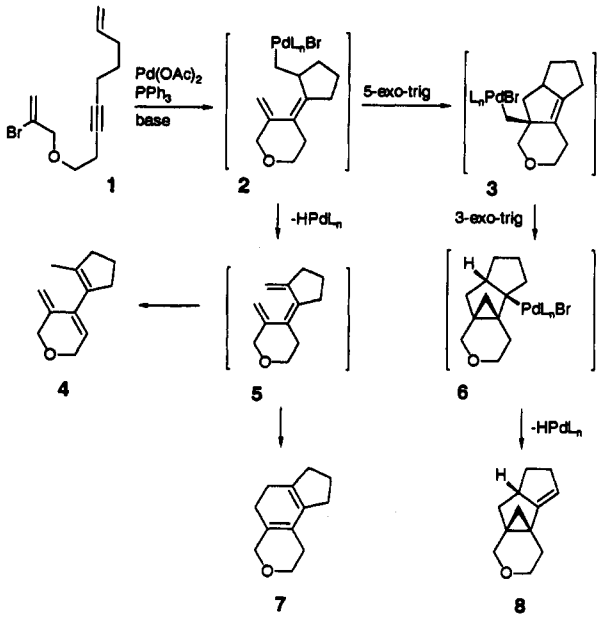
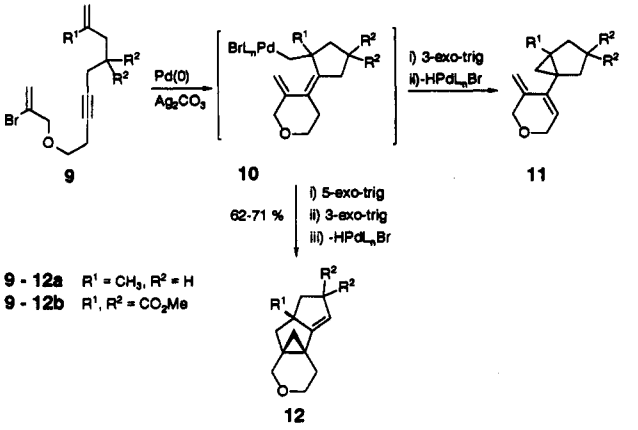
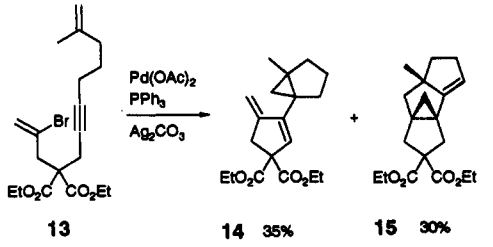


tricyclic intermediate **3**, which has no other choice but to cyclize again with formation of a three-membered ring. The secondary alkylpalladium bromide **6** thus formed can eventually eliminate β-hydride and give the byproduct **8**. The β-hydride elimination in **2** can be prevented with a suitable substituent in the 12-position of the starting dienyne **1**. Thus, dienyne **9a**,[9] when treated with 3–5 mol % $Pd(PPh_3)_4$ and 2 equiv of silver carbonate in refluxing acetonitrile for 3 days, formed the tetracyclic compound **12a** as the sole product, isolated in 62% yield. Moreover, when dienyne **9b** was subjected to similar cyclization conditions (3–5 mol % $Pd(OAc)_2$, 12–20 mol % PPh_3, and 2 equiv of silver carbonate, acetonitrile, 3 h, 130 °C), tetracycle **12b** was obtained in 71% isolated yield.[10]



Under these circumstances, the alkylpalladium species **10** apparently prefers to undergo a 5-*exo-trig* ring closure to an intermediate of type **3** over a 3-*exo-trig* process eventually leading to **11**.[2a] With potassium carbonate instead of silver carbonate, however, small amounts (4%) of the isomeric product **11** could be detected by ^1H NMR spectroscopy.

These observations tempted us to try the assembly of a linear triquinane skeleton containing no heteroatoms. For example, dienyne **13** was cyclized by using 3 mol % $Pd(OAc)_2$, 12 mol % PPh_3, and 2 equiv of silver carbonate in DME/acetonitrile (1:1) at 60 °C for 2 days. After workup, compounds **14**, **15**, and an as yet unidentified product were isolated in 35, 30, and 10% yield, respectively.



