Hydrolysis and Methanolysis of the Small *closo*-Carbaboranes 1,5-Dicarbapentaborane, 1,6-Dicarbahexaborane, and 2,4-Dicarbaheptaborane. The Preparation of 1,3,5-Triborapentane Derivatives, $(RO)_2BCH_2B(OR)CH_2B(OR)_2$ (R = Me or H)

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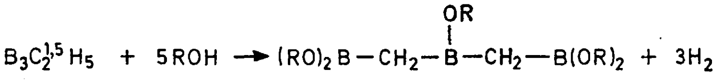
The order of reactivity toward methanol and water for three small *closo*-carbaboranes is $B_3C_2^{1,5}H_5 > B_5C_2^{2,4}H_7 > B_4C_2^{1,6}H_6$. The smallest carbaborane reacts rapidly with both reagents to form initially the triborapentane, $(RO)_2BCH_2B(OR)CH_2B(OR)_2$ (R = Me or H). The other two carbaboranes give smaller cleavage products, $B(OR)_3$, $BMe(OR)_2$, and $(RO)_2BCH_2B(OR)_2$. The mass spectra of $(MeO)_2BCH_2B(OMe)CH_2B(OMe)_2$ and $(MeO)_2BCH_2B(OMe)_2$ are discussed.

THE common polyboranes (*e.g.* B_2H_6, B_4H_{10}, B_5H_9, and B_5H_{11}) generally react under mild conditions with methanol or water (the latter sometimes requiring a solvent common with the borane) to give hydrogen and trimethoxyborane, $B(OMe)_3$, or boric acid, $B(OH)_3$, respectively.[1] While *closo*-carbaboranes are thought to be much more stable toward these reagents,[2] very little definitive work has been carried out on the small carbaboranes. The present study was carried out to rectify this and also to investigate the possibility of preparing new organopolyboranes by hydrolysis or methanolysis of carbaboranes.

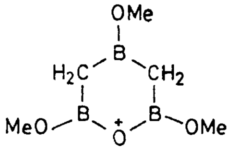


$$B_3C_2^{1,5}H_5 + 5ROH \rightarrow (RO)_2B-CH_2-\overset{OR}{\overset{|}{B}}-CH_2-B(OR)_2 + 3H_2 \qquad (1)$$

(I) R=H or Me

RESULTS AND DISCUSSION

The reactivity of the small carbaboranes towards both methanol and water follows the order $B_3C_2^{1,5}H_5 > B_5C_2^{2,4}H_7 > B_4C_2^{1,6}H_6$, the smallest carbaborane reacting vigorously under ambient conditions whereas octahedral $B_4C_2^{1,6}H_6$ requires temperatures close to 100 °C for partial reaction. For $B_3C_2^{1,5}H_5$ the reaction gave nearly quantitative methanol consumption and hydrogen evolution for the stoicheiometry implied by equation (1) (R = Me). The structure of the boron-containing product in the case of R = Me was confirmed by (*a*) the presence of two mid-field methoxy- and one upfield CH_2 resonance in the ^1H n.m.r. spectrum with area ratios of 12 : 3 : 4, and (*b*) two ^{11}B n.m.r. resonances, one at δ −52 p.p.m. (area 1) in the region expected for two carbons and one oxygen surrounding a three-co-ordinate boron,[3] and the other at δ −32 p.p.m. (area 2) in the chemical-shift region of a three-co-ordinate boron atom surrounded by one carbon and two oxygens. Also consistent with the formulation is a mass-spectrum cut off at *m/e* 170 corresponding to $[P - (OMe)_2]^+$ which may well be the cyclic radical ion (II). Further evidence for the nature of the ester (I) comes from a study of the methanolysis using MeOD. Almost pure HD was liberated in place of H_2, and the ^1H n.m.r. spectrum now showed singlets with area ratios 12 : 3 : 2. The ^{11}B n.m.r. spectrum was not significantly changed.



