for orifice thickness. The data obtained are shown in Fig. 1 where the solid lines are derived from a least squares analysis of the data. The corresponding equations, temperature ranges and heats of vaporization derived from the simple Clausius–Clapeyron equation are shown below. The uncertainties in the constants represent standard deviations calculated from the residuals of the experimental data. The data of Burg and Schlesinger for trimethylamine–borane did not overlap the present data but appeared consistent on a log P *vs.* $1/T$ plot. They were consequently included in the least squares determination of the vapor pressure equation which as a result covers the longest temperature range of the four compounds.

Two measurements were carried out on ammonia–borane, NH_3BH_3 but, because of the small weight loss, their precision is low. They are sufficient to set an upper limit of one micron for the vapor pressure at 25°.

Methylamine–borane, $(CH_3)NH_2{:}BH_3$

$$\log P\,(\text{mm.}) = -\frac{4114(1 \pm 0.053)}{T} + 11.411(1 \pm 0.063)$$

Range = 0 to 45° $\Delta H = 18.8 \pm 1.0$ kcal./mole

Dimethylamine–borane, $(CH_3)_2NH{:}BH_3$

$$\log P\,(\text{mm.}) = -\frac{4034(1 \pm 0.038)}{T} + 12.544(1 \pm 0.042)$$

Range = 0 to 35° $\Delta H = 18.5 \pm 0.7$ kcal./mole

Trimethylamine–borane, $(CH_3)_3N{:}BH_3$

$$\log P\,(\text{mm.}) = -\frac{2962(1 \pm 0.014)}{T} + 9.894(1 \pm 0.014)$$

Range = 0 to 90° $\Delta H = 13.6 \pm 0.2$ kcal./mole

Ammonia triborane, $NH_3B_3H_7$

$$\log P\,(\text{mm.}) = -\frac{3739(1 \pm 0.0075)}{T} + 9.200(1 \pm 0.0096)$$

Range = 30 to 55° $\Delta H = 17.1 \pm 0.1$ kcal./mole

Inspection of the heats of sublimation shows that the values for the monomethyl- and dimethylamine compounds are approximately the same and are appreciably higher than the value for the trimethylamine compound. This observation agrees with the greater polar nature to be expected for these substances despite their lower melting points. The magnitude of the difference, approximately 5 kcal./mole, suggests that hydrogen bonding may be of importance in the crystal structure of the first two compounds.

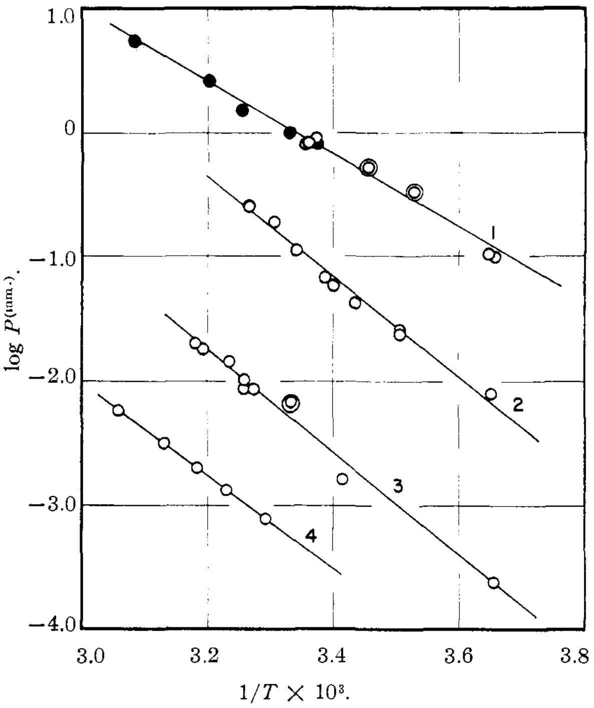

Fig. 1.—Vapor pressure–temperature relationships for some amine–boranes: 1, $(CH_3)_3N{:}BH_3$; 2, $(CH_3)_2NH{:}BH_3$; 3, $(CH_3)NH_2{:}BH_3$; 4, $NH_3{:}B_3H_7$ (●, ref. 1).

