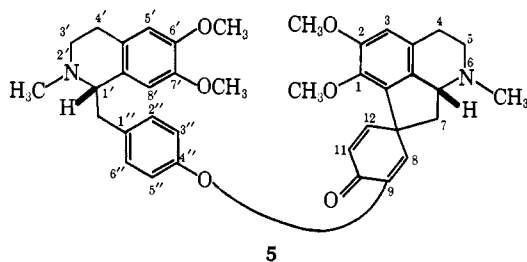
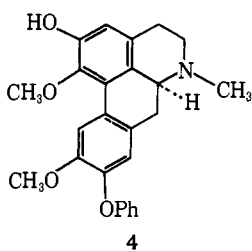


mp 137°. Comparison of the mass spectrum of **1a**,  $m/e$  608 ( $M^+$ ), 402 ( $M - x$ ), 312 ( $M - y$ ), 296 ( $M - z$ ), and 206 ( $x$ , base), with that of **1b**,  $m/e$  636 ( $M^+$ ), 430 ( $M - x$ ), 340 ( $M - y$ ), 324 ( $M - z$ ), and 206 ( $x$ , base), established that **1a** possessed a diphenolic aporphine moiety bonded through a diphenyl ether linkage to an armapavine-like residue.

The nmr spectrum of pakistanine **1a** in  $CDCl_3$  exhibits a high-field methoxyl singlet absorption at  $\delta$  3.61 (3 H, C-7'OCH<sub>3</sub>), two methoxyl singlets at 3.83 and 3.88 (6 H), two *N*-methyl singlets at 2.51 and 2.56 (6 H), a singlet at 6.06 (1 H, C-8'*H*), three aromatic proton singlets at 6.54, 6.56, and 6.71 (3 H), an A<sub>2</sub>B<sub>2</sub> quartet (4 H) at 7.00 ( $J = 8.5$  and 10 Hz) (C-2'', -3'', -5'', and -6''*H*), and a one-proton singlet at 8.11 (C-11*H*). The nmr spectrum of *O,O*-dimethylpakistanine (**1b**) shows two additional methoxy resonances, one at  $\delta$  3.72 and the other at 3.90, the former being characteristic of a C-1 methoxyl.<sup>3</sup> The remainder of the spectrum of **1b** is very similar to that of **1a**. The phloroglucinol test for an *o*-diphenol function<sup>4</sup> was negative for pakistanine **1a**, so that a methoxyl group could be assigned to the C-2 position.

Sodium in liquid ammonia cleavage of *O,O*-dimethylpakistanine (**1b**) yielded (–)-2,10-dimethoxyaporphine (**2**)<sup>1</sup> and L-(+)-armepavine (**3**).

The uv spectrum of pakistanine **1a** ( $\lambda_{max}^{EtOH}$  218, 270 sh, 277, and 307 nm (log  $\epsilon$  4.61, 4.13, 4.21, and 4.07)) resembles that of 9-phenylboldine (**4**),<sup>5</sup> C<sub>25</sub>-



H<sub>25</sub>O<sub>4</sub>N, mp 155–157° ( $\lambda_{max}^{EtOH}$  218, 270 sh, 276, and 303 nm (log  $\epsilon$  4.30, 4.23, 4.25, and 3.82)).

From the same plant source we have also obtained the first known proaporphine benzyloquinoline alkaloid, pakistanamine (**5**): C<sub>38</sub>H<sub>42</sub>O<sub>6</sub>N<sub>2</sub>; mp 158–162° dec (picrate); mp 215° (hydrochloride); amorphous free base;  $[\alpha]^{25D} + 20^\circ$  (MeOH).

