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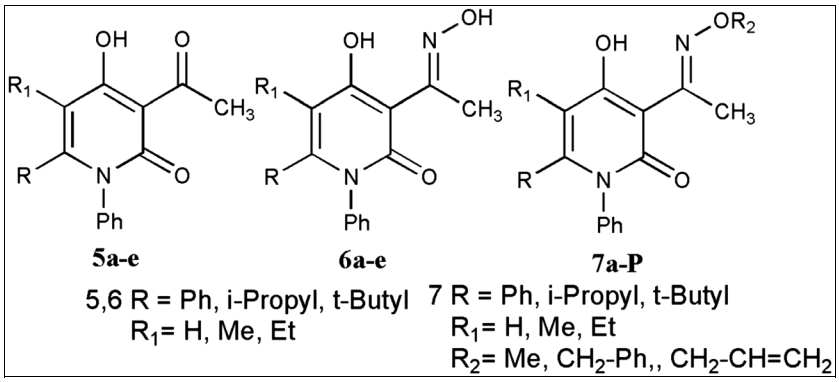

The cyclization of aryl ketone anilides **3** with diethyl malonate to affords 4-hydroxy-6-phenyl-6*H*-pyrano[3,2-*c*]-pyridin-2,5-diones **4** in good yields. 3-Acetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-ones **5** are obtained by ring-opening reaction of 4-hydroxy-6-phenyl-6*H*-pyrano[3,2-*c*]-pyridin-2,5-diones **4** in the presence of 1,2-diethylene glycol. The reaction of 3-acetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-ones **5** with hydroxylamine hydrochloride produces 4-hydroxy-3-[*N*-hydroxyethanimidoyl]-1-phenylpyridin-2(1*H*)-ones **6** from which 3-alkyloxyiminoacetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-ones **7** are obtained by reacting with alkyl bromides or iodides in the presence of anhydrous potassium carbonate with moderate yields. The similar compounds can be synthesized on refluxing 3-acetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-ones **5** with substituted hydroxylamine hydrochloride in the presence of sodium bicarbonate with good yields. Most of the synthesized compounds are characterized by IR and NMR spectroscopic methods.

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INTRODUCTION

4-Hydroxy-pyridon-2-ones are the important class of pyridine derivatives. These compounds are found in natural products having important biological activities such as flavipucin [1], tenellin [2], mocimycin [3], ilicicolin [4], and sambutoxin [5]. 4-Hydory-pyridon-2-ones are also used for the synthesis of nucleotides [6]. Some of them are showing herbicidal activity [7]. The earlier X-ray diffraction studies [8–10] have confirmed that 4-hydroxy-2-pyridones are more stable than 2-hydroxy-4-pyridones. The synthesis of 4-hydroxy-2-pyridones via condensation of enamines or azomethines with reactive malonic acid derivatives has been reported [11,12].

4-Hydroxy-2-pyridones with aliphatic acyl groups in position 3 have found much interest because of their biological properties [13]. A number of methods have been reported for the acylation of 4-hydroxy-2-pyridinones [14]. To obtain 3-acetyl-4-hydroxy-2-pyridinones, we have adopted the method in which azomethines are refluxed with two equivalents of diethyl malonate to yield pyranopyridinones. These pyronopyridones are excellent intermediates in the synthesis of number of compounds having potential biological activity [15]. The degradation of pyranopyridinones in sodium hydroxide and diethylene glycol yields the 3-acetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-ones.

These remarkable importance of 4-hydroxy-pyridin-2-one derivatives and part of developing new synthetical compounds enthused us to develop new methods for the synthesis of 3-acetyl-4-hydroxy-1-phenylpyridin-2 (1*H*)-one derivatives.

RESULTS AND DISCUSSION

The new method for the synthesis of strecker type intermediates **2a–e** is introduced. The nitrile compounds **2a–e** (Table 1) are obtained in excellent yields (85–100%) by adding sodium cyanide to a cooled mixture of ketone **1a–e** and aniline in glacial acetic acid [15]. Elimination of HCN from **2a–e** to yield **3a–e** has been accomplished with potassium hydroxide in refluxing methanol [16]. This method yields the pure azomethines **3a–e** (Table 2), which can be used for further reactions. The mechanism of this reaction has been investigated in kinetic study [17]. It has also been reported that nitrile compounds of type **2a–e**

Table 1

Experimental data of nitriles **2**.

	R	R_1	Yield (%)	mp (°C) solvent	Molecular formula
2a	Ph	H	85	158–160°C petroleum ether	$C_{15}H_{14}N_2$
2b	Ph	Methyl	96	141–142°C petroleum ether	$C_{16}H_{16}N_2$
2c	Ph	Ethyl	100	159–160°C petroleum ether	$C_{17}H_{18}N_2$
2d	i-Propyl	H	98	55–57°C petroleum ether	$C_{12}H_{16}N_2$
2e	i-Butyl	H	92	105–106°C petroleum ether	$C_{13}H_{18}N_2$

Table 2

Experimental data of azomethines **3**.

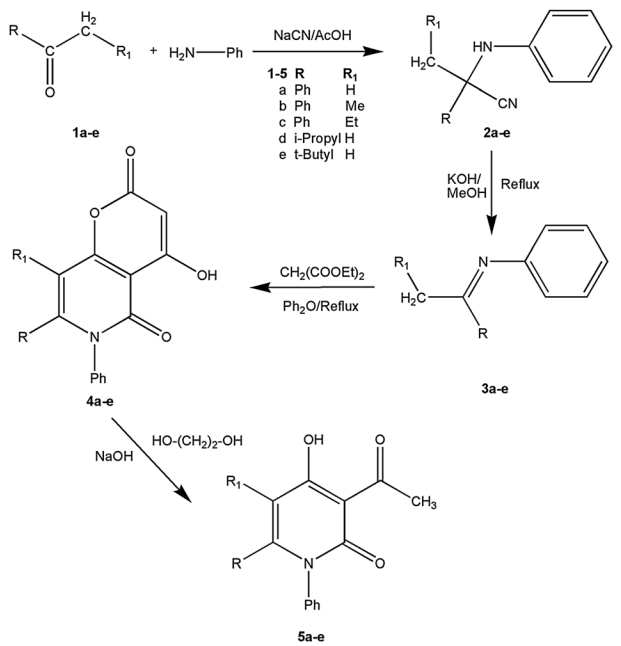
	R	R_1	Yield (%)	mp (°C) solvent	Molecular formula
3a	Ph	H	90	42–43°C petroleum ether	$C_{14}H_{13}N$
3b	Ph	Methyl	91	50–52°C petroleum ether	$C_{15}H_{15}N$
3c	Ph	Ethyl	81	K_{p30}185–187°C petroleum ether	$C_{16}H_{17}N$
3d	i-Propyl	H	91	K_{p30}153–154°C petroleum ether	$C_{11}H_{15}N$
3e	i-Butyl	H	77	K_{p30}160–162°C petroleum ether	$C_{12}H_{17}N$

undergo pyrolytic elimination of HCN at 210°C to afford the azomethines **3a–e** [18].

