ORGANIC LETTERS

2010
Vol. 12, No. 13
3066−3069

Preparation of Tetrasubstituted Furans via Intramolecular Wittig Reactions with Phosphorus Ylides as Intermediates

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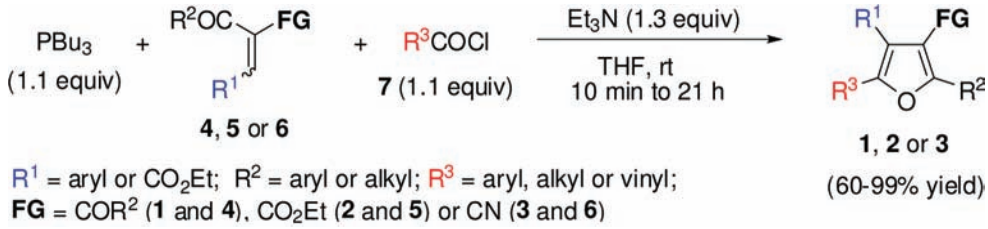
Received May 11, 2010

ABSTRACT



Novel preparation of tetrasubstituted furans, starting from the Michael acceptors, tributylphosphine, and acyl chlorides, is realized. A broad range of highly functional furans can be efficiently generated in one step at room temperature within 10 min to 21 h in moderate to high yields (60−99%). The reaction was proposed to proceed via intramolecular Wittig-type reactions, using phosphorus ylides as intermediates.

Multisubstituted furans are of great importance because numerous interesting compounds bearing such a heterocyclic ring exhibit a wide array of activity and are also building blocks for organic synthesis.[1,2] Many synthetic routes toward furan rings with specific substitution patterns have been designed and well applied,[1c,3] such as direct functionalization of furan rings,[4] cyclocondensation of 1,4-dicarbonyl compounds (Paal−Knorr synthesis),[5] Feist−Bénary synthesis,[6] and transition metal-catalyzed cycloisomerization of alkynyl or allenyl substrates.[3a,7] To our surprise, of these developed strategies,[1−8] there are few literature reports for the syntheses of tetrasubstituted furans with three aryl groups and a ketone,

(1) For recent reviews, see: (a) Hou, X. L.; Yang, Z.; Wong, H. N. C. *Progress in Heterocyclic Chemistry*; Gribble, G. W., Joule, J. A., Eds.; Pergamon: Oxford, UK, 2008; Vol. 19, p 176. (b) Keay, B. A.; Dibble, P. W. *Comprehensive Heterocyclic Chemistry II*; Katritzky, A. R., Rees, C. W., Scriven, E. F. V., Eds.; Elsevier: Oxford, UK, 1997; Vol. 2, p 395. (c) Hou, X. L.; Cheung, H. Y.; Hon, T. Y.; Kwan, P. L.; Lo, T. H.; Tong, S. Y. T.; Wong, H. N. C. *Tetrahedron* **1998**, *54*, 1955. (d) Keay, B. A. *Chem. Soc. Rev.* **1999**, *28*, 209. (e) Gilchrist, T. L. *J. Chem. Soc., Faraday Trans. 1* **1999**, 2849.

(2) (a) Lipshutz, B. H. *Chem. Rev.* **1986**, *86*, 795. (b) Wong, H. N. C.; Yu, P.; Yick, C.-Y. *Pure Appl. Chem.* **1999**, *71*, 1041. (c) Lee, H.-K.; Chan, K.-F.; Hui, C.-W.; Yim, H.-K.; Wu, X.-W.; Wong, H. N. C. *Pure Appl. Chem.* **2005**, *77*, 139. (d) Heaney, H. *Natural Products Chemistry*; Nakanishi, K., Ed.; Kodansha: Tokyo, Japan, 1974; p 297.

(3) For recent reviews, see: (a) Kirsch, S. F. *Org. Biomol. Chem.* **2006**, *4*, 2076. (b) Brown, R. C. D. *Angew. Chem., Int. Ed.* **2005**, *44*, 850. (c) Cacchi, S. *J. Organomet. Chem.* **1999**, *576*, 42.

