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STUDIES OF HETEROAROMATICITY—LXIV
CHARACTERIZATION OF PYRIDINIUM N-ALLYLIDES

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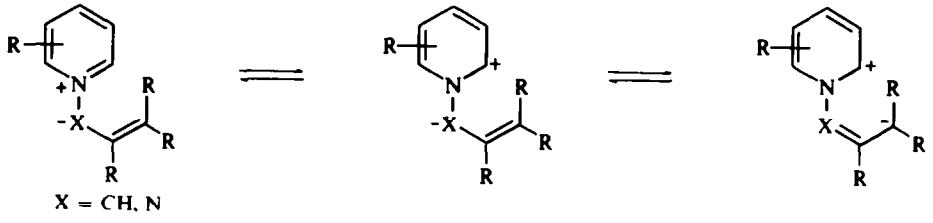
Abstract—Pyridinium N-allylides, readily obtainable from pyridinium salts **4–11** in the presence of alkaline, react together or with acetylenic compounds to give 3-ethenyl-indolidine derivatives (**12–16** respectively). N-Allylide **4** with pyridinium N-phenacylide afforded 3-benzoyl-indolidine (**20**) indicating 1,3-dipolarophilicity of such N-allylides. N-Allylide **5** with diphenylcyclopropenone gave 3,4-diphenyl salicylate (**22**). N-Allylides derived from salts **8–11** cyclized intramolecularly to give 3-unsubstituted indolidine derivatives (**23–27**). Structural elucidations were accomplished by physical and spectral means. The reactivities of pyridinium N-allylides and the formation mechanisms for the indolidine derivatives are also discussed.

PYRIDINIUM N-ALLYLIDES (X = CH), readily available by dehydrohalogenation of the corresponding pyridinium salts, are intriguing molecules since they have two powerful activating groups, an azomethine ylide and a double bond conjugated with a carbonyl group. Thus, N-allylides are not only expected to react intermolecularly at the two functional groups, but also react as 1,5-dipoles as indicated by the following resonance structures (Scheme 1).



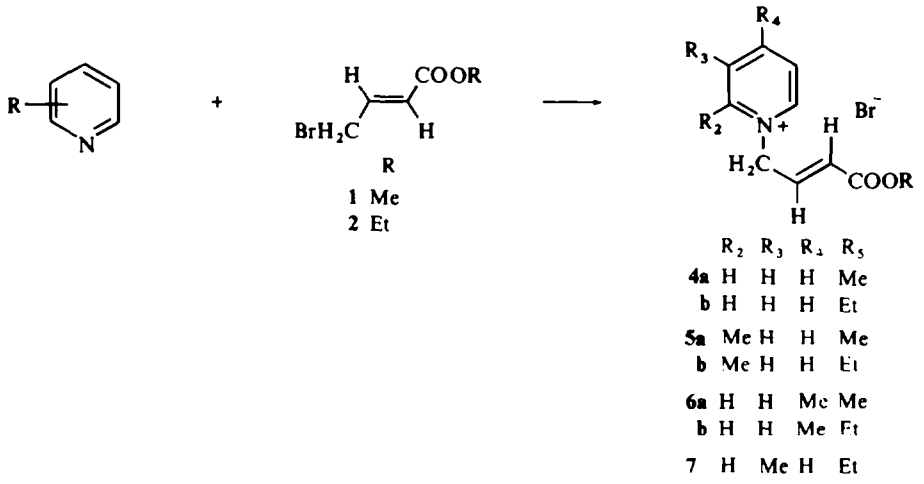
SCHEME 1.

Independently, Sasaki *et al.*[2] and Tamura *et al.*[3] have reported intramolecular reactions of N-vinyliminopyridinium ylides (X = N) and characterized them as 1,5-dipoles. In particular, we have shown that such pyridinium ylides are unstable in $CHCl_3$ and tend to cyclize intramolecularly to afford primarily dihydro-cycloadducts, which have not yet been isolated.

Here we report intermolecular reactions of pyridinium N-allylides with various reagents and also their intramolecular cyclizations.

RESULTS AND DISCUSSION

Preparations of pyridinium salts. Excess of the pyridine derivatives and γ-bromo-crotonates (**1**, **2**) were mixed in $CHCl_3$ at room temp. to afford the corresponding pyridinium salts (**4–7**) in quantitative yields (Scheme 2).



	R_2	R_3	R_4	R_5
4a	H	H	H	Me
b	H	H	H	Et
5a	Me	H	H	Me
b	Me	H	H	Et
6a	H	H	Me	Me
b	H	H	Me	Et
7	H	Me	H	Et

SCHEME 2.

Similar treatment of the pyridine and γ-bromo-β-methylcrotonate (**3**) gave hygroscopic salts (**8–11**) (Scheme 3). These compounds were used in subsequent reactions without further purification.