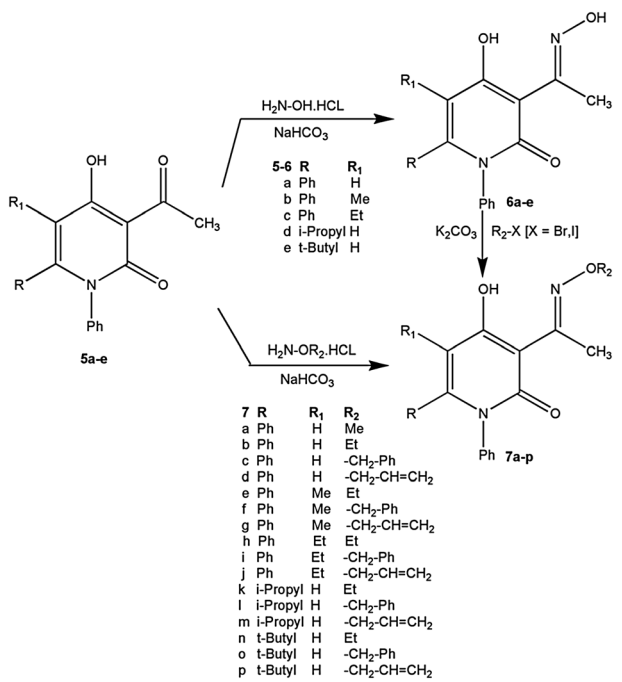
The azomethines can be condensed with substituted dialkylmalonates to give 4-hydroxy-2-pyridones in moderate to good yields [19–21]. The present paper shows that the condensation of azomethines **3a–e** with diethyl malonate satisfactory yields the substituted 4-hydroxy-6-phenyl-6*H*-pyrano[3,2-*c*]pyridin-2,5-diones **4a–e**. Because of lower reactivity of alkyl malonates, reaction time had to rise to several hours. A solution of pyranopyridinedione compounds **4a–e** with 1,2-diethylene glycol was refluxed with sodium hydroxide and acidified strongly with hydrochloric acid to afford the important 3-acetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-ones **5a–e** (Scheme 1) with good yields.

The pyridine compounds **5a–e** were directly converted into 3-alkyloxyiminoacetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-ones **7a–e** or via the conversion into 4-hydroxy-3-hydroxyliminoacetyl-1-phenylpyridin-2(1*H*)-ones **6a–e**. The pyridine compounds **5a–e** on reaction with hydroxyl amine hydrochloride in the presence of sodium bicarbonate yield 4-hydroxy-3-hydroxyliminoacetyl-1-phenylpyridin-2(1*H*)-ones **6a–e**. The oxime compounds **6a–e** on reaction with alkyl bromides or iodides in the presence of anhydrous potassium carbonate convert into 3-alkyloxyiminoacetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-ones **7a–e** with moderate yield. To improve the yield of compounds **7a–e**, the pyridine compounds **5a–e** were directly converted into 3-alkyloxyiminoacetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-ones **7a–e** (Scheme 2) by refluxing with substituted

Scheme 1



Scheme 2



hydroxyl amines in presence of sodium bicarbonate with better yields.

CONCLUSION

In conclusion, we have reported an efficient method for synthesizing variety of substituted 3-acetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-ones and its derivatives. The main practical importance of this reaction is that to report a better method to synthesize 3-alkyloxyiminoacetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-ones.

EXPERIMENTAL

The melting points were determined in open capillary tubes on a Gallenkamp melting point apparatus, MFB 595. The IR spectra were recorded on PerkinElmer 298 spectrophotometer using samples in potassium bromide disks. The ^{1}H NMR spectra (200 MHz) were obtained on a Varian EM-360 spectrometer at 60 MHz with TMS as an internal standard and are given in δ units. The solvent for ^{1}H NMR was DMSO-d_6. Microanalysis was performed on a Carlo Ebra 1106 analyzer. All reactions were monitored by thin layer chromatography and carried out on 0.2 mm silica gel 60 F-254 (Merck) plates using UV light (254 and 366 nm) for detection. Commercially available (E. Merck) chemicals were used without further purification. The UV, IR and C, H, and N analysis were carried out at Institute of Organic Chemistry, Karl-Franzens University, Graz (Austria).

Synthesis of 2-(phenylamino)-nitriles (2a–e). General method: a solution of ketone **1a–e** (0.3 mole) and aniline (33.5 g, 0.36 mole) in 125 mL glacial acetic acid was cooled to 5°C, and then, sodium cyanide (26.7 g) was added portion-wise with stirring for a period of 1 h. Then, it was stirred for 12 h at room temperature. It was then poured on ice–water. The product comes out, which was then filtered, washed with petroleum ether, and dried in air to yield respective nitriles **2a–e**. The results of this reaction are reported in Table 1.

Synthesis of *N*-alkylidene-benzamines (3a–e). General method: 2-(phenyamino)-nitrile **2a–e** (0.3 mole) was boiled in 400 mL of methanol, and then, potassium hydroxide (64.4 g) dissolved in 500 mL methanol was added through the condenser. The solution was then heated for 1 h under reflux. Then, it was diluted with 800 mL water and extracted with 4 × 500 mL petroleum ether. Organic layer is then dried over sodium sulfate for overnight and evaporated on rotary vapor until dryness. The results of this reaction are reported in Table 2.

