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THE SYNTHESIS OF PYRIMIDOQUINAZOLONES[1]

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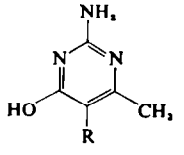
Abstract—The synthesis of a series of 1-amino-3-methyl-(4-alkyl)-pyrimido-[4,3-b]-quinazol-10-ones has been investigated. Using a series of 2-amino-4-chloro-(5-alkyl)-6-methylpyrimidines it has been shown that these compounds condense readily with methyl anthranilate, anthranilic acid and with anthranilamide, either by heating the reactants together or by allowing the reaction to take place in solution in the presence of a trace of acid.

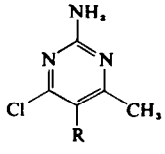
The resulting 2-amino-4-(*o*-substituted-anilino)-(5-alkyl)-6-methylpyrimidines were all obtained in good yields and could be made to undergo ring-closure with formation of the corresponding pyrimido-[4,3-b]-quinazol-10-ones, by the action of heat.

A synthesis of the latter compounds directly from the 4-chloro-pyrimidines and anthranilamide was achieved.

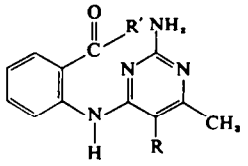
AS PART of a program[2–6] concerned with the synthesis of quinazolones from imidoyl chlorides and anthranilic acid derivatives the results with 2-amino-4-chloropyrimidines (II) are reported. Reaction of II with excess methyl anthranilate at 95° for several hours furnished 2-amino-4-(*o*-carbomethoxyanilino)pyrimidine (III) hydrochloride in high yield. The ester (III) was more conveniently and rapidly obtained in 90–95% yields by refluxing equimolar amounts of the reactants in aqueous ethanol containing a trace of hydrochloric acid; in the absence of the acid catalyst[7] condensation was extremely slow.



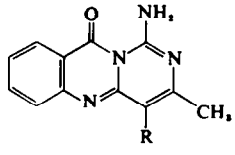
I



II



III, R′ = OCH_3
IV, R′ = OH
V, R′ = NH_2



VI

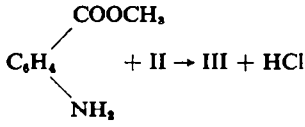
a, R = H; b, R = CH_3; c, R = C_2H_5; d, R = *n*-C_3H_7

When heated above its m.p. III decomposed with loss of methanol and formed 1-amino-3-methylpyrimido-[4,3-b]-quinazol-10-one (VI) in almost quantitative yield. The ester (III) while stable in hot dilute hydrochloric acid was hydrolysed rapidly by more concentrated acid to 2-amino-4-(*o*-carboxyanilino)-pyrimidine (IV). The same compound IV, in the form of its sodium salt, resulted also on heating III with 2N NaOH. In this respect is is noteworthy that the pyrimidoquinazolone (VI) likewise underwent ready hydrolysis in hot strong hydrochloric acid or in 2N NaOH affording

[1] Abstracted from the Ph.D. Dissertation of C. A. R. Hurt, University of the Witwatersrand, October (1952).
[2] P. R. Levy and H. Stephen, *J. Chem. Soc.* 985 (1956).
[3] T. Stephen and H. Stephen, *J. Chem. Soc.* 4173 (1956).
[4] H. Stephen and B. Staskun, *J. Chem. Soc.* 980 (1956).
[5] E. Stephen and H. Stephen, *J. Chem. Soc.* 490 (1957).
[6] H. Stephen and G. Wadge, *J. Chem. Soc.* 4420 (1956).
[7] C. K. Banks, *J. Amer. Chem. Soc.* **66**, 1127 (1944).

