Monatshefte für Chemie 114, 615—624 (1983)

Monatshefte für Chemie
Chemical Monthly

Syntheses of Some Azolopyridopyrimidines

Andrej Petrič, M. Tišler*, and **Branko Stanovnik**

Department of Chemistry, E. Kardelj University,
YU-61000 Ljubljana, Yugoslavia

(Received 30 December 1982. Accepted 19 January 1983)

Syntheses of isomeric azolopyridopyrimidines are described. As starting material the corresponding pyridopyrimidines were used. It could be established that in many cases a transformation of a functional group with the purpose to form an annelated five-membered ring proceeded with ring opening at the pyrimidine part. Subsequent ring closure with one carbon synthons gave then the desired tricyclic heterocycles.

(Keywords: Cyclization with C—N *and* N—N *bond formation; Tricyclic heterocyclic compounds)*

Synthesen einiger Azolopyridopyrimidine

Synthesen von isomeren Azolopyridopyrimidinen werden beschrieben. Als Ausgangsverbindungen dienten die entsprechenden Pyridopyrimidine. Es wurde beobachtet, daß in manchen Fällen der Ringschluß unter Beteiligung einer geeigneten funktionellen Gruppe zu gleichzeitiger Ringöffnung des Pyrimidinringes führte. Durch nachfolgende Cyclisierungen konnten die entsprechenden tricyclischen Heterocyclen dargestellt werden.

Introduction

As a continuation of our studies on condensed pyrimidines[1–5] it was necessary to prepare some tricyclic systems with an annelated azolo ring on the pyrimidine part. In order to facilitate the investigations of their stability and ring opening reactions only at the N_1—C_2 bond of the pyrimidine ring, we have devised simple blocking of position 4 with an annelated ring, involving also the N_3 atom of the pyrimidine part.

Results and Discussion

An imidazole ring annelated to a heterocycle and with a nitrogen atom common to both rings can be formed in several ways. Most common methods use an aminoheterocycle and an α-halo aldehyde,

0026-9247/83/0114/0615/$ 02.00

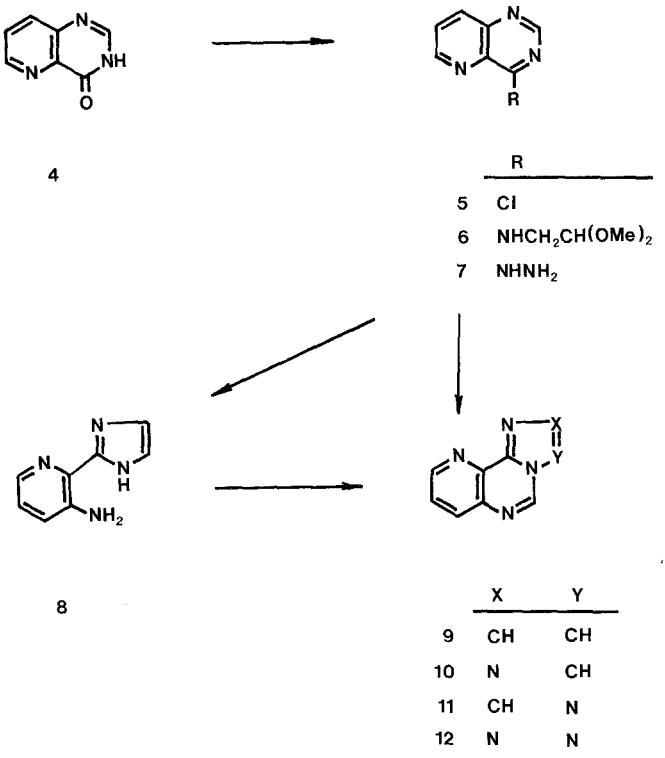
usually in the acetal form, or α-halo ketone. Since this reaction, when applied to some bi- and polycyclic analogs does not give satisfactory results, we have elaborated a new approach: 4-Chloroquinazoline (**1**) was transformed with aminoacetaldehyde dimethyl acetal into the corresponding amino derivative **2** which was subsequently cyclized in the presence of acid into imidazo(1,2-c)quinazoline (**3**) in 85% yield. It should be noted that this compound was prepared previously from 4-chloroquinazoline and ethylene imine[6] or from 2-(*o*-nitrophenyl)-1-hydroxyimidazole 3-oxide[7].

