# A practical and rapid synthesis of 2-aryloxymethylene-6-arylimidazo [2,1-*b*][1,3,4]thiadiazole in aqueous media

**Xi-Cun Wang*, Man-Gang Wang, Zhi Yang, Zheng-Jun Quan, Fang Wang and Zheng Li**

*GanSu Key Laboratory of Polymer Materials, College of Chemistry and Chemical Engineering, Northwest Normal University, Lanzhou, Gansu, 730070, P. R. China*

A practical and rapid procedure is reported for the synthesis of a variety of 2-aryloxymethylene-6-arylimidazo[2,1-*b*] [1,3,4]thiadiazole **3a–m** by condensation reaction of 2-amino-5-aryloxymethylene-1,3,4-thiadiazole **1a–g** with ω-bromoacetophenone derivatives **2a–b** in aqueous media under microwave irradiation and yielded a series of novel compounds. The procedure is simple and the yields are good to excellent.

**Keywords:** imidazo[2,1-b][1,3,4]thiadiazole, aqueous media

In past years much effort has been devoted to the synthesis of imidazo[2,1-*b*][1,3,4]thiadiazole derivatives due to their diverse biological activity, such as anticancer,[1] antitubercular,[2] antibacterial,[3] antifungal[4] anticonvulsant, analgesic[5] and antisecretory[6] activities. Meanwhile, much interest has also been focused on the antiinflammatory,[7] cardiotonic,[8] diuretic,[9] herbicidal[10] activities displayed by compounds incorporating this heterocyclic system. Since the imidazo [2,1-*b*][1,3,4]thiadiazole system is similar in part to Levamisole, a well-known immunomodulator,[11] the possibility of reducing the harmful effects of the cytotoxic agents on the immune system also appears to be very challenging. These applications prompt us to develop a simple and rapid synthetic method to prepare a series of new compounds, with the object of obtaining new biological active compounds.

2,6-Disubstituted imidazo[2,1-b][1,3,4]thiadiazole and their derivatives have been synthesised by traditional methodology via several approaches, one of the more popular being the condensation of 2-amino-1,3,4-thiadiazole with ω-bromoacetophenone using acid as catalysis in ethanol solvent at reflux temperature for at least 12 h, giving only middle yield.[2] So, long reaction procedure, extended reaction period at elevated temperature and poor yield are usually encountered. In recent years, organic reactions in aqueous media have attracted much attention of synthetic organic chemists as an environmentally benign synthetic process.[12] On the other hand, the application of microwave technology in organic chemistry has been explored extensively within the last decade. Microwave irradiation often leads to a remarkable decrease in reaction time, increased yields, and easier work up matching with green chemistry protocols.[13] Moreover, the most successful example of microwave applications are necessarily found to be related to the use of aqueous systems, in which microwaves could be absorbed easily by water (polar solvents) and, therefore, can more efficiently drive chemical reactions.[14] In our current work to develop rapid and efficient methods for the synthesis of compound libraries we have sought to speed up reactions using focused microwave sources.[15] We describe here our results achieved when applying this approach to the synthesis of 2,6-disubstituted imidazo[2,1-*b*][1,3,4]thiadiazole in aqueous media.

Reaction of 2-amino-5-aryloxymethylene-1,3,4-thiadiazole **1a–g** with ω-bromoacetophenone derivatives **2a–b** using water as the only solvent under 350 W microwave irradiation without any catalysts or dehydrating reagent for 6–10 min to give 2-aryloxymethylene-6-arylimidazo[2,1-b][1,3,4]thiadiazole **3a–m** in excellent yields (Scheme 1). The reaction is clean and efficient. When the same reaction was carried out in an oil bath at 90–110 °C which was the observed temperature after the completion of microwave irradiation, even after 10 h the reactions were not complete (TLC) and yields were poor.

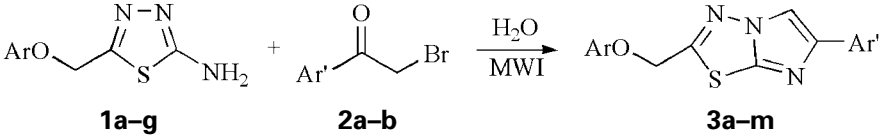
In conclusion, we have demonstrated a practical, rapid and simple process for preparation of 2,6-disubstituted imidazo[2,1-b][1,3,4]thiadiazole in aqueous media under microwave irradiation without any catalysts or dehydrating reagent and given a series of novel compounds. The simplicity of the isolation procedure, short reaction time and no pollution by using water as the only solvent instead of organic solvent are interesting features of this methodology.