The ir spectrum,  $\nu^{CHCl_3}$  1640 (C=C) and 1670 cm<sup>-1</sup> (conjugated C=O), and the uv spectrum,  $\lambda_{max}^{EtOH}$  225 sh, 280, and 310 sh nm (log  $\epsilon$  4.63, 4.12, and 3.61), of pakistanamine (**5**) were suggestive of the presence of a dienone moiety.<sup>6</sup> The nmr spectrum of pakistan-

(3) M. Tomita, H. Furukawa, S.-T. Lu, and S. M. Kupchan, *Chem. Pharm. Bull.*, **15**, 959 (1967); M. Tomita, S.-T. Lu, and Y.-Y. Chen, *Yakugaku Zasshi*, **86**, 763 (1966); and S. M. Kupchan and N. Yokoyama, *J. Amer. Chem. Soc.*, **85**, 1361 (1963).

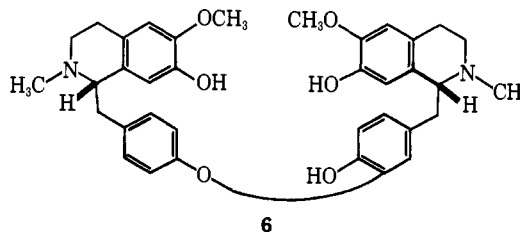
(4) H. A. Schroder, *J. Chromatogr.*, **30**, 537 (1967).

(5) Synthesized by us from (+)-boldine and bromobenzene via an Ullman condensation.

amine (**5**) in  $CDCl_3$  showed four methoxyl singlets at  $\delta$  3.57, 3.64, 3.80, and 3.82 (12 H), two *N*-methyl singlets at 2.32 and 2.35 (6 H), a singlet at 6.07 (1 H, C-8'*H*), a doublet at 6.16 (1 H,  $J_{8,12} = 2.5$  Hz, C-8*H*), a doublet at 6.32 (1 H,  $J_{11,12} = 10$  Hz, C-11*H*), a doublet of doublets at 7.06 (1 H,  $J = 2.5$  and 10 Hz, C-12*H*), singlets at 6.55 and 6.62 (2 H, C-3 and C-5'*H*), and finally a singlet at 7.02 (4 H, C-2'', -3'', -5'' and -6''*H*).

Dienone-phenol rearrangement of pakistanamine **5** using dilute HCl gave rise to the phenolic aporphine benzyloquinoline dimer **1c**: C<sub>38</sub>H<sub>42</sub>O<sub>6</sub>N<sub>2</sub>; mp 117°;  $[\alpha]^{25D} - 103^\circ$  (MeOH). The uv spectrum of **1c**,  $\lambda_{max}^{EtOH}$  225 sh, 267 sh, 277, and 304 nm (log  $\epsilon$  4.75, 4.28, 4.37, and 4.16), closely resembles that of pakistanine **1a**, and indeed *O*-methylation of **1c** with diazomethane furnished the product **1b**, identical in terms of melting point, mixture melting point, uv, ir, nmr, and mass spectra, tlc  $R_f$  values, and ORD curve with a sample of *O,O*-dimethylpakistanine derived by *O*-methylation of pakistanine **1a**.

The fact that pakistanine and pakistanamine are found in the same plant lends substantial support to the biogenetic scheme involving the sequence: bisbenzyloquinoline dimer → proaporphine benzyloquinoline dimer → aporphine benzyloquinoline dimer.<sup>7</sup> Noteworthy also is the observation that the required bisbenzyloquinoline precursor in the present instance corresponds to the alkaloid (+)-berbamunine (**6**) found



in *B. amurensis* Rupr. var. *japonica*,<sup>8</sup> a close relative of *B. baluchistanica*.

(6) K. Bernauer and W. Hofheinz, *Fortschr. Chem. Org. Naturst.*, **26**, 245 (1968).

(7) K. L. Stuart and M. P. Cava, *Chem. Rev.*, **68**, 321 (1968).

(8) M. Tomita and T. Kugo, *Yakugaku Zasshi*, **77**, 1075, 1079 (1967).