(II)

The addition of a trace of methanol to (I; R = Me) caused the two MeO resonances to collapse to a singlet implying rapid exchange of MeO groups. However, no further B–C bond cleavage resulted. In contrast, (I; R = H), formed in the hydrolysis of $B_3C_2^{1,5}H_5$, is unstable towards water, and there is evidence for some cleavage [equation (2)].

$$\begin{array}{l} (HO)_2BCH_2B(OH)CH_2B(OH)_2 \xrightarrow{H_2O} \\ \qquad (HO)_2BCH_2B(OH)_2 + BMe(OH)_2 \\ \qquad \Big\downarrow {\scriptstyle H_2O} \\ \qquad BMe(OH)_2 + B(OH)_3 \qquad (2) \end{array}$$

The structure of the smallest carbaborane $B_3C_2^{1,5}H_5$ has sometimes been described as a simple [1.1.1]bicyclic compound with little B–B bonding about the non-bridging boron atoms.[4,5] Such a structure contains strained four-membered B–C–B–C rings that are expected to be more susceptible to hydrolytic cleavage than acyclic organoboron compounds.[6] Two overall modes of ring cleavage, both of which remove ring strain, are possible [equations (3) and (4)]. With both

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[1] Primary refs. in R. M. Adams, 'Boron, Metallo-Boron Compounds and Boranes,' Interscience, New York, 1964, pp. 507—692.

[2] R. N. Grimes, 'Carboranes,' Academic Press, New York, 1970.

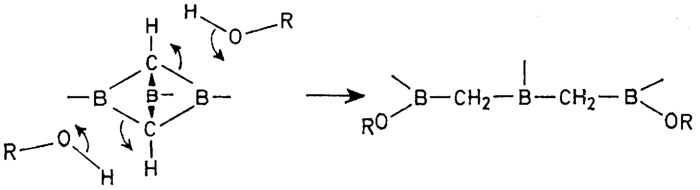
[3] G. R. Eaton and W. N. Lipscomb, 'NMR Studies of Boron Hydrides and Related Compounds,' W. A. Benjamin, Inc., New York, 1969, pp. 437—516.

[4] C. S. Cheung, R. A. Beaudet, and G. A. Segal, *J. Amer. Chem. Soc.*, 1970, **92**, 4158.

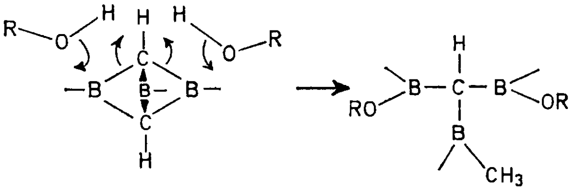
[5] T. Onak and E. Wan, *J.C.S. Dalton*, 1974, 665.

[6] T. Onak, 'Organoborane Chemistry,' Academic Press, New York, 1975.

the methanolysis and hydrolysis reactions, equation (3) predominates with only a suggestion in the ^{1}H n.m.r. spectrum of the boron product expected from (4).



(3)



(4)

The largest of the three *closo*-carbaboranes studied, $B_5C_2^{2,4}H_7$, is surprisingly more reactive than the smaller octahedral $B_4C_2^{1,6}H_6$. Methanolysis of the former gave $(MeO)_2BCH_2B(OMe)_2$, $BMe(OMe)_2$, and $B(OMe)_3$ in a stoicheiometry that suggests a *ca.* 1 : 2 combination of equations (5) and (6). It is felt that two different cage degradative mechanisms are operating in

$$B_5C_2^{2,4}H_7 + 12MeOH \longrightarrow (MeO)_2BCH_2B(OMe)_2 + BMe(OMe)_2 + 2B(OMe)_3 + 7H_2 \quad (5)$$

$$B_5C_2^{2,4}H_7 + 13MeOH \longrightarrow 2BMe(OMe)_2 + 3B(OMe)_3 + 7H_2 \quad (6)$$

this pentagonal-bipyramidal carbaborane rather than that in equation (5) followed by further cleavage of the diboron compound $(MeO)_2BCH_2B(OMe)_2$; for, under the conditions of the methanolysis reaction, the latter compound appears to be rather stable.[7]

Both methanolysis and hydrolysis of $B_4C_2^{1,6}H_6$ were very slow and incomplete under the conditions employed and resulted in the cleavage products in equation (7).

$$B_4C_2^{1,6}H_6 + 10\,ROH \longrightarrow 2BMe(OR)_2 + 2B(OR)_3 + 5H_2 \quad (7)$$