Synthesis of 4-hydroxy-6-phenyl-*6H*-pyrano[3,2-*c*]pyridine-2,5-diones (4a–e). General method: a solution of *N*-alkylidene-benzamine **3a–e** (0.35 mole) and diethyl malonate (112 g, 0.7 mole) in diphenyl ether (150 mL) placed in 500 mL round bottom flask fitted with vigorous distillation column and distillation assembly. It was then heated to 250–270°C in oil bath until 75–80 mL of ethanol was collected. It was then cooled, digested with methanol, filtered, and washed with methanol and diethyl ether. The solid product was then crystallized to yield respective 4-hydroxy-6-phenyl-*6H*-pyrano[3,2-*c*]pyridine-2,5-diones **4a–e**. Compounds **4a–e** together with their physical constants are listed in the succeeding text.

4-Hydroxy-6,7-diphenyl-6H-pyrano[3,2-c]pyridine-2,5-dione (4a). 58 g (50%), mp 293–296°C (DMF); ^{1}H NMR (DMSO): δ 6.13 (m, 1, Ar-H), 6.4 (s, 1, Ar-H), 7.00–7.30 (m, 10, Ar-H), 15.0 (d, 1, OH). *Anal*. Calcd for $C_{20}H_{13}NO_4$ (331.32): C, 72.50; H, 3.95; N, 4.23. Found: C, 72.48; H, 3.94.40; N, 4.22%.

4-Hydroxy-8-methyl-6,7-diphenyl-6H-pyrano[3,2-c]pyridine-2,5-dione (4b). 45.8 g (70%), mp 271–272°C (acetic acid); IR (KBr) ν_{max}/cm^{-1}: 3060 w, 1725 s, 1670 s, 1600 s, 1550 m, 1480 m; ^{1}H NMR (DMSO): δ 1.89 (s, 3, CH_3), 5.63 (s, 1, Ar-CH_3), 7.20–7.30 (m, 10, Ar-H). *Anal*. Calcd for $C_{21}H_{15}NO_4$ (345.35): C, 73.04; H, 4.38; N, 4.06. Found: C, 73.16; H, 4.40; N, 4.01%.

8-Ethyl-4-hydroxy-6,7-diphenyl-6H-pyrano[3,2-c]pyridine-2,5-dione (4c). 33.5 g (58%), mp 218–219°C (acetic acid); IR (KBr) ν_{max}/cm^{-1}: 3060–2940 w, 1725 s, 1670 s, 1600 w, 1550 s; ^{1}H NMR (DMSO): δ 1.00 (t, J = 7 Hz, 3, CH_3), 2.26 (q, J = 7.5 Hz, 2, CH_2), 5.60 (s, 3, Ar-CH_3), 7.20–7.40 (m, 10, Ar-H), 13.38 (s, 1, OH). *Anal*. Calcd for $C_{22}H_{17}NO_4$ (359.37): C, 73.53; H, 4.77; N, 3.90. Found: C, 73.22; H, 4.87; N, 3.80%.

4-Hydroxy-7-isopropyl-6-phenyl-6H-pyrano[3,2-c]pyridine-2,5-dione (4d). 20 g (31%), mp 205–207°C (acetic acid).

7-tert-Butyl-4-hydroxy-6-phenyl-6H-pyrano[3,2-c]pyridine-2,5-dione (4e). 6.2 g (40%), mp 243–244°C (acetic acid); IR (KBr) ν_{max}/cm^{-1}: 2980 w, 1760 s, 1680 s, 1600 w, 1580 m, 1480 s; ^{1}H NMR (DMSO): δ 1.10 (s, 9, 3CH_3), 5.50 (s, 1, Ar-H), 6.80 (s, 1, Ar-H_8), 7.40–7.70 (m, 5, Ar-H), 12.80 (s, 1, OH). *Anal*. Calcd for $C_{18}H_{17}NO_4$ (311.33): C, 69.44; H, 5.50; N, 4.50. Found: C, 69.34; H, 5.44; N, 4.42%.

Synthesis of 3-acetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-ones (5a–e). General method: a solution of 4-hydroxy-6-phenyl-*6H*-pyrano[3,2-*c*]pyridine-2,5-dione **4a–e** (0.045 mole) and 1,2-diethylene glycol (270 mL), and 18 g sodium hydroxide in 27 mL water was placed in 500 mL round bottom flask and refluxed with stirring for 1 h. The solution was then cooled and added to 1.5 L of ice–water. The solution was acidified with conc. hydrochloric acid to pH = 1. The product was then filtered, dried, and crystallized to yield respective 3-acetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-ones **5a–e**. The compounds **5a–e** together with their physical constants are listed in the succeeding text.

3-Acetyl-4-hydroxy-1,6-diphenylpyridin-2(1H)-one (5a). 12.96 g (95%), mp 120–122°C (ethanol).

3-Acetyl-4-hydroxy-5-methyl-1,6-diphenylpyridin-2(1H)-one (5b). 22 g (95%), mp 203–204°C (ethanol); IR (KBr) ν_{max}/cm^{-1}: 3030–2930 w, 1675 s, 1620 s, 1600 m, 1550 m; ^{1}H NMR (DMSO): δ 1.70 (s, 3, CH_3), 2.68 (s, 3, CH_3 acetyl), 7.10–7.30 (m, 10, Ar-H). *Anal*. Calcd for $C_{20}H_{17}NO_3$ (319.35): C, 75.22; H, 5.37; N, 4.39. Found: C, 75.23; H, 5.33; N, 4.31%.

3-Acetyl-5-ethyl-4-hydroxy-1,6-diphenylpyridin-2(1H)-one (5c). 15 g (90%), mp 163–164°C (ethanol). *Anal*. Calcd for $C_{21}H_{19}NO_3$ (333.38): C, 75.66; H, 5.75; N, 4.20. Found: C, 75.78; H, 5.70; N, 4.15%.

3-Acetyl-4-hydroxy-6-isopropyl-1-phenylpyridin-2(1H)-one (5d). 12.4 g (85%), mp 160–161°C (ethanol).

6-tert-Butyl-3-acetyl-4-hydroxy-1-phenylpyridin-2(1H)-one (5e). 10 g (91%), mp 160–161°C (ethanol); IR (KBr) ν_{max}/cm^{-1}: 2980 w, 1680 s, 1620 s, 1600 m, 1520 m; ^{1}H NMR (DMSO): δ 1.10 (s, 9, 3CH_3), 2.50 (s, 3, CH_3 acetyl), 6.20 (s, 1, Ar-H_5), 7.30–7.60 (m, 5, Ar-H), 15.43 (s, 1, OH). *Anal*. Calcd for $C_{17}H_{19}NO_3$ (285.34): C, 71.56; H, 6.71; N, 4.91. Found: C, 71.78; H, 6.70; N, 4.88%.