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## Reduction of Acetylene and Nitrogen by a Cobalt-Porphyrin System

Sir:

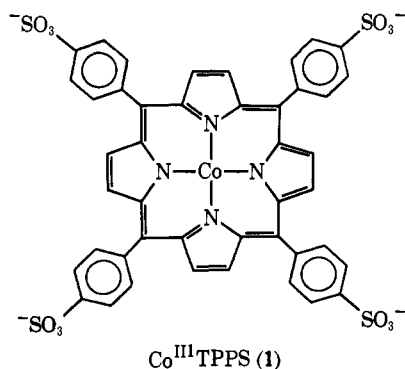
Since the successful isolation and purification of nitrogenase, the enzyme system that fixes molecular nitrogen to ammonia in living organisms, many studies have been made to find a model for nitrogenase action.<sup>1-3</sup> We present in this paper the description of a

(1) R. Murray and D. C. Smith, *Coord. Chem. Rev.*, **3**, 429 (1968); J. Chatt, *Proc. Roy. Soc., Ser. B*, **172**, 317 (1969); E. van Tamelen, *Accounts Chem. Res.*, **3**, 361 (1970).

(2) G. N. Schrauzer and P. A. Doemeny, *J. Amer. Chem. Soc.*, **93**, 1608 (1971); G. N. Schrauzer and A. Schlesinger, *ibid.*, **92**, 1808 (1970); R. E. Hall and R. L. Richards, *Nature (London)*, **233**, 144 (1971).

(3) W. E. Newton, J. L. Corbin, P. W. Schneider, and W. A. Bulen, *J. Amer. Chem. Soc.*, **93**, 268 (1971).

model system that reduces nitrogenase substrates; all the reactions of our model system are carried out in water at room temperature and 1 atm of pressure, including the nitrogen fixation. Since strong reducing agents such as sodium borohydride have been found to reduce cobalt(III) in porphyrins to Co(I),<sup>4</sup> and Co(I) complexes of vitamin B<sub>12</sub>, dimethylgloxime, and bipyridyl have been found to reduce nitrous oxide to molecular nitrogen,<sup>5</sup> we tried a model system consisting of the water-soluble *meso*-tetra(*p*-sulfonatophenyl)porphinatocobalt(III)<sup>6</sup> (Co<sup>III</sup>TPPS, 1) and sodium



borohydride and found it to be a very effective catalyst for the reduction of the substrates of nitrogenase.

Nitrogenase reduces acetylene specifically to ethylene and this reaction has generally been used as an assay for nitrogen-fixing activity.<sup>7</sup> Besides reducing acetylene, our system also catalytically reduces azide, cyanide, acetonitrile, propargyl alcohol, and methylacetylene; so far the system does *not* catalytically reduce nitrogen.

A typical experiment for the acetylene reduction is as follows. In a 50-ml erlenmeyer flask were placed 0.5 ml of CoTPPS stock solution ( $8 \times 10^{-3} M$ ), 2 ml of pH 10 buffer (borax-NaOH), and 6.5 ml of distilled water. Then the sealed flask was flushed with water-washed acetylene. Freshly prepared 1 *M* sodium borohydride (1 ml) was injected to initiate the reaction and after 10 min a 10% conversion of the acetylene to ethylene had taken place; some ethane was also produced.<sup>8</sup> The ratio of ethylene/ethane was dependent on pH; at a pH of 10 this ratio was >10. The introduction of carbon monoxide in the system (3 parts acetylene-1 part carbon monoxide) was found to completely inhibit the catalysis just as the nitrogenase action is inhibited by carbon monoxide. The catalytic properties of this system can be regenerated again and again by a new injection of fresh borohydride without loss of activity.<sup>9</sup>

The reduction of molecular nitrogen was carried out by passing pure dry *air* (pure nitrogen also works) through a solution containing 2 ml of the CoTPPS stock solution, 6 ml of pH 10 buffer, and 20 ml of distilled water in a filter flask. The effluent gas was

(4) H. W. Whitlock and B. K. Bower, *Tetrahedron Lett.*, **52**, 4827 (1965).

(5) R. Banks, R. J. Henderson, and J. M. Pratt, *Chem. Commun.*, 387 (1967).

(6) E. B. Fleischer, J. M. Palmer, T. S. Srivastava, and A. Chatterjee, *J. Amer. Chem. Soc.*, **93**, 3162 (1971); E. B. Fleischer and M. Krishnamurthy, *ibid.*, **93**, 3784 (1971).

