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Organic Preparations and Procedures International: The New Journal for Organic Synthesis

Publication details, including instructions for authors and subscription information: http://www.tandfonline.com/loi/uopp20

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Dilip R. Birari^a, Maruti G. Ghagare^a, Muddassar A. Kazi^a, Sandeep M. Bagul^a, Bhausaheb K. Ghotekar^a, Raghunath B. Toche^a & Madhukar N. Jachak^a

^a Organic Chemistry Research Centre, Department of Chemistry, KTHM College, Nashik, 422002, M. S., India Published online: 20 Nov 2009.

To cite this article: Dilip R. Birari , Maruti G. Ghagare , Muddassar A. Kazi , Sandeep M. Bagul , Bhausaheb K. Ghotekar , Raghunath B. Toche & Madhukar N. Jachak (2009) Synthesis of Cytosine Derivatives and Study of their Alkylation under Mild Conditions, Organic Preparations and Procedures International: The New Journal for Organic Synthesis, 41:6, 515-532

To link to this article: <u>http://dx.doi.org/10.1080/00304940903324390</u>

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Synthesis of Cytosine Derivatives and Study of their Alkylation under Mild Conditions

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Organic Chemistry Research Centre, Department of Chemistry, KTHM College, Gangapur Road, Nashik 422002, (M. S.), India

The isolation of cytosine from the hydrolysis of calf thymus tissue in 1894 and its structure determination in 1903,¹ followed by its first synthesis from 2-ethylthiopyrimidin-4(3*H*)-one are a few of the first important steps of current nucleotide chemistry. Cytosine derivatives have gained paramount importance in the pharmaceutical sphere, particularly in cases where no vaccination is available and prompt medical treatment is required. Hence these derivatives are widely used as antiviral,² anti-neoplastic,^{3–5} and anti-AIDS agents.⁶

Cytosine derivatives have been synthesized through various processes. One of the most frequently used involves the conversion of uracil into 2,6-dichloropyrimidine, followed by monoaminolysis to a mixture of chloropyrimidinamines. These chloropyrimidinamines are converted into mixtures of methoxypyrimidinamines by treatment with sodium methoxide. Isolation of the desired components is accomplished by taking advantage of the different solubility of the individual components in dioxane followed by acid hydrolysis of isolated methoxypyrimidinamines into cytosine.⁷ Another route starts with the conversion of 2-thiouracil into dithiouracil followed by its aminolysis into 2-thiocytosine, which upon heating with aqueous chloroacetic acid or warming in DMSO containing a little sulfuric acid leads to cytosine.⁸ Most of the synthetic endeavors towards the preparation of cytosine involve the corresponding thio analogues.⁹ This suggests that there is a need for the development of alternative efficient, commercially viable approaches towards the synthesis of cytosine derivatives.

Several pyrimidine derivatives have recently emerged as the integral backbone of calcium channel blockers, antihypertensive agents and antagonists.^{10–14} We previously reported the synthesis of 4-aryl/4-aminopyrimidines,^{15, 16} fused pyrimidines¹⁷ and pyrazolo-[3,4-*b*]pyridines.¹⁸ Heterocycles such as pyrazoles,^{19–22} pyrimidines^{23–26} and 1,2,4 triazolo-[1,5-*a*] pyrimidines²⁷ have been the subject of chemical and biological studies

Received March 20, 2009; in final form September 3, 2009.

Address correspondence to Madhukar N. Jachak, Organic Chemistry Research Centre, Department of Chemistry, KTHM College, Gangapur Road, Nashik 422002, (M. S.), India. E-mail: mn_jachak@hotmail.com



Figure 1

due to their interesting pharmacological properties such as antipyretic, $^{28, 29}$ analgesic, 30 anti-inflammatory, 31 potential herbicidal, 32 fungicidal $^{33, 34}$ and leishmanicidal activity. $^{35, 36}$ Recently diethyl ethoxymethyl-enemalonate (EMME) and 2-ethoxymethylene-3-oxobutanenitrile (*Figure 1*) have been used as attractive building blocks for the synthesis of biologically relevant heterocyclic or carbocyclic compounds. $^{37-42}$

Pyrimidines and triazines can easily be accessed by reaction of **EMME** with aliphatic or aromatic amidines.⁴³ Similarly pyrazoles, pyrimidines and bicyclic triazolopyrimidines are readily synthesized by reaction of 2-ethoxymethylene-3-oxobutanenitrile with hydrazines, amidines and aminotriazoles respectively.⁴⁴ Stimulated by these findings, we explored the application of 2-dimethyaminomethylene-3-oxobutanenitrile (1) a synthetic equivalent of **EMME** and 2-ethoxymethylene-3-oxobutanenitrile where the two ester groups in **EMME** are replaced by ketone and nitrile moieties and the ethoxy group is replaced by the dimethylamino moiety (2-ethoxymethylene-3-oxobutanenitrile). We now report an efficient, commercially viable synthesis of some novel cytosines and studied their alkylation.

A previous communication¹⁶ described the one-pot synthesis of ureidopropenenitriles from benzoylacetonitrile, triethyl orthoformate and urea (and substituted ureas). Considering the biological activities of cytosine derivatives, we undertook the synthesis of new cytosine derivatives having an acetyl group at the 5-position. Thus, treatment of 3-oxobutanenitrile, obtained by acid hydrolysis of β -aminocrotononitrile,⁴⁵ with dimethylformamide dimethyl acetal (DMF-DMA) furnished 2-dimethyaminomethylene-3-oxobutanenitrile (1). Condensa-tion of 1 with urea or substituted ureas (**2a–f**) furnished open-chain ureidopropenitriles **3** in 50–80% yields (*Scheme 1*). Compounds **3** were characterized by spectral and analytical data (*Tables 1 and 2*).



Scheme 1

It is to be noted that the protons attached to nitrogen and the olefinic carbon in compound 3 showed doubling of signals in ¹H NMR. This type of doubling of signals

has already been reported and discussed in the literature.⁴⁶ Cyclization of **3a** and **3b** using triethylamine in acetonitrile yielded 5-acetylcytosine derivatives **4a** and **4b** in 43–47% yields (*Scheme 2*). However, **3c–f** could not be cyclized to 5-acetylcytosines under these conditions. Hence **3c–f** were cyclized to **4c-f** using a strong base such as *t*-BuOK in ethanol at reflux temperature in 75–85% yields (*Scheme 2*). Treatment of **3a** and **3b** with sodium methoxide in MeOH furnished **5a** and **5b** respectively, in 77–78% yields (*Scheme 2*). The presence of the hydroxy group in **5** was detected by the FeCl₃ color test for phenol and confirmed by ¹³C NMR.



