrN_4O_2$	45.33 (45.14	2.21 2.19	17.68 17.55)
8f	170-72	60%	$C_{13}H_{10}BrN_3O$	51.21 (51.31	3.24 3.28	13.81 13.81)
8g	180-82	60%	$C_{13}H_{10}BrN_3$	54.21 (54.16	3.63 3.47	14.72 14.58)

Table II — ^{13}C NMR chemical shifts (δ, ppm) of 3-bromo-2-arylimidazo[1,2-*a*]pyrimidines, **8a-g**

Compd	C-2	C-3	C-5	C-6	C-7	C-8a	Others
8a	131.48	90.36	144.11	109.40	148.13	150.21	Ph-1': 132.18; Ph-2', 6': 128.50; Ph-3', 5': 128.05; Ph-4': 128.84
8b	131.47	90.39	143.08	109.51	148.15	150.44	Ph-1': 130.71; Ph-2', 6': 129.26; Ph-3', 5': 128.75; Ph-4': 134.85
8c	131.48	90.66	143.18	109.52	148.19	150.47	Ph-1': 131.18; Ph-2', 6': 129.54; Ph-3', 5': 131.73; Ph-4': 123.22
8d	131.47	90.04	143.35	109.45	148.13	150.30	Ph-1': 128.34; Ph-2', 6': 129.88-129.99 (d, ^3J=8.25 Hz); Ph-3', 5': 115.41-115.70 (d, ^2J=21.75 Hz); Ph-4': 161.47-164.76 (q, ^1J=246.75 Hz)
8e	131.82	91.87	143.1	109.99	148.21	149.18	Ph-1, 5': 130.21; Ph-2', 4': 123.42; Ph-3': 151.24; Ph-6': 133.79
8f	130.21	88.38	143.07	108.15	147.09	148.80	OCH_3: 54.31; Ph-1': 123.78; Ph-2', 6': 128.39; Ph-3', 5': 112.93; Ph-4': 159.13
8g	131.39	90.06	144.27	109.36	148.07	150.12	CH_3: 21.41; Ph-1': 129.28; Ph-2', 6': 128.00; Ph-3', 5': 129.21; Ph-4': 138.98

8g: ^{1}H NMR ($CDCl_3$): δ 2.44 (s, 3H, CH_3), 7.00-7.04 (dd, 1H, *J*=6.9 Hz, *J*=4.2 Hz, H-6), 7.31-7.34 (d, 2H, *J*=8.1 Hz, Ph-3', 5'-H), 8.13-8.16 (d, 2H, *J*=8.1 Hz, Ph-2', 6'-H), 8.47-8.49 (dd, 1H, *J*=6.6 Hz, *J*=2.1 Hz, H-5), 8.59-8.61 (dd, 1H, *J*=4.2 Hz, *J*=2.1 Hz, H-7).

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