One-pot synthesis of thiazolo[3,2-α]pyridine derivatives catalysed by ionic liquids

Hai-Liang Chen and Hong-Yun Guo*

College of Chemical Engineering and Materials, Zhejiang University of Technology, Hangzhou 310014, P. R. China

A green method for the synthesis of thiazolo[3,2-α]pyridine derivatives via the coupling of malononitrile, an aryl aldehyde and methyl thioglycolate in an ionic liquid has been developed. The advantages of this protocol are that it is non-toxic, no by-products are formed, short reaction times are required and high yields are obtained. Thiazolo [3,2-α]pyridines have important biological and medical applications.

Keywords: one-pot synthesis, ionic liquids, thiazolo[3,2-a]pyridine

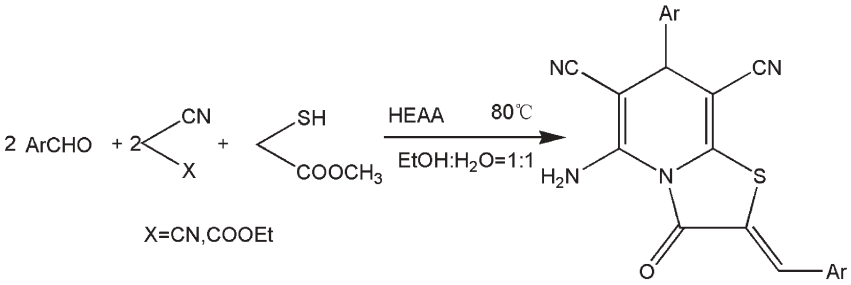
Thiazolo[3,2-α]pyridines are an important class of compounds possessing notable biological activites including the inhibition of beta-amyloid production,[1] potent CDK2-cyclin A inhibition,[2] α-glucosidase inhibitor,[3] potent uterine stimulation,[4] together with antibacterial and antifungal activities.[5,6] Therefore, the synthesis of thiazolo[3,2-α]pyridines has attracted much attention, and numerous procedures have been developed[7,8]. However these methods all have drawbacks, so the attempts to develop green and facile approaches are important.

Recently, room temperature ionic liquids have attracted interest due to their unique properties including non-volatility, high ionic conductivity, nonflammability, high thermal stability, wide electrochemical window and their recyclable uses. Ionic liquids have been used in organic reactions such as the Friedel–Crafts reaction,[9] Diels–Alder reaction,[10] Heck reaction,[11] Pechmann condensations,[12] Biginelli reaction,[13] Beckmann rearrangement[14] and other reactions.[15] As part of our research on thiazolo[3,2-α]pyridine derivatives, we report a green synthesis of these compounds by coupling malononitrile, aromatic aldehydes and methyl thioglycolate in an ionic liquid.

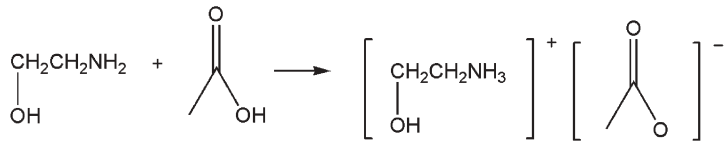
First, the three-component reaction of benzaldehyde (**1a**, 2 mmol), malononitrile (**2**, 2 mmol) and methyl thioglycolate (**3**, 1 mmol)was carried out in different solvents in the presence of HEAA(2-hydroxyethylammonium acetate) under conventional heating and the reaction was followed by TLC. As shown in Table 1, various solvents were screened for their efficiency in the reaction. Among them, ethanol:H_2O was superior to other solvents and was chosen for the further investigations of the reaction.

The optimised reaction conditions were then tested for the construction of a library with 14 aldehydes and two acetonitrile derivatives (malonitrile and ethyl cyanoacetate). The corresponding fused thiazolo[3,2-α]pyridine derivatives were obtained in good yields. The aldehydes, acetonitrile derivatives and methyl thioglycolate were all commercially available materials. The results are summarised in Table 2.