Mass Spectra.—As remarked above the mass spectrum cut off for (I; R = Me) corresponds to $[P - (OMe)_2]^+$. The compounds $BMe(OMe)_2$ and $(MeO)_2BCH_2B(OMe)_2$ also gave very weak parent ions in the mass spectra; fragmentation to $[P{-}Me]^+$ in both instances appears to be the first step in sequential removal of Me and O, or MeO, groups. Facile loss of Me and MeO groups in organoboron systems is not considered unusual.[8]

EXPERIMENTAL

The carbaboranes used in the experiments were contaminated with trace amounts of hydrocarbon impurities which could not be easily removed but which did not take part in any of the reactions reported below. All materials were handled using conventional vacuum-line techniques.

Methanolysis of 1,5-*Dicarba*-closo-*pentaborane*(5).—The carbaborane (0.110 g, 1.80 mmol) reacted slowly at −40 °C with pure dry methanol (0.256 g, 8.00 mmol) to give hydrogen, identified mass spectrometrically (4.78 mmol), excess of the carbaborane (0.0135 g, 0.22 mmol), trimethoxyborane (0.008 g, 0.08 mmol), identified by i.r. and n.m.r. spectroscopy, and the ester, $(MeO)_2BCH_2B(OMe)CH_2B(OMe)_2$, (I; R = Me) (0.356 g, 1.65 mmol) (Found: C, 38.4; H, 8.85. $C_7H_{19}B_3O_5$ requires C, 39.0; H, 8.90%), a colourless liquid which was trapped completely at −20 °C. The ^{11}B n.m.r. spectrum of the ester showed absorptions at δ −32.2 and −52.2 p.p.m. (intensities 2 : 1 respectively); in the ^{1}H n.m.r. spectrum, measured against internal $CHCl_3$, there were singlets at τ 6.72 (intensity 3), 6.86 (12), and 9.85 (4). The stoicheiometry of the reaction was not noticeably changed by the use of excess of methanol in place of excess of carbaborane. The presence of traces of methanol caused the collapse of the two singlets at τ 6.72 and 6.86 in the spectrum of the ester to a single sharp line through rapid exchange of MeO groups.

[$^2H_1$]Methanol (0.215 g, 6.52 mmol) and $B_3C_2^{1,5}H_5$ (0.068 g, 1.11 mmol) were allowed to react together below 0°C in a small sealed tube. The non-condensable gas (3.10 mmol) was totally consumed when passed over heated copper oxide in a furnace, demonstrating the absence of methane. Examination by mass spectrometry showed the gas to be at least 95% HD. The remaining products were excess of [$^2H_1$]methanol (0.046 g, 1.39 mmol), a trace amount of $B(OMe)_3$, and the ester $(MeO)_2BCHDB(OMe)CHDB(OMe)_2$ (0.219 g, 1.01 mmol) with ^{11}B n.m.r. signals at δ −31.5 and −53.3 p.p.m. (intensities 2 : 1) and ^{1}H n.m.r. signals at τ 6.76 (3 H), 6.89 (12 H), and 9.91 (2 H, br).

Hydrolysis of $B_3C_2^{1,5}H_5$.—A trial reaction on a small scale between $B_3C_2^{1,5}H_5$ (0.036 g, 0.58 mmol) and H_2O (0.010 g, 0.56 mmol) at room temperature in tetrahydrofuran (thf) gave hydrogen (0.59 mmol) and excess of carbaborane (0.23 g, 0.38 mmol). The semicrystalline residue dissolved in water gave ^{11}B n.m.r. peaks at δ −54, −33, and −20 p.p.m., the middle peak being the strongest, and ^{1}H n.m.r. singlets at τ 9.39, 9.65, and 9.80 (external $SiMe_4$). The pH of the solution was *ca.* 4.

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[7] R. B. Castle and D. S. Matteson, *J. Organometallic Chem.*, 1969, **20**, 19.

