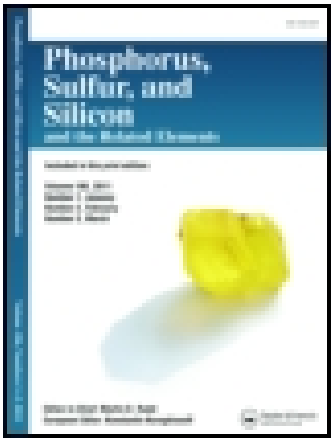

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The Synthesis of 2-Alkylthio-3-alkyl-5-arylmethylidene4H-imidazol-4-ones

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2-alkylthio-3-alkyl-5-arylmethylidene-4H-imidazol-4-ones were synthesized by the S-alkylation and N-alkylation of 2-thioxo-5-arylmethylidene-4-imidazolidinones, which were obtained via a tandem aza-Wittig reaction of vinyliminophosphoranes, carbon disulfide, and excess ammonium hydroxide (28% NH_3 in water).

Keywords 4H-imidazol-4-ones; alkylation; aza-Wittig reaction; synthesis

INTRODUCTION

4H-imidazol-4-ones are important heterocycles having biological and pharmaceutical activities,[1–10] and some 2-alkylthio-4H-imidazol-4-ones show significant fungicidal activities.[11–13] Until now, many of the new derivatives of 2-alkylthio-4H-imidazol-4-ones have been synthesized to evaluate their biological and pharmaceutical activities. However, most of the 2-alkylthio-4H-imidazol-4-ones reported are of the 5,5-disubstituted type and were generally synthesized from the corresponding α-amino acetic acid[13,14] (Scheme 1). Regrettably, 2-alkylthio-5-arylmethylidene-4H-imidazol-4ones cannot be prepared by this general method, for the corresponding starting material needed would

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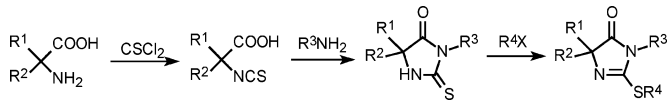
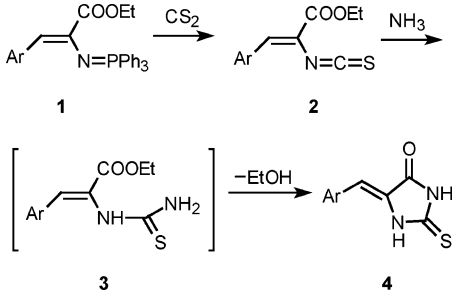


SCHEME 1

be unstable vinyl amino acetic acids. Recently, we have become interested in the synthesis of new imidazolone derivatives, especially in 2-alkylthio-5-arylmethylidene-4H-imidazol-4ones, via a tandem aza-Wittig reaction, and some of them have been shown potential fungicidal activities.[15–24] In the present work, we wish to report further a new efficient synthesis method of some new 2-alkylthio-3-alkyl-5-arylmethylidene-4H-imidazol-4-ones derivatives **5** from the stable vinyliminophosphoranes **1**.

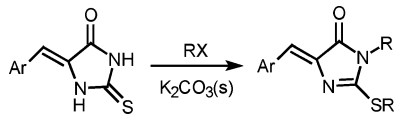
RESULTS AND DISCUSSION

The easily accessible iminophosphoranes **1**[25,26] reacted with carbon disulfide to give vinyl isothiocyanates **2**,[13–18] which were allowed to react with excess ammonium hydroxide (28% NH_3 in water) smoothly at r.t. to give 2-thioxo-5-arylmethylidene-4-imidazolidinones **4**[27,28] in 86–90% yields (Scheme 2 and Table I). The formation of **4** can be rationalized in terms of an initial nucleophilic addition of ammonia to give the intermediates **3**, which cyclize to give **4**.



SCHEME 2

The S- alkylation[13–18] and N-alkylation of **4** with excess alkyl halides in the presence of solid potassium carbonate provided 2-alkylthio-3-alkyl-5-arylmethylidene-4H-imidazol-4-ones **5** in 55–86% yields (Scheme 3). When activated alkylating reagents (RI, $BrCH_2COR$) were used, the alkylation could be carried out at r.t. When other alkylating reagents were applied, the alkylation had to be carried out at 50–70°C (Table I).



