Synthesis of 5-Amino-9-benzyl-6-formyl-4-methoxy-2-pivaloylamino-7,8-dihydropyrimido[4,5-*b*]azepine. A Potentially Useful Intermediate Towards The Synthesis of Pyrimidoazepine Based Folic Acid Derivatives

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Synthesis of the title compound is described *via* an intramolecular 1,3-dipolar cycloaddition reaction between the pyrimidine nitrile oxide **24** and an alkyne dipolarophile tethered to the 6-position of the pyrimidine ring. The resulting isoxazolopyrimidoazepine cycloadduct **25** was treated with molybdenum hexacarbonyl to provide the titled enamino aldehyde **26**.

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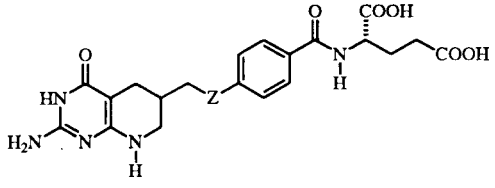
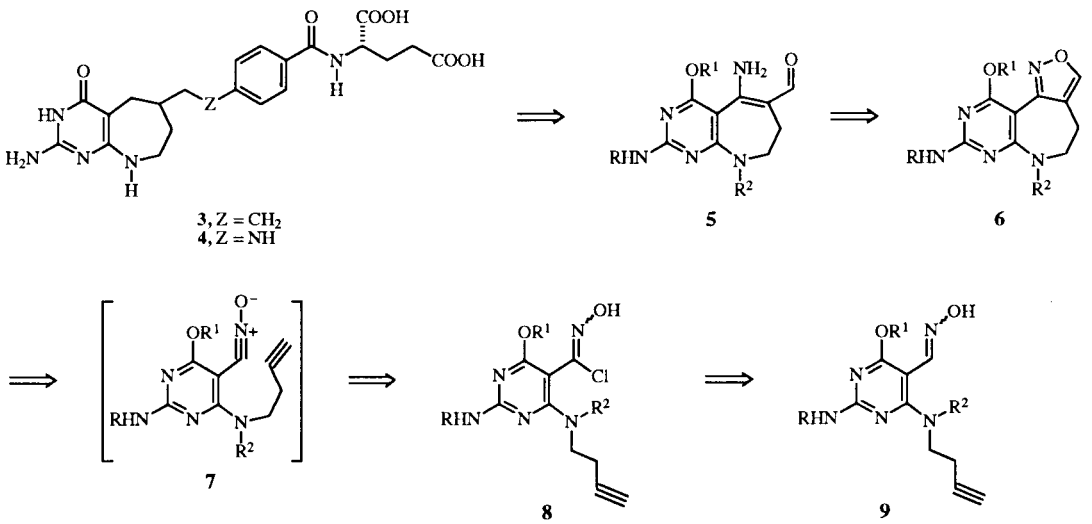
The discovery that the folate antimetabolites 5,10-dideaza-5,6,7,8-tetrahydrofolic acid (**1**) [1] and 5-deaza-5,6,7,8-tetrahydrofolic acid (**2**) [2], exhibit potent antitumor activities against leukemic cells in culture and transplantable murine solid tumors *in vivo* has led to the synthesis and biological evaluation of many related analogs [3]. Both 5,10-dideaza-5,6,7,8-tetrahydrofolic acid and 5-deaza-5,6,7,8-tetrahydrofolic acid exert their cytostatic effects by inhibition of the enzyme glycinamide ribonucleotide formyltransferase which catalyzes the transfer of the formyl group from 10-formyl-5,6,7,8-tetrahydrofolic acid to the amino group of glycinamide ribonucleotide during *de novo* purine biosynthesis. Although 5,10-dideaza-5,6,7,8-tetrahydrofolic acid shows a broad spectrum of antitumor activity in experimental animals it has been reported to be highly toxic in phase I clinical trials [4]. It is therefore of considerable importance to prepare further structurally modified analogs of 5,10-dideaza-5,6,7,8-tetrahydrofolic acid with the aim of discovering a more selective antitumor agent.



