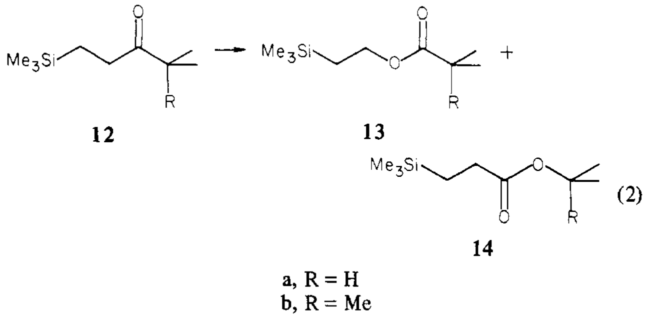
The formation of lactone **9**, although unexpected, is easily explained in terms of acid-catalyzed ring opening of lactone **6b** with 1,2-migration of Me_3Si and ring closure to the more stable **9**. 1,2-Shifts of Me_3Si groups to cationic centers are known in simple systems,[28] but their stereochemistry has not been investigated. The fact that both cis and trans olefinic acids were obtained in high purity from lactone **9** indicates that the silicon migration observed here was highly stereospecific.

The rearrangement of lactone **6b** to lactone **9** occurs under very mild conditions. A mixture of **6b** and **9** (approximately 3:2 by NMR) in CCl_4 solution (with $CHCl_3$ internal standard) was almost completely transformed into lactone **9** after 9 days at room temperature. The lability of lactone **6b** may in part be due to the fact that in a preferred conformation of **6b** (by analysis of molecular models) the C–Si and C–O bonds are in the anti conformation favored for acid-catalyzed β elimination. In lactone **9**, the Me_3Si group is not conformationally constrained, and the anti conformation, although easily achieved, should not be particularly favored. It should be noted that this rearrangement transfers the stereochemistry easily established in a cyclic system to a system having silicon attached to a side chain, which otherwise would not be easily prepared with defined stereochemistry.

In order to test the magnitude of the directive effect of silicon in the Baeyer–Villiger reactions of β-silyl ketones, we have studied the Baeyer–Villiger reactions of silyl ketones **12a** and **12b**.[29] Ketone **12a** gave a 2:1 ratio of esters **13a**[8] and **14a**[8] in 80% yield, while ketone **12b** gave a 1:2 ratio of esters **13b**[8] and **14b**[8] in 79% yield (eq 2).[31] Thus, in these compounds, the migratory aptitude of $Me_3SiCH_2CH_2$– is intermediate between that of secondary and tertiary alkyl groups.[32]

This work demonstrates that silicon can direct the Baeyer–Villiger reactions of β-silyl ketones[33] and that the products resulting from cyclic β-silyl ketones can be transformed into olefinic acids and esters. We expect these reactions will have a number of applications in organic synthesis, for example, for insect pheromones and for the olefinic acid side chain in many prostaglandins.



12 **13** **14** (2)

a, R = H
b, R = Me

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(22) The product obtained from lactone **9** was identified as 5-ketoheptanoic acid by comparison of its IR and NMR spectra with those of an independently prepared sample of 5-ketoheptanoic acid[23] and by its mp (50–51 °C) and the mp of its semicarbazone (179–180 °C, not depressed when mixed with an independently prepared sample[23]). The product obtained from lactone **6b** contaminated with lactone **9** (from the buffered Baeyer–Villiger reaction of **5b**) was identified as a mixture of 5- and 6-ketoheptanoic acids (with some olefinic acid) by comparison of its IR and NMR spectra and VPC retention times with those of independently prepared samples of 5-[23] and 6-ketoheptanoic[25] acids.

(23) A comparison sample of 5-ketoheptanoic acid was prepared from cyclopentanone by (1) treatment with EtMgBr, (2) dehydration of the product with oxalic acid, and (3) oxidation with $KMnO_4$. The final product had mp 50–51 °C (lit.[24] mp 51–52 °C); semicarbazone mp 180–181 °C (lit.[24] mp 186–188 °C).

(24) S. G. Levine, *J. Am. Chem. Soc.*, **82**, 2556–2559 (1960).

(25) A comparison sample of 6-ketoheptanoic acid was prepared from 2-methylcyclohexanone by (1) Baeyer–Villiger oxidation with MCPBA, (2) saponification with $KOH/MeOH/H_2O$, and (3) oxidation with CrO_3/pyridine. The final product had mp 31–32 °C (lit.[26] mp 33–34 °C); semicarbazone mp 142–143 °C (lit.[27] mp 144–145 °C).