3

	R
1	Cl
2	$NHCH_2CH(OMe)_2$

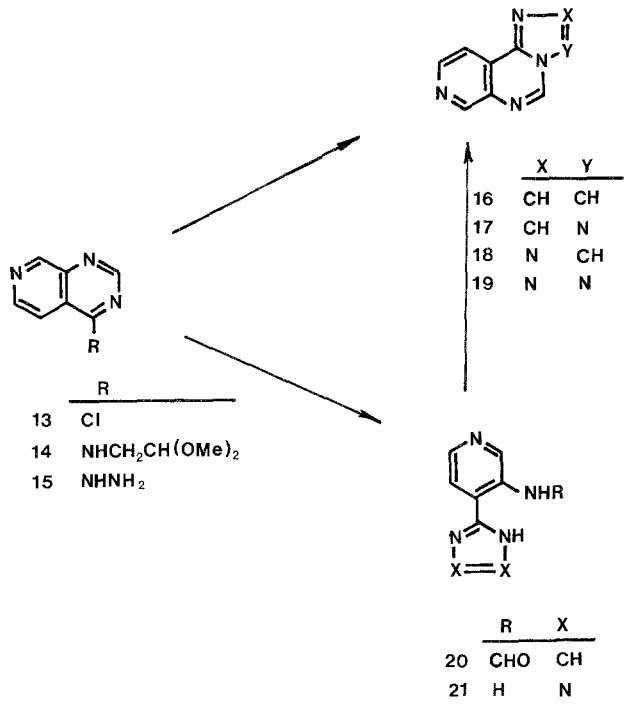
In a similar manner the synthesis of other aza analogs has been performed: 4-Chloropyrido(3,2-d)pyrimidine (**5**) was obtained from the corresponding oxo precursor and oxalyl chloride. This method is superior to the conventional reaction between the oxo compound and phosphorus pentachloride[8] which afforded the chloro compound in very low yield or not at all. The amino derivative **6** was then prepared with aminoacetaldehyde dimethyl acetal, but the attempted cyclization yielded only the corresponding imidazolylpyridine **8**. The formation of this product indicates that the initial cyclization to the imidazo (2,1-f)pyrido(3,2-d)pyrimidine was followed by ring opening of the pyrimidine part of the tricyclic compound. Nevertheless, the obtained derivative **8** could be subsequently cyclized in the presence of triethyl orthoformate to give **9**.

The synthesis of a compound with m.p. 130–131 °C, to which the structure of pyrido(3,2-d)-*s*-triazolo(3,4-f)pyrimidine (**10**) has been assigned, was described without experimental details and with incomplete NMR data[9]. We have now synthesized this tricyclic system, m.p. 286–288 °C, from 4-hydrazinopyrido (3,2-d)pyrimidine (**7**) and triethyl orthoformate in boiling diphenyl ether. The recorded NMR spectrum is in full accord with the tricyclic structure **10** and the



problem of the eventual isomerization of the fused triazolo ring will be discussed later. The isomeric triazolo tricyclic compound **11** could be prepared from the same starting compound, but diethoxymethyl acetate in hot toluene was used instead of triethyl orthoformate. From the chloro compound **5** also the tetrazolo analog **12** was obtained in fair yield.

