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Reactivity of bis(Arylcarbamoyl)-*N*-arylphenacylamine Oximes. Synthesis of 1,3-Dihydroimidazol-2-ones and *N*-Unsubstituted *O*-Arylcarbamoylhydroxylamines

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Abstract: *N*-Arylphenacylamine oximes **1** reacted with aryl isocyanates in refluxing benzene to give bis(arylcarbamoyl)-*N*-arylphenacylamine oximes **2** in good yields. Compounds **2** reacted with equimolar amount of $TsOH.H_2O$ in THF at room temperature to give imidazol-2-ones **5** and *O*-arylcarbamoylhydroxylammonium tosylates **6** in high yields. Compounds **2** were heated under vacuum to induce Beckmann fragmentation but the resulting products were 1,3,4-triaryl-1,3-dihydroimidazol-2-ones **5** instead of the expected 1,3-diazetidinones **4**.

Recently we have reported the synthesis of 5,6,7,7a-tetrahydroimidazo[1,5-*b*][1,2,4]oxadiazol-2(1*H*)-ones by the reaction of 1,4-diaryl- and 1,2,4-triaryl-imidazoline 3-oxides with aryl isocyanates.[1] In a continuing investigation on the reaction of *N*-aryl-*N,N*-diphenacylamine dioximes with aryl isocyanates,[2] we have observed that iminium intermediates formed as a result of Beckmann fragmentation[3,4] of the dicarbamoylated *N*-aryl-*N,N*-diphenacylamine dioximes cyclize to give corresponding imidazoline 3-oxides which react with aryl isocyanate to give corresponding tetrahydroimidazooxadiazolone. To exploit the same reactivity in the synthesis of 1,3-diazetidin-2-ones **4** we aimed to prepare compounds **2** starting from the corresponding *N*-arylphenacylamine oximes **1** (Scheme 1).

To our knowledge, no investigations on the Beckmann fragmentation of bis(arylcarbamoyl)-*N*-arylphenacylamine oximes **2** nor on the intramolecular cyclization of these compounds to imidazol-2-ones have been reported.

We report herein the synthesis of bis(arylcarbamoyl)-*N*-arylphenacylamine oximes **2** which were proved to be excellent precursors in the synthesis of *N*-unsubstituted *O*-arylcarbamoylated hydroxylamines and 1,3-dihydroimidazol-2-ones. Compounds **6** are inaccessible by the reaction of hydroxylamine with aryl isocyanates.

Oximes **1a-e**, prepared by the reaction of corresponding *N*-arylphenacylamines with hydroxylamine hydrochloride in the presence of sodium acetate in ethanol, were refluxed in benzene in the presence of two equivalents of aryl isocyanate to give corresponding bis(arylcarbamoyl)-*N*-arylphenacylamine oximes **2**.

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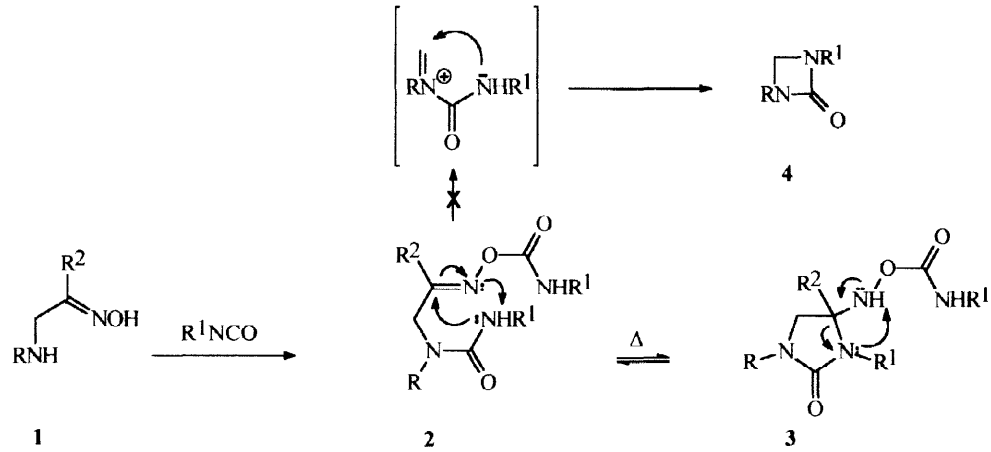