IV or the sodium salt of IV respectively. This instability of the "quinazolone" ring to acids precluded the possibility of effecting ring-closure of the ester (III) by means of hot strong hydrochloric acid as can be done with the corresponding N-pyridyl and N-quinolyl derivatives of methyl anthranilate.[3]

On reacting the 4-chloropyrimidine (II) with methyl anthranilate at 165°, methyl chloride was evolved and the compound isolated from the resulting tar proved to be the carboxyanilinopyrimidine (IV) which is considered to arise as follows:



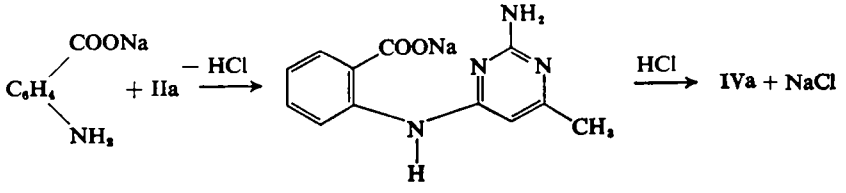
(1) $C_6H_4(COOCH_3)(NH_2) + II \rightarrow III + HCl$

(2) $III + HCl \rightarrow IV + CH_3Cl$

In this respect 2-amino-4-(*o*-carbomethoxyanilino)-6-methylpyrimidine (IIIa) hydrochloride when heated at 165° gave methyl chloride and IVa and a similar result was obtained also in the presence of an equimolar amount of methyl anthranilate. The direct synthesis of the pyrimidoquinazolones (VI) from methyl anthranilate and II could not be achieved because of this effect of the hydrogen chloride produced; employment in the reaction of a suitable dehydrohalogenating agent might however lead to successful results.

Anthranilic acid condensed smoothly and rapidly with the chloropyrimidine (II) at 165° forming the hydrochloride of the corresponding carboxypyrimidine (IV) as the main product without noticeable decomposition of the anthranilic acid taking place. In each case the condensation was accomplished by a subsequent reaction in which part of the product was converted by loss of water into the corresponding pyrimidoquinazolone (VI). When heated *in vacuo* above 200°, the acid (IV) underwent cyclization to VI; however a considerable amount of decomposition occurred and the yields were never much more than 50%. Ring-closure of IV to VI could also be effected by hot acetic anhydride; prolonged reaction however afforded the acetyl derivative of VI.

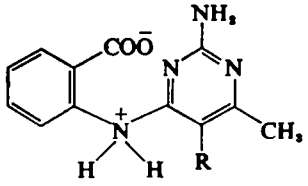
Equimolar amounts of II and anthranilic acid when refluxed in aqueous medium in the presence of a trace of hydrochloric acid as catalyst readily furnished the carboxypyrimidine (IV). In the case of sodium anthranilate which with 2-amino-4-chloro-6-methylpyrimidine (IIa) yielded IVa, the reaction is considered to proceed as indicated below; a Mumm and Hesse type of rearrangement[8] would have lead to a 3-anthranolypyrimidine intermediate which, however, was not formed.



$C_6H_4(COONa)(NH_2) + IIa \xrightarrow{-HCl}$ [intermediate] $\xrightarrow{HCl}$ IVa + NaCl

The carboxypyrimidines (IV) were all very weak acids and did not dissolve readily in sodium carbonate solution possibly because the substances existed as stable internal salts VII—as evidenced also by their relatively high m.ps—and were reactive only towards stronger bases.

[8] O. Mumm, H. Hesse and H. Volquartz, *Ber. Dtsch. Chem. Ges.* **48**, 379 (1915).



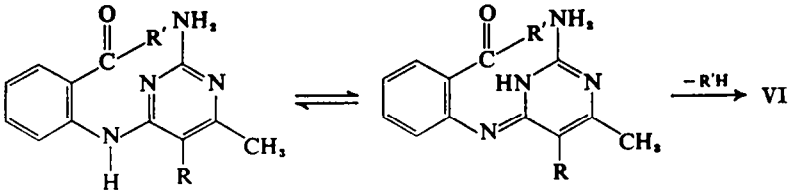
VII

Anthranilamide likewise condensed readily with II at 110° and afforded the hydrochloride of 2-amino-4-(*o*-carbamylanilino)-pyrimidine (V) in high yield. At 150° the carbamylpyrimidine (V) underwent ring-closure to VI quantitatively. The pyrimidoquinazolones (VI) could in fact be synthesized directly in excellent yields by heating together equimolar amounts of anthranilamide and 4-chloropyrimidine (II) at 125°.

