

**FACTORS INFLUENCING THE PATHWAY OF REACTIONS OF 1-HYDRAZINO-
PHthalAZINE WITH DI- AND TRICARBONYL COMPOUNDS**

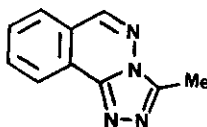
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Abstract — The reaction of 1-hydrazinophthalazine (hydralazine) (1) with substituted ethyl benzoylpyruvates (3) leads under neutral reaction conditions to 3-[2-oxo-2-(substituted phenyl)ethyl]-4H-as-triazino[3,4-a]phthalazin-4-ones (4). Under acidic but otherwise identical conditions depending on the substituent of 3, 4 and/or 3-carbethoxy-s-triazolo[3,4-a]phthalazine (5) with simultaneous elimination of the appropriate acetophenone were obtained. 1-HCl and substituted ethyl cinnamoylpyruvates (6a-e) give rise to formation of 1-(1-phthalazinyl)-3-carbethoxy-5-(3- or 4-substituted styryl)pyrazoles (7). Under neutral conditions the reaction of 1 with 6 gives as-triazino[3,4-a]phthalazine (8). The structures of 4, 5, 7, and 8 are based on elemental analysis and extensive ms- and ¹H-nmr investigations.

Introduction

Reactions of the antihypertensive compound 1-hydrazinophthalazine (1) with carbonyl compounds are well known and also play an important role in the human metabolism of this drug. Thus, a prominent urinary metabolite of 1 is 3-methyl-s-triazolo[3,4-a]phthalazine (2) as was found independently by two research groups.^{1,2,3} Compound 2 also was obtained in attempts to acylate 1.⁴ Similar results were obtained by reacting 1 with a number of monocarboxylic acids^{5,6} as well as with di-, tri- and tetracarboxylic acids.⁶ The latter acids only yielded the corresponding 2-analogs at all as well as in good yields if, instead of the appropriate acid chlorides,⁵ the corresponding p-nitrophenyl esters were employed.⁶



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1,2-Dicarbonyl compounds, such as di-p-nitrophenyl oxalate or pyruvic acid yielded with 1 an annelated six-membered ring. In the case of the oxalate 1-H-as-triazino[3,4-a]phthalazine-3,4-dione was formed;⁶ whereas pyruvic acid yielded by heating the originally formed 3-methyl-1-H-as-triazino[3,4-a]phthalazin-4-one the triazolo compound 2.⁴

Reactions of 1 with 1,3-dicarbonyl compounds lead to formation of 2 by loss of acetone in the case of 2,4-pentanedione and by loss of ethyl acetate with ethyl acetoacetate.^{4,7} The corresponding pyrazolones were not formed as was reported erroneously.⁸

Attempts to annelate seven or eight-membered rings to phthalazine by reacting 1 with β -ketoglutaric acid, β -acetylacrylic acid, or levulinic acid failed. In the case of β -ketoglutaric acid the hydralazone of acetone was obtained as the only product, whereas the other two ketoacids either gave only the corresponding hydrazones or decomposition products.⁶ The reaction between 1 and phthalaldehydic acid only leads to a pair of ring-chain tautomers in which the seven-membered ring tautomer, 6,7-dihydro-7-hydroxy-12H-phthalazino[2,1-b][2,4]-benzodiazocin-12-one and the chain-tautomer, 2-(2-formylbenzoyl)-1-hydrazono-1,2-dihydrophthalazone could not be separated.⁹

The present paper describes the results of further attempts to annelate a seven-membered ring to phthalazine by reacting 1 with a number of 1,2,4-tricarbonyl compounds such as substituted ethyl benzoylpyruvates (3). This reaction initially yields hydrazones involving the benzoyl carbonyl group. These hydrazones then could conceivably undergo a ring-closure reaction along four different pathways. a: The N-2 atom of the hydralazine moiety attacks the keto group leading to formation of a pyrazole ring. b: The N-3 atom attacks the ester group with displacement of ethanol yielding the fused 1,2,4-triazino system. c: The N-3 atom attacks the carbon atom of the azomethine group with displacement of an alkyl group, yielding a fused 1,2,4-triazolo ring. d: Attack of the N-3 atom on the keto group to yield the fused 1,2,4-triazepino ring system (Scheme I).

