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1-Azaacepleiadylene—A Novel *peri*-Fused Heterocyclic System

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Abstract—Treatment of 2-aryl-1*H*,4*H*-2,3,7,8-tetrahydroacenaphtho[5,6-*bc*]azepin-4-one with hydrazine hydrate on heating in ethylene glycol in the presence of alkali resulted in the reduction of the oxo group to CH_2. Dehydrogenation of the reduction product with 2,3,5,6-tetrachloro-1,4-benzoquinone gave 2-aryl-1-azaacepleiadylene which is the first representative of a novel *peri*-fused heterocyclic system.

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In the previous communication [1] we reported on the synthesis of 2-aryl-1*H*,4*H*-2,3,7,8-tetrahydroacenaphtho[5,6-*bc*]azepin-4-ones (**I**) which may be convenient starting compounds in the transformation of carbonyl group into methylene according to Wolff–Kishner (reaction sequence **I** → **II** → **III** in Scheme 1) [2].

For this purpose, ketones **Ia** and **Ib** were heated for a short time with hydrazine hydrate in alcohol. We thus obtained the corresponding hydrazones **IIa** and **IIb** in good yield. However, our attempt to reduce hydrazones **IIa** and **IIb** with potassium *tert*-butoxide in dimethyl sulfoxide at room temperature according to the procedure described in [3] was unsuccessful: only the initial compounds were isolated.

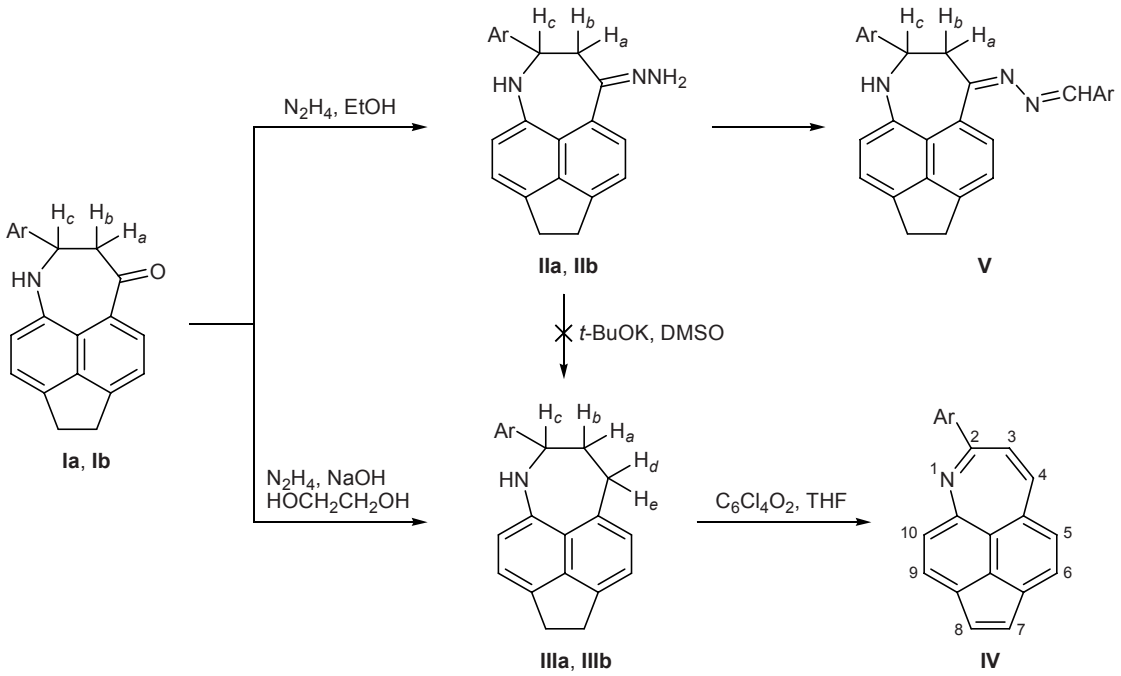
As applied to ketones **Ia** and **Ib**, a successful version of the Kishner reaction turned out to be Huang–Minlon modification [4], i.e., heating of ketone **I** with hydrazine hydrate and sodium hydroxide in ethylene glycol. This procedure ensured preparation of naphtho[5,6-*bc*]azepines **IIIa** and **IIIb** in high yield.

