Tetrabutylammonium Fluoride Promoted Intramolecular Nucleophilic Attack of a Carbodiimide Group on an α,β-Unsaturated Ester Group

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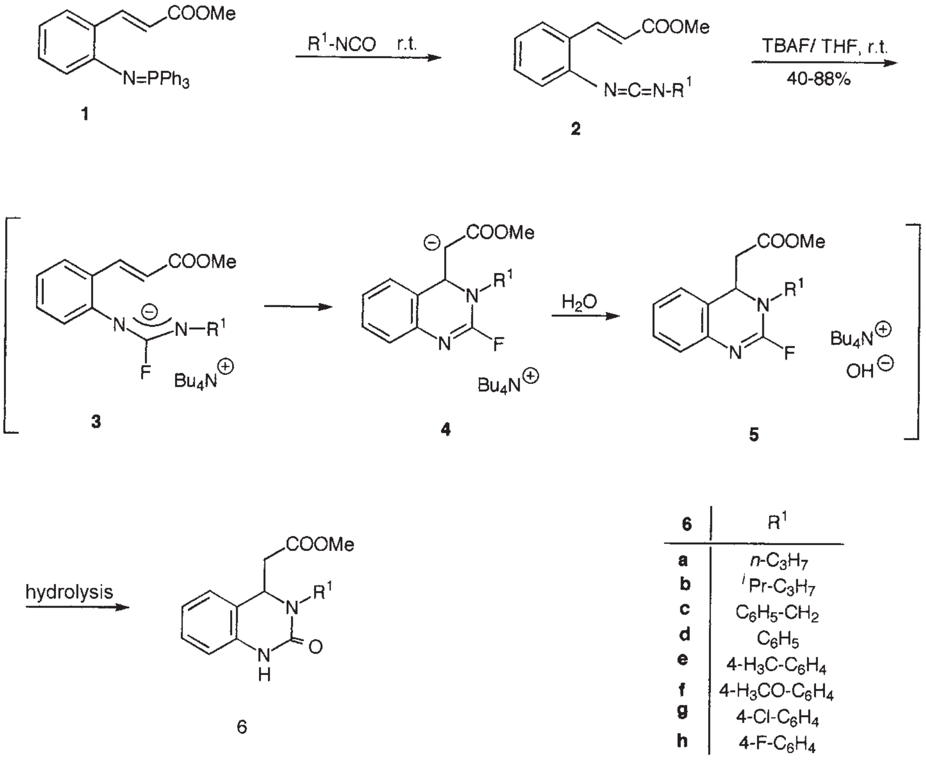
Abstract: (*N*-Aryl, *N'*-aryl/alkyl) carbodiimides bearing an α,β-unsaturated ester moiety in the *ortho* position undergo cyclization in the presence of tetrabutylammonium fluoride under mild conditions to give dihydroquinazoline derivatives in synthetically useful yields.

Key words: carbodiimide, tetrabutylammonium fluoride, Michael Addition, dihydroquinazolines, aza Wittig reaction

We have reported[1] that *N,N'* -diarylcarbodiimides bearing one *o*-vinyl substituent undergo a thermally induced 6π-electrocyclization to give quinoline derivatives. In this context, the so-called tandem aza-Wittig/carbodiimide mediated electrocyclization methodology is successfully utilized in heterocyclic synthesis.[2] We report here the direct conversion of carbodiimides **2**, readily available from iminophosphorane **1**, into dihydroquinazoline derivatives **5** promoted by tetrabutylammonium fluoride (TBAF) in synthetically useful yields. Formally, this conversion involves an intramolecular N-conjugate addition to an α,β-unsaturated ester moiety. In contrast to the various Michael addition methods, there are few examples of heteroconjugated addition of nitrogen compounds such as amines,[3] amides,[4] thiolactam,[5] and guanidines.[6] To the best of our knowledge no examples dealing with heteroconjugate addition of carbodiimides to α,β-unsaturated esters have been previously reported.

