perature and then cooling it in an ice bath to give **8b**: 78% yield; mp 88–90 °C; ^{1}H NMR ($CDCl_3$) δ 1.25 (3 H, t), 1.4 (9 H, s), 4.25 (2 H, s), 4.35 (1 H, d, $J \approx 5$ Hz), 5.0–5.2 (4 H, m, overlapping NH, CH, CH_2Ph), 7.42 (5 H, s); IR (neat oil before recrystallization) 1790, 1745–1750, 1725 cm^{-1}; mass spectrum (FD), *m/e* 336 (M^+ – 28, loss of ethylene from the ethyl ester by McLafferty rearrangement), 308 (M^+ – 56, loss of isobutylene from the Boc group); $[\alpha]_D$ +46° (*c* 0.49, EtAc).

***N*-(Benzyloxy)-DL-*trans*-2-azetidinone (12).** The hydroxamate **11** (250 mg, 0.679 mmol) was cyclized with DEAD/TPP in the usual manner. After 3 h, the solvent was evaporated, and the residue was chromatographed on silica gel with CH_2Cl_2–IPA (99.5:0.5). The solvent was evaporated from the β-lactam-containing fractions to provide 160 mg (67%) of **12**. Recrystallization from ethyl acetate–hexanes gave the analytical sample: mp 93.5–95 °C; ^{1}H NMR ($CDCl_3$) δ 1.4 (9 H, s), 3.75 (3 H, s), 4.23 (1 H, d, J = 2.5 Hz), 4.43 (1 H, br d), 5.05 (2 H, s), 5.65 (NH), 7.4 (m, 5 H); IR (neat oil before recrystallization) 1800, 1755, 1715 cm^{-1}. Anal. Calcd for $C_{17}H_{22}N_2O_6$: C, 58.28; H, 6.33; N, 8.00. Found: C, 58.11; H, 6.03; N, 7.87.

***N*-Hydroxy-2-azetidinone (9a).** Compound **8a** (62.7 mg, 0.18 mmol) was dissolved in 10 mL of CH_3OH and 13 mg of 5% Pd/C was added. H_2 gas was bubbled through the solution for 1 h. The catalyst was removed by filtration and the filtrate evaporated to give 46.5 mg (100% yield) of **9a**. Recrystallization from ethyl acetate–hexanes gave the analytical sample: mp 125–126 °C; ^{1}H NMR ($CDCl_3$) δ 1.43 (9 H, s), 4.13 (3 H, s), 4.63–4.73 (1 H, d, J = 4 Hz), 5.0–5.3 (2 H, br), 6.3–6.7 (1 H, br); IR (KBr) 3365 (NOH), 1785, 1730, 1705 cm^{-1}; $[\alpha]^{20}_D$ +46° (*c* 1.5, ethyl acetate). Anal. Calcd for $C_{10}H_{16}N_2O_6$: C, 46.15; H, 6.15; N, 10.78. Found: C, 46.18; H, 6.30; N, 10.60.