This procedure was applied to aryl aldehydes with either electron-donating groups(hydroxyl, alkoxyl) or electron-withdrawing groups(nitro, halide) and heterocyclic aldehydes. It can be seen from Table 2 that the electronic effect of the



1 **2** **3** **4**



HEAA 2-hydroxyethyl ammonium acetate

Scheme 1

* Correspondent. E-mail: guohy63@163.com

Table 1 Reaction conditions optimisation for the synthesis of **4a**

Entry	Solvent	Temp./°C)	Time/h	Yield[a]/%
1	acetonitrile	r.t.	4	trace
2	piperidine	r.t.	3	66
3	H_2O	r.t.	3	34
4	DMF	r.t.	0.5	70
5	Glycol	r.t.	0.5	55
6	EtOH	r.t.	0.5	76
7	EtOH:H_2O=1:1	r.t.	0.5	82
8	EtOH:H_2O=1:1	60	0.5	83
9	EtOH:H_2O=1:1	80	0.5	86
10	EtOH:H_2O=1:1	100	0.5	81

[a] Isolated yield.

substituted benzaldehydes play an important role in the reaction. Substrates bearing electron-withdrawing groups have higher reactivity (higher yields and shorter reaction time) than those bearing electron-donating groups. In this investigation, all the products were characterised by m.p.., IR, ^{1}H NMR.

Finally, we examined the ionic liquid (HEAA). Because of the possible toxicity of organic catalytic agents, we chose the ionic liquid (HEAA) to catalyse the reaction. Therefore, we tested the recovery and reuse of the ionic liquid. As shown in Table 3, the ionic liquid HEAA could be successfully recovered and reused four times without any notable loss in catalytic activity.

A plausible mechanism for the formation of compounds **4** is suggested in Scheme 2. Firstly, the nucleophilic addition of methyl thioglycolate 3 to the acetonitrile derivative **2** yielded the intermediate **5**, which was converted to the thiazolinone derivatives **6** via intramolecular dehydration. Then, the intermediate **6** underwent a Michael addition with **7** which was formed from Knoevenagel condensation of **1** and **2**. After the open-chain intermediate **8** was formed, a series of intramolecular cyclisations and isomerisations occurred to form compound **10**. Finally, compound **4** was formed by another Michael addition between 1 and 10.

Table 2 Synthesis of thiazolo[3,2-α]pyridine derivatives **4** using HEAA as catalyst in ethanol

Entry	Ar	X	Products	Time/h	Yield[a]/%
1	C_6H_5-	CN	**4a**	0.5	89
2	4-CH_3-C_6H_4-	CN	**4b**	0.7	90
3	4-NO_2-C_6H_4-	CN	**4c**	1.5	78
4	2-Cl-C_6H_4-	CN	**4d**	0.4	86
5	4-Cl-C_6H_4-	CN	**4e**	1.0	76
6	4- OCH_3-C_6H_4-	CN	**4f**	1.5	77
7	4-F-C_6H_4-	CN	**4g**	0.5	91
8	4-OH-C_6H_4-	CN	**4h**	0.5	86
9	3-Br-C_6H_3-	CN	**4i**	1.5	73
10	4-OH-3-OCH_3-C_6H_3-	CN	**4j**	0.6	82
11	2,4- Cl_2-C_6H_3-	CN	**4k**	0.5	84
12	2-NO_2-C_6H_4-	CN	**4l**	0.5	79
13	2-Furyl-	CN	**4m**	3	78
14	n-Butyl-	CN	**4n**	12	NR
15	4-Cl-C_6H_4-	COOEt	**4o**	5	77
16	4-F-C_6H_4-	COOEt	**4p**	5.5	73

[a] Isolated yield.