Indeed, preliminary experiments investigating the reaction between 1 and 1,2,4-tricarbonyl compounds showed that different structures were obtained. It was found that the conditions (acidic or neutral) were responsible for the pathway the reaction followed.¹⁰

Neutral Condition

Thus, refluxing 1 with 3 3-[2-oxo-2-(substituted phenyl)ethyl]-4H-as-triazino[3,4-a]phthalazin-4-ones (4) were obtained. Electronic effects of the substituents have little, if any, effect on the yields (Table I).

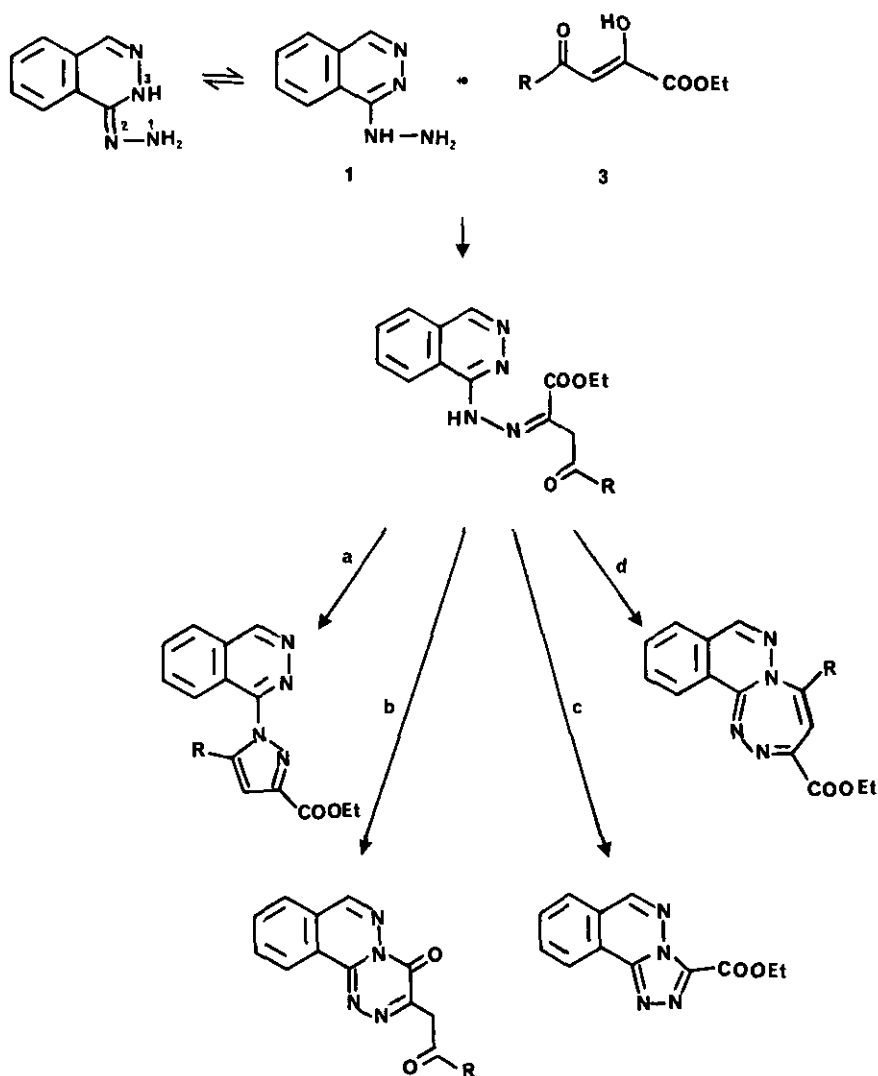
Table I

3-[2-Oxo-2-(3-or 4-substituted phenyl)ethyl]-4H-as-triazino[3,4-a]phthalazin-4-ones (4)

	R	mp°C	yield, %
4a	p-OMe	270-4	69
4b	p-Me	259-61	49
4c	m-OMe	257-9	50
4d	p-NO ₂	315-6	59

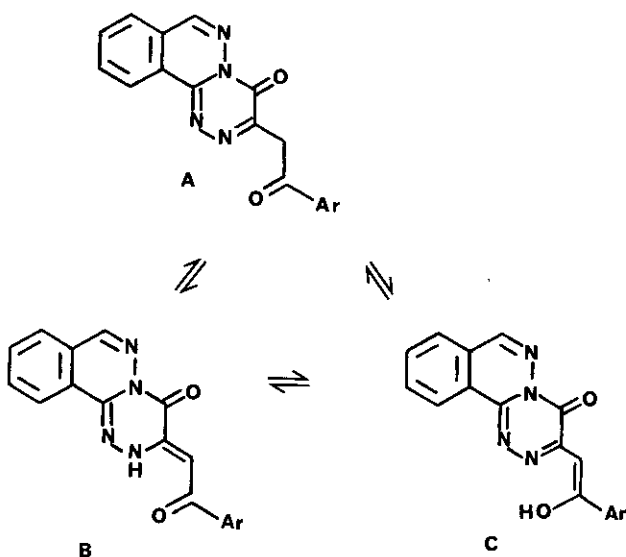
The structures of type-4 compounds are based on ¹H-nmr, ir- and ms-spectroscopic data (see experimental). Type-4 compounds can occur in a number of tautomers;¹⁰⁻¹³ structures A, B, and C might represent the most probable ones (Scheme II).