ANN ARBOR, MICHIGAN

[CONTRIBUTION FROM THE DEPARTMENT OF CHEMISTRY, UNIVERSITY OF MICHIGAN]

The Crystal Structure of $(NH_3)_2BH_2Cl$[1]

BY C. E. NORDMAN AND CHARLES R. PETERS

RECEIVED SEPTEMBER 8, 1958

An X-ray diffraction study of $(NH_3)_2BH_2Cl$ indicates that the compound forms orthorhombic crystals, $a = 10.20 \pm 0.04$ Å., $b = 10.20 \pm 0.04$ Å., $c = 17.42 \pm 0.08$ Å. A very prominent subcell exists, for which $a = b = 10.20$ Å., $c = 8.71$ Å. The approximate structure based on the subcell is consistent with the space group D_{2h}^{18}-Bbcm and consists of puckered layers of chloride ions interleaved with layers of $(NH_3)_2BH_2^+$ ions. The latter contain two B–N bonds of nearly equal length, 1.58 ± 0.02 Å. forming a nearly tetrahedral N–B–N angle.

Introduction

The compound $(NH_3)_2BH_2Cl$ was first prepared by Schultz and Parry[2] by allowing $B_2H_6{\cdot}2NH_3$ ("diammoniate of diborane") to react with NH_4Cl. More recently it has been obtained as a product of the reaction of $B_4H_{10}{\cdot}2NH_3$ with HCl.[3] In a recent series of papers[2,4] Parry and co-workers already have presented strong chemical evidence that the structure of the "diammoniate of diborane," long the subject of conflicting theories, is $[H_2B(NH_3)_2][BH_4]$ and that the structure of the corresponding halide salts is $[H_2B(NH_3)_2]X$, where X = Cl^- or Br^-. These conclusions have been supported by the results of Raman spectroscopic

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[1] Presented, in part, at the 133rd Meeting of the American Chemical Society, San Francisco, Calif., April, 1958.

[2] D. R. Schultz and R. W. Parry, THIS JOURNAL, **80**, 4 (1958).

[3] R. W. Parry and G. Kodama, Proc. XVI Congress, IUPAC, Inorganic Section, Paris, 1957, p. 483; G. Kodama and R. W. Parry, THIS JOURNAL, in press.

[4] S. G. Shore and R. W. Parry, THIS JOURNAL, **80**, 8 (1958); **80**, 12 (1958); R. W. Parry and S. G. Shore, *ibid.*, **80**, 15 (1958); S. G. Shore, P. R. Girardot and R. W. Parry, *ibid.*, **80**, 20 (1958).

studies of $B_2H_6 \cdot 2NH_3$ in liquid ammonia solution.[5] The present X-ray diffraction study was undertaken in order to gain additional independent information relating to these problems.

Experimental

Samples of $(NH_3)_2BH_2Cl$ prepared according to methods previously described were generously supplied by Dr. Goji Kodama and Mr. Masanobu Yamauchi, both of this department. The preparation of a single crystal suitable for X-ray work proved difficult due to the fact that the compound is decomposed by most solvents containing protonic hydrogen, as it is by traces of moisture. Acetone, diethyl ether and hydrocarbons do not dissolve $(NH_3)_2BH_2Cl$, and crystallization from liquid ammonia yields a microcrystalline powder. A few imperfect crystals were obtained, however, by dissolving the compound in Ansul 141 (diethylene glycol dimethyl ether) and layering the solution with diethyl ether. These crystals were sealed in thin walled glass capillaries for X-ray examination. In spite of repeated attempts only one crystal suitable for single crystal work was obtained.

Zero, first and second level precession or Weissenberg patterns of the three principal nets were recorded using Mo Kα or Cu Kα radiation. This yielded a total of 186 independent intensities. The X-ray work was discontinued at this point due to the deterioration of the crystal.

