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Synthesis of Some New Heterocyclic Compounds Derived from 2-Amino-4-Hydrazino-6-Substituted Pyrimidines

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A number of some new heterocyclic compounds containing 2-amino-6-substituted pyrimidin-4-yl moiety have been synthesized from reaction of 2-amino-4-hydrazino-6-substituted pyrimidines (I c, d) with some condensation cyclization reagents. Their structures have been established on the basis of elemental analysis and spectral data.

INTRODUCTION

In view of the significant biological activities of pyrimidines[1] having an amino or a (substituted)-amino group at C-2 or C-4 position, we have synthesized some new pyrimidine derivatives from 2-amino-4-hydrazino-6-substituted pyrimides, and it therefore appears likely that the new synthesized pyrimidine derivatives will exhibit interesting biological properties.

RESULTS AND DISCUSSION

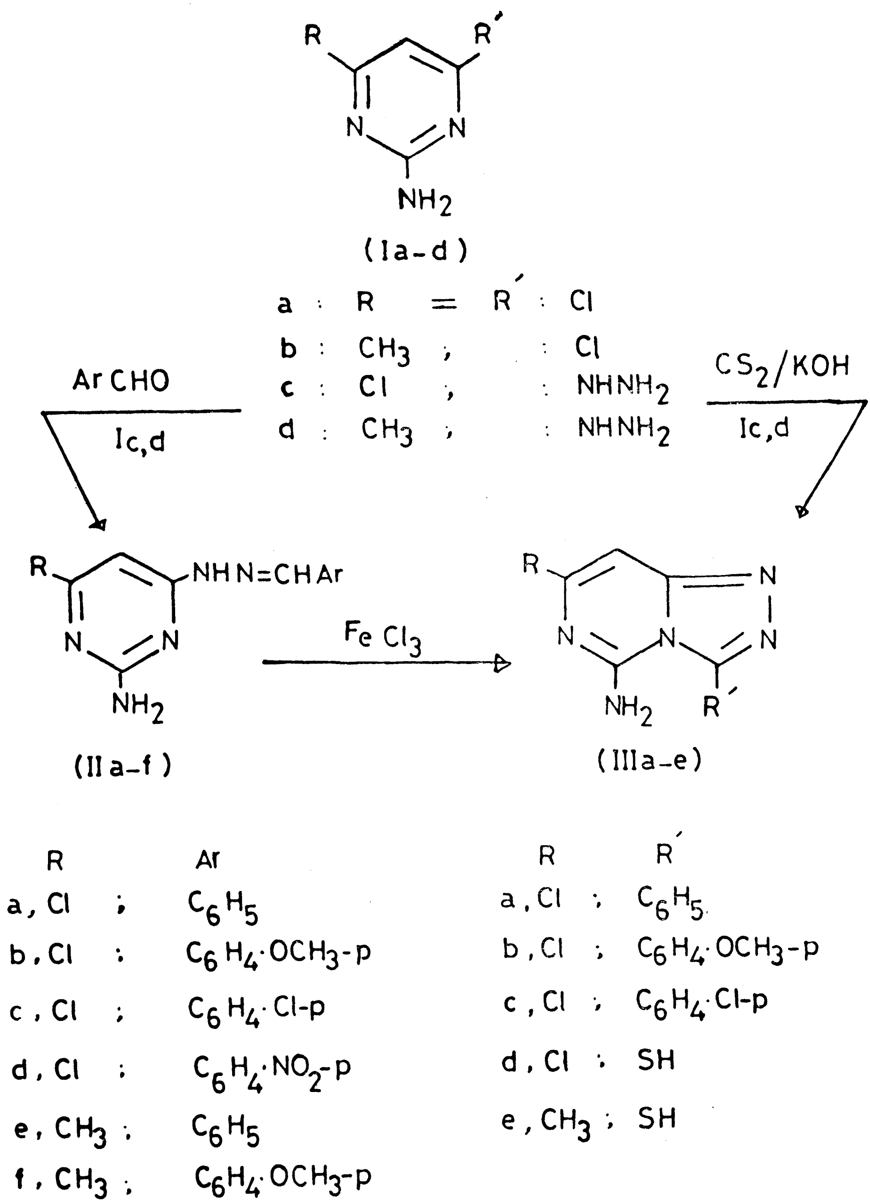
Treatment of 2-amino-4-hydrazino-6-substituted pyrimidines[2] (I c,d) with aromatic aldehydes gave the corresponding arylidene-hydrazino-pyrimidines (II a-f) which on oxidative cyclization using $FeCl_3$ yielded the respective fused *s*-triazolo[4,3-c] pyrimidines (IIIa-c), while *s*-triazolo [4,3-c] pyrimidine derivatives (IIId,e)[3] were obtained by refluxing of (Ic) or (Id) with CS_2 in ethanolic KOH (Scheme I).