Also the cyclization of the amine **14** did not proceed straighthforward. The attempted cyclization yielded the tricyclic compound **16** together with the corresponding imidazolylpyrimidine **20** in a ratio of about 2 : 1. This demonstrates again the susceptibility of the pyrimidine part in the tricyclic system for ring opening involving the C_5—N_6 bond. The isomeric pyridotriazolopyrimidines **17** and **18** were both synthesized from 4-hydrazinopyrido(3,4-d)pyrimidine (**15**). Compound **17** was prepared with diethoxymethyl acetate as the precursor for one carbon unit, whereas for the synthesis of **18** triethyl orthoformate in hot diphenyl ether was used. The differentiation between a triazolo ring fused in the 1,2,4- or 1,3,4-manner (cf. **18** and **17**) is possible on hand of



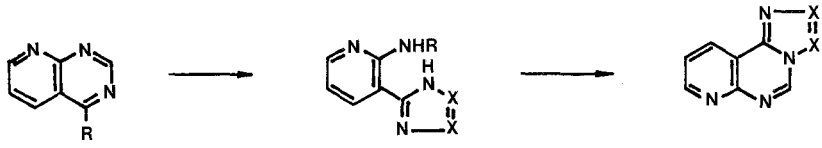
^{1}H NMR spectra. The isomerization can occur as a consequence of a *Dimroth* or ANRORC type rearrangement[10]. The differences in chemical shifts for H_3 of compound **18** and H_2 of compound **17**, when compared with some other related systems (Table 1) clearly demonstrate the correct structural assignment.

Table 1. *Chemical shifts of the isomeric triazoloazines* (δ)

Compound	H_3	H_2
s-Triazolo(4,3-c)quinazoline[22]	9.50	—
s-Triazolo(1,5-c)quinazoline[22]	—	8.60
18	9.27	—
17	—	8.56
10	9.25	—
11	—	8.83
s-Triazolo(4,3-a)pyrimidine[23]	9.28	—
s-Triazolo(1,5-a)pyrimidine[23]	—	8.76

Cyclization with simultaneous ring opening was also observed when 4-chloropyrido(3,4-d)pyrimidine (**13**) was treated with sodium azide in *N*,*N*-dimethylformamide (*DMF*). Instead of the anticipated tricyclic compound **19** a tetrazolylpyridine **21** was obtained. This compound could be used for the synthesis of the tricyclic compound **19** and the ring closure was accomplished with diethoxymethyl acetate as the one carbon synthon.

The instability of the pyrimidine part has been observed also with the isomeric pyrido(2,3-d)pyrimidine system. The attempted cyclization of the amino derivative **23** in hot polyphosphoric acid resulted in the formation of the corresponding imidazolylpyridine **24**. Upon deformylation to **25**, the tricyclic compound **27** could be synthesized in boiling triethyl orthoformate. Similarly, the related tetrazolo analog **28** was prepared from the corresponding tetrazolylpyridine **26**, but using diethoxymethyl acetate as the reagent for cyclization. This tricyclic compound was claimed to be obtained previously from 2-amino-3-(tetrazolyl-5')pyridine and triethyl orthoformate[11], but on hand of lacking experimental description and uncomplete NMR data no firm conclusion about the structure of this product can be made.



	R
22	Cl
23	$NHCH_2CH(OMe)_2$

	R	X
24	CHO	CH
25	H	CH
26	H	N

	X
27	CH
28	N

All these transformations show the relatively high susceptibility of a fused pyrimidine for ring opening and this can be used for the preparation of functionalized heterocycles, this work being in progress.

Acknowledgement

This work has been financially supported in part by the Research Council of Slovenia.

Experimental

Melting points were determined on a *Kofler* hot plate m.p. apparatus. ^{1}H NMR spectra were recorded on a JEOL JNM C-60 HL spectrometer (*TMS* as internal standard, δ-values in ppm) and mass spectra were recorded on a Hitachi-Perkin-Elmer RMU-6L spectrometer. Elemental analysis (C, H, N) are in agreement with the formulas given for **3**, **8**, **9**, **10**, **11**, **12**, **16**, **17**, **18**, **19**, **20**, **21**, **23**, **24**, **26**, **27**, and **28**.