Aza-Wittig reaction of iminophosphorane **1**[1] with aliphatic and aromatic isocyanates in dry tetrahydrofuran at room temperature gave the corresponding carbodiimides **2**, which were used for the next step without further purification. When compounds **2** were treated with a solution of TBAF in tetrahydrofuran (1:4 molar ratio) at room temperature for a short period of time (10 min) the dihydroquinazoline derivatives **6** were obtained in yields ranging from 56 to 88% (Scheme 1). When a 1:1 molar ratio was used, the reaction proceeded very slowly and the yields were lower. The best results were obtained when the reactions were carried out in the presence of a mixture of anhydrous $MgSO_4/Na_2SO_4$. When the reaction was carried out in the absence of this dehydrating agent the corresponding ureas derived from **2** were found to be the major product along with a small amount of **5.**

Iminophosphorane **1**, also reacted with several vinyl isocyanates **7** to give the expected carbodiimides **8** (as evidenced by IR) which by treatment with TBAF under the same reaction conditions afforded the corresponding 3-vinyl-3,4-dihydroquinazoline derivatives **9** in yields ranging from 57 to 65%. Attempts to cyclize compounds **9** by the action of TBAF/THF at reflux temperature failed, except for compound **9c** which under the above conditions provided, after dehydrogenation, the tricyclic compound **10** in 60% yield (Scheme 2).