The carbamylpyrimidines (V) were fairly stable in hot 2N NaOH and underwent ready hydrolysis in acid solution to the carboxypyrimidine (IV) but could nevertheless be prepared by refluxing anthranilamide and II with very dilute hydrochloric acid provided the reaction time was short; on utilization of ethanol as solvent containing a trace of hydrochloric acid, the hydrochloride of V was quickly deposited in good yield.

Generally, the rate of reaction between the chloropyrimidines (II) and anthranilic acid and its derivatives in solution was found to be increased by the presence of traces of hydrochloric acid and to be inhibited by small amounts of alkali. The acid activation of the chloropyrimidines is adequately explained on the basis of the observations of Banks[7] and others.[9]

The process whereby the compounds III, IV and V undergo ring-closure to VI is considered to involve the following tautomerism:



Thus N-methylanthranilamide was condensed with IIa to give 2-amino-4-(*o*-carbamyl-N-methylanilino)-6-methylpyrimidine; this compound, unlike the corresponding Va, would not undergo ring-closure when heated above its m.p. but was recovered unchanged. 2-Amino-4-(*o*-carbomethoxy-N-methylanilino)-6-methylpyrimidine likewise resisted thermal cyclization. This is considered evidence that a tautomerism involving the 2-amino group in III and V did not occur and that this grouping played no part in the ring-closure reactions.

EXPERIMENTAL[10]

4-Hydroxypyrimidines (I). These compounds were prepared by heating under reflux guanidine carbonate with the appropriate ethyl acetoacetate in EtOH as described in the literature.[11]

2-Amino-4-hydroxy-5-n-propyl-6-methylpyrimidine (Id) was obtained in 55% yield from guanidine carbonate (12·0 g) and ethyl n-propyl-acetoacetate (16·8 g) in EtOH (25 ml) by heating under reflux for 12 hr; crystals from dil EtOH, m.p. 293–294°. (Found: N, 25·06. Calc. for $C_8H_{13}ON_3$: N, 25·06%.)

[9] A. J. Tomisek and B. E. Christensen, *J. Amer. Chem. Soc.* **67**, 2112 (1945); J. S. Morely and J. C. E. Simpson, *J. Chem. Soc.* 1014 (1949).

[10] M.ps are uncorrected. All nitrogen analyses were carried out by the micro Dumas method.

[11] A. Byk, *Ber. Dtsch. Chem. Ges.* **36**, 1915 (1903); J. Jaeger, *Liebigs Ann.* **262**, 365 (1891).

4-Chloropyrimidines (II). The 4-hydroxypyrimidine (I) was heated under reflux with excess $POCl_3$[13] until the evolution of HCl ceased. The unreacted oxychloride was removed by distillation under red. press. and the residue treated with crushed ice and made slightly alkaline with ammonia. The precipitated material was filtered off and recrystallized from EtOH. Compounds Ia (m.p. 182–183°), Ib (m.p. 215–216°), and Ic (m.p. 154–156°) are reported in the literature.

2-Amino-4-chloro-5-n-propyl-6-methylpyrimidine (IId) prepared in 77% yield had m.p. 175–176°. (Found: N, 22·64. Calc. for $C_8H_{13}N_3Cl$: N, 22·64%.)

