Treatment of **9** with 5% aqueous NH_4OH in CH_2Cl_2 at room temperature overnight resulted in a quantitative recovery of the material.

9-Methoxy-4,4a,10b-trimethyl-1,2,3,4,4a,10b-hexahydrobenzo[*f*]quinoline (10) Hydrobromide.—**7a** (regenerated from 2.92 g of the picrate) was brominated as described previously. Evaporation of CH_2Cl_2 at room temperature gave **8a** as an amorphous powder. Ethereal methylmagnesium iodide (100 ml of 0.43 *M*) was added to a suspension of **8a** in ether (70 ml) and refluxed for 15 hr. The cooled mixture was poured into ice–water containing NH_4Cl, basified with NH_4OH, and extracted with ether. Evaporation of the dried extracts gave the residue which was chromatographed over silica gel (80 g) and eluted with chloroform–methanol (95:5). Conversion of the eluate into the hydrobromide and recrystallization from acetone–methanol–ether gave **10** hydrobromide (0.38 g, 18%): mp 254–256° dec; uv max (MeOH) 282 mμ (ε 14,500); nmr (D_2O) 1.39 (s, 3, CCH_3), 1.44 (s, 3, CCH_3), 3.05 (s, 3, N^+CH_3), 3.95 (s, 3, OCH_3), 6.24 (d, 1, J = 10 Hz, C_5H), 6.75 (d, 1, J = 10 Hz, C_6H).

Anal. Calcd for $C_{17}H_{24}NOBr$: C, 60.36; H, 7.15; N, 4.15; Br, 23.66. Found: C, 60.15; H, 7.26; N, 4.09; Br, 23.52.

Elution with chloroform–methanol (9:1) and conversion of the eluate into the hydrochloride gave **9-methoxy-4,4a,10b-trimethyl-1,2,3,4,4a,5,6,10b-octahydrobenzo[*f*]quinoline (11) hydrochloride** (0.28 g, 13%): mp 227–230° dec; needles from acetone–methanol–ether; ir 3380, 3440 cm^{-1} (hydrate H_2O); nmr (D_2O) 1.37 (s, 3, CCH_3), 1.54 (s, 3, CCH_3), 3.08 (s, 3, N^+CH_3), 4.05 (s, 3, OCH_3).

Anal. Calcd for $C_{17}H_{26}NOCl \cdot H_2O$: C, 65.15; H, 8.99; N, 4.46. Found: C, 65.41; H, 8.95; N, 4.52.

Reaction of **7a** perchlorate with ethereal methylmagnesium iodide also gave **11** in 40% yield.

Registry No.—**3a**, 28360-42-1; **3b** hydrochloride, 34887-93-9; **6a**, 34887-94-0; **6b**, 34887-95-1; **7a**, 34887-96-2; **7a** picrate, 34887-97-3; **7a** perchlorate, 34917-95-8; **7b** picrate, 34887-98-4; **7b** perchlorate, 34887-99-5; **8b**, 34887-61-1; **9** perchlorate, 34887-62-2; **10** hydrobromide, 34887-63-3; **11** hydrochloride, 34887-64-4.

Acknowledgment.—The authors express their gratitude to Director Michio Yamazaki of this laboratory and Professor Emeritus S. Sugasawa of the Tokyo University for their interest and encouragement. Thanks are also extended to the staff of the analytical section presided over by Dr. K. Kotera for elemental and spectral analyses.

Studies on Heterocyclic Compounds. XI. 1,3-Dipolar Cycloaddition of Benzimidazolium Ylide with Acetylenic Compounds