6	R^1
a	n-C_3H_7
b	^{i}Pr-C_3H_7
c	C_6H_5-CH_2
d	C_6H_5
e	4-H_3C-C_6H_4
f	4-H_3CO-C_6H_4
g	4-Cl-C_6H_4
h	4-F-C_6H_4

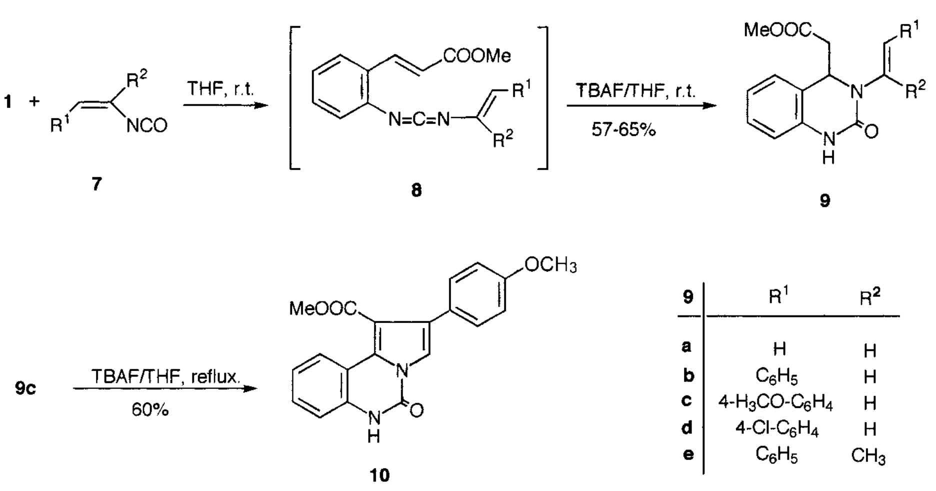
Scheme 1

Table 1. Dihydroquinazolines **6**, **9**, **10**, and **13** Prepared

Product[a]	R^1	R^2	mp (°C)	Yield[b] (%)	MS (70 eV) m/z (%)
6a	n-C_3H_7	–	172–173	64	262 (M^+, 10), 219 (8), 189 (100), 147 (86), 129 (11), 91 (7), 77 (10)
6b	^{i}Pr-C_3H_7	–	133–134	72	262 (M^+, 3), 189 (64), 147 (100), 129 (7)
6c	C_6H_5–CH_2	–	130–131	88	429 (M^+, 1), 237 (59), 219 (12), 91 (100), 65 (25)
6d	C_6H_5	–	205–206	75	296 (M^+, 5), 223 (100), 117 (13), 104 (22), 91 (12), 77 (53)
6e	4-H_3C–C_6H_4	–	167–168	73	310 (M^+, 8), 237 (100), 118 (16), 91 (33), 65 (16)
6f	4-H_3CO–C_6H_4	–	170–171	77	326 (M^+, 6), 253 (100), 181 (6), 162 (7), 149 (9), 134 (32), 117 (26), 107 (14), 92 (27), 77 (43)
6g	4-Cl–C_6H_4	–	211–212	56	332 (M^+ +2, 2), 330 (M^+, 6), 257 (100), 138 (24), 111 (36), 90 (20)
6h	4-F–C_6H_4	–	179–180	40	314 (M^+, 4), 241 (100), 122 (27), 117 (14), 95 (38), 90 (12), 75 (12)
9a	H	H	128–129	60	260 (M^+, 10), 187 (100), 147 (31), 118 (7), 57 (7)
9b	C_6H_5	H	229–230	57	322 (M^+, 17), 249 (100), 204 (18), 147 (16), 132 (18), 117 (22), 103 (18), 91 (17), 77 (31)
9c	4-H_3CO-C_6H_4	H	208–210	60	352 (M^+, 40), 279 (100), 132 (22), 117 (14), 89 (14), 59 (6)
9d	4-Cl–C_6H_4	H	213–214	59	358 (M^+, + 2, 9), 356 (M^+, 27), 283 (100), 162 (16), 147 (29), 132 (24), 102 (41), 89 (38), 77 (18)
9e	C_6H_5	CH_3	157–158	65	336 (M^+, 10), 263 (37), 219 (19), 117 (40), 91 (45), 77 (24), 592 (22)
10	4-H_3CO–C_6H_4	H	160–161	60	349 (M^+ + 1, 1), 336 (17), 263 (50) 247 (18), 219 (25), 147 (100), 132 (10), 128 (22), 117 (35), 115 (55), 91 (33)
13	–	–	234–235	40	380 (M^+, 17), 321 (8), 26 (15), 247 (100), 234 (10), 128 (33), 115 (11), 77 (8)

[a] Satisfactory microanalyses obtained: C ± 0.30, H + 0.28, N + 0.29.

[b] Yield of isolated pure product after chromatographic separation.