Scheme I



Indeed, according to nmr-evidence of compounds 4a and 4b when dissolved in $CDCl_3$, a mixture of tautomers was observed. Their 1H -nmr spectra showed a peak between δ 14.2-14.3 which integrated to almost one proton. This peak could be attributed to the NH-group of B or the OH-group of C. A singlet peak appearing at δ 6.88 or 6.87 respectively is due to the $-CH=$ proton of the side chain of either B or C. The weak peak at δ 4.7 is attributed to the $-CH_2-$ group of the side chain in A. Compound 4c did not show a peak for the $-CH_2-$ group, therefore, tautomer A is present to only traces or not at all. Due to solubility problems with compound 4d no 1H -nmr-spectrum could be obtained. Thus, on the basis of the 1H -nmr-spectra, a definite structure assignment for tautomer B or C could not be made. However, the long wave uv-absorption of compounds 4a-4d with λ_{max}

Scheme II

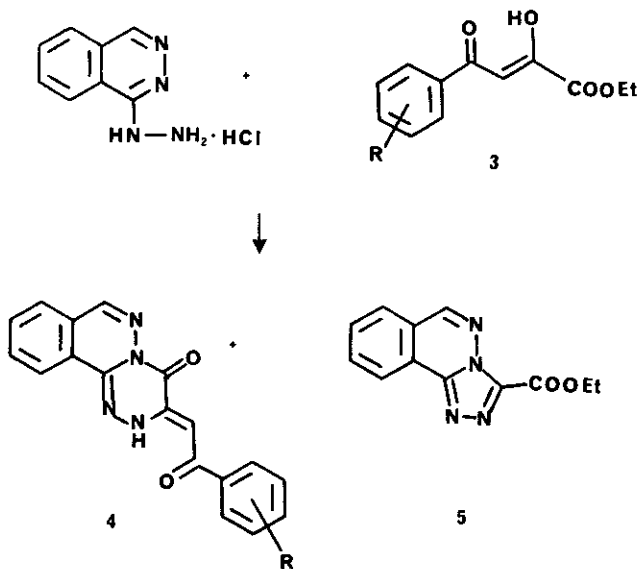


values between 475 nm and 486 nm and especially the $\lambda_{\max}$ of 486 nm for 4d, seems to indicate that structure C is probably the correct one for the predominate tautomer in highly dilute solution.

Acidic Conditions

If 1-hydrochloride was reacted with type-3 compounds a mixture of type-4 compounds and 3-carbethoxy-s-triazolo[3,4-a]phthalazine (5) was obtained (Scheme III).

Scheme III



Formation of **5** should be accompanied by an elimination of the appropriate acetophenone. That this is the case was shown by isolation of the corresponding acetophenones. They were characterized by their 2,4-dinitrophenylhydrazones.

The reaction under acidic conditions is apparently sensitive towards electronic effects. Thus, electron donating groups in type-3 compounds favor formation of **5** whereas, the strongly electronic withdrawing p-nitro group leads to formation of the corresponding type **4** compound and only traces of **5** (Table II).