(7) M. J. Dilworth, *Biochim. Biophys. Acta*, **127**, 285 (1966).

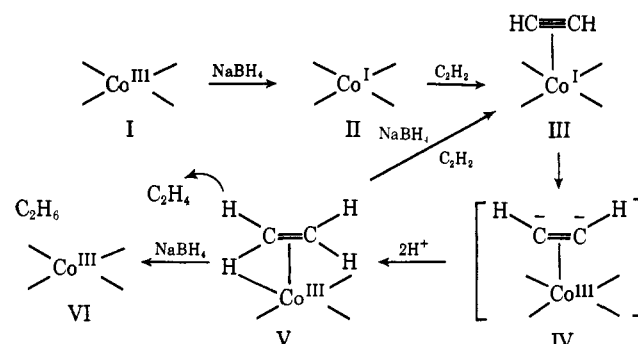
(8) The analyses of the gases was done by gas chromatography using a FM 700 gc with 25-ft DMS column at ambient temperatures and the carrier gas flow rate of 30 ml/min.

(9) The initial turnover number (first 10 min) for the catalysis with respect to acetylene reduction is 60 mol of acetylene  $\text{min}^{-1} \text{mol}^{-1}$  of catalyst.

bubbled through 20 ml of 0.1 *M* HCl in another flask. To initiate the reaction, 2 ml of freshly prepared 0.25 *M* sodium borohydride was injected with further 1-ml injections of borohydride every day for a week. The ammonia produced was determined by distilling a few milliliters of the solution in the reaction flask (after addition of NaOH) into the HCl solution in the second receiver flask. The ammonia was then determined employing Nessler's reagent.<sup>10,11</sup> This system produced between 0.8 and 1.0 mol of ammonia per mol of porphyrin catalyst. We are now searching for conditions to make the N<sub>2</sub> fixation catalytic.<sup>12,13</sup>

Scheme I outlines a possible mechanism for our catalytic system.<sup>14</sup> The reduction of the Co(III) to

Scheme I



Co(I) (species II) has previously been confirmed.<sup>4</sup> IV only portrays a method of counting electrons and does not imply an actual species in the reaction. When the reaction is carried out in D<sub>2</sub>O only *cis*-D<sub>2</sub>H<sub>2</sub>C<sub>2</sub> is produced with no trans isomer present in the acetylene reduction.

The catalytic acetylene reduction does not take place when either the Fe<sup>III</sup>TPPS or Mn<sup>III</sup>TPPS is employed in place of the Co<sup>III</sup>TPPS; on the other hand the acet-

(10) K. R. Middleton, *J. Appl. Chem.*, **10**, 281 (1960).

(11) To ascertain that the NH<sub>3</sub> did *not* come from the porphyrin catalyst, a total N by Kjeldahl digestion was carried out on samples both before and after catalysis. At least duplicate runs were made on the total nitrogen determination. In an experiment in which  $1.46 \times 10^{-5}$  mol of cobalt porphyrin was used in preparing the catalyst and in which  $1.5 \times 10^{-5}$  mol of ammonia (as determined from Nessler's reagent) was produced by the nitrogen fixation, the total nitrogen in the catalyst system before fixation was  $5.84 \times 10^{-5}$  mol and the total nitrogen in the catalyst after the nitrogen fixation and distillation of the ammonia from the catalyst system is  $5.90 \times 10^{-5}$  mol (this is equivalent to 7 ml of base in the titration). If our error in titration is 0.05 ml, this could result in an error of  $0.04 \times 10^{-5}$  mol in the nitrogen determination. We have also qualitatively detected hydrazine in the system after the nitrogen fixation, and we are now carrying out quantitative determinations of the hydrazine present. Several other types of blanks were carried out; all these runs yielded either no or a low blank amount of ammonia as determined by Nessler's reagent. Since the level of ammonia production is so low, we are planning a <sup>15</sup>N<sub>2</sub> experiment that will definitely prove the origin of the ammonia.