Table 3 Studies on the reuse of HEAA in the preparation of **4a**

Round	1	2	3	4	5
Yield (%)	93	91	88	85	83

In conclusion we have developed an efficient protocol for the synthesis at highly functionalised thiazolo[3,2-α]pyridine derivatives which are of great interest because of their biological activity. The procedure has several advantages including higher yields, shorter reaction time, no side-product, minimal environmental impact, and is a useful and attractive method for the synthesis of those compounds.

Experimental

Melting points were determined with a X-4 microscopic melting-point apparatus and were uncorrected. IR spectra were recorded on a Nexus 670 spectrometer in KBr. ^{1}H NMR were measured on a Bruker Avance-II 500 MHz spectrometer using TMS as internal standard and DMSO-d_6 as solvent.

The synthesis of HEAA was carried out by a similar method to the literature.[16] The ionic liquid was formed quantitatively and in high purity as assessed by ^{1}H NMR. All other chemicals (AR grade) were commercially available and were used without further purification.

Synthesis of thiazolo[3,2-α]pyridine derivatives **4***; general procedure*
The mixture of the aromatic aldehyde **1** (2 mmol), malononitrile **2** (2 mmol), methyl thioglycolate **3** (1 mmol), HEAA (0.1 mmol) in EtOH:H_2O=1:1 (5 mL) was stirred at 80 °C for the appropriate time (monitored by TLC). After the reaction was finished, the mixture was cooled and poured into ice water (10 mL). The product was collected by filtration and recrystallised from EtOH. The filtrate was extracted with diethyl ether several times to retrieve the HEAA for subsequent use.

5-Amino-2,3-dihydro-3-oxo-7-phenyl-2-(phenylmethylene)-7H-thiazolo[3,2-a]pyridine-6,8-dicarbonitrile, (**4a**): Yellow powder; m.p. 249–250 °C (lit.[17] 245–248 °C); IR (KBr) ν_{max}: 3422, 3320, 3236, 2982, 2940, 2208, 1730, 1661 cm^{-1}; ^{1}H NMR (DMSO-d_6, 500 MHz) δ: 4.56 (s, 1H, pyridine-4 H), 7.34 (m, 1H, ArH), 7.38 (m, 4H, ArH), 7.50 (m, 1H, ArH), 7.52 (s, 2H, NH_2-H), 7.56 (m, 2H, ArH), 7.63 (m, 2H, ArH), 7.82 (s, 1H, olefin-H).

