satisfactory spectroscopic data. Commercial alcohols, benzene, cyclohexane, and piperylenes were purified by distillation.

Authentic Samples for Identification of Reaction Products. The syntheses of the alkoxyacetates **2**–**4** and the β-hydroxybutyrate **5** have been published[31] elsewhere. α-Methoxy ketones (**12**, **15**, **19a**, and **19b**) were prepared[32] by treatment of the corresponding α-diazo ketone with boron trifluoride in methanol. Methyl *tert*-butylacetate (**14**) was prepared by esterification of the corresponding acid, obtained[33] by bromoform reaction of neopentyl methyl ketone.[34] Cycloalkylformates (**17**) were obtained from the corresponding cycloalkylformic acids.[35] Satisfactory spectroscopic data have been obtained for all authentic compounds. All other reagents were obtained from commercial sources.

Photochemical Reactions and Analyses. All irradiations were conducted by using a Halos 300-W high-pressure mercury lamp with a water-cooled quartz jacket. In the general procedure, ca. 0.05 mmol of diazo compound was added to 2 mL of solvent in a Pyrex tube of 5-mL capacity. The tube was suspended in a transparent Pyrex Dewar flask fitted with coolant and irradiated until all of the precursor molecule was destroyed. The coolants were water (24 °C), solid CO_2–EtOH (−78 °C), liquid nitrogen–isopentane (−110 and −155 °C), and liquid nitrogen (−196 °C). Control experiments ruled out the interconversion of the products during the irradiation and also showed that no reaction occurred in the absence of light. Product identification and absolute yields were determined by standard GC techniques.

Sensitized experiments were performed with a monochromatic light in the presence of 5–10 molar excess benzophenone so as to assure that >98% of the incident light was absorbed by the sensitizer.

Acknowledgment. Support of this work by the Asahi Glass Foundation is gratefully acknowledged.

Registry No. 1, 6832-16-2; **2**, 17639-73-5; **3**, 6149-45-7; **4**, 17640-21-0; **5**, 17639-74-6; **6**, 79-20-9; **9**, 4250-02-6; **10**, 103-25-3; **11**, 103-79-7; 12, 29417-89-8; 13, 6832-15-1; 14, 10250-48-3; 15, 39195-77-2; **16a**, 3242-56-6; **16b**, 18202-04-5; **17a**, 4630-80-2; **17b**, 4630-82-4; **18a**, 108-94-1; **18b**, 502-42-1; **19a**, 7429-44-9; **19b**, 1121-18-2; **20a**, 930-68-7; **20b**, 1121-66-0; **21**, 120-92-3; 2-propanol, 67-63-0; methanol, 67-56-1.

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Nonconvergent Cycloaddition Pathways for Thermal vs. Photochemical Reaction of *N*-Isobutenylpyrrolidine with Dimethyl Fumarate

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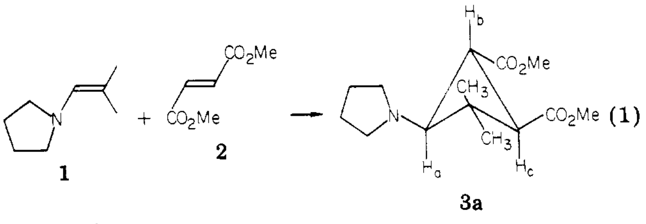
Received May 16, 1980

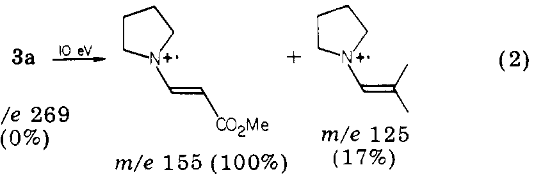
The thermal reaction of the title compounds proceeds via a reversibly formed 1,4-zwitterion intermediate to yield the thermodynamically most stable [2 + 2] cycloadduct. The rate of the thermal reaction is substantially faster in moderately polar vs. nonpolar solvent. In contrast, the photochemical reaction of the title compounds occurs only in nonpolar solvent and yields a stereoisomer of the thermal adduct. Photochemical cycloaddition is not observed for several other α,β-unsaturated esters and enamines. The mechanism of the photochemical reaction and the failure to observe photochemical ionic [2 + 2] cycloaddition are discussed.