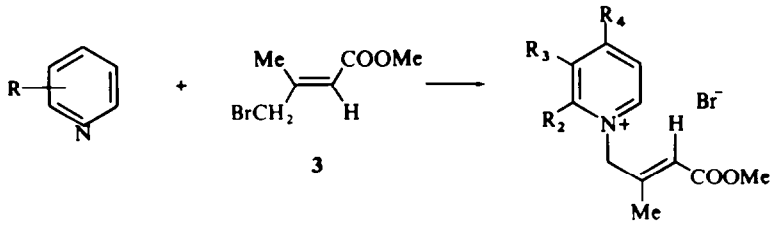
Reactions of the pyridinium salts in the presence of alkaline. Pyridinium salts (**4–7**) were treated with excess K_2CO_3 in $CHCl_3$ at room temp. (1 day) to afford the corresponding 3-ethenylindolidine derivatives,* **12–16**, respectively. In the reaction using unsymmetrically substituted pyridinium salt **7**, two isomeric compounds (**15** and **16**) were obtained in the ratio 1 :18 by NMR inspection. The results are in Scheme 4.

With a view to obtaining mechanistic information on the above reactions, the reaction of the pyridinium salt with an acetylenic compound in the presence of K_2CO_3 was investigated. Treatment of **4a** and **4b** with ethyl propiolate in the presence of K_2CO_3 afforded 1-ethoxycarbonyl-3-ethenylindolidine derivatives, **17** and **12b**, respectively (no isomeric compounds were detected (Scheme 5)).

Interestingly, reactions of salts **4a** and **4b** and pyridinium N-phenacylide (**18**) in the presence of K_2CO_3 gave 1-alkoxycarbonyl-3-benzoylindolidine compounds, **20a** and **20b**, in about 20% yields, while, in the reaction of **4a** and 4-methylpyridinium N-phenacylide (**19**), a mixture of **21a** and **12a** was obtained in 12 and 48% yields. Similar reactions of **4b** and **19** gave **21b** in 26% yield. Only starting materials were recovered in the absence of K_2CO_3. These results indicate that the double bond attached to the carbonyl group in the N-allylides behaves as a 1,3-dipolarophile to give 1-alkoxycarbonyl-3-benzoylindolidine derivatives.

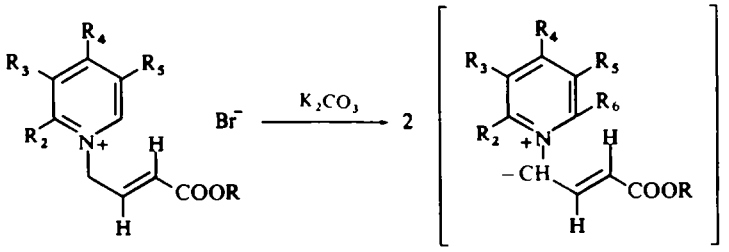
One of the most interesting reactions of the N-allylide is that with diphenylcyclopropenone (DPP), since similar reactions of N-alkoxycarbonyliminopyridinium ylides or pyridinium N-phenacylide with DPP have been described by us[4] and by Eicher *et al.*[5] In the reaction of **5a** and **5b** with DPP in the presence of K_2CO_3, the

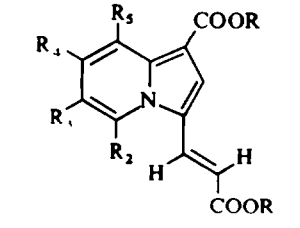
* In the course of our investigation, similar results were obtained by the reactions of the pyridinium allylides; private communication from Professor Y. Tamura of Osaka University.



	R_2	R_3	R_4
8	H	H	H
9	Me	H	H
10	H	Me	H
11	H	H	Me

SCHEME 3.



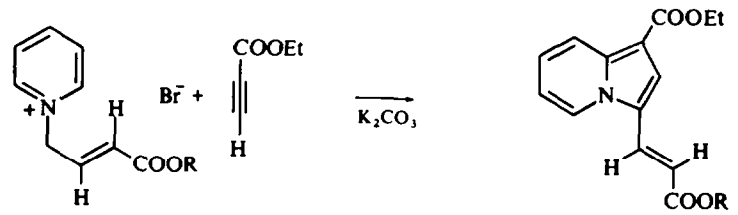


	R_2	R_3	R_4	R_5	R
12a	H	H	H	H	Me
b	H	H	H	H	Et
13a	Me	H	H	H	Me
b	Me	H	H	H	Et
14a	H	H	Me	H	Me
b	H	H	Me	H	Et
15	H	Me	H	H	Et
16	H	H	H	Me	Et

SCHEME 4

3,4-diphenyl salicylates, **22a** and **22b**, were obtained in 72 and 82% yields, respectively (Scheme 7).