Further studies to use this tetrakiscyclization process for the single-step construction of triquinane systems and applications toward natural product synthesis are in progress.

Acknowledgment. Financial support of this work by the "Volkswagen-Stiftung" and the "Fonds der Chemischen Industrie" is gratefully acknowledged. We are indebted to BASF, BAYER, HOECHST, and DEGUSSA AG for generous gifts of chemicals. F.E.M. thanks the "Studienstiftung des Deutschen Volkes" for a scholarship.

(9) Dienyne **9a** can be prepared from 8-methylnon-8-en-3-yn-1-ol and 2,3-dibromopropene under phase-transfer conditions in 60% yield.

(10) Typical experimental procedure for the preparation of trimethyl 10-oxatetracyclo[6.4.1.$0^{1,8}$.$0^{2,6}$]tridec-2-ene-4,4,6-tricarboxylate (**12b**): A mixture of 0.5 g (1.16 mmol) of **9b**, $Pd(OAc)_2$ (0.015 g; 0.058 mmol), PPh_3 (0.065 g, 0.232 mmol), and Ag_2CO_3 (0.638 g; 2.318 mmol) in 10 mL of MeCN was heated in a sealed tube at 130 °C for 3 h. Extractive workup and flash column chromatography (silica gel; 1:12 ether–petroleum ether) provided 0.288 g (71%) of **12b**. IR (neat): 3090, 3020, 1960, 1860, 1725, 1670, 1440, 1390, 1250, 1095, 1055, 975, 920, 840 cm^{-1}. ^1H NMR (250 MHz, $CDCl_3$): δ 5.70 (1 H, s, H-12), 4.03 (1 H, d, J = 11.2 Hz, H-6), 3.76 (3 H, s, CO_2Me-11), 3.74 (3 H, s, CO_2Me-11), 3.64 (3 H, s, CO_2Me-9), 3.60 (2 H, m, H-4,6), 3.08 (1 H, m, H-4), 3.00 (1 H, d, J = 13.5 Hz, H-10), 2.69 (1 H, d, J = 12.73 Hz, H-8), 2.46 (1 H, d, J = 13.5 Hz, H-10), 2.02 (2 H, m, H-3), 1.58 (1 H, d, J = 12.76 Hz, H-8), 1.03 (1 H, d, J = 6.01 Hz, H-13), 0.90 (1 H, d, J = 6.00 Hz, H-13). ^{13}C NMR (62.9 MHz, $CDCl_3$): δ 175.3, 171.1, 170.3, 158.4, 118.4, 70.2, 70.0, 63.6, 60.6, 52.8, 52.6, 52.1, 47.0, 39.5, 31.2, 25.8, 25.1, 21.3. Anal. Calcd for $C_{18}H_{21}O_7$: C, 61.71; H, 6.33. Found: C, 61.78; H, 6.53.

Palladium-Catalyzed Coupling of 2-Bromonaphthoquinones with Stannanes: A Concise Synthesis of Antibiotics WS 5995 A and C and Related Compounds†

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Summary: The syntheses of antibiotics WS 5995 A and C and a hypothetical intermediate in the biosynthesis of the kinamycin antibiotics have been completed by using as the key step the palladium-catalyzed coupling of 2-bromo-1,4-naphthoquinones with stannanes.

The kinamycin antibiotics (e.g. **1**, Kinamycin D), isolated from *Streptomyces murayamaensis*, are biosynthesized from benz[*a*]anthraquinones, such as dehydrorabelomycin

†Dedicated to Professor Francisco Fariña on the occasion of his 65th birthday.