Unit Cell and Space Group.—The diffraction patterns showed nearly tetragonal symmetry and the axes were labeled accordingly. On the nets perpendicular to the *c* axis, however, systematic intensity variations were found which violated the fourfold symmetry and led to the conclusion that the crystals were orthorhombic with two axes, *a* and *b*, of equal length. It was further observed that the only systematic absences in the *hk*0 net occurred when *h* and *k* were both odd. Since this is not a space group extinction, it was concluded that the crystal was a twin consisting of two fragments turned with respect to each other by 90 degrees about the *c*-axis. The observed absences in *hk*0 could then be explained as occurring for odd *h* in each of the fragments. Since the *a* and *b* axes were equal in length, all *hk*0 reflections with *h* and *k* even were overlaps of, in general, non-equivalent reflections from the two fragments. This was not the case for reflections with *k* odd, in view of the systematic absences for odd *h*. Thus, *hk*0 reflections with $h + k$ odd were "pure" and afforded a means of determining the relative size or diffractive power, of the two fragments. The ratio of the intensity of *hkl*, divided by the Lorentz and polarization factors, to that of *khl* was found to be very nearly constant for all *hk*0 with $h + k$ odd, as well as for corresponding upper level reflections. This constituted strong evidence that the crystal was indeed a twin and yielded a value for the ratio of the sizes of the fragments.

The unit cell parameters are $a = 10.20 \pm 0.04$ Å., $b = 10.20 \pm 0.04$ Å., $c = 8.71 \pm 0.04$ Å., where the limits of error are taken as three times the estimated standard deviation. The calculated density is 1.21 g. cm.$^{-3}$ assuming eight formula units per cell.

Systematic absences, referred to the untwinned crystal, were observed in *hkl* for $h + l$ odd, in 0*kl* for *k* odd and in *h*0*l* for *h* odd. The possible space groups are then D_{2h}^{18}-Bbcm and C_{2v}^{17}-Bba2. The first of these has sixteen general positions and consequently requires at least the chlorine and boron atoms to lie in special positions.

In addition to the reflections leading to the above unit cell assignment a set of very weak and somewhat diffuse reflections was observed calling for a doubling of the *c*-axis. The true unit cell parameters are then $a = 10.20 \pm 0.04$ Å., $b = 10.20 \pm 0.04$ Å., $c = 17.42 \pm 0.08$ Å. In view of the relatively small number of clearly observable reflections in this category, no attempt was made to determine the space group applicable to the doubled cell. The reflections demanding the doubled cell were ignored in the structure analysis.

Structure Determination.—Intensity data were obtained from the diffraction photographs by visual comparison with a series of timed exposures of spots of similar shape. Application of the Lorentz and polarization factors yielded a set of raw values of the squares of the structure factors. For every spot observed on two films the ratios of these raw F^2-values were calculated and averaged to give the ratio of the scale factors to be applied to the two films. Through appropriate averaging of such ratios the scale factors themselves were found and the raw F^2-values placed on a common scale.

Due to the twinning of the crystal most of the values obtained were derived from spots which were overlaps of contributions from the two fragments, whereas the rest of the values arose from one or the other of the two fragments.

From the nature of the twinning and the symmetry of the reciprocal lattice, it follows that a reflection of *hkl* from one of the fragments (A) will appear as "*khl*" when produced by the other fragment (B). If $h + k$ is odd, either the reflection *hkl* or *khl* produced by A must be absent, since all reflections with $h + l$ odd are absent. For example, if *h* is even and *k* odd, a reflection with even *l* indexed as "*khl*" must in fact be the *hkl* reflection from fragment B. Furthermore, it must be "pure" inasmuch as it cannot contain a contribution from an overlapping reflection from A. For the same reason the reflection indexed as *hkl* must be a pure reflection from fragment A.

Twenty-seven independent reflections with $h + k$ odd were observed. The ratios of the raw F^2-values of these reflections *hkl* to the corresponding values of the reflections "*khl*" were found to be constant within the error of measurement. The average value of this ratio, $1/r$, expresses the ratio, A to B, of the two crystal fragments. The value of *r* was found to be 0.26.

All reflections for which $h + k$ is even are overlaps of contributions from both fragments. If the raw F^2-values of these reflections are denoted $I(hkl)$, we have

$$I(hkl) = F^2(hkl) + rF^2(khl)$$

and

$$I(khl) = F^2(khl) + rF^2(hkl)$$

Consequently, the expression

$$F^2(hkl) = [I(hkl) - rI(khl)]/(1 - r^2)$$

yields the magnitudes of the individual structure factors expressed on the scale of the large fragment.

