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Reactions of 9-Alkyl-3-aminocarbazoles with Ethyl-3-oxo-butanoate and Identification of the Products Obtained

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Abstract: The reactions in benzene of 9-alkyl-3-aminocarbazoles with ethyl-3-oxobutanoate yielded ethyl-3-[(9-alkyl-9H-carbazol-3-yl)amino]but-2-enoate condensation products or N-(9-ethyl-9H-carbazol-3-yl)-3-oxobutanamide acylation products. The condensation products were cyclized to the corresponding 4,7-dihydro-pyrido[2,3-c]-carbazol-1-ones upon heating in mineral oil at 240-250 °C. The structures of the synthesized compounds were investigated by IR, mass spectrometry, ^{1}H- and ^{13}C-NMR spectroscopy and MM2 molecular mechanics and AM1 semi-empirical quantum mechanical methods.

Keywords: Condensation, intramolecular cyclization, substituted pyrido[2,3-c]carbazol-1-ones, NMR spectra, molecular modeling.

Introduction

We have chosen to study the synthesis of carbazole ring-containing amino acids and their derivatives for two main reasons: carbazole fragments are present in the structure of some alkaloids [1-3] and carbazole ring-containing compounds usually exhibit semiconductor properties and are used in electrography [4,5]. It was also expected that their combination with amino acid fragments would enable the synthesis of new biologically active substances.

The goal of this study was to clear up the peculiarities of the synthesis procedure and to investigate the structural features of products obtained. It is known that in the reaction of aromatic amines with ethyl-3-oxobutanoate two types of products can form: amides and condensation products – ethyl-β-arylaminocrotonates [6], and that the direction of the reaction depends on the temperature and catalyst.

Results and Discussion

Reaction of 3-amino-9-ethylcarbazole (**1b**) with ethyl-3-oxobutanoate in refluxing toluene provided only the acylation reaction product - N-(9-ethylcarbazol-3-yl)-3-oxobutanamide (**3**, Scheme 1). Addition of a catalytic amount of hydrochloric acid to the reaction solution led to the formation of a mixture of two products – the amide **3** and the condensation product ethyl-3-[(9-ethylcarbazol-3-yl)amino]-2-butenoate (**2b**) in a 1:4 ratio. Pure condensation products **2a-c** were obtained by reaction of amines **1a-c** with ethyl-3-oxobutanoate in refluxing benzene in the presence of a catalytic amount of hydrochloric acid with azeotropic separation of the water formed.

Intramolecular cyclizations of compounds **2a-c** were carried out in mineral oil at 240-250°C. The reaction according to the synthesis methods of described in literature [7] took 15-20 min, giving compounds **4a-c** in quite moderate yields of 46-69%. Prolonged reaction times led to decreased yields of the products.

^{1}H-, ^{13}C-NMR spectra and INEPT ^{13}C-NMR experiments proved the structure of the compounds obtained. The assignment of the resonances in the NMR spectra was based on chemical shift theory [8,9] and signal intensity arguments, multiplicities and comparison with structurally related compounds [10-16]. The carbon atoms are identified following the arbitrary numbering given in Scheme 1. The results of NMR studies are presented in Table 1.

The spectral lines intrinsic to the carbazolyl and butenoate fragment have been observed in the ^{13}C-NMR spectra of compounds **2a-c**. Changes of solvent and the 9N-substituents in the carbazolyl moiety resulted in weak changes of the chemical shifts of carbon resonances, therefore the structural features of the side chain were studied in more detail. The chemical shifts of the double bond of the butenoate moiety were successfully calculated using the additivity rules of substituent influence on the double bond. It was noticed that the double bond in compounds **2a-c** is quite polarized due to the different electronic effects of the substituents present [8,9]. The C-11 atom of the double bond adjacent to the NHR group is strongly deshielded and resonates approximately at 160 ppm. The signal of the C-12 atom of the double bond adjacent to the COOCH$_2$CH$_3$ group is observed much further upfield – approximately at 84 ppm.

Compound **3** is distinguished by the characteristic fragments of the side chain moiety, which were specifically reflected in its ^{13}C-NMR spectra. The resonances of the aliphatic carbons of the COCH$_3$ and COCH$_2$CO fragments and the resonances of both CO group carbons in the downfield region reliably confirmed the formation of compound **3**.