## Experimental

IR spectra were recorded using KBr pellets on a Nicolet AVATAR 360 FT-IR spectrophotometer and NMR spectra on a Varian Mercury Plus-400 instrument using DMSO-$d_6$ as solvent and $Me_4Si$ as internal standard. Mass spectra were recorded on a QP-1000A GC-MS using the impact mode (70eV). Elemental analyses were performed on a Vario EI Elemental Analysis instrument. Melting points were determined with an electrothermal micromelting point apparatus and uncorrected. 2-Amino-5-aryloxymethylene-1,3,4-thiadiazole[16] were prepared according to literature procedure.

*General procedure for the synthesis of 2-aryloxymethylene-6-arylimidazo[2,1-b][1,3,4]thiadiazole* **3a–m**: To a suspension of 2 amino-5-aryloxymethylene-1,3,4-thiadiazole **1** (5 mmol) in water (5 ml) was added 1-aryl-2-bromoethan-1-one **2** (5 mmol), and the mixture was stirred for 10 min at r.t. Then this mixture was irradiated in a microwave oven for 6–10 min irradiated time at 350W power, and completion of reactions was monitored by TLC using ethyl acetate, acetone and petroleum ether (1:1:2) as eluent. The precipitate was isolated by filtration and recrystallised from ethanol to give the products. The physical and spectral data of compounds **3a–m** are shown below:

**3a:** White solid, $^1$H NMR (δ ppm) 5.54 (s, 2H), 7.00–7.02 (t, $J$ = 7.6 Hz, 1H), 7.09–7.11 (dd, $J$ = 7.6, 0.8 Hz, 2H), 7.32–7.36 (m, 2H), 7.47–7.49 (d, $J$ = 8.0 Hz, 2H), 7.88–7.90 (d, $J$ = 8.0 Hz, 2H), 8.77 (s, 1H); $^{13}$C NMR (δ ppm): 161.29, 157.13, 145.54, 139.39, 134.81, 132.78, 130.05, 128.74, 126.43, 121.94, 115.04, 110.89,




**1a–g** **2a–b** **3a–m**

**Scheme 1**

* Correspondent. E-mail: wangxicun@nwnu.edu.cn

**Table 1** Synthesis of 2-aryloxymethylene-6-arylimidazo[2,1-b][1,3,4]thiadiazole **3a–m**

| Compound number | Ar | Ar′ | Reaction period/ min | Yield/% | M.p./°C |
|---|---|---|---|---|---|
| **3a** | $C_6H_5$ | 4-$ClC_6H_4$ | 10 | 82 | 210–212 |
| **3b** | 2-$CH_3C_6H_4$ | 4-$ClC_6H_4$ | 7 | 80 | 195–197 |
| **3c** | 3-$CH_3C_6H_4$ | 4-$ClC_6H_4$ | 6 | 80 | 187–189 |
| **3d** | 4-$CH_3C_6H_4$ | 4-$ClC_6H_4$ | 6 | 80 | 230–231 |
| **3e** | 4-$ClC_6H_4$ | 4-$ClC_6H_4$ | 8 | 85 | 223–225 |
| **3f** | 4-$CH_3OC_6H_4$ | 4-$ClC_6H_4$ | 7 | 80 | 218–220 |
| **3g** | $C_6H_5$ | 3-$NO_2C_6H_4$ | 9 | 83 | 152–154 |
| **3h** | 2-$CH_3C_6H_4$ | 3-$NO_2C_6H_4$ | 6 | 78 | 158–160 |
| **3i** | 3-$CH_3C_6H_4$ | 3-$NO_2C_6H_4$ | 8 | 85 | 171–173 |
| **3j** | 4-$CH_3C_6H_4$ | 3-$NO_2C_6H_4$ | 6 | 75 | 162–163 |
| **3k** | 4-$ClC_6H_4$ | 3-$NO_2C_6H_4$ | 7 | 80 | 166–168 |
| **3l** | 4-$CH_3OC_6H_4$ | 3-$NO_2C_6H_4$ | 7 | 78 | 145–147 |
| **3m** | 2,4-$Cl_2C_6H_3$ | 3-$NO_2C_6H_4$ | 8 | 80 | 183–185 |

64.73. MS: *m/z,* 341($M^+$). IR (KBr, δ, $cm^{-1}$): 1592, 1474 (C=N), 1244 ( N–N=C), 688 (C–S–C). Anal. Calcd. for $C_{17}H_{12}ClN_3OS$: C, 59.74; H, 3.54; N, 12.29. Found: C, 59.80; H, 3.51; N, 12.33.