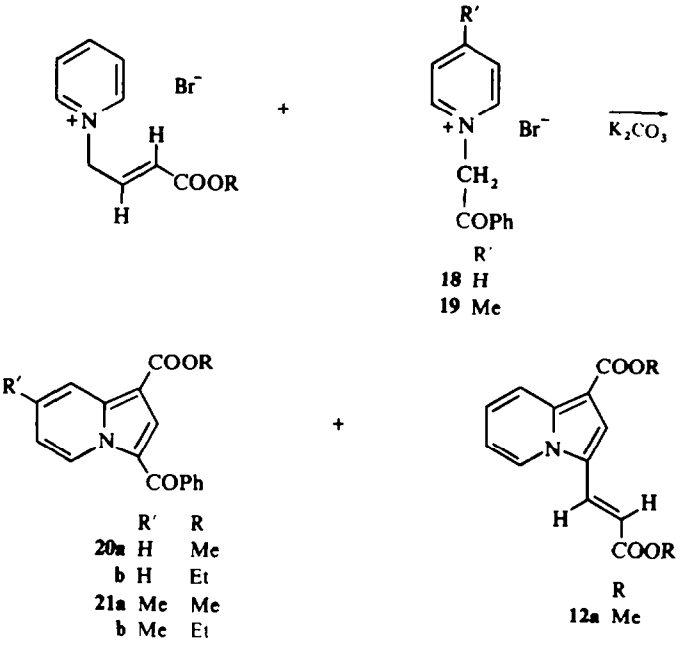
Treatment of pyridinium salts (**8–11**) with excess K_2CO_3 in $CHCl_3$ at room temp. for 1 day afforded the corresponding 3-unsubstituted indolidine derivatives (**23–27**),



R
4a Me
b Et

R
17 Me
12b Et

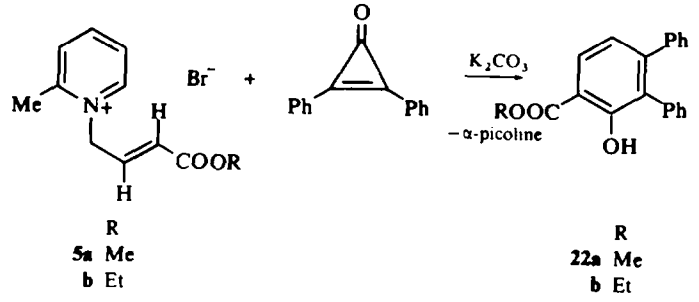
SCHEME 5.



R′ R
20a H Me
b H Et
21a Me Me
b Me Et

R
12a Me

SCHEME 6.

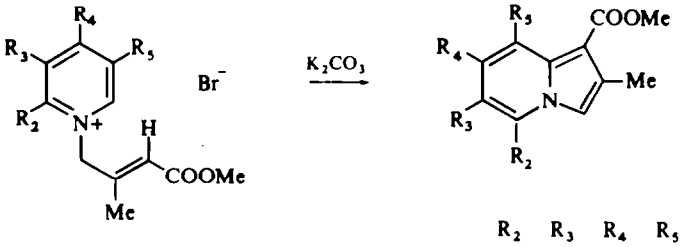


R
5a Me
b Et

R
22a Me
b Et

SCHEME 7.

but no 3-ethenylindolidines were detected. Using unsymmetrically substituted pyridinium salt (**10**), two isomeric compounds, **25** and **26**, were obtained in the ratio of 1:11 as determined by NMR; cyclization of the more sterically hindered site on the pyridine ring predominated.



	R_2	R_3	R_4	R_5
23	H	H	H	H
24	Me	H	H	H
25	H	Me	H	H
26	H	H	H	Me
27	H	H	Me	H

SCHEME 8.

Structural elucidations of the pyridinium salts, indolidine derivatives and phenol compounds. The pyridinium salts (**4–5** and **7–11**) were very hygroscopic oily compounds, only **6a** was obtained as crystals. The NMR spectrum of **6a** in $CDCl_3$ exhibited two signals at τ 2·06 and 3·02 attributable to four protons on a pyridine ring, an AB-quartet signal coupled with 15·0 Hz at τ 2·90 and 3·78, indicative of a disubstituted double bond in a *trans* configuration, and a doublet at τ 0·57 assignable to the N-substituted methylene protons.

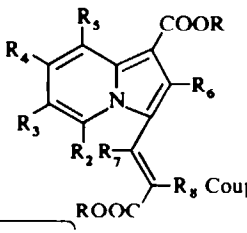
The structures of the indolidine derivatives (**12–17**, **20–21**, and **23–27**) were established by elemental and spectral analyses and independent syntheses. The elemental analyses were in good agreement with proposed structures, and the NMR spectral data of the indolidine derivatives were listed in Tables 1–3.