Scheme 1

a: R=CH$_3$, **b**: R=C$_2$H$_5$, **c**: R=C$_3$H$_7$

Compounds **4a-c** were obtained by cyclization of the butenoate fragment. Particular attention was paid to the structure determination of the cyclization products. The ^{13}C-NMR spectra of compounds **4a-c** were assigned on the basis of the model spectrum of 2-methyl-4-quinolone [17]. The respective changes to the spectral information of this compound using the chemical shift additivity rules enabled us to evaluate the chemical shifts of the carbons of the formed ring and its influence on carbazolyl fragment moiety, which is adjacent to the newly formed cycle. The C-11 atom of the double bond adjacent to the NH group in the cyclic compounds **4a-c** is more shielded (146.5 ppm), and the C-12 atom of double bond is more deshielded (108.5 ppm), respectively, in comparison with the chemical shifts of the corresponding carbons in acyclic compounds **2a-c**.

In order to gain further insight about the cyclization of butenoate fragment the ^{1}H-NMR spectra of the carbazole fragments were investigated in detail. The hydrogen resonances of this moiety in compounds **2a-c** were completely and unambigously assigned by the concerted examination of their chemical shifts and spin-spin couplings. It is important to note that all hydrogen resonances of the fragment mentioned above were reliably distinguished by their characteristic splittting patterns. Spin-spin couplings over three (^{3}J$_{HCCH}$) bonds were derived for all hydrogens of the carbazole fragment with

the exception of the H-4 atom. The narrow multiplicty induced by the detectable long-range – over four bond coupling ($^4J_{HCCCH}$) was observed for all hydrogens of the carbazole fragment with the exception of the H-1 atom. The formation of compounds **4a-c** from **2a-c** led to changes in the chemical shifts and multiplicities of some of the hydrogen atoms. For instance, the disappearance of the narrow splitting doublet resonating at 8 ppm and attributed to H-4 was detected, notwithstanding the fact that the doublet with three bond splitting attributed to H-2 was found to resonate in almost the same place. The chemical shifts of the resonances of H-6, H-7, H-8, which were remote from the changes in the molecule were less perturbed. The largest alternation in the chemical shift occurred for the H-5 atom.

A singlet corresponding to the CH group hydrogen of the side chain double bond is seen at 4.7 ppm for compounds **2a-c**, though for compounds **4a-c** it appears at 6.1 ppm after ring formation. The singlet of the CH_3 group hydrogens of the acyclic chain is observed at 1.9 ppm, but the one in the ring is at 2.4 ppm. Finally, compounds **4a-b** have no ester group, whose characteristic triplet and quadruplet were observed at 1.1 ppm and 4.1 ppm, respectively, in the spectra of compounds **2a-c**. The proton of the NH group in cycle is also more deshielded. It resonates at 11.6 ppm, whereas the signal of the proton of the NH group of the acyclic chain in compounds **2a-c** is at 10.4 ppm.

The data obtained from computer molecular modeling are in reasonable agreement with the NMR findings. The models of molecules of studied compounds were optimized to the total steric energy (Figure 1) by MM2 molecular mechanics and AM1 semi-empirical quantum mechanical methods. The computer molecular models demonstrated the turn of side chain towards the carbazole ring for compounds **2a-c**, but the preference of the side chain for the straight structure in compound **3**.

Figure 1. View of optimized to the total steric energy molecular models of compounds **2b**, **3** and **4b**