Scheme 1

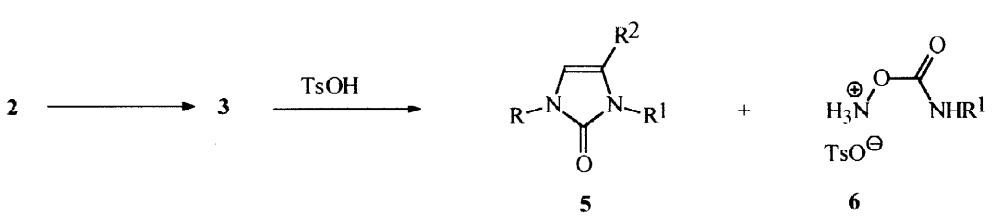
Compounds **2** were refluxed in benzene for 24 h but no conversion to **4** occurred. The starting oximes **2** were recovered unchanged. There are at least two possible reasons why the compounds **2** did not undergo Beckmann fragmentation. The urea type nitrogen may be not basic enough to favour the formation of iminium species or the heating of compounds **2** may lead to the formation of imidazolidinones **3** which are unable to undergo the expected fragmentation. The ^{1}H NMR spectra of the compounds **2** recorded in $CDCl_3$ revealed singlets at approximately δ 5.25 assigned to CH_2 next to the oxime C=N double bond. However, the ^{1}H NMR spectrum of **2a** dissolved in DMSO-d_6 heated on a water bath shows the presence of **2a** together with the main product **3a** in 1:2 ratio. Beside the singlet at δ 5.25 characteristic for compounds **2** an AB system centered at δ 4.25 (J_{AB} = 11.0 Hz) indicates the presence of imidazolidinone **3a**.[5] This fact confirms our assumption that compounds **2** could not undergo fragmentation because the favoured process is the intramolecular nucleophilic addition of the amide nitrogen to the oxime C=N double bond (Scheme 1). It is probable that at temperatures higher than room temperature the compounds **2** are in equilibrium with **3** . The conversion of **2** into **3** on heating may proceed most probably as a simultaneous process as illustrated in Scheme 1. The formation of imidazol-2-ones by heating the compounds **2** in solvent-free conditions under vacuum can be rationalised by assuming the thermal conversion of **2** to **3** (see Table 2, method B for the yields). It was impossible to isolate the corresponding *O*-arycarbamoylated hydroxylamine in this method due to their instability. However the reaction of compounds **2a-g** with $TsOH.H_2O$ in THF at room temperature gave almost quantitatively an easily separable mixture of the corresponding *O*-arylcarbamoylhydroxylammonium tosylates and imidazol-2-ones (Table 2). Probably, in this reaction the TsOH shifts the equilibrium to **3** which on protonation eliminates **6** (Scheme 2). *N*-Unsubstituted *O*-acylhydroxylamines are accessible by the hydrolysis of *O*-acylated hydroxymates or by HCl-cleavage of *O*-acylated *tert*-butyl-*N*-hydroxycarbamates.[6] *O*-Arylcarbamoyl-hydroxylamine tosylates were characterised by spectral means as well as converting them to compounds

known in the literature. *O*-Phenylcarbamoylhydroxylammonium tosylate reacted with phenyl isocyanate to give *N,O*-bis(phenylcarbamoyl)hydroxylamine.[7] *O*-Carbamoylhydroxylamine derivatives are known to possess antifungal and antimalarial activity.[8]

Table 1. 4-Arylcarbamoyloxaminoimidazolidin-2-one equivalents.

Starting Material	Product	R	R^1	R^2	Yield (%)	mp (°C)	IR (KBr) ν_{NH}	IR (KBr) $\nu_{C=O}$
1a	**2a**	Ph	Ph	Ph	65	161.5	3355, 3309	1670, 1736
1b	**2b**	p-MeC_6H_4	Ph	Ph	75	135.5	3355, 3309	1670, 1736
1c	**2c**	p-MeO C_6H_4	Ph	Ph	70	146-147	3355, 3309	1670, 1736
1d	**2d**	p-MeO C_6H_4	Ph	p-$MeOC_6H_4$	62	165-166	3355, 3309	1670, 1736
1e	**2e**	p-MeC_6H_4	Ph	p-$MeOC_6H_4$	66	166.5	3355, 3309	1670, 1736
1a	**2f**	Ph	p-$MeOC_6H_4$	Ph	65	156-157	3419, 3253	1673, 1728
1b	**2g**	p-MeC_6H_4	p-$MeOC_6H_4$	Ph	70	137-138	3419, 3253	1673, 1728

To prove the structure of compounds **5** by a chemical method we have prepared them by heating *N*-arylphenacylamines with aryl isocyanates.[9] The products obtained in this way were identical with those obtained by the reaction of compounds **2** with TsOH as well as with those obtained by heating them under vacuum.



