[4] Crystal structure of **2**: Crystals from cyclopentane by slow concentration at room temperature over a period of some days; $C_{54}H_{69}Al_7$, orthorhombic, *P*nma; $a = 2028.45(11)$, $b = 1713.07(6)$, $c = 1490.23(6)$ pm, $V = 5178.4(4)$ Å^3, $Z = 4$, $\rho_{calcd} = 1.163$ g cm^{-3}, crystal dimensions: $0.75 \times 0.46 \times 0.34$ mm, diffractometer Stoe IPDS, $Mo_{K\alpha}$ radiation, 193 K, measurement range: $4.2 < 2\Theta < 52.0^\circ$, 168 exposures, $\Delta\varphi = 1.1^\circ$, 5221 independent reflections, 3512 reflections $F > 4\sigma(F)$, $\mu = 0.175$ mm^{-1}, programs SHELXTL PLUS REL 4.1 and SHELXL-93, 307 parameters, $R1 = 0.052$ and w$R2$ (all data) = 0.156, max/min residual electron density: $0.77/-0.37 \times 10^{30}$ e m^{-3}. Crystal structure of **3**: Crystals from toluene; $C_{54}H_{69}Al_7$, triclinic, *P*1, $a = 1082.39(7)$, $b = 1440.15(12)$, $c = 1648.2(2)$ pm, $\alpha = 80.516(11)$, $\beta = 89.883(11)$, $\gamma = 81.348(9)^\circ$, $V = 2504.6(4)$ Å^3, $Z = 2$, $d_{calc} = 1.203$ g cm^{-3}, crystal dimensions: $0.80 \times 0.39 \times 0.26$ mm, diffractometer Stoe IPDS, $Mo_{K\alpha}$ radiation, 193 K, measurement range $4.2 < 2\Theta < 51.8^\circ$, 282 exposures, $\Delta\varphi = 1.1^\circ$, 9016 independent reflections, 6515 reflections $F > 4\sigma(F)$, $\mu = 0.181$ mm^{-1}, 561 parameters, $R1 = 0.035$ and w$R2$ (all data) = 0.107; max/min residual electron density: $0.30/-0.31 \times 10^{30}$ e m^{-3}. The positions of the hydrogen atoms H1 were obtained from a difference Fourier synthesis, they were refined isotropically. Crystallographic data (excluding structure factors) for the structures reported in this paper have been deposited with the Cambridge Crystallographic Data Centre as supplementary publication nos. CCDC 127469 (**2**) and CCDC 127470 (**3**). Copies of the data can be obtained free of charge on application to CCDC, 12 Union Road, Cambridge CB2 1EZ, UK (fax: (+44) 1223-336-033; e-mail: deposit@ccdc.cam.ac.uk).

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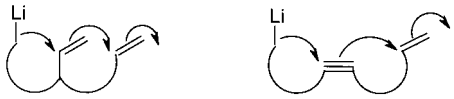
Tandem Intermolecular–Intramolecular Carbolithiation: A Versatile Method for Synthesis of Cyclopentanes**