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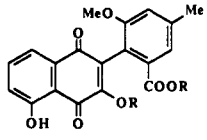
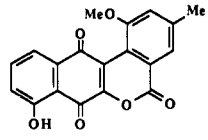
(2), via the intermediate prekinamycin (3) (Scheme I).[1,2] The cyanamide carbon has been proposed to be derived from C-5 of 2 via oxidation and nitrogen insertion to give the hypothetical intermediate benzo[*b*]phenanthridinone 4, which rearranges to give the ring-contracted benzo[*b*]-carbazole 3.[1] Very recently, a natural compound related to 4, the aglycon of phenanthroviridin, has been described.[3]

In the retrosynthetic analysis for both prekinamycin[4,5] and benzo[*b*]phenanthridinones 4 and 5 the key carbon–carbon bond between the naphthoquinone nucleus and the 2-aryl can be derived by a palladium-catalyzed Stille reaction[6] from an electrophilic 2-bromo-5-hydroxy-1,4-naphthoquinone (2-bromojuglone) synthon and a 2,4,6-trisubstituted arylstannane. The use of the unprotected quinone nucleus in the coupling reaction should minimize the protection, deprotection, and oxidation steps usually required for the synthesis of this type of compounds.[7] The desired heterocyclization was expected to take place on a C-3 substituted derivative with a suitable leaving group.

In this paper, we report the successful use of 2-bromonaphthoquinones as electrophiles in the palladium-catalyzed cross-coupling reaction with stannanes as well as the application of this method to the synthesis of antibiotics WS 5995 C (6) and A (7)[8,9] and benzo[*b*]phenanthridinones 4 and 5.



6, R = H
16, R = Me



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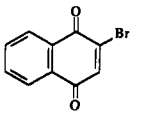
Selected results from the coupling of 2-bromonaphthoquinone (8),[10] α-bromojuglone (9),[10,11] and 2-bromo-8-hydroxy-6-methoxynaphthoquinone (10)[12] are shown in Table I.[13,14] The coupling reactions proceed smoothly with tetraalkyl,[15,16] alkenyl, and aryltrialkyl stannanes by using either $Pd(PPh_3)_4$ or $PdCl_2(dppf)$[17] catalysts. In most cases (entries 1–3, 6, 9, 10, and 12) higher yields and/or shorter reaction times were obtained by the addition of Cu(I) as cocatalyst, as has been shown in a number of recently reported palladium-catalyzed couplings.[18] In the reaction of 9 with [2,6-dimethoxy-4-methylphenyl]trimethylstannane (11)[19] (entry 7), 2-methyljuglone (plumbagin) was also obtained in low yield, even in the presence of CuBr. Related transfers of the alkyl group have also been observed in the couplings of some trialkylaryl stannanes with anthraquinone[17] and 4-quinolyl triflates.[18] However, quinone 9 coupled with other sterically hindered stannanes such as 12[20] and 13[21] (entries 9 and 10) to give 14 and 15 in good to excellent yields. On the other hand, the corresponding reaction of 9 with phenylboronic acid in the presence of $Pd(PPh_3)_4$ catalyst[22] led only to decomposition of the naphthoquinone.



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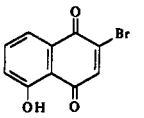

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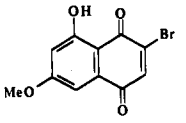

10

Coupled product 15 was transformed into antibiotic WS 5995 C (6) in two steps by epoxidation (12 equiv of *t*-BuOOH, 40% aqueous $[PhCH_2NMe_3]OH$ in THF, 23 °C, 1 h) to give a mixture of unstable epoxides, which, without

