

Synthesis, Structure, and Reactivity of $\{\eta^3\text{-HB(3-Bu}^t\text{pz)}_3\}\text{BeCH}_3$, a Terminal Beryllium Alkyl Complex Supported by Tris(3-*tert*-butylpyrazolyl)hydroborato Ligation

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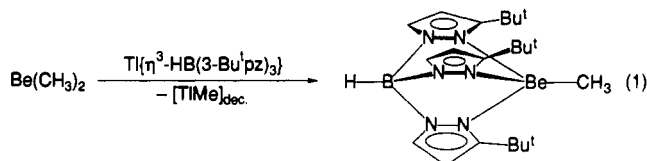
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Introduction

We recently described the use of the sterically-demanding tris(3-*tert*-butylpyrazolyl)hydroborato ligand, $[\text{HB(3-Bu}^t\text{pz)}_3]^-$ ($3\text{-Bu}^t\text{pz} = 3\text{-C}_3\text{N}_2\text{Bu}^t\text{H}_2$)¹ to provide a well-defined coordination environment for stabilizing monomeric alkyl and hydride derivatives of the s- and p-block metals.^{2–5} One such example is $\{\eta^3\text{-HB(3-Bu}^t\text{pz)}_3\}\text{BeH}$, the first terminal beryllium hydride complex to be structurally characterized by X-ray diffraction.⁶ In this paper we report the synthesis, structure, and reactivity of the related beryllium methyl derivative, $\{\eta^3\text{-HB(3-Bu}^t\text{pz)}_3\}\text{BeCH}_3$.

Results

The [tris(pyrazolyl)hydroborato]beryllium methyl complex, $\{\eta^3\text{-HB(3-Bu}^t\text{pz)}_3\}\text{BeCH}_3$, is readily prepared by the reaction of $\text{Ti}\{\eta^3\text{-HB(3-Bu}^t\text{pz)}_3\}$ with $\text{Be(CH}_3)_2$ (eq 1). The molecular



structure of $\{\eta^3\text{-HB(3-Bu}^t\text{pz)}_3\}\text{BeCH}_3$ has been determined by X-ray diffraction, as shown in Figure 1. Selected bond lengths and angles are presented in Tables I and II, and atomic coordinates, in Table III. $\{\eta^3\text{-HB(3-Bu}^t\text{pz)}_3\}\text{BeCH}_3$ reacts (i) with H_2S to give $\{\eta^3\text{-HB(3-Bu}^t\text{pz)}_3\}\text{BeSH}$ and CH_4 and (ii) with I_2 to give $\{\eta^3\text{-HB(3-Bu}^t\text{pz)}_3\}\text{BeI}$ and CH_3I , as illustrated in Scheme I. However, $\{\eta^3\text{-HB(3-Bu}^t\text{pz)}_3\}\text{BeCH}_3$ does not react with CH_3I at 100 °C.

Discussion

Our recent studies have described the synthesis of a series of tris(3-*tert*-butylpyrazolyl)hydroborato half-sandwich beryllium complexes $\{\eta^3\text{-HB(3-Bu}^t\text{pz)}_3\}\text{BeX}$ ($\text{X} = \text{H, Cl, Br, I, SH}$),⁶ which complements the cyclopentadienyl and pentamethylcyclopentadienyl series (C_5H_5) BeX ($\text{X} = \text{F, Cl, Br, I, H, R, BH}_4$)⁷ and (C_5Me_5) BeX ($\text{X} = \text{H,}^8 \text{Cl,}^9 \text{PBu}_2^{10}$). However, although the hydride derivative $\{\eta^3\text{-HB(3-Bu}^t\text{pz)}_3\}\text{BeH}$ was readily obtained by the reaction of $\{\eta^3\text{-HB(3-Bu}^t\text{pz)}_3\}\text{BeX}$ ($\text{X} = \text{Cl, Br}$) with LiAlH_4 , we were unable to synthesize the corresponding methyl