(26) R. M. Acheson, *J. Chem. Soc.*, 4232–4237 (1956).

(27) G. Foster and W. J. Hickinbottom, *J. Chem. Soc.*, 215–218 (1960).

(28) M. A. Cook, C. Eaborn, and D. R. M. Walton, *J. Organomet. Chem.*, **24**, 301–306 (1970); A. J. Bourne and A. W. P. Jarvie, *ibid.*, **24**, 335–340 (1970); see also ref 2a, p 188.

(29) Ketones **12a**[8a,30] and **12b**[8] were prepared from the cyclohexylamine imines of methyl isopropyl ketone and pinacolone, respectively, by reaction with *i*-Pr_2NLi, Me_3SiCH_2Cl, and acetic acid (hydrolysis) (cf. the preparation of ketone **1**).

(30) D. N. Andreev and E. V. Kukharskaya, *Izv. Akad. Nauk SSSR, Ser. Khim.*, 1397-1398 (1958); *Chem. Abstr.*, **53**, 6992 (1959); see also V. Bažant, V. Chvalovský, and J. Rathouský, "Organosilicon Compounds", Academic Press, New York, 1965, Volume 2, Part 1, p 347.

(31) The Baeyer–Villiger reaction of **12b** with MCPBA at room temperature was quite sluggish and was more conveniently carried out at reflux (in CH_2Cl_2) in the presence of 4,4′-thiobis(6-*tert*-butyl-3-methylphenol) to minimize decomposition of the peracid [cf. Y. Kishi, M. Aratani, H. Tanino, T. Fukuyama, T. Goto, S. Inoue, S. Sugiura, and H. Kakoi, *J. Chem. Soc., Chem. Commun.*, 64–65 (1972)]. We thank Dr. R. H. Ellison for bringing this reference to our attention and for a sample of this compound.

(32) Baeyer–Villiger reactions of methyl isopropyl ketone and pinacolone under similar conditions gave isopropyl acetate and *tert*-butyl acetate, respectively, as expected.

(33) The possibility that a β-trimethylsilyl group might accelerate the Baeyer–Villiger reaction is also of interest. Rate acceleration might be expected if rearrangement were rate determining. [This might be the case for some ketones under some conditions—see ref 4b, and B. W. Palmer and A. Fry, *J. Am. Chem. Soc.*, **92**, 2580–2581 (1970).] In our work, the Me_3Si group appears to have little effect on the rate of reaction. The reactivities of silyl ketones **1** and **5b** were quite similar to that of 2-methylcyclohexanone, and the reactivity of silyl ketone **12a** was quite similar to that of methyl isopropyl ketone. Silyl ketone **5a** was slightly more reactive than cyclohexanone (for example, after 10 min at room temperature, 65% and 18% reacted, respectively), and silyl ketone **12b** was somewhat less reactive than pinacolone.

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Competitive Condensation and Proton-Transfer Processes in the Reaction of $t\text{-}C_4H_9^+$ Ions with Ammonia in Gaseous Systems at Atmospheric Pressure

Sir:

The gas-phase reaction of alkyl cations with ammonia and amines has been recently developed into a general mechanistic model for a wide class of ion–polar molecule interactions, as a result of detailed mass spectrometric and theoretical studies.[1–5]

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In particular, attention has been focused on the reaction of *tert*-butyl ions with ammonia, characterized by two competitive, exothermic channels (eq 1 and 2; $\Delta H_1° = -8.3$ kcal mol^{-1}, $\Delta H_2° = -38.3$ kcal mol^{-1}). The main object has been to ascertain, in

$$t\text{-}C_4H_9^+ + NH_3 \rightarrow i\text{-}C_4H_8 + NH_4^+ \qquad (1)$$

$$t\text{-}C_4H_9^+ + NH_3 \rightarrow [(C_4H_{12}N)^+]^* \xrightarrow[-M^*]{+M} C_4H_{12}N^+ \qquad (2)$$