**3b:** White solid, $^1$H NMR (δ ppm) 2.16 (s, 3H), 5.44 (s, 2H), 7.00–7.06 (m, 2H), 7.12–7.15 (q, *J* = 7.2 Hz, 2H), 7.44–7.46 (d, *J* = 8.0 Hz, 2H), 7.86–7.88 (d, *J*=8.0 Hz, 2H), 8.76 (s, 1H); $^{13}$C NMR (δ ppm): 163.16, 155.31, 148.50, 139.48, 134.59, 132.61, 130.47, 128.37, 127.24, 126.58, 126.29, 122.01, 121.86, 110.96, 64.99, 16.05. MS: *m/z,* 355($M^+$). IR (KBr, δ, $cm^{-1}$): 1592, 1472 (C=N), 1246 (N–N=C), 730 (C–S–C). Anal. Calcd. for $C_{18}H_{14}ClN_3OS$: C, 60.76; H, 3.97; N, 11.81. Found: C, 60.72; H, 3.94; N, 11.76.

**3c:** White solid, $^1$H NMR (δ ppm) 2.21 (s, 3H), 5.51 (s, 2H), 6.82–6.92 (m, 3H), 7.18–7.22 (t, *J* = 8.0 Hz, 1H), 7.46–7.48 (d, *J* = 8.0 Hz, 2H), 7.88–7.90 (d, *J* = 8.0 Hz, 2H), 8.78 (s, 1H); $^{13}$C NMR (δ ppm): 162.83, 157.21, 148.49, 139.89, 139.26, 134.51, 132.58, 129.60, 128.47, 126.31, 122.89, 115.82, 112.03, 110.89, 64.78, 21.16. MS: *m/z,* 355($M^+$). IR (KBr, δ, $cm^{-1}$): 1604, 1470 (C=N), 1260 (N–N=C), 688 (C–S–C). Anal. Calcd. for $C_{18}H_{14}ClN_3OS$: C, 60.76; H, 3.97; N, 11.81. Found: C, 60.72; H, 4.01; N, 11.84.

**3d:** White solid, $^1$H NMR (δ ppm) 2.30 (s, 3H), 5.52 (s, 2H), 7.03–7.05(dd, *J* = 4.8, 2.4 Hz, 2H), 7.20–7.22(dd, *J* = 4.8, 2.4 Hz, 2H), 7.45–7.47 (d, *J* = 8.0 Hz, 2H), 7.87–7.89 (d, *J* = 8.0 Hz, 2H), 8.88 (s, 1H); $^{13}$C NMR (δ ppm): 163.09, 157.67, 148.51, 139.32, 134.63, 132.59, 131.68, 130.77, 128.40, 126.51, 115.40, 110.89, 65.07, 24.21. MS: *m/z,* 355($M^+$). IR (KBr, δ, $cm^{-1}$): 1590, 1476 (C=N), 1244 (N–N=C), 702 (C–S–C). Anal. Calcd. for $C_{18}H_{14}ClN_3OS$: C, 60.76; H, 3.97; N, 11.81. Found: C, 60.80; H, 3.94; N, 11.84.

**3e:** White solid, $^1$H NMR (δ ppm) 5.61 (s, 2H), 6.88–6.90(d, *J*=2.0 Hz, 2H), 7.21–7.23 (d, *J* = 2.0 Hz, 2H), 7.47–7.49(d, *J* = 2.0 Hz, 2H), 7.88–7.90 (d, *J* = 2.0 Hz, 2H), 8.79 (s, 1H); $^{13}$C NMR (δ ppm): 161.48, 156.69, 151.08, 139.40, 134.58, 132.37, 130.49, 128.47, 126.66, 126.28, 116.40, 110.87, 65.36. MS: *m/z,* 376($M^+$). IR (KBr, δ, $cm^{-1}$): 1598, 1488 (C=N), 1282 (N–N=C), 670 (C–S–C). Anal. Calcd. for $C_{17}H_{11}Cl_2N_3OS$: C, 54.27; H, 2.95; N, 11.17. Found: C, 54.32; H, 2.90; N, 11.22.