1, Z = CH_2
2, Z = NH

We have become interested in preparing novel, ring expanded pyrimidoazepine analogs of 5,10-dideaza-5,6,7,8-tetrahydrofolic acid, **3** and 5-deaza-5,6,7,8-tetrahydrofolic acid **4**. One possible approach to the synthesis of these compounds is from the enamino aldehyde **5**, which should be readily available from the reductive cleavage of the corresponding isoxazole **6**. Retrosynthetic analysis led us to the overall strategy shown in Scheme 1. Thus, we reasoned that the isoxazolopyrimidoazepine **6** should become available *via* an intramolecular 1,3-dipolar cycloaddition reaction between an appropriately substituted pyrimidine-5-nitrile oxide **7** and

Scheme 1



3, Z = CH_2
4, Z = NH

5

6

7

8

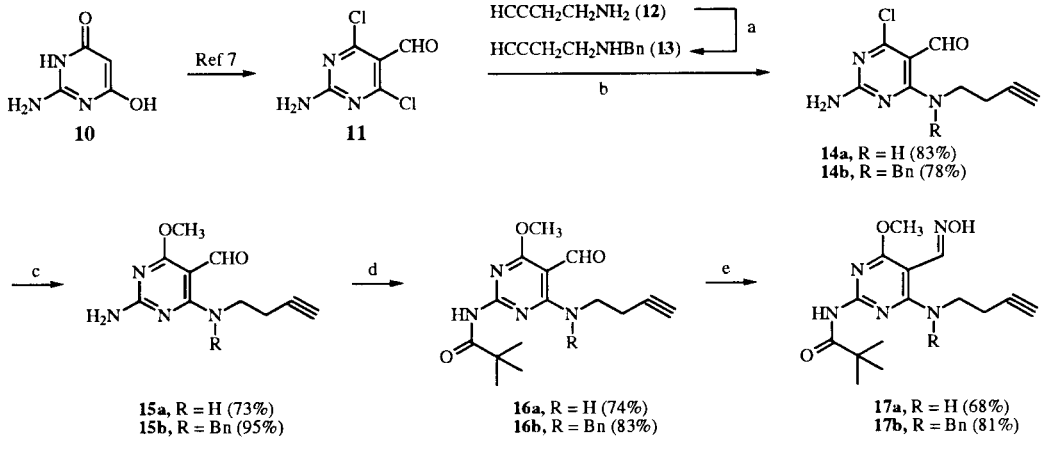
9

an alkyne dipolarophile tethered to the 6-position of the pyrimidine ring [5]. The nitrile oxide **7** could be generated *in situ* from the corresponding oxime **9**, *via* the α-chloro oxime **8**. In this paper we describe the utilization of the above strategy and report the synthesis of the title compound **26**, with which we hope to prepare our desired targets **3** and **4**.

Our synthetic studies began with the conversion of commercially available 3-butyn-1-ol to 4-amino-1-butyne (**12**) as described in the literature [6]. Reaction of 4-amino-1-butyne with 2-amino-4,6-dichloro-5-formylpyrimidine (**11** [7], prepared from 2-amino-6-hydroxypyrimidin-4(3*H*)-one **10**) in the presence of triethylamine in refluxing ethanol gave **14a** in 83% yield (Scheme 2). Reaction of **14a** with sodium methoxide in refluxing methanol gave the methoxy pyrimidine **15a** in 73% yield. Treatment of **15a** with pivalic anhydride in the presence of a catalytic amount of 4-dimethylaminopyridine in refluxing toluene provided the pivaloylamino derivative **16a** in 74% yield and reaction of **16a** with hydroxylamine gave the corresponding oxime **17a** in 68% yield.