4 5

SCHEME 3

The structures of **4** and **5** have been determined through spectroscopic characterization. For example, the ^{1}H NMR spectroscopic data in **5a** show the signals of=CH, $-NCH_3$, and $-SCH_3$ at 6.88 ppm, 3.17 ppm, and 2.74 ppm as single peaks, respectively. The chemical shift of the aryl hydrogens were in the range 8.10–7.26 ppm and appear as a multiplet. In the IR spectrum data of **5a**, the strong stretching peak of imidazolone C=O appears at 1726 cm^{-1}. The stretching vibration of C=C shows a strong absorption band at about 1642 cm^{-1} due to a resonance effect. The MS of **5a** shows a molecular ion peak at m/z 266 with 100% abundance.

EXPERIMENTAL

Melting points were uncorrected. MS were measured on a Finnigan Trace spectrometer. IR were recorded on a PE-983 infrared

TABLE I The Preparation of Derivatives of 2-Thioxo-5-Arylmethylidene-4-Imidazolidinones 4 and 2-Alkylthio-3-Alkyl-5-Arylmethylidene-4H-imidazol-4-ones 5

Entry	Ar	RX	Conditions	Masses (g)	Yield (%)*	m.p. (°C)
4a[a]	4-Cl—C_6H_4		r.t./2 h	1.07	90	286–287
4b	2-Cl—C_6H_4		r.t./3 h	1.02	86	256–258
5a[b]	4-Cl—C_6H_4	MeI	r.t./2 h	0.82	77	159–161
5b	4-Cl—C_6H_4	EtBr	60°C/4 h	0.85	72	114–116
5c	4-Cl—C_6H_4	*n*-PrBr	70°C/6 h	0.85	66	102–104
5d	4-Cl—C_6H_4	*n*-BuBr	70°C/8 h	0.77	55	76–78
5e	4-Cl—C_6H_4	$PhCH_2Cl$	50°C/3 h	1.44	86	183–185
5f	4-Cl—C_6H_4	$PhCOCH_2Br$	r.t./3 h	1.59	84	197–199
5g	4-Cl—C_6H_4	$BrCH_2COOMe$	r.t./3 h	1.10	72	156–158
5h	4-Cl—C_6H_4	$ClCH_2COOEt$	50°C/4 h	1.13	69	134–136
5i	2-Cl—C_6H_4	MeI	r.t./3 h	0.79	74	181–182
5j	2-Cl—C_6H_4	$PhCH_2Cl$	50°C/4 h	1.36	81	121–123

[a] Isolated yields of **4** based on vinyliminophosphoranes **1**.
[b] Purified yields of **5** based on 2-thioxo-5-arylmethylidene-4-imidazolidinones **4**.

spectrometer as KBr pellets with absorption in cm^{-1}. NMR were recorded in $CDCl_3$ for **5** or DMSO-d_6 for **4** on a Varian Mercury 400 spectrometer, and resonances are given in ppm (δ) relative to tetramethylsilane (TMS). Elemental analyses were recorded on a Vario EL III elementary analysis instrument. CS_2 is poisonous, and a good hood should be used. Vinyliminophosphoranes **1** were prepared by the literature report.[25,26]

The Preparation of 2-Thioxo-5-arylmethylidene-4-imidazolidinones 4

To a solution of vinyliminophosphoranes **1** (2.43 g, 5 mmol) in dry methylene chloride (15 mL) was added excess carbon disufide (5 mL). After the reaction mixture was refluxed for 28 h, the solvent was removed under reduced pressure, and ether/petroleum ether (1:2, 20 mL) was added to precipitate triphenylphosphine sulfide, which was removed by filtration. The filtrate was evaporated to give vinyl isothiocyanate **2**, which was used directly without further purification. To the solution of crude **2** in CH_3CN (15 mL) was added excess ammonium hydroxide (28% NH_3 in water) (2 mL, 30 mmol). The mixture was allowed to stand for 2–3 h at r.t., and the precipitated solid was collected and washed with water and ethanol and recrystallized from ethanol to give **4**.