 $R = CH_3$

a) $R' = CH_2Ph$; b) $R' = CH_2CH_2Ph$; c) R' = H; d) $R' = CH_3$; e) $R' = CH_2CH_3$; f) R' = Ph

Scheme 2

It is interesting to note that treatment of **3a** and **3b** with AcOH yielded the 5-cyanopyrimidines **6a,b** in 40–48% yields; however, this cyclization failed to occur with **3c-f**. Compounds **4–6** were characterized by IR, mass spectra, ¹H NMR and ¹³C NMR (*Table 2*). In a manner analogous to **4a**, compound **4g** ($\mathbf{R} = \mathbf{H}$, $\mathbf{R}^1 = \mathbf{CH}_2\mathbf{Ph}$) was prepared from 3-dimethylamino-2-formyl-acrylonitrile^{47–49} a useful synthon for the synthesis of various heterocycles. The *N*-alkylation of **4a** and **4g** was studied. Of the several alkylation conditions tested, including bases such as sodium bicarbonate, potassium carbonate, sodium carbonate, triethylamine, *t*-BuOK in solvents such as acetonitrile, dimethylformamide, dimethyl sulfoxide, acetone, tetrahydrofuran, toluene, the combination of DMF and K₂CO₃ proved to be superior and scalable. Thus reaction of compounds **4a** and **4g** (1.05 equiv) with **7** proceeded smoothly (monitored by TLC) and furnished exclusively *N*(1) alkylated products **8a–n** in 60–95% yields. Neither O-alkylated nor N-imino alkylated products were obtained. The presence of the imine nitrogen in **4a** and **4g** was established by *N*-alkylation with 2-bromo-*N*-phenyacetamides (**7**) (*Scheme 3*). Compounds **8a–n** were characterized by spectral and analytical data (*Tables 1 and 2*).



a) $R = CH_3$, $Ar = 4-Cl-3-CF_3-C_6H_3$; b) $R = CH_3$, $Ar = 4-F-C_6H_4$; c) $R = CH_3$, $Ar = 4-Me-C_6H_4$; d) $R = CH_3$, $Ar = 2-Cl-6-F-C_6H_3$; e) $R = CH_3$, $Ar = 2,5-di-CF_3-C_6H_3$; f) $R = CH_3$, $Ar = 2,4-di-Cl-C_6H_3$; g) $R = CH_3$, $Ar = 4-Cl-C_6H_4$; h) R = H, $Ar = 4-Cl-3-CF_3-C_6H_3$; i) R = H, $Ar = 4-F-C_6H_4$; j) R = H, $Ar = 4-Me-C_6H_4$; k) R = H, $Ar = 2-Cl-6-F-C_6H_3$; l) R = H, $Ar = 2,5-di-CF_3-C_6H_3$; m) R = H, $Ar = 2,4-di-Cl-C_6H_3$; n) R = H, $Ar = 4-Cl-C_6H_4$; h) R = H, $Ar = 4-Cl-C_6H$

Scheme 3

Under similar conditions, the alkylation of cytosine 4g with alkyl halides 9a-e gave, after quenching in ice-cold water, solids which were recrystallized from ethanol. Clean monoalkylation of 4g occurred to afford 10a-e in 78–90% yields. Thus selectivity of the alkylation at the N(1) position and not at the imino nitrogen was confirmed by spectroscopic analysis (*Scheme 4, Table 2*).



Scheme 4

The feasibility of our approach was tested by reaction of a known cytosine, 5-fluorocytosine **11**, with a slight excess of different anilides (**7**) (1.05 mmole) in DMF/K₂CO₃ at room temperature for 8–12 h; the sole products **12a–g** obtained were characterized by IR, Mass, ¹H NMR and ¹³C NMR, which showed that the alkylation was at N(1) nitrogen and not at the imino nitrogen. No by-products were detected when the reaction was monitored by TLC (*Scheme 5*).

In summary, we have reported a simple and convenient method to synthesize some novel cytosine derivatives. We have reported a new series of N(1) alkylated-3-benzyl-4-imino-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carbaldehyde (**4g**) and 3-benzyl-4-imino-2-oxo-1,2,3,4-tetrahydro-5-acetylpyrimidine (**4a**) by a simple, convenient and scalable method. This transformation could be of importance to synthetic and combinatorial chemists to generate a library of cytosine derivatives.

Experimental Section

Melting points were determined on a Buchi melting point apparatus, Mod. B-545 and are uncorrected. The ¹H (300 MHz) and ¹³C (75 MHz) NMR spectra (*Table 2*) were



Scheme 5

recorded on a Varian XL-300MHz spectrometer. Chemical shifts are reported in parts per million (δ ppm) relative to tetramethylsilane (*TMS*), and multiplicities are given as s (singlet), d (doublet), t (triplet), q (quartet), or m (multiplet). The solvents for NMR spectra were *DMSO-d*₆ and *CDCl*₃ unless otherwise stated. Infrared spectra were taken on a Thermo Electron Corporation Nicolet 380 FTIR instrument as potassium bromide pellets unless otherwise stated. Mass spectra were recorded on Shimadzu GC-MS QP 2010A mass spectrometer with an ionization potential of 70 eV (*Table 2*). All reactions were monitored by thin layer chromatography, carried out on 0.2 mm silica gel 60 F₂₅₄ (Merck) plates using UV light (254 and 366 nm) for detection. Common reagent grade chemicals were either commercially available or used without further purification or prepared by standard literature procedures.

Synthesis of 3-Oxobutanenitrile

 β -Aminocrotononitrile (10.0 g, 0.12 mol) was slurried in water (12.5 mL) and conc. hydrochloric acid (12.5 mL) was added dropwise below 15°C within 1 h. The reaction mixture was then heated to 80°C for 2 h. After completion of reaction (TLC check, chloroform:methanol 9:1), the reaction mixture was cooled to room temperature and diluted with ethyl acetate (100 mL). The insoluble materials were filtered through hyflo supercel, the biphasic clear filtrate was separated, and the aqueous layer was extracted twice with ethyl acetate (100 mL × 2). The combined ethyl acetate extracts were washed with saturated sodium chloride solution and the organic layer was dried over sodium sulfate. The solvent was removed under reduced pressure to afford crude 3-oxobutanenitrile as a light brown oil (9.0 g, 90%), which was used in the next step for the synthesis of compound **1**.