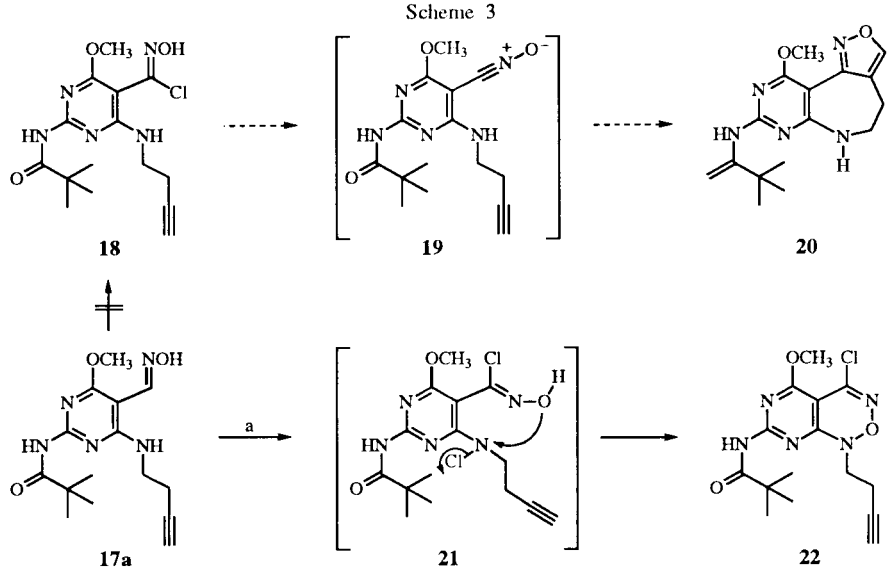
We wanted to convert **17a** to the α-chloro oxime **18** (Scheme 3) with the intention of generating the nitrile oxide **19**, which contains an alkyne dipolarophile tethered to the 6-position of the pyrimidine ring. It was our expectation that an intramolecular 1,3-dipolar cycloaddition reaction between the nitrile oxide and the tethered alkyne would lead to the isoxazole cycloadduct **20**. In the event, however, treatment of the oxime **17a** with either *tert*-butyl hypochlorite or *N*-chlorosuccinimide gave the unexpected product **22**. None of the α-chloro oxime **18** or the

Scheme 2



Reagents: (a) PhCHO, MeOH, rt, 1 hour; then $NaBH_4$ (65%); (b) **12** or **13**, Et_3N, EtOH, Δ; (c) NaOMe, MeOH, Δ; (d) $(Me_3CO)_2O$, toluene, DMAP (cat), Δ; (e) $NH_2OH{\cdot}HCl$, pyridine, EtOH, Δ.

Scheme 3

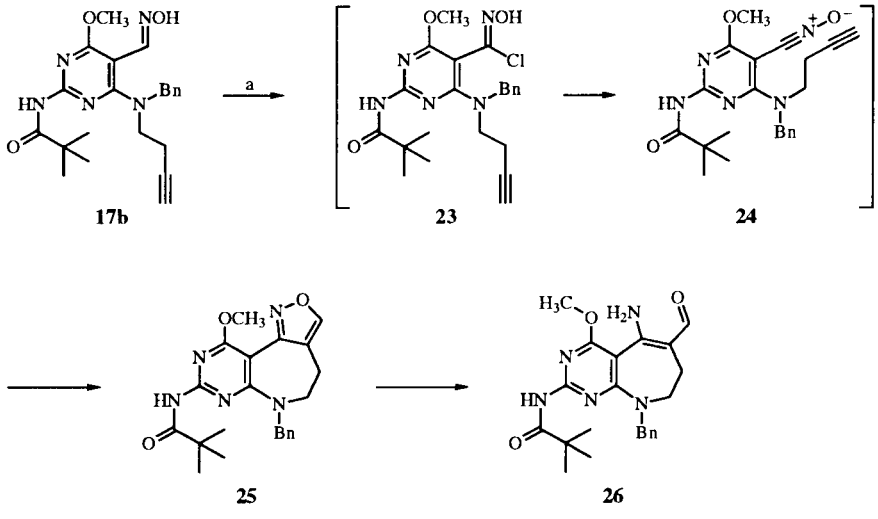


Reagents: (a) NCS, CH_2Cl_2 then Et_3N (45%).

cycloadduct **20** was isolated. Apparently, reaction of **17a** with the chlorinating reagent takes place at the amine nitrogen as well as alpha to the oxime which results in the formation of intermediate **21** which cyclizes *in situ*, as shown in Scheme 3, to give **22**.