Synthesis of 4-hydroxy-3-[*N*-hydroxyethanimidoyl]-1-phenylpyridin-2(1*H*)-ones (6a–e). General method: a solution of 3-acetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-one **5a–e** (0.015 mole), hydroxylamine hydrochloride (1.25 g, 0.018 mole), and sodium bicarbonate (1.51 g, 0.018 mole) was refluxed in 75 mL EtOH–H_2O (2:1) mixture for 1 h. On cooling, the product crystallizes, which was then filtered, dried, and crystallized to yield respective 4-hydroxy-3-[1-(hydroxyimino)ethyl]-1,6-diphynyl-1,2-dihydropyridin-2-ones **6a–e**. The compounds **6a–e** together with their physical constants are listed in the succeeding text.

4-Hydroxy-3-[1-(hydroxyimino)ethyl]-1,6-diphynyl-1,2-dihydropyridin-2-one (6a). 4.1 g (86%), mp 228–229°C (ethanol); IR (KBr) ν_{max}/cm^{-1}: 3190 w, 1645 m, 1605 w, 1560 w, 1400 m; ^1H NMR (DMSO): δ 2.25 (s, 3, CH_3 acetyl), 6.05 (s, 1, Ar-H_5), 7.10–7.30 (m, 10, Ar-H), 11.25 (s, 1, OH). *Anal*. Calcd for $C_{19}H_{16}N_2O_3$ (320.34): C, 71.24; H, 5.03; N, 8.75. Found: C, 71.53; H, 5.04; N, 8.69%.

4-Hydroxy-3-[N-hydroxyethanimidoyl]-5-methyl-1,6-diphenylpyridin-2(1H)-one (6b). 5.38 g (81%), mp 221–222°C (ethanol); IR (KBr) ν_{max}/cm^{-1}: 3170 m, 1640 s, 1600 s, 1550 m, 1490 s; ^1H NMR (DMSO): δ 1.71 (s, 3, CH_3), 2.40 (s, 3, CH_3 acetyl), 7.05–7.30 (m, 10, Ar-H). *Anal*. Calcd for $C_{20}H_{18}N_2O_3$ (334.37): C, 71.84; H, 5.43; N, 8.38. Found: C, 71.22; H, 5.41; N, 8.23%.

5-Ethyl-4-hydroxy-3-[N-hydroxyethanimidoyl]-1,6-diphenylpyridin-2(1H)-one (6c). 2.5 g (72%), mp 208–209°C (acetic acid); ^1H NMR (DMSO): δ 0.94 (t, $J = 7$ Hz, 3, CH_3), 2.12 (q, $J = 7.5$ Hz, 2, CH_2), 2.38 (s, 3, CH_3 acetyl), 7.00–7.30 (m, 10, Ar-H), 11.40 (s, 1 OH), 14.0 (s, 1, OH). *Anal*. Calcd for $C_{21}H_{20}N_2O_3$ (348.4): C, 72.40; H, 5.79; N, 8.04. Found: C, 72.27; H, 5.81; N, 7.97%.

4-Hydroxy-3-[N-hydroxyethanimidoyl]-1-phenyl-6-(propan-2-yl)pyridin-2(1H)-one (6d). 5.62 g (53%), mp 194–195°C (ethanol); IR (KBr) ν_{max}/cm^{-1}: 3140-2970 w, 1640 s, 1600 m, 1560 m, 1400 s; ^1H NMR (DMSO): δ 1.30 (d, $J = 8$ Hz, 6, $2CH_3$), 2.25 (s, 3, CH_3 acetyl), 2.35 (m, 1, CH), 6.05 (s, 1, Ar-H_5), 7.25–7.60 (m, 5, Ar-H), 11.20 (s, 1, OH). *Anal*. Calcd for $C_{16}H_{18}N_2O_3$ (286.33): C, 67.12; H, 6.34; N, 9.78. Found: C, 67.14; H, 6.35; N, 9.67%.

6-tert-Butyl-4-hydroxy-3-[N-hydroxyethanimidoyl]-1-phenylpyridin-2(1H)-one (6e). 3.5 g (58%), mp 195–196°C (ethanol); IR (KBr) ν_{max}/cm^{-1}: 3200-2980 w, 1640 s, 1610 w, 1550 m, 1490 m, 1370 s, 1030 s; ^1H NMR (DMSO): δ 1.04 (s, 9, $3CH_3$), 2.19 (s, 3, CH_3 acetyl), 6.15 (s, 1, Ar-H_5), 7.20–7.60 (m, 5, Ar-H), 11.20 (s, 1, OH), 12.90 (s, 1, OH). *Anal*. Calcd for $C_{17}H_{20}N_2O_3$ (300.35): C, 67.98; H, 6.71; N, 9.33. Found: C, 68.12; H, 6.74; N, 9.20%.

Synthesis of 3-alkyloxyiminoacetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-one compounds by reacting with alkyl bromides or iodides

4-Hydroxy-3-(1-methoxyiminoethyl)-1,6-diphenylpyridin-2(1H)-one (7a). A solution of 4-hydroxy-3-[1-(hydroxyimino) ethyl]-1,6-diphynyl-1,2-dihydropyridin-2-one **6a** (3.2 g, 0.01 mole), methyl iodide (1.42 g, 0.01 mole), and anhydrous potassium carbonate (1.38 g, 0.01 mole) in ethanol (30 mL) was refluxed for 8 h. The solution was then cooled; 25 mL water was added and neutralized with glacial acetic acid. The solid product separated on keeping was filtered, washed with EtOH–H_2O (2:1), dried, and crystallized to yield 1.17 g (50%), mp 116–117°C (ethanol); IR (KBr) ν_{max}/cm^{-1}: 1660 s, 1570 m, 1490 s, 1390 m; ^1H NMR (DMSO): δ 2.20 (s, 3, CH_3 acetyl), 3.88 (s, 3, CH_3), 6.07 (s, 1, Ar-H_5), 7.10–7.30 (m, 10, Ar-H). *Anal*. Calcd for $C_{20}H_{18}N_2O_3$ (334.37): C, 71.84; H, 5.43; N, 8.38. Found: C, 71.33; H, 5.10; N, 8.54%.