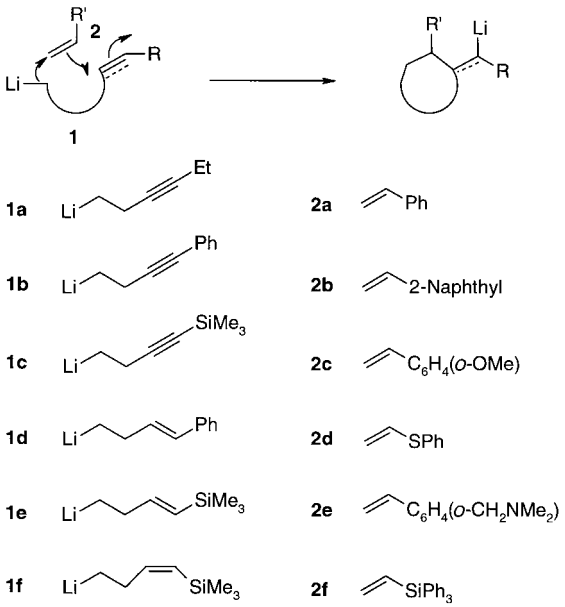
Xudong Wei and Richard J. K. Taylor*

The carbolithiation of alkenes and alkynes is a particularly useful synthetic transformation in that the generation of a new carbon–carbon bond is accompanied by the formation of an organolithium species which can be elaborated directly or after transmetalation.[1–4] To date, this chemistry has been largely confined to alkenes and alkynes activated by conjugation to carbonyl and related electron-withdrawing groups, but within this sub-group a rich diversity of examples have been described. However, in recent years, the development of improved procedures for the generation of organolithium reagents[5] has meant that major advances have been made in the carbolithiation of unactivated alkenes and alkynes.[2–4] Intramolecular variants have attracted particular attention and a range of synthetically useful 4-, 5-, and 6-*exo* carbolithiation procedures have been reported.[2f–2n] In addition, the groups of Bailey and Krief have extended this research to develop tandem intramolecular carbolithiation procedures (Scheme 1).[3]



Scheme 1. The principle of tandem intramolecular carbolithiations.

Encouraged by our research in the intermolecular carbolithiation area[4] we designed a tandem intermolecular–intramolecular carbolithiation sequence (Scheme 2). This procedure employs organolithium reagents as difunctional, conjunctive reagents and represents a versatile anionic [3+2] approach to substituted cyclopentanes.[6] This convergent



Scheme 2. Tandem intermolecular–intramolecular carbolithiation.

sequence is especially attractive as, in contrast to many tandem processes, the precursors are readily available. After the completion of our studies, we discovered that one isolated example of this procedure had been described: Krief and Barbeaux trapped a homoallylic lithium reagent with ethene, with the resulting organolithium intermediate undergoing cyclization followed by protonation to give a methylcyclopentane.[3b] Our plan was to establish a general and versatile method for the preparation of complex cyclopentanes with a high level of functionality in a stereocontrolled mannner from functionalized organolithium reagents and alkenes. Herein we report the successful realization of this plan.

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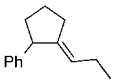
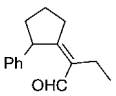
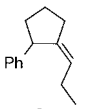
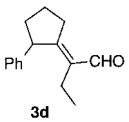
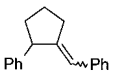
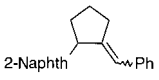
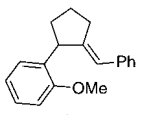
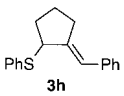
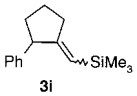
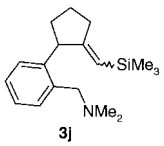
Several design features were incorporated to facilitate the desired process: 1) homoallylic and homopropargylic systems were chosen to preclude cyclization of the initial organolithium reagents **1**; 2) "activated" alkenes **2** (R = Ar, Ph_3Si, PhS) were employed to facilitate the initial intermolecular carbolithiation and ensure regioselectivity; 3) alkenes **2** were added by syringe-pump over extended times to minimize polymerization processes. The successful reactions are summarized in Table 1 and Table 2. All new compounds were fully characterized by high-field ^1H and ^{13}C NMR spectroscopy and by elemental analysis or high-resolution mass spectrometry. Reactions of the acetylenic reagents **1a–c**,[7] generated by lithium–iodine exchange using *tert*-butyllithium at $-78\,^\circ C$,[5] were studied first (Table 1). Alkenes **2a–d** were slowly (0.5–1.5 h) added to the organolithium reagents and we were delighted to observe that, after protonation, the anticipated tandem sequence to give cyclopentanes **3a, c, e–j** proceeded in reasonable to high yields (50–82%), often with excellent stereocontrol. Thus treatment of styrene with hexynyllithium **1a** (entry 1) gave cyclopentane **3a** in 61% yield (*E*:*Z* = 97:3). The preponderance of the *E*-alkene is consistent with the accepted *syn*-carbolithiation mechanism.[2h] However, when a diethyl ether/THF (3/1) solvent system was employed the corresponding *Z*-isomer **3c** was obtained as the major product (entry 3: *E*:*Z* = 2:98; structures confirmed by NOE studies). Similar results were obtained when DMF was employed as the electrophilic trapping agent (entries 2 and 4): **3b** was obtained in diethyl ether, **3d** in diethyl ether/THF. We are currently carrying out further reactions to rationalize this unprecedented result, although we assume that the effective size of the lithium substituent is increased in THF and that this leads to vinyl inversion.

Table 1. Tandem intermolecular–intramolecular carbolithiation processes with the lithiated alkynes **1a–c**.