When 2-(4-methoxyphenyl)-1*H*,4*H*-2,3,7,8-tetrahydroacenaphtho[5,6-*bc*]azepine (**IIIa**) was heated with 2,3,5,6-tetrachloro-1,4-benzoquinone in tetrahydrofuran, we isolated 2-(4-methoxyphenyl)-1-azaacepleiadylene **IV** with a small yield, whereas 2-(3,4-dimethoxyphenyl) derivative **IIIb** under analogous conditions gave rise to a mixture of products which we failed to separate and identify. Our attempts to obtain 2-aryl-1-azaacepleiadylenes **IV** by heating their potential precursors **IIIa** and **IIIb** in *p*-xylene or mesitylene over Pd/C were also unsuccessful: only contaminated initial compounds were recovered from the reaction mixtures.

The structure of the newly synthesized compounds was confirmed by spectral methods, and the structure of **IV** was proved by X-ray analysis.

Protons on C^2 and C^3 in the molecules of hydrazones **IIa** and **IIb** form a strongly coupled spin system. Owing to nonequivalence of the geminal C^3H_2 protons in the seven-membered ring, the H_a and H_c (2-H) signals appear as doublets of doublets at δ 2.70 and 4.70 ppm (**IIa**) and 2.73 and 4.70 ppm (**IIb**), respectively. The other methylene proton signal (H_b) is obscured by the wide multiplet in the region δ 3.2–3.4 ppm, which belongs to C^7H_2 and C^8H_2. The ^{1}H NMR spectra of reduction products **IIIa** and **IIIb** in the region δ 2.2–4.5 ppm are even more complex. Although some signals were identified on the basis of chemical shifts of the corresponding protons, it was difficult to determine spin–spin coupling constants for methylene protons, for their signals were complex multiplets. We were able to identify only the H_c signal as a doublet of doublets. Broadened signal from the NH proton was overlapped by one (**IIIa**) or two (**IIIb**) singlets from the methoxy protons. In the aromatic region, the 5-H and 6-H protons resonated as one two-proton singlet.

Scheme 1.



I–**III**, Ar = 4-$MeOC_6H_4$ (**a**), 3,4-$(MeO)_2C_6H_3$ (**b**); **IV**, **V**, Ar = 4-$MeOC_6H_4$.

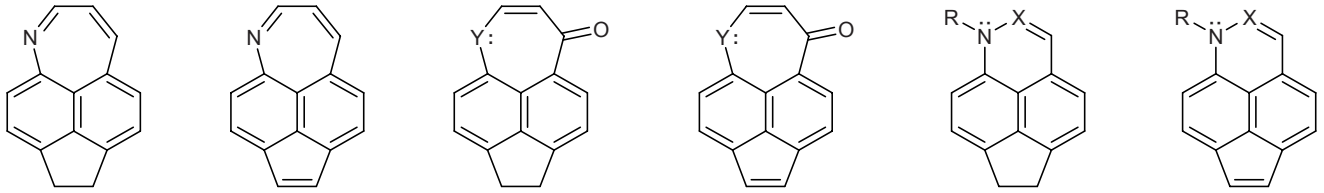
Apart from sufficiently informative spectral data, the presence of a reactive amino group in the molecule of hydrazone **IIa** was confirmed by its reaction with 4-methoxybenzaldehyde which resulted in the formation of azine **V** (Scheme 1).