**3f:** White solid, $^1$H NMR (δ ppm) 3.71 (s, 3H), 5.49 (s, 2H), 6.89–6.92 (dd, *J* = 4.8, 2.4 Hz, 2H), 7.04–7.06 (dd, *J* = 4.8, 2.4 Hz, 2H), 7.47–7.49 (d, *J* = 8.0 Hz, 2H), 7.88–7.90 (d, *J* = 8.0 Hz, 2H), 8.75 (s, 1H); $^{13}$C NMR (δ ppm): 161.87, 154.33, 151.03, 148.37, 139.29, 134.61, 132.38, 128.79, 126.40, 115. 69, 114.31, 110.94, 65.63, 55.90. MS: *m/z,* 371($M^+$). IR (KBr, δ, $cm^{-1}$): 1604, 1476 (C=N), 1236 (N–N=C), 698 (C–S–C). Anal. Calcd. for $C_{18}H_{14}ClN_3O_2S$: C, 58.14; H, 3.79; N, 11.30. Found: C, 58.19; H, 3.83; N, 11.25.

**3g:** Yellow solid, $^1$H NMR (δ ppm) 5.53 (s, 2H), 7.00–7.02 (t, *J* = 7.6 Hz, 1H), 7.08–7.10 (dd, *J* = 7.6, 0.8 Hz, 2H), 7.33–7.37 (m, 2H), 7.69–7.73 (t, *J*=8.0 Hz, 1H), 8.14–8.16 (m, 1H), 8.30–8.32 (d, *J* = 7.6 Hz, 1H), 8.67–8.68 (t, *J* = 2.0 Hz, 1H), 8.97 (s, 1H); $^{13}$C NMR (δ ppm): 161.74, 154.96, 148.52, 145.48, 143.16, 135.61, 130.92, 130.49, 129.08, 128.99, 124.31, 122.15, 118.96, 112.15, 65.16. MS: *m/z,* 352($M^+$). IR (KBr, δ, $cm^{-1}$): 1600, 1494 (C=N), 1242 (N–N=C), 728 (C–S–C). Anal. Calcd. for $C_{17}H_{12}N_4O_3S$: C, 57.95; H, 3.43; N, 15.90. Found: C, 57.90; H, 3.39; N, 15.95.

**3h:** Yellow solid, $^1$H NMR (δ ppm) 2.10 (s, 3H), 5.57 (s, 2H), 6.93–6.97 (m, 1H), 7.11–7.13 (d, *J* = 7.6 Hz, 1H), 7.19–7.23 (dd, *J* = 7.6, 1.2 Hz, 2H), 7.70–7.74 (t, *J* = 8.0 Hz, 1H), 8.12–8.14 (m, 1H), 8.29–8.31 (m, 1H), 8.67–8.68 (t, *J* = 2.0 Hz, 1H), 8.97 (s, 1H); $^{13}$C NMR (δ ppm): 161.16, 155.31, 148.50, 145.41, 143.11, 135.60, 130.95, 130.48, 127.25, 126.29, 122.01, 121.86, 118.99, 112.27, 112.15, 64.99, 16.03. MS: *m/z,* 366($M^+$). IR (KBr, δ, $cm^{-1}$): 1592, 1494 (C=N), 1248 (N–N=C), 722 (C–S–C). Anal. Calcd. for $C_{18}H_{14}N_4O_3S$: C, 59.01; H, 3.85; N, 15.29. Found: C, 59.07; H, 3.81; N, 15.26.

**3i:** Yellow solid, $^1$H NMR (δ ppm) 2.31 (s, 3H), 5.54 (s, 2H), 6.85–6.94 (m, 3H), 7.21–7.25 (t, *J* = 8.0 Hz, 1H), 7.69–7.73 (t, *J* = 8.0 Hz, 1H), 8.12–8.14 (m, 1H), 8.28–8.30 (d, *J* = 7.6 Hz, 1H), 8.66–8.67 (t, *J* = 2.0 Hz, 1H), 8.95 (s, 1H); $^{13}$C NMR (δ ppm): 162.83, 157.21, 148.49, 145.46, 143.13, 139.56, 135.59, 130.94, 130.49, 129.61, 122.88, 122.03, 118.99, 115.82, 112.14, 112.01, 64.78, 21.16. MS: *m/z,* 366($M^+$). IR (KBr, δ, $cm^{-1}$): 1586, 1488 (C=N), 1256 (N–N=C), 762 (C–S–C). Anal. Calcd. for $C_{18}H_{14}N_4O_3S$: C, 59.01; H, 3.85; N, 15.29. Found: C, 59.07; H, 3.81; N, 15.33.