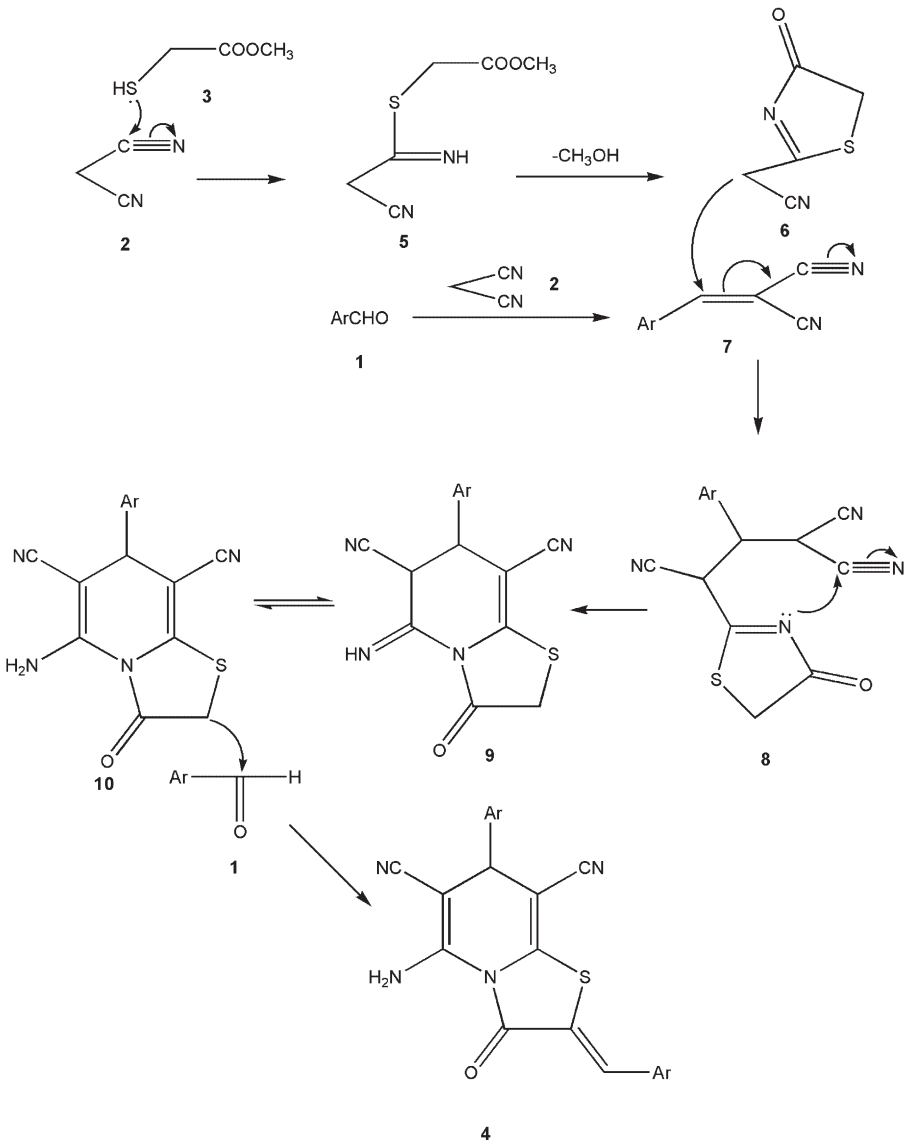
5-Amino-2,3-dihydro-7-(4-methylphenyl)-2-[(4-methylphenyl) methylene]-3-oxo-7H-thiazolo[3,2-a]pyridine-6,8-dicarbonitrile (**4b**): Yellow powder; m.p. 239–240 °C (lit.[18] 239–240 °C); IR (KBr) ν_{max}: 3420, 3380, 3270, 2960, 2940, 2200, 1740, 1650 cm^{-1}; ^{1}H NMR (DMSO-d_6, 500 MHz) δ: 2.32 (s, 3H, CH_3-H), 2.38 (s, 3H, CH_3-H), 4.55 (s, 1H, pyridine-4 H), 7.21 (d, 2H, ArH, J = 7.2 Hz), 7.28 (d, 2H, ArH, J = 7.2 Hz), 7.40–7.41 (m, 2H, NH_2-H), 7.54 (d, 4H, ArH, J = 7.2 Hz), 7.82 (s, 1H, olefin- H).

5-Amino-2,3-dihydro-7-(4-nitrophenyl)-2-[(4-nitrophenyl) methylene]-3-oxo-7H-thiazolo[3,2-a]pyridine-6,8-dicarbonitrile (**4c**): Yellow powder; m.p. 259–261 °C (lit.[18] 255–257 °C; IR (KBr) ν_{max}: 3380, 3282, 3075, 2855, 2204, 2198, 1727, 1656, 1561, 1520, 1410, 1339, 1169, 845 cm^{-1}; ^{1}H NMR (DMSO-d6, 500 MHz) δ: 4.94 (s, 1H, pyridine-4 H), 7.69 (s, 2H, NH_2-H), 7.76 (d, 2H, ArH, J = 8.8 Hz), 7.94 (d, 2H, ArH, J = 8.8 Hz), 8.00 (s, 1H, olefin-H), 8.30 (d, 2H, ArH, J = 8.8 Hz), 8.34 (d, 2H, ArH, J = 8.8 Hz).