2b (E = 14.42 kcal/mol)	**3** (E = 3.69 kcal/mol)	**4b** (E = 14.81 kcal/mol)
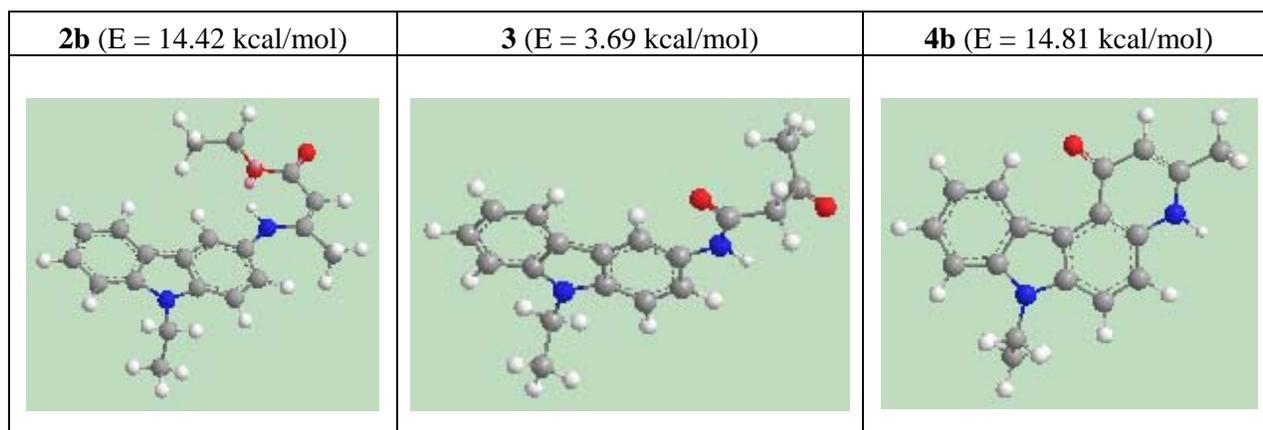
Extended Hückel partial charges derived from the optimized models of compounds **2a-c** revealed more negative C-4 (-O.144 a. v) and H-4 (0.023 a.v.) atoms in comparison ones with C-2 (-0.122 a.v.) and H-4 (0.031 a. v.) of compounds **4a-c**. This fact was one more argument for the cyclization onto the C-4 atom, with no participation of the carbazole ring C-2 atom. The close contact perceived between the CO group oxygen of the new heterocycle and the H-5 atom of the carbazole ring related to the deshielding of H-5 atom in compounds **4a-c.**

Conclusions

It was determined that, depending on the reaction conditions, 9-alkyl-3-aminocarbazoles upon reaction with ethyl-3-oxobutanoate formed acylation or condensation products, whose intramolecular cyclization at 240-250 °C took place onto the 4-position of the carbazole ring providing substituted pyrido[2,3-c]carbazol-1-ones. Taking into consideration that carbazoles are structurally similar to benzenes, the synthesized compounds were successfully studied by NMR spectra using the substituent effect rules as for benzenes. The results obtained during the NMR investigation fully substantiated the structural distinctions of studied compounds. Computer molecular modeling data supported the findings deduced from the ^{13}C-NMR and the ^{1}H-NMR spectra.

Experimental

General

^{1}H- and ^{13}C-NMR spectra were recorded on a *Bruker AC 250-P* (250 MHz), *Bruker DRX 500* (500 MHz) and *Varian Unity Inova* 300 MHz spectrometers operating in Fourier transform mode with TMS as an internal standard. The IR spectra were measured on a *Perkin-Elmer Spectrum GX FT-IR system*. Silica gel plates (*Silufol UV-254*) were used for analytical tlc. Mass spectral data were obtained on *Waters (Micromass) ZQ 2000 Spectrometer*. Physical properties, analytical data and yields of the prepared compounds are given in Table 2. The molecular modeling of the studied compounds was carried out using Chem 3D Ultra 9.0 [18].

Ethyl-3-[(9-methyl-9H-carbazol-3-yl)amino]but-2-enoate (**2a**). A mixture of 3-amino-9-methyl-carbazole (**1a**, 7.85 g, 0.04 mol), benzene (100 mL), ethyl-3-oxobutanoate (7.6 mL, 0.06 mol) and hydrochloric acid (0.5 mL) was refluxed for 5 h with azeotropic separation of the water formed. The benzene was evaporated under reduced pressure and the residue was poured into hexane (100 mL) and was kept cold for crystallization. The precipitate was filtered, washed with hexane and recrystallized from hexane.

Ethyl-3-[(9-ethyl-9H-carbazol-3-yl)amino]but-2-enoate (**2b**) was obtained from 3-amino-9-ethyl-carbazole **1b** (15 g, 0.07 mol) in the same way as described for **2a**. MS (m/z): 323.2 [M$^+$+1].