(4) For recent examples, see: (a) Melzig, L.; Rauhut, C. B.; Knochel, P. *Chem. Commun.* **2009**, 3536. (b) Snégaroff, K.; L'Helgoual'ch, J.-M.; Bentabed-Ababsa, G.; Nguyen, T. T.; Chevallier, F.; Yonehara, M.; Uchiyama, M.; Derdour, A.; Mongin, F. *Chem.—Eur. J.* **2009**, *15*, 10280. For selected reviews, see: (c) Ila, H.; Baron, O.; Wagner, A. J.; Knochel, P. *Chem. Commun.* **2006**, 583. (d) *Handbook of functionalized organometallics*; Knochel, P., Ed.; Wiley-VCH: Weinheim, Germany, 2005.

(5) Minetto, G.; Raveglia, L. F.; Sega, A.; Taddei, M. *Eur. J. Org. Chem.* **2005**, 5277, and references cited therein.

(6) For selected examples, see: (a) Mross, G.; Holtz, E.; Langer, P. *J. Org. Chem.* **2006**, *71*, 8045. (b) Feist, F. *Chem. Ber.* **1902**, *35*, 1537. (c) Bénary, E. *Chem. Ber.* **1911**, *44*, 489.

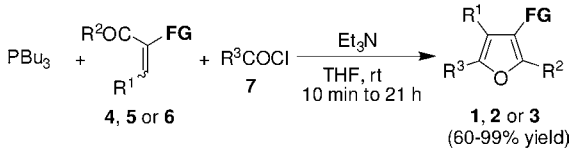
(7) For selected examples starting from allenyl ketones, see: (a) Dudnik, A. S.; Gevorgyan, V. *Angew. Chem., Int. Ed.* **2007**, *46*, 5195. (b) Hashmi, A. S. K. *Angew. Chem., Int. Ed. Engl.* **1995**, *34*, 1581. For examples from alkynyl ketone, see: (c) Hashmi, A. S. K.; Schwarz, L.; Choi, J.-H.; Frost, T. M. *Angew. Chem., Int. Ed.* **2000**, *39*, 2285. (d) Marshall, J. A.; Bartley, G. S. *J. Org. Chem.* **1994**, *59*, 7169. (e) Ma, S.; Zhang, J.; Lu, L. *Chem.—Eur. J.* **2003**, *9*, 2447. For examples from alkynyl epoxide, see: (f) Hashmi, A. S. K.; Sinha, P. *Adv. Synth. Catal.* **2004**, *346*, 432. For electrophilic cyclization, see: (g) Sniady, A.; Wheeler, K. A.; Dembinski, R. *Org. Lett.* **2005**, *7*, 1769. For examples from alkynyl alcohols, see: (h) Liu, Y.; Song, F.; Song, Z.; Liu, M.; Yan, B. *Org. Lett.* **2005**, *7*, 5409. For examples from alkynyl cyclopropyl ketones, see: (i) Zhang, J.; Schmalz, H.-G. *Angew. Chem., Int. Ed.* **2006**, *45*, 6704. For examples from other substrates, see (j) Peng, L.; Zhang, X.; Ma, M.; Wang, J. *Angew. Chem., Int. Ed.* **2007**, *46*, 1905. (k) Zhang, M.; Jiang, H.-F.; Neumann, H.; Beller, M.; Dixneuf, P. H. *Angew. Chem., Int. Ed.* **2009**, *48*, 1681.

10.1021/ol101080q
Published on Web 06/03/2010

an ester, or a cyano group.[9] Therefore, a strong demand remains to develop an efficient approach.