9	R^1	R^2
a	H	H
b	C_6H_5	H
c	4-H_3CO-C_6H_4	H
d	4-Cl-C_6H_4	H
e	C_6H_5	CH_3

Scheme 2

Table 2. Spectral Data of Compounds **6, 9, 10**, and **13**

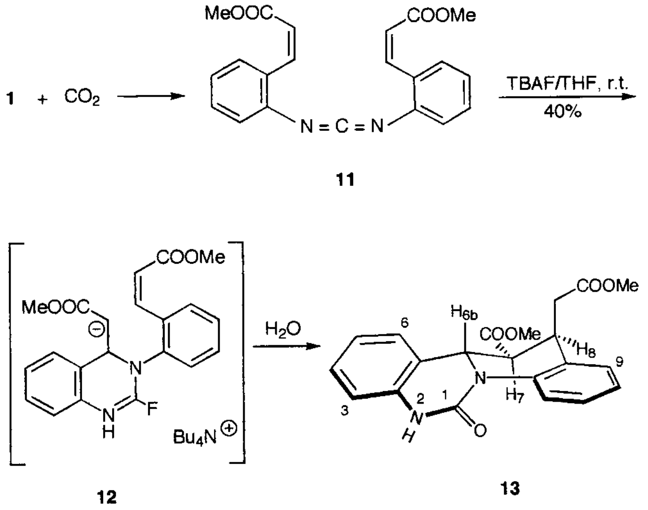
Product	IR (Nujol) ν (cm^{-1})	^{1}H NMR (300 MHz) ($CDCl_3$/TMS) δ, J (Hz)	^{13}C NMR (75 Hz) ($CDCl_3$/TMS) δ
6a	3194, 1728, 1669, 1471, 1297, 749	9.22 (s, 1H, NH), 7.10 (m, 1H), 6.99 (d, 1H, ^{3}J = 7.1), 6.78 (m, 2H), 4.76 (dd, J = 8.2, 5.2, H4), 3.90 (m, 1H), 3.54 (s, 3H, CH_3O), 2.85 (m, 1H), 2.63 (m, 2H, CH_2–COO), 1.57 (m, 2H), 0.83 (t, 3H, ^{3}J = 7.1, CH_3)	170.97 (q, COO), 155.11 (q, C2), 136.80 (q, C8a), 128.43, 125.17, 121.62, 120.98 (q, C4a), 114.15, 55.84 (C4), 51.70 (CH_3O), 47.17 (CH_2), 39.52 ($\mathit{C}H_2$–COO), 21.39 (CH_2), 11.09 (*C*H3-CH_2)
6b	3194, 1737, 1666, 1601, 1453, 1291, 1203, 746	9.04 (s, 1H, NH), 7.10 (m, 2H), 6.82 (m, 2H), 4.83 (dd, 1H, ^{3}J = 8.2, 5.1, H4), 4.53 [m, 1H, ^{3}J = 6.8, $CH(CH_3)_2$], 3.51 (s, 3H, CH_3O), 2.61 (m, 2H), 1.28 (d, 3H, ^{3}J = 6.8, CH_3), 1.18 (d, 3H, ^{3}J = 6.8, CH_3)	170.81 (q, COO), 155.28 (q, C2), 136.82 (q, C8a), 128.35, 124.92, 122.11 (q, C4a), 121.55, 114.07, 51.60 (CH_3O) 51.12 (C4), 47.01 [$\mathit{C}H(CH_3)_2$], 41.25 (CH_2), 21.33 (CH_3), 20.70 (CH_3)
6c	3211, 1732, 1681, 1602, 1504, 1454, 1292, 1149, 756	9.25 (s, 1H, NH), 7.11 (m, 7H), 6.79 (m, 2H), 5.18 (d, 1H ^{3}J = 15.4 H4), 4.73 (dd, 1H, ^{3}J = 7.4, 5.4), 4.15 (d, 1H, ^{3}J = 8.2, 5.1), 3.49 (s, 3H, CH_3O), 2.59 (m, 2H, CH_2–COO)	170.78 (q, COO), 155.28 (q, C2), 137. 10 (q, C_i–Ph), 136.52 (q, C8a), 128.59, 128.53, 127.66, 127.41, 125.27, 121.87, 120.86 (q, C8a), 114.27, 55.11 (C4), 51.72 (CH_3O), 48.59 (CH_2-Ph), 39.40 (CH_2–COO)
6d	3190, 1736, 1673, 1440, 1417, 1216, 1149, 753	8.69 (s, 1H, NH), 7.34 (m, 5H), 7.21 (m, 2H), 6.87 (m, 1H), 6.72 (d, 1H, ^{3}J = 7.9), 5.21 (dd, 1H, ^{3}J = 8.2, 5.1, H4), 3.42 (s, 3H, CH_3O), 2.70 (m, 2H, CH_2–COO)	170.38 (q, COO), 153.73 (q, C2), 140.73 (q, C_i–Ph), 136.48 (q, C8a), 129.15, 128.76, 127.41, 126.85, 125.53, 122.12, 121.04 (q, C4a), 114.44, 59.88 (C4), 51.65 (CH_3O), 39.78 (CH_2)
6e	3187, 1734, 1674, 1595, 1455, 1293, 1146, 763	8.62 (s, 1H, NH), 7.13 (m, 6H) 6.86 (m, 1H), 6.72 (d, 1H, ^{3}J = 1 7.9), 5.15 (dd, 1H, ^{3}J = 8.2, 5.1, H4), 3.42 (s, 3H, CH_3O), 2.73 (m, 2H, CH_2), 2.28 (s, 3 H, CH_3–Ar)	170.39 (q, COO), 153.72 (q, C2), 40.69 (q, C_i–Ar), 136.72 (q), 136.49 (q), 129.17 (C_m–Ar), 128.78, 127.42 (C_0–Ar), 126.89, 125.55, 122.16, 121.03 (q, C4a), 114.15, 59.87 (C4), 51.69 (CH_3O), 39.76 (CH_2–COO), 20.98 (CH_3-Ar)