2-Amino-4-(o-carbomethoxyanilino)-6-methylpyrimidine (IIIa). *A*: Compound IIa(1 mole) and methyl anthranilate (4 moles) were heated together on a water bath for 4 hr. The reaction mixture was cooled and mixed with ether (25 ml) which precipitated the hydrochloride of IIIa. This was filtered off and washed with ether and found to be very soluble in hot water and hot EtOH from which solvents it crystallized as white needles. The crude IIIa hydrochloride was dissolved in warm water and the solution made just alkaline with ammonia when III separated as a white emulsion which soon solidified to a white crystalline mass, m.p. 175–190°, 0·92 g (95%). It crystallized from EtOH or aqueous dioxan in colourless rhombs, m.p. 195–195·5° dec. (Found: N, 21·70; mol. wt., 297. Calc. for $C_{13}H_{14}O_2N_4$: N, 21·71%; mol. wt., 294·5.)

Compound IIa (0·5 g, 1 mole) and methyl anthranilate (1·05 g, 2 moles) were heated together at 165° until all effervescence ceased (about 2 hr). The residue was cooled and extracted with cold dil. NaOHaq leaving a small amount of insoluble tar. Neutralization of the resulting solution with dil. HClaq precipitated IVa (0·68 g, 79%).

Compound IIIa hydrochloride (0·5 g) when heated at 160° melted to a pale brown viscous liquid which effervesced with the evolution of methyl chloride and gradually darkened. After 2 hr, effervescence ceased and the residue was extracted with 2N NaOH, filtered from insoluble tar and the filtrate neutralized with dil. HClaq which precipitated IVa (0·28 g, 68%).

B: Equimolar amounts of IIa (1·0 g) and the ester (1·05 g) were added to water (25 ml) containing EtOH (2 ml) and one drop of conc. HClaq. The mixture was heated under reflux for 15 min and the resulting solution filtered and cooled. The cold filtrate was neutralized with dil. ammonia when crude IIIa separated, 1·79 g (99·5%); m.p. 195 dec. (from EtOH).

Heating the reactants under reflux in water–ethanol containing one drop of 2N NaOH gave no IIIa after 4 hr; the 4-chloropyrimidine (IIa) was recovered unchanged. The following 2-amino-4-(*o*-carbomethoxyanilino)-pyrimidines (III) were obtained using the appropriate 4-chloropyrimidines (II). The method and crude yield are given.

2-Amino-4-(o-carbomethoxyanilino)-5,6-dimethylpyrimidine (IIIb). *A*: Heating for 36 hr gave 85%; *B*: 92%; colourless needles from EtOH, m.p. 203–204° dec. (Found: N, 20·62. Calc. for $C_{14}H_{16}O_2N_4$: N, 20·60%.)

2-Amino-4-(o-carbomethoxyanilino)-5-ethyl-6-methylpyrimidine (IIIc). *A*: After heating for 20 hr, the reaction mixture when diluted with ether gave no precipitate and was extracted with water and the aqueous extract made ammoniacal, yield 88%; *B*: Heating under reflux for 30 min, gave 95%; colourless needles from EtOH, m.p. 183–184°. (Found: N, 19·63. Calc. for $C_{15}H_{18}O_2N_4$: N, 19·57%.)

2-Amino-4-(o-carbomethoxyanilino)-6-methyl-5-n-propylpyrimidine (IIId). *A*: Heating for 40 hr gave 88%; *B*: heating for 40 min under reflux, 90%; crystals from ethanol, m.p. 164–165°. (Found: N, 18·67. Calc. for $C_{16}H_{20}O_2N_4$: N, 18·66%.)

2-Amino-4-(o-carboxyanilino)-6-methylpyrimidine (IVa). *A*: Compound IIa (1·05 g) and anthranilic acid (1·0 g) were added to water (50 ml) containing one drop of conc. HClaq and the mixture heated under reflux for 15 min. Most of the reactants dissolved but before solution was complete a pale brown compound separated. The mixture was cooled and filtered and the residue then dried; 1·62 g (90%), m.p. above 280°. (Found: N, 22·92. Calc. for $C_{12}H_{11}O_2N_4$: N, 22·94%.) The acetyl derivative had m.p. 180°.