3-[1-Ethoxyiminoethyl]-4-hydroxy-1,6-diphenylpyridin-2(1H)-one (7b). A solution of 4-hydroxy-3-[1-(hydroxyimino)ethyl]-1,6-diphynyl-1,2-dihydropyridin-2-one **6a** (3.2 g, 0.01 mole), ethyl iodide (1.56 g, 0.01 mole), and anhydrous potassium carbonate (1.38 g, 0.01 mole) in ethanol (50 mL) was refluxed for 6 h. The solution was then cooled; 25 mL water was added and neutralized with glacial acetic acid. The solid product separated on keeping was filtered, washed with EtOH–H_2O (2:1), dried, and crystallized to yield 1.87 g (54%), mp 131–132°C (ethanol); IR (KBr) ν_{max}/cm^{-1}: 3400 w, 3050 w, 1650 m, 1600 w, 1570 s, 1490 s; ^1H NMR (DMSO): δ 1.30 (t, $J = 7$ Hz, 3, CH_3), 2.15 (s, 3, CH_3 acetyl), 4.15 (q, $J = 7$ Hz, 2, CH_2), 6.08 (s, 1, Ar-H_5), 7.10–7.30 (m, 10, Ar-H), 11.70 (s, 1, OH). *Anal*. Calcd for $C_{21}H_{20}N_2O_3$ (348.4): C, 72.40; H, 5.79; N, 8.04. Found: C, 72.57; H, 5.74; N, 8.07%.

3-[1-Benzyloxyiminoethyl]-4-hydroxy-1,6-diphenylpyridin-2(1H)-one (7c). A solution of 4-hydroxy-3-[1-(hydroxyimino) ethyl]-1,6-diphynyl-1,2-dihydropyridin-2-one **6a** (3.2 g, 0.01 mole), benzyl bromide (1.71 g, 0.01 mole), and anhydrous potassium carbonate (1.38 g, 0.01 mole) in ethanol (25 mL) was refluxed for 6 h. The solution was then cooled; 25 mL water was added and neutralized with glacial acetic acid. The solid product separated on keeping was filtered, washed with EtOH–H_2O (2:1), dried, and crystallized to yield 2.65 g (65%), mp 137–138°C (ethanol); IR (KBr) ν_{max}/cm^{-1}: 3060 w, 1660 m, 1570 w, 1500 w; ^1H NMR (DMSO): δ 2.20 (s, 3, CH_3 acetyl), 5.15 (s, 2, CH_2), 6.05 (s, 1, Ar-H_5), 7.10–7.50 (m, 15, Ar-H), 11.50 (s, 1, OH). *Anal*. Calcd for $C_{26}H_{22}N_2O_3$ (410.46): C, 76.08; H, 5.40; N, 6.83. Found: C, 76.11; H, 5.53; N, 6.68%.

3-[1-Allyloxyiminoethyl]-4-hydroxy-1,6-diphenylpyridin-2(1H)-one (7d). A solution of 4-hydroxy-3-[1-(hydroxyimino)ethyl]-1,6-diphynyl-1,2-dihydropyridin-2-one **6a** (3.2 g, 0.01 mole), allyl bromide (1.21 g, 0.01 mole), and anhydrous potassium carbonate (1.38 g, 0.01 mole) in ethanol (25 mL) was refluxed for 6 h. The solution was then cooled; 25 mL water was added and neutralized with glacial acetic acid. The solid product separated on keeping was filtered, washed with EtOH–H_2O (2:1), dried, and crystallized to yield 2.53 g (79%), mp 97–98°C (ethanol + water); IR (KBr) ν_{max}/cm^{-1}: 3080 w, 3060 s, 2920 w, 2870 w, 1650 m, 1610 m, 1570 w, 1500 s; ^1H NMR (DMSO): δ 2.15 (s, 3, CH_3 acetyl), 4.61 (d, $J = 5$ Hz, 2, CH_2 allyl), 5.29 (t, $J = 7$ Hz, 2, CH_2), 5.40 (m, 1, CH), 6.05 (s, 1, Ar-H_5), 7.10–7.30 (m, 10, Ar-H), 11.65 (s, 1, OH). *Anal*. Calcd for $C_{22}H_{20}N_2O_3$ (360.41): C, 73.32; H, 5.59; N, 7.77. Found: C, 73.52; H, 5.62; N, 7.71%.

3-[1-Ethoxyiminoethyl]-4-hydroxy-5-methyl-1,6-diphenylpyridin-2(1H)-one (7e). A solution of 4-hydroxy-3-[*N*-hydroxyethanimidoyl]-5-methyl-1,6-diphenylpyridin-2(1*H*)-one **6b** (3.34 g, 0.01 mole), ethyl iodide (1.55 g, 0.01 mole), and anhydrous potassium carbonate (0.7 g, 5 mmole) in ethanol (25 mL) was refluxed for 6 h. The solution was then cooled; 25 mL water was added and neutralized with glacial acetic acid. The solid product separated on keeping was filtered, washed with EtOH–H_2O (2:1), dried, and crystallized to yield 2.5 g (69%), mp 117–118°C (ethanol); ^1H NMR (DMSO): δ 1.31 (t, $J = 7$ Hz, 3, CH_3), 1.72 (s, 3, CH_3), 2.34 (s, 3, CH_3 acetyl), 4.22 (q, $J = 7$ Hz, 2, CH_2), 7.10–7.30 (m, 10, Ar-H), 12.95 (s, 1, OH). *Anal*. Calcd for $C_{22}H_{22}N_2O_3$ (362.42): C, 72.91; H, 6.12; N, 7.73. Found: C, 73.12; H, 6.23; N, 7.63%.