Synthesis of Dimethylaminomethylene-3-oxobutanenitrile (1)

The crude, 3-oxobutane- nitrile (9.0 g, 0.11 mol) obtained above, was dissolved in dimethylformamide dimethyl acetal (20 mL) at 25°C and stirred for 30 min. The reaction mixture was then heated to 70–80°C and maintained for 1 h. After completion of reaction (TLC check, chloroform: methanol, 9:1), the reaction mixture was cooled to room temperature and the excess dimethylformamide dimethyl acetal was distilled to give dark brown oil. The crude oil was purified by column chromatography on silica gel (elution with dichloromethane)

 Table 1

 Yield, mps, and Elemental Analyses of Compounds

			Elemental Analysis (Found)		
Cmpd	Yield (%)	mp (°C)	С	Н	N
1	84	65–66	60.85 (60.85)	7.30 (7.25)	20.28 (20.25)
3a	76	145-146	64.19 (64.02)	5.39 (5.42)	17.27 (17.46)
3b	80	164–165	65.35 (65.30)	5.88 (6.02)	16.33 (16.54)
3c	76	196–198	47.06 (47.23)	4.61 (4.60)	27.44 (27.37)
3d	63	200-201	50.29 (50.35)	5.43 (5.60)	25.14 (25.30)
3e	50	199–200	53.03 (52.79)	6.12 (6.03)	23.19 (22.94)
3f	67	224-225	62.87 (62.82)	4.84 (4.83)	18.33 (18.26)
4 a	43	215-216	64.19 (63.99)	5.39 (5.15)	17.27 (17.48)
4b	47	220-221	65.35 (65.30)	5.88 (6.02)	16.33 (16.56)
4 c	75	218-219	47.06 (47.26)	4.61 (4.71)	27.44 (27.66)
4d	88	225-226	50.29 (50.46)	5.43 (5.26)	25.14 (24.86)
4 e	76	164–165	53.03 (53.23)	6.12 (6.34)	23.19 (23.42)
4f	85	113–114	62.87 (62.82)	4.84 (4.83)	18.33 (18.26)
5a	78	195–196	64.19 (64.44)	5.39 (5.13)	17.27 (17.06)
5b	77	197–198	65.35 (65.47)	5.88 (5.95)	16.33 (16.51)
6a	48	211-212	69.32 (69.53)	4.88 (4.77)	18.66 (18.43)
6b	40	220-221	70.28 (70.08)	5.48 (5.24)	17.56 (17.84)
8a	60	145–146	55.18 (55.43)	3.79 (4.06)	11.70 (11.55)
8b	80	158–160	63.95 (63.82)	4.86 (4.95)	14.21 (14.13)
8c	78	186–187	67.68 (67.52)	5.68 (5.79)	14.35 (14.60)
8d	96	230-231	58.82 (59.04)	4.23 (4.14)	13.06 (13.25)
8e	74	168-170	53.91 (54.15)	3.54 (3.36)	10.93 (11.16)
8f	91	130–131	56.64 (56.62)	4.07 (4.08)	12.58 (12.32)
8g	92	168–170	53.96 (54.05)	4.06 (4.16)	11.99 (12.07)
8h	86	115-117	54.26 (54.39)	3.47 (3.58)	12.05 (12.31)
8i	89	112–114	63.15 (63.38)	4.50 (4.68)	14.73 (14.97)
8j	91	170–172	67.01 (67.31)	5.36 (5.57)	14.88 (15.16)
8k	87	185–187	57.91 (58.20)	3.89 (4.16)	13.51 (13.73)
81	85	194–196	53.02 (53.27)	3.24 (3.50)	11.24 (11.49)
8m	92	125–127	55.70 (55.43)	3.74 (3.49)	12.99 (12.81)
8n	88	113–115	60.53 (60.29)	4.32 (4.47)	14.12 (14.25)
10a	78	116–118	65.36 (65.19)	5.88 (6.07)	16.33 (16.50)
10b	90	105–107	67.35 (67.50)	6.71 (6.49)	14.73 (14.50)
10c	82	94–96	67.35 (67.19)	6.71 (6.45)	14.73 (14.61)
10d	88	112–114	68.21 (68.00)	7.07 (7.29)	14.04 (14.26)
10e	88	155–158	62.63 (62.48)	4.43 (4.55)	15.38 (15.00)
12a	83	288-290	48.58 (48.80)	3.40 (3.57)	18.88 (19.11)

			Elemental Analysis (Found)		
Cmpd	Yield (%)	mp (°C)	С	Н	N
12b	86	297–298	42.82 (43.06)	2.49 (2.27)	15.36 (15.50)
12c	87	278-280	51.43 (51.32)	3.60 (4.45)	19.99 (20.20)
12d	85	288-290	56.52 (56.74)	4.74 (4.95)	20.28 (20.10)
12e	85	253-255	45.80 (46.04)	2.88 (3.01)	17.80 (18.00)
12f	89	240-242	42.22 (42.49)	2.28 (2.47)	14.07 (14.26)
12g	79	235-237	43.53 (43.79)	2.74 (2.87)	16.92 (17.14)

 Table 1

 Yield, mps, and Elemental Analyses of Compounds (Continued)

to afford light yellow oil, which was triturated with hexane to yield a yellow solid. It was crystallized from isopropyl alcohol (12.6 g, 84%), mp. 65–68°C, *lit*.⁵⁰ mp. 68°C.

Synthesis of 2-Acetyl-3-uredopropenenitriles 3a-f. General Procedure

To a solution of **1** (10.0 g, 0.074 mol) and urea/substituted ureas **2a–f** (0.079 moles) in ethanol (250 mL), was added 7.5 mL conc. hydrochloric acid and the reaction mixture was heated to $60-65^{\circ}$ C for 3 h. After completion of the reaction (TLC check, chloroform: methanol, 9:1), the solvent was evaporated to dryness under reduced pressure. The resulting solid obtained was stirred in cold ethanol (25 mL), collected, dried and recrystalized from methanol: chloroform (3:2). All the compounds were obtained as colorless solids.

Synthesis of 5-Acetyl-3,4-dihydro-4-iminopyrimidin-2(1H)-one Derivatives 4a, b. General Procedure

To a solution of **3a** or **3b** (0.004 mol) in acetonitrile (20 mL) was added triethylamine (1.2 mL), and the mixture was refluxed for 24–30 h. After completion of reaction (TLC check, chloroform: methanol, 9:1), the solvent was evaporated to dryness under reduced pressure. The resulting solid was stirred in cold ethanol (10 mL), collected, dried and recrystalized from methanol:chloroform (3:2). Compounds **4a** and **4b** were obtained as pale yellow solids.

Synthesis of 5-Acetyl-3,4-dihydro-4-iminopyrimidin-2(1H)-one Derivatives 4c–f. General Procedure

A solution of 3c-f (0.0021 mol) and sodium tert-butoxide (0.28 g, 0.0025 mol) in ethanol (10 mL) was refluxed for 5–6 hrs. After completion of reaction (TLC check, chloro-form:methanol, 9:1). The solvent was removed under reduced pressure and the oily residue obtained was treated with acetic acid to adjust pH up to 6.5. The solids obtained were collected, washed with ethanol and crystallized from ethanol.