the first place, the nature of the adduct from eq 2 and, secondly, to establish whether the proton-transfer and condensation products arise from the unimolecular fragmentation and collisional stabilization, respectively, of a common, excited intermediate corresponding to the σ-bonded ion $(CH_3)_3CNH_3^+$. In a recent mass spectrometric study,[5] Meot-Ner has investigated the nature of the condensation product from eq 2 by a careful proton affinity (PA) bracketing technique and has suggested a protonated *tert*-butylamine structure. Mass spectrometric studies on related reactions, e.g., between *sec*-$C_3H_7^+$ and NH_3, seem to exclude the possibility that proton-transfer and condensation processes proceed via competitive fragmentation and collisional deactivaton of an excited alkylammonium ion, but no such evidence has been adduced for reactions 1 and 2. A crucial mechanistic criterion is based on the measurement of the product ratio $[C_4H_{12}N^+]/[NH_4^+]$ that should increase linearly with pressure if a common intermediate is involved, since reaction 1 is bimolecular and reaction 2 termolecular. Unfortunately, the intensity ratio of the products, $C_4H_{12}N^+$ and NH_4^+, fails to level off in the very restricted pressure range (below 1.5 torr) accessible to mass spectrometry, and previous results have been inconclusive, owing to large experimental errors.[5]

The tentative nature of structural assignments based on m/e or PA values is not particularly vexing in this case, as the protonated *tert*-butylamine structure of the ionic product from eq 2 appears quite reasonable. However, the pressure range available to mass spectrometry is clearly too restricted in the specific case to provide a meaningful mechanistic discrimination, as shown by the failure of the $C_4H_{12}N^+$ and NH_4^+ intensity ratios to level off below 1.5 torr.[5]

A useful route to gaseous t-$C_4H_9^+$ ions in a much wider pressure range is provided by a technique based on the radiolysis of neopentane, introduced by Ausloos[6-8] and subsequently applied in gas-phase ionic chemistry.[9] The technique has been actually exploited in the study of eq 1, but not of eq 2, although the formation of *tert*-butylamine had been specifically postulated[7,10] in the irradiation of neopentane–ammonia mixtures. This neglect can probably be traced to the analytical difficulties encountered in the determination of minute traces of the highly basic and polar product t-$C_4H_9NH_2$ due to adsorption onto the walls of the irradiation vessel and to troublesome "memory" effects.

The first goal of the present study has been therefore to develop a reliable analytical procedure. After several unsuccessful attempts, it was found that preliminary treatment of Pyrex vessels with gaseous $(CH_3)_3N$, followed by thorough outgassing under vacuum, prevented irreversible adsorption of *tert*-butylamine. This product could be recovered and analyzed by condensing the gaseous contents of the irradiated vessels at −196 °C, followed by rinsing the walls with aqueous ammonia containing a suitable internal standard (e.g., 2-butanol). Aliquots of the solution were subsequently injected into a 2 m long, 2-mm i.d. glass column packed with Carbowax 20 M (4%) and KOH (0.8%) on Carbopack GB. The column, whose operation is totally unaffected by the presence of traces of water, gives a clean separation of the amines of interest.[11]

Control experiments carried out on synthetic mixtures containing a few μg/L *tert*-butylamine in neopentane resulted in recoveries exceeding 90% with the absence of "memory" effects. Once the analytical prcedure had been established, a large number of gaseous systems, containing neopentane as the bulk constituent (650–720 torr), ammonia (10–80 torr), a radical scavenger (O_2; 5–10 torr) and, in a few samples, trimethylamine (3 torr), was irradiated at 25 °C in a 220 Gammacell (Nuclear Canada Ltd.) to a dose of 2.4 Mrad. Blank analyses, carried out both on unirradiated gaseous mixtures of the above composition and on irradiated mixtures containing no ammonia, excluded the presence of detectable amounts of *tert*-butylamine.

The results of the radiolytic experiments can be summarized as follows: (i) *tert*-Butylamine is formed in all irradiated systems containing NH_3. (ii) The ratio of the yields of *tert*-butylamine and 2-butene, expressed as a mean value of a large number of irradiations, was found to be 0.040 ± 0.015. The ratio is independent of the presence of $(CH_3)_3N$, and of the NH_3 pressure.

The actual isolation of the amine provides compelling evidence for the protonated *tert*-butylamine structure of the condensation adduct from reaction 2. Furthermore, the insensitivity of the yield ratio to the presence of $(CH_3)_3N$, which has a PA of 222 kcal mol^{-1} (compared to 218.8 of *tert*-butylamine and 202.3 for ammonia[12]), shows that deprotonation of the *tert*-butylammonium ions is fast, even in the absence of a base stronger than t-$C_4H_9NH_2$. This is a possible consequence of a cooperative (clustering) process involving more than one NH_3 molecule.