Entry	1	2	Solvent	Electrophile	Product	Yield [%] (*E*:*Z*)
1	**1a**	**2a**	diethyl ether	MeOH	 **3a**	61 (97:3)
2	**1a**	**2a**	diethyl ether	DMF/H_3O^+	 **3b**	48 (97:3)
3	**1a**	**2a**	diethyl ether:THF (3:1)	MeOH	 **3c**	50 (2:98)
4	**1a**	**2a**	diethyl ether:THF (3:1)	DMF/H_3O^+	 **3d**	43 (2:98)
5	**1b**	**2a**	diethyl ether	MeOH	 **3e**	53 (70:30)
6	**1b**	**2b**	diethyl ether	MeOH	 **3f**	62 (56:44)
7	**1b**	**2c**	diethyl ether	MeOH	 **3g**	60 (100:0)
8	**1b**	**2d**	diethyl ether	MeOH	 **3h**	62[a] (100:0)
9	**1c**	**2a**	diethyl ether	MeOH	 **3i**	82 (76:24)
10	**1c**	**2e**	diethyl ether	MeOH	 **3j**	56 (85:15)

[a] Yield based on recovered starting material.

The remaining reactions in Tables 1 and 2 were all carried out in diethyl ether. The reactions of the lithiated alkynes **1b** and **1c** with styrene (entries 5 and 9, respectively) gave the corresponding cyclopentanes **3e** and **3i** as *E*/*Z* mixtures, presumably as a consequence of fast isomerization of phenyl- and trimethylsilyl-stabilized vinyl anions.[2h] Other alkene acceptors were also employed. Thus, 2-vinylnaphthalene (**2b**), *ortho*-methoxystyrene (**2c**), phenylthioethene (**2d**), and *ortho*-(dimethylaminomethyl)styrene (**2e**) were all converted into the corresponding cyclopentanes (entries 6–8 and 10). The stereoselective formation of **3g** and **3h** presumably reflects intramolecular coordination of the intermediate vinyllithium to oxygen and sulfur, respectively, and demonstrates that stereocontrol can be achieved even with phenyl-stabilized vinyl anions.

The reactions of the lithiated alkenes **1d–f**[7] (Table 2) to give the cyclopentanes **3k–3m** proceeded in reasonable yield, and, in reactions 2–4, excellent stereocontrol. At first sight, the low stereoselectivity observed in the reaction of styrene and **1d** (entry 1) is surprising: we ascribe this to a π-stacking effect, which provides added stabilization for *cis*-**3k**.

These reactions can also be carried out on disubstituted alkenes (Scheme 3). Thus, reaction of alkynyllithium reagent **1a** with (*E*)-1-phenylpropene (**4**) in diethyl ether/THF followed by protonation produced cyclized adduct **5** in 50% yield. The corresponding carboxylic acid **6** was obtained when the intermediate vinyllithium was trapped with carbon dioxide. In a similar manner, reaction of alkynyllithium reagent **1a** with 1,2-dihydronaphthalene (**7**) in diethyl ether/THF produced a single adduct **8** in 41% yield with the expected predominance of the *Z*-alkene. The corresponding reaction using lithiated alkene **1d** produced tricycle **9** as the only isolable product (although minor diastereomeric impurites were visible in the ^1H NMR spectrum of the crude product). The *cis*,*cis* stereochemistry of **9**, which was assigned using NOE experiments, presumably reflects the π-stacking

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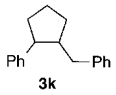
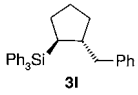
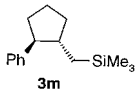
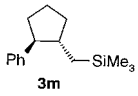
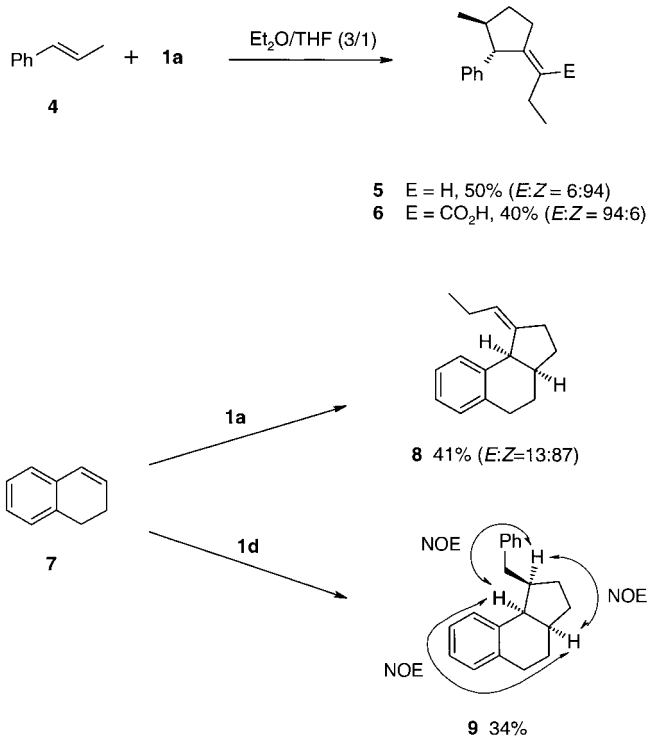
Table 2. Tandem intermolecular–intramolecular carbolithiation processes with lithiated alkenes **1d–f**.