3-[(*tert*-Butoxycarbonyl)amino]-4-(methoxycarbonyl)-2-azetidinone (1a). The *N*-(benzyloxy)-2-azetidinone (**8a**; 67.6 mg, 0.19 mmol) was dissolved in 10 mL of absolute CH_3OH and hydrogenated over 5% Pd/C as above for 40 min. The catalyst was removed by filtration and washed with 5 mL more of CH_3OH. The combined methanolic solutions of the *N*-hydroxy β-lactam **9a** were added to a solution of $TiCl_3$ (MCB, 20% aqueous, 0.8 mL, 1 mmol) and $NaHCO_3$ (0.28 g, 3.4 mmol) in 10 mL of water adjusted to pH 6.5 with 10% aqueous Na_2CO_3. After being stirred for 1 h under argon, the solution was extracted with three 50-mL portions of ethyl acetate. The combined extracts were quickly washed with ice cold 10% Na_2CO_3 (20 mL), ice cold 0.1 N HCl (20 mL), and brine (25 mL). After the mixture was dried over $MgSO_4$, evaporation gave 46.5 mg of the crude product. TLC (silica; ethyl acetate–hexanes, 1:1) revealed two components with R_f 0.75 and 0.1, respectively. Chromatography (Chromatotron, 1-mm silica plate with ethyl acetate–hexanes, 1:1) separated the unidentified nonpolar (R_f 0.75) component [7.4 mg; ^{1}H NMR δ 1.5 (9 H, s), 3.7 (3 H, s), 4.7 (1 H, s), 5.23 (0.5 H, s) and 5.37 (0.5 H, s), 6.5–7.0 (2 H, br), 7.2–8.0 (4 H, br m); IR (CCl_4) 1750, 1710, 164 cm^{-1}] and the desired β-lactam **1a** (28.5 mg, 61%). Crystallization from ethyl acetate–hexanes gave the solid: mp 112–114.5 °C; ^{1}H NMR δ 1.43 (9 H, s), 3.8 (3 H, s), 4.40–4.47 (1 H, d, J = 5.5 Hz), 5.1–5.6 (2 H, br m), 6.5–6.6 (1 H, br); IR (KBr) 1780, 1730 (shoulder), 1720, 1705 (shoulder); mass spectrum (FD), *m/e* 244 (M^+), 245 (M + 1); $[\alpha]^{20}_D$ +86° ±10% (*c* 0.275, ethyl acetate).

Acknowledgment. We gratefully acknowledge support from the National Institutes of Health (Grant GM 26867), Eli Lilly and Co., and Alfred P. Sloan Foundation (M.J.M.) D. Schifferl and K. Peterson provided instrumental assistance. The 300-MHz NMR system used was provided by funds from the NIH and the University of Notre Dame. The FD mass spectra were performed at Eli Lilly and Co.

Registry No. **1a**, 86832-64-6; **2**, 17015-08-6; **3**, 7298-98-8; **4a**, 20790-73-2; **4b**, 86767-40-0; **5a**, 86767-41-1; **5b**, 86767-42-2; **6a**, 86767-43-3; **6b**, 86767-44-4; **8a**, 86767-45-5; **8b**, 86767-46-6; **9a**, 86767-47-7; **10**, 4294-45-5; **10** monomethyl β-ester, 84035-03-0; **10** monomethyl β-ester/*N*-*tert*-butoxycarbonyl derivative, 86767-48-8; **11**, 86832-65-7; **12**, 86832-66-8; diethyl (–)-*trans*-epoxysuccinate, 74243-85-9; *O*-benzylhydroxylamine hydrochloride, 2687-43-6; *O*-benzylhydroxylamine, 622-33-3.

Influence of Steric Effects upon the Rate Constants for Competing $B_{AC}2$ and E1cB Ester Hydrolyses

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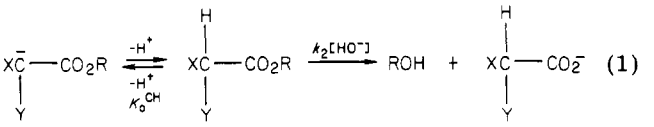
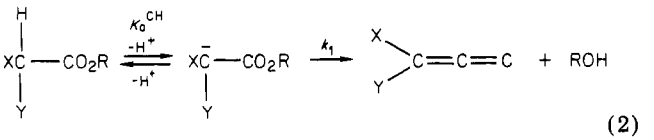
The rates of hydrolysis of carboxylic acid esters which possess an α-ionizable proton (pK_a^{CH}) are proportional to hydroxide concentration ($k_{obsd} = k'[HO^-]$) when pH ≪ pK_a^{CH} and independent of pH ($k_{obsd} = k_{pl}$) when pH ≫pK_a^{CH}. There are two mechanisms which account for this pH dependence. For the $B_{AC}2$ mechanism (eq 1) k'