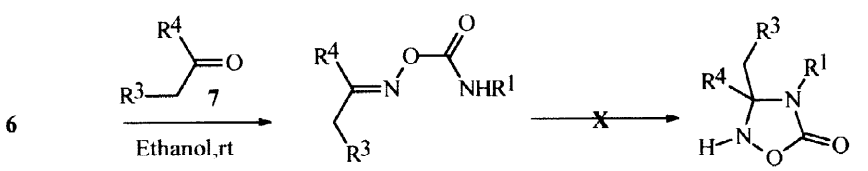
5 **6**

Scheme 2

Table 2. 1,3,4-Triaryl-1,3-dihydroimidazol-2-ones.

Starting Material	Product	Mol. Formula or lit. mp	Yield (%) Method A	Method B	mp (°C)	IR (KBr) $\nu_{C=O}$	MS *m/z* (M^+)
2a	**5a**	164-165[9]	90	85	164-165	1700	312
2b	**5b**	165-166[9]	93	80	165-166	1700	326
2c	**5c**	$C_{22}H_{18}N_2O_2$ (342.39)	90	78	170	1700	342
2d	**5d**	$C_{23}H_{20}N_2O_3$ (372.42)	95	75	180-181	1700	372
2e	**5e**	$C_{23}H_{20}N_2O_2$ (356.42)	92	77	106-107	1700	356
2f	**5f**	$C_{22}H_{18}N_2O_2$ (342.39)	93	85	168-168.5	1700	342
2g	**5g**	$C_{23}H_{20}N_2O_2$ (356.42)	94	87	196-197	1700	356

O-Arylcarbamoylated hydroxylammonium tosylates react with ketones **7** in ethanol at room temperature to give corresponding carbamoylated oximes **8** in almost quantitative yields. We expected that at higher temperature this reaction could lead to the formation of oxadiazolidin-5-ones **9**. However, the products obtained from the reaction of carbamoylated hydroxylamine tosylates with ketones at reflux in THF were the same as those obtained at room temperature. Compounds **8a,g** were prepared from the reaction of the corresponding oximes with phenyl isocyanate and proved to be identical with those obtained from the reaction of carbamoylated hydroxyl amines with ketones.



Scheme 3

	R^1	R^3	R^4		R^1	R^3	R^4
a	Ph	H	Me	**e**	4-$MeOC_6H_4$	$-CH_2(CH_2)_2CH_2-$	
b	Ph	Me	Me	**f**	4-$MeOC_6H_4$	H	3,4-$(MeO)_2C_6H_3$
c	Ph	H	Ph	**g**	Ph	Br	Ph
d	Ph	$-CH_2(CH_2)_2CH_2-$		**h**	Ph	H	3,4-$(MeO)_2C_6H_3$

EXPERIMENTAL

N-Arylphenacylamines were prepared by the reaction of the amine with phenacyl bromide (2:1 mole ratio) in benzene at room temperature. These compounds were converted into oximes in ethanol at reflux using hydroxylamine hydrochloride in the presence of sodium acetate. Aryl isocyanates were purchased from Aldrich and used without additional purification. Melting points were determined on a Electrothermal digital melting point apparatus and are uncorrected. IR spectra of the compounds were taken on a Mattson FTIR. ^{1}H NMR spectra of the compounds were recorded on a Bruker instrument (200 MHz). Mass spectra were recorded with Hewlett-Packard GC-MS.