6f	3220, 1730, 1679, 1602, 1515, 1454 1298, 1247	8.96 (s, 1H, NH), 7.31 (d, 2H, ^{3}J = 8.7), 7.14 (m, 2H), 6.95 (m, 3H), 6.78 (d, 1H, ^{3}J = 7.8), 5.20 (dd, 1H, ^{3}J = 8.2, 5.1, H4), 3.81 (s, 3H, CH_3O), 3.51 (s, 3H, CH_3O), 2.80 (m, 2H, CH_2)	170.40 (q, COO), 158.21 (q, C_p–Ar), 153.93 (q, C2), 136.56 (q, C8a), 133.36 (q, C_0–Ar), 128.91, 128.62, 125.40, 121.91, 120.82 (q, C4a), 114.44, 114.34, 60.11 (C4), 55.35 (CH_3O–Ar), 51.60 (CH_3O), 39.69 (CH_2)
6g	3187, 1731, 1673, 1595, 1462, 1293, 1152, 769	9.04 (s, 1H, NH), 7.30 (s, 4H) 7.10 (m, 2H), 6.89 (m, 1H), 6.71 (d, 1H, ^{3}J = 7.9), 5.18 (dd, 1H, ^{3}J = 8.2, 5.2, H4), 3.44 (s, 3H, CH_3O), 2.66 (m, 2H, CH_2)	170.25 (q, COO), 153.66 (q, C2), 139.21 (q), 136.24 (q, C8a), 132.35 (q), 129.24, 128.85, 128.70, 125.45, 122.28, 120.75 (q, C4a), 114.54, 59.80 (C4), 51.74 (CH_3O), 39.78 $(CH)_2$
6h	3216, 1734, 1680, 1604, 1506, 1458, 1296, 1155, 841, 760	9.07 (s, 1H, NH), 7.31 (m, 2H), 7.08 (m, 4H), 6.87 (m, 1H), 6.70 (d, 1H, ^{3}J = 7.9), 5.14 (dd, 1 H, ^{3}J = 5.2, 5.1, H4), 3.43 (s, 3H, CH_3O), 2.66 (m, 2H, CH_2)	170.30 (q, COO), 161.10 (q,$^{1}J_F$ = 246.6 C_p–Ar), 153.85 (q, C2), 136.58 (q, $^{4}J_F$ = 3.2, C_i–Ar), 136.37 (q, C8a), 129.44 ($^{3}J_F$ = 8.5), 128.78, 128.78, 125.43, 122.18, 120.71 (C4a), 115.97 ($^{2}J_F$ = 22.65), 114.53 (CH_3O) 60.06 (C4), 51.70 (CH_3O), 39.77 (CH_2)
9a	3204, 1740, 1679, 1604, 1295, 1217, 1200, 993, 953, 760	9.40 (s, 1H, NH), 7.15 (m, 1H), 7.02 (d, 1H, ^{3}J = 7.2), 6.85 (m, 2H), 5.77 (m, 1H), 5.14 (m, 2H), 4.73 (dd, 1H, ^{3}J = 8.1, 4.6, H4), 4.45 (m, 1H), 3.61 (m, 1H), 3.51 (s, 3H, CH_3), 2.79 (dd, 1H, ^{2}J = 14.6, 4.6), 2.52 (dd, 1H, ^{2}J = 14.6, 8.1)	170.00 (q, COO), 153.20 (C2), 137.29 (q, C8a), 134.23, 128,34, 125.44, 121.21, 120.92 (q, C4a), 116.94 (CH_2), 113.76, 54.50 (C4), 51.52 (CH_3O), 46.56 (CH_2), 39.09 (CH_2–COO)
9b	3207, 1728, 1693, 1682, 1644, 1609, 1506, 1454, 1326, 1134, 950, 755	10.05 (s, 1H, NH), 7.80 (d, 1H, ^{3}J = 15.3), 7.28 (m, 7H), 6.95 (m, 2H), 6.19 (d, 1H, ^{3}J = 15,3), 5.55 (dd, 1H, ^{3}J = 7.5, 4.9, H4), 3.52 (s, 3H, CH_3), 2.76 (m, 2H, CH_2)	169.98 (q, COO), 150.65 (C2), 136.90 (q, C_i–Ph), 135.66 (q, C8a), 128.70, 125.94, 125.45, 125.12, 121.86, 119.96 (q, C4a), 114.04, 108.04, 52.26 (C4), 51.54 (CH_3O), 38.56 (CH_2)