Ethyl-3-[(9-propyl-9H-carbazol-3-yl)amino]but-2-enoate (**2c**) was obtained from 3-amino-9-propylcarbazole **1c** (15 g, 0.07 mol) in the same way as described for **2a**.

N-(9-ethyl-9H-carbazol-3-yl)-3-oxobutanamide (**3**). The reaction mixture of 3-amino-9-ethylcarbazole (**1b**, 2.1 g, 0.01 mol), ethyl-3-oxobutanoate (3.79 mL, 0.03 mol) and toluene (25 mL) was refluxed for 2 h and was kept in cold for crystallization. The precipitate formed was filtered off, washed with hexane and recrystallized from toluene.

3,7-Dimethyl-4,7-dihydro-1H-pyrido[2,3-c]carbazol-1-one (**4a**). A mixture of compound **2a** (2 g, 0.006 mol) and mineral oil (15 mL) was heated at 240-250 °C for 20 min, then cooled down to 20 °C and diluted with hexane. The precipitate formed was filtered off, washed with hexane and recrystallized from dioxane.

7-Ethyl-3-methyl-4,7-dihydro-1H-pyrido[2,3-c]carbazol-1-one (**4b**) was obtained from compound **2b** (1 g, 0.003 mol) in the same way as described for **4a**. The product formed was purified by column chromatography (*Silikagel L 40/100*, eluent: acetone). MS (m/z): 277.2 [M^{+}+1].

3-Methyl-7-propyl-4,7-dihydro-1H-pyrido[2,3-c]carbazol-1-one (**4c**) was obtained from compound **2c** (4 g, 0.01 mol) in the same way as described for **4a**. The product formed was purified by column chromatography (*Silikagel L 40/100*, eluent: acetone).

Table 1. Spectroscopic data of the prepared compounds (NMR: δ, ppm; J, Hz; IR: cm^{-1})