5-Amino-7-(2-chlorophenyl)-2-[(2-chlorophenyl)methylene]-2, 3-dihydro-3-oxo-7H-thiazolo[3,2-a]pyridine-6,8-dicarbonitrile (**4d**): Yellow powder; m.p. 293–295 °C (lit.[19] 293–295 °C); IR (KBr) ν_{max}: 3434, 3402, 3270, 2960, 2956, 2212, 1742, 1650 cm^{-1}; ^{1}H NMR (DMSO-d_6, 500 MHz) δ: 4.48 (s, 1H, pyridine-4 H), 6.68 (s, 2H, NH_2-H), 7.24 (d, 2H, ArH, J = 8.5 Hz), 7.40 (d, 2H, ArH, J = 8.5 Hz), 7.47–7.53 (d, 4H, ArH, J = 8.5 Hz), 7.80 (s, 1H, olefin-H).

5-Amino-7-(4-chlorophenyl)-2-[(4-chlorophenyl)methylene]-2, 3-dihydro-3-oxo-7H-thiazolo[3,2-a]pyridine-6,8-dicarbonitrile (**4e**): Yellow powder; m.p. 254–255 °C (lit.[19] 253–254 °C); IR (KBr) ν_{max}: 3382, 2925, 2208, 1718, 1654, 1617, 1560, 1336, 1092, 823 cm^{-1}; ^{1}H NMR (DMSO-d_6, 500 MHz) δ: 4.69 (s, 1H, pyridine-4 H), 7.47 (d, 2H, ArH, J = 8.6 Hz), 7.51 (d, 2H, ArH, J = 8.6 Hz), 7.65 (d, 4H, ArH, J = 8.6 Hz), 7.71 (d, 2H, ArH, J = 8.6 Hz)7.60 (s, 2H, NH_2-H), 7.87 (s, 1H, olefin-H).

5-Amino-2,3-dihydro-7-(4-methoxyphenyl)-2-[(4-methoxyphenyl) methylene]-3-oxo-7H-thiazolo[3,2-a]pyridine-6,8-dicarbonitrile (**4f**): Yellow powder; m.p. 242–243 °C (lit.[19] 243–244 °C); IR (KBr) ν_{max}: 3463, 3350, 2982, 2950, 2208, 1720, 1662 cm^{-1}; ^{1}H NMR (DMSO-d_6, 500 MHz) δ: 3.78 (s, 3H, CH_3-H), 3.85 (s, 3H, CH_3-H), 4.54 (s, 1H, pyridine-4 H), 6.95 (d, 2H, ArH, J = 8.9 Hz), 7.15 (d, 2H, ArH, J = 8.9 Hz), 7.54 (d, 2H, ArH, J = 8.9 Hz), 7.63 (d, 2H, ArH, J = 8.9 Hz), 7.31-7.34 (m, 2H, NH_2-H), 7.82 (s, 1H, olefin-H).



Scheme 2

5-Amino-7-(4-fluorophenyl)-2-[(4-fluorophenyl)methylene]-2,3-dihydro-3-oxo-7H-thiazolo[3,2-a]pyridine-6,8-dicarbonitrile (**4g**): Yellow powder; m.p. 255–257 °C (lit.[18] 255–257 °C); IR (KBr) ν_{max}: 3420, 3379, 3270, 2940, 2936, 2200, 1740, 1660 cm^{-1}; ^{1}H NMR (DMSO-d$_6$, 500 MHz) δ: 4.67 (s, 1H, pyridine-4 H), 7.24–7.27 (t, 2H, ArH, J = 8.9 Hz), 7.42–7.46 (t, 2H, ArH, J = 8.9 Hz), 7.58 (s, 2H, ArH), 7.74–7.77 (m, 2H, ArH) 7.48–7.50 (m, 2H, NH$_2$-H), 7.89 (s, 1H, olefin-H).