We, therefore, decided to protect the offending nitrogen with a benzyl group. Thus, reductive alkylation of 4-amino-1-butyne (**12**) with benzaldehyde followed by treatment with sodium borohydride gave the benzyl derivative **13**. Reaction of 2-amino-4,6-dichloro-5-formylpyrimidine (**11**) with **13** gave **14b** which was converted to the oxime **17b** (Scheme 1) using a similar sequence of reactions as described above for the conversion of **14a** to **17a**. The oxime **17b** was obtained in 50% overall yield starting from **11**. Reaction of **17b** with *N*-chlorosuccinimide followed by treatment with triethylamine gave the isoxazole cycloadduct **25** in 48% yield. We did not attempt to isolate the intermediate α-chloro oxime **23** (Scheme 4). Treatment of **25** with molybdenum hexacarbonyl with hot aqueous acetonitrile resulted in cleavage of the N-O bond of the isoxazole and gave the desired enamino aldehyde **26** in 49% yield after purification by chromatography on silica gel. Further manipulation of **26** to provide our desired targets is currently under investigation.

Scheme 4



Reagents: (a) NCS, CH_2Cl_2 then Et_3N (48%); (b) $Mo(CO)_6$, MeCN, Δ, (49%).

EXPERIMENTAL

Melting points were determined in open capillary tubes using a Thomas-Hoover apparatus and are uncorrected. The proton (300 MHz) and carbon (75 MHz) nmr spectra were recorded on a Varian VXR-300 spectrometer. Chemical shifts are expressed in parts per million (ppm) downfield from internal tetramethylsilane. Column chromatography was performed on Merck silica gel 60 (240-400) mesh; silica gel plates were routinely used for tlc determinations. Elemental analyses were performed by Desert Analytics, Tucson, AZ. High resolution mass spectral (hrms) data were obtained on a VG Analytical 7070 E-HF double focusing mass spectrometer by peak matching technique with known PFK peaks.

2-Amino-6-(3-butynyl-1-amino)-4-chloro-5-formylpyrimidine (**14a**).

A mixture of 2-amino-4,6-dichloro-5-formylpyrimidine [7] (13.04 g, 0.0683 mole), 4-amino-1-butyne [6] (4.7 g, 0.0683 mole), triethylamine (9.5 ml, 0.0683 mole) and ethanol (200 ml) was heated at reflux for 5 hours. The solvent was removed by evaporation under reduced pressure, the residue was stirred in water (200 ml) and the solid was collected by vacuum filtration. The filter cake was dried *in vacuo* over calcium chloride to give 12.57 g (83%) of a pale yellow solid, mp 158-160°. This material was used without further purification; ^{1}H nmr (deuteriochloroform): δ 2.05 (t, 1H, J = 2.6 Hz), 2.51 (dt, 2H, J = 2.6, 6.8 Hz), 3.66 (q, 2H, J = 6.8 Hz), 5.52 (br s, 2H), 9.45 (s, 1H), 10.08 (s, 1H); ^{13}C nmr (deuteriochloroform): δ 19.1, 39.4, 70.3, 81.0, 102.8, 162.2, 162.6, 166.4, 188.9; hrms: ei m/z for $C_9H_9ClN_4O$. Calcd: 224.0477. Found: 224.0465.

2-Amino-6-(3-butynyl-1-amino)-5-formyl-4-methoxypyrimidine (**15a**).

To a freshly prepared solution of sodium methoxide (1.26 g, 0.0548 mole of sodium in 300 ml of anhydrous methanol) was added **14a** (11.0 g, 0.049 mole) and the mixture was heated at reflux under nitrogen for 4.5 hours. The precipitated sodium chloride was removed by filtration and the filtrate was evaporated under reduced pressure. The residue was partitioned between methylene chloride and water and the organic layer was separated, dried over anhydrous magnesium sulfate and the solvent was removed by evaporation under reduced pressure to give 7.8 g (72 %) of a cream colored solid, mp 129-130°, which was used without further purification; ^{1}H nmr (deuteriochloroform): δ 2.04 (t, 1H, J = 2.6 Hz), 2.49 (dt, 2H, J = 2.6, 6.9

Hz), 3.63 (q, 2H, J = 6.9 Hz), 3.91 (s, 3H), 5.28 (br s, 2H), 9.4 (s, 1H), 9.94 (s, 1H); ^{13}C nmr (deuteriochloroform): δ 19.4, 39.3, 53.7, 70.0, 81.5, 93.2, 163.5, 163.6, 172.8, 186.5; hrms: ei m/z for $C_{10}H_{12}N_4O_2$. Calcd: 220.0960. Found: 220.0961.