Imidazo(1,2-c)quinazoline (**3**)

A solution of 5 g 4-chloroquinazoline[12] (**1**) in 50 ml of anhydrous benzene was treated with 3.6 g of aminoacetaldehyde dimethyl acetal and 3.5 g of triethylamine and the mixture was stirred at room temperature for 48 h. The solvent was evaporated in vacuo, the residue was dissolved in 50 ml of $CHCl_3$ and the solution washed with water. Upon drying, the solvent was evaporated and the crude 4-(1,1-dimethoxyethylene-2-yl)aminoquinazoline (6.2 g) (**2**) was dissolved in 60 ml of glacial acetic acid, 10 drops of concentrated hydrochloric acid were added and the reaction mixture was heated under reflux for 1 h. The mixture was evaporated to dryness, the residue was treated with 100 ml of water and the mixture was neutralized with solid $NaHCO_3$. The aqueous layer was extracted five times with 50 ml of $CHCl_3$ and from the dried extracts upon evaporation of the solvent 4.38 g (85%) of imidazo(1,2-c)quinazoline (**3**) was obtained. M.p. 140–140.5 °C (Lit.[7] m.p. 127–129 °C). MS (*m*/e): 169 (M^+). NMR ($CDCl_3$): 8.84 (s, H_5), 7.50–8.00 and 8.35–8.60 (m, H_2, H_3, H_7, H_8, H_9 and H_{10}).

2-(Imidazolyl-2′)-3-aminopyridine (**8**)

A suspension of 1 g pyrido(3,2-d)pyrimidin-4(3*H*)-one[13] (**4**) in 25 ml of $CHCl_3$ was treated with 2 drops of *DMF* and 2 ml of oxalyl chloride and the mixture was heated under reflux for 5 h. Upon evaporation to dryness the residue was dissolved in 80 ml of $CHCl_3$ and the solution was shaken with 40 ml of a saturated aqueous solution of Na_2CO_3. The $CHCl_3$ layer was dried and the solvent distilled off. The obtained chloro compound **5** was dissolved in 50 ml of $CHCl_3$, 1.52 g of aminoacetyldehyde dimethyl acetal and 0.732 g of triethylamine were added and the mixture was heated under reflux for 1.5 h. Upon evaporation of the solvent, the residue **6** was treated with 20 ml of glacial acetic acid and 3 drops of concentrated hydrochloric acid and the mixture was heated under reflux for 1 h. The reaction mixture was then evaporated to dryness, a saturated aqueous solution of $NaHCO_3$ was added and the mixture was again heated under reflux for 45 min. Upon cooling, neutralization with hydrochloric acid (1 : 1) and extraction with $CHCl_3$ there were obtained 0.495 g (45%) of the product. NMR ($CDCl_3$): 7.79 (m, H_6), 7.0 (m, H_4 and H_5 and $H_{4'}$ and $H_{5'}$).

Imidazo(2,1-f)pyrido(3,2-d)pyrimidine (**9**)

A mixture of 0.49 g of the above compound **8**, 1 ml of triethyl orthoformate and 3 ml diphenyl ether was heated under reflux for 4 min. The cold reaction mixture was poured into 60 ml of stirred petroleum ether and the separated product filtered to give 0.348 g (67%) of **8**, m.p. 241–244 °C. MS (*m*/e): 170 (M^+). NMR ($CDCl_3$): 7.74 (s, H_2 and H_3), 8.99 (s, H_5), 8.23 (dd, H_7), 7.58 (dd, H_8), 8.97 (dd, H_9), $J_{7,8} = 8.6$, $J_{8,9} = 4.8$, $J_{7,9} = 1.7$ Hz.