5-Amino-2,3-dihydro-7-(4-hydroxyphenyl)-2-[(4-hydroxyphenyl)methylene]-3-oxo-7H-thiazolo[3,2-a]pyridine-6,8-dicarbonitrile (**4h**): Black powder; m.p 269–270 °C (lit.[1] 271–272 °C); IR (KBr) ν_{max}: 3430, 3415, 3270, 2950, 2930, 2208, 1743, 1637 cm^{-1}; ^{1}H NMR (DMSO-d$_6$, 500 MHz) δ: 4.46 (s, 1H, pyridine-4 H), 6.76–6.78 (d, 2H, ArH, J = 8.6 Hz), 6.96–6.98 (d, 2H, ArH, J = 8.7 Hz), 7.51–7.54 (t, 4H, ArH), 7.18–7.19 (d, 2H, NH$_2$-H), 7.76 (s, 1H, olefin-H), 9.51 (s, 1H, OH-H), 10.42 (s, 1H, OH-H).

5-Amino-7-(3-bromophenyl)-2-[(3-bromophenyl)methylene]-2,3-dihydro-3-oxo-7H-thiazolo[3,2-a]pyridine-6,8-dicarbonitrile (**4i**): Yellow powder; m.p. 261–263 °C (lit.[1] 261–262 °C); IR (KBr) ν_{max}: 3411, 3393, 3358, 2963, 2960, 2200, 1752, 1650 cm^{-1}; ^{1}H NMR (DMSO-d$_6$, 500 MHz) δ: 4.68 (s, 1H, pyridine-4 H), 7.36 (d, 1H, ArH, J = 8.3 Hz), 7.44 (d, 1H, ArH, J = 7.8 Hz), 7.54 (d, 2H, ArH, J = 8.3 Hz), 7.61–7.64 (t, 3H, ArH, J = 8.3 Hz), 7.70 (d, 2H, NH$_2$-H), 7.85 (s, 1H, ArH), 7.91 (s, 1H, olefin-H).

5-Amino-2,3-dihydro-7-(4-hydroxy-3-methoxyphenyl)-2-[(4-hydroxy-3-methoxyphenyl)methylene]-3-oxo-7H-thiazolo[3,2-a]pyridine-6,8-dicarbonitrile (**4j**): Blackish green powder, m.p. 277–278 °C (lit.[1] 278–279 °C); IR (KBr) ν_{max}: 3400, 3370, 3250, 2910, 2880, 2175, 1734, 1620 cm^{-1}; ^{1}H NMR (DMSO-d$_6$, 500 MHz) δ: 3.78 (s, 3H, CH$_3$-H), 3.84 (s, 3H, CH$_3$-H), 4.47 (s, 1H, pyridine-4 H), 6.78 (d, 2H, ArH, J = 8.3 Hz), 6.93 (d, 1H, ArH, J = 8.3 Hz), 7.00 (d, 1H, ArH, J = 8.3 Hz), 7.11 (d, 1H, ArH, J = 8.3 Hz), 7.27 (d, 1H, ArH, J = 8.3 Hz), 7.53 (s, 2H, NH$_2$-H), 7.78 (s, 1H, olefin-H), 9.07 (s, 1H, OH-H), 10.08 (s, 1H, OH-H).

5-Amino-7-(2,4-dichlorophenyl)-2-[(2,4-dichlorophenyl)methylene]-2,3-dihydro-3-oxo-7H-thiazolo[3,2-a]pyridine-6,8-dicarbonitrile (**4k**): Yellow powder; m.p. >300 °C (lit.[1] >300 °C); IR (KBr) ν_{max}: 3400, 3380, 3273, 2910, 2883, 2195, 1738, 1647 cm^{-1}; ^{1}H NMR (DMSO-d$_6$, 500 MHz) δ: 5.10 (s, 1H, pyridine-4 H), 7.54–7.56 (m, 2H, ArH, J = 8.4 Hz), 7.62 (d, 2H, ArH, J = 8.4 Hz) 7.68 (d, 2H, ArH, J = 8.4 Hz), 7.69–7.71 (m, 1H, olefin-H), 7.90–7.91 (m, 2H, NH$_2$-H).