Signals from protons in the naphthalene fragment of compounds **IIa**, **IIb**, **IIIa**, and **IIIb** were located in their ^{1}H NMR spectra in the regions δ 6.6–7.6 (**II**) and 6.6–7.1 ppm (**III**), whereas naphthalene protons in 1-azapleiadylene **IV** resonated at δ 7.33–8.25 ppm. The observed downfield shift may be interpreted in terms of increased diamagnetic constituent of the ring current in **IV**, i.e., in terms of higher aromaticity of the latter compound. In fact, 1-azapleiadylene can be regarded as an aromatic ring system: its 16π-electron structure can be represented as a superposition of 14π-electron peripheral contour and 10π-electron naphthalene fragment. Taking into account that each skeletal carbon atom in molecule **IV** is included into 14π- or 10π-electron cyclic system, each conforming to the $4n + 2$ constraint, such molecular entities may be considered to constitute a specific "supplement to Hückel's rule."* Formalistically, structure **IV** contains 16π ($4n$) electrons, so that it should be classed with antiaromatic systems according to Hückel.

However, it seems inappropriate to presume enhanced aromaticity of the 1-azapleiadylene system (**IV**) compared to compounds **I**–**III** on the basis of "boundary" chemical shifts of protons in the naphthalene ring (most downfield 5-H and most upfield 10-H), for the position of their signals strongly depends on the electron-withdrawing or electron-donating effect of the neighboring functional groups. In other words, the chemical shifts of 5-H and 10-H are determined not so much by the ring current as by the electron density on C^5 and C^{10}, which depends on the above effects and disturbs the ring current. It would be more appropriate to compare molecular systems differing by the presence or absence of the double bond at the *peri* positions opposite to the heteroring, e.g., structures **A** and **B**, **C** and **D**, or **E** and **F**. In these cases, structural variations induced by the double bond are minimal, while differences in the electronic structures are quite significant. Isolated heteroring in structures **A**, **C**, and **E** becomes involved in aromatic conjugation in going to 14π-electron peripheral contour in structures **B**, **D**, and **F**.** Taking into account the above stated, the downfield shift of naphthalene proton signals, as well as of signals from protons in the heteroring and substit-

* The term "supplement to Hückel's rule" is introduced by us for the first time.

** Divinyl (isolated) character of the seven-membered ring in the molecule of acepleiadiene and aromaticity of acepleiadylene were discussed in [5, 6].

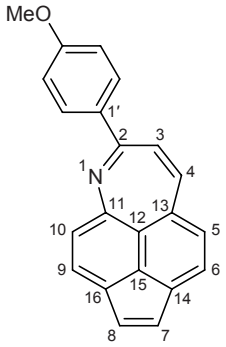


A **B** **C** **D** **E** **F**

Y = O, RN; X = CH, N.

uents therein, observed by us previously in going from structure **C** to **D** [7] or from **E** to **F** [8] may be interpreted as unambiguous evidence in favor of increased aromaticity of the whole *peri*-fused heteroaromatic system. Unfortunately, analogous comparison of structures **A** and **B** cannot be performed in the framework of the present study because of the lack of compounds having structure **A**.

The structure of 2-(4-methoxyphenyl)-1-azaacepleiadylene (**IV**) was determined by X-ray analysis. This compound forms non-centrosymmetric crystals belonging to space group *P*1 with two independent molecules in a unit cell. The main geometric parameters of both molecules were similar within the experimental error, so that the parameters of only one of them will be considered here. Molecule **IV** is essentially planar, except for the 4-methoxyphenyl substituent which is turned through a dihedral angle of 23.6° about the C^2–$C^{1'}$ bond. However, the $N^1C^2C^3C^4$ fragment slightly deviates from the acenaphthene plane, the minimal deviation being 0.05 Å for C^2, while the other atoms in that fragment deviate from the acenaphthene plane to a considerably stronger extent (0.11–0.13 Å). The observed bend of the seven-membered ring cannot be induced only by the effect of crystal packing. In fact, deviations of the corresponding atoms from the acenaphthene plane in the second independent molecule, which appears in a different crystal environment, range from 0.06 to 0.11 Å. Therefore, we presume that the



Atom numbering in the molecule of 2-(4-methoxyphenyl)-acenaphtho[5,6-*bc*]azepine (**IV**).

N^1=C^2–C^3=C^4 fragment is slightly forced out from conjugation with the other part of the molecule. In addition, steric repulsion between hydrogen atoms on C^4 and C^5, which should hamper flattening of the molecule, cannot be ruled out.