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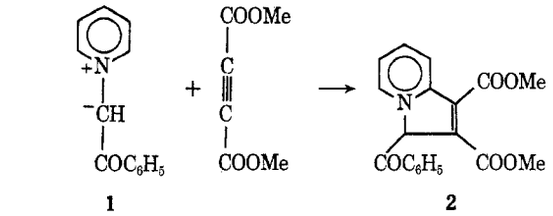
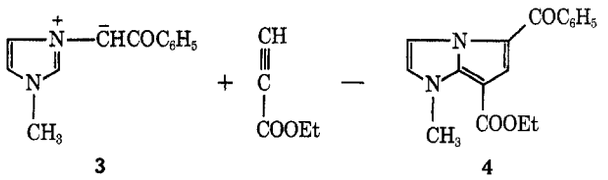
1,3-Dipolar cycloaddition of 3-substituted 1-alkylbenzimidazolium ylides with ethyl propiolate gave 3-substituted 9-alkyl-1-ethoxycarbonylpyrrolo[1,2-*a*]benzimidazoles. Reaction of 1-alkyl-3-phenacylbenzimidazolium ylides with dimethyl acetylenedicarboxylate afforded 4-alkyl-2,3-bis(methoxycarbonyl)-1-phenacylpyrrolo[1,2-*a*]-benzimidazole (**7**) and an open-chain compound (**8**). On the other hand, reaction of 1-alkyl-3-methoxycarbonylmethylbenzimidazolium ylides with dimethyl acetylenedicarboxylate gave 4-alkyl-1,2,3-tris(methoxycarbonyl)-pyrrolo[1,2-*a*]benzimidazole (**9**) and 5-alkyl-3,4-bis(methoxycarbonyl)-1-oxopyrido[1,2-*a*]benzimidazole (**10**).

For the purpose of obtaining potential physiologically active compounds, we synthesized compounds of the tricyclic azole system, such as thiazolo[3,2-*a*]benzimidazoles,[1] thiazolo[2,3-*b*]benzothiazoles,[2] imidazo[2,1-*b*]-benzothiazoles,[3] imidazo[2,1-*b*]benzoxazoles,[4] pyrimido-[1,2-*a*]benzazoles,[5] and imidazo[1,2-*a*]benzimidazoles.[6] In our previous report,[7] 9-alkylamino-2-arylimidazo-[1,2-*a*]benzimidazole showed a strong analgesic activity. We also suggested that pyrrolo[1,2-*a*]benzimidazole systems would have potential physiological activities.

Recently, Boekelheide and coworkers[8] prepared pyrrocoline (**2**) by the reaction of pyridinium ylide (**1**) and methyl acetylenedicarboxylate, and they found that 3-phenacylimidazolium ylide (**3**) reacted with ethyl propiolate to yield 4-benzoyl-6-ethoxycarbonyl-1-methyl-1,3a-diazapentalene[9] (**4**). These facts suggest



1 **2**



3 **4**

that the reaction of benzimidazolium ylide with acetylenic compounds might offer a useful synthesis for pyrrolo[1,2-*a*]benzimidazoles.[10]

Reaction of 3-substituted 1-alkylbenzimidazolium ylides, which were prepared from bromides **5**, with ethyl propiolate gave 1-substituted 4-alkyl-3-ethoxycarbonyl-4*H*-pyrrolo[1,2-*a*]benzimidazoles (**6**) (Chart I). Their structures were confirmed by the ir and nmr spectra. The chemical shift of the C-8 proton in **6a–f** is summarized in Table I, and the values show the para-

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(2) H. Ogura, T. Itoh, M. Ogiwara, and T. Okamoto, *Yakugaku Zasshi*, **89**, 469 (1969).

(3) H. Ogura and T. Itoh, *Chem. Pharm. Bull.*, **18**, 1981 (1970).

(4) H. Ogura, T. Itoh, and S. Sugimoto, *ibid.*, **18**, 2204 (1970).

(5) H. Ogura and M. Kawano, Abstracts of Papers, 91st Annual Meeting of the Pharmaceutical Society of Japan, 1971, p 669.

(6) H. Ogura and T. Itoh, *Kitasato Arch. Exp. Med.*, **42**, 65 (1969).

(7) H. Ogura, M. Kawano, K. Kikuchi, and T. Itoh, Abstracts of Papers, 3rd International Congress of Heterocyclic Chemistry, 1971, p 506.

(8) V. Boekelheide and N. A. Fedoruk, *J. Org. Chem.*, **32**, 2062 (1967).

(9) V. Boekelheide and N. A. Fedoruk, *J. Amer. Chem. Soc.*, **90**, 3830 (1968).

(10) H. Ogura, T. Itoh, K. Kikuchi, and H. Sekine, Abstracts of Papers, 91st Annual Meeting of the Pharmaceutical Society of Japan, 1971, p 670.