Herein, we wish to report a novel preparation of tetrasubstituted furans **1**, **2**, or **3** starting from Bu_3P, the corresponding Michael acceptors **4**, **5**, or **6**, and acid chlorides **7** in the presence of Et_3N in one step (Scheme 1). To the best of our knowledge,

Scheme 1. Preparation of Fully Functionalized Furans **1**, **2**, or **3** from **4**, **5**, or **6**, Acid Chlorides **7**, and Bu_3P in the Presence of Et_3N

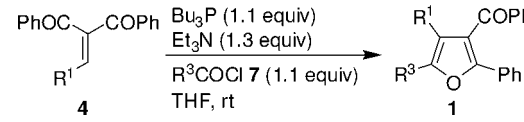


R^1 = aryl or CO_2Et
R^2 = aryl or alkyl
R^3 = aryl, alkyl or vinyl
FG = COR^2 (**1** and **4**), CO_2Et (**2** and **5**), or CN (**3** and **6**)

there are no reports of successful reactions or related studies that utilize this strategy.[10,11] In addition, different kinds of **4**, **5**, or **6** in combination with various acid chlorides **7** should make this methodology an attractive approach toward a wide diversity of substitution patterns in the furan rings.

Thus, the Michael acceptor **4a**, Bu_3P (1.1 equiv), Et_3N (1.3 equiv), and the acid chloride **7a** (1.1 equiv) reacted smoothly at room temperature within 1.5 h, furnishing the highly substituted furan **1aa** in 91% yield (Table 1, entry 1).[12] Similarly, the reaction of **4a** and the other aryl-substituted acid chlorides bearing an electron-withdrawing group, such as **7b** or **7c**, or that with an electron-donating group, like **7d** or **7e**, proceeded efficiently at room temperature within 1.5 h, providing **1ab**, **1ac**, **1ad**, or **1ae** in 82%, 86%, 97%, or 92% yield, respectively (entries 2−5). A heteroaryl-substituted acid chloride, like **7f**, and benzoyl chloride (**7g**) also worked nicely with **4a** in the presence of Bu_3P and Et_3N within 1.5 or 3 h, giving **1af** or **1ag** in 75% or 98% yield, respectively (entries 6 and 7). Other Michael acceptors, such as **4b**−**i** (R^1 = 4-BrC_6H_4, 4-ClC_6H_4, 4-CNC_6H_4, 2-BrC_6H_4, 2-ClC_6H_4, 2-furyl, 2-thienyl, or CO_2Et), reacted successfully with various acid chlorides (**7b**,**c**, **7e**,**f**, or **7h**,**i**) within 2−21 h, leading to the corresponding

Table 1. Syntheses of Furans **1** from **4** and Acid Chlorides **7**[a]



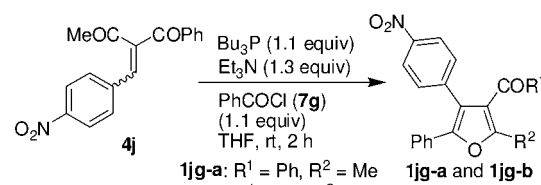
entry	R^1 (**4**)	R^3 (**7**)	time (h)	yield of **1**[b] (%)
1	4-$NO_2C_6H_4$ (**4a**)	4-$NO_2C_6H_4$ (**7a**)	1.5	**1aa**, 91
2	4-$NO_2C_6H_4$ (**4a**)	4-BrC_6H_4 (**7b**)	1.5	**1ab**, 82
3	4-$NO_2C_6H_4$ (**4a**)	4-ClC_6H_4 (**7c**)	1.5	**1ac**, 86
4	4-$NO_2C_6H_4$ (**4a**)	4-MeC_6H_4 (**7d**)	1.5	**1ad**, 97
5	4-$NO_2C_6H_4$ (**4a**)	4-$MeOC_6H_4$ (**7e**)	1.5	**1ae**, 92
6	4-$NO_2C_6H_4$ (**4a**)	2-furyl (**7f**)	1.5	**1af**, 75
7	4-$NO_2C_6H_4$ (**4a**)	Ph (**7g**)	3	**1ag**, 98
8	4-BrC_6H_4 (**4b**)	4-ClC_6H_4 (**7c**)	4	**1bc**, 84
9	4-BrC_6H_4 (**4b**)	2-ClC_6H_4 (**7h**)	9	**1bh**, 80
10	4-BrC_6H_4 (**4b**)	4-$MeOC_6H_4$ (**7e**)	19	**1be**, 85
11	4-ClC_6H_4 (**4c**)	2-furyl (**7f**)	21	**1cf**, 71
12	4-ClC_6H_4 (**4c**)	4-ClC_6H_4 (**7c**)	19	**1cc**, 81
13	4-CNC_6H_4 (**4d**)	4-$MeOC_6H_4$ (**7e**)	2	**1de**, 84
14	2-BrC_6H_4 (**4e**)	4-BrC_6H_4 (**7b**)	18	**1eb**,[c] 79
15	2-BrC_6H_4 (**4e**)	2-$NO_2C_6H_4$ (**7i**)	7	**1ei**, 80
16	2-ClC_6H_4 (**4f**)	2-$NO_2C_6H_4$ (**7i**)	4	**1fi**, 84
17	2-ClC_6H_4 (**4f**)	4-ClC_6H_4 (**7c**)	17	**1fc**, 70
18	2-furyl (**4g**)	4-$MeOC_6H_4$ (**7e**)	2	**1ge**, 80
19	2-furyl (**4g**)	2-ClC_6H_4 (**7h**)	2	**1gh**, 93
20	2-furyl (**4g**)	2-furyl (**7f**)	21	**1gf**, 71
21	2-thienyl (**4h**)	4-$MeOC_6H_4$ (**7e**)	6	**1he**, 72
22	CO_2Et (**4i**)	2-$NO_2C_6H_4$ (**7i**)	2	**1ii**, 82