Addition of allyl/phenyl isothiocyanate to (Ic) or (Id) in absolute ethanol afforded 1-(2′-amino-6′-substituted pyrimidin-4′-yl)-4-allyl/phenyl-3-thiosemicarbazides (IVa-d), respectively. Some of them (IVb,d) underwent cyclocondensation with malonic acid and acetyl chloride giving 1-[2′-amino-6′-substituted pyrimidin-4′-ylimino]-3-phenyl 2,3-dihydro-2-thioxo-4,6(1H,5H)-pyrimidinediones (Va,b), respectively[4]. The presence of methylene group in compound (Va) was established by its condensation with *p*-bromobenzaldehyde in the presence of glacial acetic acid and sodium acetate to give the arylidene derivative (VI).

β-Dicarbonyl reagents[5] such as acetylacetone, benzoylacetone and dibenzoylmethane reacted with (Ic) or (Id) to give the corresponding pyrazolo derivatives (VIIa-f), while the pyrazolo derivative (VIIg) was synthesized from (Id) by the action of ethyl ethoxymethylenecyanoacetate[6]. Moreover, (Ic) on refluxing with a mixture of acetic anhydride and ethyl cyanoacetate[7] afforded 1-[2′-amino-6-chloropyrimidin-4′-yl]-4-carbethoxy-3-methylpyrazole-5(4H)-one (VIII) (Scheme 2).

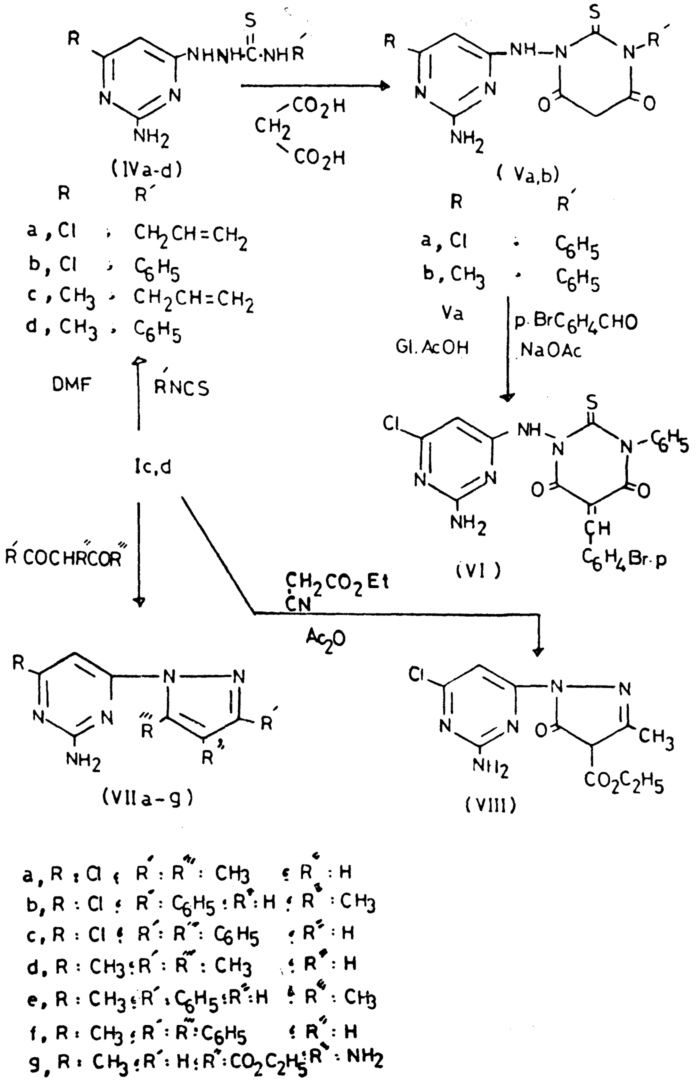
Compound (Ic) or (Id) condensed with α-oxoglutaric acid to give 2-[2′-amino-6′-substituted pyrimidin-4′-yl]-6-carboxy-4,5-dihvdro-3(2H)-pyridazinones (IXa,b), respectively.