Entry	1	2	Solvent	Electrophile	Product	Yield [%] (*cis*:*trans*)
1	**1d**	**2a**	diethyl ether	MeOH	 **3k**	52 (50:50)
2	**1d**	**2f**	diethyl ether	MeOH	 **3l**	45 (100:0)
3	**1e**	**2a**	diethyl ether	MeOH	 **3m**	52 (100:0)
4	**1f**	**2a**	diethyl ether	MeOH	 **3m**	45 (100:0)



Scheme 3. Tandem intermolecular–intramolecular carbolithiation of disubstituted alkenes.

effect observed earlier. Although optimization studies have to be carried out, these results illustrate the potential of this new methodology for the synthesis of polysubstituted cyclopentanes and related annelated systems.

In summary we have demonstrated that tandem intermolecular–intramolecular carbolithiations provide an extremely convenient and versatile route to substituted cyclopentanes from readily available starting materials. In many cases excellent stereocontrol can be achieved by solvent chelation, intramolecular coordination, or π-stacking effects. Studies to develop asymmetric variants[1h, 2l–m, 4c, 8] of this methodology and to apply it to natural product synthesis are in progress.

Experimental Section

tert-Butyllithium (1.24 mL, 1.7 M solution in hexanes, 2.1 mmol) was added dropwise to a stirred solution of the corresponding iodoalkene or -alkyne (1.0 mmol) in diethyl ether (9 mL) at −78 °C and under an inert atmosphere. The cooling bath was removed after 15 min and the solution of organolithium reagent **1** was allowed to warm up to room temperature. A solution of alkene **2** (0.50 mmol) in diethyl ether (3 mL; or THF in those cases specified in the table) was added slowly with a syringe pump over 1 h at room temperature. Upon completion, the reaction mixture was stirred for another 5 min and quenched with the specified electrophilic reagent. The reaction mixture was diluted to 70 mL with diethyl ether and was washed with water (15 mL), brine (15 mL), and the organic layer was dried over sodium sulfate. After evaporation of the solvent and column chromatograghy (silica gel) the cyclized product **3** was obtained, for example, **3g** as a white solid (60 %, *E*:*Z*, 100:0): m.p. 69.5–70.5 °C; thin-layer chromatography $R_f = 0.29$ (petrol ether 40–60 °C); ^{1}H NMR (270 MHz, $CDCl_3$, 25 °C, TMS): $\delta = 1.67–1.83$ (m, 2H), 1.90–2.03 (m, 1H), 2.03–2.19 (m, 1H), 2.70–2.90 (m, 2H), 3.83 (s, 3H), 4.17 (t, ^3J (H,H) = 8 Hz, 1H), 5.99 (m, 1H), 6.87–6.94 (m, 2H), 7.10–7.30 (m, 7H); ^{13}C NMR (67.9 MHz, $CDCl_3$): $\delta = 25.7$, 32.3, 34.2, 46.6, 55.5, 110.7, 120.6, 122.4, 125.6, 127.1, 128.06, 128.10, 129.1, 133.7, 138.9, 150.0, 157.5; IR (film): $\tilde{\nu}_{max} = 2954$, 2866, 1597, 1585, 1491, 1241, 1029, 753 cm^{-1}; MS (EI) *m*/*z*: 264, [M^+], 173, 91; HR-MS (EI): *m*/*z*: 264.1518 (calcd for $C_{19}H_{20}O$: 264.1514, error -1.5×10^{-6}); elemental analysis calcd for $C_{19}H_{20}O$: C 86.32, H 7.63; found: C 85.96, H 7.78.