[a] Reactions were carried out with **4** (0.5 mmol) in THF (2.5 mL) under nitrogen at rt. [b] Yield of isolated products. [c] The structure of **1eb** was confirmed by X-ray analysis (CCDC no. 769220).

products **1** in 70−93% yields (entries 8−22). The steric effect was observed when an acid chloride, like **7c** (R^3 = 4-ClC_6H_4) or **7h** (R^3 = 2-ClC_6H_4), participated in our designed reaction with **4b**, furnishing the corresponding furan **1bc** or **1bh** in 84% or 80% yield within 4 or 9 h, respectively (entries 8 and 9). Surprisingly, the expected more challenging case, such as the reaction of **4e** (R^1 = 2-BrC_6H_4) or **4f** (R^1 = 2-ClC_6H_4) with **7i** (R^3 = 2-$NO_2C_6H_4$), was successfully achieved within 7 or 4 h, providing the corresponding furan **1ei** or **1fi** in 80% or 84% yield, respectively (entries 15 and 16).

Instead of using the Michael acceptor with the same ketone functionality (Table 1), the substrate bearing two different ketone functions, such as **4j**, was also studied (Scheme 2). The reaction of **4j** and **7g** in the presence of Bu_3P and Et_3N

Scheme 2. Preparation of Furans **1jg-a** and**1jg-b**



1jg-a: R^1 = Ph, R^2 = Me
1jg-b: R^1 = Me, R^2 = Ph

1jg-a and **1jg-b** (60% yield; 84:16)

(8) For recent selected examples of other tetrasubstituted furans with 2-(1-alkynyl)-2-alken-1-ones as substrates catalyzed by transition metal, see: (a) Liu, R.; Zhang, J. *Chem.—Eur. J.* **2009**, *15*, 9303. (b) Xiao, Y.; Zhang, J. *Adv. Synth. Catal.* **2009**, *351*, 617. (c) Xiao, Y.; Zhang, J. *Chem. Commun.* **2009**, 3594. (d) Liu, F.; Zhang, J. *Angew. Chem., Int. Ed.* **2009**, *48*, 5505. (e) Gao, J.; Xhao, X.; Yu, Y.; Zhang, J. *Chem.—Eur. J.* **2010**, *16*, 456. (f) Zhang, Y.; Chen, Z.; Xiao, Y.; Zhang, J. *Chem.—Eur. J.* **2009**, *15*, 5208.

