

4,6-Bis(1-morpholino)-2-(1-phenylethylideneamino)-1,3,5-oxadiazinium Hexachloroantimonate (5h). Reaction time 3 h, yield 71%, orange prisms, mp 203–204 °C (dec). IR (CH₂Cl₂): 1535, 1575, 1670 cm⁻¹.

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Synthesis of Some New 2,4-Diaryl-6-(β -phenylvinyl)pyridines via Phenacylpyridinium Bromides

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Phenacylpyridinium bromide, *p*-chlorophenacylpyridinium bromide, and *p*-methylphenacylpyridinium bromide were reacted with *para*-substituted dibenzalacetones in presence of ammonium acetate in glacial acetic acid to give 2,4-diaryl-6-(β -phenylvinyl)pyridines in 45–65% yields. Ammonium acetate in acetic acid was used as an aza cyclization agent. The structures of the resulting pyridines were confirmed by IR and NMR spectral data and elemental analyses.

Pyridinium salts and their ylides have gained considerable importance in the synthesis of indoles (1), tetrazine (2), pyridines (3), and polynuclear hydrocarbon (4, 5). Recently we have studied the synthetic and mechanistic aspects of ylides and their salts of group V elements. We now report herein the reactions of some *para*-substituted phenacylpyridinium bromides with substituted dibenzalacetones in the presence of ammonium acetate in glacial acetic acid with a view to examine the aza ring closure ability of pyridinium salts with dibenzalacetones (Scheme I).

Experimental Section

Phenacylpyridinium salts (1a–c) were prepared by the reaction of substituted phenacyl bromides with pyridine at reflux (9, 10). Substituted dibenzalacetones were prepared by the condensation of acetone with substituted benzaldehydes in the presence of aqueous NaOH solution (11).

The IR spectra of pyridines in general showed two characteristic bands in the region 1500 and 1600 cm⁻¹ due to stretching vibrations of C=N and C=C of the pyridine nucleus. In the NMR spectra olefin and aromatic protons were observed in the range δ 6.75–7.10 and δ 7.05, respectively.

Preparation of 2,4-Diaryl-6-(β -phenylvinyl)pyridines (5a–k). **General Procedure.** A mixture of phenacylpyridinium salt (1a–c, 3 mmol), ammonium acetate (3 g), and glacial acetic acid (50 mL) was stirred at 80 °C for 2–3 h. The dibenzalacetone (3.3 mmol) in glacial acetic acid (20 mL) was added dropwise during an interval of 1 h. The temperature was raised