Also relevant to the purposes of the investigation is the k_2/k_1 ratio, deduced from the relative yields of 2-butene and *tert*-butylamine,[13] i.e., 0.020 at 760 torr. Comparison with the rough estimate from the mass spectrometric experiments, i.e., $k_2/k_1 \sim 0.025$ in a pressure region ($\sim$1 torr) where the ratio failed to show a significant leveling off, shows that competition between reactions 1 and 2 is largely unaffected by an almost thousandfold increase of the third body density.

The result is of mechanistic significance, since it positively excludes the possibility that the proton-transfer and condensation products arise respectively from the unimolecular fragmentation and the collisional stabilization of a common, excited precursor and shows that, on the contrary, the branching ratio depends on some intrinsic feature (geometrical constraints, charge distribution, formation of proton-bound adducts, etc.), which is largely unaffected by the reaction environment.

As a whole, the results of the present study confirm the structure assigned to the condensation adducts from eq 2 on the grounds of indirect mass spectrometric evidence and theoretical considerations. Further, these results are consistent with the mechanistic model suggested for other alkyl cations.

Finally, the formation of *tert*-butylamine in the irradiation of neopentane–ammonia mixtures, postulated by Ausloos and Lias,[7] is confirmed, even though the yields are much lower than anticipated by these authors.

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(11) The analysis has been carried out by using a Sigma 1 gas chromatograph (Perkin-Elmer Co.) equipped with a FID unit. At 85 °C, at a flow rate of 20 mL min^{-1}, the column ensured a clean separation of the relevant amines, as shown by injection of authentic samples of $(CH_3)_3N$, i-$C_3H_7NH_2$, n-$C_3H_7NH_2$, $(C_2H_5)_2NH$, *sec*-$C_4H_9NH_2$, i-$C_4H_9NH_2$, and n-$C_4H_9NH_2$. In particular, secondary and tertiary amines such as i-$C_3H_7(NH)CH_3$ and $(CH_3)_2C_2H_5N$ have appreciably shorter retention times than t-$C_4H_9NH_2$. The identity of the product has been unequivocally confirmed by using, in addition to the Carbopack column, a 10 m long, 3-mm i.d. Pennwalt 231 column from Applied Sciences Laboratories Inc., especially suitable for the amines separation, connected to a selective, nitrogen-sensitive detector. The column was operated at 70 °C, at a flow rate of 25 mL min^{-1}.

(12) Kebarle, P. *Annu. Rev. Phys. Chem.* **1977**, *28*, 445–476.

(13) The ratio of the yields has been corrected for the small amount of 2-butene that is known to arise from nonionic processes; cf. ref 7 and 10. From the mass spectrometric data of ref 2 and 5, the rate constant k_2 can be estimated to be $\sim 8 \times 10^7$ cm^3 molecule^{-1} s^{-1}.

Sparapani for skilful experimental help.

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Visible Light Driven Generation of Chlorine and Bromine. Photooxidation of Chloride and Bromide in Aqueous Solution at Illuminated n-Type Semiconducting Molybdenum Diselenide and Molybdenum Disulfide Electrodes

Sir:

We report the sustained, visible light driven oxidation of Br^- and Cl^- according to eq 1 and 2 in aqueous solutions with the use of n-type semiconducting MoY_2 (Y = S, Se) photoanode-based

$$2Cl^-(aq) \xrightarrow{-2e^-} Cl_2(g)\ [E^\circ(Cl_2/Cl^-) = +1.11 \text{ V vs. SCE}] \quad (1)$$

$$2Br^-(aq) \xrightarrow{-2e^-} Br_2(aq)\ [E^\circ(Br_2/Br^-) = +0.86 \text{ V vs. SCE}] \quad (2)$$

cells. On the basis of results from previous studies, the oxidizing power of the photogenerated holes at the MoY_2/liquid interface is great enough [more positive than $E^\circ(Cl_2/Cl^-)$] to effect Cl_2 or Br_2 generation in aqueous solution.[1-3] However, in aqueous solutions, photoanodic corrosion of the electrodes is the dominant process in the presence of Br^- or Cl^-,[1-3] while photooxidation of I^- to I_3^- has been shown to be quite efficient at MoY_2 photoanodes in aqueous media.[3,4] This investigation of the oxidation of aqueous Cl^- and Br^- was prompted by the findings in this laboratory that Cl^-, Br^-, or I^- could be photooxidized in nonaqueous (CH_3CN) solution without deterioration of the MoY_2 photoanode.[5-7] We thus sought conditions where the oxidation of Cl^- or Br^- could be achieved in aqueous solutions, starting with the notion that the solvent (H_2O) could play both the role of (i) kinetic competitor with X^- for the photogenerated oxidizing equivalents and (ii) partially determining the energetics for the photocorrosion process.[8-10]