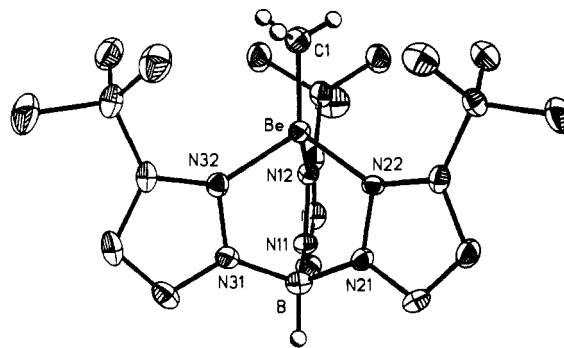


Figure 1. Molecular structure of $\{\eta^3\text{-HB(3-Bu}^t\text{pz)}_3\}\text{BeCH}_3$.

Table I. Selected Bond Lengths (Å)

Be–C(1)	1.708(6)	Be–N(12)	1.860(5)
Be–N(22)	1.855(6)	Be–N(32)	1.882(4)
N(11)–N(12)	1.382(6)	N(11)–B	1.526(7)
N(21)–N(22)	1.369(5)	N(21)–B	1.524(7)
N(31)–N(32)	1.360(6)	N(31)–B	1.529(7)

Table II. Selected Bond Angles (deg)

C(1)–Be–N(12)	120.1(4)	C(1)–Be–N(22)	120.3(3)
C(1)–Be–N(32)	118.8(2)	N(12)–Be–N(22)	96.8(2)
N(12)–Be–N(32)	98.2(2)	N(22)–Be–N(32)	97.7(3)
N(12)–N(11)–B	119.0(4)	Be–N(12)–N(11)	111.7(3)
N(22)–N(21)–B	120.4(4)	Be–N(22)–N(21)	110.9(3)
N(32)–N(31)–B	119.7(3)	Be–N(32)–N(31)	111.4(3)
N(11)–B–N(21)	108.1(4)	N(11)–B–N(31)	106.6(4)
N(21)–B–N(31)	107.8(4)		

Table III. Atom Coordinates ($\times 10^4$) and Temperature Factors ($\text{Å}^2 \times 10^3$)

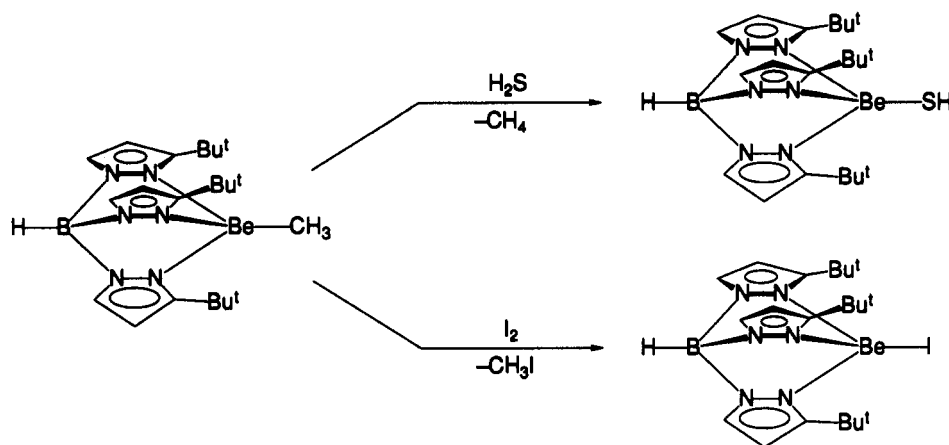
atom	x	y	z	U^a
Be	1960	2386(4)	7510	41(2)
C(1)	62(6)	2130(3)	7742(5)	61(2)
N(11)	4633(5)	1912(2)	6561(4)	47(1)
N(12)	3101(5)	1608(2)	6596(4)	41(1)
N(21)	3909(5)	3473(2)	6472(4)	43(1)
N(22)	2303(4)	3367(2)	6480(4)	38(1)
N(31)	5021(4)	2764(2)	8754(4)	46(1)
N(32)	3572(5)	2602(2)	9118(4)	44(1)
C(11)	5429(6)	1324(3)	5951(5)	55(2)
C(12)	4451(7)	629(3)	5555(5)	62(2)
C(13)	2987(6)	815(3)	5949(4)	48(2)
C(14)	1512(7)	244(3)	5693(5)	60(2)
C(15)	1177(8)	–71(4)	7105(6)	89(3)
C(16)	1813(9)	–557(4)	4810(8)	111(3)
C(17)	88(7)	733(4)	4864(6)	81(2)
C(21)	4142(6)	4187(3)	5762(5)	54(2)
C(22)	2680(6)	4571(3)	5275(5)	55(2)
C(23)	1531(6)	4049(3)	5720(4)	45(2)
C(24)	–263(6)	4188(3)	5474(5)	50(2)
C(25)	–1167(7)	3401(4)	4740(6)	71(2)
C(26)	–766(7)	4385(4)	6899(6)	73(2)
C(27)	–714(7)	4964(4)	4481(6)	83(2)
C(31)	6148(6)	2860(3)	9920(5)	58(2)
C(32)	5481(7)	2763(3)	11064(5)	63(2)
C(33)	3836(7)	2611(3)	10555(5)	50(2)
C(34)	2520(7)	2495(3)	11417(5)	62(2)
C(35)	3274(9)	2589(5)	12993(6)	104(3)
C(36)	1269(8)	3199(4)	11046(6)	86(3)
C(37)	1821(8)	1583(4)	11208(6)	82(3)
B	5138(7)	2806(4)	7190(6)	52(2)