SCHEME 1



On the other hand, reaction of compound (Ic) with phthalic anhydride[8] with *o*-acetobenzoic acid[9] in glacial acetic acid led to the formation of 2-[2′-amino-6′-chloropyrimidin-4′-yl]-1,4(2H,3H)-phthalazinedione(X) and 2-[2’-amino-6′-chloropyrimidin-4′-yl]-4-methyl-1(2H)-phthalazinone (XI), respectively.

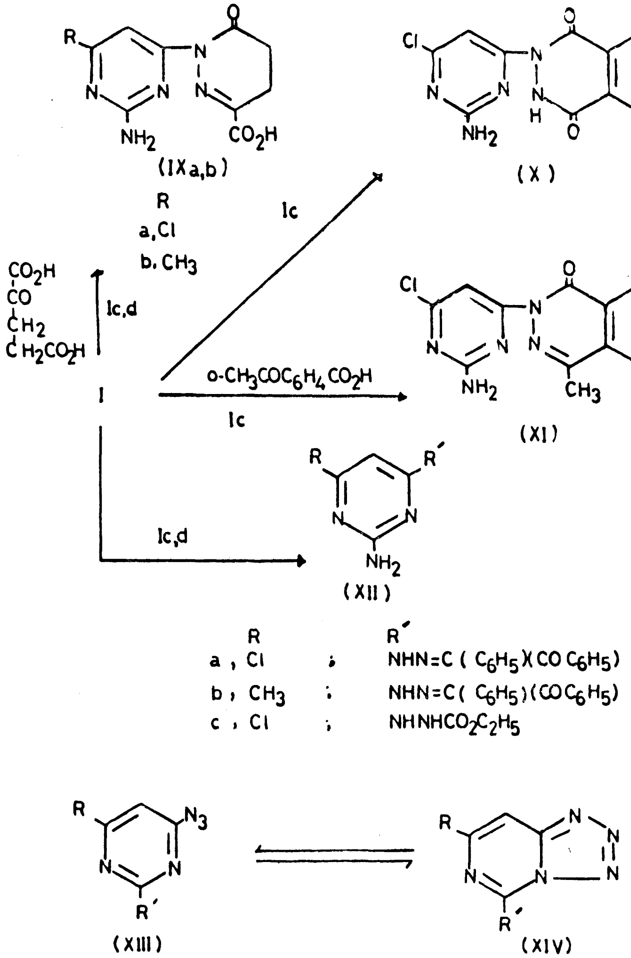
SCHEME 2



Condensation of (Ic) or (Id) with benzil in 20% acetic acid gave the corresponding benzilmonohydrazones (XIIa,b) respectively. Further compound (Ic) when refluxed with ethyl chloroformate in aqueous KOH afforded 2-amino-4-(β-carbethoxyhydrazino)-6-chloropyrimidine (XIIc).