(9) For the only example of tetrasubstituted furans bearing three phenyl groups and a ketone function with poor yield, see: (a) Trisler, J. C.; Doty, J. K.; Robinson, J. M. *J. Org. Chem.* **1969**, *34*, 3421. For the only example of tetrasubstituted furans having three phenyl groups and an ester function synthesized in multiple steps, see: (b) Tseng, J.-C.; Chen, J.-H.; Luh, T.-Y. *Synlett.* **2006**, 1209.

(10) To the best of our knowledge, there is no report with phosphorous ylides for the syntheses of furans.

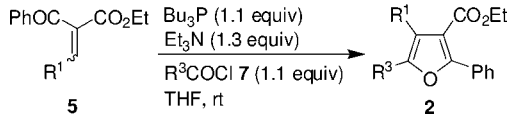
(11) For interesting phosphine-mediated reductive cyclizations of γ-acyloxy butynoates to furans, see: Jung, C.-K.; Wang, J.-C.; Krische, M. J. *J. Am. Chem. Soc.* **2004**, *126*, 4118.

(12) In our one-step protocol, the addition sequence of reactants has no influence on the results of the formation of furans.

proceeded smoothly within 2 h at room tempearture, giving rise to the fully functionalized furans **1jg-a** and **1jg-b** in 60% yield (**1jg-a**:**1jg-b** = 84:16).

The broad reaction scope of our protocol was demonstrated by further studies disclosed in Table 2. It showed that the

Table 2. Syntheses of Furans **2** from **5** and Acid Chlorides **7**[a]



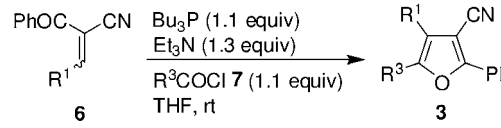
entry	R^1 (**5**)	R^3 (**7**)	time (h)	yield of **2**[b] (%)
1	4-$NO_2C_6H_4$ (**5a**)	4-$NO_2C_6H_4$ (**7a**)	2	**2aa**, 88
2	4-$NO_2C_6H_4$ (**5a**)	4-BrC_6H_4 (**7b**)	2	**2ab**, 94
3	4-$NO_2C_6H_4$ (**5a**)	4-ClC_6H_4 (**7c**)	2	**2ac**, 97
4	4-$NO_2C_6H_4$ (**5a**)	4-MeC_6H_4 (**7d**)	2	**2ad**, 99
5	4-$NO_2C_6H_4$ (**5a**)	4-$MeOC_6H_4$ (**7e**)	2	**2ae**, 93
6	4-$NO_2C_6H_4$ (**5a**)	2-$NO_2C_6H_4$ (**7i**)	2	**2ai**, 87
7	4-$NO_2C_6H_4$ (**5a**)	2-ClC_6H_4 (**7h**)	2	**2ah**, 92
8	4-$NO_2C_6H_4$ (**5a**)	3-ClC_6H_4 (**7j**)	2	**2aj**, 94
9	4-$NO_2C_6H_4$ (**5a**)	Ph (**7g**)	2	**2ag**,[c] 93
10	4-$NO_2C_6H_4$ (**5a**)	CH_3 (**7k**)	4	**2ak**, 70
11	4-BrC_6H_4 (**5b**)	4-ClC_6H_4 (**7c**)	12	**2bc**, 95
12	4-BrC_6H_4 (**5b**)	2-ClC_6H_4 (**7h**)	12	**2bh**, 90
13	4-BrC_6H_4 (**5b**)	2-furyl (**7f**)	12	**2bf**, 75
14	4-BrC_6H_4 (**5b**)	2-thienyl (**7*l***)	8	**2b*l***, 82
15	4-CNC_6H_4 (**5c**)	4-ClC_6H_4 (**7c**)	4	**2cc**, 90
16	4-CNC_6H_4 (**5c**)	4-$MeOC_6H_4$ (**7e**)	4	**2ce**, 85
17	4-CNC_6H_4 (**5c**)	2-$NO_2C_6H_4$ (**7i**)	4	**2ci**, 92
18	4-CNC_6H_4 (**5c**)	2-thienyl (**7*l***)	4	**2c*l***, 91
19	2-BrC_6H_4 (**5d**)	4-ClC_6H_4 (**7c**)	12	**2dc**, 86
20	2-BrC_6H_4 (**5d**)	2-ClC_6H_4 (**7h**)	12	**2dh**, 80
21	2-furyl (**5e**)	4-ClC_6H_4 (**7c**)	12	**2ec**, 85
22	2-furyl (**5e**)	4-$MeOC_6H_4$ (**7e**)	4	**2ee**, 80
23	2-furyl (**5e**)	2-thienyl (**7*l***)	9	**2e*l***, 80
24	2-thienyl (**5f**)	4-ClC_6H_4 (**7c**)	12	**2fc**, 80
25	1-naphthyl (**5g**)	4-ClC_6H_4 (**7c**)	12	**2gc**, 84
26	2-naphthyl (**5h**)	4-ClC_6H_4 (**7c**)	12	**2hc**, 88
27	CO_2Et (**5i**)	4-ClC_6H_4 (**7c**)	4	**2ic**, 83