The NMR spectra of 3-ethenylindolidine derivatives (**12–17**) showed an AB-quartet signal with J = 15·0 Hz attributable to the *trans* olefinic protons attached to the C-3 position on the indolidine skeleton, in addition to other signals attributable to indolidine ring and ester protons at C-1 (Table 1). Furthermore, compound **12b** was identical with a 1,3-dipolar cycloadduct prepared by the reaction of **4b** with ethyl propiolate in the presence of K_2CO_3 (Scheme 5). An attempt to cyclize 3-ethenylindolidine to [3.2.2]cyclazine was unsuccessful even in refluxing toluene in the presence of Pd/C as dehydrogenating agent.

The structures of 3-benzoylindolidine derivatives (**20** and **21**) were determined by NMR comparison with an authentic sample (**20b**) prepared by 1,3-dipolar cycloaddition of pyridinium N-phenacylide with ethyl propiolate.[6]

The structures of 3-unsubstituted 1-methoxycarbonyl-2-methylindolidine derivatives (**23–27**) were also determined by NMR. Thus, the spectra of **23** in $CDCl_3$ exhibited a singlet at τ 3·12 (1H) attributable to a proton at C-3 on the indolidine skeleton (Table 2).

TABLE 1. NMR SPECTRAL DATA OF INDOLIDINE DERIVATIVES (12–17)

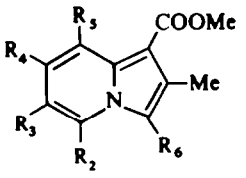


Compd	Proton Signals (τ)							R				Coupling Constants
	R_2	R_3	R_4	R_5	R_6	R_7	R_8	Me	Me	Et CH_2	Et Me	
12a	1·75 bd	3·12 bq	2·77 bq	1·78 bd	2·38 s	2·12 d	3·70 d	6·11 s	6·21 s			$J_{7,8}$ = 15·0 Hz
12b	1·75 bd	3·13 bq	2·78 bq	1·78 bd	2·37 s	2·13 d	3·68 d			5·63 q	8·58 t	
										5·75 q	8·65 t	
13a	7·13 s	3·47 d	2·97 dd	1·81 d	2·40 s	1·72 d	3·84 d	6·13 s	6·23 s			$J_{7,8}$ = 15·0 Hz, $J_{3,4}$ = 7·0 Hz
												$J_{4,5}$ = 8·0 Hz
13b	7·12 s	3·44 bd	2·99 dd	1·51–1·97†		1·51–1·97†				5·65 q	8·59 t	$J_{3,4}$ = 7·5 Hz, $J_{4,5}$ = 9·0 Hz
					2·36 s		3·82 d			5·77 q	8·67 t	$J_{7,8}$ = 15·0 Hz
14a	1·90 d	3·32 dd	7·61 s	2·04 bd	2·48 s	2·20 d	3·77 d	6·15 s	6·23 s			$J_{7,8}$ = 15·0 Hz, $J_{2,3}$ = 6·0 Hz
14b	1·88 d	3·33 bdd	7·59 s	1·99 bd	2·45 s	2·18 d	3·76 d			5·66 q	8·59 t	$J_{7,8}$ = 15·0 Hz, $J_{3,5}$ = 1·5 Hz
										5·77 q	8·66 t	$J_{2,3}$ = 7·5 Hz
16*	1·86 bd	3·23 bt	2·29 bd	7·24 s	3·14 s	2·12 d	3·67 d			5·66 q	8·59 t	$J_{7,8}$ = 15·0 Hz
										5·77 q	8·66 t	
17	1·70 bd	3·10 bt	2·76 bt	1·70 bd	2·31 s	2·06 d	3·64 d	6·19 s		5·61 q	8·59 t	$J_{7,8}$ = 15·0 Hz, $J_{2,3}$ = 7·5 Hz
												$J_{3,4}$ = 7·0 Hz, $J_{4,5}$ = 8·0 Hz

* Isomer **15** shows methyl signals (R_3) at τ 7·62 (s).

† Overlapped each other.