6-(Butynyl-1-amino)-5-formyl-4-methoxy-2-pivaloylamino-pyrimidine (**16a**).

A mixture of **15a** (7.81 g, 0.0355 mole) trimethylacetic anhydride (8.11 ml, 0.0391 mole), 4-dimethylaminopyridine (50 mg, 4.1 mmoles) and toluene (400 ml) was heated at reflux for 12 hours. The solvent was removed by evaporation under reduced pressure, the residue was triturated with pentane, and the pentane was decanted. The residue was chromatographed on silica gel, eluting with 2% methanol in methylene chloride to give 8.0 g (74%) of a colorless solid, mp 113-114°; ^1H nmr (deuteriochloroform): δ 1.32 (s, 9H), 2.05 (t, 1H, J = 2.6 Hz), 2.53 (dt, 2H, J = 2.6, 6.8 Hz), 3.62 (q, 2H, J = 6.8 Hz), 4.02 (s, 3H), 7.85 (s, 1H), 9.42 (s, 1H), 10.06 (s, 1H); ^{13}C nmr (deuteriochloroform): δ 19.4, 27.4, 39.5, 40.5, 54.4, 70.2, 81.3, 95.1, 158.7, 163.1, 172.8, 175.7, 187.6.

Anal. Calcd. for $C_{15}H_{20}N_4O_3$: C, 59.20; H, 6.62; N, 18.41. Found: C, 59.05; H, 6.40; N, 18.21.

6-(Butynyl-1-amino)-4-methoxy-5-oximinomethyl-2-pivaloyl-aminopyrimidine (**17a**).

A mixture of **16a** (3.94 g, 0.013 mole), hydroxylamine hydrochloride (1.0 g, 0.014 mole), pyridine (1.14 ml, 0.016 mole) and ethanol (50 ml) was heated at reflux for 35 minutes. The solvent and excess pyridine was removed by evaporation under reduced pressure and the residue was partitioned between methylene chloride and 0.1 *N* hydrochloric acid. The organic layer was separated, dried over anhydrous magnesium sulfate and the solvent was removed *in vacuo* to give 2.84 g (68%) of a colorless solid. An analytical sample was obtained by chromatography on silica gel, eluting with 50% ethyl acetate in hexane to give a colorless solid, mp 123-125°; ^1H nmr (deuteriochloroform): δ 1.29 (s, 9H), 1.98 (t, 1H, J = 2.6), 2.48 (dt, 2H, J = 2.6, 6.6 Hz), 3.68 (q, 2H, J = 6.6 Hz), 3.95 (s, 3H), 7.91 (br s, 1H), 8.28 (s, 1H), 8.44 (s, 1H), 8.73 (br s, 1H); ^{13}C nmr (deuteriochloroform): δ 19.5, 27.4, 39.9, 40.4, 54.2, 70.0, 82.0, 88.3, 145.2, 155.8, 160.8, 168.2, 176.2.

Anal. Calcd. for $C_{15}H_{21}N_5O_3$: C, 56.41; H, 6.63; N, 21.93. Found: C, 56.04; H, 6.46; N, 21.67.

1-(3-Butynyl)-3-chloro-4-methoxy-7-pivaloylaminopyrimido-[4,5-*c*][1,2,6]oxadiazine (**22**).