5-Amino-2,3-dihydro-7-(2-nitrophenyl)-2-[(2-nitrophenyl)methylene]-3-oxo-7H-thiazolo[3,2-a]pyridine-6,8-dicarbonitrile (**4l**): Brown powder, m.p. 246–248 °C (lit.[1] 245–246 °C); IR (KBr) ν_{max}: 3480, 3454, 3267, 2960, 2940, 2213, 1740, 1670 cm^{-1}; ^{1}H NMR (DMSO-d$_6$, 500 MHz) δ: 5.18 (s, 1H, pyridine-4 H), 7.61–7.63 (m, 1H, ArH, J = 8.5 Hz), 7.65 (s, 2H, NH$_2$-H), 7.68–7.85 (m, 3H, ArH, J = 8.5 Hz), 7.91-8.25 (m, 3H, ArH, J = 8.5 Hz), 8.25 (s, 1H, ArH), 8.26-8.26 (m, 1H, olefin-H).

5-amino-7-(2-furanyl)-2-(2-furanylmethylene)-2,3-dihydro-3-oxo-7H-thiazolo[3,2-a]pyridine-6,8-dicarbonitrile (**4m**): Yellow powder; m.p. 266-268 °C (lit. [21] 266–268 °C); IR (KBr) ν_{max}: 3403, 3306, 3229, 2213, 1720, 1620 cm^{-1}; ^{1}H NMR (DMSO-d_6, 500 MHz) δ: 4.62 (s, 1H, pyridine-4 H), 6.36–6.39 (m, 2H, NH_2-H), 6.64–6.67 (m, 3H, furan-H), 6.88–7.58 (m, 3H, furan-H), 7.77 (m, 1H, olefin-H).

5-amino-7-(4-chlorophenyl)-2-[(4-chlorophenyl)methylene]-2,3-dihydro-3-oxo-7H-Thiazolo[3,2-a]pyridine-6,8-dicarboxylic acid 6,8-diethyl ester (**4o**): Yellow powder; m.p. 230–231 °C (lit.[20] 230–231 °C); IR (KBr) ν_{max}: 3430, 3387, 3320, 2910, 2883, 2195, 1738, 1647 cm^{-1}; ^{1}H NMR (DMSO-d_6, 500 MHz) δ: 1.19–1.22 (t, 3H, CH_3-H), 1.25–1.28 (t, 3H, CH_3-H), 4.07–4.11 (t, 2H, CH_2-H), 4.19–4.23 (m, 2H, CH_2-H), 4.91 (s, 1H, pyridine-4 H), 7.15 (d, 2H, ArH, J = 8.6 Hz), 7.19 (d, 2H, ArH, J = 8.6 Hz), 7.45 (d, 2H, ArH, J = 8.6 Hz), 7.56 (d, 2H, ArH, J = 8.6 Hz), 7.70 (s, 1H, olefin-H), 8.69 (s, 2H, NH_2-H).

5-Amino-7-(4-fluorophenyl)-2-[(4-fluorophenyl)methylene]-2,3-dihydro-3-oxo-7H-thiazolo[3,2-a]pyridine-6,8-dicarboxylic acid 6,8-diethyl ester (**4p**): Yellow powder; m.p. 225–226 °C (li.[20] 226–267 °C); IR (KBr) ν_{max}: 3390, 3382, 3200, 2916, 2874, 2210, 1743, 1650 cm^{-1}; ^{1}H NMR (DMSO-d_6,500MHz) δ: 1.18–1.21 (t, 3H, CH_3-H), 1.25–1.28 (t, 3H, CH_3-H), 4.07–4.11 (m, 2H, CH_2-H), 4.18–4.24 (m, 2H, CH_2-H), 4.92 (s, 1H, pyridine-4 H), 6.90–6.93 (t, 2H, ArH, J = 8.7 Hz), 7.17–7.21(dd, 4H, ArH, J_1 = 8.0, J_2 = 1.9 Hz), 7.63–7.65 (m, 2H, ArH), 7.72 (s, 1H, olefin-H), 8.68 (s, 2H, NH_2-H).