Distribution of bond lengths in the $C^3C^4C^{13}C^{12}$ fragment (Table 1) is almost the same as in the molecular complex of acepleiadylene with trinitrobenzene, studied previously [9]. An exception is the C^2–C^3 bond which is longer [1.451(4) Å] than in the above complex (1.425 Å). The N^1–C^2 bond [1.302(4) Å] in molecule **IV** is considerably shorter than the N^1–C^{11} bond [1.395(3) Å]; on the other hand, the latter is appreciably shorter than the corresponding bond distance in, e.g., *N*-benzylidenenaphthalen-1-amine (1.422 Å) [10]. The bond angles at the bridgehead carbon atoms in the naphthalene ring (Table 2) are distorted to a considerable extent: the $C^{11}C^{12}C^{13}$ angle (seven-membered ring) is 128.3(3)°, while the $C^{14}C^{15}C^{16}$ angle in the five-membered ring is much smaller, 109.0(2)°. Thus, mutual approach of carbon atoms in the *peri* positions at the side of the five-membered ring is accompanied by extension of the distance between the opposite *peri*-carbon atoms belonging to the seven-membered heteroring. In other words, we observed consistent contributions of the five-membered carbocycle and seven-membered heteroring to the in-plane distortion of the naphthalene ring.

Unlike electron-rich compounds like **F** (X = N) described previously, we now report on the synthesis of the first representative of 16π-electron π-deficient *peri*-fused heteroaromatic compounds having a 1-azaacepleiadylene skeleton and pyridine type nitrogen atom. 1-Azaacepleiadylene **IV** and 1-azapleiadylen-4-one derivatives **D** (Y = NR) synthesized previously should display some similarity in chemical properties to pyridine (quinoline) and pyridin-4-one (quinolin-4-one), respectively. 1- and 2-Azapyrenes [11, 12] may also be regarded as compounds that are structurally related and isoelectronic to 16π-electron π-deficient *peri*-fused heteroaromatic system. 1-Azaacepleiadylene and 1-azapyrene differ from each other as

aromatic hydrocarbons acepleiadylene [6] and azulene differ from pyrene and naphthalene, respectively.

Analysis of the spectral parameters and crystallographic data of compound **IV**, as well as comparison with other carbo- and heteroaromatic structures, led us to presume essential delocalization of π-electron density in the molecule of 2-(4-methoxyphenyl)-1-azaacepleiadylene and hence aromatic character of this novel heterocyclic system.

EXPERIMENTAL

The ^{1}H NMR spectra were recorded on Bruker DPX-250 and Varian Unity-300 spectrometers at 250 and 300 MHz, respectively, using hexamethyldisiloxane as internal reference. The IR spectra were obtained on a Specord 75IR instrument from samples dispersed in mineral oil. Chromatographic purification was performed using aluminum oxide as sorbent and chloroform as eluent.

X-Ray diffraction study on 2-(4-methoxyphenyl)-acenaphtho[5,6-*bc*]azepine (IVa). $C_{22}H_{15}NO$. *M* 309.35. Single crystals suitable for X-ray analysis were obtained by isothermal evaporation of a solution of **IVa** in toluene. Triclinic crystals with the following unit cell parameters (120 K): a = 5.1780(4), b = 12.2346(10), c = 12.4865(10) Å; α = 105.2135(15), β = 99.6982(16), γ = 90.5036(17)°; V = 751.23(10) Å^3; d_{calc} = 1.368 g/cm^3; μ = 0.84 cm^{-1}; Z = 2 (Z' = 2); space group $P1$. Intensities of 7364 reflections were measured at 120 K on a Smart 1000 CCD automatic diffractometer (MoK_α irradiation, graphite monochromator, ω-scannibg, $2\theta_{max}$ = 59°); 4055 independent reflection intensities (R_{int} = 0.0189) were used in subsequent calculations. The structure was solved by the direct method and was refined by the full-matrix anisotropic–isotropic approximation with respect to F^2. Hydrogen atoms were localized by difference syntheses of electron density, and their positions were refined according to the riding model. The final divergence factors were wR_2 = 0.1105 for all reflections and R_1 = 0.0480 for 3272 reflections with $I > 2\sigma(I)$; goodness of fit 0.967. Calculations were performed using SHELXTL PLUS 5.0 software package [13].