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$Ca_{11}N_6(CN_2)_2$ and $Ca_4N_2(CN_2)$: The True Nature of an "Unusual Binary Nitride" Is Finally Revealed**

Olaf Reckeweg and Francis J. DiSalvo*

The compound reported in the literature as the "electron deficient, binary nitride $Ca_{11}N_8$" has been an object of discussion for many years.[1] Both its electronic and crystal structure are not yet fully understood. The only reported structure determination of "$Ca_{11}N_8$" is based solely on X-ray crystallographic methods.[1] Because of their similar X-ray scattering factors, boron, carbon, nitrogen, and oxygen are often hard to distinguish with X-ray methods. It has been suggested[2] an oxygen contamination during the reaction led to the ionic compound $(Ca^{2+})_{11}(N^{3-})_6(O^{2-})_2$ and the net electrical neutrality simplifies the electronic situation. However, this model requires an unusual coordination sphere for one of the nitrogen atoms. The average Ca–N interatomic distance in "$Ca_{11}N_8$" is 248 pm for N1 (numbering after ref. [1]) and 242 pm for N2 (both in a distorted octahedral environment), but 263 pm (two 250 pm bonds and one 290 pm bond) for N3 (247 pm with coordination number six in Ca_3N_2[3]). For this unusual nitrogen (or oxygen) coordination sphere, or for the even more exceptional electron-deficient binary nitride, no satisfying explanation had been found yet.

While we were pursuing research in the ternary system Ca-Cu-N, a reaction of Ca_3N_2, Cu_3N, and NaN_3 in a stainless steel ampoule yielded metallic copper and two different kinds of crystals. Both crystals appeared as very fine needles, one being a yellow-green color and transparent, the other being dark red, nearly black, but also transparent if intensely illuminated. Both crystals are air and moisture sensitive.

Single crystal X-ray structure analysis showed surprising results.[4] The crystal structure of the dark-red crystals has a striking resemblance to the one reported for "$Ca_{11}N_8$". The crystallographic data ($P4_2nm$ (No. 102), $a = 1445(2)$ pm, and $c = 360(1)$ pm for "$Ca_{11}N_8$", compared to $P4_2/mnm$ (No. 136), $a = 1452.3(2)$ pm, and $c = 360.83(4)$ pm for the dark-red crystals) and the coordinates of almost all atoms of "$Ca_{11}N_8$" and our compound are closely related to each other (see Figure 1). The only difference (except the missing mirror plane $\perp c$ in "$Ca_{11}N_8$") is, instead of the single (and already under doubt) nitrogen atom on the N3 position of "$Ca_{11}N_8$", our data showed a triatomic unit with three crystallographically independent sites. Refinements with nitrogen atoms on all three sites showed a very large atomic displacement factor for the middle nitrogen atom. Refining a carbon atom for this position instead of a nitrogen atom decreased both R values

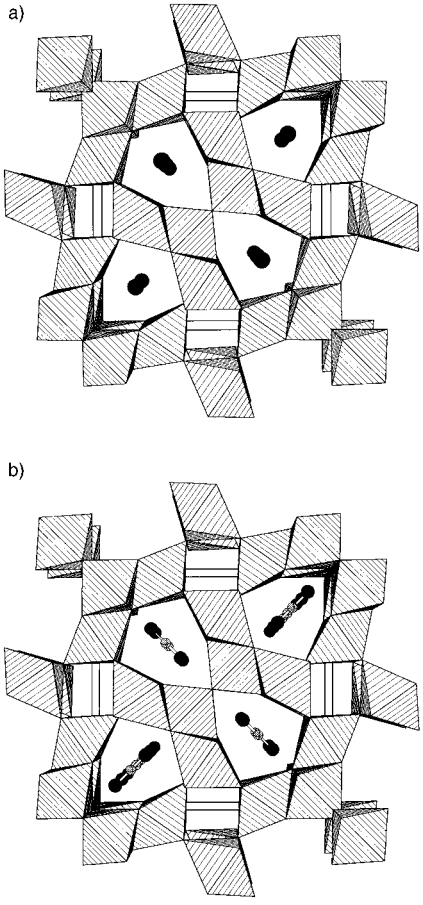

Figure 1. Comparison of the structures of a) "$Ca_{11}N_8$" and b) **1**. The views parallel to the c axes are shown. The distorted Ca_6N octahedra are displayed as hatched polyhedra, nitrogen atoms are displayed as black circles, carbon atoms as gray spheres.

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