Compound	^{1}H-NMR (solvent)	^{13}C-NMR (solvent)	IR (KBr tabl.)
2a	$(CD_3)_2CO$: 1.24 (t, 3H, J^3 = 7.1, $COOCH_2\underline{CH_3}$), 1.97 (s, 3H, $C\underline{CH_3}$), 3.92 (s, 3H, $N\underline{CH_3}$), 4.11 (q, 2H, J = 7.1, $COO\underline{CH_2}CH_3$), 4.67 (s, 1H, CH=), 7.21 (td, 1H, J^3 = 6.8, J^3 = 7.9, J^4 = 1.3, Ar-6C-H), 7.30 (dd, 1H, J^3 = 8.6, J^4 = 2.0, Ar-2C-H), 7.48 (td, 1H, J^3 = 6.8, J^3 = 8.2, J^4 = 1.3, Ar-7C-H), 7.53 (d, 1H, J^3 = 8.6, Ar-1C-H), 7.54 (dd, IH, J^3 = 8.2, J^4 = 1.3, Ar-8C-H), 7.98 (d, 1H, J^4 = 2.0, Ar-4C-H), 8.17 (dd 1H, J^3 = 7.9, J^4 = 1.3, Ar-5C H), 10.48 (s, 1H, NH).	$(CD_3)_2CO$: 14.96 ($COOCH_2\underline{CH_3}$), 20.18 ($C\underline{CH_3}$), 29.48 ($N\underline{CH_3}$), 58.80 ($COO\underline{CH_2}CH_3$), 85.09 (CH=), 109.80 (Ar-C-8), 109.89 (Ar-C-1), 118.01 (Ar-C-4), 119.71 (Ar-C-6), 121.22 (Ar-C-5), 123.17 (Ar-C-5a), 123.78 (Ar-C-4a), 124.77 (Ar-C-2), 126.92 (Ar-C-7), 131.73 (Ar-C-3), 139.91 (Ar-C-1a), 142.52 (Ar-C-8a), 161.26 (C=), 170.94 (CO).	3250(NH), 1656(CO).
2b	$(d_6$-DMSO): 1.22 (t, 3H, J = 7.1, $COOCH_2\underline{CH_3}$), 1.32 (t, 3H, J = 7.1, $NCH_2\underline{CH_3}$), 1.96 (s, 3H, $C\underline{CH_3}$), 4.08 (q, 2H, J = 7.1, $COO\underline{CH_2}CH_3$), 4.43 (q, 2H, J = 7.1, $N\underline{CH_2}CH_3$), 4.68 (s, 1H, CH=), 7.19 (td, 1H, J^3 = 7.4, J^3 = 7.6, J^4= 1.3, Ar-6C-H), 7.29 (dd, 1H, J^3 = 8.6, J^4 = 2.0, Ar-2C-H), 7.46 (td, 1H, J^3 = 6.6, J^3 = 7.4, J^4 = 1.3, Ar-7C-H), 7.58 (d, 1H, J^3 = 8.6, Ar-1C-H), 7.59 (dd, 1H, J^3 = 6.6, J^3 = 1.3, Ar-8C-H), 8.02 (d, 1H, J^4 = 2.0, Ar-4C-H), 8.18 (dd, 1H, J^3 = 7.6, J^4 =1.3, Ar-5C-H), 10.38 (s, 1H, NH).	$(d_6$-DMSO): 13.63 ($NCH_2\underline{CH_3}$), 14.49 ($COOCH_2\underline{CH_3}$), 19.76 ($C\underline{CH_3}$), 36.99 ($N\underline{CH_2}CH_3$), 57.90 ($COO\underline{CH_2}CH_3$), 83.93 (C=), 109.13 (Ar-C-8), 109.25 (Ar-C-1), 116.99 (Ar-C-4), 118.65 (Ar-C-6), 120.64 (Ar-C-5), 121.86 (Ar-C-5a), 122.42 (Ar-C-4a), 123.57 (Ar-C-2), 125.98 (Ar-C-7), 130.21 (Ar-C-3), 137.37 (Ar-C-1a), 140.03 (Ar-C-8a), 160.29 (C=), 169.48 (CO).	3267(NH), 1652(CO).
2c	$(CDCl_3)$: 0.98 (t, 3H, J=7.5, $COOCH_2\underline{CH_3}$), 1.31 (t, 3H, J = 6.9, $NCH_2CH_2\underline{CH_3}$), 1.89 – 1.92 (m, 2H, $NCH_2\underline{CH_2}CH_3$), 1.94 (s, 3H, $C\underline{CH_3}$), 4.18 (q, 2H, J = 7.5, $COO\underline{CH_2}CH_3$), 4.26 (q, 2H, J = 6.9, $N\underline{CH_2}CH_2CH_3$), 4.69 (s, 1H, CH=), 7.21 (dd, 1H, J^3 = 8.2, J^4 = 2.0, Ar-2C-H), 7.22 (td, 1H, J^3 = 7.6, J^3 = 8.2, J^4 = 1.3, Ar-6C-H), 7.34 (dd, 1H, J^3 = 6.9, J^4 = 1.3, Ar-8C-H), 7.40 (d, 1H, J^3 = 8.2, Ar-1C-H), 7.47 (td, 1H, J^3 = 8.2, J^3 = 6.9, J^4 = 1.3, Ar-7C-H), 7.83 (d, 1H, J^4 = 2.0, Ar-4C-H), 8.03 (dd, 1H, J^3 = 7.6, J^4 = 1.3, Ar-5C-H), 10.38 (s, 1H, NH).	$(CDCl_3)$: 11.82 ($NCH_2CH_2\underline{CH_3}$), 14.68 ($COOCH_2\underline{CH_3}$), 20.29 ($C\underline{CH_3}$), 22.34 ($NCH_2\underline{CH_2}CH_3$), 44.78 $N\underline{CH_2}CH_2CH_3$), 58.60 ($COO\underline{CH_2}CH_3$), 84.22 (C=), 108.79 (Ar-C-8), 108.93 (Ar-C-1), 117.79 (Ar-C-4), 118.93 (Ar-C-6), 120.39 (Ar-C-5), 122.37 (Ar-C-5a), 122.99 (Ar-C-4a), 124.26 (Ar-C-2), 126.04 (Ar-C-7), 130.70 (Ar-C-3), 138.58 (Ar-C-1a), 141.05 (Ar-C-8a), 160.76 (C=), 170.61 (CO).	3257(NH), 1651(CO).

Table 1. Cont.