Pyrido(3,2-d)-s-triazolo(3,4-f)pyrimidine (**10**)

A mixture of 0.5 g 4-hydrazinopyrido(3,2-d)pyrimidine[14] (**7**) 5 ml of triethyl orthoformate and 15 ml of diphenyl ether was heated under reflux for 7 min, cooled and poured into 200 ml of petroleum ether. The separated product was filtered, washed with petroleum ether and crystallized from *EtOH—DMF*, m.p. 286–288 °C (lit.[8] m.p. 130–131 °C) (0.41 g, 77%). MS (*m/e*): 171 (M^+). NMR (*DMSO-d*$_6$, 150 °C): 9.25 (s, H_3), 9.30 (s, H_5), 8.25 (dd, H_7), 7.72 (dd, H_8), 8.91 (dd, H_9), $J_{7,8} = 8.4$, $J_{8,9} = 4.5$, $J_{7,9} = 1.8$ Hz.

Pyrido(3,2-d)-s-triazolo(5,1-f)pyrimidine (**11**)

The compound was prepared by heating a mixture of 0.25 g of 4-hydrazinopyrido(3,2-d)pyrimidine (**7**)[14], 0.5 g of diethoxymethyl acetate and 10 ml of anhydrous toluene for 2.5 h. The reaction mixture was filtered hot and the filtrate evaporated in vacuo to give 0.191 g (72%) of the product, m.p. 171–174 °C (from toluene). MS (*m/e*): 171 (M^+). NMR (*DMSO-d*$_6$): 8.83 (s, H_2), 9.80 (s, H_5), 8.53 (dd, H_7), 7.99 (dd, H_8), 9.11 (dd, H_9), $J_{7,8} = 8.9$, $J_{8,9} = 3.4$, $J_{7,9} = 1.4$ Hz.

Pyrido(3,2-d)tetrazolo(5,1-f)pyrimidine (**12**)

A mixture of 0.97 g 4-chloropyrido(3,2-d)pyrimidine[15] (**5**), 0.45 g of NaN_3 and 20 ml of *DMF* was stirred at room temperature for 12 h. The solvent was evaporated in vacuo, 50 ml of water were added and the solution was extracted with $CHCl_3$. From the dried $CHCl_3$ extract upon evaporation 0.46 g (46%) of the product was obtained, m.p. 204–207 °C (dec.) (from ethanol). MS (*m/e*): 172 (M^+). NMR (*DMSO-d*$_6$): 10.14 (s, H_5), 8.59 (dd, H_7), 8.04 (dd, H_8), 9.16 (dd, H_9), $J_{7,8} = 8.4$, $J_{8,9} = 4.2$, $J_{7,9} = 1.7$ Hz.

4-Chloropyrido(3,4-d)pyrimidine (**13**)

A mixture of 1 g pyrido(3,4-d)pyrimidin-4(3*H*)-one[16], 1 g of pyridine and 15 ml of $POCl_3$ was heated for 1 h. The mixture was evaporated to a pasty residue and this was poured onto 100 g of ice. Upon extraction with $CHCl_3$ and evaporation of the solvent the chloro compound was obtained (0.622 g, 55%), m.p. 108–110 °C (Lit.[16] m.p. 110–112 °C). NMR ($CDCl_3$) 9.08 (s, H_2), 7.91 (dd, H_5), 8.81 (d, H_6), 9.42 (d, H_8), $J_{5,6} = 5.9$, $J_{5,8} = 0.9$ Hz.