The magnitudes of the structure factors with all indices even are, on the average, considerably larger than the rest. This observation led to the conclusion that the chlorine atoms, which dominate the scattering, must lie at or near the points of a simple tetragonal lattice having one-half the axial lengths of the assumed cell. The *hk*0 and *h*0*l* electron density projections therefore were computed using structure factors with even indices only, with signs based on the chlorine contributions. The *h*0*l* projection showed the nitrogen and boron atoms to lie in layers perpendicular to the *c*-axis half-way between the assumed chlorine layers. This arrangement is consistent with Bbcm, the more symmetrical of the two possible space groups, if the boron and nitrogen atoms lie in mirror planes at $z = 0$ and $^1/_2$ and the chlorine atoms on twofold axes parallel to the *c*-axis at $(x,y) = (0,0), (0,^1/_2)\ (^1/_2,0)$ and $(^1/_2,^1/_2)$. Since any violation of these conditions was, at most, very slight, the space group was tentatively assumed to be Bbcm.

The eightfold chlorine positions are then $(0,0,0;\ ^1/_2,0,^1/_2) + 0,0,z;\ 0,0,\bar{z};\ ^1/_2,^1/_2,z;\ ^1/_2,^1/_2,\bar{z}$ where the parameter *z* is approximately 0.25. The nitrogen and boron atoms are located at eightfold positions of the type $(0,0,0;\ ^1/_2,0,^1/_2) + x,y,0;\ \bar{x},\bar{y},0;\ x,^1/_2 - y,^1/_2;\ \bar{x},^1/_2 + y,^1/_2$. Approximate *x*- and *y*-coördinates of the two nitrogen atoms and one boron atom were derived from the *hk*0 electron density projection. The suggested configuration was an angular N–B–N group with two approximately equal B–N distances.

The seven atomic coördinates, the scale factor k and the isotropic temperature factor *B* were now refined by least squares methods. Specifically, the quantity

$$\Sigma w(hkl)[\mathrm{k}F_0(hkl) - F_c(hkl)]^2/\Sigma w(hkl)\mathrm{k}^2F_0^2(hkl)$$

was minimized, where the weight $w(hkl)$ of each reflection was taken as $3F_{\min}/F(hkl)$ if $F(hkl) > 3F_{\min}$ and equal to unity otherwise.

The least squares refinement proceeded to $R_1 = \Sigma|\mathrm{k}|F_0| - |F_c||/\Sigma \mathrm{k}|F_0| = 0.17$ at which point several Fourier sections and $(F_0 - F_c)$ syntheses of the electron density were computed. The chief reason for this was to check for indications of lower symmetry than that of Bbcm. Since no such indications were found, it must be concluded that the space group is Bbcm, at least to the accuracy of this deter-

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[5] R. C. Taylor, D. R. Schultz and A. R. Emery, THIS JOURNAL, **80**, 27 (1958).

