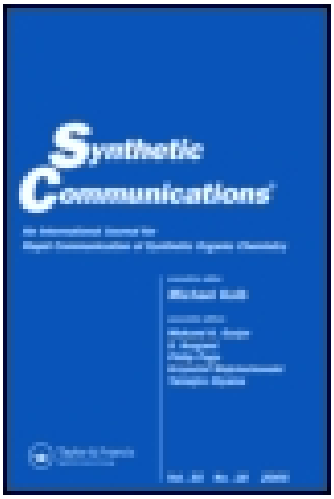

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Efficient Synthesis of 6-(1H-1,2,4-Triazol-1-yl)-thieno[2,3-d]pyrimidin-4(3H)-ones via an Iminophosphorane

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EFFICIENT SYNTHESIS OF 6-(1*H*-1,2,4-TRIAZOL-1-YL)-THIENO[2,3-d]PYRIMIDIN-4(3*H*)-ONES VIA AN IMINOPHOSPHORANE

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Ethyl 2-amino-4-methyl-5-(1H-1,2,4-triazol-1-yl)thiophene-3-carboxylate 2, obtained from Gewald reaction of 1-(1H-1,2,4-triazol-1-yl)acetone 1 with ethyl cyanoacetate and sulfur, was transferred into iminophosphorane 3. Further reaction of 3 with aromatic isocyanates gave carbodiimides 4, which were treated subsequently with a secondary amine to give 6-(1H-1,2,4-triazol-1-yl)-thieno[2,3-d]pyrimidin-4(3H)-ones 6 in good yields in the presence of a catalytic amount of sodium ethoxide.

Keywords: Aza-Wittig reaction; Gewald reaction; iminophosphorane; thieno[2,3-d]pyrimidin-4(3*H*)-one; 1,2,4-triazole

The derivatives of heterocycles containing the thienopyrimidine system are of great importance because of their remarkable biological properties. They have shown significant antifungal, antibacterial, antimalarial, and antiallergic activities.[1–6] The chemistry of thienopyrimidinones has also received attention because their starting materials, 2-amino-3-carboxythiophenes, can conveniently be synthesized by the Gewald reaction.[7] On the other hand, many 1-substituted 1,2,4-triazole compounds show good fungicidal and plant-growth-regulative activities.[8,9] The introduction of 1,2,4-triazole to the thienopyrimidine system is expected to influence the biological activities significantly. However, there are few reports about the 1,2,4-triazole-substituted thienopyrimidinone system, which is of considerable interest as potential agricultural or pharmaceutical fungicides.

Recently, we have been interested in the syntheses of imidazolinones, quinazolinones, and thienopyrimidinones via the aza-Wittig reaction of α- or β-ethoxycarbonyl iminophosphorane with aromatic isocyanate and subsequent reaction with various nucleophiles under mild conditions, with the aim of evaluating their fungicidal activities.[10–14] Herein we report an efficient synthesis of 6-(1*H*-1,2,4-triazol-1-yl)-thieno[2,3-d]pyrimidin-4(3*H*)-ones via an iminophosphorane **3**.

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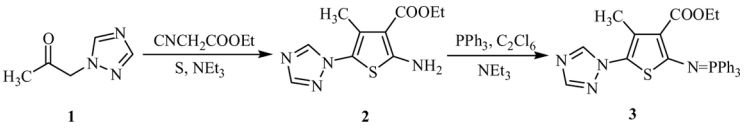


1 2 3

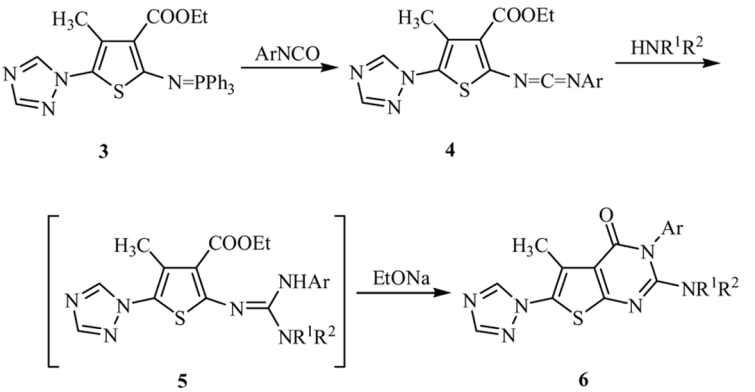
Scheme 1. Preparation of iminophosphorane **3**.