Imidazo(2,1-f)pyrido(3,4-d)pyrimidine (**16**) *and 3-Formylamino-4[2(1H)-imidazolyl]pyridine* (**20**)

To a stirred solution of 0.56 g 4-chloropyrido(3,4-d)pyrimidine in 15 ml of dry benzene a mixture of 0.443 g of aminoacetaldehyde dimethyl acetal and 0.342 g of triethylamine in 5 ml of dry benzene was added. The reaction mixture was heated under reflux for 15 min, the solvent was evaporated, the residue treated with 30 ml of water and extracted with $CHCl_3$. The crude intermediate **14** (0.68 g) was dissolved in 12 ml of glacial acetic acid, 5 drops of concentrated hydrochloric acid were added and the mixture was heated under reflux for 1 h. The reaction mixture was thereafter evaporated in vacuo, the residue was treated with 20 ml of water, neutralized with solid $NaHCO_3$ and extracted with $CHCl_3$. There were obtained 0.2 g (35%) of the tricycle **16**, m.p. 202–205 °C (from ethanol). MS (*m/e*): 170 (M^+). NMR: ($CDCl_3$): 7.66 (s, H_2 and H_3), 8.89 (s, H_5), 9.22 (d, H_7), 8.73 (d, H_9), 8.26 (dd, H_{10}), $J_{9,10} = 5.3$, $J_{7,10} = 0.9$ Hz.

The residue, obtained after extraction, was filtered and crystallized from *Et*OH to give compound **20** in 17% yield (0.11 g), m.p. 239–240 °C. MS (*m/e*): 188 (M^+). NMR (*DMSO-d*$_6$): 8.60 (s, H_2), 7.89 (d, H_5), 8.35 (d, H_6), 9.73 (broad s, CHO), 7.34 (s, $H_{4'}$ and $H_{5'}$), $J_{5,6} = 5.3$ Hz.

Pyrido(3,4-d)-s-triazolo(5,1-f)pyrimidine (**17**)

A mixture of 1 g **15**[17], 4 g of diethoxymethyl acetate and 25 ml of anhydrous toluene was heated under reflux for 1.5 h. The solvent was evaporated and the residue was crystallized from toluene (0.9 g, 85%), m.p. 209–211 °C. MS (*m/e*): 171 (M^+). NMR (*DMSO-d*$_6$, 150 °C): 8.56 (s, H_2), 9.47 (s, H_5), 9.24 (d, H_7), 8.76 (d, H_9), 8.15 (dd, H_{10}), $J_{9,10} = 5.4$, $J_{7,10} = 0.9$ Hz.

Pyrido(3,4-d)-s-triazolo(3,4-f)pyrimidine (**18**)

A mixture of 0.34 g **15**[17], 3 ml of triethyl orthoformate and 4 ml of diphenyl ether was heated under reflux for 5 min. The cold mixture was poured into 50 ml of *n*-hexane and the separated product was filtered and crystallized from *n*-propanol (0.36 g, yield almost quantitative), m.p. 275–287 °C (Lit.[17] m.p. 170 °C). MS (*m/e*): 171 (M^+). NMR (*DMSO-d*$_6$, 132 °C): 9.27 (s, H_3), 9.35 (s, H_5), 9.16 (s, H_7), 8.79 (d, H_9), 8.24 (dd, H_{10}), $J_{9,10} = 8.3$, $J_{7,10} = 0.9$ Hz.

3-Amino-4-(tetrazolyl-5')pyridine (**21**)

A mixture of 1.0 g 4-chloropyrido(3,4-d)pyrimidine (**13**)[16], 0.45 g of NaN_3 and 15 ml of *DMF* was left at room temperature overnight. The solvent was evaporated in vacuo and the residue was treated with water to give crude 3-formylamino-4-(tetrazolyl-5')pyridine. However, if the crude product, obtained after evaporation of *DMF* was treated with 20 ml of saturated aqueous $NaHCO_3$ solution and the mixture heated under reflux for 1 h, cooled and neutralized with aqueous HCl (1 : 1) compound **21** was obtained (0.736 g, 75%), m.p. 294–296 °C (from *Et*OH—*DMF*). MS (*m/e*): 162 (M^+). NMR (*DMSO-d*$_6$, 144 °C): 8.19 (broad s, H_2), 7.65 (dd, H_5), 7.80 (d, H_6), $J_{5,6} = 5.1$, $J_{2,5} = 0.6$ Hz.