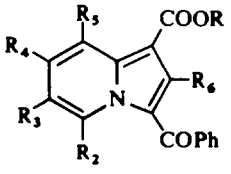
TABLE 2. NMR SPECTRAL DATA OF INDOLIDINE DERIVATIVES (23–27)



Compd	Proton Signals (τ) R_2	R_3	R_4	R_5	R_6	COOMe	Me	Coupling Constants
23	2·26 d	3·54 bt	3·18 bt	1·95 d	3·12 s	6·21 s	7·60 s	$J_{2,3}$ = 7·0 Hz, $J_{4,5}$ = 9·3 Hz
24	7·51 s	3·57 d	3·09*	1·96 d	3·09*	6·15 s	7·57 s	$J_{3,4}$ = 7·0 Hz, $J_{4,5}$ = 9·0 Hz
25†	2·25‡	7·75 s	3·09–3·40 m*		3·09–3·40 m*		7·53 s	
				1·98 bd		6·13 s		
26†	2·25 b dd	3·44 q	3·09–3·40 m*		3·09–3·40 m*		7·60 s	$J_{2,3}$ = $J_{3,4}$ = 6·8 Hz
				7·42 s		6·13 s		
27	2·36 d	3·70 dd	7·68 s	2·21 b s	3·21 s	6·24 s	7·65 s	$J_{2,3}$ = 7·0 Hz, $J_{1,3}$ = 2·0 Hz

* Overlapped each other.
† The proton signals were assigned from a mixture of **25** and **26**.
‡ Overlapped with R_2-proton of **26**.

TABLE 3. NMR SPECTRAL DATA OF INDOLIDINE DERIVATIVES (20–21)

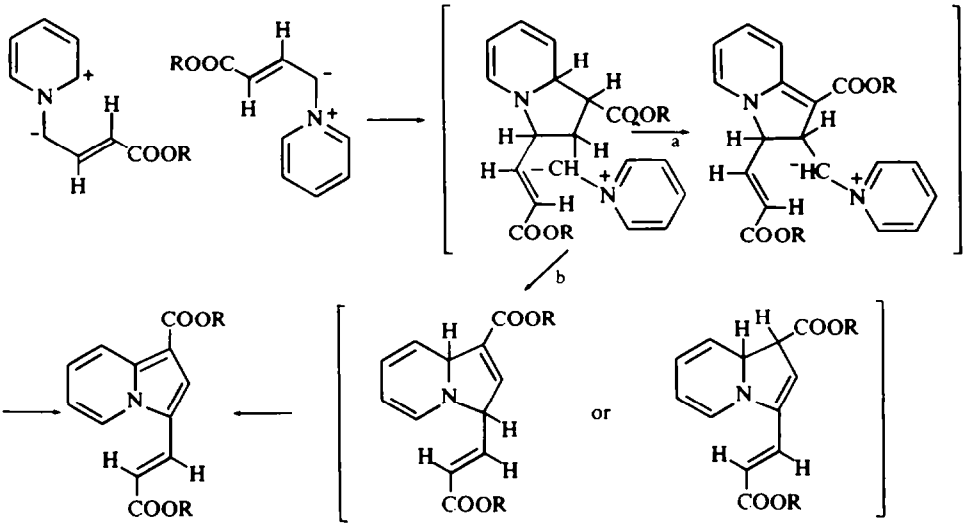


Compd	Proton Signals (τ) R_2	R_3	R_4	R_5	R_6	R	Ph	Coupling Constants (Hz)
20a	0·08 bd	2·09	2·09	1·61 b dd	2·09–3·20 m*		2·09–3·20 m*	
		3·20 m*	3·20 m*			6·13 s		$J_{2,3}$ = 7·5 Hz, $J_{4,5}$ = 9·0 Hz
								$J_{5,3}$ = 1·5 Hz
20b	0·15 bd	3·05 bt	2·22–2·82 m*		2·22–2·82 m*		2·22–2·82 m*	
				1·72 bd		5·70 q		$J_{2,3}$ = 7·5 Hx, $J_{3,4}$ = 7·5 Hz
						8·62 t		$J_{4,5}$ = 7·5 Hz
21a	0·24 d	3·14 b dd	7·51 s	1·87 bs	2·15–2·85 m*		2·15–2·85 m*	
						6·15		$J_{2,3}$ = 7·5 Hz, $J_{3,5}$ = 1·5 Hz
21b	0·20 d	3·12 b dd	7·51 s	1·85 bs	2·10–1·82 m*		2·10–2·82 m*	
						5·65 q		$J_{2,3}$ = 7·5 Hz, $J_{3,5}$ = 1·5 Hz
						8·62 t		

* Overlapped each other.

The structures of compounds **22a** and **22b** could be determined by their IR and NMR spectra. Compounds **22a** and **22b** exhibited carbonyl absorption at 1665 cm^{-1} and hydroxyl absorption at 3080 cm^{-1} in the IR spectra, the latter shifted strongly to lower frequency due to hydrogen bonding between these groups. While, the NMR spectra showed signals at τ −1·09 (1H) in **22a** and at τ −0·97 (1H) in **22b**, indicative of a hydrogen-bonding hydroxyl group. From these facts, the structures of **22a** and **22b** were assigned as 3,4-diphenyl salicylates.