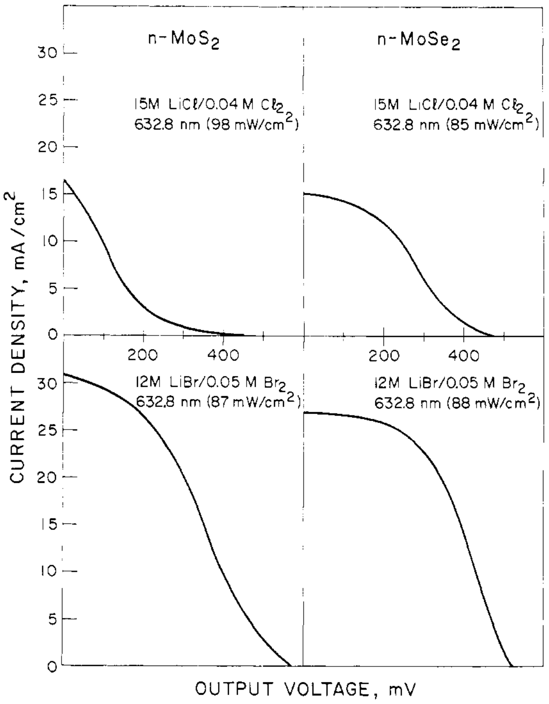

Figure 1. Steady-state photocurrent–voltage curves for n-type MoS_2 (left frames) and n-type $MoSe_2$ (right frames) photoanode-based cells. Illumination was at 632.8 nm under the conditions indicated.

Our strategy for oxidation of Cl^- or Br^- in aqueous solution has been to employ super high concentrations of these ions by using aqueous electrolyte solutions of LiCl (15 M) or LiBr (12 M). The rationale is twofold: (i) the effective activity of X^- can be much higher than the concentration (the Cl^- activity of 15 M LiCl is ~480 M and the Br^- activity of 12 M LiBr is ~560 M[11]) to provide the kinetic advantage for productive photooxidation of X^-, and (ii) the activity of H_2O can be reduced significantly by the high ionic strength[11] to reduce the efficiency for photocorrosion.

Visible light irradiation of the single-crystal, n-type MoY_2 anode of an electrochemical cell employing aqueous 15 M LiCl or 12 M LiBr (pH ≈6) results in the anodic processes represented by eq 1 or 2, respectively.[12] Light of higher energy than the band gap, ~1.1 eV, is effective.[13,14] Quantitative measurements have been made by using 514.5- or 632.8-nm light from an Ar ion or He–Ne laser, respectively. The cathode process is either the reduction of X_2 to $2X^-$ and/or the reduction of H_2O to form H_2, depending on the amount of X_2 present.

Two facts are significant. First, the photoanodes are durable when X_2 is being generated. Second, photooxidation occurs at MoY_2 potentials up to ~0.5 V more negative than $E^\circ(X_2/X^-)$, showing that light can contribute up to ~0.5 V toward the energy needed to produce X_2. Steady-state photocurrent–voltage curves for MY_2 photoanodes in X_2/LiX aqueous solutions are given in Figure 1. Under these conditions, the dark Pt cathode process is X_2 to $2X^-$ reduction, and there is no net chemical change in the cell. Data from such curves are summarized in Table I. When the MoY_2 potential is more negative than the electrochemical

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(12) Electrodes were prepared by using materials and procedures previously described.[5-7] LiCl and LiBr solutions were prepared by adding distilled H_2O to the solid to achieve a concentration of 15 M LiCl and 12 M LiBr. Small amounts of insoluble materials were filtered to achieve optical clarity. Solutions for steady-state photocurrent–voltage curves were prepared by adding Cl_2 or Br_2 to the LiCl or LiBr, respectively, and were stoppered to prevent escape of the X_2 from the electrochemical cell. All data are for 298 K. Samples of photogenerated gas (Cl_2) or authentic Cl_2 were added to a 9 mM $IrCl(CO)(PPh_3)_2$ (from Pressure Chemical Co.) $CHCl_3$ solution under Ar via a gas-tight Hamilton syringe with Teflon fittings. Infrared spectra of $IrCl(CO)(PPh_3)_2$ and $IrCl_3(CO)(PPh_3)_2$ were obtained by using a Perkin-Elmer Model 180 spectrometer. Br_2 was monitored spectrophotometrically with a Cary 17 UV–vis–near-IR spectrophotometer.

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