[8] R. W. Law and J. L. Margrave, *J. Chem. Phys.*, 1956, **25**, 1086; Y. Wada and R. W. Kiser, *J. Phys. Chem.*, 1964, **68**, 1588; P. J. Fallon, P. Kelly, and J. C. Lockhart, *J. Mass Spectrometry Ion Phys.* 1968, **1**, 133; P. J. Fallon and J. C. Lockhart, *ibid.*, 1969, **2**, 247; R. H. Cragg, J. F. J. Todd, and A. F. Weston, *J.C.S. Dalton*, 1972, 1373; R. H. Cragg and A. F. Weston, *J. Organometallic Chem.*, 1974, **67**, 161.

Reaction of $B_3C_2^{1,5}H_5$ (1.46 mmol, 0.48 mmol recovered) and water (0.100 g, 5.54 mmol) at room temperature gave hydrogen (2.91 mmol), $BMe(OH)_2$ (τ 9.86; ^{11}B at δ −32.2 p.p.m.; 0.18 mmol), and a crystalline involatile residue (0.14 g). Rapid recrystallisation of the residue from water gave the pure acid $(HO)_2BCH_2B(OH)CH_2B(OH)_2$, (I; R = H) (0.065 g, 0.44 mmol), with one ^1H n.m.r. absorption in the CH_nB region at τ 9.49 (Found: C, 15.35; H, 6.25; B, 23.15. $C_2H_9B_3O_5$ requires C, 16.5; H, 6.20; B, 22.3%).

In another reaction a mixture of $B_3C_2^{1,5}H_5$ (1.09 mmol used, 0.32 recovered) and H_2O (0.077 g, 4.30 mmol) evolved hydrogen (2.38 mmol), and $BMe(OH)_2$ (0.012 g, 0.20 mmol), easily recovered by fractionation, was identified by n.m.r. spectroscopy (τ 9.89). The residue in MeOH gave ^{11}B singlets at δ −19 (area 1), −32 (7.5), −53 (3) p.p.m. immediately on making up the solution, but after standing for 1 week at room temperature the peak at δ −53 p.p.m. had disappeared. Similar results were obtained in water and *NN*-dimethylformamide–water solvents. It is concluded that further hydrolysis of the initially formed product can occur, giving eventually $B(OH)_3$ and $BMe(OH)_2$.

Methanolysis of 2,4-*Dicarba*-closo-*heptaborane* (7).—Into a 5 mm n.m.r. tube was condensed $B_5C_2^{2,4}H_7$ (0.0425 g, 0.50 mmol) and dry methanol (0.016 g, 0.50 mmol). Monitoring of the ^{11}B n.m.r. spectrum at room temperature indicated that 4.2% of the carbaborane in the mixture disappeared after 10 min at 25 °C with the concomitant appearance of two moderately broad singlets at δ −18 and −32 p.p.m., area ratio 1.15 : 1, respectively. After 3 h, 8.7% of the carbaborane had reacted, with the two lower-field singlets growing in area and continuing to maintain the same area ratio. Very little further change in the ^{11}B n.m.r. intensities was evident after 1 week, and after subsequent heating to 100 °C (18 h) the relative areas had settled to 20.1 (summation of all $B_5C_2^{2,4}H_7$ resonances): 1.15 (δ −18): 1.00 (δ −32 p.p.m.). The ^1H n.m.r. spectrum showed, in addition to the unchanged carbaborane pattern,[9] singlet resonances at τ 6.65 (OMe), 6.70 (OMe), 9.86 (BCH_2B), and 10.00 (MeB) with no OH peak evident (expected at τ *ca.* 5.5—6.1).

In a separate experiment a mixture of $B_5C_2^{2,4}H_7$ (0.042 g, 0.48 mmol) and methanol (0.165 g, 5.16 mmol) was allowed to stand at room temperature for 4 weeks. After measuring the liberated hydrogen (3.02 mmol), cold-column [10] fractionation of the condensable materials gave $B(OMe)_3$ (0.099 g, 0.95 mmol; *ca.* −75 °C from column), $BMe(OMe)_2$ (0.044 g, 0.50 mmol; −88 °C), and $(MeO)_2BCH_2B(OMe)_2$ (0.018 g, 0.11 mmol; −40 °C). A trace amount of non-volatile residue remaining in the original reaction flask gave a very weak ^{11}B resonance at δ −31 p.p.m. when dissolved in thf.