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References

1 M.J. Mullan, D. Paris and P. Bakshi, WO 2008070875, 2008, *Chem. Abstr.* 2008, **149**, 45259.
2 S. Vadivelan, B.N. Sinha, S.J. Irudayam and S.A.R.P. Jagarlapudi, *J. Chem. Inf. Model.*, 2007, **47**, 1526.
3 H. Park, K.Y. Hwang, K.H. Oh, Y.H. Kim, J.Y. Lee and K. Kim, *Bioorg. Med. Chem.*, 2008, **16**, 284.
4 F.M. Manhi and G.A. Soliman, *Chem. Abstr.*, 1993, **121**, 50029.
5 G. El-Hag Ali, A. Khalil, R. Lamphon and A. El-Maghraby, *Phosphorus, Sulfur Silicon Relat. Elem.*, 2005, **180**, 1909.
6 A.A. El-Maghraby, G.A.M. El-Hag Ali, A.H.A. Ahmed and M.S.A. El-Gaby, *Phosphorus, Sulfur Silicon Relat. Elem.*, 2002, **177**, 293.
7 F. Shi, C.M. Li, M. Xia, K.J. Miao, Y.X. Zhao, S.J. Tu, W.F. Zheng, G. Zhang and N. Ma *Bioorg. Med. Chem.*, 2009, **19**, 5565.
8 F.F. Abdel-latif, Y.S. Mohammed, H. Abdel-ghani, E.Kh. Ahmed and E.H. El-gawish, *Phosphorus, Sulfur, Silicon*, 1993, **78**, 251.
9 L. He, G.H. Tao, W.S. Liu, Xiong, W. Xiong, T. Wang and Y. Kou, *Chin. Chem. Lett*., 2006, **17**, 321.
10 T. Fischer, A. Sethi, T. Welton and J. Woolf. *Tetrahedron Lett.*, 1999, **40**, 793.
11 H. Hagiwara, Y. Shimizu, T. Hoshi, T. Suzuki, M. Ando, K. Ohkubo and C. Yokoyama, *Tetrahedron Lett*., 2001, **42**, 4349.
12 M.K. Potdar, S.S. Mohile and M.M. Salunkhe, *Tetrahedron Lett.*, 2001, **42**, 9285.
13 J. Peng and Y. Deng, *Tetrahedron Lett.*, 2001, **42**, 5917.
14 X. X. Xu, Z.L. Wei and X. Bo, *Chin. J. Org. Chem*., 2006, **3**, 354.
15 B. Wang, Y. Gu, C. Luo, T. Yang, L. Yang and J. Suo, *Tetrahedron Lett.*, 2004, **45**, 3417.
16 X.L. Yuan, S.J. Zhang and X.M. Lu, *J. Chem. Eng. Data*, 2007, **52,** 596.
17 H.M. Al-Matar, K.D. Khalil, H. Meier, H. Kolshorn and M. H. Elnagdi, *Arkivoc*, 2008, 288.
18 A.A. El-Maghraby, A.M. Gameel, E.H. Ali, A.H.A. Ahmed and M.S.A. El-Gaby, *Phosphorus, Sulfur Silicon*, 2002, **177**, 293.
19 F.F. Abdel-latif, Y.S. Mohammed, H. Abdel-ghani, E.Kh. Ahmed and E.H. El-gawish. *Phosphorus, Sulfur, Silicon*, 1993, **78**, 251.
20 M.H. Elnagdi, M.R.H. Elmoghayar, A.H.H. Elghandour and K. U. Sadek, *Egypt. Sulfur Lett*., 1989, **9**, 109.
21 M.H. Elnagdi, M.R.H. Elmoghayar, A.H.H. Elghandour and K.U. Sadek, *Coll. Czech. Chem. Commun.*, 1990, **55**, 745.