Pyrido(3,4-d)tetrazolo(5,1-f)pyrimidine (**19**)

A mixture of 0.58 g of the above compound (**21**), 1.1 g of diethoxymethyl acetate and 15 ml of anhydrous toluene was heated under reflux for 6.5 h. Upon evaporation of the solvent in vacuo the crude product was crystallized from *Et*OH to give the product, m.p. 168–172 °C (0.53 g, 86%). MS (*m/e*): 172 (M^+). NMR (*DMSO-d*$_6$): 10.16 (s, H_5), 9.46 (d, H_7), 8.99 (d, H_9), 8.47 (dd, H_{10}), $J_{9,10} = 5.6$, $J_{7,10} = 0.9$ Hz.

4-(1,1-Dimethoxyethylen-2-yl)aminopyrido(2,3-d)pyrimidine (**23**)

A mixture of 0.95 g 4-chloropyrido(2,3-d)pyrimidine[18] (**22**), 0.66 g of aminoacetaldehyde dimethyl acetal, 0.178 g of triethylamine and 40 ml of anhydrous benzene was heated under reflux for 105 min. The solvent was evaporated, 30 ml of water were added and the mixture was extracted with $CHCl_3$. From the extracts 1.06 g (74%) of **23** were obtained, m.p. 173–176 °C (from *n*-propanol). MS (*m/e*): 250 (M^+). NMR ($CDCl_3$): 8.72 (s, H_2), 8.24 (dd, H_5), 7.26 (dd, H_6), 8.94 (dd, H_7), 3.39 (s, *Me*), 3.78 (dd, CH_2), 4.56 (t, CH), 6.55 (broad t, NH), $J_{5,6} = 8.1$, $J_{6,7} = 4.7$, $J_{CH_2,CH} = J_{CH_2NH} = 4.9$, $J_{5,7} = 1.7$ Hz.

2-Formylamino-3-(imidazolyl-2')pyridine (**24**)

2 g of **22** and 30 g of polyphosphoric acid were heated at 120 °C for 5 h. The cooled mixture was treated with 100 g of ice and neutralized with solid $NaHCO_3$. The separated product was filtered and crystallized from ethanol (1.5 g, 98%), m.p. 167–180 °C with formation of new crystals which had m.p. 206–218 °C. MS (*m/e*): 188 (M^+). NMR (*DMSO-d*$_6$): 8.25 (m, H_4 and H_6), 7.16 (dd, H_5), 7.27 (m, $H_{4'}$ and $H_{5'}$), 9.48 (d, CH), 12.4 (broad d, NHCHO), 12.85 (broad s, NH), $J_{4,5} = 7.8$, $J_{5,6} = 5.1$, $J_{NH,CH} = 9.8$ Hz.

2-Amino-3-(imidazolyl-2')pyridine (**25**)

1 g of **24** and 50 ml of saturated aqueous solution of $NaHCO_3$ were heated under reflux for 30 min. Upon cooling and neutralization with 2 *N* HCl the solution was continuously extracted with $CHCl_3$ for 2 h, the solvent was evaporated and the product crystallized from $CHCl_3$ (0.606 g, 71%), m.p. 167–169 °C. MS (*m/e*) 160 (M^+). NMR (*DMSO-d*$_6$): 7.90 (m, H_4 and H_6), 6.51 (dd, H_5), 7.08 (s, $H_{4'}$ and $H_{5'}$), $J_{4,5} = 7.2$, $J_{5,6} = 5.6$ Hz.