Reactions of 4-hydrazinopyrimidines (Ic,d) with nitrous acid and of 2-amino-4-chloro-6-substituted pyrimidines[2, 10] (Ia,b) with sodium azide in DMF gave azidopyrimidines which may exist as true azides (XIIIa-d) or as tetrazolo[1,5-c] pyrimidines (XIVa-d) in analogy with other related heterocyclic azides[11-13] having the azido group attached to the cyclic carbon atom adjacent to an annular nitrogen atom (Scheme 3). The infrared spectra (KBr) of XIII(a, b and c) exhibited a strong band at 1190 cm^{-1} with the absence of the band which could be assigned to azido function and it was thought to be indicative of the predominance of the proposed tetrazolo structures XIV(a, b and d). However, compound (XIIIc)

SCHEME 3



	R		R′
a	Cl	;	NHN=C(C$_6$H$_5$)(COC$_6$H$_5$)
b	CH$_3$	;	NHN=C(C$_6$H$_5$)(COC$_6$H$_5$)
c	Cl	;	NHNHCO$_2$C$_2$H$_5$

	R		R′
a	Cl	;	OH
b	CH$_3$	;	OH
c	Cl	;	NH$_2$
d	CH$_3$	;	NH$_2$

showed a strong azide band at 2150 cm^{-1} with the absence of the tetrazolo band at 1190 cm^{-1} and this favours the proposed azido structure (XIIIc). In addition, the low intensity of the absorption band (n-π^*) in UV spectra (DMF) at λ_{max} 262 nm is due to the cyclic tetrazolo structure (XIVd), while its intensity in compound (XIIIc) was 350 nm which indicates the acyclic structure of azido derivative XIIIc.

EXPERIMENTAL

Melting points reported are uncorrected. UV spectra were recorded on a Perkin-Elmer 550 S spectrophotometer, IR spectra (KBr) on a Perkin-Elmer 598 spectrophotometer and H^1 nmr spectra on an EM-390 (90 MHz) spectrometer using TMS as internal standard.

2-Amino-4-Hydrazino-6-Methylpyrimidine(Id)

A mixture of (Ib) (0.01 mole) in 50 ml ethanol and hydrazine hydrate (5 ml) was refluxed for 3 hrs. The solid obtained was filtered off and recrystallized from DMF to give (Id) as colourless crystals (yield 95%), m.pt. 240°C. (Calcd: C, 43.17; H, 6.47; N, 50.36; $C_5H_9N_5$, Found: C, 43.10; H, 6.50; N, 50.40).

IR: (Id) 1600 (C=C and C=N), 1650 (δNH_2), 3400–3140 cm^{-1} (NH and NH_2).

2-Amino-4-Arylidenehydrazino-6-Substituted Pyrimidines (IIa-f)

A mixture of (Ic) or (Id) (0.01 mole) and the corresponding aldehydes (0.012 mole) in DMF (30 ml) or acetic acid (20%, 30 ml) was refluxed for 1 hr, the solid obtained was filtered off and recrystallized from the proper solvent to give (IIa-f) as colourless crystals (except IId as yellow crystals).

TABLE 1

Comp.	M.pt. °C	Yield %	Solvent	Formula	Calcd./Fouud C	H	N	Cl
IIa	235	85	Ethanol	$C_{11}H_{10}N_5Cl$	53.33 53.20	4.04 4.10	28.28 28.20	14.34 14.40
IIb	229	72	n-Butanol	$C_{12}H_{12}N_5OCl$	51.89 51.90	4.32 4.30	25.23 24.90	12.79 12.80
IIc	260	80	DMF	$C_{11}H_9N_5Cl_2$	46.81 46 78	3.19 3.20	24.82 24.70	25.18 25.20
IId	264	75	DMF-Water	$C_{11}H_9N_6O_2Cl$	45.13 45.22	3.08 3.10	28.72 28.70	12.14 12.10
IIe	257	70	DMF-Ethanol	$C_{12}H_{13}N_5$	63.44 63.50	5.73 5.70	30.84 30.80	
IIf	183	80	Ethanol	$C_{13}H_{15}N_5O$	60.70 60.60	5.84 5.80	27.24 27.10	

IR: (IIa) 1610–1590 (C=C and C=N), 1660 (δNH_2), 3480–3160 cm^{-1} (NH and NH_2).