The ethyl 2-amino-4-methyl-5-(1*H*-1,2,4-triazol-1-yl)thiophene-3-carboxylate **2** was obtained by the Gewald method from 1-(1*H*-1,2,4-triazol-1-yl)acetone **1**, ethyl cyanoacetate, and sulfur in the presence of triethylamine. Compound **2** was easily converted to iminophosphorane **3** by treatment with triphenylphosphine, hexachloroethane, and triethylamine in dry acetonitrile in good yield (Scheme 1).

The iminophosphorane **3** reacted with an equimolar quantity of the aromatic isocyanates to give the carbodiimides **4**, which were allowed to react with secondary amines to provide guanidine intermediates **5**. By treatment with sodium ethoxide in ethanol at room temperature, the intermediates **5** underwent intramolecular heterocyclization to give the expected 6-(1*H*-1,2,4-triazol-1-yl)-thieno[2,3-d]pyrimidin-4(3*H*)-ones **6** in good yields (Scheme 2). The results are listed in Table 1.

Owing to the instability of carbodiimide **4**, the preparation of carbodiimides **4** must be carried out at low temperature (0–5 °C) for a long reaction time (12 h) under dry conditions; otherwise, hydrolysis or polymeric reaction of carbodiimide **4** takes place, which results in poor yields. In preparation of products **6**, all of the reactions between carbodiimide **4** and the secondary amine proceeded smoothly at room temperature though the reactivity of the carbodiimides **4** varied with the substituent on the benzene ring. This implied the high reactivity of the carbodiimide **4**.

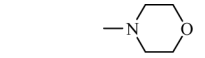
The structure of the synthesized compound **6** was confirmed by their spectral data and elemental analyses. For example, the ^{1}H NMR spectral data of **6a** show the signals of –NCH_3 and CH_3 at 2.70 and 2.41 ppm as singlets. The triazole ring's signals appeared at 8.33 ppm (3-CH) and 8.13 (5-CH) as singlets. The phenyl signals appeared at 7.53–7.26 ppm. The mass spectrum (MS) of **6a** shows a molecule ion peak (M^+) at m/z 352 with 100% abundance.



3 4 5 6

Scheme 2. Preparation of thieno[2,3-d]pyrimidin-4(3*H*)-ones **6**.

Table 1. Preparation of thieno[2,3-d]yrimidine-4(3*H*)-ones **6**

Compound	Ar	NR^1R^2	Conditions	Yield (%)[a]
6a	Ph	NMe_2	rt/4 h	81
6b	Ph	$N(n\text{-}Bu)_2$	rt/4 h	78
6c	Ph	$N(i\text{-}Pr)_2$	rt/8 h	77
6d	Ph		rt/4 h	91
6e	Ph	N(Me)Ph	rt/6 h	72
6f	3-CH_3-C_6H_4	NEt_2	rt/4 h	85
6g	3-CH_3-C_6H_4	$N(n\text{-}Pr)_2$	rt/5 h	87
6h	3-CH_3-C_6H_4	$N(n\text{-}Bu)_2$	rt/7 h	80
6i	4-Cl-C_6H_4	NMe_2	rt/4 h	76
6j	4-Cl-C_6H_4	NEt_2	rt/5 h	88
6k	4-Cl-C_6H_4	$N(n\text{-}Pr)_2$	rt/5 h	83

[a]Isolated yields based on iminophosphorane **3**.

In conclusion, we have developed an efficient synthesis of 2-substituted 6-(1*H*-1,2,4-triazol-1-yl)-thieno[2,3-d]pyrimidin-4(3*H*)-ones via reaction of functionalized carbodiimides with various secondary amines. Because of the easily accessible and versatile starting material, this method has potential for the syntheses of many biologically and pharmaceutically active 1,2,4-triazolyl-containing thienopyrimidinone derivatives.

EXPERIMENTAL

Melting points were uncorrected. MS were measured on Finnigan Trace MS spectrometer. ^{1}H NMR spectra were recorded in $CDCl_3$ on a Varian Mercury 400 spectrometer, and resonances are given in parts per million (δ) relative to tetramethylsilane (TMS). Elemental analyses were performed on a Vario EL III elementary analysis instrument. 1-(1*H*-1,2,4-Triazol-1-yl)acetone **1** was obtained quantitatively by the reaction of 1-chloroacetone with 1*H*-1,2,4-triazole (1.5 equiv.) and solid potassium carbonate in acetone at 60 °C for 20 h.