= k_2 and $k_{pl} = k_2K_w/K_a^{CH}$ while for the E1cB mechanism (eq 2) $k' = k_1K_a^{CH}/K_w$ and $k_{pl} = k_1$.[1] Because the two mechanisms are kinetically equivalent they cannot be distinguished by use of their pH–rate profiles (but see footnote 2). Many criteria have been proposed to distinguish between the mechanisms of eq 1 and 2. These include the use of (i) dependence of log k' or log k_{pl} on proton basicity of the leaving group,[3] (ii) trapping of intermediate ketene,[3] (iii) deuterium isotope effects,[3,4] (iv) curvature in buffer plots,[5] and (v) steric effects.[6,7] As the departure of the leaving group from the carbon atom adjacent to the carbanion in the E1cB reaction is a dissociative process, one might expect that a sterically bulky group in the molecule would enhance the rate of the departure step.[8] However, it has been concluded that expulsion of the leaving group in E1cB alkene-forming reactions is "remarkably insensitive" to substituent effects.[9,10]

(1) (a) Holmquist, B.; Bruice, T. C. *J. Am. Chem. Soc.* **1969**, *91*, 2993, 3003. (b) Another mechanism, nucleophilic attack of water on the ester moiety of the substrate carbanion, is also kinetically equivalent to the two mechanisms mentioned in the text. However, because of the strong electron-donating effects of the carbanion lone pair, nucleophilic attack of water on the ester moiety of the carbanion ($k_{anion}^{H_2O}$) must be much slower than nucleophilic attack of water on the ester moiety of the unionized ester ($k_o^{H_2O}$). Therefore, the possibility that $k_{pl} = k_{anion}^{H_2O}$ is ruled out, as discussed in ref 1 and 3.

(2) For the E1cB hydrolysis of *p*-nitrophenyl acetoacetate, for example, a pH–rate profile with two plateau regions was observed. This pH–rate profile cannot be explained by a $B_{AC}2$ mechanism but can be explained on the basis of an E1cB mechanism with a change from preequilibrium to irreversible in carbanion formation.[3]

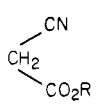
(3) Pratt, R. F.; Bruice, T. C. *J. Am. Chem. Soc.* **1970**, *92*, 5956.
(4) Tobias, P. S.; Kezdy, F. J. *J. Am. Chem. Soc.* **1969**, *91*, 5171.
(5) Kirby, A. J.; Lloyd, G. J. *J. Chem. Soc., Perkin Trans. 2* **1976**, 1762. Thea, S.; Williams, A. *J. Chem. Soc., Chem. Commun.* **1979**, 715.
(6) Remers, W. A.; Roth, R. H.; Weiss, M. J. *J. Org. Chem.* **1965**, *30*, 2910. Thea, S.; Cuanti, G.; Williams, A. *J. Chem. Soc., Chem. Commun.* **1981**, 535.
(7) Douglas, K. T. *Prog. Bioorg. Chem.* **1976**, *4*, 216.
(8) For example: Bartlett, P. D.; Tidwell, T. T. *J. Am. Chem. Soc.* **1968**, *90*, 4421. Note, however, solvolytic substitution is a charge- (carbenium ion) forming reaction while E1cB ester hydrolysis is a charge-(carbanion) consuming reaction.
(9) Redman, R. P.; Thomas, P. J.; Stirling, C. J. M. *J. Chem. Soc., Perkin Trans. 2* **1978**, 1135.