5-Amino-3-Aryl-7-Chloro-s-Triazolo [4,3-c] Pyrimidines (IIIa-c)

These were obtained as colourless crystals by oxidative cyclization of the corresponding arylidenehydrazinopyrimidines (IIa-c) with ferric chloride as described earlier[14].

TABLE 2

Comp.	M.pt. °C	Yield %	Solvent	Formula	Calcd./Found			
					C	H	N	Cl
IIIa	290	50	Ethanol	$C_{11}H_8N_5Cl$	53.77 53.70	3.26 3.30	28.51 28.40	14.46 14.50
IIIb	279	45	Ethanol	$C_{12}H_{10}N_5OCl$	52.27 52.30	3.63 3.60	25.41 25.30	12.89 12.80
IIIc	> 300	54	Ethanol	$C_{11}H_7N_5Cl_2$	47.14 47.20	2.50 2.50	25.00 24.90	25.36 25.40

IR: (IIIb), 1600 (C=C and C=N), 1670 (δNH$_2$), 3440–3110 cm^{-1} (NH$_2$).

5-Amino-3(2H)-Thioxo-7-Substituted-s-Triazolo [4,3-c] Pyrimidines (IIId-e)

Compound (Ic) or (Id) (0.01 mole) in ethanol (30 ml), 1.52 g. of carbon disulphide and 0.56 g. of potassium hydroxide in water (5 ml) were refluxed for 3 hrs, the excess ethanol was distilled off, 20 ml of potassium hydroxide solution (5%) was added and filtred. The filtrate on acidification with hydrochloric acid gave (IIId) as colourless crystals and (IIIe) as pale yellow crystals.

TABLE 3

Comp.	m.pt. °C	Yield %	Solvent	Formula	Calcd./Found			
					C	H	N	S
IIId	> 300	72	DMF	$C_5H_4N_5SCl$	29.78 29.80	1.99 2.00	34.74 34.60	15.88 15.80
IIIe	289	75	DMF-Water	$C_6H_7N_5S$	39.78 39.60	3.87 3.90	38.67 38.60	17.68 17.60

IR: (IIId) 1275 (C=S), 1610 (C=C and C=N), 1680 (δNH$_2$) 3280–3130 cm^{-1} (NH and NH$_2$).

1-[2′-Amino-6′-Substituted Pyrimidin-4′-yl]-4-Substituted-3-Thiosemicarbazides (IVa-d)

A mixture of (Ic) or (Id) (0.01 mole) and allyl/phenylisothiocyanate (0.01 mole) in methanol (50 ml) was refluxed for 4 hrs. The solid obtained was filtered off and recrystallized from the proper solvent to give the corresponding thiosemicarbazide derivatives (IVa-d) as colourless crystals (expect IVb as brown crystals).

TABLE 4

Comp.	m.pt. °C	Yield %	Solvent	Formula	Calcd./Found				
					C	H	N	S	Cl
IVa	198	80	DMF-Water	$C_8H_{11}N_6SCl$	37.14	4.26	32 50	12.38	13.73
					37.10	4.30	32.50	12.40	13.70
IVb	207	65	DMF	$C_{11}H_{11}N_6SCl$	44.82	3.74	28.52	10.87	12.05
					44.80	3.70	28.60	10.80	12.00
IVc	215	70	Water	$C_8H_{12}N_6S$	42.86	5.36	37.50	14.29	
					42.90	5.30	37.50	14.30	
IVd	185	75	Ethanol	$C_{12}H_{14}N_6S$	52.55	5.11	30.66	11.68	
					52.60	5.10	30.60	11.70	

UV, λ_{max} DMF nm: (IVa) 260, 324; (IVc) 270, 310; (IVd) 260, 330.