CHART I

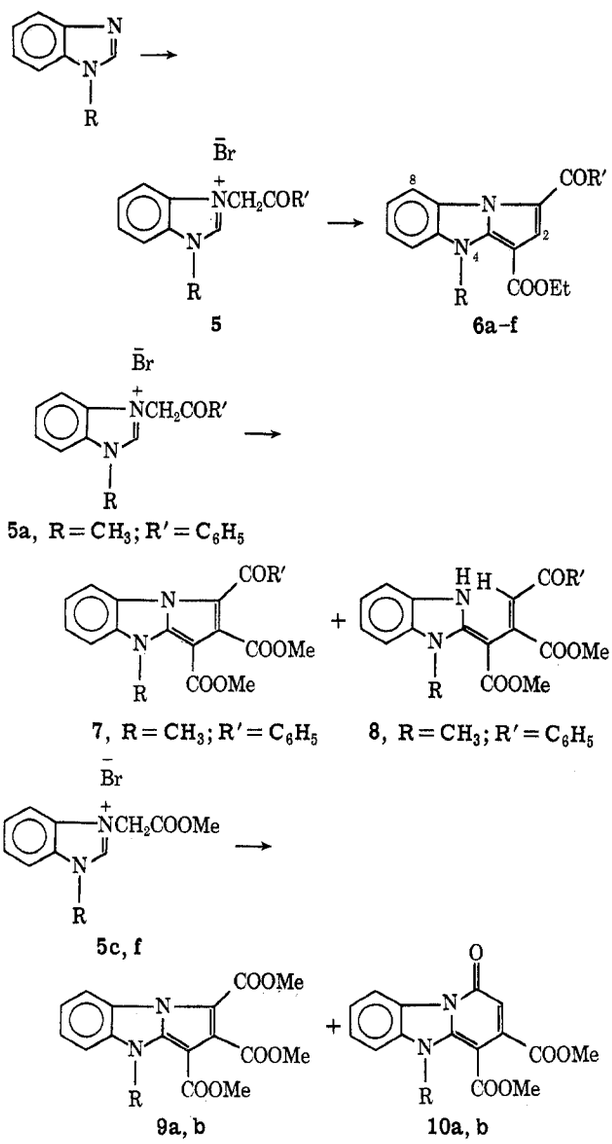


TABLE I

NMR DATA OF THE C-8 PROTON IN 6

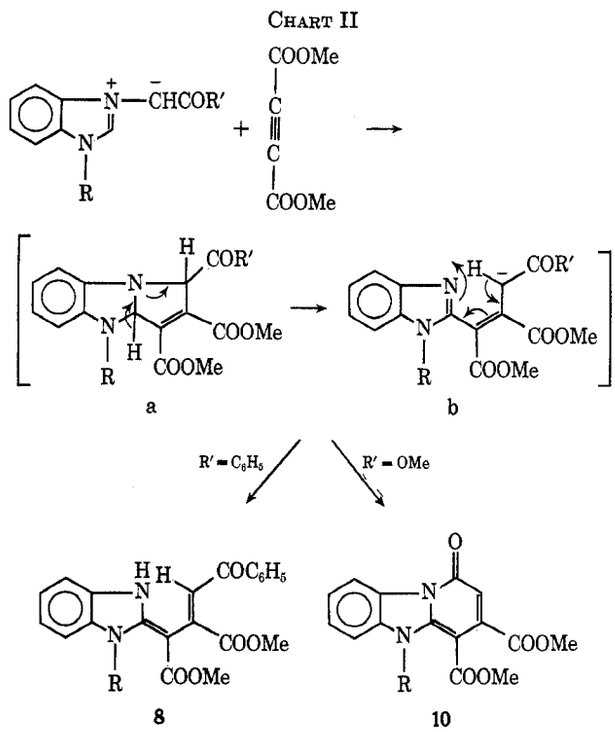
6	R	R′	δ, ppm (CDCl₃)
a	Me	C_6H_5	8.90
b	Me	$C_6H_4Br(p)$	9.80
c	Me	OMe	8.13
d	Et	C_6H_5	8.90
e	Et	$C_6H_4Br(p)$	9.10
f	Et	OMe	8.21

magnetic anisotropic effect of the carbonyl group in the C-1 position. Moreover, there is no nuclear Overhauser effect between the N^4-methyl group and the proton at the C-2 position in **6a** (R = CH_3, R′ = C_6H_5). Although there is a possibility of cyclization in another direction, the product in that case should have a nuclear Overhauser effect (*ca.* 15%), from the distance between protons in the N^4-methyl group and in the C-3 position (3).[11]