[a] Reactions were carried out with **5** (0.5 mmol) in THF (2.5 mL) under nitrogen at rt. [b] Yield of isolated products. [c] The structure of **2ag** was confirmed by X-ray analysis (CCDC no. 768639).

reactions of Michael acceptors bearing a ketone and an ester group, like **5a**−**i** (R^1 = 4-$NO_2C_6H_4$, 4-BrC_6H_4, 4-CNC_6H_4, 2-BrC_6H_4, 2-furyl, 2-thienyl, 1-naphthyl, 2-naphthyl, or CO_2Et), and acid chlorides **7a**−***l*** (1.1 equiv) in the presence of Bu_3P (1.1 equiv) and Et_3N (1.3 equiv) took place in 2−12 h, leading to the corresponding adducts **2** in 70−99% yields (entries 1−27).

Remarkably, the Michael acceptor **6** having a ketone and a cyano group exhibited a significant enhancement in reactivity for the formation of the corresponding furan **3** in comparison with **4** or **5** for the formation of the corresponding furan **1** or **2**. Excellent results, for example, using **6a**−**h** and **7c**, **7g**, or **7h** for syntheses of furans **3**, were shown in Table 3. Under the same reaction condition for the preparation of **1** or **2**, the corresponding furans **3** were efficiently

Table 3. Syntheses of Furans **3** from **6** and Acid Chlorides **7**[a]



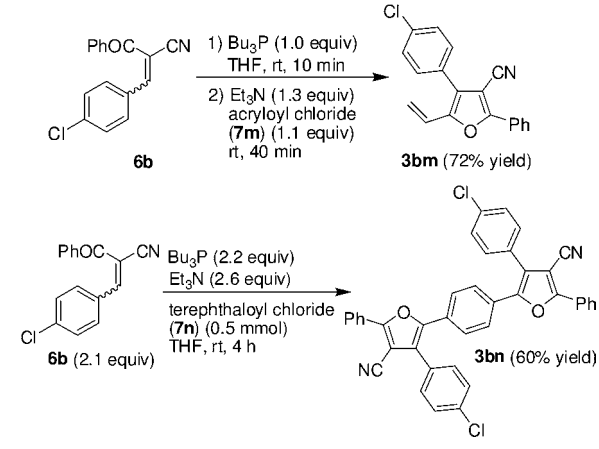
entry	R^1 (**6**)	R^3 (**7**)	time (min)	yield of **3**[b] (%)
1	4-$NO_2C_6H_4$ (**6a**)	4-ClC_6H_4 (**7c**)	10	**3ac**,[c] 95
2	4-$NO_2C_6H_4$ (**6a**)	2-ClC_6H_4 (**7h**)	10	**3ah**, 89
3	4-$NO_2C_6H_4$ (**6a**)	Ph (**7g**)	10	**3ag**, 88
4	4-ClC_6H_4 (**6b**)	4-ClC_6H_4 (**7c**)	10	**3bc**, 93
5	4-CNC_6H_4 (**6c**)	4-ClC_6H_4 (**7c**)	10	**3cc**, 95
6	4-CNC_6H_4 (**6c**)	2-ClC_6H_4 (**7h**)	10	**3ch**, 85
7	4-$CH_3C_6H_4$ (**6d**)	4-ClC_6H_4 (**7c**)	40	**3dc**, 84
8	2-BrC_6H_4 (**6e**)	4-ClC_6H_4 (**7c**)	40	**3ec**, 85
9	Ph (**6f**)	2-ClC_6H_4 (**7h**)	240	**3fh**, 84
10	2-furyl (**6g**)	4-ClC_6H_4 (**7c**)	10	**3gc**, 99
11	2-thienyl (**6h**)	4-ClC_6H_4 (**7c**)	10	**3hc**, 91