Bis(Arylcarbamoyl)-*N*-arylphenacylamine oximes 2. General Procedure. To a solution of *N*-arylphenacylamine oxime (10 mmol) in benzene (50 mL) aryl isocyanate (20 mmol) was added and the mixture stirred at reflux for 45 min. The solvent was removed under vacuum. Ether (50 mL) was added to the residue and heated under reflux for 5 min. The solution was left to cool at room temperature. The crystalline product was filtered, washed with ether and dried under vacuum at room temperature.

2a. ^{1}H NMR ($CDCl_3$) δ 5.40 (2H, s), 6.15 (1H, s, NH), 7.00-7.85 (20H, m), 8.12 (1H, s, NH). Anal. Calcd for $C_{28}H_{24}N_4O_3$ (464.52): C, 72.40; H, 5.20; N, 12.06. Found: C, 72.30; H, 5.24; N, 11.91.

2b. ^{1}H NMR ($CDCl_3$) δ 2.32 (3H, s), 5.40 (2H, s), 6.20 (1H, s, NH), 6.90-7.80 (19H, m), 8.12 (1H, brs, NH). Anal. Calcd for $C_{29}H_{26}N_4O_5$ (478.53): C, 72.78; H, 5.48; N, 11.71. Found: C, 72.80; H, 5.52; N, 11.68.

2c. ^{1}H NMR ($CDCl_3$) δ 3.77 (3H, s), 5.35 (2H, s), 6.15 (1H, s, NH), 6.84-7.80 (19H, m), 8.10 (1H, s, NH). Anal. Calcd for $C_{29}H_{26}N_4O_4$ (494.54): C, 70.43; H, 5.30; N, 11.33. Found: C, 70.35; H, 5.40; N, 11.35.

2d. ^{1}H NMR ($CDCl_3$) δ 3.77 (3H, s), 3.87 (3H, s), 5.35 (2H, s), 6.17 (1H, s, NH), 6.80-7.80 (18H, m), 8.10 (1H, brs, NH). Anal. Calcd for $C_{30}H_{28}N_4O_5$ (524.57): C, 68.69; H, 5.38; N, 10.68. Found: C, 68.75; H, 5.39; N, 10.74

2e. ^{1}H NMR (DMSO-d_6) δ 2.22 (3H, s), 3.80 (3H, s), 5.24 (2H, s), 6.82-7.78 (19H, m), 9.60 (1H, s, NH). Anal. Calcd for $C_{30}H_{28}N_4O_4$ (508.57): C, 70.85; H, 5.55; N, 11.01. Found: C, 70.80; H, 5.49; N, 10.05

2f. ^{1}H NMR (DMSO-d_6) δ 3.74 (3H, s), 3.78 (3H, s), 5.40 (2H, s), 6.94-7.98 (19H, m), 9.60 (1H, s, NH). Anal. Calcd for $C_{30}H_{28}N_4O_5$ (524.57): C, 68.69; H, 5.37; N, 10.68. Found: C, 68.37; H, 5.52; N, 10.34.

2g. ^{1}H NMR ($CDCl_3$) δ 2.35 (3H, s), 3.77 (3H, s), 3.80 (3H, s), 5.40 (2H, s), 6.05 (1H, s, NH), 6.75-7.85 (17H, m), 8.05 (1H, brs, NH). Anal. Calcd for $C_{31}H_{30}N_4O_5$ (538.60): C, 69.13; H, 5.61; N, 10.40. Found: C, 69.17; H, 5.55; N, 10.43

Reaction of compounds 2a-g with TsOH.H_2O. General Procedure. Method A. To a solution of compound **2** (2 mmol) in THF (12 mL) TsOH.H_2O (dissolved in 2 mL of THF) was added dropwise at room temperature. The white solid precipitated after 3-5 min. The mixture was stirred for 3 h at room temperature and filtered. The solid was characterized to be *O*-arylcarbamoylhydroxylamine tosylate **6**.[10] The filtrate was evaporated and the residue was dissolved in hot ethanol and left to crystallize. The needle shaped crystals were filtered to give the corresponding 1,3-dihydroimidazol-2-one in almost quantitative yield.