A repeat of the reaction using $B_5C_2^{2,4}H_7$ (0.0425 g, 0.50 mmol) and dry methanol (0.160 g, 5.0 mmol) gave, after 2 weeks at room temperature, $B(OMe)_3$ (0.088 g, 0.85 mmol), $BMe(OMe)_2$ (0.029 g, 0.33 mmol), $(MeO)_2BCH_2B(OMe)_2$ (0.027 g, 0.17 mmol), and unchanged $B_5C_2^{2,4}H_7$ (0.006 g, 0.07 mmol).

The identity of $B(OMe)_3$ was established by the ^{11}B n.m.r. singlet at δ −18.5 p.p.m.,[11] the ^1H n.m.r. singlet at τ 6.75 [12] (internal $SiMe_4$), the i.r. pattern,[13] and the mass spectrum with a cut off at *m/e* 104 ($[^{12}C_3{}^1H_9{}^{16}O_3{}^{11}B]^+$) which is 58% of the base peak at *m/e* 73 $[B(OMe)_2]$.[8] The ester $BMe(OMe)_2$ was identified by an ^{11}B singlet at δ −31.5 p.p.m.,[14] two singlets at τ 6.69 (OMe) and 10.00 (CH_3B) [15] with a measured area ratio of 2.1 : 1.0 in the ^1H n.m.r. spectrum, an i.r. pattern very similar to that of $B(OMe_3)$ but with much less absorption in the region 14.2—15.7 µm and additional medium-intensity absorption at 11.12, 9.54, and 7.8—8.0 µm, and a mass spectrum which exhibited important contributions at *m/e* 43 (26, $[BO_2]^+$), 56 {5.3, $[BO(OCH)]^+$}, 57 {14, $[BMe(OMe)]^+$ or $[BO(OCH_2)]^+$}, 72 {29, $[BEt(O_2)]^+$}, 73 {100, $[BMe_2(O_2)]^+$}, 88 (1—2%, $[^{12}C_3{}^1H_9{}^{16}O_2{}^{11}B]^+$), and *M* (gas density) 86 ± 2 (calc.: 87.8). The compound $(MeO)_2BCH_2B(OMe)_2$ exhibited a ^{11}B singlet at δ −31.2 p.p.m., two ^1H n.m.r. singlets at τ 6.66 (OMe) and 9.86 (CH_2) in an area ratio 6 : 1,[7,16] an i.r. pattern similar to that described by Castle and Matteson [7] with the difference that the absorptions at 9.00 and 12.8—12.9 µm in the gas phase were very weak, and a mass spectrum with important contributions at *m/e* 43 (34, $[BO_2]^+$), 55 (15), 56 {54, $[BCH_2(OMe)]^+$ or $[BO(OCH)]^+$}, 57 {39, $[BMe(OMe)]^+$ or $[BO(OCH_2)]^+$}, 72 (28), 73 {100, $[B(OMe)_2]^+$}, 98 (11), 99 {21%, $[CH_2B_2H(OMe)_2]^+$}, 114 (12), 115 {22, $[(MeO)_2$-$BCH_2B(OH)]^+$}, 128 (7), 129 {9, $[(MeO)_2BCH_2B(OMe)]^+$}, 144 (42), and 145 {81%, $[(MeO)_2BCH_2B(OMe)O]^+$}. This mass-spectroscopic pattern was, within experimental error, identical to that of an authentic sample donated by D. S. Matteson.