Table 2	
IR, ¹ H NMR, ¹³ C NMR, and Mass Spectra of Compounds	

Cmpd	$IR (cm^{-1})$	¹ H NMR (δ)	¹³ C NMR (δ)	Mass Spectra
1	3469, 2241, 1711, 1667	2.42(s, 3H,-CH ₃), 3.20(s, 3H,-NCH ₃), 3.4(s, 3H, -NCH ₃), 7.78(s, 1H,CH).	20.11, 24.67, 108.37, 114.86 154.0, 194.9	138[M] ⁺ , 123, 95, 81, 42.
3a	3334, 3308, 2222, 1726, 1667, 1544	2.32(s, 3H, CH ₃), 4.45(t, 2H, CH ₂), 7.30(m, 5H, Ar-H), 8.40 & 8.00 (d, 1H, olefinic CH, $J = 12.3 & 9.9$ Hz), 8.78 & 7.68(t, 1H, NH, $J =$ 5.3 Hz & 9.3 Hz), 11.42 & 10.31(d, 1H, NH, $J = 12.3$ Hz & 9.9 Hz).	26.4, 48.9, 89.4, 119.3, 126.2, 127.1, 127. 8, 128.2, 128.8, 141.7, 150.2, 151.7, 195.2.	243[M] ⁺ , 144, 110, 95, 68, 43.
3b	3312, 3278, 2219, 1725, 1669, 1561	2.32(s, 3H, CH ₃), 2.78(t, 2H, CH ₂ , $J = 4.8$ Hz), 3.45(dt, 2H, CH ₂ , $J = 6.0$ Hz, & $J = 4.8$ Hz), 7.23 (m, 5H, Ar-H), 8.40 & 8.00(d, 1H, olefinic CH, $J = 12.6$ & 12.9 Hz), 8.79 & 7.70(t, 1H, NH, J = 6.0 Hz & 4.8 Hz), 11.32 & 10.21(d, 1H, NH, $J = 12.6$ Hz & 12.9 Hz).	26.3, 38.1, 42.9, 89.5, 115.6, 125.4, 126.3, 126.5, 128.3, 128.6, 140.4, 150.2, 151.3, 195.5.	257[M] ⁺ , 144, 110, 95, 68, 43
3c	3371, 3212, 2219, 1739, 1670, 1552	2.30(s, 3H, CH ₃), 6.70 (bs,1H, NH), 7.70(bs, 1H, NH), 8.02 & 8.40(d, 1H, olefinic CH, J = 12.9 Hz)10.20 & 11.30(d, 1H, NH, J = 12.9 Hz).	25.6, 98.2, 115.0, 155.2, 164.7, 194.5	153[M] ⁺ , 144, 110, 95, 68, 43.
3d	3317, 3276, 2218, 1711, 1661, 1573	2.3 (s, 3H, CH ₃), 2.7(d, 3H, J = 4.5Hz, CH ₃), 8.20 and 7.11 (bq, 1H, NH, $J = 4.5$ and 4.2 Hz), 8.4 and 8.0 (d, 1H, olefinic CH, $J = 11.1$ and 12.6 Hz), 11.4 and 10.3 (d, 1H, NH, $J = 10.5$ Hz).	26.2, 28.2, 88.9, 115.6, 151.2, 152.1, 195.5,	167[M] ⁺ , 110, 95, 58, 43.
3e	3325, 3198, 2219, 1723, 1666, 1554	1.07(t, 3H, CH ₃), 2.30(s, 3H, CH ₃), 3.17(dq, 2H, CH ₂), 8.10 (d, 1H, olefinic CH, J = 12.3 Hz, & 13.2 Hz), 8.32(t, 1H, NH), 11.37 & 10.23 (d, 1H, NH, J = 12.6 Hz).	14.1, 26.3, 40.6, 88.7, 119.4, 150.1, 151.6, 195.3.	181[M] ⁺ , 144, 115, 106, 91, 44.

Cmpd	IR (cm^{-1})	¹ H NMR (δ)	13 C NMR (δ)	Mass Spectra
3f	3296, 3224,	2.32(s, 3H, CH ₃),	35.5, 89.2, 118.2,	229[M] ⁺ , 137,
	2218, 1721,	7.50-7.47(m, 5H, Ar-H), 8.	124.6, 125.1,	119, 110, 95,
	1671, 1544	40 & 8.10(d, 1H, olefinic	125.1, 127.1,	77, 68, 43.
		CH, $J = 2.9$ Hz &	127.6, 137.4,	
		12.3 Hz), 9.57(s,1H, NH),	151.2, 152.2,	
		11.5 & 10.25(d, 1H, NH, J	195.3.	
		= 12.3 Hz and 12.9 Hz)		
4a	3405, 3134,	2.42(s, 3H, CH ₃), 5.18(s, 2H,	26.6, 44.5, 101.5,	243[M] ⁺ , 242,
	1671, 1575,	N-CH ₂), 7.36–7.15 (m, 5H,	126.8, 127.2,	159, 138,
	1370	Ar-H), 8.79 (s, 1 H, olefinic	127.8, 128.2,	106, 91, 77,
		CH), 8.50(bs, 1H, NH),	129.3, 138.4,	55, 43.
		10.0(bs, 1H, NH).	143.4, 150.1,	
			152.4, 197.2.	
4b	3403, 3144,	2.40(s, 3H, CH ₃), 2.80(t, 2H,	26.1, 36.2, 45.0,	257[M] ⁺ , 166,
	1670, 1570,	$CH_2, J = 5.4 Hz), 4.10 (t,$	119.2, 125.4,	137, 104,
	1376	2H, N-CH ₂ , $J = 5.3$ Hz),	126.8, 128.3,	95, 77.
		7.26–7.29	128.5, 130.2,	
		(m, 5H, Ar -H), 8.57 (bs,	140.5, 154.2,	
		1H, NH), 8.72(s, 1H,	157.7, 195.2.	
		olefinic, CH), 10.05(bs, 1H,		
	2525 2460	\mathbf{NH}	20 4 00 2 154 6	1520 84 120
4c	3527, 3469,	$2.37(s, 3H, CH_3), 8.72(s, 1H, 1H)$	29.4, 99.2, 154.6,	153[M] ⁺ , 139,
	3282, 3131,	olefinic CH), 8.57 (bs, 1H,	157.1, 164.2,	124, 110,
	1711.	NH), 9.90 (bs, 2H, NH ₂).	195.3.	95, 57, 44.
4d	3527, 3469,	2.40(s, 3H,-CH ₃), 3.22(s, 3H,	26.1, 29.4, 99.2,	167 [M] ⁺ ,
	3282, 3131,	$N-CH_3$), 8.69(s,1H, olefinic	154.6, 157.1,	152, 139,
	1711, 1667	CH), 8.42(bs, 1H, NH),	164.2, 195.3.	124, 110,
		9.80(bs, 1H, NH).		95, 57, 44.
4 e	3325, 3198,	$1.10(t, 3H, CH_3), 2.40(s, 3H,$	15.0, 26.1. 38.4,	181[M] ⁺ , 153,
	2219, 1723,	CH_3), 3.93(q, 2H, CH_2),	119.2, 140.1,	138, 83.
	1666,	8.69(s, 1H, olefinic	154.6, 157.1,	
		CH), 10.05 (bs, 2H, NH ₂).	195.3	
4f	3527, 3469,	2.42(s, 3H, CH ₃), 7.42–7.32	26.3, 115.2,	229[M] ⁺ , 186,
	3282, 3131,	(m, 5H, Ar-H), 7.82 (s, 1H,	119.4, 120.1,	171, 143,
	1711, 1667	olefinic CH), 8.42 (bs, 1H,	125.1, 130.2,	77, 44.
		NH). 9.8(bs, 1H, NH).	131.2 134.2,	
			140.1, 150.1,	
			165.2, 198.4	