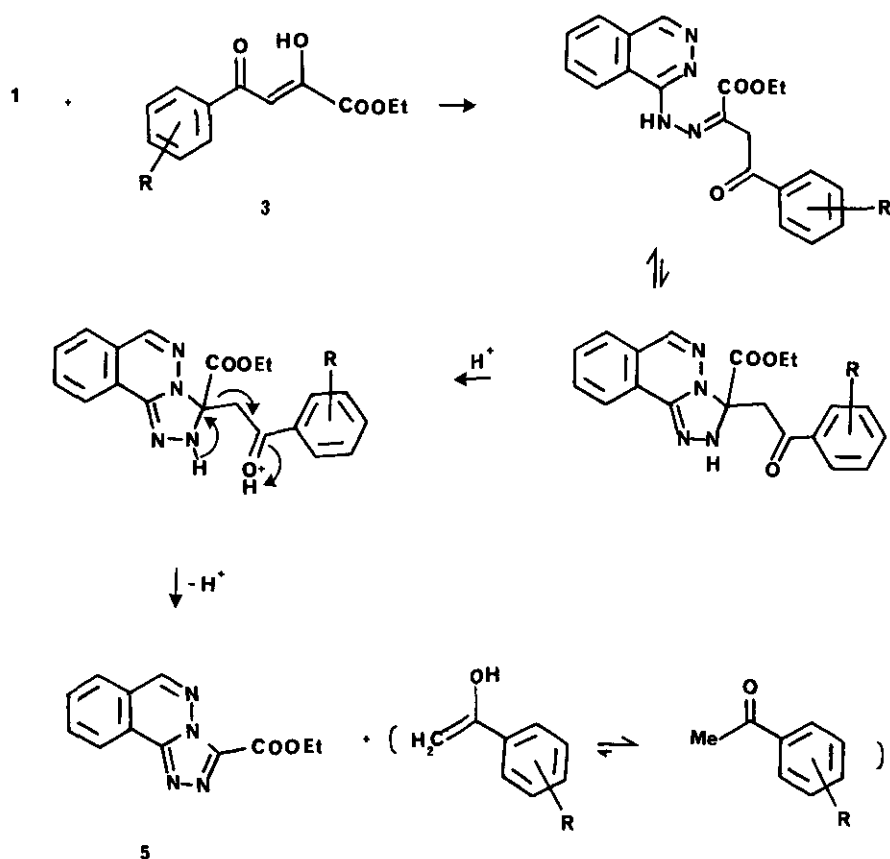
Table II

Products from the Reaction of Hydralazine Hydrochloride with Ethyl Benzoylpyruvates (**3a-d**)

	R	yield, % of 4	yield, % of 5
3a	p-OCH ₃	small amount	62
3b	p-CH ₃	small amount	53
3c	m-OCH ₃	10	23
3d	p-NO ₂	41	small amount

A mechanism which could account for formation of **5** is assumed as follows (Scheme IV):

Scheme IV



Reaction with Ethyl Cinnamoylpyruvates

When 1-hydrochloride was reacted with a number of different ethyl cinnamoylpyruvates (**6**) the reaction proceeded differently from the one using type-3 compounds. The products obtained in this case were proven to be pyrazoles (**7a-e**) (Table III). They are formed by the intramolecular attack of the N-2 atom on the keto group via the transiently occurring hydrazone. This finding is in agreement with a recent publication¹⁴, in which only a tentative structure proof for **7** was given (Scheme V).

Scheme V

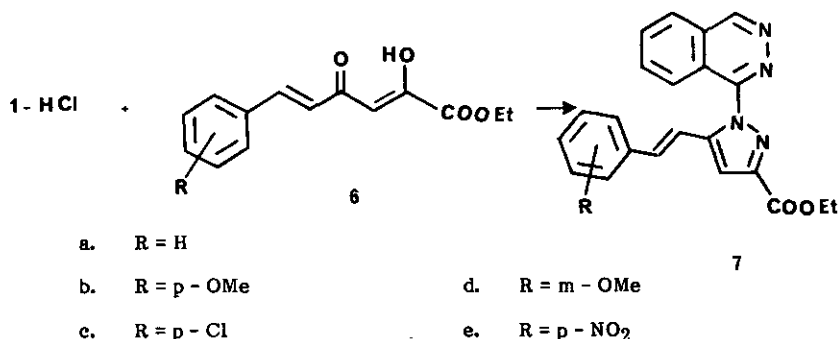


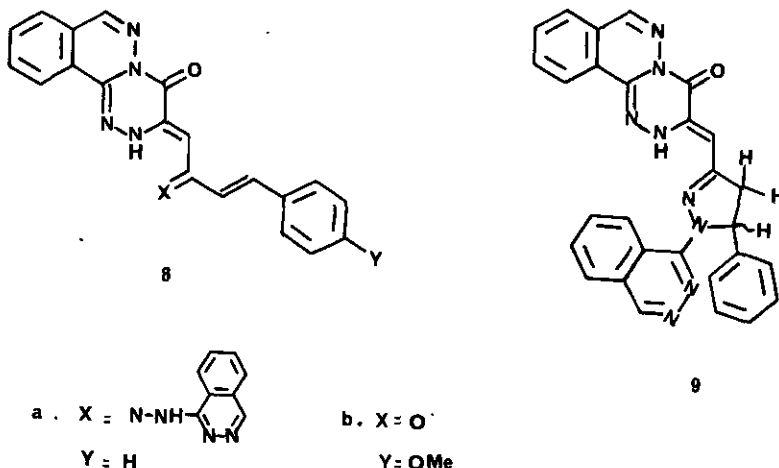
Table III

1-(1-Phthalazinyl)-3-carbomethoxy-5-(3 or 4-substituted styryl) pyrazoles (**7**)