Compound IIa (1·0 g, 1 mole) and sodium anthranilate (1·11 g, 1 mole) were added to water (50 ml) containing one drop conc. HClaq and heated under reflux for 30 min. The mixture was cooled and the insoluble material (IVa) filtered off; 1·52 g (89%), m.p. above 280°; acetyl derivative, m.p. 180°. On heating the reactants in water containing one drop of 2N NaOH under reflux for 4 hr, the 4-chloro-pyrimidine (IIa) was recovered unchanged.

B: Equimolar amounts of IIa and anthranilic acid were mixed together and heated at 160–165°. The mixture melted and almost immediately set to a pasty solid which on cooling became hard and

[13] S. Gabriel and J. Coleman, *Ber. Dtsch. Chem. Ges.* **32**, 2921 (1899).

brittle. The product (IVa·HCl) was treated with cold 2N NaOH when practically all dissolved. The solution was filtered from a small amount of a yellow compound (VIa) and neutralized with dil. HClaq when IVa separated in 87% yield.

C: Compound IIIa (1·0 g) was treated with 20% HClaq (5 ml) and boiled for 2 min. A clear solution resulted and almost immediately precipitated a white compound. The mixture was cooled, neutralized with ammonia and the carboxy derivative (IVa), m.p. >280° (acetyl derivative, m.p. 180°), obtained in 92% yield.

D: Compound IIIa (1·0g) was heated with 2N NaOH (10 ml) and EtOH (2 ml) on the water bath for 3 hr, a clear solution resulting. This was neutralized with HClaq when crude IVa separated in 85% yield.

E: Compound VIa on heating under reflux with 20% HClaq or with 2N NaOH likewise afforded IVa in high yield. The following carboxypyrimidines (IV) were prepared from the appropriate starting materials (method and yield given).

2-*Amino*-4-(o-*carboxyanilino*)-5,6-*dimethylpyrimidine* (IVb). *A*: Ethanol (10 ml) containing one drop of conc. HClaq as solvent was heated under reflux for 15 min, 84%; *B*: 88%; *C*: 97%; *D*: 98%; white crystals, m.p. above 280°. (Found: N, 21·71. Calc. for $C_{13}H_{14}O_2N_4$: N, 21·70%.) The acetyl derivative had m.p. 182–183°.

2-*Amino*-4-(o-*carboxyanilino*)-5-*ethyl*-6-*methylpyrimidine* (IVc). *A*: Heating 15 min under reflux gave 86%; *B*: 52%, together with 8% VIc, m.p. 244°; *C*: 95%; *D*: 96%; white crystals, m.p. above 280°. (Found: N, 20·62. Calc. for $C_{14}H_{16}O_2N_4$: N, 20·60%.) The acetyl derivative melted at 177–178°.

2-*Amino*-4-(o-*carboxyanilino*)-6-*methyl*-5-*n*-*propylpyrimidine* (IVd). *A*: Heating 50 min under reflux gave 77%; *B*: at 160°, 23% IVd together with 37% VId; *C*: 90%; *D*: 90%; crystals, m.p. above 280°. (Found: N, 19·62. Calc. for $C_{15}H_{18}O_2N_4$: N, 19·57%.) The acetyl derivative had m.p. 158–159°.

2-*Amino*-4-(o-*carbamylanilino*)-6-*methylpyrimidine* (Va). *A*: Compound IIa (0·5 g) and anthranilamide (0·48 g) were heated at 110°. The mixture formed a clear melt which rapidly set to a pale brown solid. This was cooled, ground up with water and made just alkaline with dil. ammonia. The resulting pale yellow insoluble material (Va) was filtered off and dried; 0·71 g (84%); yellow needles from dioxan, m.p. 253–254°. (Found: N, 28·80. Calc. for $C_{13}H_{13}ON_5$: N, 28·79%.) At 125° the product was 1-amino-3-methylpyrimido-[4,3-b]-quinazol-10-one (VIa), m.p. 274°, in 91% yield (see below).