1-[2′-Amino-6′-substituted Pyrimidin-4′-ylimino]-3-Phenyl-2,3-Dihydro-2-Thioxo-4,6 (1H,5H)-Pyrimidinediones (Va,b)

A mixture of IVb or IVd (0.01 mole) and malonic acid (0.02 mole) in acetyl chloride (7 ml) was refluxed for 6 hrs on a steam bath, then poured onto crushed ice. The solid obtained was filtered off and recrystallized from the proper solvent to give (Va) as orange crystals and (Vb) as brown crystals, respectively.

TABLE 5

Comp.	m.pt. °C	Yield %	Solvent	Formula	Calcd./Found				
					C	H	N	S	Cl
Va	> 300	85	Ethanol	$C_{14}H_{11}N_6O_2SCl$	46.34	3.03	23.17	8.83	9.79
					46.30	3.00	23.20	8.80	9.80
Vb	205	70	Ethanol	$C_{15}H_{14}N_6O_2S$	52.63	4.09	24.56	9.36	
					52.60	4.00	24.50	9.40	

IR: (Va) 1250 (C=S), 1650 (δNH_2), 1700 (C=O). 3500-3400 cm^{-1} (NH and NH_2). H^1NMR, $\delta(DMSO\text{-}d_6)$: (Va) 3.5 (s, 2H, CH_2), 6.5-7.8 ppm (m, 9H, NH, NH_2, C_5'-H, C_6H_5).

1-[2′-Amino-6′-Chloropyrimidin-4′-ylimino]-3-Phenyl-5-(p-Bromobenzylidene)-2,3-Dihydro-2-Thioxo-4,6(1H, 5H)-Pyrimidinedione (VI)

A mixture of Va (0.01 mole), *p*-bromobenzaldehyde (0.012 mole) and sodium acetate (0.015 mole) in acetic acid (20 ml) was refluxed for 4 hrs, then poured onto cold water. The solid obtained was filtered off and recrystallized from DMF to give (VI) as yellow crystals, (yield 75%), m.pt. 265°C. (Calcd: C, 47.59; H, 2.64; N, 15.86; S, 6.04; Cl, 6.70; $C_{21}H_{14}N_6SO_2ClBr$, found: C, 47.60; H, 2.60; N, 15.90; S, 6.00; Cl, 6.70).

IR: (VI) 1280 (C=S), 1590 (C=C and C=N), 1650 (δNH_2), 1690 (C=O), 3500–3420 cm^{-1} (NH and NH_2).

1-[2′-Amino-6′-Substituted Pyrimidin-4′-yl]-3,5-Disubstituted Pyrazoles (VIIa-f)

To a solution of (Ic) or (Id) (0.01 mole) in ethanol (50 ml) or acetic acid (20%, 50 ml) was added (0.012 mole) of the appropriate β-dicarbonyl compound (acetylacetone; benzoylacetone; dibenzoylmethane) and the resulting solution was refluxed for 5 hrs. The reaction mixture was poured onto cold water and the solid obtained was filtered off and recrystallized from the proper solvents to give (VIIa-f) as colourless crystals.

TABLE 6

Comp.	m.pt. °C	Yield %	Solvent	Formula	Calcd./Found			
					C	H	N	Cl
VIIa	190	85	Ethanol	$C_9H_{10}N_5Cl$	48.32 48.30	4.47 4.50	31.32 31.30	15.88 15.90
VIIb	179	78	Ethanol	$C_{14}H_{12}N_5Cl$	58.84 58.90	4.20 4.20	24.52 24.50	12.43 12.40
VIIc	200	60	Ethanol	$C_{19}H_{14}N_5Cl$	65.61 65.70	4.03 4.00	20.14 20.10	10.22 10.30
VIId	120	65	Water	$C_{10}H_{13}N_5$	59.11 59.10	6.40 6.40	34.48 34.50	— —
VIIe	196	80	Ethanol	$C_{15}H_{15}N_5$	67.92 68.00	5.66 5.70	26.42 26.40	— —
VIIf	199	70	Ethanol	$C_{20}H_{17}N_5$	73.39 73.40	5.20 5.20	21.41 21.40	— —

IR: (VIIb) 1595 (C=C and C=N), 1650 (δNH_2), 3390-3200 cm^{-1} (NH_2); (VIIc) 1595 (C=C and C=N), 1660 (δNH_2), 3340-3160 cm^{-1} (NH_2).