2-Thioxo-5-(4-chlorophenylmethylidene)-4-imidazolidinone (4a)

Yellow crystals, ^{1}H NMR (DMSO-d_6, 400 MHz): δ 12.43 (s, 1H, O=CNH), 12.21 (s, 1H, C=CNH), 7.78–7.47 (m, 4H, Ar-H), 6.48 (s, 1H, =CH); IR (cm^{-1}), 3336 (N–H), 3310 (N–H), 1716 (C=O), 1649 (C=C); MS (*m/z*, %), 240 (M^+, 27), 238 (M^+, 70), 203 (2), 150 (62), 122 (23), 115 (31), 88 (71), 58 (100). Elemental anal. calcd. for $C_{10}H_7N_2OSCl$: C, 50.31; H, 2.94; N, 11.74. Found: C, 50.44; H, 3.10; N, 11.93.

2-Thioxo-5-(2-chlorophenylmethylidene)-4-imidazolidinone (4b)

Light yellow crystals, ^{1}H NMR (DMSO-d_6, 400 MHz): δ 12.49 (s, 1H, O=CNH), 12.29 (s, 1H, C=CNH), 7.84–7.40 (m, 4H, Ar–H), 6.60 (s, 1H, =CH); IR (cm^{-1}), 3345 (N–H), 3321 (N–H), 1719 (C=O), 1653 (C=C); MS (m/z, %), 240 (M^+, 2), 238 (M^+, 6), 203 (6), 151 (12), 124 (14), 115 (18), 85 (62), 58 (100). Elemental anal. calcd. for $C_{10}H_7N_2OSCl$: C, 50.31; H, 2.94; N, 11.74. Found: C, 50.57; H, 3.18; N, 12.01.

The Preparation of 2-Alkylthio-3-alkyl-5-arylmethylidene-4H-imidazol-4-ones 5

A mixture of **4** (4 mmol), excess alkyl halides (16 mmol), and solid potassium carbonate (2.22 g, 16 mmol) in CH_3CN (30 mL) was stirred for 2–8 h at r.t. or 50–70°C and filtered; the filtrate was condensed, and the residue was recrystallized from methylene chloride/petroleum ether to give **5**.

2-Methylthio-3-methyl-5-(4-chlorophenylmethylidene)-4H-imidazol-4-one (5a)

Light yellow crystals, ^{1}H NMR ($CDCl_3$, 400 MHz): δ 8.10–7.26 (m, 4H, Ar-H), 6.88 (s, 1H, =CH), 3.17 (s, 3H, NCH_3), 2.74 (s, 3H, SCH_3); IR (cm^{-1}), 1726 (C=O), 1642 (C=C); MS (*m/z*, %), 268 (M^+, 35), 266 (M^+, 100), 251 (1), 233 (5), 221 (11), 176 (13), 149 (24), 87 (87). Elemental anal. calcd. for $C_{12}H_{11}N_2OSCl$: C, 54.03; H, 4.13; N, 10.51. Found: C, 53.99; H, 3.98; N, 10.73.

2-Ethylthio-3-ethyl-5-(4-chlorophenylmethylidene)-4H-imidazol-4-one (5b)

Yellow crystals, ^{1}H NMR ($CDCl_3$, 400 MHz): δ 8.09–7.27 (m, 4H, Ar—H), 6.85 (s, 1H, =CH), 3.64 (q, 2H, NCH_2), 3.36 (q, 2H, SCH_2), 1.52 (t, 3H, $NCH_2\underline{CH_3}$), 1.26 (t, 3H, $SCH_2\underline{CH_3}$); IR (cm^{-1}), 1729 (C=O), 1638 (C=C); MS (*m/z*, %), 296 (M^+, 29), 294 (M^+, 100), 278 (3), 266 (57), 261 (44), 238 (11), 182 (21), 149 (30). Elemental anal. calcd. for $C_{14}H_{15}N_2OSCl$: C, 57.05; H, 5.09; N, 9.51. Found: C, 57.11; H, 5.18; N, 9.71.