(12) Some other cobalt porphyrins were found to be catalytically active with respect to acetylene reduction; the cobaloxime system, on the other hand, did not show any activity.

(13) The nitrogen fixation has now been shown to take place in less than 12 hr if the temperature is elevated to 70°; the ammonia has also been produced in a closed system without the necessity of continually passing N<sub>2</sub> through the solution. We have also confirmed the presence of hydrazine in the nitrogen experiments.

(14) For discussion of the homogeneous catalytic reduction and hydration of acetylene, see the following: R. F. Heck, *Accounts Chem. Res.*, **2**, 11 (1969); J. P. Candlin, K. A. Taylor, and D. T. Thompson in "Reactions of Transition Metal Complexes," Elsevier, Amsterdam, 1968, pp 238-250, 334-347.

ylene reduction does take place quite efficiently when a molybdenum porphyrin is used.<sup>15,16</sup>

(15) E. B. Fleischer and T. S. Srivastava, *Inorg. Chim. Acta*, **5**, 151 (1971).

(16) This work was supported by the NIH and NSF.

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### Cleavage of Silicon-Silicon and Germanium-Germanium Bonds with 1,2-Dibromoethane by a Free-Radical Mechanism. Evidence for Bimolecular Homolytic Substitution at Silicon and Germanium<sup>1</sup>

Sir:

Recent stereochemical, kinetic, and esr studies have shown that bimolecular homolytic substitution (SH<sub>2</sub>) reactions may take place at a multivalent metallic center of the group II-V elements.<sup>2</sup> However, no



example of this process for organosilicon and organogermanium compounds has been reported to date in spite of the fact that such compounds are very common. This is probably due to their possessing stronger element-carbon bonds than those of other organometallic compounds. For example, while benzylstannanes undergo extensive Sn-C bond cleavage,<sup>2h</sup> SH<sub>2</sub> reactions will take place only at benzylic hydrogen<sup>3</sup> when the corresponding benzylsilanes are treated with *N*-bromosuccinimide. We thought that the silicon-silicon bond, like oxygen-oxygen<sup>4</sup> and sulfur-sulfur<sup>5</sup> bonds, could be susceptible to the SH<sub>2</sub> reaction since the bond energy of Si-Si is considerably lower than that of Si-C.<sup>6</sup>

After several unsuccessful attempts,<sup>7,8</sup> we have now obtained clear-cut evidence for the SH<sub>2</sub> reaction at silicon and germanium in solution. The reaction stems from chain reactions involving attack of a bromine atom generated from 1,2-dibromoethane on an organodisilane to give a bromosilane and a silyl radical.

(1) Silyl Radicals. IX. For part VIII, see: H. Sakurai and A. Hosomi, *J. Amer. Chem. Soc.*, **93**, 1709 (1971).

(2) (a) A. G. Davies and B. P. Roberts, *J. Chem. Soc. B*, 17 (1967); (b) *ibid.*, 1074 (1968); (c) *ibid.*, 311 (1969); (d) *ibid.*, 317 (1969); (e) *Chem. Commun.*, 699 (1969); (f) *J. Organometal. Chem.*, **19**, P17 (1969); (g) A. G. Davies and S. C. W. Hook, *J. Chem. Soc. B*, 735 (1970); (h) A. G. Davies, B. P. Roberts, and J. M. Smith, *Chem. Commun.*, 557 (1970); (i) A. G. Davies, D. Griller, B. P. Roberts, and R. Tudor, *ibid.*, 640 (1970); (j) A. G. Davies, S. C. W. Hook, and B. P. Roberts, *J. Organometal. Chem.*, **22**, C37 (1970); (k) W. G. Benitude and R. A. Wielesek, *J. Amer. Chem. Soc.*, **91**, 2406 (1969); (l) K. U. Ingold, *Chem. Commun.*, 911 (1969); (m) P. G. Allies and P. B. Brindly, *J. Chem. Soc. B*, 1126 (1969); (n) P. J. Krusic and J. K. Kochi, *J. Amer. Chem. Soc.*, **91**, 3942 (1969); (o) *ibid.*, **91**, 3944 (1969); (p) H. C. Brown and G. W. Kabalka, *ibid.*, **92**, 744 (1970); (q) M. M. Midland and H. C. Brown, *ibid.*, **93**, 1506 (1971); (r) A. Suzuki, S. Nozawa, M. Harada, M. Itoh, H. C. Brown, and M. M. Midland, *ibid.*, **93**, 1508 (1971).