UV, λ_{max} DMF nm: (VIIC) 270, 305.

H^1NMR, δ (DMSO-d_6): (VIIb) 2(s, 3H, CH_3), 6.5 (s, 1H, C_4-H), 6.7 (s, 1H, C_5'-H), 7 (s, 2H, NH_2), 7.4 ppm (m, 5H, C_6H_5).

1-[2′-Amino-6′-Methylpyrimidin-4′-yl]-5-Amino-4-Carbethoxypyrazole (VIIg)

A mixture of (1d) (0.01 mole) and ethyl ethoxymethylenecyanoacetate (0.01 mole) in absolute ethanol (50 ml) was refluxed for 3 hrs. The solid obtained upon evaporating the excess solvent was recrystallized from ethanol to give (VIIg) as yellow crystals, (Yield 75%), m.pt. 210°C. (Calcd: C, 50.38; H, 5.34; N, 32.06; $C_{11}H_{14}N_6O_2$, found: C, 50.40; H, 5.30; N, 32.10.

1-[2′-Amino-6′-chloropyrimidin-4′-yl]-4-carbethoxy-3-methylpyrazole-5(4H)-one (VIII)

A mixture of 1c (0.01 mole) and ethyl cyanoacetate (0.01 mole) in acetic anhydride (20 ml) was refluxed for 3 hrs, the reaction mixture was cooled, then poured onto cold water and the solid obtained was filtered off and recrystallized from ethanol to give (VIII) as colourless crystals, (yield 55%), m.pt. 250°C. (Calcd: C, 44.37; H, 4.03; N, 23.53; Cl, 11.93; $C_{11}H_{12}N_5O_3Cl$, found: C, 44.40; H, 4.00; N, 23.50; Cl, 12.00).

IR: 1600 (C=C and C=N), 1650 (δ NH_2), 1700–1680 (C=O), 3280–3140 cm^{-1} (NH_2).

2[2′-Amino-6′-Substituted Pyrimidin-4′-yl]-6-Carboxy-4,5-Dihydro-3(2H)-Pyridazinones (IXa,b)

A mixture of (1c) or (1d) (0.01 mole) and α-oxoglutaric acid (0.01 mole) in glacial acetic acid (30 ml) was refluxed for 1/2 h. The solid obtained was filtered off and recrystallized from the proper solvent to give (IXa,b) as colourless crystals.

TABLE 7

Comp.	m.pt. °C	Yield %	Solvent	Formula	Calcd./Found			
					C	H	N	Cl.
IXa	250	73	DMF	$C_9H_8N_5O_3Cl$	40.07 40.10	2.97 3.00	25.97 25.90	13.17 13.20
IXb	270	80	DMF	$C_{10}H_{11}N_5O_3$	48.19 48.20	4.42 4.40	28.11 28.10	

IR: (IXb) 1590 (C=C and C=N), 1650 (δ NH_2), 1720–1680 (C=O), 3200 (NH_2), 3480 cm^{-1} (OH).

Formation of Phthalazinone Derivatives (X) and (XI)

A mixture of (1c) (0.01 mole) and phthalic anhydride or *o*-acetobenzoic acid (0.01 mole) in glacial acetic acid (50 ml) was heated under reflux for 3 hrs, cooled, poured onto cold water and filtered. The solid obtained was filtered off and recrystallized from the proper solvent to give (X) as yellow crystals and (XI) as brownish crysials.