Preparation of Ethyl 2-Amino-4-methyl-5-(1*H*-1,2,4-triazol-1-yl)thiophene-3-carboxylate 2

Triethyl amine (1.2 mL) was added to a stirred mixture of 1-(1*H*-1,2,4-triazol-1-yl)acetone **1** (0.62 g, 5 mmol), sulfur (0.16 g, 5 mmol), and ethyl cyanoacetate (0.57 g, 5 mmol) in dimethylformamide (DMF; 10 mL). After the mixture was stirred at room temperature for 12 h, it was poured into water (50 mL), and the formed solid was filtered and recrystallized from ethanol/petroleum ether (1:1, v/v) to give **2** as light yellow needles: 0.67 g (53%), mp 122–124 °C; ^{1}H NMR ($CDCl_3$, 400 MHz): δ 8.24 (s, 1H, triazolyl-3-H), 8.08 (s, 1H, triazolyl-5-H), 6.60 (br, 2H, NH_2), 4.31 (q, 2H, $J = 7.2$ Hz, OCH_2), 2.13 (s, 3H, CH_3), 1.36 (t, $J = 7.2$ Hz, 3H, CH_3). MS (*m*/*z*, %): 252 (M^+, 100), 224 (11), 206 (81), 178 (51), 124 (17). Anal. calcd. for $C_{10}H_{12}N_4O_2S$: C, 47.61; H, 4.79; N, 22.21. Found: C, 47.85; H, 4.71; N, 22.05.

Preparation of Iminophosphorane 3

NEt_3 (2.42 g, 24 mmol) was added dropwise to a mixture of 2-amino-4-methyl-5-(1*H*-1,2,4-triazol-1-yl)thiophene-3-carboxylate **2** (2.02 g, 8 mmol), PPh_3 (3.14 g, 12 mmol), and C_2Cl_6 (2.84 g, 12 mmol) in dry CH_3CN (40 mL) at room temperature. The color of the reaction mixture quickly turned yellow. After the mixture was stirred for 4 h, the solvent was removed under reduced pressure and the residue was recrystallized from EtOH to give iminophosphorane **3** as light yellow needles: 3.32 g (81%), mp 179–181 °C; ^{1}H NMR ($CDCl_3$, 400 MHz): δ 8.10–7.48 (m, 17H, Ar-H), 4.35 (q, 2H, $J = 7.2$ Hz, OCH_2), 2.13 (s, 3H, CH_3), 1.38 (t, $J = 7.2$ Hz, 3H, CH_3). MS (*m*/*z*, %): 512 (M^+, 100), 467 (6), 320 (9), 261 (86), 182 (54), 107 (24). Anal. calcd. for $C_{28}H_{25}N_4O_2SP$: C, 65.61; H, 4.92; N, 10.93. Found: C, 65.87; H, 4.68; N, 10.97.

Preparation of Carbodiimides 4

Aromatic isocyanate (2 mmol) was added to a solution of iminophosphorane **3** (1.02 g, 2 mmol) in anhydrous CH_2Cl_2 (10 mL) under a nitrogen atmosphere at room temperature. After the reaction mixture was left unstirred for 6–12 h at 0–5 °C, the iminophosphorane **3** had disappeared (monitored by thin-layer chromatography, TLC). The solvent was removed under reduced pressure, and Et_2O/petroleum ether (1:2, 20 mL) was added to precipitate triphenylphosphine oxide. Removal of the solvent gave carbodiimides **4**, which were used directly without further purification. Carbodiimide **4a** was also isolated from the reaction mixture by column chromatography on silicon gel as a light yellow solid. Mp: 82–84 °C, IR (KBr): 2253, 1701, 1656, 1530, 1438, 1190 cm^{-1}. ^{1}H NMR ($CDCl_3$, 600 MHz): δ 8.23 (s, 1H, triazolyl-3-H), 8.10 (s, 1H, triazolyl-5-H), 7.40–7.21 (m, 5H, Ar-H), 4.33 (q, 2H, $J = 7.2$ Hz, OCH_2), 2.10 (s, 3H, CH_3), 1.37 (t, $J = 7.2$ Hz, 3H, CH_3). MS (*m*/*z*, %): 353 (M^+, 100), 307 (62), 293 (34), 265 (74), 178 (67), 124 (47). Anal. calcd. for $C_{17}H_{15}N_5O_2S$: C, 57.78; H, 4.28; N, 19.82. Found: C, 57.54; H, 4.01; N, 19.95.