	R	mp°C	yield, %
7a	H	254	53
7b	p-OMe	185-6	40
7c	p-Cl	152-5	49
7d	m-OMe	202-4	39
7e	p-NO ₂	272-4	32

The structure assignments of type **7** compounds are based on nmr-, ir-, and ms-data (see experimental). The ester group attached to the pyrazole ring can easily be recognized by the characteristic ester ir frequency appearing between 1716 and 1736 cm⁻¹, while the weak absorption band between 1631 and 1638 cm⁻¹ can be assigned to the C=C double bond of **7a-e**. Also, their ¹H-nmr spectra show a triplet and quartet at about δ 1.4 and 4.5 ppm respectively, which correspond to the three methyl and two methylene hydrogen atoms of the ethyl ester. The hydrogen atom adjacent to the phthalazine ring nitrogen gives rise to a singlet at low field with a chemical shift of about δ 9.6 ppm. The mass spectrum exhibits the typical fragmentation of an ester group leading to the ions [M.⁺.OEt] and [M.⁺.COOEt]. The most intense peak of the pyrazoles **7a-e**, however, appears for the fragment ion with m/z = 293. This ion is formed by cleavage of the aryl group from the ionized molecule (see experimental for further fragmentation pattern). However, we found that if **1** was reacted as free base with an equimolar amount of **6a** (R=H), no pyrazole was formed. Instead, only a highly insoluble purple

compound could be isolated. Based on elemental analysis and mass spectral data, the compound seems to be a bis-condensation product formed by condensation of 1 with ethyl cinnamoylpyruvate 6a in a 2:1 ratio. According to its analytical data, structure 8a or its tautomeric one 9 was tentatively assigned to this product. To avoid bis-condensation to occur, the p-methoxy substituted 6b was chosen since it was thought that the electron donating ability of the methoxy group in p-position would decrease the electrophilicity of the γ -carbonyl C-atom of 6. The outcome of this experiment seems to confirm this assumption because the "normal" product 3-[2-oxo-4-(4-methoxyphenyl)but-3-enylidene]-3,4-dihydro-4-oxo-2H-as-triazino[3,4-a]phthalazine 8b was obtained in 64% yield.



In conclusion, it can be stated that these results of the reactions between 1 and a number of 1,2,4-tricarbonyl compounds can again best be rationalized on the high nucleophilicity of the N-3 atom, a feature of hydralazine reactivity which was mentioned earlier by us and others.^{10,15} Moreover, these results again point to the high tendency of 1 to annelate a third ring only as a five- or six-membered moiety.⁶

EXPERIMENTAL

General: Melting points were determined with a Mel-Temp melting point apparatus and are uncorrected. IR spectra were recorded using a Perkin-Elmer Model 599 spectrometer calibrated against the 1601 cm^{-1} band of polystyrene. ^1H -nmr spectra were recorded on a Varian T-60 spectrometer or a Nicolet NT300 narrow-bore spectrometer (300 MHz) with a 1180-E data system. Chemical shifts are expressed in δ relative to tetramethylsilane as internal standard. Mass spectral data were obtained on a Perkin-Elmer RMU-7 mass spectrometer and/or a Kratos MS-80 instrument with a DS-55 data system. Ultraviolet spectra were recorded on a Unicam SP800 spectrometer. Elemental analyses were performed at M-H-W Laboratories, Phoenix, Arizona.

General Procedure for the Preparation of 3-[2-Oxo-2-(3- or 4-substituted phenyl)ethyl]-4H-as-triazino[3,4-a]phthalazin-4-one (4a-b)

The appropriate ethyl benzoylpyruvate (0.004 mole) and 0.004 mole of hydralazine were refluxed for 30 min. The solid that separated was filtered off after cooling and recrystallized from a suitable solvent.