^a Equivalent isotropic U defined as one-third of the trace of the orthogonalized U_{ij} tensor.

derivative $\{\eta^3\text{-HB(3-Bu}^t\text{pz)}_3\}\text{BeCH}_3$ by the related reaction with CH_3Li . Nevertheless, we have now found that the methyl derivative $\{\eta^3\text{-HB(3-Bu}^t\text{pz)}_3\}\text{BeCH}_3$ can be synthesized cleanly by the direct reaction of $\text{Be(CH}_3)_2$ with the thallium reagent $\text{Ti}\{\eta^3\text{-HB(3-Bu}^t\text{pz)}_3\}$ (eq 1). The reaction is similar to those used previously to prepare magnesium and zinc alkyl derivatives $\{\eta^3\text{-$

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Scheme I



$\text{HB}(3\text{-Bu}^1\text{pz})_3\text{MR}$, in which the deposition of I due to decomposition of unstable $[\text{TR}]$ provides an effective driving force for the reaction.^{2-5,11}

The molecular structure of $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeCH}_3$ has been determined by X-ray diffraction (Figure 1), confirming both the monomeric nature of the complex and the η^3 -coordination mode of the tris(pyrazolyl)hydroborato ligand. In this regard it is interesting to note that the beryllium derivatives $\{\eta^2\text{-B}(\text{pz})_4\}_2\text{Be}$ and $[\{\eta^2\text{-HB}(\text{pz})_3\}\text{Be}(\mu\text{-OH})]_3$ were recently reported, but in these complexes the poly(pyrazolyl)borate ligands adopt only an η^2 -coordination mode.¹² The trigonally distorted tetrahedral coordination environment about beryllium in $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeCH}_3$ is similar to those in the derivatives $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeH}$ and $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeBr}$. However, the average Be-N bond length [1.86(2) Å] in $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeCH}_3$ is longer than those in both $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeH}$ [1.778(4) Å] and $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeBr}$ [1.80(2) Å] and is presumably a reflection of increased steric interactions between the Be -methyl group and the *tert*-butyl substituents of the tris(3-*tert*-butylpyrazolyl)hydroborato ligand. Indeed, the van der Waals radius of CH_3 (2.0 Å) is greater than those of Br (1.95 Å) and H (1.2 Å).¹³

The Be-CH_3 bond length in $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeCH}_3$ [1.708(6) Å] is similar to the sum of the covalent radii of Be and C (1.66 Å)¹³ and also compares well with that of the cyclopentadienyl analogue $(\text{C}_5\text{H}_5)\text{BeCH}_3$ [1.706(3) Å] measured in the gas phase by electron diffraction.¹⁴ However, the Be-C bond length in $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeCH}_3$ is substantially shorter than those in (i) "ate" derivatives such as $\text{Li}_2\text{Be}(\text{CH}_3)_4$ [1.84(10) Å]¹⁵ and (ii) bridging methyl derivatives such as solid $[(\text{CH}_3)_2\text{Be}]_n$ [1.93(2) Å].¹⁶ For reference, a selection of Be-C bond lengths is presented in Table IV, indicating a relatively large range of Be-CH_3 bond lengths from 1.698(5) Å, in gaseous $(\text{CH}_3)_2\text{Be}$, to 1.93(2) Å, in solid $[(\text{CH}_3)_2\text{Be}]_n$.