[a] Reactions were carried out with **6** (0.5 mmol) in THF (2.5 mL) under nitrogen at rt. [b] Yield of isolated products. [c] The structure of **3ac** was confirmed by X-ray analysis (CCDC no. 769770).

furnished at room temperature within 10−40 min in 84−99% yields (entries 1−8, 10, and 11), except for **3fh** (84% yield; 4 h; entry 9).

Furthermore, acryloyl chloride (**7m**), which was prone to undergo polymerization catalyzed by Bu_3P, reacted successfully with **6b**, Bu_3P, and Et_3N at room temperature within 40 min, leading to the corresponding furan **3bm** in 72% yield (Scheme 3). Interestingly, the reaction of terephthaloyl

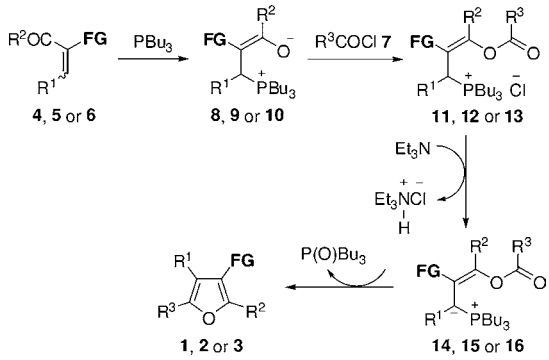
Scheme 3. Preparation of Highly Functionalized Furans **3bm** and **3bn**



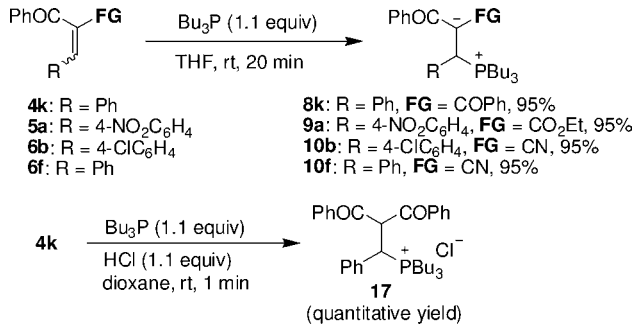
chloride (**7n**) and **6b** in the presence of Bu_3P and Et_3N occurred smoothly at room temperature within 4 h, providing the corresponding furan **3bn** in 60% yield.

On the basis of experimental results (Tables 1−3, Schemes 2 and 3), a plausible reaction mechanism was proposed (Scheme 4). First, the Michael addition of Bu_3P toward **4**,