3-[2-Oxo-2-(4-methoxyphenyl)ethyl]-4H-as-triazino[3,4-a]phthalazin-4-one(4a)

Yield 69%, mp 270-274°C (EtOH/CHCl₃); ¹H-nmr (CDCl₃): δ 3.90 (s, 3H, -OCH₃), 4.76 (s, 1H, -CH₂-) 6.88 (s, 1H, =CH-), 6.98, 7.27, 7.73, 8.05, 8.37 (m,s,m,m,m, 9H, ArH), 14.2 (s, 1H, NH) ppm; uv(CHCl₃): 476 (18,600), 448 (30,700), 424 (34,200), 294 (21,600), 283 (19,900) nm; ir (KBr): 1720, 1606, 1580 cm⁻¹; ms: 346 (M⁺, 16), 345 (26), 318 (4), 317 (16), 239 (3), 238 (7), 211 (5), 175 (19), 149 (17), 136 (12), 135 (94), 129 (20), 128 (100) m/z; Anal. calcd. for C₁₉H₁₄N₄O₃: C, 65.89; H, 4.07; N, 16.18. Found: C, 66.00; H, 4.19; N, 16.15.

3-[2-Oxo-2-(4-methylphenyl)ethyl]-4H-as-triazino[3,4-a]phthalazin-4-one (4b)

Yield 49%, mp 259-261°C (DMF); ¹H-nmr (CDCl₃): δ 2.43 (s, 3H, -CH₃), 4.78 (s, 0.2 H, -CH₂-) 6.87 (s, 0.9 H, =CH-), 7.29, 7.74, 7.97, 8.40 (m,m,m,m, 9H, ArH), 14.25 (s, 0.9H, NH) ppm; uv (CHCl₃): 475 (16,500), 446 (25,000), 424 (26,600), 294 (18,700), 283 (16,900) nm; ir (KBr): 1720, 1602, 1580 cm⁻¹; ms: 330 (M⁺, 51), 329 (75), 302 (6), 301 (22), 211 (4), 170 (12), 135 (9), 102 (8) m/z; Anal. calcd. for C₁₉H₁₄N₄O₂: C, 69.08; H, 4.27; N, 16.96. Found: C, 69.15; H, 4.37; N, 16.75. Exact mass calcd. for C₁₉H₁₄N₄O₂: 330.1117; Found: 330.1102.

3-[2-Oxo-2-(3-methoxyphenyl)ethyl]-4H-as-triazino[3,4-a]phthalazin-4-one (4c)

Yield 50%, mp 257-259°C (DMF); ¹H-nmr (CDCl₃): δ 3.94 (s, 3H, H₃CO-), 6.94 (s, 1H, =CH-), 7.12, 7.31, 7.43, 7.75, 8.47 (m,s,m,m,m, 9H, ArH) 14.33 (s, 1H, NH) ppm; uv (CHCl₃): 477 (21,400), 449 (31,200), 426 (31,700), 294 (20,200), 283 (19,400) nm; ir (KBr): 1722, 1595, 1580 cm⁻¹; ms: 346 (M⁺, 62), 345 (100), 318 (5), 317 (39), 251 (9), 239 (6), 211 (6), 175 (20), 163 (57), 135 (48), 129 (30) m/e; Anal. calcd. for C₁₉H₁₄N₄O₃: C, 65.89; H, 4.07; N, 16.18. Found: C, 65.82; H, 4.22; N, 15.99. Exact mass calcd. for C₁₉H₁₄N₄O₃: 346.1066; Found: 346.1069.

3-[2-Oxo-2-(4-nitrophenyl)ethyl]-4H-as-triazino[3,4-a]phthalazin-4-one (4d)

Yield 59%, mp 315-316°C (DMF); uv (CHCl₃): 486 (32,800), 458 (39,200), 434 (31,200), 292 (25,600), 283 (26,100) nm; ir (KBr): 1728, 1600, 1570 cm⁻¹; ms: 361 (M⁺, 22), 360 (12), 332 (2), 311 (14), 293 (21), 283 (21), 239 (13), 238 (7), 163 (55), 135 (16), 113 (90); Anal. calcd. for C₁₈H₁₁N₅O₄: C, 59.84; H, 3.07; N, 19.38. Found: C, 59.66; H, 3.33; N, 19.47.