(3) (a) A. G. Brook, *ibid.*, **79**, 4373 (1957); (b) A. G. Brook, M. A. Quigley, G. J. D. Peddle, N. V. Schwartz, and C. M. Warner, *ibid.*, **82**, 5102 (1960).

(4) K. Nozaki and P. D. Bartlett, *ibid.*, **68**, 1686 (1946).

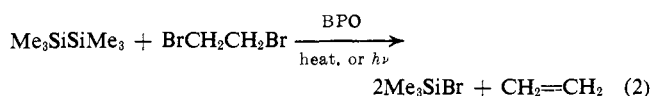
(5) (a) W. A. Pryor, *Tetrahedron Lett.*, 1201 (1963); (b) W. A. Pryor and H. Guard, *J. Amer. Chem. Soc.*, **86**, 1150 (1964).

(6) E. A. V. Ebsworth in "Organometallic Compounds of the Group IV Elements," Vol. 1, Part 1, A. G. MacDiarmid, Ed., Marcel Dekker, New York, N. Y., 1968, p. 1.

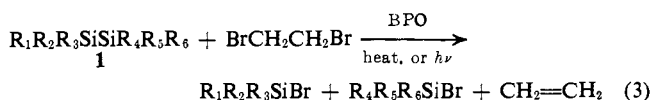
(7) H. Sakurai, A. Hosomi, J. Nakajima, and M. Kumada, *Bull. Chem. Soc. Jap.*, **39**, 2263 (1966).

(8) H. Sakurai, T. Kishida, A. Hosomi, and M. Kumada, *J. Organometal. Chem.*, **8**, 65 (1967).

In a typical experiment, a mixture of 3.64 mmol of hexamethyldisilane (**1a**) and 6.35 mmol of 1,2-dibromoethane was irradiated for 0.5 hr by a 450-W high-pressure mercury arc lamp at room temperature in the presence of a catalytic amount (0.1 mmol) of dibenzoyl peroxide (BPO). The course of the reaction was characterized by the evolution of almost the theoretical amount (70.3 ml at normal temperature and pressure, 96.6%) of ethylene. Examination of the mixture by vpc and nmr revealed that 6.51 mmol (100%) of trimethylbromosilane was the sole product, along with small amounts of unchanged **1a** (0.39 mmol) and 1,2-dibromoethane (3.10 mmol). A similar result was obtained by a thermally induced reaction. Thus, 16.01 mmol (100%) of trimethylbromosilane and 7.99 mmol (99.8%) of ethylene together with 2.02 mmol of unchanged **1a** and 2.10 mmol of 1,2-dibromoethane were obtained from a mixture of **1a** (10.01 mmol) and 1,2-dibromoethane (10.11 mmol) by heating at 90° for 45 min in the presence of 0.088 mmol of BPO.

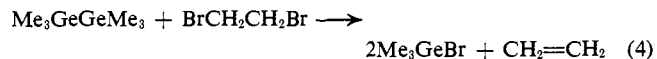


A number of organodisilanes were then examined. All disilanes examined gave the corresponding bromosilanes quantitatively.