The reaction of dimethyl acetylenedicarboxylate with 1-methyl-3-phenacylbenzimidazolium ylide, prepared from the corresponding bromide (**5a**), gave a normal 1,3-dipolar cycloaddition product (**7**, 11%) and an open-chain 1-methyl-2-[1,2-bis(methoxycarbonyl)-3-benzoyl-2-propenylidene]benzimidazoline (**8a**, 6%). The reaction of 1-alkyl-3-methoxycarbonylmethylbenzimidazolium ylide (**5c,f**) and dimethyl acetylenedicarboxylate afforded 5-alkyl-3,4-bis(methoxycarbonyl)-1-oxo-1,5(2*H*)pyrido[1,2-*a*]benzimidazole (**10a,b**; both 6%) besides the normal product (**9a,b**; 7 and 2%, respectively). Structures of these compounds were determined by nmr, mass, and ir spectra. The ir spectrum of **8a** showed an NH band at 3432 cm^{-1} (10^{-4} mol in CCl_4), and the nmr spectrum of 3,4-bis(methoxycarbonyl)-5-methyl-1-oxo-1,5(2*H*)-pyrido-[1,2-*a*]benzimidazole (**10a**, R = CH_3) showed two ester methyl groups at 3.91 (s, 3 H) and 4.04 ppm (s, 3 H).

The mechanisms of these reaction may be represented by the sequence shown in Chart II. The initial 1,3-

CHART II



dipolar addition product a was oxidized by excess reagent to normal 1,3-addition products (**7**, **9**), but another possible route may proceed to a ring opening to yield the second intermediate b. In the compound **5c,f** with R′ = OCH_3, cyclization occurred in a manner similar to the reaction of 2-aminobenzazole with acetylenic compounds.[5] In the case of R′ = phenyl (**5a,b,d,e**), the second intermediate b was not cyclized. Further cyclization was not effected even on heating **8** in polyphosphoric acid. This result is similar to that of the reaction of dimethyl acetylenedicarboxylate and 1-methyl-3-imidazolium dicyanomethylide.[9]

Experimental Section

Temperatures are uncorrected. Nmr spectra were measured in $CDCl_3$ with a Varian T-60 spectrometer with Me_4Si as an internal standard. Mass spectra were measured with JEOL-01S spectrometer by a direct inlet system at 75 eV. Elemental analyses (C, H, N) of the compounds gave values corresponding to their formula within ±0.4%: Ed.

(11) R. A. Bell and J. K. Saunders, *Can. J. Chem.*, **48**, 1114 (1970).

TABLE II

1-SUBSTITUTED 3-ALKYLBENZIMIDAZOLIUM BROMIDES (5a–f)

Compd No.	R	R′	Solvent	Mp, °C form	Yield, %	Ir ν_{CO}^{KBr}, cm^{-1}
5a	CH_3	C_6H_5	Dichloromethane	205 white needles (EtOH)	78	1690
5b	CH_3	$C_6H_4Br(p)$	Me_2CO–MeOH	241 white needles (MeOH)	91	1685
5c	CH_3	OCH_3	Et_2O	109 white needles (EtOH)	88	1740
5d	C_2H_5	C_6H_5	Dichloromethane	119 white prisms (EtOH)	98	1685
5e	C_2H_5	$C_6H_4Br(p)$	Me_2CO	139 white needles (MeOH)	90	1685
5f	C_2H_5	OCH_3	Et_2O	129 white needles (EtOH)	84	1745

TABLE III

1-BENZOYL-3-ETHOXYCARBONYL-4-METHYL-4*H*-PYRROLO[1,2-*a*]BENZIMIDAZOLES (6a–f)

Compd No.	R	R′	Mp, °C	Yield, %	Ir ν_{CO}^{KBr}, cm^{-1}	Uv λ_{max}^{EtOH}, nm (log ϵ)	Mass *m/e* (M^+)
6a	CH_3	C_6H_5	164	4	1690, 1920		346
6b	CH_3	$C_6H_4Br(p)$	224	5	1685, 1610	227 (4.41) 333 (3.77)	425
6c	CH_3	OCH_3	185	5	1705, 1650	246 (4.73) 313 (4.07) 326 (4.02)	300
6d	C_2H_5	C_6H_5	155–157	4	1695, 1620		360
6e	C_2H_5	$C_6H_4Br(p)$	186	4	1680, 1610	228 (4.42) 332 (3.81)	439
6f	C_2H_5	OCH_3	160	2	1675, 1650	246 (4.63) 294 (4.18) 313 (4.10) 327 (4.17)	314