***O*-Phenylcarbamoylhydroxylammonium tosylate.** Mp 149-150 °C dec. (from ethanol). Lit.[11] mp for *O*-phenylcarbamoylhydroxylammonium chloride is 110-112 °C. IR (KBr) ν $_{C=O}$ 1778 cm^{-1}. ^{1}H NMR DMSO-d_6 δ 2.24 (3H, s), 7.10-7.90 (12H, m), 10.90 (1H, s). Anal. Calcd for $C_{14}H_{16}N_2O_5S$ (324.35): C, 51.84; H, 4.97; N, 8.64. Found: C, 51.80; H, 5.05; N, 8.70

***O*-4-Methoxyphenylcarbamoylhydroxylammonium tosylate.** Mp 140 °C dec. IR (KBr) ν $_{C=O}$ 1792 cm^{-1}. ^{1}H NMR DMSO-d_6 δ 2.24 (3H, s), 3.70 (3H, s) 6.80-7.90 (11H, m), 10.34 (1H, s). Anal. Calcd for $C_{15}H_{18}N_2O_6S$ (354.37): C, 50.84; H, 5.12; N, 7.90. Found: C, 50.75; H, 5.10; N, 7.93

Reaction of *O*-phenylcarbamoylhydroxylammonium tosylate with phenyl isocyanate. *O*-Phenylcarbamoylhydroxylammonium tosylate (0.323 g, 1 mmol) was suspended in THF (10 mL). Phenyl isocyanate (0.238 g, 2 mmol) was added dropwise and the mixture refluxed for 2 h. The reaction mixture was poured into water and the amorphous solid was filtered. The compound was dried under vacuum and redissolved in hot chloroform. The solution was filtered and left to crystallize at room temperature. The needle shaped crystals were collected by filtration. Yield 60%. The compound melts at 170 °C lit.[7] mp 164-165. The IR spectrum of the compound was identical with the spectrum of known *N,O*-bisphenylcarbamoylhydroxylamine.[10]

Thermal treatment of compounds 2a-g: 1,3,4-Triaryl-1,3-dihydroimidazol-2-ones 5a-g. General Procedure. Method B.

Compound **2** (0.043 mmol) was placed in a sample vial and heated in a vacuum oven at 175 °C under vacuum for 10 min. The mixture was cooled and dissolved in chloroform (0.5 mL), then separated on a silica gel by preparative TLC, developed with chloroform. The band containing the imidazol-2-one was collected, the solvent was evaporated and the product was recrystallized from ether or ethanol.

5c. ^{1}H NMR $CDCl_3$ δ 3.80 (3H, s, MeO), 6.80 (1H, s, CH-5), 6.85 (2H, d, J = 8.0, Ar), 7.10-7.50 (10H, m, Ar), 7.71 (2H, d, J = 8.0, Ar). MS *m/z* 342 (M^+, 100). Anal. Calcd for $C_{22}H_{18}N_2O_2$ (342.39): C, 77.17; H, 5.30; N, 8.18. Found: C, 77.10; H, 5.35; N, 8.10

5d. ^{1}H NMR $CDCl_3$ δ 3.78 (3H, s, MeO), 3.86 (3H, s, MeO), 6.69 (1H, s, CH-5), 6.81 (2H, d, J = 8.0, Ar), 6.93-7.14 (3H, m, Ar), 7.24-7.50 (5H, m, Ar), 7.59 (2H, d, J = 8.0, Ar). MS *m/z* 372 (M^+, 100). Anal. Calcd for $C_{23}H_{20}N_2O_3$ (372.42): C, 74.17; H, 5.41; N, 7.52. Found: C, 74.11; H, 5.38; N, 7.58

5e. ^{1}H NMR $CDCl_3$ δ 2.40 (3H, s, Me), 3.80 (3H, s, MeO), 6.70 (1H, s, CH-5), 6.80 (2H, d, J = 8.0, Ar), 7.05 (2H, d, J = 8.0), 7.20-7.40 (5H, m, Ar), 7.60 (2H, d, J = 8.0, Ar). MS *m/z* 356 (M^+, 100). Anal. Calcd for $C_{23}H_{20}N_2O_2$ (356.42): C, 77.50; H, 5.66; N, 7.86. Found: C, 77.45; H, 5.60; N, 7.81

5f. ^{1}H NMR $CDCl_3$ δ 3.80 (3H, s, MeO), 6.80 (1H, s, CH-5), 6.85 (2H, d, J = 8.0, Ar), 7.10-7.50 (10H, m, Ar), 7.71 (2H, d, J = 8.0, Ar). MS *m/z* 342 (M^+, 100). Anal. Calcd for $C_{22}H_{18}N_2O_2$ (342.39): C, 77.17; H, 5.30; N, 8.18. Found: C, 77.10; H, 5.35; N, 8.10