**3j:** Yellow solid, $^1$H NMR (δ ppm) 2.34 (s, 3H), 5.56 (s, 2H), 6.97–6.99 (dd, *J* = 4.8, 2.4 Hz, 2H), 7.16–7.18 (dd, *J* = 4.8, 2.4 Hz, 2H), 7.68–7.72 (t, *J* = 8.0 Hz, 1H), 8.12–8.14 (m, 1H), 8.28–8.30 (m, 1H), 8.67–8.68 (t, *J* = 2.0 Hz, 1H), 8.87 (s, 1H); $^{13}$C NMR (δ ppm): 163.24, 157.08, 148.50, 145.47, 143.14, 135.58, 131.16, 130.94, 130.48, 129.69, 122.03, 118.99, 115.34, 112.16, 64.99, 24.31. MS: *m/z,* 366($M^+$). IR (KBr, δ, $cm^{-1}$): 1590, 1510 (C=N), 1242 (N–N=C), 724 (C–S–C). Anal. Calcd. for $C_{18}H_{14}N_4O_3S$: C, 59.01; H, 3.85; N, 15.29. Found: C, 58.96; H, 3.89; N, 15.24.

**3k:** Yellow solid, $^1$H NMR (δ ppm) 5.63 (s, 2H), 6.89–6.91 (d, *J* = 8.0 Hz, 2H), 7.22–7.24 (d, *J* = 8.0 Hz, 2H), 7.70–7.74 (t, *J* = 8.0 Hz, 1H), 8.13–8.15 (m, 1H), 8.30–8.32 (m, 1H), 8.67–8.68 (t, *J* = 2.0 Hz, 1H), 8.95 (s, 1H); $^{13}$C NMR (δ ppm): 162.89, 156.41, 151.74, 145.49, 143.15, 135.61, 130.93, 130.48, 128.96, 126.32, 122.06, 118.94, 116.74, 112.15, 65.02. MS: *m/z,* 386($M^+$). IR (KBr, δ, $cm^{-1}$): 1586, 1488 (C=N), 1284 (N–N=C), 716 (C–S–C). Anal. Calcd. for $C_{17}H_{11}ClN_4O_3S$: C, 52.79; H, 2.87; N, 14.48. Found: C, 52.82; H, 2.84; N, 14.43.

**3l:** Yellow solid, $^1$H NMR (δ ppm) 3.71 (s, 3H), 5.52 (s, 2H), 6.88–6.91 (dd, *J* = 4.8, 2.4 Hz, 2H), 7.04–7.06 (dd, *J* = 4.8, 2.4 Hz, 2H), 7.69–7.73 (t, *J* = 8.0 Hz, 1H), 8.13–8.15 (m, 1H), 8.29–8.31 (d, *J*=7.6 Hz, 1H), 8.68–8.69 (t, *J*=2.0 Hz, 1H), 8.96 (s, 1H); $^{13}$C NMR (δ ppm): 162.36, 157.42, 151.36, 150.10, 145.44, 143.16, 135.62, 130.91, 130.49, 122.16, 118.74, 115.71, 114.64, 112.16, 64.78, 39.74. MS: *m/z,* 382($M^+$). IR (KBr, δ, $cm^{-1}$): 1540, 1506 (C=N), 1236 (N–N=C), 724 (C–S–C). Anal. Calcd. for $C_{18}H_{14}N_4O_4S$: C, 56.54; H, 3.69; N, 14.65. Found: C, 56.58; H, 3.73; N, 14.69.

**3m:**Yellow solid, $^1$H NMR (δ ppm) 5.50 (s, 2H), 7.15–7.39 (m, 3H), 7.68–7.72 (t, *J* = 8.0 Hz, 1H), 8.12–8.14 (m, 1H), 8.30–8.32 (m, 1H), 8.68–8.69 (t, *J* = 2.0Hz, 1H), 8.95 (s, 1H); $^{13}$C NMR (δ ppm): 161.47, 154.08, 147.94, 145.46, 143.13, 135.60, 133.09, 132.47, 131.61, 130.95, 130.50, 122.08, 118.98, 117.63, 114.32, 112.15, 66.42. MS: *m/z,* 421($M^+$). IR (KBr, δ, $cm^{-1}$): 1604, 1478 (C=N), 1262 (N–N=C), 696 (C–S–C). Anal. Calcd. for $C_{17}H_{10}Cl_2N_4O_3S$: C, 48.47; H, 2.39; N, 13.30. Found: C, 48.51; H, 2.35; N, 13.34.

The authors thank the Scientific and Technological Innovation Engineering of Northwest Normal University (NWNU-KJCXGC-02-08), Natural Science Foundation of Gansu Province (ZS021-A25-006-Z), Environmental Protection Foundation of Gansu Province (GH2003-19) for the financial support of this work..

*Received 4 July 2005; accepted 1 August 2005*
*Paper 05/3310*

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