TABLE IV

1-ALKYL-2-[1,2-BIS(METHOXYCARBONYL)-3-BENZOYL-2-PROPENYLIDENE]BENZIMIDAZOLINES (8a,b,d,e)

Compd No.	R	R′	Mp, °C	Yield, %	Ir ν_{CO}^{KBr}, cm^{-1}	Uv λ_{max}^{EtOH}, nm (log ϵ)	Mass *m/e* (M^+)
8a	CH_3	C_6H_5	184 (white prisms)	6	1730, 1680	234 (4.63)	392
8b	CH_3	$C_6H_4Br(p)$	233 (white prisms)	9	1740, 1735, 1675	233 (4.59) 325 (3.87)	469
8d	C_2H_5	C_6H_5	229 (white prisms)	4	1720, 1690	235 (4.87) 332 (4.22)	406
8e	C_2H_5	$C_6H_4Br(p)$	199 (white needles)	12	1730, 1670	234 (4.60) 333 (3.89)	483

General Procedure for 1-Substituted 3-Alkylbenzimidazolium Bromides (5a–f) (Table II).—To a solution of 1-alkylbenzimidazole (0.03 mol) in a suitable organic solvent (50–100 ml), acyl bromide (0.04 mol) was added. After standing for 2–3 days at room temperature, the mixture deposited white crystals. Recrystallization from alcohol gave white needles.

Reaction of 1-Substituted 3-Alkylbenzimidazolium Bromide and Ethyl Propiolate (Table III).—To an orange-red solution of the ylide prepared from 1-substituted 3-alkylbenzimidazolium bromide (**5a–f**) (3 mmol) and K_2CO_3 (3 mmol) in dimethylformamide (40–50 ml), ethyl propiolate (6 mmol) was added at room temperature and the mixture stood for 2 days. After filtration, the organic solvent was removed under reduced pressure. Addition of EtOH gave **6a–f** as white needles after recrystallization from the same solvent.

Reaction of 3-Alkyl-1-phenacylbenzimidazolium Ylides and Dimethyl Acetylenedicarboxylate (Table IV).—To an orange-red solution of the ylide prepared from 3-alkyl-1-phenacylbenzimidazolium bromide (**5a,b,d,e**) (3 mmol) and K_2CO_3 (3 mmol) in dimethylformamide (40–50 ml), dimethyl acetylenedicarboxylate (6 mmol) was added at room temperature and the mixture was allowed to stand for 3 days. After filtration, the organic solvent was evaporated under reduced pressure. The residue was extracted with Me_2CO and the solvent was evaporated from the extract to obtain the open-chain compound **8** as white crystals after recrystallization from EtOH.

Chromatography of the Me_2CO-insoluble part of **8a** on silica gel afforded 1-benzoyl-2,3-bis(methoxycarbonyl)-4-methyl-4*H*-pyrrolo[1,2-*a*]benzimidazole (**7**) in 11% yield as white needles: mp 130–131°; ir (KBr) 1735 ($COOCH_3$), 1685 cm^{-1} (CO); uv λ_{max}^{EtOH} 234 nm (log ϵ 4.51), 330 (3.83); nmr δ 2.90 (singlet, NCH_3), 3.76, 3.83 [singlet, $(COOCH_3)_2$], 7.23 (multiplet, aromatic protons), 7.73 ppm (singlet, 5-H); mass spectrum *m/e* 390 (M^+). *Anal.* Calcd for $C_{22}H_{18}N_2O_5$: C, H, N.

Reaction of 1-Methoxycarbonylmethyl-3-methylbenzimidazolium Ylide with Dimethyl Acetylenedicarboxylate.—To a yellow

solution of the ylide prepared from 1-methoxycarbonylmethyl-3-methylbenzimidazolium bromide (**5c**) (3 mmol) and K_2CO_3 (3 mmol) in dimethylformamide (50 ml), dimethyl acetylenedicarboxylate (6 mmol) was added at room temperature and the mixture was stirred for 3 days. The black reaction mixture was filtered, and the filtrate was evaporated under reduced pressure. The residue was extracted with acetone and the extract was evaporated to obtain 5-methyl-3,4-bis(methoxycarbonyl)-1-oxo-1,5(2*H*)-pyrido[1,2-*a*]benzimidazole (**10a**) in 6% yield as white prisms: mp 255° (EtOH); ir (KBr) 1740, 1710 ($COOCH_3$), 1655 cm^{-1} (CO); uv λ_{max}^{EtOH} 243 nm (log ϵ 4.23), 314 (3.71), 328 (3.62); nmr δ 3.64 (singlet, NCH_3), 3.91, 4.04 [singlet, $(COOCH_3)_2$], 7.42 (singlet, 3-H), 7.50 (multiplet, aromatic protons), 8.21 (doublet, 6-H); mass spectrum *m/e* 314 (M^+). *Anal.* Calcd for $C_{16}H_{14}N_2O_5$: C, H, N.