B: Equimolar amounts of IIa (0·5 g) and amide (0·48 g) were added to water (25 ml) containing one drop HClaq and the mixture heated under reflux. After 5 min, a clear yellow solution was obtained and a small amount of material precipitated. Heating was discontinued immediately, the solution filtered and made alkaline with dil. ammonia, when crude pale yellow Va, m.p. 238° separated in 91% yield.

Heating the acid solution of the reactants under reflux for 1 hr furnished IVa in 88% yield. The latter compound was produced also (98% yield) by boiling Va (0·5 g) with 5 ml 10% HClaq for 1 min. The following carbamyl derivatives (V) were obtained using the appropriate reactants (method and yield given).

2-*Amino*-4-(o-*carbamylanilino*)-5,6-*dimethylpyrimidine* (Vb). *A*: Heating for 10 min gave 97%. Heating at 160° for 2 min the product was VIb in 90% yield. *B*: Heating for 15 min gave 95%; yellow needles from dioxan, m.p. 257–257·5°. (Found: N, 27·21. Calc. for $C_{13}H_{15}ON_5$: N, 27·22%.)

2-*Amino*-4-(o-*carbamylanilino*)-5-*ethyl*-6-*methylpyrimidine* (Vc). *A*: Heating for 10 min gave 87%. Heating at 160° for 2 min the product was 1-amino-4-ethyl-3-methylpyrimido-[4,3-b]-quinazol-10-one in 92% yield. *B*: Heating for 10 min gave 86%; pale yellow needles from dioxan, m.p. 246–246·5°, (Found: N, 25·82. Calc. for $C_{14}H_{17}ON_5$: N, 25·82%.)

2-*Amino*-4-(o-*carbamylanilino*)6-*methyl*-5-*n*-*propylpyrimidine* (Vd). *A*: Heating for 15 min gave 43%. Heating at 130° for 10 min the yield was 83%; at 200° for 2 min the product was 1-amino-3-methyl-4-*n*-propylpyrimido-[4,3-b]-quinazol-10-one in 82% yield. *B*: Heating for 40 min under reflux gave 91%; pale yellow needles from dioxan, m.p. 247–247·5°. (Found: N, 24·54. Calc. for $C_{15}H_{19}ON_5$: N, 24·55%.)

1-*Amino*-3-*methylpyrimido*-[4,3-b]-*quinazol*-10-*one* (VIa). *A*: Compound IIIa (1·0 g) was heated at 200° until all effervescence ceased. The evolved vapour was collected and found to be MeOH.

The residue, a pale brown solid, 0·86 g (98%), crystallized from EtOH in yellow needles, m.p. 272°. (Found: N, 24·76. Calc. for $C_{18}H_{10}ON_4$: N, 24·77%.)

B: Compound IVa (1·0 g) was heated *in vacuo* at 200–220°. Water was evolved and long yellow needles slowly sublimed (0·60 g, 65%) leaving a black carbonaceous residue. The yellow needles were crystallized from dioxan and had m.p. 273°.

C: Compound IVa (1·0 g) was boiled with acetic anhydride (5 ml) until it turned yellow and started to dissolve (2 min). The mixture was cooled immediately, filtered, and the residue extracted with cold dil. NaOHaq. The insoluble residue (0·56 g, 60%) consisted of fine yellow needles of VIa, m.p. 272°. On heating the acetic anhydride solution of IVa under reflux for 15 min and cooling, yellow needles of the acetyl derivative of VIa, m.p. 180°, separated.

D: Compound IIa (1·0 g) and anthranilamide (0·95 g) were heated together at 125°. The mixture melted and quickly set to a pale-brown solid. This was ground up with water and made alkaline with dil. ammonia yielding insoluble yellow crystalline VIa (1·54 g, 91%) m.p. 264–272°; yellow needles from dioxan, m.p. 274°.