3	(CDCl$_3$): 1.41 (t, 3H, J = 7.2, NCH$_2$<u>CH$_3$</u>), 2.36 (s, 3H, COCH$_3$), 3.65 (s, 2H, COCH$_2$), 4.32 (q, 2H, J = 7.2, N<u>CH$_2$</u>CH$_3$), 7.23 (td, 1H, J^3 = 7.0, J^3 = 7.8, J^4 = 1.1, Ar-6C-H), 7.31 (d, 1H, J^3 = 8.7, Ar-1C-H), 7.40 (dd, 1H, J^3 = 8.2, J^4 = 1.1, Ar-8C-H), 7.49 (td, 1H, J^3 = 8.2, J^3 = 7.0, J^4 = 1.1, Ar-7C-H), 7.55 (dd, 1H, J^3 = 8.7, J^4 = 2.0, Ar-2C-H), 8.08 (dd, 1H, J^3 = 7.8, J^4 = 1.1, Ar-5C-H), 8.36 (d, 1H, J^4 = 2.0, Ar-4C-H), 9.23 (s, 1H, NH).	(CDCl$_3$): 14.06 (NCH$_2$<u>CH$_3$</u>), 31.45 (CO<u>CH$_3$</u>), 37.81 (N<u>CH$_2$</u>CH$_3$), 50.30 (CO<u>CH$_2$</u>CO), 108.71 (Ar-C-8), 108.79 (Ar-C-1), 113.26 (Ar-C-4), 119.01 or 119.86 (Ar-C-6), 119.01 or 119.86 (Ar-C-2), 120.96 (Ar-C-5), 122.99 (Ar-C-5a), 123.21 (Ar-C-4a), 126.12 (Ar-C-7), 129.60 (Ar-C-3), 137.55 (Ar-C-1a), 140.66 (Ar-C-8a), 163.93 (NHCO), 205.54 (<u>CO</u>CH$_3$).	3276(NH), 1721(CO), 1640(CO).
4a	(d$_6$-DMSO): 2.41 (s, 3H, CCH$_3$), 3.97 (s, 3H, NCH$_3$), 6.09 (CH=), 7.18 (td, 1H, J^3 = 7.1, J^3 = 8.2, Ar-6C-H), 7.46 (td, 1H, J^3 = 8.2, J^3 = 7.1, Ar-7C-H), 7.60 (d, 1H, J^3 = 8.2, Ar-8C-H), 7.69 (d, 1H, J^3 = 8.9, Ar-1C-H), 8.02 (d, 1H, J^3 = 8.9, Ar-2C-H), 9.94 (d, 1H, J^3 = 8.2, Ar-5C-H), 11.69 (s, 1H, NH).	(d$_6$-DMSO): 18.87 (CCH$_3$), 29.01 (NCH$_3$), 108.60 (CH=), 109.93 (Ar-C-1), 109.93 (Ar-C-8), 114.72 (Ar-C-2), 116.57 (Ar-C-4), 117.91 (Ar-C-6), 121.04 (Ar-C-4a), 121.04 (Ar-C-5a), 122.75 (Ar-C-5), 125.28 (Ar-C-7), 128.90 (Ar-C-3), 136.33 (Ar-C-1a), 140.70 (Ar-C-8a), 146.68 (C=), 177.97 (CO).	3267(NH), 1623(CO).
4b	(d$_6$-DMSO): 1.28 (t, 3H, J = 7.2, NCH$_2$<u>CH$_3$</u>), 2.45 (s, 3H, CCH$_3$), 4.52 (q, 2H, J = 7.2 N<u>CH$_2$</u>CH$_3$), 6.05 (s, 1H, CH=), 7.14 (t, 1H, J^3 = 7.5, J^3 = 8.2, Ar-6C-H), 7.42 (t, 1H, J^3 = 7.5, J^3 = 8.2, Ar-7C-H), 7.59 (d, 1H, J^3 = 8.2, Ar-8C-H), 7.65 (d, 1H, J^3 = 8,8, Ar-1C-H), 8.02 (d, 1H, J^3 = 8.8, Ar-2C-H), 9.90 (d, 1H, J^3 = 8.2, Ar-5C-H), 11.65 (s, 1H, NH).	(d$_6$-DMSO): 13.91 (NCH$_2$<u>CH$_3$</u>), 18.89 (CH$_3$), 36.86 (N<u>CH$_2$</u>CH$_3$), 108.52 (CH=), 109.95 (Ar-C-1), 109.95 (Ar-C-8), 114.67 (Ar-C-2), 116.62 (Ar-C-4), 117.93 (Ar-C-6), 121.19 (Ar-C-4a), 121.19 (Ar-C-5a), 122.93 (Ar-C-5), 125.29 (Ar-C-7), 129.10 (Ar-C-3), 136.34 (Ar-C-1a), 139.58 (Ar-C-8a), 146.58 (C=), 177.97 (CO).	3281(NH), 1619(CO).
4c	(d$_6$-DMSO): 0.85 (t, J = 7.2, 3H, NCH$_2$CH$_2$<u>CH$_3$</u>), 1.75 − 1.82 (m, 2H, NCH$_2$<u>CH$_2$</u>CH$_3$), 2.37 (s, 3H, CCH$_3$), 4.55 (t, J = 7.2, 2H, N<u>CH$_2$</u>CH$_2$CH$_3$), 6.04 (s, 1H, CH=), 7.13 (t, 1H, J^3 = 7.6, J^3 = 8.2, Ar-6C-H), 7.41 (t, 1H, J^3 = 7.6, J^3 = 8.2, Ar-7C-H), 7.60 (d, 1H, J^3 = 8.2, Ar-8C-H), 7.64 (d, 1H, J^3 = 8.8, Ar-1C-H), 8.03 (d, 1H, J^3 = 8.8, Ar-2C-H), 9.90 (d, 1H, J^3 = 8.2, Ar-5C-H), 11.63 (s, 1H, NH).	(d$_6$-DMSO): 11.31 (NCH$_2$CH$_2$<u>CH$_3$</u>), 18.84 (CH$_3$), 22.09 (NCH$_2$<u>CH$_2$</u>CH$_3$), 43.53 (N<u>CH$_2$</u>CH$_2$CH$_3$), 108.98 (CH=), 109.93 or 109.99 (Ar-C-1), 109.93 or 109.99 (Ar-C-8), 114.94 (Ar-C-2), 116.56 (Ar-C-4), 117.88 (Ar-C-6), 121.11 (Ar-C-5a), 121.11 (Ar-C-4a), 122.78 (Ar-C-5), 125.23 (Ar-C-7), 129.03 (Ar-C-3), 136.32 (Ar-C-1a), 140.13 (Ar-C-8a), 146.51 (C=), 177.98 (CO).	3283(NH), 1643(CO).