0022-3263/83/1948-3559$01.50/0

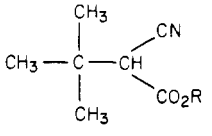
Table I. Spectral Data

compd	NMR (in $CDCl_3$), ppm	IR (liquid film), cm^{-1}
2b	1.23 (9 H, s), 3.40 (1 H, s), 4.57 (2 H, q, $J(CH_2-CF_3)$ = 82 Hz)	2580 (m), 1770 (s), 1180 (s)
2c	1.33 (9 H, s), 3.50 (1 H, s), around 7.2 (5 H, phenyl)	2240 (m), 1760 (s), 1185 (s), 1130 (s)
2d	1.32 (9 H, s), 3.50 (1 H, s), around 7.2 (4 H, phenyl)	2230 (m), 1750 (s), 1190 (s), 1125 (s)
2e	1.34 (9 H, s), 3.58 (1 H, s), ~7.5 (2 H, m), 8.0 (2 H, m)	2250 (m), 1770 (s), 1525 (s), 1350 (s), 1200 (s), 1130 (s), 732 (s)
2f	1.3 (9 H, s), 3.55 (1 H, s), 7.3 and 2.8 (4 H, AA'BB' coupling)	2220 (m), 1765 (s), 1200 (s), 1120 (s)
2g	1.33 (9 H, s), 3.51 (1 H, s), 7.3 and 8.2 (4 H, AA'BB' coupling)	2550 (m), 1760 (s), 1525 (s), 1345 (s), 1200 (s), 1125 (s)

No systematic study of steric effects on the rate of E1cB ester hydrolysis has been reported. We report herein a comparison of the kinetics of hydrolysis of 2-cyano-3,3-dimethylbutanoate esters (**2**) and cyanoacetate esters



1



2

R: **a**, Et; **b**, CF_3CH_2; **c**, Ph; **d**, *m*-ClC_6H_4; **e**, *m*-$O_2NC_6H_4$; **f**, *p*-NC-C_6H_4; **g**, *p*-$O_2NC_6H_4$

(**1**).[1,11] We show here that steric hindrance brought about by *α-tert*-butylation of cyanoacetate esters results in an increase in the pH-independent rates (observed at basic pH) for ester hydrolysis by both $B_{AC}2$ and E1cB mechanisms. In the case of the $B_{AC}2$ mechanism this is shown to be due to an *increase* in the pK_a for α-proton dissociation. For the E1cB mechanism the increase in rate is due to release of ground-state strain at the transition state.

Experimental Section

Ethyl 2-cyano-3,3-dimethylbutanoate (**2a**) was prepared from ethyl 2-cyano-3-methylbutanoate and methylmagnesium iodide.[5,12] The esters **2b–g** were prepared from 2-cyano-3,3-dimethylbutanoic acid (prepared by the alkaline hydrolysis of **2a**) and 2,2,2-trifluoroethanol or the appropriate phenol by the action of $POCl_3$ at 70–80 °C for 24 h and were purified by column chromatography (silica gel, hexane and hexane–ethyl acetate). The esters **1** were prepared from cyanoacetic acid by the same procedures. For example, for preparation of **1f**, cyanoacetic acid (56 mmol) and *p*-cyanophenol (50 mmol) were heated with $POCl_3$ (22 mmol) at 60 °C for 3 h (47% yield), while for preparation of **2f**, 2-cyano-3,3-dimethylbutanoic acid (4 mmol) and the phenol (3.5 mmol) were heated with $POCl_3$ (5 mmol) at 70–80 °C for 24 h (20% yield). Spectral data are given in Table I.

Kinetic measurements were carried out in aqueous acetonitrile (1.0% v/v) at μ = 1.0 (with KCl) and at 30 ± 0.1 °C. Buffer solutions were prepared from K_2CO_3, KH_2PO_4, 1 M KOH (carbonate free), and deionized, freshly double-distilled water. A Cary 118 spectrophotometer was employed to follow reactions by repetitive scanning. A Perkin-Elmer Lambda 3 and a Durrum-Gibson Model 13001 stopped-flow spectrophotometer were appropriately used to follow spectral changes at single wavelengths. Acetonitrile stock solutions of **1** and **2** were prepared from freshly distilled and dried acetonitrile (of spectral quality) and stored at −20 °C. Reactions carried our under other than stopped flow conditions were initiated by the addition of 30 μL of stock solution of **2** to 3 mL of aqueous buffer solution equilibrated at 30 °C. The pH of the reaction mixtures was measured in the cuvette at 30 °C by using a glass electrode after every kinetic measurement. Stopped flow experiments were carried out as follows: a mixture of 6.3 portions of a stock solution of the ester and 100 portions of 0.001 M aqueous CH_3COOH–CH_3COO^- buffer (μ = 1.0 with KCl) solution was prepared just before every experiment. This mixture and hydroxide or carbonate buffer were equilibrated at 30 °C and mixed in a ratio of 1:5.25. In this way, the reaction conditions (i.e., solvent composition, ionic strength, etc.) used in the stopped-flow experiments were kept identical with those used in ordinary spectrophotometric experiments, except for the presence of additional acetate buffer, which was necessary to prevent spontaneous hydrolysis during temperature preequilibration. The concentration of acetate buffer was more than 50 times smaller than the minimum concentration of hydroxide or carbonate used as the buffer in stopped-flow experiments, and the contribution of acetate-ion catalysis was negligible. The pHs of the reaction mixtures were measured separately.