5g. ^{1}H NMR $CDCl_3$ δ 2.37 (3H, s, Me), 3.80 (3H, s, MeO), 6.77 (1H, s, CH-5), 6.90 (2H, d, J = 8.0, Ar), 7.15-7.30 (9H, m, Ar), 7.59 (2H, d, J =8.0, Ar). MS *m/z* 356 (M^+, 100). Anal. Calcd for $C_{23}H_{20}N_2O_2$ (356.42): C, 77.50; H, 5.66; N, 7.86. Found: C, 77.92; H, 5.72; N, 7.73

Reaction of arylcarbamoylhydroxylammonium tosylates 6 with ketones:

8a. *O*-Phenylcarbamoylhydroxylammonium tosylate (0.324 g, 1 mmol) was dissolved in acetone (3 mL) and stirred for 15 min at room temperature. Water (10 mL) was added and the solution was left to crystallize at room temperature. The white needles were collected by filtration and dried under vacuum. Yield 98%. Mp 111 °C. Lit.[11] mp 109 °C. IR (KBr) ν_{NH} 3228 cm^{-1}; $\nu_{C=O}$ 1719 cm^{-1}; $\nu_{C=N}$ 1643 cm^{-1}. ^{1}H NMR ($CDCl_3$) δ 2.04 (3H, s), 2.09 (3H, s), 7.10 (1H, t, J = 6.5), 7.36 (2H, t, J = 6.5), 7.51 (2H, d, J = 6.5), 8.23 (1H, brs, NH).

8b. From the reaction of **O**-phenylcarbamoylhydroxylammonium tosylate with methylethyl ketone. Yield 97%. Mp 125-127 °C. IR (KBr) ν_{NH} 3228 cm^{-1}; $\nu_{C=O}$ 1719 cm^{-1}; $\nu_{C=N}$ 1643cm^{-1}. ^{1}H NMR ($CDCl_3$) δ 1.20 (3H, t, J =

8.0), 2.06 (3H, s), 2.42 (2H, q, J = 8.0), 7.13 (1H, t, J = 6.5), 7.37 (2H, t, J = 6.5), 7.51 (2H, d, J = 6.5), 8.32 (1H, brs, NH). Anal. Calcd for $C_{11}H_{14}N_2O_2$ (206.24): C, 64.06; H, 6.84; N, 13.58. Found: C, 64.16; H, 6.78; N, 11.61

8c. To a solution of *O*-phenylcarbamoylhydroxylammonium tosylate (0.162 g, 0.5 mmol) in ethanol (3 mL) acetophenone (0.060 g, 0.5 mmol) was added and the mixture stirred at room temperature for 1.5 h. Water (6 mL) was added to the reaction mixture and left to crystallize at room temperature. The crystals were filtered and dried under vacuum.Yield 95%. Mp 127-129 °C. IR (KBr) ν_{NH} 3228 cm^{-1}; $\nu_{C=O}$ 1719 cm^{-1}; $\nu_{C=N}$ 1618 cm^{-1}. ^1H NMR ($CDCl_3$) δ 2.51 (3H, s), 7.15 (1H, t, J = 6.5), 7.39 (2H, t, J = 6.5), 7.50 (5H, m), 7.74 (2H, m), 8.44 (1H, brs, NH). Anal. Calcd for $C_{15}H_{14}N_2O_2$ (254.28): C, 70.85; H, 5.55; N, 11.01. Found: C, 70.80; H, 5.60; N, 11.09

8d. To a solution of *O*-phenylcarbamoylhydroxylammonium tosylate (0.080 g, 0.25 mmol) in ethanol (3 mL) cyclohexanone was added (0.049 g, 0.5 mmol). The mixture was stirred at room temperature for 30 min. The white solid formed was filtered washed with ethanol and dried under vacuum. Yield 97%. Mp 153-154 °C. IR (KBr) ν_{NH} 3228 cm^{-1}; $\nu_{C=O}$ 1719 cm^{-1}; $\nu_{C=N}$ 1643cm^{-1}. ^1H NMR ($CDCl_3$) δ 1.66 (6H, m), 2.39 (2H, m), 2.66 (2H, m), 7.13 (1H, t, J = 6.5), 7.36 (2H, t, J = 6.5), 7.54 (2H, d, J = 6.5), 8.31 (1H, brs, NH). Anal. Calcd for $C_{13}H_{16}N_2O_2$ (232.28): C, 67.22; H, 6.94; N, 12.06. Found: C, 67.30; H, 6.89; N, 11.97