3-[1-Benzyloxyiminoethyl]-4-hydroxy-5-methyl-1,6-diphenylpyridin-2(1H)-one (7f). A solution of 4-hydroxy-3-[*N*-hydroxyethanimidoyl]-5-methyl-1,6-diphenylpyridin-2(1*H*)-one **6b** (2.39 g, 7 mmole), benzyl bromide (1.26 g, 7 mole), and anhydrous

potassium carbonate (1.04 g, 7 mmole) in ethanol (30 mL) was refluxed for 6 h. The solution was then cooled; 25 mL water was added and neutralized with glacial acetic acid. The solid product separated on keeping was filtered, washed with EtOH/H_2O (2:1), dried, and crystallized to yield 1.75 g (58%), mp 116–117°C (ethanol); ^{1}H NMR (DMSO): δ 1.67 (s, 3, CH_3), 2.37 (s, 3, CH_3 acetyl), 5.24 (s, 2, CH_2), 7.00–7.50 (m, 15, Ar-H), 12.55 (s, 1, OH). *Anal*. Calcd for $C_{27}H_{24}N_2O_3$ (424.49): C, 76.40; H, 5.70; N, 6.60. Found: C, 76.86; H, 5.75; N, 6.58%.

3-[1-Allyloxyiminoethyl]-4-hydroxy-5-methyl-1,6-diphenylpyridin-2(1H)-one (7g). A solution of 4-hydroxy-3-[*N*-hydroxyethanimidoyl]-5-methyl-1,6-diphenylpyridin-2(1*H*)-one **6b** (2.68 g, 8 mmole), allyl bromide (0.96 g, 8 mmole), and anhydrous potassium carbonate (1.10 g, 8 mmole) in ethanol (35 mL) was refluxed for 6 h. The solution was then cooled; 25 mL water was added and neutralized with glacial acetic acid. The solid product separated on keeping was filtered, washed with EtOH–H_2O (2:1), dried, and crystallized to yield 2.3 g (67%), mp 95–96°C (ethanol + water); IR (KBr) ν_{max}/cm^{-1}: 2920 w, 1650 s, 1610 m, 1550 m, 1220 m; ^{1}H NMR (DMSO): δ 1.70 (s, 3, CH_3), 2.35 (s, 3, CH_3 acetyl), 4.69 (d, J = 5 Hz, 2, CH_2allyl), 5.38 (m, 2, CH_2), 6.05 (m, 1, CH), 7.00–7.25 (m, 10, Ar-H), 12.70 (s, 1, OH). *Anal*. Calcd for $C_{23}H_{22}N_2O_3$ (374.43): C, 73.78; H, 5.92; N, 7.48. Found: C, 73.97; H, 5.99; N, 7.30%.

Synthesis of 3-alkyloxyiminoacetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-one compounds by reacting with alkyloxyamino hydrochlorides

3-[1-Ethoxyiminoethyl]-4-hydroxy-1,6-diphenylpyridin-2(1H)-one (7b). A solution of 3-acetyl-4-hydroxy-1,6-diphenylpyridin-2(1*H*)-one **5a** (4.58 g, 15 mmole), ethoxylamine hydrochloride (1.766 g, 18 mmole), and sodium bicarbonate (1.51 g, 18 mmole) in 75 mL EtOH–H_2O (2:1) was heated under reflux for 1 h. On cooling, the product was crystallized, filtered, dried, and recrystallized to yield 3.65 g (70%), mp 131–132°C (ethanol).

3-[1-Benzyloxyiminoethyl]-4-hydroxy-1,6-diphenylpyridin-2(1H)-one (7c). A solution of 3-acetyl-4-hydroxy-1,6-diphenylpyridin-2(1*H*)-one **5a** (4.58 g, 15 mmole), benzoxylamine hydrochloride (2.85 g, 18 mmole), and sodium bicarbonate (0.92 g, 10 mmole) in 50 mL EtOH–H_2O (2:1) was heated under reflux for 1 h. On cooling, the product was crystallized, filtered, dried, and recrystallized to yield 3.84 g (94%), mp 137–138°C (ethanol).

3-[1-Allyloxyiminoethyl]-4-hydroxy-1,6-diphenylpyridin-2(1H)-one (7d). A solution of 3-acetyl-4-hydroxy-1,6-diphenylpyridin-2(1*H*)-one **5a** (4.58 g, 15 mmole), allyloxylamine hydrochloride (1.84 g, 17 mmole), and sodium bicarbonate (1.43 g, 17 mmole) in 45 mL EtOH–H_2O (2:1) was heated under reflux for 90 min. On cooling, 20 mL water was added. The product was crystallized. It was then filtered, dried, and recrystallized to yield 4.86 g (98%), mp 97–98°C (ethanol + water).

3-[1-Ethoxyiminoethyl]-4-hydroxy-5-methyl-1,6-diphenylpyridin-2(1H)-one (7e). A solution of 3-acetyl-4-hydroxy-5-methyl-1,6-diphenylpyridin-2(1*H*)-one **5b** (3.19 g, 10 mmole), ethoxylamine hydrochloride (1.17 g, 12 mmole), and sodium bicarbonate (1.01 g, 12 mmole) in 45 mL EtOH–H_2O (2:1) was heated under reflux for 90 min. On cooling, the product was crystallized, filtered, dried, and recrystallized to yield 3.1 g (86%), mp 117–118°C (ethanol).

3-[1-Benzyloxyiminoethyl]-4-hydroxy-5-methyl-1,6-diphenylpyridin-2(1H)-one (7f). A solution of 3-acetyl-4-hydroxy-5-methyl-1,6-diphenylpyridin-2(1*H*)-one **5b** (3.19 g, 10 mmole), bezyloxylamine hydrochloride (1.9 g, 12 mmole), and sodium bicarbonate (1.01 g, 12 mmole) in 45 mL EtOH–H_2O (2:1) was heated under reflux for 2 h. On cooling, the product was crystallized, filtered, dried, and recrystallized to yield 2.70 g (64%), mp 116–117°C (ethanol).

3-[1-Allyloxyiminoethyl]-4-hydroxy-5-methyl-1,6-diphenylpyridin-2(1H)-one (7g). A solution of 3-acetyl-4-hydroxy-5-methyl-1,6-diphenylpyridin-2(1*H*)-one **5b** (3.19 g, 0.01 mole), allyloxylamine hydrochloride (1.3 g, 12 mmole), and sodium bicarbonate (1.01 g, 12 mmole) in 45 mL EtOH–H_2O (2:1) was heated under reflux for 2 h. On cooling, the product was crystallized, filtered, dried, and recrystallized to yield 3.10 g (83%), mp 95–96°C (ethanol + water).