N-Chlorosuccinimide (11 mg, 0.077 mmole) was added to a solution of **17a** (0.25 g, 0.077 mmole) dissolved in anhydrous methylene chloride (30 ml) cooled to 5°. The mixture was stirred for 0.5 hour at 5° and a mixture of triethylamine (0.156 g, 1.544 mmoles) and methylene chloride was added dropwise. The mixture was allowed to warm to room temperature and stirred for a further 1 hour. The reaction mixture was washed with water, dried over anhydrous magnesium sulfate and the solvent was removed *in vacuo*. The residue was chromatographed on silica gel eluting with 50% ethyl acetate in hexane to give 0.11 g (45%) of a colorless solid, mp 171-173°; ^1H nmr (deuteriochloroform): δ 1.33 (s, 9H), 1.93 (t, 1H, J = 2.6 Hz), 2.78 (dt, 2H, J = 2.6, 7.2 Hz), 4.12 (s, 3H), 4.15 (t, 2H, J = 7.2 Hz), 8.0 (br s, 1H); ^{13}C nmr (deuteriochloroform): δ 17.9, 27.4, 40.4, 41.0, 54.9, 71.1, 79.2, 93.2, 111.0, 147.4, 155.0, 161.7, 175.6.

Anal. Calcd. for $C_{15}H_{18}ClN_5O_3$: C, 51.21; H, 5.12; N, 19.92; Cl, 10.10. Found: C, 51.06; H, 5.30; N, 19.93; Cl, 9.92.

N-Benzyl 4-amino-1-butyne (**13**).

4-Amino-1-butyne (4.74 g, 0.0687 mole) was added dropwise to a solution of benzaldehyde (7.3 g, 0.0687 mole) in methanol (50 ml). The mixture was stirred at room temperature for 1 hour, sodium borohydride (5.18 g, 0.137 mole) was added portionwise, and the reaction mixture was stirred an additional 1 hour. Water (50 ml) was slowly added and the mixture was extracted with methylene chloride. The organic layer was dried over anhydrous magnesium sulfate and the solvent was removed by evaporation under reduced pressure to give 7.1 g (65%) of a colorless oil which was used without further purification; ^1H nmr (deuteriochloroform): δ 1.94 (t, 1H, J = 2.6 Hz), 2.34 (dt, 2H, J = 2.6, 7.0 Hz), 2.72 (q, 2H, J = 7.0 Hz), 3.74 (s, 2H), 4.50 (br s, 1H), 7.27 (m, 5H).

2-Amino-6-(*N*-benzyl-*N*-3-butynylamino)-4-chloro-5-formyl-pyrimidine (**14b**).

A mixture of 2-amino-4,6-dichloro-5-formylpyrimidine [7] (10.0 g, 0.0524 mole), *N*-benzyl-4-amino-1-butyne (8.33 g, 0.0524 mole), triethylamine (5.3 g, 0.0524 mole) and ethanol (200 ml) was heated at reflux for 5 hours. The solvent was removed by evaporation under reduced pressure, the residue was stirred in water (200 ml) and the solid was collected by vacuum filtration. The filter cake was dried *in vacuo* over calcium chloride to give 13.0 g (78%) of a pale yellow solid. An analytical sample was obtained by chromatography on silica gel, eluting with 50% ethyl acetate in hexane which gave a colorless solid, mp 96-98°; ^1H nmr (deuteriochloroform): δ 1.96 (t, 2H, J = 2.6 Hz), 2.49 (dt, 2H, J = 2.6, 7.3 Hz), 3.53 (t, 2H, J = 7.3 Hz), 4.73 (s, 2H), 5.92 (br s, 2H), 7.2 (m, 5H), 10.0 (s, 1H); ^{13}C nmr (deuteriochloroform): δ 17.5, 49.2, 54.5, 70.3, 81.1, 105.6, 127.6, 127.7, 128.7, 136.3, 160.9, 163.9, 167.4, 185.2.

Anal. Calcd. for $C_{16}H_{15}ClN_4O$: C, 61.05; H, 4.77; N, 17.81. Found: C, 61.12; H, 4.96; N, 17.75.

2-Amino-6-(*N*-benzyl-*N*-3-butynylamino)-5-formyl-4-methoxy-pyrimidine (**15b**).