Thermal [2 + 2] cycloaddition reactions are known to occur via concerted [$_\pi 2_s + {}_\pi 2_a$], stepwise-biradical, and stepwise-zwitterionic mechanisms. Photochemical concerted [$_\pi 2_s + {}_\pi 2_s$] and stepwise-biradical (singlet and triplet) cycloaddition reactions are also well-known; however, there is at present no bona fide example of a photochemical zwitterionic [2 + 2] cycloaddition reaction. Epiotis and Shaik[1] recently predicted the occurrence of photochemical ionic [$_\pi 2_s + {}_\pi 2_s$] cycloadditions and noted the absence of examples. In an attempt to obtain experimental evidence for ionic photochemical cycloaddition, we have investigated the photochemical reactions of several enamines with α,β-unsaturated esters. Previous investigations have established the formation of a zwitterionic intermediate in the thermal cycloaddition reaction between these classes of alkenes.[2,3] We report that the cycloaddition of *N*-isobutenylpyrrolidine (**1**) and dimethyl fumarate (**2**) can be effected either thermally or photochemically and that a zwitterionic intermediate is formed in the thermal but not in the photochemical reaction.

Results

Mixing of **1** and **2** in dry acetonitrile results in formation of a yellow charge-transfer complex (Figure 1), which slowly fades to yield a colorless solution from which the adduct **3a** can be isolated (eq 1). The structure **3a** is assigned on the basis of spectral data. Mass spectral fragmentation of **3a** yields predominantly the two possible aminum radical retro [2 + 2] fragments (eq 2). The stereochemistry of **3a** is assigned on the basis of the large





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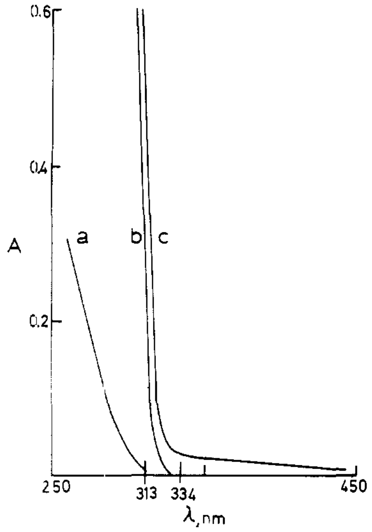


Figure 1. Absorption spectra of 0.01 M **2** (a), 0.36 M **1** (b), and 0.01 M **2** + 0.36 M **1** (c).

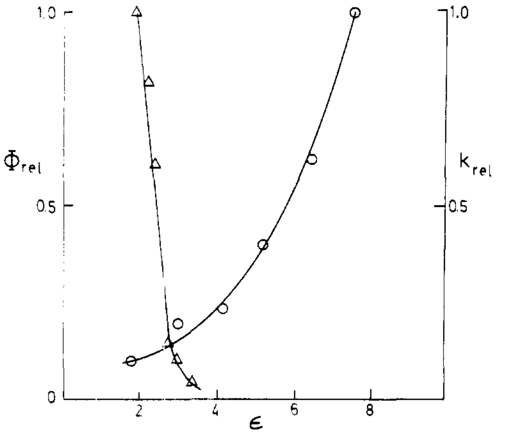


Figure 2. Solvent dependence of the relative rate of thermal cycloaddition (○) and the relative quantum yield of photochemical cycloaddition (△). Dielectric constant $\epsilon = \sum_i \epsilon_i V_i$, where V_i is the volume fraction of component i in hexane/tetrahydrofuran mixed solvent.