3-[1-Ethoxyiminoethyl]-5-ethyl-4-hydroxy-1,6-diphenylpyridin-2(1H)-one (7h). A solution of 3-acetyl-5-ethyl-4-hydroxy-1,6-diphenylpyridin-2(1*H*)-one **5c** (3.33 g, 10 mmole), ethoxylamine hydrochloride (1.17 g, 12 mmole), and sodium bicarbonate (1.01 g, 12 mmole) in 45 mL EtOH–H_2O (2:1) was heated under reflux for 1 h. On cooling, the product was crystallized, filtered, dried, and recrystallized to yield 2.95 g (79%), mp 145–146°C (ethanol + water); IR (KBr) ν_{max}/cm^{-1}: 2980 w, 1650 s, 1605 w, 1550 m, 1210 s; ^{1}H NMR (DMSO): δ 0.94 (t, J = 7 Hz, 3, CH_3 ethyl), 1.30 (t, J = 7 Hz, 3, CH_3 ethyloxy), 2.12 (q, J = 7.5 Hz, 2, CH_2), 2.33 (s, 3, CH_3 acetyl), 4.22 (q, J = 7 Hz, 2, CH_2 ethyloxy), 7.00–7.25 (m, 10, Ar-H), 12.95 (s, 1, OH). *Anal*. Calcd for $C_{23}H_{24}N_2O_3$ (376.45): C, 73.38; H, 6.43; N, 7.44. Found: C, 73.49; H, 6.63; N, 7.12%.

3-[1-Benzyloxyiminoethyl]-5-ethyl-4-hydroxy-1,6-diphenylpyridin-2(1H)-one (7i). A solution of 3-acetyl-5-ethyl-4-hydroxy-1,6-diphenylpyridin-2(1*H*)-one **5c** (3.33 g, 0.01 mole), benzyloxylamine hydrochloride (1.9 g, 12 mmole), and sodium bicarbonate (1.01 g, 12 mmole) in 45 mL EtOH–H_2O (2:1) was heated under reflux for 1 h. On cooling, the product was crystallized, filtered, dried, and recrystallized to yield 3.44 g (79%), mp 129–130°C (ethanol + water); IR (KBr) ν_{max}/cm^{-1}: 3060 w, 1650 s, 1605 m, 1550 m, 1400 m; ^{1}H NMR (DMSO): δ 0.91 (t, J = 7 Hz, 3, CH_3), 2.10 (q, J = 7.5 Hz, 2, CH_2), 2.36 (s, 3, CH_3 acetyl), 5.25 (s, 2, CH_2 benzyl), 7.00–7.50 (m, 15, Ar-H), 12.57 (s, 1, OH). *Anal*. Calcd for $C_{28}H_{26}N_2O_3$ (438.52): C, 76.69; H, 5.98; N, 6.39. Found: C, 76.56; H, 5.99; N, 6.27%.

3-[1-Allyloxyiminoethyl]-5-ethyl-4-hydroxy-1,6-diphenylpyridin-2(1H)-one (7j). A solution of 3-acetyl-5-ethyl-4-hydroxy-1,6-diphenylpyridin-2(1*H*)-one **5c** (3.33 g, 10 mmole), allyloxylamine hydrochloride (1.31 g, 12 mmole), and sodium bicarbonate (1.01 g, 12 mmole) in 45 mL EtOH–H_2O (2:1) was heated under reflux for 1 h. On cooling, the product was crystallized, filtered, dried and recrystallized to yield 2.58 g (67%), mp 128–129°C (ethanol + water); IR (KBr) ν_{max}/cm^{-1}: 3060 w, 1650 s, 1605 w, 1550 s, 1400 s; ^{1}H NMR (DMSO): δ 0.93 (t, J = 7 Hz, 3, CH_3), 2.10 (q, J = 7.5 Hz, 2, CH_2), 2.35 (s, 3, CH_3 acetyl), 4.70 (d, J = 5.5 Hz, 2, CH_2 allyl), 5.35 (m, 2, CH_2), 6.05 (m, 1, CH), 7.00–7.30 (m, 10, Ar-H), 12.70 (s, 1, OH). *Anal*. Calcd for $C_{24}H_{24}N_2O_3$ (388.46): C, 74.21; H, 6.23; N, 7.21. Found: C, 74.44; H, 6.39; N, 6.98%.

3-[1-Ethoxyiminoethyl]-4-hydroxy-6-isopropyl-1-phenylpyridin-2(1H)-one (7k). A solution of 3-acetyl-4-hydroxy-6-isopropyl-1-phenylpyridin-2(1*H*)-one **5d** (2.71 g, 10 mmole), ethoxylamine hydrochloride (1.17 g, 0.012 mole), and sodium bicarbonate (1.01 g, 12 mmole) in 50 mL EtOH–H_2O (2:1) was heated under reflux for 2 h. On cooling, the product was crystallized, filtered, dried and recrystallized to yield 1.58 g (50%), mp 107–108°C (ethanol + water); IR (KBr) ν_{max}/cm^{-1}: 3040–2980 w, 1660 s, 1620 s, 1590 s, 1400 s; ^{1}H NMR (DMSO): δ 1.05 (d, J = 7 Hz, 6, 2CH_3), 1.25 (t, J = 7 Hz, 3, CH_3 ethyloxy), 2.11 (s, 3, CH_3 acetyl), 2.35 (m, 1, CH), 4.10 (q, J = 7 Hz, 2, CH_2), 6.05 (s, 1, Ar-H_5),

7.20–7.60 (m, 5, Ar-H), 11.05 (s, 1, OH). *Anal*. Calcd for $C_{18}H_{22}N_2O_3$ (314.38): C, 68.77; H, 7.05; N, 8.91. Found: C, 68.76; H, 7.11; N, 8.82%.