2-(4-Methoxyphenyl)-1,2,3,4,7,8-hexahydroacenaphtho[5,6-*bc*]azepin-4-one hydrazone (IIa). A mixture of 0.718 g (2.2 mmol) of compound **Ia**, 0.12 ml (2.4 mmol) of hydrazine hydrate, and 5 drops of acetic acid in 10 ml of ethanol was heated for 2 h under reflux. A part of the solvent (5–6 ml) was distilled off, the remaining solution was diluted with water, the precipitate was filtered off, dried, and dissolved in chloroform, and the solution was subjected to chromatography on aluminum oxide, a fraction with R_f 0.3 being collected. Yield 0.668 g (89%), light yellow powder, mp 135–136°C (from ethanol). IR spectrum, ν, cm^{-1}: 3287, 3180, 3154 (NH, NH$_2$); 1630 (C=N). ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 2.70 d.d (1H, H$_a$, J = 2.83, 3.14, 16.33 Hz), 3.24–3.43 m (5H, CH_2CH_2, H$_b$), 3.80 s (3H, CH_3O), 4.10 br.s (1H, NH), 4.70 d.d (1H, H$_c$, J = 3.14, 10.68 Hz), 5.30 br.s (2H, NH$_2$), 6.64 d (1H, 10-H, $J_{10,9}$ = 7.22 Hz), 6.92 d (2H, 3′-H, 5′-H, ^3J = 8.48 Hz), 7.03 d (1H, 9-H, $J_{9,10}$ = 7.22 Hz), 7.23 d (1H, 6-H, $J_{6,5}$ = 7.22 Hz), 7.37 d.d (2H, 2′-H, 6′-H, ^3J = 8.48 Hz), 7.67 d (1H, 5-H, $J_{5,6}$ = 7.22 Hz). Found, %: C 76.73; H 5.98; N 12.43. $C_{22}H_{21}N_3O$. Calculated, %: C 76.94; H 6.16; N 12.24.

2-(3,4-Dimethoxyphenyl)-1,2,3,4,7,8-hexahydroacenaphtho[5,6-*bc*]azepin-4-one hydrazone (IIb) was synthesized in a similar way. Yield 87%, light yellow powder, mp 198–199°C (from ethanol). IR spectrum, ν, cm^{-1}: 3327, 3170, 3150 (NH, NH$_2$); 1630 (C=N). ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 2.73 d.d (1H, H$_a$, J_{ab} = 16.17, J_{ac} = 3.14 Hz), 3.21–3.44 m (5H,

Table 1. Principal bond lengths d in one independent molecule of 2-(4-methoxyphenyl)acenaphtho[5,6-*bc*]azepine (**IV**)

Bond	d, Å	Bond	d, Å
N^1–C^2	1.302(4)	C^7–C^{14}	1.453(5)
N^1–C^{11}	1.395(3)	C^8–C^{16}	1.451(4)
C^2–C^3	1.451(4)	C^9–C^{16}	1.378(4)
C^2–$C^{1''}$	1.492(4)	C^9–C^{10}	1.401(4)
C^3–C^4	1.348(4)	C^{10}–C^{11}	1.406(4)
C^4–C^{13}	1.436(4)	C^{11}–C^{12}	1.448(4)
C^5–C^{13}	1.401(4)	C^{12}–C^{15}	1.385(4)
C^5–C^6	1.409(4)	C^{12}–C^{13}	1.445(4)
C^6–C^{14}	1.372(4)	C^{14}–C^{15}	1.430(4)
C^7–C^8	1.371(5)	C^{15}–C^{16}	1.428(4)

Table 2. Selected bond angles ω in one independent molecule of 2-(4-methoxyphenyl)acenaphtho[5,6-*bc*]azepine (**IV**)