Chromatography of the acetone-insoluble part of **10a** on silica gel afforded 1,2,3-tris(methoxycarbonyl)-4-methyl-4*H*-pyrrolo[1,2-*a*]benzimidazole (**9a**) in 7% yield as white prisms: mp 177–178° (EtOH); ir (KBr) 1742, 1690, 1655 cm^{-1} ($COOCH_3$); uv λ_{max}^{EtOH} 247 nm (log ϵ 4 49), 292 (4.29), 331 (4.22); nmr δ 3.86, 3.93, 4.00 [singlet, $(COOCH_3)_3$], 4.23 (singlet, NCH_3), 7.36 (multiplet, aromatic protons), 3.40 (doublet, 8-H); mass spectrum *m/e* 344 (M^+). *Anal.* Calcd for $C_{17}H_{16}N_2O_6$: C, H, N.

Reaction of 3-Ethyl-1-methoxycarbonylmethylbenzimidazolium Ylide with Dimethyl Acetylenedicarboxylate.—A similar reaction occurred with the *N*-ethyl compound. 5-Methyl-3,4-bis(methoxycarbonyl)-1-oxo-1,5(2*H*)-pyrido[1,2-*a*]benzimidazole (**10b**) was obtained in 6% yield as white leaflets (EtOH): mp 202–203°; ir (KBr) 1740, 1710 ($COOCH_3$), 1655 cm^{-1} (CO); uv λ_{max}^{EtOH} 243 nm (log ϵ 4.72), 312 (4.09), 325 (3.99); nmr δ 1.31 (triplet, NCH_2CH_3), 3.88, 4.05 [singlet, $(COOCH_3)_2$], 4.36 (quartet, NCH_2CH_3), 7.36 (singlet, 3-H), 7.50 (multiplet, aromatic protons), 8.12 (doublet, 6-H); mass spectrum *m/e* 328 (M^+). *Anal.* Calcd for $C_{17}H_{16}N_2O_5$: C, H, N.

4-Ethyl-1,2,3-tris(methoxycarbonyl)-4*H*-pyrrolo[1,2-*a*]benzimidazole (**9b**) was obtained as white prisms (2% yield): mp 134–135° (EtOH); ir (KBr) 1740, 1710, 1690 cm^{-1} ($COOCH_3$); uv λ_{max}^{EtOH} 215 nm (log ϵ 4.43), 247 (4.48), 292 (4.38), 331 (4.31); nmr δ 1.43 (triplet, NCH_2CH_3), 3.90, 3.95, 4.02 [singlet, $(COOCH_3)_3$], 4.76 (quartet, NCH_2CH_3), 7.33 (multiplet, aromatic protons), 8.40 (doublet, 8-H); mass spectrum *m/e* 358 (M^+). *Anal.* Calcd for $C_{18}H_{18}N_2O_6$: C, H, N.

Registry No.—**5a**, 34910-61-7; **5b**, 34910-62-8; **5c**, 34910-63-9; **5d**, 34910-64-0; **5e**, 34910-65-1; **5f**, 34910-66-2; **6a**, 34910-67-3; **6b**, 34934-78-6; **6c**, 34910-68-4; **6d**, 34910-69-5; **6e**, 34910-70-8; **6f**, 34910-71-9; **7**, 34910-72-0; **8a**, 34910-73-1; **8b**, 34910-74-2; **8d**, 34910-75-3; **8e**, 34910-76-4; **9a**, 14882-70-3; **9b**, 34910-78-6; **10a**, 34910-79-7; **10b**, 34915-98-5.