The beryllium methyl group in $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeCH}_3$ has also been characterized by ^1H , ^{13}C , and ^9Be NMR spectroscopies, as summarized in Table V. Notably, the ^1H NMR resonance of the beryllium methyl group is observed as a relatively sharp singlet at δ 0.57 ppm (Figure 2), whereas the ^{13}C NMR resonance at δ 4.7 ppm is broadened considerably as a result of quadrupolar relaxation by the beryllium nucleus (Figure 3). The ^9Be NMR resonance of $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeCH}_3$ is observed at δ 5.2 ppm (relative to $[\text{Be}(\text{H}_2\text{O})_4]^{2+}$), close to the range δ 1–4 ppm established previously for the four-coordinate beryllium complexes $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeX}$ ($\text{X} = \text{H}, \text{Cl}, \text{Br}, \text{I}, \text{SH}$)⁶ but substantially

Table IV. Be-C Bond Lengths

compound	method ^a	$d(\text{Be-C})$, Å	ref
$(\text{CH}_3)_2\text{Be}$	ED	1.698(5)	<i>d</i>
Bu^1_2Be	ED	1.699(3)	<i>e</i>
$(\text{C}_5\text{H}_5)\text{BeCH}_3$	ED	1.706(3) ^b	<i>f</i>
$\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeCH}_3$	XD	1.708(6)	this work
$[(\text{CH}_3)\text{Be}(\mu\text{-OSiMe}_3)]_4$	XD	1.71(2)	<i>g</i>
$[\text{CH}_3\text{Be}(\mu\text{-CCMe})(\text{NMe}_3)]_2$	XD	1.748(7) ^c	<i>h</i>
$[\text{Na}(\text{OEt}_2)]\{(\text{CH}_2\text{CH}_3)_2\text{Be}(\mu\text{-H})\}_2$	XD	1.766(8)	<i>i</i>
		1.810(8)	
$(\text{CH}_3)_2\text{Be}(\text{quinuclidine})_2$	XD	1.81	<i>j</i>
		1.85	
$\text{Li}_2\text{Be}(\text{CH}_3)_4$	XD	1.84(10)	<i>k</i>
LiBeBu^1_3	XD	1.812(4)	<i>l</i>
		1.854(4)	
		1.864(3)	
$[(\text{CH}_3)_2\text{Be}]_n$	XD	1.93(2)	<i>m</i>

^a ED = electron diffraction; XD = X-ray diffraction. ^b The $\text{Be-cyclopentadienyl}$ bond lengths are 1.923(3) Å. ^c The $\text{Be}(\mu\text{-CCMe})_2\text{B}$ bond lengths are 1.852(6) and 1.890(6) Å. ^d Almenningen, A.; Haaland, A.; Morgan, G. L. *Acta Chem. Scand.* **1969**, *23*, 2921–2922. ^e Almenningen, A.; Haaland, A.; Nilsson, J. E. *Acta Chem. Scand.* **1968**, *22*, 972–976. ^f Drew, D. A.; Haaland, A. *Acta Chem. Scand.* **1972**, *26*, 3079–3084. ^g Mootz, D.; Zinnius, A.; Böttcher, B. *Angew. Chem., Int. Ed. Engl.* **1969**, *8*, 378–379. ^h Morosin, B.; Howatson, J. J. *Organomet. Chem.* **1971**, *29*, 7–14. ⁱ Adamson, G. W.; Bell, N. A.; Shearer, H. M. M. *Acta Crystallogr.* **1981**, *B37*, 68–71. ^j Whitt, C. D.; Atwood, J. L. *J. Organomet. Chem.* **1971**, *32*, 17–25. ^k Weiss, E.; Wolfrum, R. *J. Organomet. Chem.* **1968**, *12*, 257–262. ^l Wermer, J. R.; Gaines, D. F.; Harris, H. A. *Organometallics* **1988**, *7*, 2421–2422. ^m Snow, A. I.; Rundle, R. E. *Acta Crystallogr.* **1951**, *4*, 348–352.