TABLE 8

Comp.	m.pt °C	Yield %	Solvent	Formula	Calcd/Found			
					C	H	N	Cl
X	295	75	DMF	$C_{12}H_8N_5O_2Cl$	49.74 49.70	2.76 2.80	24.18 24.10	
XI	272	80	DMF	$C_{13}H_{10}N_5OCl$	54.26 54.20	3.48 3.50	24.35 24.40	

IR: (X) 1600 (C=C and C=N), 1640 (δ NH_2), 1790–1740 (C=O), 3340 (NH and NH_2), 3480 cm^{-1} (0H).
UV, λ_{max} DMF nm: (X) 270, 320

2-Amino-4-(α-Benzoylbenzylidenehydrazino)-6-Substituted Pyrimidines (XIIa,b)

A mixture of (1c) or (1d) (0.01 mole) and benzil (0.01 mole) in acetic acid (20%, 50 ml) was refluxed for 4 hrs, the reaction mixture was cooled, then poured onto cold water and the solid obtained was filtered off and recrystallized from the proper solvent to give (XIIa,b) as yellow crystals.

TABLE 9

Comp.	m.pt °C	Yield %	Solvent	Formula	Calcd./Found			
					C	H	N	Cl
XIIa	> 300	40	DMF	$C_{18}H_{14}N_5OCl$	61.45 61.50	3.98 4.00	19.91 19.80	10.10 10.00
XIIb	292	50	DMF	$C_{19}H_{17}N_5O$	68.88 68.80	5.14 5.10	21.15 21.00	

IR: (XIIa) 1585 (C=C and C=N), 1640 (δ NH_2), 1660 (C=O), 3340–3120 cm^{-1} (NH and NH_2).

2-Amino-4-(β-Carbethoxyhydrazino)-6-Chloropyrimidine (XIIc)

Compound (1c) (0.01 mole), 1.3 g. of ethyl chloroformate and 0.84 g. of potassium hydroxide in 3 ml. of water were refluxed for 3 hrs, ethanol was distilled off, and the solid obtained was filtered off and recrystallized from DMF to give (XIIc) as colourless crystals (Yield 60%), m.pt. 240°C. (Calcd: C, 36.29; H, 4.32; N, 30.24; Cl, 15.33; $C_7H_{10}N_5O_2Cl$; found: C, 36.30; H, 4.30; N, 30.20; Cl, 15.40).

IR: 1620 (C=C and C=N), 1650 (δ NH_2), 1720 (C=O), 3440–3200 cm^{-1} (NH and NH_2).

Tetrazolo [1,5-c] Pyrimidines (XIVa, b and d)

A solution of (1c) or (1d) (0.01 mole) in acetic acid (5 ml) and water (1 ml) was treated with a solution of sodium nitrite (0.015 mole) in water (2 ml) at 0–5°C. The solid obtained upon diluting the reaction mixture with 10 ml of water was filtered off and recrystallized from ethanol to give (XIVa,b) as colourless crystals. While compound (XIVd) was synthesized by refluxing of a mixture of (Ib) (0.01 mole) in DMF (30 ml) and sodium azide (0.01 mole) in water (2 ml) for 3 hrs, then the reaction mixture was cooled, poured onto cold water and the resulting precipitate was filtered off, recrystallized from DMF to give (XIVd) as colourless crystals.

TABLE 10

Comp.	m.pt °C	Yield %	Formula	Calcd./Found		
				C	H	N
XIVa	230	48	$C_4H_2N_5OCl$	27.99 28.00	1.17 1.20	40.82 40.80
XIVb	215	55	$C_5H_5N_5O$	39.74 39.70	3.31 3.30	46.36 46.40
XIVd	235	70	$C_5H_6N_6$	40.00 39.90	4.00 4.00	56.00 56.10

2-Amino-4-Azido-6-Chloropyrimidine (XIIIc)

A mixture of (1a) (0.01 mole) in DMF (30 ml) and sodium azide (0.01 mole) in water (2 ml) was refluxed for 3 hrs, then poured onto cold water. The solid obtained was filtered off and recrystallized from DMF to give (XIVc) as yellow crystals, (yield 65%), m.pt. 193°C. (Calcd: C, 28.15; H, 1.76; N, 49.27; Cl, 20.82; $C_4H_3N_6Cl$, found: C, 28.20; H, 1.80; N, 49.30; Cl, 20.80).

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