 Table 2

 IR, ¹H NMR, ¹³C NMR, and Mass Spectra of Compounds (*Continued*)

(Continued on next page)

Table 2
IR, ¹ H NMR, ¹³ C NMR, and Mass Spectra of Compounds (<i>Continued</i>)

Cmpd	IR (cm^{-1})	¹ H NMR (δ)	¹³ C NMR (δ)	Mass Spectra
5a	3148, 1682,	2.42(s, 3H, CH ₃), 5.10(s,	26.6, 40.2, 125.0,	243[M] ⁺ , 242,
	1564, 1288	2H, N-CH ₂), 7.31–7.34	126.8, 127.2,	226, 183,
		(m, 5H, Ar-H), 8.72(s,	127.4, 128.2,	159, 138,
		1H, olefinic CH),	128.6, 145.3,	106, 91.
		9.92(s, 1H, NH),	149.4, 154.2,	
		11.2(s,1H,OH).	164.4, 198.2	
5b	3405, 3143,	2.49(s, 3H, CH ₃), 2.80(t,	27.1, 31.2, 40.1,	257[M] ⁺ , 166,
	1671, 1575,	$2H, CH_2, J = 7.5 Hz),$	126.8, 127.4,	137, 104,
	1366, 1287	$4.20(t, 2H, N-CH_2, J =$	127.8, 128.1,	95, 77.
		7.8 Hz), 7.20–7.34 (m,	128.4, 128.6,	
		5H, Ar-H), 8.82(s, 1H,	139.5, 148.3,	
		olefinic CH), 10.42(bs,	153.2, 167.7, 198.2	
		1H, NH), 11.60(s, 1H, OH)		
6a	3415 3134	$1.71(s, 3H, CH_2), 5.18(s)$	16 6 48 5 54 5	225[M]+ 183
va	2240	2H N-CH ₂) 7.06–7.50	97.6 120.9 126.8	159 138
	2240,	(m 5H Ar-H) 8.79(s)	127.0, 128.2	106 91
		(III, 511, 711, 11), 0.75(3, 1H, ArH)	129.3 141 5 155 0	100, 91.
		····, · ····).	160.0 164.7	
6b	3453 3150	1 78(s 3H CH ₂) 2.82 (t	20 1 33 7 50 02	239[M]+ 166
0.0	2242, 1670.	$2H$, CH_2 , $J = 5.4$ Hz).	58.4. 95.4. 117.8.	137, 104.
	1570 1376	$4 15 (t 2H N-CH_2) I =$	127.1 128.0	95 77
	1070, 1070	6.0 Hz, $7.10-7.60(m.)$	128.4 128.8	,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,,
		5H. Ar-H). 8.60(s. 1H.	130.2, 140.5,	
		ArH).	145.2, 154.2,	
8a	3464, 3268,	$2.50(s, 3H, CH_3), 4.84(s, $	30.2, 44.3, 52.8,	477(M-1).
	3196, 3122,	NCH ₂), 5.00(s, 2H,	115.1, 117.7,	278, 221,
	3075, 1696,	-N CH ₂), 7.26(m, 5H,	120.2, 123.2,	195, 133, 91,
	1658, 1597	ArH), 7.29–7.70 (m,	124.1, 126.2,	77, 65, 43.
	,	3H, ArH) 8.15 (bs, 1H,	126.4, 127.0,	, ,
		NH), 8.59(s, 1H,	127.2, 127.7,	
		olefinic CH),	128.5, 132.5,	
		10.85(bs,1H,NH).	137.1, 141.2,	
			150.0, 151.8,	
			160.4, 165.5, 198.2	

Cmpd	$IR (cm^{-1})$	¹ H NMR (δ)	¹³ C NMR (δ)	Mass Spectra
8b	3455, 3266,	2.36(s, 3H,-CH ₃), 4.65 (s,	26.2, 43.3, 51.8,	393(M-1),
	3147, 3115,	2H, N-CH ₂), 5.13(s, 2H,	112.2, 115.3, 115.7,	242, 133,
	3075, 1697,	-CH ₂), 7.25(m, 7H, Ar-H),	120.5, 121.1, 126.0,	111, 91,
	1657	7.58(m, 2H, Ar-H),	126.8, 127.4, 128.2,	77, 43.
		8.50(bs, 1H, -NH), 9.63(s,	128.4, 134.5, 137.1,	
		$1H_{,} = CH_{,} 10.68$ (bs,	149.2, 153.2, 156.2,	
		1H,-NH).	159.2, 165.2, 196.2	
8c	3451, 3274,	2.24(s, 3H, CH ₃), 2.36(s,	26.2, 45.3, 55.8,	389(M-1),
	3149, 3111,	3H,CH ₃), 4.65(s, 2H,	114.2, 115.8, 115.6,	242, 106,
	3077, 1691,	N-CH ₂), 5.13(s, 2H,	121.5, 122.4, 125.8,	91, 77, 65,
	1654,	-CH ₂), 7.10(d, 2H, $J = 8.1$	126.0, 127.2, 127.8,	43.
		Hz, Ar-H), 7.25(m, 5H,	129.4, 135.5, 139.1,	
		Ar-H), $7.46(d, 2H, J = 8.1)$	148.2, 154.2, 158.2,	
		Hz, Ar-H), 8.51(bs,	160.2, 165.2, 198.2	
		1H,-NH), 9.62 (s, 1 H, =		
		CH), 10.30(bs, 1H,-NH).		
8d	3457, 3280,	2.30(s, 3H,-CH ₃), 4.74(s,	28.2, 45.8, 53.6,	427(M-1),
	3144, 3118,	2H, N-CH ₂), 5.14(s, 2H,	113.5, 116.6, 115.2,	242, 145,
	3078, 1695,	-CH ₂), 7.20(m, 5H, Ar-H),	118.4, 122.4, 126.6,	106, 91,
	1654	7.46(m, 3H, ArH),	127.2, 127.8, 128.5,	77, 65, 43.
		8.54(bs,1H, -NH), 9.60 (s,	128.8, 131.2, 132.4,	
		$1H_{,} = CH_{,} 10.15(bs, 1H_{,})$	144.2, 149.4, 153.2,	
		-NH).	160.5, 168.3, 195.8	
8e	3485, 3288,	2.32(s, 3H, -CH ₃), 4.82 (s,	26.2, 44.8, 54.6,	511(M-1),
	3145, 3110,	2H, N-CH ₂), 5.19(s, 2H,	114.2, 115.8, 117.2,	285, 257,
	1690, 1655,	-CH ₂), 7.17(m, 5H, Ar-H),	121.8, 123.8, 124.2,	132, 124,
	1591	7.52(m, 3H, Ar-H),	124.9, 125.8, 129.2,	91, 82, 65,
		8.56(bs, 1H, -NH), 9.65(s,	133.1, 133.8, 134.2,	43.
		1H = CH), 10.22 (bs, 1H,	138.2, 141.4, 148.8,	
		NH).	150.9, 151.3, 158.5,	
			166.8, 192.2	
8f	3485, 3288,	2.36(s, 3H, -CH ₃) 4.76(s,	26.9, 43.5, 51.3,	443(M-1),
	3145, 3110,	2H, N-CH ₂), 5.13 (s, 2H,	116.9, 124.2, 126.8,	242, 161,
	3085, 1690,	-CH ₂), 7.23(m, 5H, Ar-H),	127.2, 128.8, 129.6,	133, 123,
	1655,	7.42(d, 1H, J = 8.7 Hz	130.6, 131.2, 131.9,	106, 91,
		Ar-H), 7.68(s, 1H,Ar-H)	133.5, 137.1, 141.2,	77, 65, 43.
		7.78(d, 1H, J = 8.4 Hz,	148.1, 149.3, 151.9,	
		Ar-H), 8.51(bs, 1H,-NH),	153.5, 168.8, 198.1.	
		9.60(s, 1H, = CH), 10.05		
		(bs,1H,-NH).		