Table V. ^1H , ^{13}C , and ^9Be NMR Parameters for the Be-CH_3 Group in $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeCH}_3$

nucleus	δ	nucleus	δ	nucleus	δ
^1H	0.56	^9Be	5.2	^{13}C	4.7

different from that for the cyclopentadienyl derivative $(\text{C}_5\text{H}_5)\text{BeCH}_3$ (δ –20.4 ppm).¹⁷

The reactivity of the methyl complex $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeCH}_3$ toward H_2S and I_2 (Scheme I) is analogous to that of the hydride derivative $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeH}$.⁶ Thus, the Be-C bond is cleaved by H_2S to give the thiol derivative $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeSH}$ and CH_4 , while I_2 cleaves the Be-CH_3 bond giving $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeI}$ and CHI_3 . However, whereas the hydride derivative $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeH}$ reacts with CH_3I at room temperature, to give $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeI}$, the methyl derivative $\{\eta^3\text{-HB}(3\text{-Bu}^1\text{pz})_3\}\text{BeCH}_3$ does not react with CH_3I at 100°C and is stable under these conditions.

Experimental Section

General Considerations. All manipulations were performed using a combination of glovebox, high-vacuum, and Schlenk techniques.¹⁸

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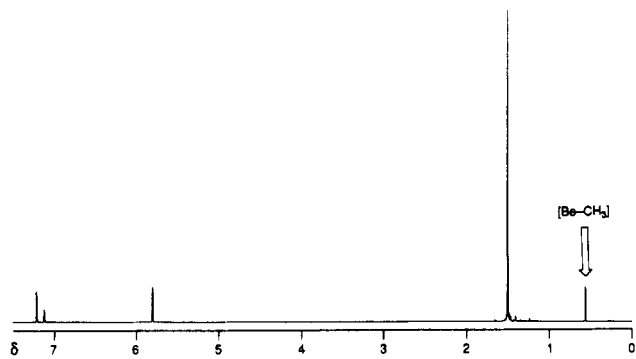


Figure 2. ^1H NMR spectrum of $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeCH}_3$ in C_6D_6 (δ 7.15).

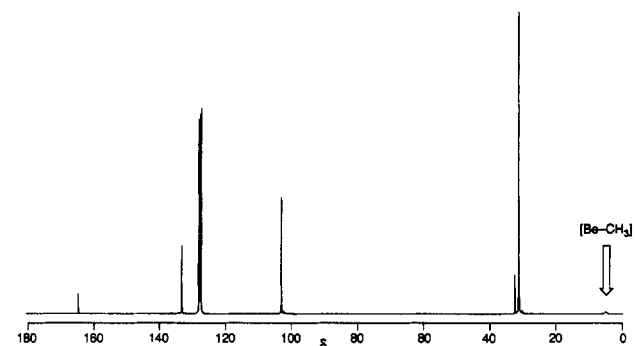


Figure 3. $^{13}\text{C}\{^1\text{H}\}$ NMR spectrum of $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeCH}_3$ in C_6D_6 (δ 128).

Solvents were purified and degassed by standard procedures. ^1H and ^{13}C NMR spectra were obtained on Varian VXR 200, 300, and 400 spectrometers, and ^9Be NMR spectra, on a Varian VXR 300 spectrometer at 42.16 MHz. All J values are reported in hertz. IR spectra were recorded as Nujol mulls between KBr disks on a Perkin-Elmer 1420 spectrophotometer and are reported in cm^{-1} . Mass spectra were obtained on a Nermag R10-10 mass spectrometer using chemical ionization (NH_3 or CH_4) techniques. Elemental analyses were obtained by using a Perkin-Elmer 2400 CHN elemental analyzer. $\text{Be}(\text{CH}_3)_2$ ¹⁹ and $\text{Tl}\{\eta^3\text{-HB(3-Bu'pz)}_3\}$ ¹ were prepared by the literature methods. **Caution:** Beryllium and thallium complexes are extremely toxic and should be handled with due care.