8e. To a solution of *O*-4-methoxyhenylcarbamoylhydroxylamine tosylate (0.088 g, 0.25 mmol) in ethanol (3.5 mL) cyclohexanone was added (0.049 g, 0.5 mmol). The mixture was stirred at room temperature for 30 min then water was added (6 mL). The crystals were collected by filtration and dried under vacuum. Yield 98%. Mp 103-104 °C. IR (KBr) ν_{NH} 3228 cm^{-1}; $\nu_{C=O}$ 1719 cm^{-1}; $\nu_{C=N}$ 1643cm^{-1}. ^1H NMR ($CDCl_3$) δ 1.66 (6H, m), 2.36 (2H, m), 2.72 (2H, m), 3.81 (3H, s), 6.87 (2H, d, J = 9.0), 7.39 (2H, d, J = 9.0), 8.19 (1H, brs, NH). Anal. Calcd for $C_{14}H_{18}N_2O_3$ (262.30): C, 64.10; H, 6.92; N, 10.68. Found: C, 64.15; H, 6.87; N, 10.55

8f. To a solution of *O*-4-methoxyphenylcarbamoylhydroxylamine tosylate (0.088 g, 0.25 mmol) in ethanol (3.5 mL) 3,4-dimethoxyacetophenone (0.045 g, 0.25 mmol) was added and the reaction mixture stirred at room temperature for 1.5 h. Water (20 mL) was added to the mixture which was then extracted with chloroform (2 x 15 mL). The combined extracts were dried over anhydrous Na_2SO_4 and filtered. The solvent was evaporated and the residue was dissolved at heating in a mixture of benzene and petroleum ether (1:1) and left to crystallize at room temperature. Yield 60%. Mp 76-77 °C. IR (KBr) ν_{NH} 3250 cm^{-1}; $\nu_{C=O}$ 1730 cm^{-1}. ^1H NMR ($CDCl_3$) δ 2.48 (3H, s), 3.81 (3H, s), 3.93 (3H, s), 3.99 (3H, s), 6.89 (3H, m), 7.39 (4H, m), 8.32 (1H, brs, NH). Anal. Calcd for $C_{18}H_{20}N_2O_5$ (344.36): C, 62.78; H, 5.85; N, 8.13. Found: C, 62.70; H, 5.75; N, 8.10

8g. The procedure is as for **8f**. The compound was recrystallized from ether petroleum ether (1:1). Yield 89%. Mp 113-114 °C. IR (KBr) ν_{NH} 3250 cm^{-1}; $\nu_{C=O}$ 1733 cm^{-1}; $\nu_{C=N}$ 1620 cm^{-1}. ^1H NMR ($CDCl_3$) δ 4.50 (2H, s), 7.10 (1H, t, J = 6.5), 7.39 (2H, t, J = 6.5), 7.59 (5H, m), 7.81 (2H, d, J = 9.0), 8.28 (1H, brs, NH). Anal. Calcd for $C_{15}H_{13}N_2O_2Br$ (333.18): C, 54.07; H, 3.93; N, 8.41. Found: C, 54.10; H, 3.85; N, 8.38

8h. The procedure is as for **8c**. Recrystallized from benzene. Yield 97%. Mp 121-122 °C. IR (KBr) ν_{NH} 3336 cm^{-1}; $\nu_{C=O}$ 1745 cm^{-1}. ^1H NMR ($CDCl_3$) δ 2.48 (3H, s), 3.93 (3H, s), 3.99 (3H, s), 6.92 (1H, d, J = 9.0), 7.15 (1H, t, J = 6.5), 7.37-7.60 (6H, m), 8.40 (1H, brs, NH). Anal. Calcd for $C_{17}H_{18}N_2O$ (314.34): C, 64.96; H, 5.77; N, 8.91. Found: C, 64.90; H, 5.68; N, 8.88

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REFERENCES AND NOTES

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