(1) Seaton, P. J.; Gould, S. J. *J. Am. Chem. Soc.* **1988**, *110*, 5912.
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(3) (a) Isolation: Fendrich, G.; Zimmermann, W.; Gruner, J.; Auden, J. A. L. Eur. Pat. Appl. EP 304,400 (Cl C07D221/18), 22 Feb 1989, CH Appl. 87/3, 196, 20 Aug 1987; *Chem. Abstr.* **1990**, *112*, 75295q. (b) Synthesis: Gore, M. P.; Gould, S. J.; Weller, D. D. *J. Org. Chem.* **1991**, *56*, 2289.
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(5) For recent syntheses of substituted benzo[*b*]carbazole-1,6-diones, see: (a) Bittner, S.; Krief, P.; Massil, T. *Synthesis* **1991**, 215. (b) Liebeskind, L. S. *Tetrahedron* **1989**, *45*, 3053 and references cited therein. (c) See also: Bergman, J.; Pelcman, B. *Pure Appl. Chem.* **1990**, *62*, 1967.
(6) Stille, J. K. *Angew. Chem., Int. Ed. Engl.* **1986**, *25*, 508.
(7) (a) For related work, see: Jung, M. E.; Jung, Y. H. *Tetrahedron Lett.* **1988**, *29*, 2517. Kubiak, G. G.; Confalone, P. N. *Tetrahedron Lett.* **1990**, *31*, 3845. Confalone, P. N. *J. Heterocycl. Chem.* **1990**, *27*, 31. (b) The coupling of stannylquinones with allylic halides also proceeds directly without protection of the quinone: Liebeskind, L. S.; Foster, B. S. *J. Am. Chem. Soc.* **1990**, *112*, 8612.
(8) Isolation from *Streptomyces auranticolor* and structure: (a) Ikushima, H.; Iguchi, E.; Kohsaka, M.; Aoki, H.; Imanaka, H. *J. Antibiot.* **1980**, *33*, 1103. (b) Ikushima, H.; Okamoto, M.; Tanaka, H.; Ohe, O.; Kohsaka, M.; Aoki, H.; Imanaka, H. *J. Antibiot.* **1980**, *33*, 1107.
(9) Synthesis: (a) Tanaka, H.; Itoh, Y.; Ikushima, H.; Okamoto, M.; Kawai, Y.; Imanaka, H. *Tetrahedron Lett.* **1980**, *21*, 4359. (b) Ikushima, H.; Takase, S.; Kawai, Y.; Itoh, Y.; Okamoto, M.; Tanaka, H.; Imanaka, H. *Agric. Biol. Chem.* **1983**, *47*, 2231. (c) Watanabe, M.; Date, M.; Furukawa, S. *Chem. Pharm. Bull.* **1989**, *37*, 292.
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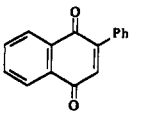
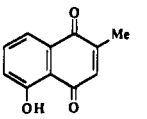
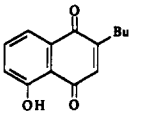
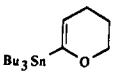
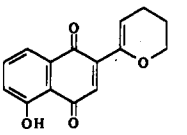
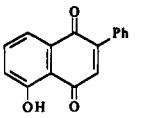
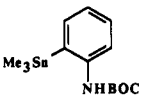
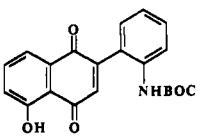
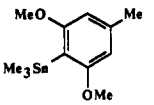
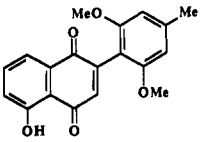
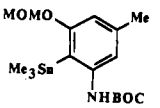
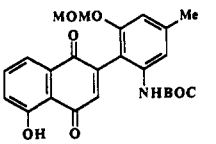
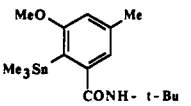
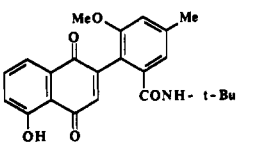