TABLE I

OBSERVED AND CALCULATED STRUCTURE FACTORS

hkl	kF_0	F_c	*hkl*	kF_0	F_c	*hkl*	kF_0	F_c
000	..	+352	244	43	+ 50	608	12	+ 6
002	22	+ 37	246	20	− 24	610	29	−28
004	117	+126	248	13	+ 14	612	23	−24
006	18	− 3	2.4.10	8	− 5	620	62	+57
008	36	+ 32	250	19	− 18	622	38	−45
0.0.10	8	+ 1	252	15	− 16	624	31	+36
020	87	+ 72	260	74	+ 61	626	19	−20
022	173	−130	262	43	− 42	628	11	+11
024	35	+ 42	264	37	+ 38	630	14	+11
026	38	− 40	266	16	− 19	640	40	+38
028	13	+ 9	268	13	+ 12	642	50	−48
0.2.10	14	− 10	280	53	+ 49	650	14	−14
040	179	+141	282	21	− 24	652	13	−13
042	34	− 33	284	36	+ 34	660	67	+46
044	57	+ 67	286	8	− 11	662	20	−24
046	18	− 16	288	12	+ 11	680	35	+28
048	20	+ 20	2.10.0	28	+ 26	682	23	−23
0.4.10	6	− 2	2.10.2	24	− 24	6.10.0	30	+22
060	23	+ 26	2.10.4	10	+ 18	6.10.2	13	−16
062	92	− 76	2.10.6	10	− 12	6.12.2	11	−11
064	17	+ 18	2.12.0	20	+ 19	711	15	−12
066	22	− 33	2.12.2	10	− 13	751	11	− 9
068	10	+ 5	2.14.2	14	− 11	771	10	+ 6
080	41	+ 39	311	21	− 19	800	36	+37
082	37	− 37	321	26	+ 25	802	40	−39
084	19	+ 25	323	13	+ 17	804	25	+23
086	11	− 18	331	9	− 8	806	21	−19
0.10.0	25	+ 22	341	22	+ 19	820	56	+50
0.10.2	27	− 29	351	8	− 7	822	18	−23
0.10.4	13	+ 16	400	135	+144	824	29	+35
0.10.6	12	− 13	402	37	− 32	826	11	−10
0.12.0	22	+ 18	404	72	+ 68	840	30	+27
0.12.2	15	− 16	406	23	− 16	842	42	−37
113	34	+ 28	408	22	+ 20	860	35	+29
115	28	− 18	410	17	− 19	862	23	−23
117	23	+ 18	412	14	− 13	880	16	+17
119	18	− 10	420	74	+ 76	882	28	−23
121	47	− 48	422	57	− 66	8.10.0	19	+14
123	25	− 31	424	46	+ 49	8.10.2	11	−13
125	10	− 16	426	27	− 24	8.12.2	11	− 8
131	20	− 18	428	11	+ 13	10.0.0	18	+23
141	19	− 20	430	25	+ 22	10.0.2	31	−28
161	23	− 24	432	14	+ 18	10.0.4	16	+17
171	16	− 12	440	71	+ 72	10.0.6	13	−13
181	9	+ 8	442	49	− 49	10.2.0	24	+25
200	66	+ 75	460	40	+ 36	10.2.2	24	−25
202	109	−127	462	57	− 51	10.2.4	17	+17
204	49	+ 43	480	27	+ 29	10.4.0	26	+22
206	45	− 40	482	37	− 36	10.4.2	22	−22
208	14	+ 9	4.10.0	26	+ 22	10.6.0	28	+22
2.0.10	8	− 9	4.10.2	22	− 23	10.6.2	15	−16
210	45	− 50	4.12.0	9	+ 15	10.8.0	19	+14
212	31	− 40	4.12.2	13	− 14	10.8.2	12	−13
220	76	+ 57	521	15	− 17	12.0.0	17	+16
222	132	−121	523	13	− 13	12.0.2	18	−18
224	33	+ 34	531	7	− 7	12.2.0	22	+20
226	32	− 37	561	11	− 12	12.2.2	13	−12
228	17	+ 11	571	11	− 10	12.4.0	15	+14
2.10	7	− 7	600	30	+ 31	12.4.2	17	−15
230	27	+ 27	602	72	− 72	12.6.2	13	−10
232	19	+ 22	604	20	+ 21	12.8.2	9	− 9
240	89	+ 79	606	37	− 31	14.2.2	11	−11
242	68	− 64						

mination. Anisotropy was observed in the thermal motion of the chlorine atoms, the peaks being somewhat elongated in the z-direction. An anisotropic temperature factor $\exp[\alpha(h^2 + k^2) + \beta l^2]$ was therefore applied to f_{Cl^-}, and by successive difference syntheses the best values $\alpha = -0.0030$ and $\beta = +0.0020$ were found. To account for the somewhat stronger thermal motion in the nitrogen atoms than in the boron atom, an additional temperature factor $\exp(-0.75 \sin^2\theta/\lambda^2)$ was applied to f_N in calculating the structure factors.

It was not possible to locate the hydrogen atoms independently; however, their presence was assumed and an approximate hydrogen contribution included in the calculation of structure factors. Two hydrogen atoms were assumed to be attached to the boron atom, and six half-hydrogens to each of the nitrogen atoms. The hydrogen scattering factors were multiplied by an additional temperature factor $\exp[-1.0 \sin^2\theta/\lambda^2]$.