- 1a**, R<sub>1</sub> = R<sub>2</sub> = R<sub>3</sub> = R<sub>4</sub> = R<sub>5</sub> = R<sub>6</sub> = CH<sub>3</sub>  
**b**, R<sub>1</sub> = H; R<sub>2</sub> = R<sub>3</sub> = R<sub>4</sub> = R<sub>5</sub> = R<sub>6</sub> = CH<sub>3</sub>  
**c**, R<sub>1</sub> = R<sub>4</sub> = H; R<sub>2</sub> = R<sub>3</sub> = R<sub>5</sub> = R<sub>6</sub> = CH<sub>3</sub>  
**d**, R<sub>1</sub> = Ph; R<sub>2</sub> = R<sub>3</sub> = R<sub>4</sub> = R<sub>5</sub> = R<sub>6</sub> = CH<sub>3</sub>  
**e**, R<sub>1</sub> = R<sub>2</sub> = Ph; R<sub>3</sub> = R<sub>4</sub> = R<sub>5</sub> = R<sub>6</sub> = CH<sub>3</sub>  
**f**, R<sub>1</sub> = R<sub>4</sub> = Ph; R<sub>2</sub> = R<sub>3</sub> = R<sub>5</sub> = R<sub>6</sub> = CH<sub>3</sub>  
**g**, R<sub>1</sub> = R<sub>2</sub> = R<sub>3</sub> = Ph; R<sub>4</sub> = R<sub>5</sub> = R<sub>6</sub> = CH<sub>3</sub>  
**h**, R<sub>1</sub> = R<sub>2</sub> = R<sub>4</sub> = R<sub>5</sub> = Ph; R<sub>3</sub> = R<sub>6</sub> = CH<sub>3</sub>  
**i**, R<sub>1</sub> = PhCH<sub>2</sub>; R<sub>2</sub> = R<sub>3</sub> = R<sub>4</sub> = R<sub>5</sub> = R<sub>6</sub> = CH<sub>3</sub>  
**j**, R<sub>1</sub> = *c*-C<sub>3</sub>H<sub>5</sub>; R<sub>2</sub> = R<sub>3</sub> = R<sub>4</sub> = R<sub>5</sub> = R<sub>6</sub> = CH<sub>3</sub>  
**k**, R<sub>1</sub> = R<sub>4</sub> = *c*-C<sub>3</sub>H<sub>5</sub>; R<sub>2</sub> = R<sub>3</sub> = R<sub>5</sub> = R<sub>6</sub> = CH<sub>3</sub>

Hexamethyldigermane also reacts with 1,2-dibromoethane in the presence of BPO at 90°, or at room temperature under irradiation, to give a quantitative yield of trimethylbromogermane and ethylene.



Many bonds of group IVb elements to the transition metals are cleaved by halogens, halogen acids, and a variety of organic halides including 1,2-dibromoethane.<sup>9</sup> Complexes having germanium bonded to Cu, Ag, Au, Pd, or Pt all react quantitatively with 1,2-dibromoethane,<sup>10</sup> the mechanism being believed to proceed through a multicenter transition state.<sup>11</sup> Group IVb mercurials of the type (R<sub>3</sub>M)<sub>2</sub>Hg (R = Me, M = Si;<sup>12</sup> R = Ph, M = Si;<sup>13</sup> R = Me, M = Ge;<sup>14</sup> and R = Ph, M = Ge<sup>15</sup>), antimony derivatives, (Ph<sub>3</sub>Si)<sub>3</sub>Sb<sup>16a</sup> and

(9) E. H. Brooks and R. J. Cross, *Organometal. Chem. Rev., Sect. A*, **6**, 227 (1970).

(10) F. Glockling, *Quart. Rev., Chem. Soc.*, **20**, 45 (1966).

(11) E. H. Brooks and F. Glockling, *J. Chem. Soc. A*, 1241 (1966).

(12) S. W. Bennett, C. Eaborn, R. A. Jackson, and R. W. Walsingham, *J. Organometal. Chem.*, **27**, 195 (1971).

(13) N. S. Vyazankin, O. A. Kruglaya, G. S. Kalinina, and E. N. Gladyshev, *Zh. Obshch. Khim.*, **38**, 1544 (1968).

(14) N. S. Vyazankin, G. A. Razuvaev, S. P. Korneva, O. A. Kruglaya, and R. F. Galilulina, *Dokl. Akad. Nauk SSSR*, **158**, 1002 (1964).

(15) N. S. Vyazankin, V. T. Bychkov, O. V. Linzina, and G. A. Razuvaev, *Zh. Obshch. Khim.*, **39**, 950 (1969).