Table 2. (continued)

Product	IR (Nujol) ν (cm^{-1})	^{1}H NMR (300 MHz) (CDCl$_3$/TMS) δ, J (Hz)	^{13}C NMR (75 Hz) (CDCl$_3$/TMS) δ
9c	3198, 1729, 1682, 1602, 1507, 1247, 1178, 1035, 950, 839, 749	10.00 (s, 1H, NH), 7.67 (d, 1H, ^3J = 15.2), 7.26 (m, 4H), 6.92 (m, 4H, ^3J = 7.4), 6.14 (d, 1H, ^3J = 15.2), 5.52 (d, 1H,^3J = 7.1, 4.4, H4), 3.72 (s, 3H, CH$_3$O), 3.51 (s, 3H, CH$_3$), 2.74 (m, 2H, CH$_2$)^a	70.09 (q, COO), 157.86 (q, C$_p$–Ar), 150.80 (q, C2), 135.82 (q), 129.39, 128.67 (q), 126.36, 125.92, 123.83, 121.85, 120.03 (q), 114.24, 114.10, 108.03, 55.05 (CH$_3$), 52.38, 51.56 (CH$_3$), 38.62 (CH$_2$)^a
9d	3190, 1732, 1689, 1424, 1324	10.06 (s, 1H, NH), 7.80 (d, 1H, ^3J = 15.2), 7,34 (m, 3H), 7.21 (d, 2H, ^3J = 7.4), 6.95 (m, 2H), 6.16 (d, 1H,^3J = 15.2), 5.53 (dd, 1H, ^3J = 7.3, 4.8, H4), 3.51 (s, 3H, CH$_3$), 2.75 (m, 2H, CH$_2$)^a	170.05 (q, COO), 150.70 (C2), 136.07 (q, C$_i$–Ph), 135.71 (q, C8a), 130.22 (q) 128.71, 126.83, 126.31, 126.01, 122.00, 120.03 (q, C4a), 114.20, 106.88, 52.39 (C4), 51.63 (CH$_3$O), 38.63 (CH$_2$)^a
9e	3192, 1735, 1166, 1597, 1285, 1168, 1035, 977, 750, 696	9.56 (s, 1H, NH), 7.39 (m, 3H), 7.19 (m, 4H), 6.89 (m, 2H), 6.56 (s, 1H, PhCH=), 5.18 (dd, 1H, ^3J = 7.2, 4.8, H4), 3.48 (s, 3H, CH$_3$O), 2.92 (dd, 1H, ^2J = 14.6, ^3J = 4.8), 2.70 (dd, 1H, ^2J = 14.6, ^3J = 7.2), 2.09 (s, 3H, CH$_3$)^a	170.25 (q, COO), 151.76 (q, C2), 138.05 (q), 137.01 (q), 136.00 (q, C8a), 128.81, 128.39, 128.33, 126.89, 126.51, 125.55, 121.30, 121.20 (q, C4a), 113.79, 57.99 (C4), 51.47 (CH$_3$O), 40.60 (CH$_2$), 18.34 (CH$_3$)^a
10	3211, 1731, 1660, 1602, 1418, 1288, 1235, 1207, 1149, 914, 771, 726, 700	12.21 (s, 1H, NH), 8.52 (s, 1H), 7.86 (d, 1H, ^3J = 7.9), 7.57 (m, 2H), 7.39 (d, 2H, ^3J = 8.6), 7.25 (m, 2H), 7.05 (d, 2H, ^3J = 8.6), 3.87 (s, 3H, CH$_3$), 3.81 (s, 3H, CH$_3$)^a	168.18 (q, COO), 158.91 (q, C$_p$–Ar), 151.11 (q), 147.00, 139.81 (q), 131.98 (q), 129.81, 129.62 (q), 127.53 (CH), 125.17 (q), 121.85, 120.06, 118.51 (q), 114.36, 111.83, 110.5 (q), 55.24 (CH$_3$), 52.78 (CH$_3$)^a
13	3191, 1734, 1680, 1593, 1169, 1127, 763	9.02 (s, 1H, NH), 7.65 (d, 1H, ^3J = 7.8), 7.24 (m, 2H), 7.11 (m, 1H), 6.95 (m, 2H), 5.02 (d, 1H, ^3J = 10.5, H6b), 3.82 (m, 1H, H8), 3.54 (dd, 1H,^3J=10.5, 4.4, H7), 2.78 (m, 2H, CH$_2$)	171.93 (q, COO), 154.45 (Cl), 136.96 (q), 136.37 (q), 131.09 (q), 128.70, 127.46, 127.03, 124.42, 123.81, 123.64, 122.58, 55.05, 52.41 (CH$_3$), 51.78, (CH$_3$), 47.98, 35.58, 35.03 (CH$_2$)

^{a} Recorded in DMSO-d_6.

