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Synthesis and Biological Activity of Some 1-Isonicotinoyl-3,5-Diphenyl-4-Substituted Phenyl azo Pyrazoles

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1-Isonicotinoyl-3,5-diphenyl-4-substituted phenyl azo pyrazoles have been synthesised by the reaction of substituted phenyl azo dibenzoyl methane with the antitubercular drug isoniazid. These compounds were screened for antibacterial and fungicidal activities. Pyrazoles have shown 30–45% fungicidal effect and inhibitory effect to the extent of 25–35% has been noticed against *E. coli, S. aureus* and *S. alvus*.

INTRODUCTION

Pyrazoles have been reported to possess various biological activities[1]. Some of the pyrazoles have been found to possess antibacterial[2] and fungicidal[3] activities. Ahluwalia and coworkers[4] have synthesised some new pyrazoles which were evaluated as antibacterial and antifungal agents. The present communication deals with the reaction of dibenzoyl methane with diazotised aromatic primary amines in presence of sodium acetate which gave 1a. Condensation of 1a with isonicotinic acid hydrazide furnished a number of new pyrazoles 2a (Scheme 1)

Antibacterial activity: Compounds 2b, 2c, 2d, 2i and 2j were tested against *E. coli, S. aureus* and *S. alvus* using cup-plate method[5]. The percent control on the infected plate was found in the range of 25–35%.

Fungicidal activity: Fungicidal activities of compounds 2b, 2c, 2d, 2h and 2j were tested by Agar plate food poisoning method[6] in 500 ppm and 1000 ppm concentrations. All compounds were found active against *Alternaria alternata* and *Rhizopus arrhizus*. The results of fungicidal activity are given in Table 1.

EXPERIMENTAL

Melting points are uncorrected. The IR spectra (KBr) were taken on the Perkin-Elmer-577 model spectrophotometer and elemental analyses were carried out by CEST-110 model analyser.

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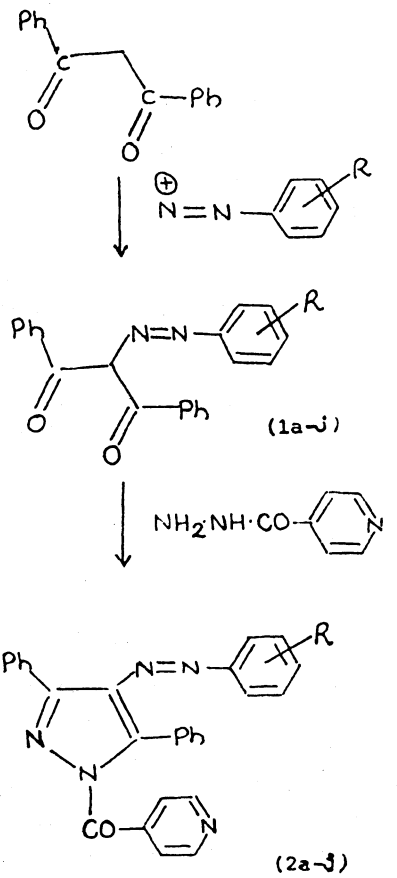

Scheme 1

TABLE 1
FUNGICIDAL ACTIVITY OF THE COMPOUNDS

Compd. No.	Percentage control*			
	Alternaria alternata		Rhizopus arrhizus	
	500 ppm	1000 ppm	500 ppm	1000 ppm
2b	30	39	35	40
2c	35	40	40	43
2d	37	42	36	44
2h	35	40	32	45
2i	30	34	36	41

*As compared to the blank plate.

Phenyl-azo dibenzoyl methane (General procedure) 1a

Aniline (2.1 ml, 0.02 mol) was diazotised using aqueous hydrochloric acid (16

ml, 1 : 1) and sodium nitrite (2.4 gm in 6 ml water) at 0°C and the diazotised solution was added to a slurry of dibenzoyl methane (4.48 gm, 0.02 mol) and sodium acetate (1.6 gm in 2 ml 50% ethanol) while stirring in an ice bath, when yellow solid separated. It was filtered under suction and recrystallised from ethanol.

Yield, 68%; m.pt. 78°C, Found: C, 76.01; H, 4.06; N, 7.73. $C_{21}H_{16}N_2O_2$ requires: C, 76.82; H, 4.87; N, 8.53%. ν_{max} (KBr) 750 (substituted phenyl), 1590 (–N=N–) and 1650 cm^{-1} (C=O)

The other compounds using appropriate amines and dibenzoyl methane were prepared by the aforementioned procedure (Table 2).