With these refinements included several more cycles of least squares adjustment of the seven atomic coördinates, scale and over-all temperature factors were carried out. The final agreement of observed and calculated structure factors is given by

$$R_1 = 0.135$$

and

$$R_2 = \Sigma(kF_0 - F_c)^2/\Sigma k^2F_0^2 = 0.036$$

The observed and final calculated structure factors are listed in Table I. An additional 96 structure factors which were part of the photographed nets were found to be unobservably small. The final calculated values of these structure factors showed no significant disagreement with their observed maximum limits.

When $h + k$ is odd, the geometrical structure factor has the form $A(hkl) = -16 \sin 2\pi hx \sin 2\pi ky \cos 2\pi lz$. Consequently, the chlorine atoms do not contribute to these structure factors. As a further check on the nitrogen and boron coördinates, a separate least squares refinement of these parameters was carried out using the 27 observed structure factors with $h + k$ odd only. This yielded coordinates very nearly equal to those obtained with the complete set of data and a value of $R_1 = 0.092$ for the set of 27 structure factors.

The final atomic coördinates (x,y,z) are (0, 0, 0.264) for Cl, (0.236, 0.064, 0) and (0.060, 0.237, 0) for N_1 and N_2, and (0.212, 0.219, 0) for B. Estimated standard deviations of these values are 0.0015, 0.002 and 0.003, respectively, for chlorine, nitrogen and boron coördinates. The reliability of these estimates, which are based on the least squares residuals, is doubtful since the effect of a possible error in r is unaccounted for. The final value of the parameter B in the over-all temperature factor $\exp(-B \sin^2\theta/\lambda^2)$ is 4.40 Å.^{2}.

Discussion

Electron density projections on (010) and (001) are shown in Figs. 1 and 2 and a perspective drawing of the structure in Fig. 3.

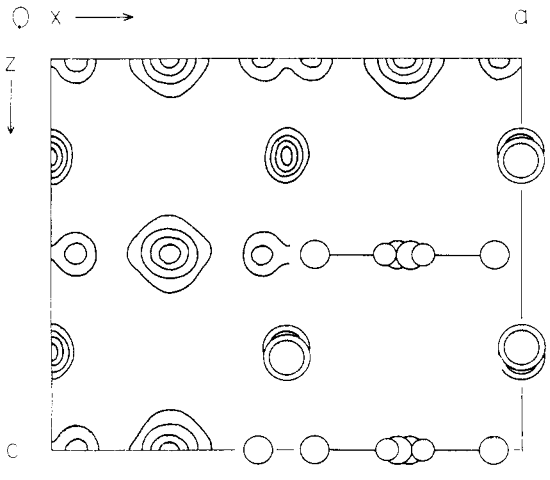

Fig. 1.—Electron density projection on (010), showing arrangement of boron (small circles), nitrogen (large circles) and chlorine atoms (double circles). Contour intervals are 4 e.Å.$^{-2}$ for B and N, 10 e.Å.$^{-2}$ for Cl, zero contour omitted.

Interatomic distances and angles are given in Fig. 2. The estimated standard deviations are 0.04 Å. for the B–N bond length and 0.02 Å. for the N . . . Cl distances.

Each NH_3 group is surrounded by four chlorine atoms at the corners of a distorted square while each chlorine is surrounded by eight NH_3 groups belonging to six different $(NH_3)_2BH_2^+$ at the corners of a strongly distorted cube. The arrangement of the chlorine atoms and the N . . . Cl distances

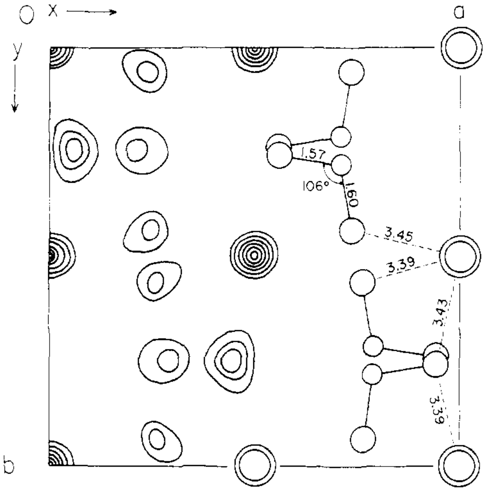

Fig. 2.—Electron density projection on (001), contours as in Fig. 1.

indicate that the compound is ionic, namely, $[(NH_3)_2BH_2^+]Cl^-$. The four symmetrically non-equivalent distances may be compared with the N . . . Cl distances in ammonium chloride (3.36 Å.) and particularly methylammonium chloride[6] (3.18 Å.) where the coördination is more comparable to that found in $[(NH_3)_2BH_2]Cl$. The lengthening of this distance in the present case is to be expected in view of the presumably smaller charge on each of the NH_3 groups in the $(NH_3)_2BH_2^+$ ion.