3-Carboethoxy-s-triazolo[3,4-a]phthalazine (5)

The appropriate substituted ethyl benzoylpyruvate (0.004 mole) and 0.78 g (0.004) l-HCl were refluxed in 20 ml of ethanol for 1.5 h. While hot, an orange precipitate which was identified as the corresponding type-4 compound was filtered off, and the filtrate concentrated. Upon cooling colorless crystals of 5 deposited, mp 235-237°C (MeOH). ¹H-nmr (Me₂SO-d₆): δ 1.43 (t, 3H, (CH₃CH₂O-), 4.5 (q, 2H, CH₃CH₂O-), 8.13-8.73 (m, 4H, ArH), 9.3 (s, 1H, N=CH); ir (KBr): 1730 cm⁻¹; ms: 242 (11.46), 197 (16.27), 171 (10.26), 170 (46.70), 142 (7.42), 141 (6.33), 129 (8.33), 115 (100.00), 114 (20.90), 102 (6.89), 88 (16.51), 63 (10.91); Anal. calcd. for C₁₂H₁₀N₄O₂: C, 59.50; H, 4.16; N, 23.13. Found: C, 59.42; H, 4.23, N, 23.08.

General Procedure for the Preparation of 1-(1-Phthalazinyl)-3-ethoxycarbonyl-5-(3- or 4-substituted styryl)-pyrazole (7a-e)

A mixture of 0.004 mole of the substituted ethyl cinnamoylpyruvate and 0.78 g (0.004 mole) of hydralazine

hydrochloride in 20 ml of ethanol were refluxed for 1.5 h. After cooling the dark precipitate was filtered off and recrystallized.

1-(1-Phthalazinyl)-3-ethoxycarbonyl-5-styrylpyrazole (7a)

Yield 53%, mp 254°C (EtOH); $^1\text{H-nmr}$ (CDCl_3): δ 1.43 (t, 3H, $\text{CH}_3\text{-CH}_2\text{-O}$); 4.48 (q, 2H, $\text{CH}_3\text{-CH}_2\text{-O}$), 7.17, 7.30, 8.00 (s, m, m, 12H, 2=CH and ArH), 9.60 (s, 1H, -N=CH of phthalazine ring); ir (KBr) 1710 cm^{-1} . Ms m/z calcd. for $[\text{M}^+, (\text{C}_{22}\text{H}_{18}\text{N}_4\text{O}_2)^+]$ 370.14314. Found: m/z 370.1418.

1-(1-Phthalazinyl)-3-ethoxycarbonyl-5-(4-methoxystyryl)pyrazole (7b)

Yield 40%, mp 185-186°C (EtOH/ CHCl_3); $^1\text{H-nmr}$ (CDCl_3): δ 1.43 (t, 3H, $\text{-OCH}_2\text{CH}_3$), 3.80 (s, 3H, -OCH_3), 4.50 (q, 2H, $\text{-OCH}_2\text{CH}_3$), 6.75-7.43, 8.07 (m, m, 11H, -CH=CH- , ArH), 9.60 (s, 1H, -N=CH-) ppm; uv (EtOH): 300 (24,000), 221 (50,500) nm; ir (KBr): 2994, 2836, 1721, 1638, 1611, 1583 cm^{-1} ; ms: 400 (M^+ , (100), 355 (5), 343 (18), 327 (12), 294 (19), 293 (96), 265 (14), 261 (18), 221 (9), 200 (10), 140 (12), 129 (15) m/z. Anal. calcd. for $\text{C}_{23}\text{H}_{20}\text{N}_4\text{O}_3$: C, 68.99; H, 5.03; N, 13.99. Found: C, 68.87; H, 5.09; N, 13.80.

1-(1-Phthalazinyl)-3-ethoxycarbonyl-5-(4-chlorostyryl)pyrazole (7c)

Yield 49%, mp 152-155°C (EtOH/ CHCl_3); $^1\text{H-nmr}$ (CDCl_3): δ 1.47 (t, 3H, $\text{-OCH}_2\text{CH}_3$), 4.50 (q, 2H, $\text{-OCH}_2\text{CH}_3$), 7.17-7.37, 8.07 (m, m, 11H, -CH=CH- , ArH), 9.62 (s, 1H, -N=CH-) ppm; uv (EtOH): 298 (29,000), 221 (51,400) nm; ir (KBr): 1716, 1631 cm^{-1} ; ms: 404 (M^+ , 52), 403 (10), 359 (5), 331 (7), 294 (23), 293 (100), 265 (12), 221 (9), 153 (9), 129 (13), 103 (9), 102 (9) m/z. Anal. calcd. for $\text{C}_{22}\text{H}_{17}\text{N}_4\text{O}_2\text{Cl}$: C, 65.27; H, 4.23; N, 13.84. Found: C, 65.09, H, 4.41; N, 13.77. Exact mass for $\text{C}_{22}\text{H}_{17}\text{N}_4\text{O}_2\text{Cl}$: 404.1040. Found: 404.1010.