Scheme I

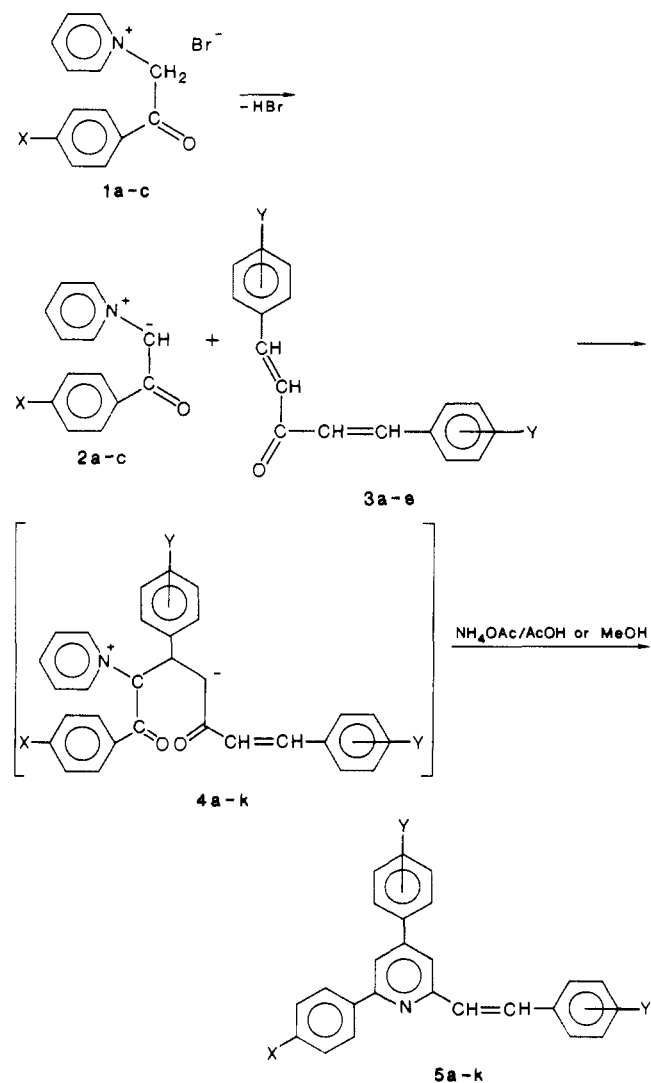


Table I. Physical and Spectral Data of Compounds 5a-k

compd	X	Y	yield	recryst solvent	mp, °C	(δ) NMR ^a data (CDCl ₃)
5a	H	H	50	A	108-10	6.85 (q, 2 H, -CH=CH-); 7.15-8.40 (m, 17 H, ArH + PyH)
5b	H	4-OCH ₃	55	B	158-60	6.80 (q, 2 H, -CH=CH-); 3.75 (s, 3 H, -OCH ₃); 3.88 (s, 3 H, -OCH ₃); 7.05-8.25 (m, 15 H, ArH + PyH)
5c	H	3,4-(OCH ₃) ₂	45	C	186-88	c-6.65 (q, 2 H, -CH=CH-); 3.75 (d <i>J</i> = 6 Hz, 6 H, (-OCH ₃) ₂); 4.05 (d <i>J</i> = Hz, 6 H, (-OCH ₃) ₂); 7.20-8.25 (m, 13 H, ArH + PyH)
5d	H	4-(CH ₃) ₂ N	58	C	170-72	6.75 (q, 2 H, -CH=CH-); 2.95 (d <i>J</i> = 5 Hz, 12 H, (-N(CH ₃) ₂)); 7.05-8.10 (7, 15 H, ArH + PyH)
5e	4Cl	H	60	B	140-42	7.00 (q, 2 H, -CH=CH-); 7.15-8.30 (m, 16 H, ArH + PyH)
5f	4Cl	4-OCH ₃	65	D	90-92	7.05 (q, 2 H, -CH=CH-); 3.85 (d, <i>J</i> = 5.5 Hz, 6 H, (-OCH ₃) ₂); 7.20-8.15 (m, 14 H, ArH + PyH)
5g	4Cl	4-(CH ₃) ₂ N	60	B	128-30	7.10 (q, 2 H, -CH=CH-); 3.05 (d, <i>J</i> = 5 Hz; 12 H, (-N(CH ₃) ₂)); 7.25-8.35 (m, 14 H, ArH + PyH)
5h	4-CH ₃	H	65	B	120-22	6.95 (q, 2 H, -CH=CH-); 2.45 (s, 3 H, -CH ₃); 7.05-8.15 (m, 16 H, ArH + PyH)
5i	4-CH ₃	4-OCH ₃	65	A	142-44	6.90 (q, 2 H, -CH=CH-); 2.35 (s, 3 H, -CH ₃); 3.90 (d, <i>J</i> = 6 Hz, 6 H, (-OCH ₃) ₂); 7.10-8.25 (m, 14 H, ArH + PyH)
5j	4-CH ₃	4-(CH ₃) ₂ N	60	D	152-54	6.80 (q, 2 H, -CH=CH-); 2.38 (s, 3 H, -CH ₃); 2.95 (D, <i>J</i> = 5 Hz) 12 H, (-N(CH ₃) ₂); 7.00-8.15 (m, 14 H, ArH + PyH)
5k	4-CH ₃	4-Cl	50	D	100-02	6.95 (q, 2 H, -CH=CH-); 2.42 (s, 3 H, -CH ₃); 7.08-8.22 (m, 14 H, ArH + PyH)

^a NMR spectra were run on Varian A-60 spectrometer using TMS as internal standard. s = singlet, m = multiplet, d = doublet, q = quartet, A = C₅H₅N-MeOH, B = CHCl₃-MeOH, C = EtOH, D = EtOH-H₂O.

to 120 °C and heating was continued for an additional 5-8 h. The reaction mixture was left overnight at room temperature. Ice water (50 mL) was added with constant stirring. The resulting solid mass was filtered, washed twice with water and then methanol, dried, and recrystallized from a suitable solvent (see Table I) to give the pyridines (5a-k).

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4-Me₂NC₆H₄CHO, 100-10-7; 4-ClC₆H₄CHO, 104-88-1; pyridine, 110-86-1; acetone, 67-64-1.

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