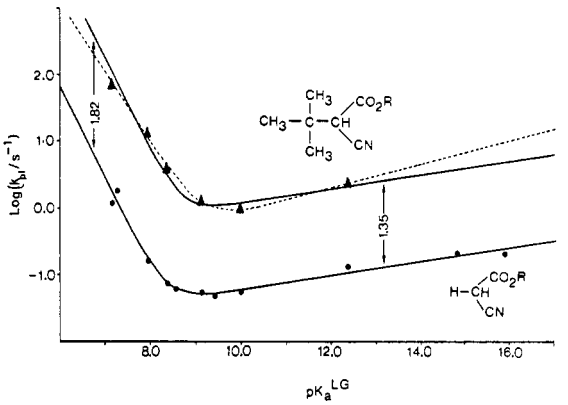


Figure 1. Plots of log k_{pl} (s^{-1}) vs. pK_a of the conjugate acid of the leaving group (pK_a^{LG}) for esters **1** and **2**. The solid line correlating the data points for esters **1** represents the best fit to $k_{pl} = 10^{-1.38pK_a^{LG}+10.09} + 10^{0.11pK_a^{LG}-2.3}$. The solid line for **2** possesses the same Brønsted slope and was generated from $k_{pl} = 10^{-1.38pK_a^{LG}+11.91} + 10^{0.11pK_a^{LG}-0.98}$. The broken line for **2** represents the best fit and was generated from $k_{pl} = 10^{-1.06pK_a^{LG}+9.42} + 10^{0.18pK_a^{LG}-1.84}$.

The values of pseudo-first-order rate constants were calculated by using a least-squares analysis in determining the slopes of plots of log $(OD_\infty - OD_0)/(OD_\infty - OD_t)$ vs. time.

All hydrolytic reactions were followed at constant pH at a total buffer concentration sufficiently larger than the concentrations of ester and were found to obey the first-order rate law. Some contribution to the rate of disappearance of ester (or carbanion derived from the ester) was made by the buffers employed. The values of the rate constants for lyate species catalysis (k_{ly}) were obtained from the intercepts of plots of the determined pseudo-first-order rate constant (k_{obsd}) vs. [total buffer] at each constant pH (eq 3). The values of the rate constants for buffer

$$k_{obsd} = k_{ly} + k_{buff}[\text{total buffer}] \quad (3)$$

catalysis (k_{buff}) were determined from the slopes of these plots. However, thus obtained k_{buff} values for **2** were small compared with buffer catalysis rates for **1**.

The wavelengths used to follow the hydrolysis rates were appropriately selected to obtain maximum optical density change (**a–d**, 260 nm; **e–f**, 275 nm; **g**, 400 nm). Identical rate constants were obtained when the optical density changes were followed at different wavelengths.

(10) Stirling's conclusion was criticized by Rappoport from the view point of electronic effects: Hoz, S.; Albeck, M.; Rappoport, Z. *Tetrahedron Lett.* **1972**, 3511.

(11) Inoue, M.; Bruice, T. C. *J. Chem. Soc., Chem. Commun.* **1981**, 884.

(12) Alexander, E. R.; McCollum, J. D.; Paul, D. E. *J. Am. Chem. Soc.* **1950**, *72*, 4791.