Reaction mechanism. As described above, the formation of 3-ethenylindolidines (**12–17**) could proceed *via* well known 1,3-dipolar cycloaddition reactions of two molecules of pyridinium N-allylide, in which one molecule acts as a 1,3-dipole and the other as a 1,3-dipolarophile as shown in Scheme 9.



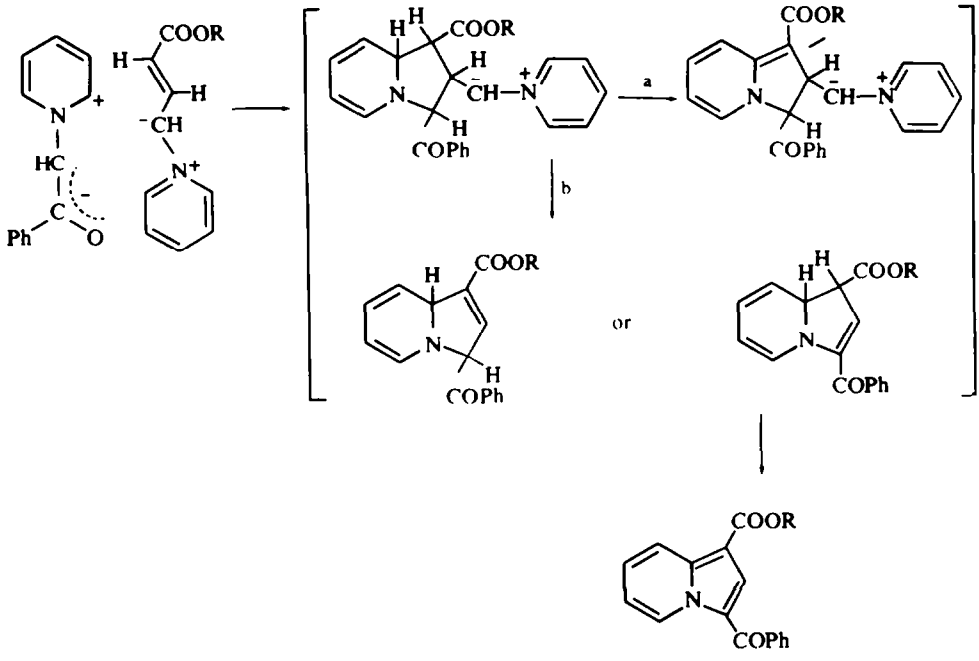
SCHEME 9.

Thus, it is concluded that the pyridinium N-allylides might be present as 1,3-dipoles rather than 1,5-dipoles in the resonance contribution in the ground state. Accordingly, mechanistic speculation leads us to consider paths **a** and **b** (Scheme 9), and in path **b**, analogous elimination at C-2 on the indolidine ring has been proposed by Krönke and Mörler.[7]

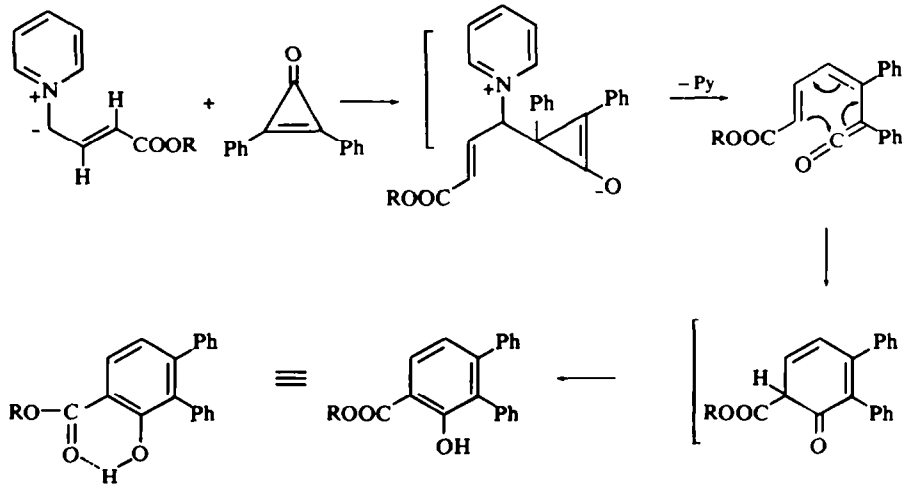
In contrast, the formation of the 1-alkoxycarbonyl-3-benzoylindolidine derivatives indicates considerable stabilization of the carbanion in pyridinium N-phenacylide (afforded by the carbonyl group) compared with that of pyridinium N-allylide. The pyridinium N-phenacylide reacts as a 1,3-dipole and the reaction mechanism through paths **a** and/or **b** could be proposed as shown in Scheme 10.