2-(n-Propylthio)-3-(n-propyl)-5-(4-chlorophenylmethylidene)-4H-imidazol-4-one (5c)

Yellow crystals, ^{1}H NMR ($CDCl_3$, 400 MHz): δ 8.09–7.27 (m, 4H, Ar—H), 6.84 (s, 1H, =CH), 3.55 (t, 2H, NCH_2), 3.32 (t, 2H, SCH_2), 1.93–1.84 (m, 2H, $NCH_2\underline{CH_2}CH_3$), 1.74–1.65 (m, 2H, $SCH_2\underline{CH_2}CH_3$), 1.10 (t, 3H, $NCH_2CH_2\underline{CH_3}$), 0.94 (t, 3H, $SCH_2CH_2\underline{CH_3}$); IR ($cm^{-1}$), 1728 (C=O), 1639 (C=C); MS (*m/z*, %), 324 (M^+, 10), 322 (M^+, 27), 307 (4), 294 (100), 280 (54), 247 (39), 238 (41), 182 (90). Elemental anal. calcd. for $C_{16}H_{19}N_2OSCl$: C, 59.53; H, 5.89; N, 8.68. Found: C, 59.69; H, 6.02; N, 8.88.

2-(n-Butylthio)-3-(n-butyl)-5-(4-chlorophenylmethylidene)-4H-imidazol-4-one (5d)

Yellow crystals, ^{1}H NMR ($CDCl_3$, 400 MHz): δ 8.10–7.26 (m, 4H, Ar—H), 6.84 (s, 1H, =CH), 3.58 (t, 2H, NCH_2), 3.33 (t, 2H, SCH_2),

1.86–1.33 (m, 8H, $NCH_2\underline{CH_2}\underline{CH_2}CH_3$ and $SCH_2\underline{CH_2}\underline{CH_2}CH_3$), 1.03–0.93 (m, 6H, $NCH_2CH_2CH_2\underline{CH_3}$ and $SCH_2CH_2CH_2\underline{CH_3}$); IR ($cm^{-1}$), 1728 (C=O), 1642 (C=C); MS (*m/z*, %), 352 (M^+, 9), 350 (M^+, 25), 321 (6), 317 (6), 303 (36), 294 (16), 261 (100), 149 (15). Elemental anal. calcd. for $C_{18}H_{23}N_2OSCl$: C, 61.63; H, 6.56; N, 7.99. Found: C, 61.89; H, 6.77; N, 8.21.

2-Benzylthio-3-benzyl-5-(4-chlorophenylmethylidene)-4H-imidazol-4-one (5e)

Light yellow crystals, ^1H NMR ($CDCl_3$, 400 MHz): δ 8.11–7.25 (m, 14H, Ar—H), 6.94 (s, 1H, =CH), 4.76 (s, 2H, NCH_2), 4.54 (s, 2H, SCH_2); IR (cm^{-1}), 1722 (C=O), 1640 (C=C); MS (*m/z*, %), 420 (M^+, 6), 418 (M^+, 14), 385 (14), 328 (8), 294 (2), 150 (6), 90 (100), 64 (25). Elemental anal. calcd. for $C_{24}H_{19}N_2OSCl$: C, 68.82; H, 4.54; N, 6.69. Found: C, 69.03; H, 4.75; N, 6.98.

2-Benzoylmethylthio-3-benzoylmethyl-5-(4-chlorophenylmethylidene)-4H-imidazol-4-one (5f)

Yellow crystals, ^1H NMR ($CDCl_3$, 400 MHz): δ 8.14–7.25 (m, 14H, Ar—H), 6.96 (s, 1H, =CH), 4.66 (s, 2H, NCH_2), 4.40 (s, 2H, SCH_2); IR (cm^{-1}), 1734 (C=O), 1696 (COPh), 1690 (COPh), 1647 (C=C); MS (*m/z*, %), 476 (M^+, 18), 474 (M^+, 49), 439 (9), 369 (35), 164 (55), 150 (87), 105 (83), 89 (100). Elemental anal. calcd. for $C_{26}H_{19}N_2O_3SCl$: C, 65.75; H, 4.00; N, 5.90. Found: C, 66.01; H, 3.96; N, 6.15.