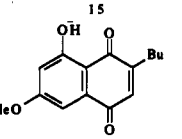
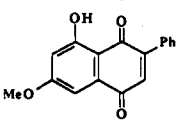
(12) Prepared from (*Z*)-1-ethoxy-3-methoxy-1-[(trimethylsilyl)oxy]-buta-1,3-diene and 2,6-dibromobenzoquinone by a procedure analogous to that of: Savard, J.; Brassard, P. *Tetrahedron* **1984**, *40*, 3455.
(13) Kvalnes, D. E. *J. Am. Chem. Soc.* **1934**, *56*, 2478.
(14) Wurm, G.; Gurka, H. J. *Arch. Pharm. (Weinheim, Ger.)* **1986**, *319*, 190.
(15) The coupling of 2,3-dichloronaphthoquinone with tetramethyl and tetrabutyl stannanes in the presence of a catalyst prepared from $PdCl_2(dppp)$ [dppp = 1,3-bis(diphenylphosphino)propane] and AlH-*i*-Bu_2 has been reported: Peet, W. G.; Tam, W. *J. Chem. Soc., Chem. Commun.* **1983**, 853.
(16) For a recent study on the reaction of 2-bromoquinones with dialkylcuprates, see: Wigal, C. T.; Grunwell, J. R.; Hershberger, J. *J. Org. Chem.* **1991**, *56*, 3759.
(17) dppf = 1,1′-bis(diphenylphosphino)ferrocene. This catalyst was shown to give superior results in the coupling of anthraquinone triflates with some stannanes: Tamayo, N.; Echavarren, A. M.; Paredes, M. C.; Fariña, F.; Noheda, P. *Tetrahedron Lett.* **1990**, *31*, 5189.
(18) (a) Gómez-Bengoa, E.; Echavarren, A. M. *J. Org. Chem.* **1991**, *56*, 3497. (b) For the use of CuI as cocatalyst, see: Liebeskind, L. S.; Fengl, R. W. *J. Org. Chem.* **1990**, *55*, 5359, and references cited therein.
(19) This stannane was prepared in 81% yield by a modification of the procedure described in: Del Valle, L.; Stille, J. K.; Hegedus, L. S. *J. Org. Chem.* **1990**, *55*, 3019.
(20) (a) Synthesized from 5-methyl-1,3-dihydroxybenzene in four steps: i. aqueous NH_4OH, NH_4Cl, 160 °C, 12 h (75%);[20b] ii. (*t*-BuOCO)$_2$O, THF, reflux, 2 h, (100%); iii. MOMCl, aqueous NaOH–CH_2Cl_2, *n*-Bu_4NBr, 23 °C, (88%); iv. *t*-BuLi, pentane–THF, −78 to −20 °C; Me_3SnCl, THF (47%). (b) Wessely, F.; Eibl, H.; Friedrich, G. *Monatsh. Chem.* **1952**, *83*, 24.
(21) (a) Synthesized in three steps from 3-methoxy-5-methylbenzoic acid:[7a,21b] i. $SOCl_2$, CH_2Cl_2, DMF (cat.), 23 °C; ii. *t*-$BuNH_2$, CH_2Cl_2, 23 °C (96%, two steps); iii. *t*-BuLi, pentane–THF, −78 to −20 °C; Me_3SnCl, THF (92%). (b) Tamura, Y.; Fukata, F.; Sasho, M.; Tsugoshi, T.; Kita, Y. *J. Org. Chem.* **1985**, *50*, 2273.
(22) (a) Miyaura, N.; Yanagi, T.; Suzuki, A. *Synth. Commun.* **1981**, *11*, 513. (b) Alo, B. I.; Kandil, A.; Patil, P. A.; Sharp, M. J.; Siddiqui, M. A.; Snieckus, V.; Josephy, P. D. *J. Org. Chem.* **1991**, *56*, 3763 and references cited therein.