TABLE 2
PHYSICAL DATA OF SUBSTITUTED PHENYL-AZO DIBENZOYL METHANES (1a–j)*

Compd. No.	R	M.pt. (°C)	Colour†	Yield (%)	Mol. Formula
1a	H	78	LY	68	$C_{21}H_{16}N_2O_2$
1b	Me(2)	85	DY	58	$C_{22}H_{18}N_2O_2$
1c	Me(4)	80	LY	66	$C_{22}H_{18}N_2O_2$
1d	OMe(2)	55	BY	38	$C_{22}H_{18}N_2O_3$
1e	OMe(4)	60	DY	48	$C_{22}H_{18}N_2O_3$
1f	OEt(2)	60	Y	53	$C_{23}H_{20}N_2O_3$
1g	OEt(4)	65	OY	67	$C_{23}H_{20}N_2O_3$
1h	NO_2(2)	60	DY	67	$C_{21}H_{15}N_3O_4$
1i	NO_2(4)	75	BY	80	$C_{21}H_{15}N_3O_4$
1j	Br(4)	82	DY	57	$C_{21}H_{15}N_2O_2Br$

*All compounds gave satisfactory elemental analysis.
†Y = Yellow; LY = Light Yellow; DY = Dark Yellow; BY = Brown Yellow; OY = Orange Yellow.

1-Isonicotinoyl-3,5-diphenyl-4-phenyl-azo-pyrazole (General procedure) 2a

A mixture of phenyl-azo-dibenzoyl methane (0.656 gm 0.002 mol), isonicotinic acid hydrazide (0.274 gm 0.002 mol), glacial acetic acid (6.5 ml) and conc. sulphuric acid (2 drops) was refluxed for 6 hrs. The contents were cooled, when the product separated. It was filtered under suction and repeatedly washed with hot acetic acid.

Yield, 20%; m.pt. 205°C; Found: C, 75.11; H, 4.01; N, 15.91. $C_{27}H_{19}N_5O$ requires: C, 75.52; H, 4.42; N, 16.31%. ν_{max} (KBr) 750 (substituted aromatic ring), 1320 (–C=N ring), 1600 (C=C phenyl) and 1650, 1730 cm^{-1} (due to exocyclic C=O).

The other pyrazoles were prepared by the aforementioned method (Table 3).

TABLE 3
PHYSICAL DATA OF 1-ISONICOTINOYL-3,5-DI-PHENYL-4-SUBSTITUTED PHENYL AZO PYRAZOLES (2a–j)*

Compd. No.	R	M.pt. °C	Colour†	Yield (%)	Mol. formula
2a	H	205	Y	20	$C_{27}H_{19}N_5O$
2b	Me(2)	208	OY	28	$C_{28}H_{21}N_5O$
2c	Me(4)	210	Y	30	$C_{28}H_{21}N_5O$
2d	OMe(2)	200	Y	11	$C_{28}H_{21}N_5O_2$
2e	OMe(4)	265	BY	27	$C_{28}H_{21}N_5O_2$
2f	OEt(2)	215	LY	12	$C_{29}H_{23}N_5O_2$
2g	OEt(4)	235	Y	23	$C_{29}H_{23}N_5O_2$
2h	NO_2(2)	195	BY	16	$C_{27}H_{18}N_6O_3$
2i	NO_2(4)	205	Y	26	$C_{27}H_{18}N_6O_3$
2j	Br(4)	220	LY	19	$C_{27}H_{18}N_5OBr$

*All compounds gave satisfactory elemental analysis.
†Y = Yellow; LY = Light Yellow; BY = Brown Yellow; OY = Orange Yellow.

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REFERENCES

1. R.B. Pathak and S.C. Bahel, *J. Indian Chem. Soc.*, **57**, 1103 (1980).
2. M. Sree Ram Murty, E. Venkata Rao and P. Ranganathan, *Indian Drugs*, **22**, 247 (1985).
3. J.N. Philips, S.D. Huppatz, B. Witrzeus and S.J. Grant, *Pestic. Biochem. Physiol.*, **25**, 163 (1986); *Chem. Abstr.*, **104**, 220643 (1986). R.S. Sharma, R.B. Pathak and S.C. Bahel, *J. Indian Chem. Soc.*, **62**, 625 (1985).
4. V.K. Ahluwalia, Uttara Dutta and H.R. Sharma, *J. Indian Chem. Soc.*, **64**, 221 (1987).
5. W.M.M. Kirbi and A.W. Bauer, *Am. J. Clin. Pathol.*, **45**, 493 (1966).
6. Y.L. Nene and P.N. Thapliyal, Fungicides in Plant Disease Control, 2nd ed., Oxford and IBH Publishing Co., New Delhi, p. 413 (1979).

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