To a freshly prepared solution of sodium methoxide (1.1 g, 0.05 mole of sodium in 300 ml of anhydrous methanol) was added **14b** (13.39 g, 0.0426 mole) and the mixture was heated at reflux under nitrogen for 4 hours. The precipitated sodium chloride was removed by filtration and the filtrate was evaporated under reduced pressure. The residue was partitioned between methylene chloride and water and the organic layer was separated, dried over anhydrous magnesium sulfate, and the solvent was removed by evaporation under reduced pressure to give 12.5 g (95%) of a cream colored solid. An analytical sample was obtained by chromatography on silica gel, eluting with 50% ethyl acetate in hexane which gave a colorless solid, mp 105-106°; ^1H nmr (deuteriochloroform): δ 1.94 (t, 1H, J = 2.6 Hz), 2.48 (dt, 2H, J = 2.6, 7.3 Hz), 3.61 (t, 2H, J = 7.3 Hz), 3.94 (s, 3H), 4.78 (s, 2H), 5.17 (s, 2H), 7.22 (m, 5H), 10.0 (s, 1H); ^{13}C nmr (deuteriochloroform): δ 17.5, 48.7, 54.0, 54.2, 69.8, 81.5, 95.7, 127.1, 127.4, 128.4, 137.2, 162.1, 164.5, 174.3, 183.6.

Anal. Calcd. for $C_{17}H_{18}N_4O_2$: C, 65.79; H, 5.85; N, 18.05. Found: C, 65.79; H, 5.82; N, 17.96.

6-(*N*-Benzyl-*N*-3-butynylamino)-5-formyl-4-methoxy-2-pivaloylaminopyrimidine (**16b**).

A mixture of **15b** (12.5 g, 0.0403 mole), trimethylacetic anhydride (16.34 ml, 0.08 mole), 4-dimethylaminopyridine (50 mg, 4.1 mmoles) and toluene (400 ml) was heated at reflux for 12

hours. The solvent was removed by evaporation under reduced pressure, the residue was triturated with pentane, and the pentane was decanted. The residue was dissolved in methylene chloride and washed with a saturated solution of sodium bicarbonate, dried over anhydrous magnesium sulfate and the solvent was removed by evaporation under reduced pressure. The residue was chromatographed on silica gel, eluting with 2% methanol in methylene chloride to give 13.0 g (82%) of a colorless solid, mp 128-129°; ^{1}H nmr (deuteriochloroform): δ 1.32 (s, 9H), 1.94 (t, 1H, J = 2.6 Hz), 2.52 (dt, 2H, J = 2.6, 7.1 Hz), 3.68 (q, 2H, J = 7.1 Hz), 4.05 (s, 3H), 4.86 (s, 2H), 7.23 (m, 5H), 7.84 (br s, 1H), 10.1 (s, 1H); ^{13}C nmr (deuteriochloroform): δ 17.6, 27.4, 40.4, 49.1, 54.6, 54.8, 70.0, 81.4, 98.0, 127.4, 127.7, 128.6, 136.7, 156.7, 163.5, 174.3, 175.5, 184.8.

Anal. Calcd. for $C_{22}H_{26}N_4O_3$: C, 66.99; H, 6.64; N, 14.20. Found: C, 66.79; H, 6.45; N, 13.81.

6-(*N*-Benzyl-*N*-3-butynylamino)-4-methoxy-5-oximinomethyl-2-pivaloylaminopyrimidine (**17b**).

A mixture of **16a** (3.0 g, 0.0076 mole), hydroxylamine hydrochloride (0.581 g, 0.0084 mole), pyridine (0.74 ml, 0.009 mole) and ethanol (70 ml) was heated at reflux for 1.5 hours. The solvent and excess pyridine were removed by evaporation under reduced pressure and the residue was partitioned between methylene chloride and 0.1 *N* hydrochloric acid. The organic layer was separated, dried over anhydrous magnesium sulfate and the solvent was removed *in vacuo* to give 2.52 g (81%) of a light brown solid. An analytical sample was obtained by chromatography on silica gel, eluting with 50% ethyl acetate in hexane to give a colorless solid, mp 112-113°; ^{1}H nmr (deuteriochloroform): δ 1.30 (s, 9H), 1.95 (t, 1H, J = 2.6), 2.52 (dt, 2H, J = 2.6, 7.0 Hz), 3.62 (t, 2H, J = 7.0 Hz), 3.97 (s, 3H), 4.76 (s, 2H), 7.30 (m, 5H), 7.82 (s, 1H), 8.03 (s, 1H), 9.58 (br s, 1H); ^{13}C nmr (deuteriochloroform): δ 17.5, 27.5, 40.3, 48.9, 54.6, 54.9, 70.0, 81.9, 91.4, 127.4, 128.5, 128.6, 137.4, 144.2, 154.9, 164.3, 169.3, 175.7.