Synthesis of $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeCH}_3$. $\text{Be}(\text{CH}_3)_2$ (40 mg, 1.0 mmol) was added to a solution of $\text{Tl}\{\eta^3\text{-HB(3-Bu'pz)}_3\}$ (580 mg, 1.0 mmol) in benzene (ca. 40 mL), resulting in the immediate formation of a black deposit of Tl metal. The mixture was filtered after 20 min at room temperature and the solvent removed under reduced pressure, giving $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeCH}_3$ as a white solid (350 mg, 85%). Anal. Calcd for $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeCH}_3$: C, 65.2; H, 9.2; N, 20.7. Found: C, 64.6; H, 9.3; N, 20.1. IR data: 2524 ($\nu_{\text{B-H}}$). MS: m/z 406 ($\text{M}^+ + 1$). ^1H NMR (C_6D_6): δ 1.51 [27H, s, $\{\eta^3\text{-HB}\{\text{C}_3\text{N}_2\text{H}_2\text{C}(\text{CH}_3)_3\}_3\}$], 5.82 [3H, d, $^3J_{\text{H-H}} = 2.2$, $\eta^3\text{-HB}\{\text{C}_3\text{N}_2\text{H}_2\text{C}(\text{CH}_3)_3\}$], 7.24 [3H, d, $^3J_{\text{H-H}} = 2.2$, $\eta^3\text{-HB}\{\text{C}_3\text{N}_2\text{H}_2\text{C}(\text{CH}_3)_3\}$], 0.57 [3H, s, BeCH_3]. ^{13}C NMR (C_6D_6): δ 31.2 [q, $^1J_{\text{C-H}} = 126$, spt (partial res), $^3J_{\text{C-H}} = 4$, $\eta^3\text{-HB}\{\text{C}_3\text{N}_2\text{H}_2\text{C}(\text{CH}_3)_3\}$], 32.4 [dct (partial res), $^2J_{\text{C-H}} = 4$, $\eta^3\text{-HB}\{\text{C}_3\text{N}_2\text{H}_2\text{C}(\text{CH}_3)_3\}$], 103.3 [d, $^1J_{\text{C-H}} = 176$, d, $^2J_{\text{C-H}} = 9$, $\eta^3\text{-HB}\{\text{C}_3\text{N}_2\text{H}_2\text{C}(\text{CH}_3)_3\}$], 133.6 [d, $^1J_{\text{C-H}} = 185$, d, $^2J_{\text{C-H}} = 7$, $\eta^3\text{-HB}\{\text{C}_3\text{N}_2\text{H}_2\text{C}(\text{CH}_3)_3\}$], 165.1 [s, $\eta^3\text{-HB}\{\text{C}_3\text{N}_2\text{H}_2\text{C}(\text{CH}_3)_3\}$], 4.7 [q, $^1J_{\text{C-H}} = 112$, BeCH_3]. ^9Be NMR (C_6D_6 ; relative to $[\text{Be}(\text{H}_2\text{O})_4]^{2+}$ in aqueous $\text{Be}(\text{NO}_3)_2$): δ 5.2 ($W_{1/2} = 64$ Hz).

Reaction of $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeCH}_3$ with H_2S . A solution of $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeCH}_3$ (20 mg, 0.05 mmol) in benzene- d_6 (0.7 mL) in a 5-mm NMR tube was treated with H_2S (1 atm), and the mixture was heated at 70°C for 3 days. The reaction was monitored by ^1H NMR spectroscopy, which demonstrated the complete formation of $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeSH}^6$ and CH_4 .

Reaction of $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeCH}_3$ with I_2 . A solution of $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeCH}_3$ (20 mg, 0.05 mmol) in benzene- d_6 (0.7 mL) in a 5-mm NMR tube was treated with I_2 (15 mg, 0.06 mmol) at room temperature.