Finally, the reaction of iminophosphorane **1** with solid carbon dioxide in a sealed tube at 110°C yielded the carbodiimide **11**, which was isolated without further purification for the next step. Reaction of compound **11** with the system TBAF/THF under standard conditions (Scheme 3) gave the tetracyclic compound **13** as the sole reaction product in 41% yield. It is worth noting that the conversion **11** → **13** implies the formation of three stereogenic centers in only one-step. The structure **13** is assigned on the basis of the coupling pattern of the tetrahydropyridine ring protons in the ^{1}H NMR spectrum: it shows both axial-axial and axial-equatorial H-H relationships between the neighboring H-6b, H-7 and H-8 protons.



Scheme 3

As far as the mechanism of the conversion **2**→**6** is concerned, it must be pointed out that ureas derived from **2** were recovered unchanged after further treatment with TBAF/THF at room temperature. Hence, it is thought that the TBAF strongly increases the nucleophilic character of the nitrogen atom of the carbodiimide[7] **2** to give, presumably, an intermediate like **3**, which by attack of the nitrogen atom on the *o*-vinyl side chain in a Michael-type addition leads to the cyclized product **4**. Eventually, compound 4 by the action of the water present in the TBAF/THF solution affords tetrabutylammonium hydroxide and the fluoropyrimidine derivative **5**, which is hydrolyzed under the reaction conditions[8] to the final product **6**. Conversion **11** → **13** could be rationalized by initial formation of the intermediate **12** which undergoes intramolecular Michael addition with concomitant hydrolysis of the resulting fluoropyrimidine derivative as described above to give the final product **13**.

In conclusion, this work shows, for the first time, that the intramolecular addition of a carbodiimide group to an α,β-unsaturated ester moiety, which represents the formal addition of a urea group, can be efficiently achieved by using TBAF as cyclization agent.

Dihydroquinazoline Derivatives 6 and 9; Geueral Procedure:
To a mixture of the iminophosphorane **1** (0.498 g, 1. 14 mmol), anhyd THF (20 mL), anhyd $MgSO_4$ (1.5 g) and anhyd Na_2SO_4 (1.5 g), was added the corresponding isocyanate (1.14 mmol) under N_2. The resultant mixture was stirred at r.t. for 1 h. 1 M TBAF in THF (4.6 mL, 4.6 mmol) was added and stirring was continued for 10 min. The mixture was poured into Na_2HPO_4 buffer (pH 7, 20 mL) and extracted with EtOAc (2 × 10 mL). The organic layers were washed with sat. brine (2 × 10 mL), water (2 × 10 mL) and finally dried ($MgSO_4$). After filtration the solution was concentrated to dryness and the crude product was chromatographed (silica gel column, Et_2O) to give **6** or **9** which were recrystallized from Et_2O/hexane.

Pyrrolo[1,2-*c*]quinazoline Derivative 10:
To a solution of dihydroquinazoline **9c** (0.25 g, 0.7 mmol) in anhyd THF (20 mL) was added 1 M TBAF in THF (4.6 mL, 4.6 mmol). The resultant mixture was stirred at reflux temperature for 24 h. Workup was similar to the preparation of compounds **6** and **9** (Table 1).

Dihydroquinazoline Derivative 13:
A mixture of iminophosphorane **1** (0.498 g, 1.14 mmol), excess of solid CO_2 and dry THF (20 mL) was heated at 110°C for 4 h in a sealed tube. After cooling, the solvent was removed and the crude carbodiimide **11** was dissolved in dry THF (15 mL). To the resultant solution anhyd $MgSO_4$ (1.5 g), anhyd Na_2SO_4 (1.5 g) and 1 M TBAF in THF (4.6 mL, 4.6 mmol) were added. The resultant mixture was stirred at r.t. for 2 h, then extracted with EtOAc (3 × 15 mL). The combined organic layers were dried ($MgSO_4$), filtered and concentrated to dryness. The crude product was chromatographed on a silica gel column (40 × 3.5 cm, 70–230 mesh) using Et_2O as eluent to give **13** (Table 1).

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