Hydrolysis of $B_5C_2{}^{2,4}H_7$.—Water (0.090 g, 5.0 mmol) was mixed with $B_5C_2^{2,4}H_7$ (0.085 g, 1.0 mmol) in thf (2.0 mmol). At room temperature, slow evolution of H_2 commenced at the interface of the two phases. After 2 d a considerable quantity of white crystalline solid had settled on the walls of the container. The ^{11}B n.m.r. spectrum of the mixture gave a weak $B_5C_2^{2,4}H_7$ signal and a broad singlet at δ −30 p.p.m. The ^1H n.m.r. spectrum exhibited, in addition to a downfield OH signal, a singlet at τ 9.85. After 5 d the volatile portion of the mixture was transferred to another n.m.r. tube. The ^{11}B spectrum showed weak $B_5C_2^{2,4}H_7$ resonances together with a broad singlet δ −30.7 p.p.m.[17] $[BMe(OH)_2]$. The boron-decoupled ^1H n.m.r. spectrum gave, in addition to thf resonances, singlets at τ 3.04 (OH) and 9.85 (Me) [18] in an area ratio of 2 : 3 respectively. The purified acid $BMe(OH)_2$ was further identified by an i.r. pattern identical to a literature spectrum,[19] and a mass spectrum which approximated closely to that of the anhydride $(BMeO)_3$.[20] The ' non-volatile ' solids proved to be mostly $B(OH)_3$ and some $(HO)_2BCH_2B(OH)_2$ (i.r. and n.m.r. analysis) which gave, in water as solvent, ^{11}B resonances at δ −19 and −33 p.p.m. in a 3 : 1 ratio. The ^1H n.m.r.

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[10] J. Dobson and R. Schaeffer, *Inorg. Chem.*, 1970, **9**, 2183.

[11] T. Onak, H. Landesman, R. E. Williams, and I. Shapiro, *J. Phys. Chem.*, 1959, **63**, 1533; $\delta = -18.1 \pm 0.5$ p.p.m. for ^{11}B singlet of $B(OMe)_3$.

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[13] W. J. Lehmann, T. Onak, and I. Shapiro, *J. Chem. Phys.*, 1959, **30**, 1215.

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[16] P. Mattschei, Ph.D. Thesis, Washington State University; CH_2 shift of τ 9.88.

[17] Ref. 12, δ −31.9 p.p.m. (referenced to $Et_2O \cdot BF_3$).

[18] Ref. 12, τ 5.02 and 9.54 (in D_2O) (external $SiMe_4$).

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spectrum of the aqueous solution gave a weak singlet at τ 9.7.

Methanolysis of 1,6-*Dicarba*-closo-*hexaborane*(6).—Methanol (0.244 g, 7.0 mmol) and $B_4C_2^{1,6}H_6$ (0.073 g, 1.0 mmol) were sealed in a flask (25 cm^3) equipped with a 5 mm tube for monitoring n.m.r. spectra. No substantial reaction occurred until the mixture was heated to 100—125 °C for a 2-week period. At this time *ca.* 20% of the carbaborane had disappeared and two singlets at δ −18.7 [$B(OMe)_3$] and −31.2 p.p.m. [$BMe(OMe)_2$] (area ratio *ca.* 1.1 : 1) appeared in the ^{11}B n.m.r. spectrum together with a weak broad singlet at *ca.* τ 9.8—10.0 in the ^1H n.m.r. The flask was opened and the contents fractionated by means of a cold column. Small quantities of both $BMe(OMe)_2$ (0.07 mmol) and $B(OMe)_3$ were obtained with large amounts of unchanged $B_4C_2^{1,6}H_6$ and methanol.

Hydrolysis of $B_4C_2^{1,6}H_6$.—A mixture of water (0.35 mmol) and thf (0.3 mmol) was sealed with $B_4C_2^{1,6}H_6$ (0.07 mmol) in a 4 mm n.m.r. tube. By monitoring the ^{11}B and ^1H n.m.r. spectra it was shown that very little reaction occurred until the mixture was heated to 95 °C for 5 d. About two thirds of the carbaborane had then disappeared and two new broad ^{11}B resonances developed at δ − 19 [$B(OH)_3$] and − 32 p.p.m. [$BMe(OH_2)$] in approximately equal area. The δ − 19 p.p.m. peak was slightly weaker than the lower-field resonance, but this was attributed to crystals of $B(OH)_3$ forming on the walls of the tube. The ^1H n.m.r. spectra of the mixture showed the development of a singlet at τ 9.89 [$BMe(OH)_2$].

Physical Measurements.—The ^{11}B (32.1 MHz) ($\delta = 0$ for $Et_2O{\cdot}BF_3$) and ^1H n.m.r. spectra (100.0 and 60.0 MHz) (τ 10.00 for $SiMe_4$) were recorded using Varian HA-100 and A-60 spectrometers. Mass spectra were obtained on a Varian CH-5 high-resolution instrument and i.r. spectra were recorded using a Beckman Acculab-3 spectrometer.

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