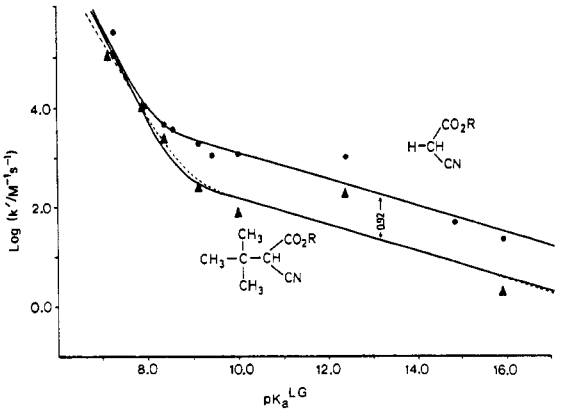
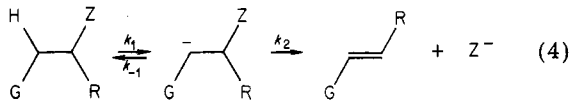


Figure 2. Plots of log k' (M^{-1} s^{-1}) vs. pK_a of the conjugate acid of the leaving group (pK_a^{LG}) for the esters 1 and 2. For ester 2, the slopes of the solid line are identical with the slopes drawn for 1 and the broken line represents the best fit.

Results and Discussion

Plots of log k_{pl} vs. pK_a of the conjugate acid of the leaving group (pK_a^{LG}) for the esters 1 and 2 are provided in Figure 1. The slopes of the plots are large and negative for the E1cb hydrolysis of the esters with good leaving groups and small and positive for the $B_{AC}2$ hydrolysis of the esters with more strongly basic leaving groups.[11,13] The values of k_{pl} for the $B_{AC}2$ hydrolysis of esters 2 are seen to be ~20 times *larger* than for esters 1. As this is an unexpected finding, we have explored the hydrolysis of 1 and 2 at pH $\ll$ pK_a^{CH}, where hydrolysis rates are proportional to the hydroxide concentration. The so-determined second-order rate constants are shown in the Brønsted-type plot of Figure 2. Inspection of Figure 2 reveals that the $B_{AC}2$ hydrolysis rate constants for esters 2 are approximately 8 times smaller than for esters 1. This finding would be anticipated on the basis of the steric bulk of the *tert*-butyl substituent α to the ester group in esters 2. Since the plateau rate of the $B_{AC}2$ reaction is given by the equation $k_{pl} = k_2K_w/K_a^{CH}$, it follows that any change in pK_a^{CH} of esters 2 compared to 1 (i.e., ΔpK_a^{CH}) is given by the equation $\Delta pK_a^{CH} = (\Delta \log k_{pl}) - (\Delta \log k_2)$, implying $\Delta pK_a^{CH} \simeq 2$. The increase in k_{pl} for the $B_{AC}2$ reaction on *tert*-butylation is then due to the accompanying increase in pK_a^{CH} by 2 units which is much larger than the magnitude of decrease in log k_2.

The rate constant for the departure step (k_1 of eq 2) of the E1cB reaction is increased by a factor of approximately 70 by *tert*-butylation of the α-carbon atom as is anticipated by the steric acceleration in dissociative reactions. This finding is in striking contrast to the deductions of Stirling et al.[9] Their conclusion was reached by examination of values of k_2/k_{-1} (eq 4) in the region pH $\ll$ pK_a^{CH} with the



(4)

assumption that k_{-1} was constant and at the diffusion-controlled limit.[14] From our observation that pK_a^{CH} is changed by 2 units on substitution of the α-hydrogen of 1 with the bulky *tert*-butyl group, it is possible that k_{-1} in eq 4 also is sensitive to steric effects of substituents and in the same manner as k_2.[15] This discussion may be supported by our observation that the second-order rate constants k' (at pH $\ll$ pK_a^{CH}) for the E1cB reaction are apparently insensitive to the steric effect.