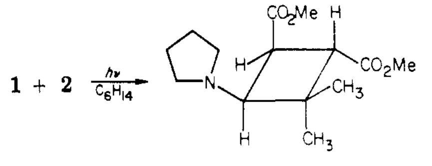
vicinal coupling constants for the cyclobutyl protons ($J_{AB} = 9.9$, $J_{BC} = 8.8$ Hz) which require a puckered cyclobutane ring with all hydrogens axial.[4,5] This conformation allows the carbomethoxy and pyrrolidine substituents to all be equatorial and thus renders **3a** the thermodynamically most stable adduct of **1** and **2**.

Adduct **3a** is also formed upon refluxing **1** and **2** in hexane for prolonged periods. The rate of adduct formation increases markedly with increasing solvent polarity as shown in Figure 2. Adduct **3a** is stable over a period of several months at room temperature in solution; however, refluxing in either hexane or acetonitrile results in cycloreversion to **1** and **2**. Addition of enamine **1** to dimethyl maleate leads to rapid and essentially quantitative isomerization of dimethyl maleate to **2** followed by slow formation of **3a**. Maleate cis–trans isomerization is catalyzed by trace impurities of pyrrolidine which can add reversibly to dimethyl maleate or fumarate.

Table I. Quantum Yields for Photochemical Cycloaddition in Hexane Solution

λ_{ex}, nm	enamine, M	dimethyl fumarate, M	$\Phi \times 10^3$
313	0.143	0.01	1.2
313	0.684	0.01	1.4
313	0.835	0.01	1.3
313	0.36	0.0065	0.84
313	0.36	0.0113	1.7
313	0.36	0.0144	2.4
313	0.36	0.0222	3.3
334	0.36	0.0226	6.4

Irradiation of a hexane solution of **1** and **2** results in the formation of an adduct which displays the same GC retention time and mass spectral fragmentation pattern as **3a**. Attempts to purify this adduct by column, thick-layer, or gas-phase chromatography led to the isolation of **3a**. However, irradiation followed by removal of solvent and most of the unreacted starting materials at room temperature yielded an adduct, the NMR spectrum of which is clearly different from that of **3a** (eq 3). Complete NMR assignments for the photoadduct proved impossible due to overlap of the cyclobutyl protons with those of residual **1**; however, structure **3b** is consistent with the available data and with the known propensity of **2** to form cycloadducts with retention of *trans*-bis(carbomethoxyl) stereochemistry in both concerted and stepwise reactions.[6,7]



(3)

3b

The efficiency of photochemical cycloaddition decreases dramatically with increasing solvent polarity (Figure 2). In solvents more polar than 20% tetrahydrofuran/hexane, irradiation causes no increase in the rate of the thermal reaction.[8] Quantum yields for the formation of **3b** in hexane solution using 313- or 334-nm light are given in Table I. The quantum yields are independent of the concentration of **1** but increase with increasing concentration of **2**. The quantum yield for 334-nm irradiation is approximately twice that for 313-nm irradiation. At the longer wavelength the charge-transfer complex is the only absorbing species, whereas at shorter wavelength the charge-transfer complex and free enamine have comparable absorbances (Figure 1). No fluorescence is observed from solutions of **1** and/or **2**.

Attempts to observe photochemical [2 + 2] cycloaddition of enamine **1** with methyl acrylate, methyl methacrylate, or dimethyl isopropylidenemalonate or of **2** with *N*-isobutenylpiperidine or *N*-isobutenyldimethylamine in hexane solution were unsuccessful.