1-(1-Phthalazinyl)-3-ethoxycarbonyl-5-(3-methoxystyryl)pyrazole (7d)

Yield 39%, mp 202-204°C (EtOH/ CHCl_3); $^1\text{H-nmr}$ (CDCl_3): δ 1.43 (t, 3H, $\text{-OCH}_2\text{CH}_3$), 3.77 (s, 3H, -OCH_3), 4.48 (q, 2H, $\text{-OCH}_2\text{CH}_3$), 6.87-7.35, 8.05 (m, m, 11H, -CH=CH- , ArH), 9.58 (s, 1H, -N=CH-) ppm; uv (EtOH): 290 (11,700), 221 (26,000) nm; ir (KBr): 2988, 2830, 1719, 1635, 1599 cm^{-1} ; ms: 400 (M^+ , 17), 327 (3), 293 (87), 265 (13), 129 (8), 128 (11) m/z. Anal. calcd. for $\text{C}_{23}\text{H}_{20}\text{N}_4\text{O}_3$: C, 68.99; H, 5.03; N, 13.99. Found: C, 68.86, H, 5.13; N, 14.09.

1-(1-Phthalazinyl)-3-ethoxycarbonyl-5-(4-nitrostyryl)pyrazole (7e)

Yield 32%, mp 272-274°C (EtOH/ CHCl_3); $^1\text{H-nmr}$ (CDCl_3): δ 1.47 (t, 3H, $\text{-OCH}_2\text{CH}_3$), 4.52 (q, 2H, $\text{-OCH}_2\text{CH}_3$), 7.25-7.59, 8.01-8.32 (m, m, 11H, -CH=CH- , ArH), 9.66 (s, 1H, -N=CH-) ppm; uv (EtOH): 339 (20,300), 221 (43,100) nm; ir (KBr): 1736, 1593 cm^{-1} ; ms: 415 (M^+ , 13), 370 (2), 342 (2), 295 (8), 294 (21), 293 (100), 265 (15), 221 (8), 129 (6). Exact mass calcd. for $\text{C}_{22}\text{H}_{17}\text{N}_5\text{O}_4$: 415.1280. Found: 415.1297.

3-(2-Phthalazinylhydrazono-4-phenylbut-3-enylidene)-3,4-dihydro-4-oxo-2H-as-triazino[3,4-a]phthalazine (8a) or Its Tautomer 9

A solution of 1 g of 1 in 20 ml of ethanol was combined with a solution of 1.54 g of 5a in 30 ml of ethanol and refluxed for 2 h. The resulting purple-colored solid was filtered, washed with ethanol and dried. Yield 93% (based on 1); mp 285-287°C (DMF); ir (KBr) 3380, 1696 cm^{-1} ; ms 484 (M^+ , 6.38), 271 (100), 212 (36.10), 184

(12.01), 145 (14.26), 129 (26.55), 128 (31.17), 116 (14.35), 115 (50.47), 114 (15.13), 103 (15.49), 102 (22.95), 89 (23.51), 88 (22.56), 77 (14.37), 76 (16.02), 75 (18.19), 64 (9.42), 63 (14.37), 62 (16.41), 51 (19.22), 50 (18.69). Anal. calcd. for $C_{28}H_{20}N_8O$: C, 69.41; H, 4.16; N, 23.13. Found: C, 69.31; H, 4.27; N, 23.35.

3-[2-Oxo-4-(4-methoxyphenyl)but-3-enylidene]-3,4-dihydro-4-oxo-2H-as-triazino[3,4-a]phthalazine (8b)

Ethyl p-methoxycinnamoylpyruvate, 1.1 g (0.004 mole) and 0.64 g (0.004 mole) of **1** were each dissolved in a small amount of hot ethanol. The two solutions were mixed and the resulting mixture allowed to stand overnight at room temperature. The red precipitate that formed was filtered off and recrystallized from $CHCl_3$. Yield 64%, mp 231-234°C ($CHCl_3$), uv ($CHCl_3$): 494 (31,100), 464 (39,100), 441 (33,000), 293 (19,700), 284 (18,600) nm; ir (KBr): 1708, 1635, 1592, 1568 cm^{-1} ; ms: 372 (M^+ , 7), 371 (2), 265 (27), 238 (19), 161 (10), 135 (16), 134 (57), 119 (13). Anal. calcd. for $C_{21}H_{16}N_4O_3$: C, 67.73; H, 4.33; N, 15.05. Found: C, 67.77; H, 4.38; N, 14.89.

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NOTES AND REFERENCES

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