The Cycloaddition of Vinyl Azides to Ketenes[1]

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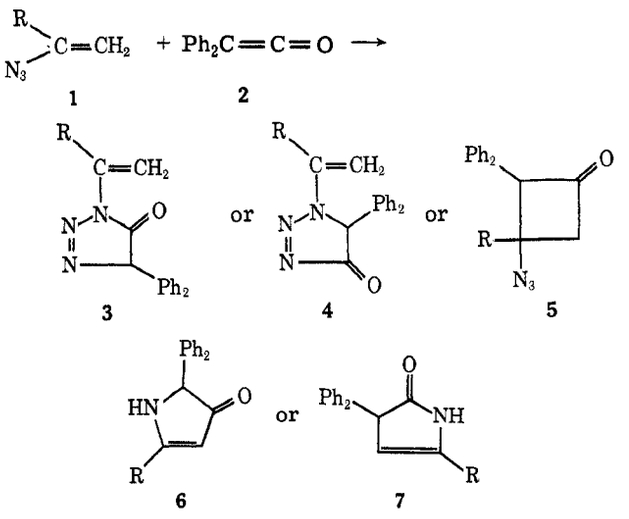
Vinyl azides undergo slow cycloaddition with diphenylketene (**2**) leading to five-membered ring enamino ketones **6** with loss of N_2. The reaction appears to involve nucleophilic attack of the β carbon from the vinyl azide upon the ketene. A substantial improvement in yield of **6** is achieved by generating **2** *in situ* from a diazo ketone in solution. Treatment of the enamino ketones with phosphorus pentachloride or chlorine leads to α chlorination. In the case of 2-azido-1-hexene, cycloaddition with **2** proceeds with formation of cyclobutanones.

The azide group represents a versatile function which can act as a nucleophile, electrophile, or 1,3 dipole.[2] An adjacent C=C, as in vinyl azides, accentuates or modifies the chemical behavior of this functional group.[3] Thus, vinyl azides exhibit a markedly greater reactivity than alkyl azides in cycloadditions with acetylenes.[4] Although the reaction of ketenes with olefins and imines has received a great deal of attention,[5] there appears to be no report of their interaction with azides.[6a] If one considers the cycloaddition of vinyl azides **1** to ketenes (*e.g.*, **2**) leading primarily to 1:1 adducts, one can envisage products of type **3**–**7** that might arise *via* a concerted or stepwise pathway.

Furthermore, in protonation and bromination of vinyl azides **1** it is difficult to distinguish whether the electrophile attacks one of the nitrogens or the β carbon of the unsaturated azide.[3] A product analysis of the reaction of **1** with ketenes offers the opportunity to establish any regiochemical preference in the cycloaddition (for instance preferential formation of **3** *vs.* **4** or **6** *vs.* **7**).



Results and Discussion

α-Azidostyrene (**1**, R = Ph) undergoes a slow reaction with diphenylketene (**2**) in ether at room temperature producing a 1:1 adduct with loss of N_2. Although the yield of this adduct was only 7% after a 3-day reaction, no other product was detected and a considerable amount of starting vinyl azide was present, together

(1) Cycloadditions. VIII. For previous paper in the series see ref 7.

(2) G. L'abbé, *Chem. Rev.*, **69**, 345 (1969).

(3) See, for instance, (a) A. Hassner, E. S. Ferdinandi, and R. J. Isbister, *J. Amer. Chem. Soc.*, **92**, 1672 (1970); (b) A. Hassner and A. B. Levy, *ibid.*, **93**, 5469 (1971).

(4) G. L'abbé, J. E. Galle, and A. Hassner, *Tetrahedron Lett.*, 303 (1970); G. L'abbé and A. Hassner, *Bull. Soc. Chim. Belg.*, **80**, 209 (1971).

(5) See, for instance, (a) W. T. Brady, *Synthesis*, 415 (1971); R. Huisgen, B. A. Davis, and M. Morikan, *Angew. Chem., Int. Ed. Engl.*, **7**, 826 (1968); (b) J. C. Martin, K. C. Brannock, R. D. Burpitt, P. G. Gott, and V. A. Hoyle, Jr., *J. Org. Chem.*, **36**, 2211 (1971).

(6) (a) Only the intramolecular decomposition of azido ketenes has been described: A. Hassner, R. J. Isbister, R. B. Greenwald, J. T. Klug, and E. C. Taylor, *Tetrahedron*, **25**, 1637 (1969). (b) A. Hassner and F. W. Fowler, *J. Amer. Chem. Soc.*, **90**, 2869 (1968).