Angle	φ, deg	Angle	φ, deg
$N^1C^{11}C^{12}$	129.2(2)	$C^8C^{16}C^9$	136.2(3)
$N^1C^{11}C^{10}$	112.94	$C^8C^{16}C^{15}$	106.4(3)
$C^{11}C^{12}C^{13}$	128.3(3)	$C^{14}C^{15}C^{16}$	109.0(3)
$C^{12}C^{13}C^4$	123.7(3)	$C^6C^{14}C^7$	135.8(3)
$C^4C^{13}C^5$	117.4(3)	$C^7C^{14}C^{15}$	106.1(3)

CH_2CH_2, H_b), 3.85 s (6H, CH_3O), 4.10 br.s (1H, NH), 4.70 d.d (1H, H_c, J_{cb} = 10.70, J_{ca} = 3.14 Hz), 5.25 br.s (2H, NH_2), 6.66 d (1H, 10-H, $J_{10,9}$ = 7.22 Hz), 6.87 d (1H, 5′-H, ^3J = 7.85 Hz), 6.94–7.05 m (3H, 9-H, 2′-H, 6′-H), 7.23 d (1H, 6-H, $J_{6,5}$ = 7.22 Hz), 7.67 d (1H, 5-H, $J_{5,6}$ = 7.22 Hz). Found, %: C 73.73; H 6.08; N 11.43. $C_{23}H_{23}N_3O_2$. Calculated%: C 73.97; H 6.21; N 11.25.

2-(4-Methoxyphenyl)-1,2,3,4,7,8-hexahydroacenaphtho[5,6-*bc*]azepine (IIIa). Hydrazine hydrate, 0.1 ml (2 mmol), was added to a suspension of 0.2 g (0.6 mmol) of compound **Ia** and 0.11 g (2.75 mmol) of sodium hydroxide in 3 ml of ethylene glycol, and the mixture was heated for 2 h under reflux. The mixture was cooled, diluted with water, acidified with dilute (1:1) hydrochloric acid to pH 1–2, and extracted with three portions of chloroform. The extracts were combined and evaporated to dryness, and the residue was dissolved in chloroform and subjected to chromatography on aluminum oxide, the first fraction with R_f 0.9 being collected. Yield 134 mg (70%), light pink powder, mp 107–108°C (from methanol). IR spectrum, ν, cm^{-1}: 3380 (NH), 1580. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 2.29 m (2H, H_a, H_b), 3.05 m (1H, H_c), 3.30 m (4H, 7-H, 8-H), 3.60 m (1H, H_d), 3.80 (4H, CH_3O, NH), 4.50 d.d (1H, H_e, J = 6.74, 9.44 Hz), 6.56 d (1H, 10-H, $J_{10,9}$ = 7.07 Hz), 6.86 d (2H, 3′-H, 5′-H, ^3J = 8.75 Hz), 6.98 br.d (1H, 9-H, $J_{9,10}$ = 7.07 Hz), 7.07 br.s (2H, 5-H, 6-H), 7.30 d (2H, 2′-H, 6′-H, ^3J = 8.75 Hz). Found, %: C 83.43; H 7.00; N 4.28. $C_{22}H_{21}NO$. Calculated, %: C 83.78; H 6.71; N 4.44.

2-(3,4-Dimethoxyphenyl)-1,2,3,4,7,8-hexahydroacenaphtho[5,6-*bc*]azepine (IIIb) was synthesized in a similar way. Yield 68%, light pink powder, mp 138–139°C (from methanol). IR spectrum, ν, cm^{-1}: 3340 (NH), 1580. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 2.27 m (2H, H_a, H_b), 3.08 m (1H, H_c), 3.30 m (4H, 7-H, 8-H), 3.62 m (1H, H_d), 3.83 br.s (1H, NH), 3.85 s and 3.89 s (3H each, CH_3O), 4.52 d.d (1H, H_e, J = 6.48, 9.53 Hz), 6.61 d (1H, 10-H, $J_{10,9}$ = 7.22 Hz), 6.84 d (1H, 5′-H, ^3J = 8.13 Hz), 6.91–6.98 m (2H, 2′-H, 6′-H), 7.01 br.d (1H, 9-H, $J_{9,10}$ = 7.22 Hz), 7.09 br.s (2H, 5-H, 6-H). Found, %: C 79.51; H 6.49; N 4.30. $C_{23}H_{23}NO_2$. Calculated, %: C 79.97; H 6.71; N 4.05.