The mechanism for the formation of **22a** and **22b** might be explained as an initial Michael type addition of the carbanion in the N-allylide to the β-carbon of the double bond in DPP (Scheme 11), as suggested by Eicher and Angerer.[8]

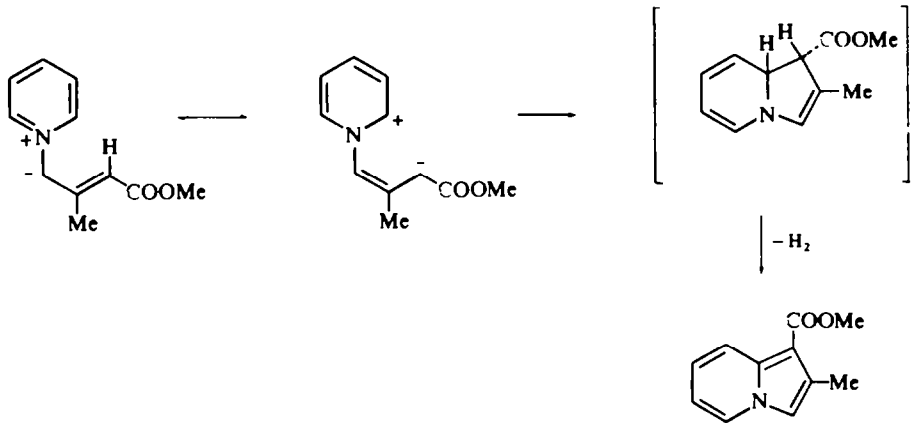
Finally, the formation of 3-unsubstituted indolidine derivatives (**23–27**) seems to proceed *via* intramolecular 1,5-dipolar cyclization followed by dehydrogenation (Scheme 12). Such a mechanism has been proposed by us for the intramolecular cyclization of N-vinyliminopyridinium ylides.[2]



SCHEME 10.



SCHEME 11.



SCHEME 12.

EXPERIMENTAL

M.ps were measured with a Yanagimoto micromelting point apparatus and are uncorrected. Microanalyses were performed on a Perkin–Elmer 240 Elemental Analyser. NMR spectra were determined with a Japan Optics Co., Model C-60-XL NMR spectrometer with TMS as internal standard (chemical shifts are in τ values). IR spectra were taken with a JASCO Model IR-S spectrophotometer and UV spectra were obtained on a JASCO Model ORD/UV-5 recorder.

Preparations of pyridinium salts (4–11) *General Method*: A mixture of γ-bromocrotonate (**1** and **2**) or γ-bromo-β-methylcrotonate (**3**) and a small excess of pyridinine derivatives was stirred in $CHCl_3$ (50 ml) at room temp for 1–2 days, and the mixture concentrated at reduced pressure. After unreacted pyridine derivatives were ether extracted, the corresponding pyridinium salts were obtained as hygroscopic brown oily materials in quantitative yield. The crude pyridinium salts (**8–11**), which were found to be mixtures of isomers in the ratio of 6:1, were used without further purifications.

Reactions of pyridinium salts (**4–7**) *in the presence of potassium carbonate. General Method*: A solution of pyridinium salt (1·0 g) and K_2CO_3 (3 g) in $CHCl_3$ (50 ml) was stirred at room temp for 1 day and then insoluble material was removed. The filtrate was concentrated *in vacuo* and the residue separated by column chromatography (silica gel, benzene as eluent). Recrystallization from benzene–ether gave the corresponding 3-ethenylindolidine derivatives (**12–16**) (Table 4).

Reactions of pyridinium N-allylides and ethyl propiolate. A solution of **4a** or **4b** (2 mmol) and ethyl propiolate (0·4 g, 4 mmol) in $CHCl_3$ (50 ml) was stirred with K_2CO_3 (3·0 g) at room temp for 1 day. Work-up as above afforded **17** (0·11 g, 20%); m.p. 131–133°, ν (KBr) 1676 cm^{-1} (C=O), or **12b** (0·13 g, 23%). Com-

TABLE 4.

Product	Yield (%)	m.p. (°C)	Formula	Calcd C	Calcd H	Calcd N	Found C	Found H	Found N
12a	43	141–142	$C_{14}H_{13}NO_4$	64·86	5·05	5·40	64·94	5·14	5·37
12b	41	131–132	$C_{16}H_{17}NO_4$	66·88	5·96	4·88	66·92	6·13	4·65
13a	37	149–150	$C_{15}H_{15}NO_4$	65·92	5·53	5·13	65·90	5·60	5·08
13b	57	111–112	$C_{17}H_{19}NO_4$	67·76	6·36	4·65	67·81	6·38	4·52
14a	50	155–156	$C_{15}H_{15}NO_4$	65·92	5·53	5·13	65·64	5·71	5·23
14b	40	118–119	$C_{17}H_{19}NO_4$	67·76	6·36	4·65	67·81	6·38	4·52
15*	3*		$C_{17}H_{19}NO_4$	67·76	6·36	4·65	67·91	6·48	4·58
16*	53*								

* The ratio of **15** to **16** was obtained from the NMR.

pound **12b** was identified with the product, prepared from the reaction of **4b** in the presence of K_2CO_3 by comparison of its physical and spectral properties. **17**: (Found: C, 66·00; H, 5·33; N, 5·20. Calc. for $C_{15}H_{15}NO_4$ C, 65·92; H, 5·53; N, 5·13%).