Anal. Calcd. for $C_{22}H_{27}N_5O_3$: C, 64.53; H, 6.65; N, 17.10. Found: C, 64.20; H, 6.43; N, 16.74.

6-Benzyl-10-methoxy-8-pivaloylamino-4,5-dihydro-6*H*-isoxazolo[3,4-*d*]pyrimido[4,5-*b*]azepine (**25**).

A mixture of **17b** (0.4 g, 0.98 mmole), *N*-chlorosuccinimide (0.156 g, 1.17 mmole), and anhydrous methylene chloride (50 ml) was stirred at room temperature, under nitrogen, for 1.5 hours. Triethylamine (0.12 g, 1.19 mmole) was then added and the mixture was stirred for a further 2 hours. Water (50 ml) was added and the organic layer separated, dried over anhydrous magnesium sulfate, and the solvent was removed in *vacuo*. The residue was chromatographed on silica gel, eluting with 75% ethyl acetate in hexane to give 0.19 g (48%) of a colorless solid, mp 158-161°; ^{1}H nmr (deuteriochloroform): δ 1.24 (s, 9H), 2.62 (m, 2H), 3.33 (m, 2H), 4.06 (s, 3H), 4.98 (s, 2H), 7.23 (m, 5H), 7.75 (br s, 1H), 8.11 (s, 1H); ^{13}C nmr (deuteriochloroform): δ 23.2, 27.4, 40.3, 50.1, 54.9, 55.1, 87.2, 116.8, 122.2, 128.2, 128.3, 137.8, 153.6, 153.7, 155.8, 163.9, 170.0, 175.6.

Anal. Calcd. for $C_{22}H_{25}N_5O_3$: C, 64.86; H, 6.14; N, 17.20. Found: C, 64.76; H, 6.24; N, 17.46.

5-Amino-9-benzyl-6-formyl-4-methoxy-2-pivaloylamino-7,8-dihydropyrimido[4,5-*b*]azepine (**26**).

A mixture of **25** (100 mg, 0.246 mmole), molybdenum hexacarbonyl (120 mg, 0.49 mmole) and acetonitrile (5 ml) was heated at reflux for 5.5 hours. The solution was filtered through a pad of Celite, the solvent was removed by evaporation under reduced pressure and the residue was chromatographed on silica gel, eluting with 65% ethyl acetate in hexane to give 50 mg (49%) of a yellow solid, mp 130-133°; ^{1}H nmr (deuteriochloroform): δ 1.3 (s, 9H), 2.56 (m, 2H), 3.51 (m, 2H), 4.06 (s, 3H), 4.86 (s, 2H), 5.79 (br s, 1H), 7.24 (m, 5H), 7.72 (br s, 1H), 9.03 (s, 1H), 10.47 (br s, 1H); ^{13}C nmr (deuteriochloroform): δ 26.1, 27.4, 40.3, 54.7, 55.5, 55.7, 58.7, 94.4, 107.9, 127.1, 128.0, 128.5, 138.0, 155.3, 157.5, 162.2, 169.0, 175.7; hrms: ei m/z for $C_{22}H_{27}N_5O_3$. Calcd: 409.2114. Found: 409.2145.

Anal. Calcd. for $C_{22}H_{27}N_5O_3$: C, 64.54; H, 6.60; N, 17.11. Found: C, 64.30; H, 6.33; N, 16.94.

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REFERENCES AND NOTES

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