2-(4-Methoxyphenyl)acenaphtho[5,6-*bc*]azepine (IV). A solution of 187 mg (0.762 mmol) of 2,3,5,6-tetrachloro-1,4-benzoquinone in 6 ml of tetrahydrofuran was added to a solution of 120 mg (0.38 mmol) of compound **IIIa** in 2 ml of tetrahydrofuran, and the mixture was stirred for 30 min at room temperature. The mixture was evaporated to dryness, the residue was dissolved in chloroform, and the solution was subjected to chromatography on aluminum oxide, the first fraction with R_f 0.8 being collected. Yield 6 mg (6%), red powder, mp 131–132°C (from toluene). IR spectrum, ν, cm^{-1}: 1600, 1590. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 3.88 s (3H, CH_3O), 7.02 d.d (2H, 3′-H, 5′-H, ^3J = 9.11, ^4J = 2.20 Hz), 7.33 d (1H, 3-H, $J_{3,4}$ = 11.93 Hz), 7.76 d (1H, 7-H, $J_{7,8}$ = 5.02 Hz), 7.81 d (1H, 4-H, $J_{4,3}$ = 11.93 Hz), 7.84 d (1H, 8-H, $J_{8,7}$ = 5.02 Hz), 7.86 d (1H, 6-H, $J_{6,5}$ = 7.22 Hz), 8.12 d.d (2H, 2′-H, 6′-H, ^3J = 9.11, ^4J = 2.20 Hz), 8.25 d (1H, 5-H, $J_{5,6}$ = 7.22 Hz), 8.32 d (1H, 9-H, $J_{9,10}$ = 7.54 Hz), 8.39 d (1H, 10-H, $J_{10,9}$ = 7.54 Hz). Found, %: C 85.50; H 4.59; N 4.92. $C_{22}H_{15}NO$. Calculated, %: C 85.41; H 4.89; N 4.53.

2-(4-Methoxyphenyl)-1,2,3,4,7,8-hexahydroacenaphtho[5,6-*bc*]azepin-4-one *N*′-(4-methoxybenzylidene)hydrazone (V). Compound **IIa**, 0.98 g (2.86 mmol), was dissolved on heating in 10 ml of ethanol, 0.35 ml (2.86 mmol) of 4-methoxybenzaldehyde was added, and the mixture was heated for 5–7 min under reflux and evaporated to dryness in air. The residue was dissolved in chloroform and subjected to chromatography on aluminum oxide using chloroform as eluent. The first fraction with R_f 0.8 was collected. Yield 1.05 g (80%), yellow powder, mp 67–68°C. IR spectrum, ν, cm^{-1}: 3340, 1610. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 3.28–3.45 m (5H, CH_2CH_2, H_a), 3.63–3.76 m (4H, 4-OCH_3, H_b), 3.84 s (3H, 4′-OCH_3), 4.10 br.s (1H, NH), 4.78 d.d (1H, 2-H, J = 4.08, 4.40, 10.21 Hz), 6.70 d (1H, 10-H, $J_{10,9}$ = 7.23 Hz), 6.83 d (2H, 3′-H, 5′-H, ^3J = 8.79 Hz), 6.91 d (2H, 3″-H, 5″-H, ^3J = 8.79 Hz), 7.07 br.d (1H, 9-H, $J_{9,10}$ = 7.22 Hz), 7.28 br.d (1H, 6-H, $J_{6,5}$ = 7.22 Hz), 7.35 d (2H, 2′-H, 6′-H, ^3J = 8.79 Hz), 7.69 d (2H, 2″-H, 6″-H, ^3J = 8.79 Hz), 7.89 d (1H, 5-H, $J_{5,6}$ = 7.22 Hz), 8.35 s (1H, N=CH). Found, %: C 78.40; H 5.59; N 8.92. $C_{30}H_{27}N_3O_2$. Calculated, %: C 78.07; H 5.90; N 9.10.

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