Table 2. The physical, analytical and yield data of the studied compounds

Compound	m. p. (°C)	Molecular Formula	Elemental analysis data (Calculated / Found) %			Yield (%)
			C	**H**	**N**	
2a	130 – 131	C$_{19}$H$_{20}$N$_2$O$_2$	74.00 / 74.08	6.54 / 6.48	9.08 / 8.96	80.0
2b	134 – 135	C$_{20}$H$_{22}$N$_2$O$_2$	74.51 / 74.46	6.88 / 7.09	8.69 / 8.43	83.0
2c	77 – 78	C$_{21}$H$_{24}$N$_2$O$_2$	74.97 / 74.71	7.19 / 7.24	8.33 / 8.43	76.0
3	157 – 158	C$_{18}$H$_{18}$N$_2$O$_2$	73.45 / 73.34	6.16 / 6.37	9.52 / 9.45	73.0

Table 2. Cont.

4a	>330	C$_{17}$H$_{14}$N$_2$O	77.84 / 77.63	5.38 / 5.10	10.68 / 10.45	63.0
4b	254 – 256	C$_{18}$H$_{16}$N$_2$O	78.24 / 78.20	5.84 / 5.82	10.14 / 10.09	46.0
4c	274 – 275	C$_{19}$H$_{18}$N$_2$O	79.90 / 79.86	4.59 / 4.62	9.81 / 9.92	69.0

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18. *Chem 3D Ultra 9.0*, Cambridge Scientific Software, Serial number: 031 406391 4800.

Sample Availability: Available from the authors.