Discussion

Both the thermal and photochemical reactions of enamine **1** and dimethyl fumarate (**2**) result in the formation of a [2 + 2] cycloadduct; however, differences in product stereochemistry and the solvent dependence of product formation clearly indicate that the two processes do not occur via a common intermediate. This result is in marked contrast to several reported stepwise-biradical [2 + 2] cycloaddition reactions in which the same products are formed thermally and photochemically.[9,10] Our data for

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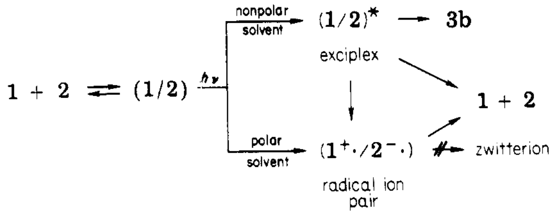
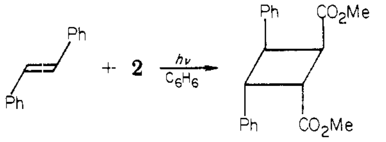
Scheme I. Photochemical Cycloaddition and Electron-Transfer Quenching



the thermal reaction are in agreement with the extensive investigations of Brannock et al.[2] and Hall and Ykman[3] on the cycloaddition reactions of enamines with electron-deficient alkenes. These investigations support the formation of a 1,4-zwitterionic intermediate in the thermal cycloaddition process. Reversible cycloaddition can account for the exclusive formation of the thermodynamically most stable adduct, **3a**.[11]

The photochemical behavior of **1** and **2** is similar to that of *trans*-stilbene with **2** (eq 4) in that cycloaddition occurs



(4)

only in nonpolar solvents and does not yield the thermodynamically most stable adduct.[6a] Excitation of either free stilbene or the stilbene–**2** charge-transfer complex leads to adduct formation via a weakly fluorescent singlet exciplex. In the case of enamine cycloaddition, the charge-transfer complex is the major photoreactive species, as evidenced by the higher quantum yield for 334- vs. 313-nm irradiation (Table I, Figure 1). Increasing the concentration of **1** (constant [**2**], [**1**] $\gg$ [**2**]) does not alter the ratio of free enamine/charge-transfer absorbance and hence leaves the quantum yield unchanged. Increasing the concentration of **2** (constant [**1**], [**1**] $\gg$ [**2**]) decreases the ratio of free enamine/charge-transfer absorbance and thus increases the quantum yield. The failure of free enamine to react with **2** is consistent with the short singlet lifetime expected for an acyclic alkene. The selective formation of a single adduct from both stilbene and **1** with **2** reflects the requirement of maximized π-orbital overlap in an exciplex intermediate.[12] Finally, the solvent dependence of the reaction of both stilbene and **1** with **2** can be attributed to solvent stabilization of a radical ion pair state which undergoes back electron transfer to regenerate starting materials but evidently does not form a 1,4-zwitterion. The greater solvent sensitivity and lower quantum yield for the reaction of **1** vs. stilbene with **2** is consistent with its lower ionization potential (7.66[13] vs. 7.94[14] eV) which should render full electron transfer exothermic in less polar solvents.

The probable mechanism for the photochemical reaction of **1** and **2** is shown in Scheme I. In polar solvents radical ion pair formation followed by back electron transfer constitutes a rapid nonradiative decay pathway, electron-transfer quenching. There is considerable current interest in the reactions of photochemically generated radical ions; however, ionic cycloaddition has not been reported. Similarly, some exceptionally strong electron-donor acceptor pairs are known to yield thermal redox products but not cycloadducts.[3] Previous failures to observe photochemical ionic cycloaddition might be due to the instability of the requisite 1,4-zwitterion or its efficient reversion to starting materials. The present investigation was designed to obviate these complications by the selection of reactants known to undergo thermal ionic cycloaddition. The absence of photochemical ionic [2 + 2] cycloaddition in this investigation raises the question of under what circumstances, if any, such a reaction might occur. A second question raised by the present investigation is why photochemical cycloaddition is not observed for enamines and α,β-unsaturated esters other than **1** and **2**. It appears likely that stronger donor–acceptor pairs may decay exclusively by electron-transfer quenching even in hexane solution.[6] Electron acceptors weaker than **2** may not form a stable enough ground-state complex with **1** to allow direct excitation.