Scheme 4. A Proposed Mechanism for the Formation of **1**, **2**, or **3**

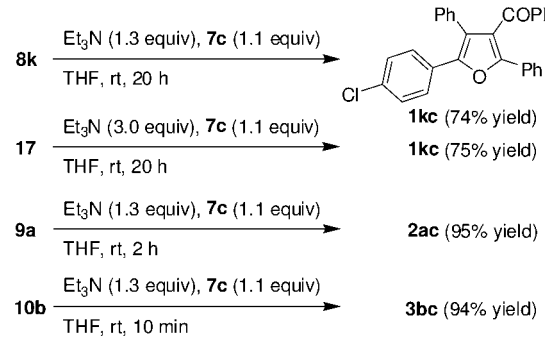


Scheme 5. Preparation of **8k**, **9a**, **10b**, **10f**, and **17**



9a, **10b**, or **10f**, resulted effectively from the addition of Bu_3P toward **4k**, **5a**, **6b**, or **6f**, and **8k**, **9a**, and **10f** were characterized with X-ray crystallography (Scheme 5).[14,15] When **4k** was treated with Bu_3P (1.1 equiv) and HCl (1.1 equiv; 1 N in dioxane), the adduct **17** was obtained in quantitative yield.[15]

Scheme 6. Preparation of Furans **1kc**, **2ac**, and **3bc** from **8k** (or **17**), **9a**, and **10b**



Besides, the reaction of the intermediate zwitterion, such as **8k**, **9a**, or **10b**, and an acid chloride, such as **7c**, in the presence of Et_3N (1.3 equiv) indeed took place smoothly at room temperature within 20 h, 2 h, or 10 min, giving the corresponding furan **1kc**, **2ac**, or **3bc** in 74%, 95%, or 94% yield, respectively (Scheme 6). The reaction of the phosphonium chloride **17** and **7c** in the presence of Et_3N (3.0 equiv) also took place smoothly, providing **1kc** in good yield (75%). All of this evidence showed that a zwitterion like **8**, **9**, or **10** was the intermediate for the formation of the corresponding furan **1**, **2**, or **3** (Schemes 5 and 6).

In conclusion, we have developed a general procedure for novel syntheses of tetrasubstituted furans **1**, **2**, or **3**. The reaction condition is very mild, and numerous Michael acceptors **4**, **5**, or **6** and acid chlorides **7** can be applied efficiently in one step to afford **1**, **2**, or **3** in high yields. The reaction mechanism is proposed to undergo the Michael reaction of Bu_3P and **4**, **5**, or **6** followed by acylation with **8**, **9**, or **10**, deprotonation of the corresponding intermediate **11**, **12**, or **13**, and finally an intramolecular Wittig reaction of **14**, **15**, or **16**. In addition, the easy access to acid chlorides **7** as well as Michael acceptors **4**, **5**, or **6**, which result from the condensation of aldehydes and 1,3-diketone, ethyl benzoylacetate, or benzoylacetonitrile, makes our protocol an attractive approach toward a wide diversity of substitution patterns in the furan rings. Further studies and the extensions of this concept in the preparation of other heterocycles are currently underway.

Acknowledgment. The authors thank the National Science Council of the Republic of China (NSC Grant No. 97-2113-M-003-003-MY2) and National Taiwan Normal University (98031013) for financial support.

Supporting Information Available: General experimental procedures, compound characterization data, and X-ray and NMR spectra. This material is available free of charge via the Internet at http://pubs.acs.org.

OL101080Q

(13) For selected reviews of Wittig reactions, see: (a) Hoffmann, R. W. *Angew. Chem., Int. Ed.* **2001**, *40*, 1411. (b) Lawrence, N. J. *Preparation of Alkenes: a Practical Approach*; Williams, J. M. J., Ed.; Oxford University Press: Oxford, UK, 1995. (c) *Phosphorus Ylides: Chemistry and Applications in Organic Chemistry*; Kolodiazhnyi, O. I., Ed.; Wiley-VCH: New York, 1999.

(14) For another method to prepare the phosphonium enolate zwitterions of type **9** starting from tertiary phosphines, 4-pyridinecarboxaldehyde (3 equiv), and alkynoates (1 equiv) within 0.5−12 h, please see: Zhu, X.-F.; Henry, C. E.; Kwon, O. *J. Am. Chem. Soc.* **2007**, *129*, 6722.

(15) The structures of adducts **8k**, **9a**, **10f**, and **17** were confirmed by ^1H, ^{13}C, and ^{31}P NMR and X-ray analyses (CCDC no. 769222 for **8k**, 771901 for **9a**, 768638 for **10f**, and 746224 for **17**).