Table I. Palladium-Catalyzed Coupling of 1,4-Naphthoquinones with Stannanes[a]

entry	quinone	stannane	reaction condns[b]	reaction time (h)	product	yield (%)
1	8	$PhSnBu_3$	A	12		66[13]
2[c]	9	Me_4Sn	A	15		100[14]
3	9	Bu_4Sn	A	30		98
4	9	Bu_4Sn	B	8.5		82
5	9		C	1		82
6	9	$PhSnBu_3$	A	5		100
7	9		C	20		70
8	9	 11	C	3		46[d]
9	9	 12	A	3	 14	100
10	9	 13	A	3	 15	82
11	10	Bu_4Sn	B	18		74
12	10	$PhSnBu_3$	A	15		100

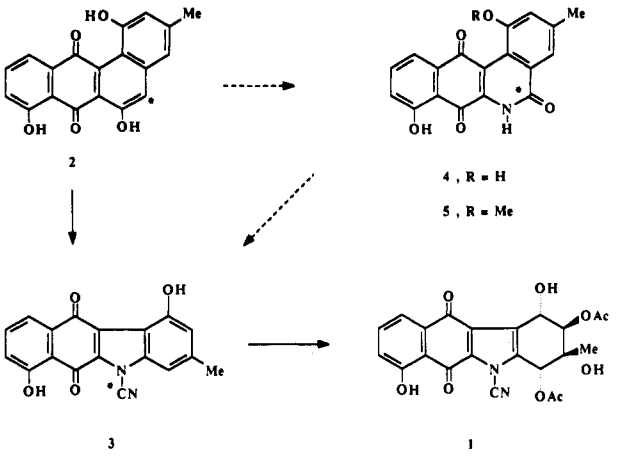
[a] Unless otherwise stated the reactions were carried out in 1,4-dioxane under reflux. [b] A = 5% each of $Pd(PPh_3)_4$ and CuBr; B = $PdCl_2$-(dppf) in DMF at 100 °C; C = 5% $Pd(PPh_3)_4$. [c] Reaction carried out in a sealed tube at 100 °C. [d] Additionally, a 6% of 2-methyljuglone was obtained.

isolation, was hydrolyzed (aqueous $HClO_4$, dioxane, 100 °C, 8 h) to **6** in 98% yield (two steps).[23] Hydrolysis of the *tert*-butyl amide is probably facilitated by the presence of the C-3 hydroxyl[9c] since **15**, lacking the C-3 substitution, was recovered unchanged after treatment under a variety of acidic conditions.[24] Treatment of **6** with excess of

(23) The synthetic compound showed identical mp, TLC mobility, and spectroscopic data (UV–vis, IR, and ^1H NMR) to those previously reported for the natural product[8b,9] (see supplementary material).

(24) Similarly fruitless was the reaction of **15** with N_2O_4 in CF_3COOH. These conditions have been successfully applied for the cleavage of *tert*-butyl amides: (a) White, E. H. *J. Am. Chem. Soc.* **1955**, *77*, 6008. (b) Kelly, T. R.; Echavarren, A.; Chandrakumar, N. S.; Köksal, Y. *Tetrahedron Lett.* **1984**, *25*, 2127. (c) Kelly, T. R.; Jagoe, C. T.; Li, Q. *J. Am. Chem. Soc.* **1989**, *111*, 4522.

Scheme I



$(CF_3CO)_2O$ at 23 °C gave antibiotic WS 5995 A (7)[23] in quantitative yield,[9] while reaction with $[Me_3O]BF_4$[25] in THF at 23 °C gave known ester 16[9b] in 76% yield.

Finally, with a concise synthesis of 16 in hand, the conversion into 4 was readily accomplished by reaction with aqueous NH_4OH in MeOH under reflux for 48 h to give 5 in 55% yield,[26] followed by demethylation with BBr_3 in CH_2Cl_2 at −78 °C (47%).[26] This described method should be also useful for the preparation of related natural products such as the gilvocarcins, which contain the reduced 2-arylnaphthalene nucleus.[27] Further work directed toward the synthesis of prekinamycin and related targets is in progress.

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Supplementary Material Available: Characterization data for all new compounds and ^1H NMR spectra for 4, its diacetate, and 5 (10 pages). Ordering information is given on any current masthead page.

(25) Methylation with diazomethane in MeOH–Et_2O as previously reported[9a,b] was difficult to reproduce and gave 16 in lower yields.

(26) The yields were determined after acetylation (Ac_2O, cat. H_2SO_4, 23 °C, 2 h) of very insoluble benzo[*b*]phenanthridinones 4 and 5. Saponification (aqueous Na_2CO_3–MeOH, 23 °C) afforded pure 4 or 5 in quantitative yield.

(27) For recent lead references on the synthesis of these family of natural products, see: (a) Kwok, D.-I.; Farr, R. N.; Daves, G. D. *J. Org. Chem.* **1991**, *56*, 3711. (b) Parker, K. A.; Coburn, C. A. *J. Org. Chem.* **1991**, *56*, 1666. (c) Hua, D. H.; Saha, S.; Maeng, J. C.; Bensoussan, D. *Synlett* **1990**, 233.