E: Compound Va (1·0 g) was heated *in vacuo* at 150° for 45 min. Almost immediately yellow needles started to sublime and the evolution of ammonia was detected. The sublimate melted at 274° and the residue at 268–270° and after recrystallization from dioxan the latter melted at 274°; total yield: 0·88 g (95%). A number of other quinazol-10-ones were prepared from the appropriate reactants (method and yield given).

1-*Amino*-3,4-*dimethylpyrimido*-[4,3-b]-*quinazol*-10-*one* (VIb). *A*: Heating at 210° for 15 min gave 93%; *B*: 59%; *C*: 62%; *D*: Heating at 160° for 2 min gave 90%; *E*: 92%; yellow needles from dioxan, m.p. 282–283°. (Found: N, 23·36. Calc. for $C_{18}H_{12}ON_4$: N, 23·32%.) The acetyl derivative (yellow needles) had m.p. 182–183°.

1-*Amino*-4-*ethyl*-3-*methylpyrimido*-[4,3-b]-*quinazol*-10-*one* (VIc). *A*: Heating at 190° for 15 min gave 91%; *B*: 53%; *C*: 43%; *D*: Heating at 160° for 2 min gave 92%; *E*: 92%; yellow needles from dioxan, m.p. 244–245°. (Found: N, 21·97. Calc. for $C_{14}H_{14}ON_4$: N, 22·04%.) Acetyl derivative: yellow needle, m.p. 177–178°.

1-*Amino*-3-*methyl*-4-*n*-*propylpyrimido*-[4,3-b]-*quinazol*-10-*one* (VId). *A*: Heating at 190° for 30 min gave 85%; *B*: 48%; *C*: 52%; *D*: Heating at 160° for 3 min gave 43%, together with 48% Vd. Heating at 200° for 2 min gave 82% yield of VId. *E*: Heating at 180° for 45 min gave 91%; yellow needles from dioxan, m.p. 223–224°. (Found: N, 20·90. Calc. for $C_{15}H_{16}ON_4$: N, 20·88%.) Acetyl derivative: yellow needles, m.p. 158–159°.

2-*Amino*-4-(o-*carbomethoxy*-N-*methylanilino*)-6-*methylpyrimidine*. Methyl N-methylanthranilate (1·0 g, 1 mole) and IIa (0·87 g, 1 mole) were added to water (25 ml) containing EtOH (2 ml) and one drop conc. HClaq and the mixture heated under reflux for 40 min. The resultant solution was filtered, cooled and neutralized with dil. ammonia when a white compound was precipitated, m.p. 165–170° (1·60 g, 97%); colourless needles from EtOH, m.p. 180–181°. (Found: N, 20·66. Calc. for $C_{14}H_{14}O_2N_4$: N, 20·60%.)

The N-methyl derivative (0·5 g) was heated at 200°; the substance melted but underwent no visible decomposition. After 20 min the material was cooled and crystallized from EtOH as white needles, m.p. 180–181°, identical (mixture m.p.) with the starting compound.

2-*Amino*-4-(o-*carbamyl*-N-*methylanilino*)-6-*methylpyrimidine*. N-methylanthranilamide (1·0 g, 1 mole) and IIa (0·96 g, 1 mole) were dissolved in EtOH (10 ml) containing one drop conc. HClaq and the mixture heated under reflux for 1 hr. The solution was cooled and neutralized with dil. ammonia when white rhombs separated, m.p. 120° (1·54 g, 92%). (Found: N, 27·18. Calc. for $C_{18}H_{16}ON_6$: N, 27·22%.)

The carbamyl derivative (0·5 g) was heated *in vacuo* at 180° for 40 min. The product was crystallized from EtOH as colourless rhombs, m.p. 120°, (0·46 g) which were identical (mixture m.p.) with the starting material.