Finally it must be noted that one should be careful in using steric effects as a criterion for distinguishing $B_{AC}2$ from E1cB mechanisms. The plateau rates of both the $B_{AC}2$ and E1cB reactions may be increased by a bulky substituent, and the pH-dependent rate can be insensitive to steric effects even though the rate of the departure step is increased in the E1cB reaction by the bulky substituent.

Acknowledgment. This work was supported by a grant from the National Institutes of Health.

Registry No. **1a**, 105-56-6; **1b**, 27827-83-4; **1c**, 6131-48-2; **1d**, 80256-96-8; **1e**, 80256-94-6; **1f**, 80256-93-5; **1g**, 80256-92-4; **2a**, 21954-81-4; **2b**, 86834-63-1; **2c**, 86834-64-2; **2d**, 86834-65-3; **2e**, 86834-66-4; **2f**, 86834-67-5; **2g**, 86834-68-6; $POCl_3$, 10025-87-3; ethyl 2-cyano-3-methylbutanoate, 3213-49-8; methylmagnesium iodide, 917-64-6; 2-cyano-3,3-dimethylbutanoic acid, 22426-28-4; cyanoacetic acid, 372-09-8; *p*-cyanophenol, 767-00-0; *p*-nitrophenol, 100-02-7.

(13) Alborz, M.; Douglas, K. T. *J. Chem. Soc., Chem. Commun.* **1980**, 728.

(14) It must be noted that for E1cB ester hydrolysis, the rate constant of departure of leaving group k_2 of eq 2 can be measured directly while for E1cB alkene formation only the rate ratio k_2/k_{-1} can be empirically determined.

(15) On the other hand, the conclusions of Stirling and co-workers may be quite valid, in which case, the difference in the sensitivities to steric effects in the departure of the leaving groups of E1cB alkene formation and ketene formation may be sought in the positions of the transition states as suggested by the standard free energies for the two elimination reactions. In the case of alkene formation (eq 3) an unstable carbanion yields a stable alkene; the transition state should be early, and the steric strain in the carbanion would be minimally *released* in the transition state. In contrast, the E1cB ester hydrolysis proceeds from a carbanion of greater stability than the ketene product and the release of strain should be fully felt in the late transition state. The observation of the insensitivity of second-order rate constants k' to the steric effect also reminds us of Williams and Douglas' early conclusion[16] that "the insensitivity of substituted azides to steric effects is consistent with the E1cB mechanism". Their conclusion was made on the basis of the data of Matier et. al.[17] for a series of azides ($RNHSO_2N_2$). As the azides have high pK_a values, only k' values (second-order rate constants) were determined.[18]

(16) Williams, A.; Douglas, K. T. *J. Chem. Soc., Perkin Trans. 2* **1974**, 1727.

(17) Matier, W. L.; Comer, W. T.; Reitchman, D. *J. Med. Chem.* **1972**, *15*, 538.

(18) See also: Casida, J. E.; Augustinsson, K. B.; Jonsson, G. *J. Econ. Entomol.* **1960**, *53*, 205.

Bis(methoxycarbonyl)sulfur Diimide, a Convenient Reagent for the Allylic Amination of Alkenes

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Despite the growing interest in this class of compounds, not many highly regio- and stereoselective procedures for the synthesis of primary 2-alkenylamines from easily accessible starting materials like alkenes are known to date. Simple examples may be laboriously prepared by allylic halogenation of an alkene and subsequent treatment with ammonia. However, the halogenation step is seldom selective enough;[1] the scope of this method is strictly limited.

Some years ago, we discovered ditosylsulfur diimide (2) to be a highly reactive enophile, converting a wide variety of alkenes (1) into *N*-(2-alkenyl)sulfinamidines under mild

(1) See for example: Stroh, R. "Methoden der organischen Chemie (Houben-Weyl)"; Georg Thieme Verlag: Stuttgart, 1962; Vol. V/3, pp 585-592, 800, 805, 806; Roedig, A. 1960, Vol. V/4, pp 221-233.

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