Imidazo(2,1-f)pyrido(2,3-d)pyrimidine (**27**)

0.1 g of **25** and 2.5 ml of triethyl orthoformate were heated under reflux for 1 h and upon cooling the separated product was filtered and crystallized from *n*-propanol (0.08 g, 75%), m.p. 222–225 °C. MS (*m/e*): 170 (M^+). NMR (*DMSO-d*$_6$): 7.53 (d, H_2), 8.00 (d, H_3), 9.33 (s, H_5), 8.81 (dd, H_8), 7.56 (dd, H_9), 8.66 (dd, H_{10}), $J_{2,3} = 1.5$, $J_{8,9} = 4.5$, $J_{9,10} = 7.8$, $J_{8,10} = 2.0$ Hz.

Pyrido(2,3-d)tetrazolo(5,1-f)pyrimidine (**28**)

A mixture of 0.5 g **26**[19] and 1 g of dimethoxyethyl acetate in 15 ml of anhydrous toluene was heated under reflux for 3 h. The mixture was left at room temperature overnight and the separated product was filtered (0.325 g, 61%), m.p. over 164 °C (dec.) (Lit.[11] m.p. 220 °C). NMR (*DMSO-d*$_6$): 10.13 (s, H_5), 7.85 (dd, H_9), 8.86-9.24 (m, H_8 in H_{10}), $J_{9,10} = 8.25$, $J_{8,9} = 4.7$ Hz.

References

1 *Kočevar M., Stanovnik B., Tišler M.*, Heterocycles **15**, 293 (1981).
2 *Verček B., Leban I., Stanovnik B., Tišler M.*, J. Org. Chem. **44**, 1695 (1979).
3 *Verček B., Leban I., Stanovnik B., Tišler M.*, Heterocycles **9**, 1327 (1978).
4 *Kočevar M., Verček B., Stanovnik B., Tišler M.*, Monatsh. Chem. **113**, 731 (1982).
5 *Kočevar M., Tišler M., Stanovnik B.*, Tetrahedron **38** (1982), in print.
6 *Claudi F., Franchetti P., Grifantini M., Martelli S.*, J. Org. Chem. **39**, 3508 (1974).
7 *Franchetti P., Grifantini M.*, J. Heterocyclic Chem. **7**, 1295 (1970).
8 *Armarego W. L. F.*, J. Chem. Soc. **1962**, 4049.
9 *Godefroy L., Queguiner G., Pastour P.*, J. Heterocyclic Chem. **10**, 1077 (1973).
10 *Tišler M.*, Pure & Appl. Chem. **52**, 1611 (1980).
11 *Godefroy L., Decormeille A., Queguiner G., Pastour P.*, C. r. Acad. Sci., Ser. C **278**, 1421 (1974).
12 *Gabriel S., Stelzner R.*, Chem. Ber. **29**, 1300 (1896).
13 *Price C. C., Curtin D. Y.*, J. Amer. Chem. Soc. **68**, 914 (1946).

[14] *Godefroy L., Queguiner G., Pastour P.*, C. r. Acad. Sci., Ser. C **277**, 703 (1973).
[15] The synthesis of 4-chloropyrido(3,2-d)pyrimidine (**5**) is described in the experimental part for compound **8**.
[16] *Gabriel S., Colman J.*, Chem. Ber. **35**, 2831 (1902).
[17] *Decormeille A., Queguiner G., Pastour P.*, Bull. soc. chim. France **1975**, 2757.
[18] *Robins R. K., Hitching G. H.*, J. Amer. Chem. Soc. **77**, 2265 (1955).
[19] 2-Amino-3-(tetrazolyl-5′)pyridine was synthesized from 2-amino-3-cyanopyridine and sodium azide in the presence of glacial acetic acid in *n*-butanol according to the general methods described in the literature[20, 21].
[20] *McManus J. M., Herbst R. H.*, J. Org. Chem. **24**, 1462 (1959).
[21] *Herbst R. M., Wilson K. R.*, J. Org. Chem. **22**, 1142 (1957).
[22] *Potts K. T., Brugel E. G.*, J. Org. Chem. **35**, 3448 (1970).
[23] *Batterham T. J.*, NMR Spectra of Simple Heterocycles, p. 288. New York: Wiley. 1973.