The lengths of the two N–B bonds in the cation are equal within experimental error, and also equal

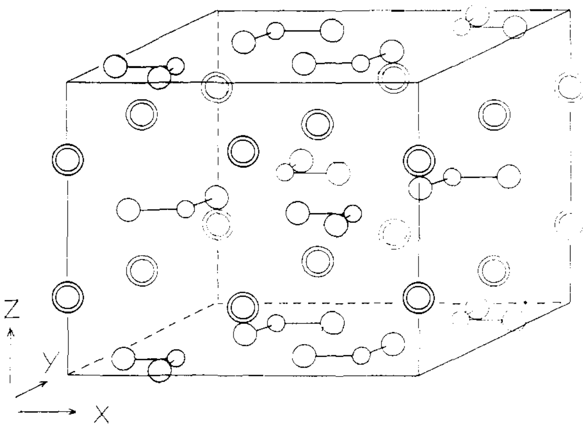

Fig. 3.—The structure of $(NH_3)_2BH_2Cl$.

to the N–B bond length found in ammonia-borane, NH_3BH_3,[7] and ammonia-triborane, NH_3-B_3H_7.[8] The N–B bond distance also is in close agreement with values found in the addition compounds of BF_3 with NH_3 and methyl amines.[9]

Acknowledgment.—This work was conducted, in part, under Contract AF33(616)-3343 with the U. S. Air Force, the sponsoring agency being the Aeronautical Research Laboratory of the Wright Air Development Center, Air Research and Development Command.

ANN ARBOR, MICHIGAN

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(7) E. W. Hughes, *ibid.*, **78**, 502 (1956); E. L. Lippert and W. N. Lipscomb, *ibid.*, **78**, 503 (1956).

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[JOINT CONTRIBUTION FROM CALLERY CHEMICAL COMPANY AND THE DEPARTMENT OF CHEMISTRY OF THE UNIVERSITY OF MICHIGAN[1]]

Systematics in the Chemistry of the Boron Hydrides

BY R. W. PARRY AND L. J. EDWARDS

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Stock originally recognized two groups of boron hydrides, the B_nH_{n+4} and B_nH_{n+6} series. This classification has been extended and used as a basis for the systematic arrangement of the boron hydrides. Four types of chemical reactions of the boron hydrides are recognized. These are: (1) symmetrical cleavage of the double bridge bond, (2) non-symmetrical cleavage of the double bridge bond, (3) loss of a proton from a hydrogen bridge bond, and (4) loss of molecular hydrogen from the molecule. The type of reaction expected from a given molecule is related to the position of the molecule in the systematic arrangement and to the geometry of the original boron hydride.

In his original reports on the boron hydrides Stock[2] suggested two series of compounds which he represented by the empirical formulas B_nH_{n+6} and B_nH_{n+4}. Early attempts to use these formulas as a basis for systematizing the chemistry of the boron hydrides were of limited success, since arguments based on analogies with the hydrocarbons were often misleading and little was known of the geometry of the molecules. In recent years the structures of nearly all of the known boron hydrides have been established with considerable certainty as a result of a number of very clever structural studies.[3] This structural information coupled with recently acquired chemical knowledge can now be used as a basis for a number of chemical correlations based on an extension of Stock's classification.

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[1] The research in boron chemistry at the University of Michigan is supported by the United States Air Force under Contract AF33(616)-3343 monitered by the Aeronautical Research Laboratory, WADC. The work at Callery is supported by the Bureau of Aeronautics of the United States Navy under contracts NOa(s)52-1024c and NOa(s) 58-454.

(2) A. Stock, "Hydrides of Boron and Silicon," Cornell University Press, Ithaca, N. Y., 1933, p. 17.

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