Experimental Section

General Procedures. Ultraviolet absorption spectra were obtained on a GCA-McPherson EU-700-32 recording spectrophotometer. Mass spectra were obtained on a Hewlett-Packard 5985 GC/MS system. NMR spectra were obtained by using a Varian CFT-20 NMR spectrometer. Gas chromatograms were obtained on a Hewlett-Packard 5750 gas chromatograph, using a 6 ft × $^1/_8$ in. column of 5% SF96 on Chromosorb G (80–100 mesh).

Hexane (Aldrich, reagent grade) was stirred over H_2SO_4 three times and distilled successively under nitrogen from H_2SO_4, K_2CO_3, and P_2O_5. Dry hexane is essential because the enamine is easily hydrolyzed by trace amounts of water. Acetonitrile and tetrahydrofuran were dried immediately prior to use. *N*-Isobutenylpyrrolidine was prepared by the method of Hall and Ykman[3] and stored under dry nitrogen. Dimethyl fumarate (Aldrich) was recrystallized twice from absolute ethanol and sublimed.

Quantum yields (313 nm) were measured by irradiating samples in a merry-go-round with a Hanovia 450-W medium-pressure mercury arc in a Pyrex cooling well. Monochromatic 313-nm light was provided by using a potassium chromate filter solution. Samples were prepared in 20-mm o.d. Pyrex ampules, degassed (5 freeze-pump-thaw cycles) and sealed under vacuum. The 334-nm quantum yield was measured on an optical bench by using a Bauch and Lomb high-pressure mercury arc and high-intensity 0.25-m monochrometer. Light intensities were measured by benzophenone–benzhydrol actinometry.[15] Product formation was determined by gas chromatography at conversions of $2 \leq 6\%$.

Dimethyl *r*-1,*cis*-3-Pyrrolidino-4,4-dimethylcyclobutane-*trans*-1,2-dicarboxylate (3a). *N*-Isobutenylpyrrolidine (3.5 g) and 5.1 g of dimethyl fumarate were mixed in 40 mL of acetonitrile and refluxed under nitrogen for 18 h. The solvent was removed and the unreacted dimethyl fumarate removed by filtration. The remaining oil was distilled in a Kugelrohr apparatus at 140 °C (1.5 mm) to give 6.4 g of a mixture of **3a** and dimethyl α-pyrrolidinosuccinate (60:40 by GC). The latter was removed by fractional crystallization from hexane, leaving 3.2 g of **3a** (42%) as a clear oil: NMR (see text for discussion) ($CDCl_3$) δ 0.98 (d, $J = 1.8$ Hz, 3 H), 1.23 (s, 3 H), 1.70 (m, 4 H), 2.39 (m, 4 H), 2.75, 3.22, 2.56 (ABC, $J_{AB} = 9.9$, $J_{BC} = 8.8$ Hz, 3 H), 3.65 (s, 6 H); IR (thin film) 2960, 1739 (s), 1438 (w), 1209 (m), 1170 cm^{-1} (m); mass spectrum (10 eV), see eq 2. Anal. Calcd for $C_{14}H_{23}NO_4$: C, 62.43; H, 8.61; N, 5.20. Found: C, 62.50; H, 8.66; N, 4.98.

Dimethyl *r*-1,*trans*-3-Pyrrolidino-4,4-dimethylcyclobutane-*trans*-1,2-dicarboxylate (3b). *N*-Isobutenylpyrrolidine (2.5 g) and 2 g of dimethyl fumarate were mixed in 250 mL of hexane. The solution was degassed by bubbling with nitrogen. After irradiation for 48 h using a Hanovia 450-W medium-pressure mercury arc in a Pyrex cooling well the solvent was removed at

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room temperature under reduced pressure. The unreacted dimethyl fumarate was removed by filtration and the residue placed under vacuum for 12 h to remove the unreacted enamine, leaving 0.46 g of **3b** as yellow oil. Attempts to further purify **3b** resulted in isomerization to **3a**: NMR ($CDCl_3$) δ 0.92 (s, 3 H), 1.30 (s, 3 H), 2.7–3.2 (m, 8 H), 3.58 (s, 6 H); IR (thin film) 1737 cm^{-1}.