 Table 2

 IR, ¹H NMR, ¹³C NMR, and Mass Spectra of Compounds (*Continued*)

(Continued on next page)

Table 2	
IR, ¹ H NMR, ¹³ C NMR, and Mass Spectra of Compounds (<i>Continued</i>)	

		, I	I V	,
Cmpd	$IR (cm^{-1})$	¹ H NMR (δ)	¹³ C NMR (δ)	Mass Spectra
8g	3489, 3281,	2.36(s, 3H,-CH ₃), 4.66(s,	26.9, 43.7, 51.5,	410[M-1], 242,
	3156, 3115,	2H, N-CH ₂), 5.13(s, 2H,	54.3, 106.9,	123, 106, 91,
	3089, 1698,	-CH ₂), 7.26 (m, 5H,	120.2, 126.9,	77, 65, 43.
	1659,	Ar-H), $7.38(d, 2H, J =$	126.9, 127.0,	
		8.7 Hz, Ar-H), 7.60(d,	127.5, 128.6,	
		2H, J = 8.7 Hz, 8.51 (bs,	128.8, 131.2,	
		1H, -NH), $9.60(s, 1H =$	131.1, 131.9,	
		CH), 10.50(bs, 1H, NH).	135.6, 137.8,	
			149.8, 153.9,	
			165.2, 195.8.	
8h	3285, 3063,	4.79(s, 2H, CH ₂), 5.18(s,	44.2, 52.1, 107.1,	464[M] ⁺
	2927, 1673,	2H, CH ₂), 7.16–7.26 (m,	117.5, 117.6,	
	1599, 1482	5H, Ar-H), 7.6881 (m,	120.6, 123.8,	
		3H,Ar-H), 8.18(s, 1H,	124.3, 126.5,	
		CH), 8.53(bs, 1H, NH),	126.9, 127.1,	
		9.52(s, 1H, CHO),	128.1, 128.3,	
		10.01(bs, 1H, NH)	132.2, 135.9,	
			137.7, 148.7,	
			153.8, 156.1,	
			165.4, 188.6	
8i	3468, 3287,	4.66(s, 2H, CH ₂), 5.11(s,	43.6, 51.8, 108.1.	380[M] ⁺
	3162, 2770.	2H, CH ₂), 7.12–7.58 (m.	115.3. 115.6.	
	2720, 1699	9H, Ar-H), 8.24(s, 1H,	120.8, 120.9,	
	,	CH), 9.22(bs. 1H, NH).	126.9, 127.3.	
		9.44 (s. 1H, CHO).	128.1. 134.7.	
		10.44(bs, 1H, NH)	136.9 149.3	
			153 2 155 3	
			156 5 159 6	
			164.9 188.9	
8i	3320 3280	2 24(s 3H CH ₂) 4 65(s	20.6 43.7 51.8	376[M]+
IJ	3032 1830	$2H_{\rm CH_2}$ 5 11(s 2H	108 2 119 1	570[11]
	1670, 1540,	$(H_2), (H_2), (J, H(3, 2H, 2H, 2H, 2H, 2H, 2H, 2H, 2H, 2H, 2H$	100.2, 119.1, 127.0, 127.4	
	1079, 1340	$2H_{2}$, 7.12(d, $J = 9$ Hz,	127.0, 127.4,	
		2H, AI-H), 7.10-7.27(III, 5H, Ar, H), 7.44(d, I = 0)	120.2, 129.3, 122.7, 125.0	
		3H, AI-H), 7.44(u, J = 9	132.7, 133.9,	
		$\Pi Z, Z\Pi, AI-\Pi),$	157.0, 149.4,	
		$0.22(8,1\Pi, U\Pi),$ $0.22(b_{0},1\Pi, V\Pi), 0.44(c_{0})$	155.4, 155.4,	
		9.22(08,1H,NH), 9.44(8,	104.8, 189.0	
		1H, CHO),10.30(bs,1H,		
		NH)		