General Preparation of 6-(1*H*-1,2,4-Triazol-1-yl)-thieno[2,3-d]pyrimidin-4(3*H*)-ones 6

Aliphatic amine (2 mmol) was added to the solution of **4** in dichloromethane (10 mL). After the reaction mixture was left unstirred for 5–6 h, the solvent was removed and anhydrous EtOH (10 mL) with several drops of EtONa was added. The mixture was stirred for 4–8 h at room temperature. The solution was condensed, and the residue was recrystallized from EtOH to give the expected cyclic compouds **6a**–**k** in good yields.

Compound 6a. Light yellow solid, mp 222–223 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ 8.33 (s, 1H, triazolyl-3-H), 8.13 (s, 1H, triazolyl-5-H), 7.53–7.26 (m, 5H, Ar-H), 2.70 (s, 6H, $2CH_3N$), 2.41 (s, 3H, CH_3). MS (*m*/*z*, %): 352 (M^+, 100%), 337 (6), 308 (12), 287 (17), 145 (70), 130 (76). Anal. calcd. for $C_{17}H_{16}N_6OS$: C, 57.94; H, 4.58; N, 23.85. Found: C, 57.80; H, 4.36; N, 23.94.

Compound 6b. Light yellow solid, mp 123–125 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ 8.32 (s, 1H, triazolyl-3-H), 8.13 (s, 1H, triazolyl-5-H), 7.51–7.26 (m, 5H, Ar-H), 3.03 (t, $J = 6.8$ Hz, 4H, $2CH_2N$), 2.41 (s, 3H, CH_3), 1.23–1.13 (m, 8H, $2CH_2CH_2$), 0.84 (t, $J = 6.8$ Hz, 6H, $2CH_3$). MS (m/z, %): 436 (M^+, 100%), 407 (13), 394 (53), 378 (78), 353 (23), 308 (40). Anal. calcd. for $C_{23}H_{28}N_6OS$: C, 63.28; H, 6.46; N, 19.25. Found: C, 63.35; H, 6.61; N, 19.48.

Compound 6c. Light yellow solid, mp 174–176 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ 8.32 (s, 1H, triazolyl-3-H), 8.13 (s, 1H, triazolyl-5-H), 7.49–7.27 (m, 5H, Ar-H), 3.60–3.53 (m, 2H, 2CHN), 2.40 (s, 3H, CH_3), 1.09 (d, $J = 6.8$ Hz, 12H, $4CH_3$). MS (m/z, %): 408 (M^+, 38%), 365 (100), 350 (20), 307 (21), 118 (16), 100 (17). Anal. calcd. for $C_{21}H_{24}N_6OS$: C, 61.74; H, 5.92; N, 20.57. Found: C, 61.52; H, 5.85; N, 20.76.

Compound 6d. Light yellow solid, mp 125–127 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ 8.35 (s, 1H, triazolyl-3-H), 8.14 (s, 1H, triazolyl-5-H), 7.55–7.27 (m, 5H, Ar-H), 3.44 (t, $J = 4.4$ Hz, 4H, $2CH_2O$), 3.16 (t, $J = 4.4$ Hz, 4H, $2CH_2N$), 2.44 (s, 3H, CH_3). MS (m/z, %): 394 (M^+, 100%), 363 (7), 349 (19), 337 (19), 308 (8), 205 (18). Anal. calcd. for $C_{19}H_{18}N_6O_2S$: C, 57.85; H, 4.60; N, 21.31. Found: C, 57.94; H, 4.74; N, 21.17.

Compound 6e. Light yellow solid, mp 222–224 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ 8.36 (s, 1H, triazolyl-3-H), 8.15 (s, 1H, triazolyl-5-H), 7.12–6.60 (m, 10H, Ar-H), 3.30 (s, 3H, CH_3N), 2.44 (s, 3H, CH_3). MS (m/z, %): 414 (M^+, 100%), 308 (32), 253 (25), 167 (35), 104 (25), 91 (24). Anal. calcd. for $C_{22}H_{18}N_6OS$: C, 63.75; H, 4.38; N, 20.28. Found: C, 63.51; H, 4.58; N, 20.33.