3-[1-Benzyloxyiminoethyl]-4-hydroxy-6-isopropyl-1-phenylpyridin-2(1H)-one (7l). A solution of 3-acetyl-4-hydroxy-6-isopropyl-1-phenylpyridin-2(1*H*)-one **5d** (2.71 g, 0.01 mole), benzyloxylamine hydrochloride (1.92 g, 12 mmole), and sodium bicarbonate (1.01 g, 12 mmole) in 45 mL EtOH:H_2O (2:1) was heated under reflux for 2 h. On cooling, the product was crystallized, filtered, dried, and recrystallized to yield 2.5 g (67%), mp 127–128°C (ethanol + water); IR (KBr) ν_{max}/cm^{-1}: 3040 w, 2980 m, 2930–2880 w, 1660 s, 1520 s, 1500 w, 1400 s; ^1H NMR (DMSO): δ 1.00 (d, J = 7 Hz, 6, $2CH_3$), 2.15 (s, 3, CH_3 acetyl), 2.35 (m, 1, CH), 5.15 (s, 2, CH_2), 6.00 (s, 1, Ar-H_5), 7.20–7.60 (m, 10, Ar-H), 11.60 (s, 1, OH). *Anal*. Calcd for $C_{23}H_{24}N_2O_3$ (376.45): C, 73.38; H, 6.48; N, 7.44. Found: C, 73.26; H, 6.39; N, 7.29%.

3-[1-Allyloxyiminoethyl]-4-hydroxy-6-isopropyl-1-phenylpyridin-2(1H)-one (7m). A solution of 3-acetyl-4-hydroxy-6-isopropyl-1-phenylpyridin-2(1*H*)-one **5d** (2.47 g, 9 mmole), allyloxylamine hydrochloride (1.12 g, 10 mmole), and sodium bicarbonate (1.01 g, 12 mmole) in 50 mL EtOH:H_2O (2:1) was heated under reflux for 2 h. On cooling, the product was crystallized, filtered, dried, and recrystallized to yield 1.5 g (51%), mp 97–98°C (ethanol + water); IR (KBr) ν_{max}/cm^{-1}: 3070 w, 1650 s, 1605 m, 1400 s, 1200 m; ^1H NMR (DMSO): δ 1.05 (d, *J* = 7 Hz, 6, $2CH_3$), 2.15 (s, 3, CH_3 acetyl), 2.35 (m, 1, CH), 4.60 (d, 2, CH_2 allyl), 5.25 (d, *J* = 7 Hz, 2, CH_2), 5.38 (m, 1, CH), 6.02 (s, 1, Ar-H_5), 7.20–7.60 (m, 5, Ar-H), 11.50 (s, 1, OH). *Anal*. Calcd for $C_{19}H_{22}N_2O_3$ (326.39): C, 69.92; H, 6.79; N, 8.58. Found: C, 69.70; H, 6.81; N, 8.43%.

6-tert-Butyl-3-[1-ethoxyiminoethyl]-4-hydroxy-1-phenylpyridin-2(1H)-one (7n). A solution of 6-*tert*-butyl-3-acetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-one **5e** (2.85 g, 0.01 mole), ethoxylamine hydrochloride (1.19 g, 0.012 mole), and sodium bicarbonate (0.93 g, 0.011 mole) in 50 mL EtOH–H_2O (2:1) was heated under reflux for 1 h. On cooling, the product was crystallized, filtered, dried, and recrystallized to yield 2.4 g (73%), mp 107–108°C (ethanol + water); IR (KBr) ν_{max}/cm^{-1}: 3080 w, 3020 w, 2980 s, 1650 s, 1570 s, 1490 m, 1410 m; ^1H NMR (DMSO): δ 1.05 (s, 9, $3CH_3$), 1.23 (t, *J* = 7 Hz, 3, CH_3), 2.07 (s, 3, CH_3 acetyl), 4.10 (q, *J* = 7 Hz, 2, CH_2), 6.19 (s, 1, Ar-H_5), 7.20–7.50 (m, 5, Ar-H), 11.60 (s, 1, OH). *Anal*. Calcd for $C_{19}H_{24}N_2O_3$ (328.41): C, 69.49; H, 7.37; N, 8.53. Found: C, 69.43; H, 7.34; N, 8.46%.

6-tert-Butyl-3-[1-benzyloxyiminoethyl]-4-hydroxy-1-phenylpyridin-2(1H)-one (7o). A solution of 6-*tert*-butyl-3-acetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-one **5e** (2.85 g, 10 mmole), benzyloxylamine hydrochloride (1.4 g, 11 mmole), and sodium bicarbonate (0.93 g, 11 mmole) in 50 mL EtOH–H_2O (2:1) was heated under reflux for 2 h. On cooling, the product was crystallized, filtered, dried, and recrystallized to yield 2.6 g (67%), mp 127–128°C (ethanol + water); IR (KBr) ν_{max}/cm^{-1}: 2980 m, 1650 s, 1580 m, 1360 s, 1010 s; ^1H NMR (DMSO): δ 1.03 (s, 9, $3CH_3$), 2.12 (s, 3, CH_3 acetyl), 5.13 (s, 2, CH_2), 6.15 (s, 1, Ar-H_5), 7.20–7.50 (m, 10, Ar-H), 11.43 (s, 1, OH). *Anal*. Calcd for $C_{24}H_{26}N_2O_3$ (390.47): C, 73.82; H, 6.71; N, 7.17. Found: C, 74.63; H, 6.79; N, 7.01%.

6-tert-Butyl-3-[1-allyloxyiminoethyl]-4-hydroxy-1-phenylpyridin-2(1H)-one (7p). A solution of 6-*tert*-butyl-3-acetyl-4-hydroxy-1-phenylpyridin-2(1*H*)-one **5e** (2.85 g, 10 mmole), allyloxylamine hydrochloride (1.24 g, 11 mmole), and sodium bicarbonate (0.93 g, 11 mmole) in 45 mL EtOH–H_2O (2:1) was heated under reflux for 1 h. On cooling, the product was crystallized, filtered, dried, and recrystallized to yield 2.4 g (47%), mp 118–119°C (ethanol + water); IR (KBr) ν_{max}/cm^{-1}: 2080 w, 1650 s, 1605 w, 1570 m, 1550 m, 1490 w; ^1H NMR (DMSO): δ 1.04 (s, 9, $3CH_3$), 2.10 (s, 3, CH_3 acetyl), 4.58 (d, *J* = 5.5 Hz, 2, CH_2 allyl), 5.26 (m, 2, CH_2), 6.00 (m, 1, CH), 6.18 (s, 1, Ar-H_5), 7.20–7.50 (m, 5, Ar-H), 11.50 (s, 1, OH). *Anal*. Calcd for $C_{20}H_{24}N_2O_3$ (340.42): C, 70.57; H, 7.11; N, 8.23. Found: C, 70.95; H, 7.17; N, 7.90%.

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