Cmpd	$IR (cm^{-1})$	¹ H NMR (δ)	¹³ C NMR (δ)	Mass Spectra
8k	3340, 3277,	4.74(s, 2H, CH ₂), 5.11(s,	43.7, 51.4, 108.3,	414[M] ⁺
	2960, 1675,	2H, CH ₂), 7.18–7.53(m,	115.3, 122.7, 125.5,	
	1649, 1583	8H, Ar-H), 8.26 (s, 1H,	127.0, 127.2, 127.4,	
		CH), 9.20(bs, 1H, NH),	128.2, 129.1, 129.2,	
		9.43(s,1H, CHO), 10.25	132.1, 137.0, 149.3,	
		(bs,1H, NH)	153.4, 156.3, 159.7,	
			165.6, 189.1	
8 1	3350, 3281,	4.75(s, 2H, CH ₂), 5.11(s,	43.7, 51.7, 108.2,	498[M] ⁺
	3052, 2847,	2H, CH ₂), 7.18–7.26(m,	120.9, 121.3, 123.6,	
	1681, 1643	5H, Ar-H), $7.84(d, J = 6$	124.5, 124.9, 125.9,	
		Hz, 1H, Ar-H), 7.88(s,	127.0, 127.4, 128.2,	
		1H, Ar-H), $8.01(d, J = 6$	128.5, 132.8, 133.3,	
		Hz, 1H, Ar-H), 8.22(s,	135.7, 136.9, 149.4,	
		1H, CH), 9.20(bs,1H,	153.2, 155.3, 167.0,	
		NH), 9.44(s,1H, CHO),	189.1	
		10.33(bs, 1H, NH)		
8m	3320, 3246,	4.77(s, 2H, CH ₂), 5.12(s,	43.6, 51.6, 93.2,	$431[M^{+1}]$
	3033, 2810,	2H, CH ₂), 7.21–7.27(m,	108.1, 126.8, 127.0,	
	1671, 1582	5H, Ar-H), 7.68–7.72(m,	127.2, 127.5, 128.0,	
		3H, Ar-H), 8.27(s, 1H,	128.9, 129.5, 133.3,	
		CH), 9.21(bs, 1H, NH),	136.9, 149.2, 153.2,	
		9.43(s, 1H, CHO),	155.3, 165.8, 188.9	
		10.11(bs, 1H, NH)		
8n	3469, 3282,	4.68(s, 2H, CH ₂), 5.13(s,	43.0, 51.1, 108.2,	396[M] ⁺
	3131, 1711,	2H, CH ₂), 7.19–7.37(m,	115.0, 115.1, 120.5,	
	1667, 1556	5H, Ar-H), $7.39(d, J =$	121.9, 126.5, 127.7,	
		6.9 Hz, 2H, Ar-H),	128.2, 132.1, 136.1,	
		7.60(d, J = 6.9 Hz, 2H,	146.9, 153.0, 155.2,	
		Ar-H), 8.25(s, 1H, CH),	157.2, 158.1, 164.7,	
		9.24(bs, 1H, NH), 9.45(s,	188.5	
		1H, CHO), 10.52(bs, 1H,		
		NH)		
10a	3298, 2995,	1.27(t, J = 6.0 Hz, 3H,	14.2, 43.4, 44.7,	257[M] ⁺
	2971, 1697,	CH ₃), $3.85(q, J = 6.0 \text{ Hz},$	108.2, 126.7, 127.3,	
	1672, 1646	2H, CH ₂), 5.13(s, 2H,	128.0, 137.1, 148.9,	
		CH ₂), 7.20–7.31(m, 5H,	153.3, 152.2, 188.7	
		Ar-H), 8.29(s, 1H, CH),		
		9.12(bs, 1H, NH), 9.43(s,		
		1H, CHO)		

 Table 2

 IR, ¹H NMR, ¹³C NMR, and Mass Spectra of Compounds (*Continued*)

(Continued on next page)

 Table 2

 IR, ¹H NMR, ¹³C NMR, and Mass Spectra of Compounds (Continued)

Cmpd	$IR (cm^{-1})$	1 H NMR (δ)	¹³ C NMR (δ)	Mass Spectra
10b	3296, 3037, 2957, 2863, 1694, 1655	$\begin{array}{l} 0.92(t, J = 5.4 \mathrm{Hz}, 3\mathrm{H}, \\ \mathrm{CH}_3), 1.26(\mathrm{m}, 2\mathrm{H}, \mathrm{CH}_2), \\ 1.67(\mathrm{m}, 2\mathrm{H}, \mathrm{CH}_2), 3.79(t, \\ J = 5.4 \mathrm{Hz}, 2\mathrm{H}, \mathrm{CH}_2), \\ 5.12(\mathrm{s}, 2\mathrm{H}, \mathrm{CH}_2), \\ 7.19-7.28(\mathrm{m}, 5\mathrm{H}, \mathrm{Ar-H}), \\ 8.26(\mathrm{s}, 1\mathrm{H}, \mathrm{CH}), 9.12(\mathrm{bs}, \\ 1\mathrm{H}, \mathrm{NH}), 9.42(\mathrm{s}, 1\mathrm{H}, \\ \mathrm{CHO}) \end{array}$	13.6, 19.2, 30.6, 43.5, 49.2, 108.1, 126.7, 127.3, 128.0, 137.1, 149.1, 153.3, 154.4, 188.8	285[M] ⁺
10c	3294, 3037, 2953, 2873, 1665, 1647	0.96(d, J = 6.4 Hz, 6H, 2CH ₃), 2.01(m, 1H, CH), 3.62(d, J = 6.4 Hz, 2H, CH ₂), 5.13(s, 2H, CH ₂), 7.19–7.28(m, 5H, Ar-H), 8.25(s, 1H, CH), 9.13(bs, 1H, NH), 9.43(s, 1H, CHO)	19.4, 27.7, 43.5, 56.0, 108.0, 126.7, 127.2, 128.0, 137.1, 149.3, 153.2, 154.6, 188.9	285[M] ⁺
10d	3298, 3037, 2960, 1695, 1666, 1649	0.88(t, $J = 6.2$ Hz, 3H, CH ₃), 1.23–1.32(m, 4H, 2CH ₂), 1.67(m, 2H, CH ₂), 3.79(t, $J = 6.2$ Hz, 2H, CH ₂), 5.12(s, 2H, CH ₂), 7.20–7.28(m, 5H, Ar-H), 8.26(s, 1H, CH), 9.12(bs, 1H, NH), 9.42(s, 1H, CHO)	13.9, 21.8, 28.0, 28.2, 43.5, 49.4, 108.1, 126.7, 127.3, 128.0, 137.1, 149.1, 153.3, 154.4, 188.8	299[M] ⁺
10e	3308, 3028, 1708, 1683, 1517,	5.11(s, 2H, CH ₂), 5.13(s, 2H, CH ₂), 7.18–7.27(m, 5H, Ar-H), 7.64(d, $J =$ 9.0 Hz, 2H, Ar-H), 8.24(d, $J =$ 9.0 Hz, 2H, Ar-H), 8.46(s, 1H, CH), 9.22(bs, 1H, NH), 9.46(bs, 1H, CHO)	43.6, 52.0, 108.6, 123.6, 126.7, 127.2, 128.0, 128.4, 136.9, 143.7, 146.8, 149.1, 153.1, 154.4, 188.9	364[M] ⁺
12a	3482, 3440, 3228, 3195, 1689	4.45(s, 2H, CH ₂), 7.38(d, J = 9.0 Hz, 2H, Ar-H), 7.48(bs, 1H, NH), 7.61(d, J = 9.0 Hz, 2H, Ar-H), 7.71(bs, 1H, NH), 7.94(d, 1H, CH), 10.37(bs, 1H, NH)	51.4, 120.4, 126.8, 128.6, 131.2, 131.6, 133.7, 136.8, 137.5, 154.1, 157.8, 166.1	296[M] ⁺