An Unusual γ-Silyl Effect in $TiCl_4$-Catalyzed Arylation of 1,4-Benzoquinones

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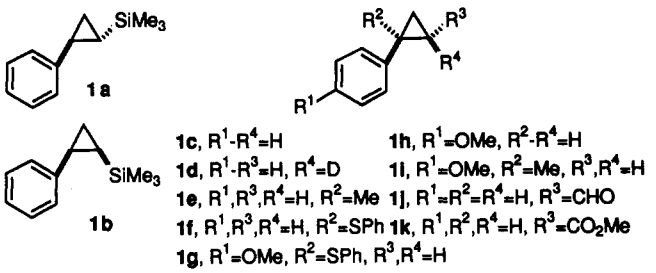
Summary: Cyclopropylbenzene (1c) reacts with 2-methoxy-1,4-benzoquinone in the presence of $TiCl_4$ to give 2-(4′-cyclopropylphenyl)-5-methoxy-1,4-benzoquinone in moderate yield (46%). Considerable improvement in yield (69%) is observed in reactions of *trans*-2-phenyl-1-(trimethylsilyl)cyclopropane (1a) with the $TiCl_4$–quinone complex.

The effects of α- and β-silicon substitution on the control and rates of formation of carbocation centers has been well-documented; α-trimethylsilyl (TMS) groups retard, relative to C, and β-TMS groups dramatically accelerate solvolytic generation of carbocations.[1] There have been fewer demonstrations of the effects of silicon substituents γ to carbocation centers; however, the effects in terms of solvolysis rates are impressive in some cases (>10^4).[1,2] To date, reports of the γ-Si effect have been limited largely to studies of the rates of solvolysis of structurally well-defined esters and sulfonates. Herein, we report one of only a few examples of the utilization of the γ-Si effect as a control element in synthesis; in this case involving a Ti(IV)-mediated arylation of 1,4-benzoquinones.[3]

We prepared a number of substituted cyclopropanes 1 and studied their Lewis acid catalyzed reactions with 2-methoxy-1,4-benzoquinone, 2. The $TiCl_4$-promoted reaction of cyclopropanes 1a and 1c with quinone 2 at −78 °C



gave 7a and 8 in 69% and 46% isolated yields, respectively (Table I and Scheme I). The products 7a/8 apparently result from electrophilic aromatic substitution on the phenyl ring of 1a/c by the Ti(IV)-bound quinone complex 3 to give 5 which then undergoes oxidation by additional Ti(IV)–quinone complex 3[4] to yield 7a/8 and 2-methoxy-

(1) (a) Lambert, J. B. *Tetrahedron* **1990**, *46*, 2677. For a listing of reviews of synthetic applications arising from these effects, see: (b) Larson, G. L. In *The Chemistry of Organic Silicon Compounds*; Patai, S.; Rappoport, Z., Eds.; John Wiley and Sons, 1989; Vol. 1, Chapter 11. (c) See also: Bassindale, A. R.; Taylor, P. G. In ref 1b, Chapter 14.

(2) (a) Shiner, V. J., Jr.; Ensinger, M. W.; Kriz, G. S. *J. Am. Chem. Soc.* **1986**, *108*, 842. (b) Davidson, E. R.; Shiner, V. J., Jr.; *J. Am. Chem. Soc.* **1986**, *108*, 3135. (c) Shiner, V. J., Jr.; Ensinger, M. W.; Rutkowske, R. D. *J. Am. Chem. Soc.* **1987**, *109*, 804. (d) Bentley, T. W.; Kirmse, W.; Llewellyn, G.; Söllenböhmer, F. *J. Org. Chem.* **1990**, *55*, 1536. (e) Kirmse, W.; Söllenböhmer, F. *J. Am. Chem. Soc.* **1989**, *111*, 4127. (f) Grob, C. A.; Gründel, M.; Sawlewicz, P. *Helv. Chim. Acta* **1988**, *71*, 1502. (g) DeLucca, G.; Paquette, L. A. *Tetrahedron Lett.* **1983**, 4931.

(3) (a) During the preparation of this manuscript, a report on the effects of γ-Si substituents in Nef reactions appeared: Hwu, J. R.; Gilbert, B. A. *J. Am. Chem. Soc.* **1991**, *113*, 5917. (b) See also Davey, A. E.; Parsons, A. F.; Taylor, R. J. K. *J. Chem. Soc., Perkin Trans. 1* **1989**, 1853.

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