2-Methoxycarbonylmethylthio-3-methoxycarbonylmethyl-5-(4-chlorophenylmethylidene)-4H-imidazol-4-one (5g)

Light yellow crystals, ^1H NMR ($CDCl_3$, 400 MHz): δ 8.07–7.26 (m, 4H, Ar—H), 6.94 (s, 1H, =CH), 4.38 (s, 2H, NCH_2), 4.35 (s, 3H, $NCH_2COO\underline{CH_3}$), 4.33 (s, 3H, $SCH_2COO\underline{CH_3}$), 4.12 (s, 2H, SCH_2); IR (cm^{-1}), 1741 (COOEt), 1738 (COOEt), 1731 (C=O), 1642 (C=C); MS (*m/z*, %), 384 (M^+, 34), 382 (M^+, 90), 351 (15), 323 (100), 310 (13), 266 (57), 250 (39), 164 (79). Elemental anal. calcd. for $C_{16}H_{15}N_2O_5SCl$: C, 50.20; H, 3.92; N, 7.32. Found: C, 50.44; H, 4.17; N, 7.15.

2-Ethoxycarbonylmethylthio-3-ethoxycarbonylmethyl-5-(4-chlorophenylmethylidene)-4H-imidazol-4-one (5h)

White crystals, ^1H NMR ($CDCl_3$, 400 MHz): δ 8.06–7.25 (m, 4H, Ar—H), 6.94 (s, 1H, =CH), 4.37 (s, 2H, NCH_2), 4.28–4.22 (m, 4H, $NCH_2COO\underline{CH_2}CH_3$ and $SCH_2COO\underline{CH_2}CH_3$), 4.11 (s, 2H, SCH_2), 1.32–1.28 (m, 6H, $NCH_2COOCH_2\underline{CH_3}$ and $SCH_2COOCH_2\underline{CH_3}$); IR ($cm^{-1}$), 1742 (COOEt), 1739 (COOEt), 1731 (C=O), 1642 (C=C); MS (*m/z*, %), 412 (M^+, 38), 410 (M^+, 100), 365 (20), 336 (99), 307 (10), 279 (82), 164

(71), 149 (76). Elemental anal. calcd. for $C_{18}H_{19}N_2O_5SCl$: C, 52.62; H, 4.63; N, 6.82. Found: C, 52.83; H, 4.76; N, 6.97.

2-Methylthio-3-methyl-5-(2-chlorophenylmethylidene)-4H-imidazol-4-one (5i)

Yellow crystals, ^{1}H NMR ($CDCl_3$, 400 MHz): δ 8.88–7.25 (m, 5H, Ar—H and =CH), 3.17 (s, 3H, NCH_3), 2.72 (s, 3H, SCH_3); IR (cm^{-1}), 1730 (C=O), 1645 (C=C); MS (*m/z*, %), 268 (M^+, 7), 266 (M^+, 17), 231 (100), 216 (10), 183 (7), 177 (9), 149 (36), 87 (83). Elemental anal. calcd. for $C_{12}H_{11}N_2OSCl$: C, 54.03; H, 4.13; N, 10.51. Found: C, 54.25; H, 4.38; N, 10.78.

2-Benzylthio-3-benzyl-5-(2-chlorophenylmethylidene)-4H-imidazol-4-one (5j)

Yellow crystals, ^{1}H NMR ($CDCl_3$, 400 MHz): δ 8.89–7.25 (m, 15H, Ar—H and =CH), 4.77 (s, 2H, NCH_2), 4.53 (s, 2H, SCH_2); IR (cm^{-1}), 1725 (C=O), 1643 (C=C); MS (*m/z*, %), 420 (M^+, 1), 418 (M^+, 3), 385 (2), 151 (3), 123 (2), 104 (2), 90 (100), 64 (21). Elemental anal. calcd. for $C_{24}H_{19}N_2OSCl$: C, 68.82; H, 4.54; N, 6.69. Found: C, 69.09; H, 4.77; N, 6.89.

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