Cmpd	$IR (cm^{-1})$	¹ H NMR (δ)	¹³ C NMR (δ)	Mass Spectra
12b	3549, 3436, 3258, 3191, 3058, 1697	4.47(s, 2H, CH ₂), 7.52(bs, 1H, NH), 7.74(d, $J = 9.6$ Hz, 1H, Ar-H), 7.75(bs, 1H, NH), 8.78(d, $J = 2.4$ Hz, 1H, Ar-H), 7.93(d, 1H, CH), 8.18(d, $J = 2.4$ Hz, 1H, Ar-H), 10.70(bs, 1H, NH)	51.3, 115.1, 121.8, 125.1, 128.3, 131.0, 131.9, 133.5, 136.0, 137.8, 154.7, 157.5, 166.5	364[M] ⁺
12c	3475, 3430, 3269, 3218, 3079, 1689	4.44(s, 2H, CH ₂), 7.12–7.18(m, 2H, Ar-H), 7.47(bs, 1H, NH), 7.56–7.60(m, 2H, Ar-H), 7.70(bs, 1H, NH), 7.94(d, 1H, CH), 10.28(bs, 1H,NH)	51.3, 115.4, 120.6, 131.6, 133.6, 135.0, 136.8, 154.1, 156.2, 157.8, 159.4, 165.9	280[M] ⁺
12d	3462, 3444, 3290, 3281, 3078, 1685	2.19(s, 3H, CH ₃), 4.44(s, 2H, CH ₂), 7.12(d, $J = 9.0$ Hz, 2H, Ar-H), 7.46(d, $J = 9.0$ Hz, 2H, Ar-H), 7.68(bs, 2H, NH2), 7.93(d, 1H, CH), 10.13(bs. 1H, NH)	20.5, 51.3, 118.8, 129.0, 131.2, 131.6, 132.1, 133.6, 136.1, 136.8, 154.1, 157.7, 165.6	276[M] ⁺
12e	3472, 3440, 3285, 3078, 1697, 1612	4.55(s, 2H, CH ₂), 7.26–7.53(m, 3H, Ar-H), 7.55(bs, 1H, NH), 7.71(bs, 1H, NH), 7.96(d, 1H, CH), 10.02(bs, 1H, NH)	51.7, 115.4, 122.9, 125.2, 128.6, 131.4, 132.0, 136.8, 157.5, 159.5, 164.2, 166.3	314[M] ⁺
12f	3402, 3269, 3108, 3020, 1687, 1650	4.54(s, 2H, CH ₂), 7.47(bs, 1H, NH), 7.71(bs, 1H, NH), 7.78–8.12(m, 3H, Ar-H), 8.26(d, 1H, CH), 10.10(bs, 1H, NH)	51.1, 121.0, 124.4, 126.8, 127.2, 128.4, 131.4, 133.7, 134.0, 136.2, 139.0, 154.1, 157.6, 167.8	398[M] ⁺
12g	3549, 3495, 3306, 3059, 1693, 1627	4.55(s, 2H, CH ₂), 7.43(d, <i>J</i> = 9.0 Hz, 1H, Ar-H), 7.56(bs, 1H, NH), 7.67(s, 1H, Ar-H), 7.70(bs, 1H, NH), 7.78(d, <i>J</i> = 9.0 Hz, 1H, Ar-H), 7.94(d, 1H, CH), 9.92(bs, 1H, NH)	51.2, 126.3, 126.5, 127.4, 128.8, 129.1, 131.5, 133.7, 136.8, 154.1, 157.7, 166.7	331[M ⁺¹]

 Table 2

 IR, ¹H NMR, ¹³C NMR, and Mass Spectra of Compounds (*Continued*)

Synthesis of 1-(3-Benzyl-1, 4-dihydro-2-hydroxy-4-iminopyrimidin-5-yl)ethanone 5a–b. General Procedure

To a solution of 3a or 3b (0.0102 mol) in methanol (50 mL), sodium methoxide (0.66 g, 0.0124 mol) was added and refluxed for 6–8 h. After completion of reaction (TLC check, chloroform:methanol, 9:1), the solvent was removed under reduced pressure and the residue obtained was dissolved in cold water and acidified with acetic acid to yield pale yellow solid. Compounds **5a** and **5b** were crystallized from ethanol.

Synthesis of 1-Benzyl-1,2-dihydro-6-methyl-2-oxopyrimidine-5-carbonitrile 6a, b. General Procedure

A solution of **3a** or **3b** (0.004 mol) in acetic acid (10 mL) was refluxed for 24–30 h. After completion of reaction (TLC check, chloroform: methanol, 9:1), acetic acid was removed under reduced pressure and the residue obtained was dissolved in ethyl acetate (10 mL) and washed with a saturated sodium chloride solution. The organic layer were dried over sodium sulfate and evaporated under reduced pressure to give **6a–b** as pale yellow solids, which were recrystalized from ethanol.

Synthesis of (5-Acetyl-3-benzyl-3, 4-dihydro-4-imino-2-oxopyrimidin-1-(2H)-yl)-N-phenylacetamide (8a-n and 12a-g). General Procedure

To a magnetically stirred suspension of **4a** or **4g** or **11** (2.1 mmol) and K_2CO_3 (0.28 g, 2.1 mmol) in *DMF* (4 mL) was added **7a–g** (2.2 mmol). This suspension was further stirred at room temperature for 9–12 hrs (TLC check, chloroform:methanol, 9:1) and the reaction mixture was then quenched in ice-cold water. The colorless solid obtained was collected, washed with water, dried and the solid residue obtained was recrystalized from dichloromethane:heptane (2:8) to give **8a–n** and **12a–g**.

General Procedure for the Synthesis of 10a-e

A suspension of 4g (0.5 g, 2.1 mmol), K₂CO₃ (0.3 g, 2.1 mmol) and alkyl halides **9a–e** (2.2 mmol) in DMF (4 mL) was stirred at room temperature for 6–8 h (TLC monitoring, mobile phase:chloroform:methanol, 9:1). Reaction mass was then quenched in ice-cold water, solid obtained was collected, washed with water, dried and was recrystalized with ethanol to give **10a–e** in 78–90% yield.

Acknowledgements

Authors thanks to CSIR and UGC, New Delhi, India, for financial support. Thanks to Dr. V. B. Gaikwad, Principal, KTHM College, Nashik, for facilities.

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