Table VI. Crystal and Intensity Collection Data

formula	$\text{C}_{22}\text{H}_{37}\text{N}_6\text{Be}$	Z	2
fw	405.4	radiation (λ , Å)	Mo K α (0.710 73)
lattice	monoclinic	space group	Pn (No. 7)
a , Å	8.453(4)	ρ (calcd), g cm^{-3}	1.10
b , Å	15.335(5)	μ (Mo K α), cm^{-1}	0.7
c , Å	9.626(3)	goodness of fit	1.112
β , deg	101.21(3)	R	0.0518
V , Å ³	1224(1)	R_w^a	0.0505

^a Weighting scheme: $w = [\sigma^2(F) + gF^2]^{-1}$.

The reaction was monitored by ^1H NMR spectroscopy, which demonstrated the immediate formation of $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeI}^6$ and CH_3I .

Attempted Reaction of $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeCH}_3$ with CH_3I . A solution of $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeCH}_3$ (20 mg, 0.05 mmol) in benzene- d_6 (0.7 mL) in a 5-mm NMR tube was treated with CH_3I (4 μL , 0.06 mmol), and the mixture was heated at 100°C for 2 days. The sample was monitored by ^1H NMR spectroscopy, which demonstrated that no reaction had occurred.

X-ray Structure Determination of $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeCH}_3$. Crystal data and data collection and refinement parameters are summarized in Table VI. A single crystal of $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeCH}_3$ grown from benzene was mounted in a glass capillary and placed on a Nicolet R3m diffractometer. The unit cell was determined by the automatic indexing of 25 centered reflections and confirmed by examination of the axial photographs. Intensity data were collected using graphite-monochromated Mo K α X-radiation ($\lambda = 0.710 73$ Å). Check reflections were measured every 100 reflections, and the data were scaled accordingly and corrected for Lorentz and polarization effects. The structure was solved using preliminary coordinates for the $[\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{Be}]$ moiety derived for the isomorphous $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeBr}$ derivative, followed by standard difference map techniques on a Data General NOVA 4 computer using SHELXTL.²⁰ Systematic absences were consistent with the space groups Pn and $P2_1/n$, but consideration of the E value statistics and the molecular symmetry suggested the choice Pn (No. 7). All non-hydrogen atoms were refined anisotropically. Most of the hydrogen atoms were located in the difference map after all the non-hydrogen atoms were located. However, only the hydrogen atoms of the Be-CH_3 and B-H groups were freely refined isotropically, while other hydrogen atoms were refined in calculated positions ($d_{\text{C-H}} = 0.96$ Å; $U_{\text{iso}}(\text{H}) = 1.2U_{\text{iso}}(\text{C})$). Block-diagonal least-squares refinement converged to $R = 0.0518$ ($R_w = 0.0505$). Inversion of configuration indicated the correct choice of enantiomorph. Atomic coordinates and thermal parameters for non-hydrogen atoms are listed in Table III, and selected bond distances and angles are listed in Tables I and II.

Conclusion

In summary, the terminal beryllium methyl complex $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeCH}_3$ has been synthesized by the reaction of $\text{Tl}\{\eta^3\text{-HB(3-Bu'pz)}_3\}$ with $\text{Be}(\text{CH}_3)_2$. The molecular structure of $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeCH}_3$ has been determined by X-ray diffraction, from which a value of 1.708(6) Å was obtained for the Be-CH_3 bond length. The complex $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeCH}_3$ is also characterized by a ^9Be NMR resonance at δ 5.2 ppm (relative to $[\text{Be}(\text{H}_2\text{O})_4]^{2+}$). The reactions of $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeCH}_3$ with H_2S and I_2 , giving $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeSH}$ and $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeI}$, respectively, are similar to those of the hydride derivative $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeH}$, whereas the absence of reaction with CH_3I indicates that the hydride derivative is substantially more reactive.

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Supplementary Material Available: Tables SI–SVI, listing crystal and intensity collection data, atomic coordinates, bond distances and angles, and anisotropic displacement parameters for $\{\eta^3\text{-HB(3-Bu'pz)}_3\}\text{BeCH}_3$ (7 pages). Ordering information is given on any current masthead page.

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