Compound 6f. Light yellow solid, mp 115–117 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ 8.34 (s, 1H, triazolyl-3-H), 8.14 (s, 1H, triazolyl-5-H), 7.41–7.10 (m, 4H, Ar-H), 3.15 (q, $J = 7.2$ Hz, 4H, $2CH_2N$), 2.41 (s, 6H, $2CH_3$), 0.85 (t, $J = 7.2$ Hz, 6H, $2CH_3$). MS (m/z, %): 394 (M^+, 100%), 365 (45), 322 (16), 275 (24), 206 (26), 119 (63). Anal. calcd. for $C_{20}H_{22}N_6OS$: C, 60.89; H, 5.62; N, 21.30. Found: C, 60.74; H, 5.45; N, 21.58.

Compound 6g. Light yellow solid, mp 150–152 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ 8.33 (s, 1H, triazolyl-3-H), 8.14 (s, 1H, triazolyl-5-H), 7.40–7.09 (m, 4H, Ar-H), 3.02 (t, $J = 7.2$ Hz, 4H, $2CH_2N$), 2.41 (s, 6H, $2CH_3$), 1.31–1.24 (m, 4H, $2CH_2$), 0.74 (t, $J = 7.2$ Hz, 6H, $2CH_3$). MS (m/z, %): 422 (M^+, 100%), 393 (36), 322 (19), 289 (15), 132 (25). Anal. calcd. for $C_{22}H_{26}N_6OS$: C, 62.53; H, 6.20; N, 19.89. Found: C, 62.47; H, 6.41; N, 19.74.

Compound 6h. Light yellow solid, mp 94–96 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ 8.33 (s, 1H, triazolyl-3-H), 8.14 (s, 1H, triazolyl-5-H), 7.39–7.08 (m, 4H, Ar-H), 3.05 (t, $J = 7.2$ Hz, 4H, $2CH_2N$), 2.41 (s, 6H, $2CH_3$), 1.23–1.11 (m, 8H, $2CH_2CH_2$), 0.84 (t, $J = 7.2$ Hz, 6H, $2CH_3$). MS (m/z, %): 450 (M^+, 84%), 393 (35), 322 (48), 294 (32), 105 (100). Anal. calcd. for $C_{24}H_{30}N_6OS$: C, 63.97; H, 6.71; N, 18.65. Found: C, 63.71; H, 6.94; N, 18.78.

Compound 6i. Light yellow solid, mp 210–212 °C. ^{1}H NMR ($CDCl_3$, 400 MHz): δ 8.34 (s, 1H, triazolyl-3-H), 8.13 (s, 1H, triazolyl-5-H), 7.50–7.27 (m,

4H, Ar-H), 2.72 (s, 6H, $2CH_3$), 2.40 (s, 3H, CH_3). MS (m/z, %): 386 (M^+, 100%), 352 (26), 308 (34), 286 (47), 130 (85). Anal. calcd. for $C_{17}H_{15}ClN_6OS$: C, 52.78; H, 3.91; N, 21.72. Found: C, 52.72; H, 3.84; N, 21.87.

Compound 6j. Light yellow solid, mp 156–158 °C. ^1H NMR ($CDCl_3$, 400 MHz): δ 8.33 (s, 1H, triazolyl-3-H), 8.13 (s, 1H, triazolyl-5-H), 7.50–7.26 (m, 4H, Ar-H), 3.13 (q, $J = 7.2$ Hz, 4H, $2CH_2N$), 2.40 (s, 3H, CH_3), 0.90 (t, $J = 6.8$ Hz, 6H, $2CH_3$). MS (m/z, %): 414 (M^+, 100%), 365 (63), 322 (63), 206 (35), 119 (76). Anal. calcd. for $C_{19}H_{19}ClN_6OS$: C, 55.00; H, 4.62; N, 20.25. Found: C, 55.16; H, 4.37; N, 20.28.

Compound 6k. Light yellow solid, mp 150–152 °C. ^1H NMR ($CDCl_3$, 400 MHz): δ 8.33 (s, 1H, triazolyl-3-H), 8.13 (s, 1H, triazolyl-5-H), 7.50–7.25 (m, 4H, Ar-H), 3.01 (t, $J = 7.6$ Hz, 4H, $2CH_2N$), 2.40 (s, 3H, CH_3), 1.38–1.29 (m, 4H, $2CH_2$), 0.77 (t, $J = 7.6$ Hz, 6H, $2CH_3$). MS (m/z, %): 442 (M^+, 74%), 408 (24), 365 (100), 307 (47), 118 (74). Anal. calcd. for $C_{21}H_{23}ClN_6OS$: C, 56.94; H, 5.23; N, 18.97. Found: C, 56.86; H, 5.25; N, 18.75.

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