Acknowledgment. Support of this work by the National Science Foundation (CHE 78-01120) is gratefully acknowledged.

Registry No. **1**, 2403-57-8; **2**, 624-49-7; **3a**, 75444-57-4; **3b**, 75494-57-4; dimethyl α-pyrrolidinosuccinate, 62626-92-0.

Pyrolysis of 1-(Trimethylsilyl)-1-alkanols. New Carbene Precursors of Silylcarbene and Phenylcarbene[1]

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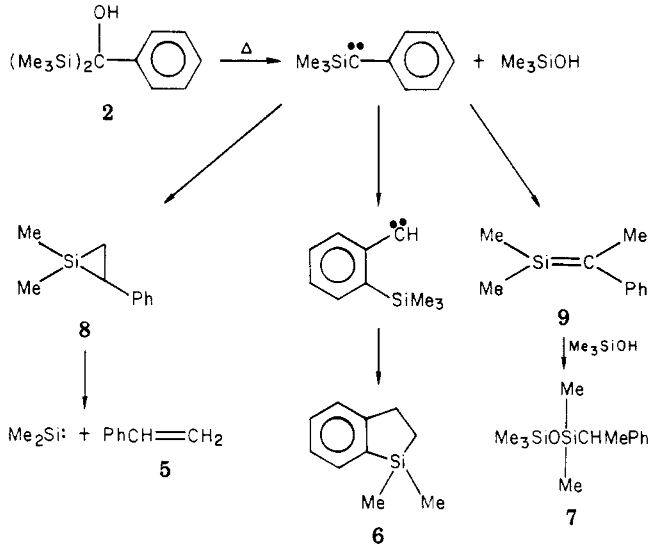
Received June 4, 1980

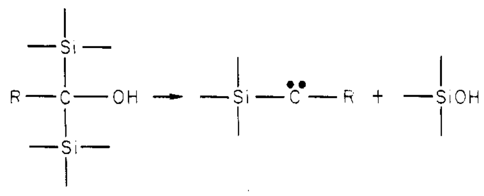
Pyrolysis of bis(trimethylsilyl)phenylmethanol gave (trimethylsilyl)phenylcarbene by the elimination of trimethylsilanol and the products were compared with those of (trimethylsilyl)phenyldiazomethane. Alkylsilylcarbenes obtained from the pyrolyses of 1,1-bis(trimethylsilyl)-1-alkanols afforded vinyltrimethylsilanes in high yields. (Trimethylsilyl)phenylmethanols were found to be excellent precursors of phenylcarbenes and their intramolecular reactions were also studied.

Introduction

In recent years, interest in the chemistry of organometallic carbenes has remarkably increased,[2] and detailed studies of silylcarbenes, (trimethylsilyl)(carboethoxy)carbene,[3-5] (trimethylsilyl)phenylcarbene,[6-9] (trimethylsilyl)carbene,[10-13] and bis(trimethylsilyl)carbene[14] have been reported. The proposed intramolecular reaction of silylcarbenes to give silicon–carbon double bonds (silenes) has also attracted interest. Silylcarbenes are generally produced by the decomposition of silyl diazo compounds, but several steps are required to synthesize silyl diazo compounds, and care must be taken to avoid the explosion of such diazo compounds. Other routes to silylcarbenes reported so far involve the decomposition of organomercury reagents such as $(Me_3SiCCl_2)_2Hg$,[15,16] base-induced α-elimination of (halomethyl)silane,[17-20] and an insertion of atomic carbon into a silicon–hydrogen bond.[21] However, some of these processes are not suitable because of several side reactions. Since silicon has a strong affinity toward oxygen, 1,1-bis(trimethylsilyl)-1-alkanols (**1**) are expected to produce silylcarbenes by the elimination of silanol.

Scheme I





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We report here that compounds **1** are very convenient precursors of silylcarbenes and also demonstrate the intramolecular reactions of (alkylsilyl)carbenes and substi-

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