Reactions of pyridinium N-allylides with pyridinium N-phenacylides. General Method: A mixture of pyridinium salts, **4a** or **4b** (2·5 mmol) and **18** or **19** (2·5 mmol), was stirred with K_2CO_3 (3·0 g) in $CHCl_3$ (50 ml) at room temp for 1 day. Work-up as above afforded the corresponding 3-benzoylindolidine derivatives (Table 5).

TABLE 5.

Reactant Compd.	Product	Yield (%)	m.p.(°C)	Formula	Calc. C	Calc. H	Calc. N	Found C	Found H	Found N
4a + **18**	**20a**	20	156–157	$C_{17}H_{13}O_3N$	73·11	4·69	5·02	72·92	4·88	5·05
4b + **18**	**20b**	17	81–82	$C_{18}H_{15}O_3N$	73·70	5·15	4·78	73·99	5·45	4·52
4a + **19**	**21a**	12*								
	12a	48*								
4b + **19**	**21b**	26	131–132	$C_{19}H_{17}O_3N$	74·25	5·58	4·56	74·05	5·83	4·47

* The ratio of **21a** to **12a** was obtained from the NMR.

Reactions of N-allylides with DPP. A solution of **5a** or **5b** (1·0 mmol) and DPP (0·21 g, 1 mmol) in benzene (50 ml) was stirred with K_2CO_3 at room temp for 1 day. Work-up as above afforded **22a** (0·22 g, 72%) or **22b** (0·26 g, 82%) as colourless needles.

22a: m.p. 112–114°, ν (KBr) 3080 (OH), 1665 cm^{-1} (C=O), λ_{max}(MeOH) 227 (ε 1·48 × 10^4), 273 (7·36 × 10^3), 318 nm (3·80 × 10^3), τ (CCl_4) 6·06 (s, 3H, OMe), 3·11 (d, 1H, $J_{5,6}$ 8·0 Hz, H-5), 2·90 (d, 10H, 2 × Ph), 2·18 (d, 1H, $J_{6,5}$ 8·0 Hz, H-6), −1·09 (s, 1H, OH). (Found: C, 79·21. H, 5·55. Calc. for $C_{20}H_{16}O_3$ C, 79·10; H, 5·55%).

22b: m.p. 126–129°, ν (KBr) 3080 (OH), 1665 cm^{-1} (C:O), λ_{max} (MeOH) 226 (ε 2·44 × 10^4), 272 (1·19 × 10^4), 316 nm (6·07 × 10^3), τ (CCl_4) 8·59 (t, 3H, J = 7·0 Hz, $OCH_2\underline{CH_3}$), 5·64 (q, J = 7·0 Hz OCH_2), 3·18 (d, 1H, $J_{5,6}$ = 8·0 Hz, H-5), 2·98 (d, 10H, 2 × Ph), 2·23 (d, 1H, $J_{6,5}$ = 8·0 Hz, H-6), −0·97 (s, 1H, OH). (Found: C, 79·14; H, 5·81. Calc. for $C_{21}H_{18}O_3$ C, 79·22; H, 5·70%).

Intramolecular cyclizations of pyridinium N-allylides derived from pyridinium salts (**8**–**11**). These reactions were carried out by the same procedure, which was used in the reactions of pyridinium salts (**4**–**7**). Recrystallization from *n*-hexane gave the corresponding indolidine derivatives (**23**–**27**) as colourless crystals (Table 6).

TABLE 6.

Compd.	Yield	m.p. (°C)	Formula	Calcd. C	Calcd. H	Calcd. N	Found C	Found H	Found N
23	46	40–41	$C_{11}H_{11}O_2N$	69·82	5·86	7·40	69·63	5·94	7·25
24	49	93–96	$C_{12}H_{13}O_2N$	70·91	6·45	6·89	70·78	6·55	6·62
25*	4*		$C_{12}H_{13}O_2N$	70·91	6·45	6·89	70·94	6·41	6·89
26*	45*								
27	54	77–78	$C_{12}H_{13}O_2N$	70·91	6·45	6·89	70·90	6·56	6·79

* The ratio of **25** to **26** was obtained from the NMR.

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