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The Derivatives of the 2a,4a-Diazacyclopenta[*c,d*]azulene

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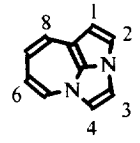
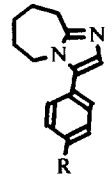
Abstract: The method of preparation of the 3-aryl-6,7,8,9-tetrahydro-5*H*-imidazo[1,2-*a*]azepines **2a-d** from *O*-methylcaprolactim and phenacylamine hydrochlorides has been developed. The bases 2 reacted with phenacylbromides to yield the quaternary salts **4a-e**. The cyclisation of **4a-e** in basic conditions resulted in a new heterocyclic system - the derivatives of 2a,4a-Diazacyclopenta[*c,d*]azulene - **5a-i**.

The heterocyclic system of the 2a,4a-diazacyclopenta[*c,d*]azulene (1) is not described in the literature. To synthesise the 5,6,7,8-tetrahydroderivatives we have chosen the method of additive construction of the pyrrole ring based on the cyclization of the α-methyl-*N*-phenacylcyclimmonium salts.[1-3] This method was originally proposed by Tshitschibabin.[4]

As starting materials we used the 3-aryl-6,7,8,9-tetrahydro-5*H*-imidazo[1,2-*a*]azepines **2a-d**, obtained in one step from *O*-methylcaprolactim and α-aminoacetophenone hydrochlorides similar to that described by us in the synthesis of the 3-aryl-6,7-dihydro-5*H*-pyrrolo[1,2-*a*]imidazoles.[5] Compound **2a** was described earlier in the work.[6] Contrary to the early procedure the simple synthesis of bases 2 is proposed in the present work, without separation of intermediate salts like 3 in the individual state.



1



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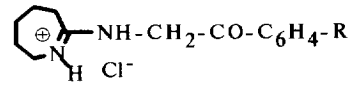


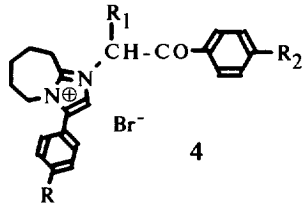
3

2a R=H, b R=OMe, c R=Cl, d R=Br

The compounds 2a-d were alkylated by the phenacylbromides in boiling polar solvents (alcohol or acetone) forming colorless high melting crystalline materials. According to the spectral characteristics we defined their structure as the imidazolium quaternary salts **4a-e**. The IR-spectra of **4a-e** have the carbonyl bond absorption at 1680-1690 cm^{-1} and CH of methylene groups at 2940-3050 cm^{-1}.

Cyclization of the quaternary salts 4a-e is carried out by boiling in water or alcohol in the presence of base (for example, sodium ethoxide, alkali, sodium carbonate). The best yields of cyclic products are observed

using the 5% water solution of sodium hydroxide. Using this method the other phenacylium salts of the 3-aryl-6.7.8.9-tetrahydro-5*H*-imidazo[1,2-*a*]azepines were subjected to cyclization without preliminary purification.



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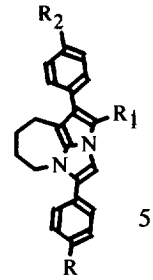

5

4	R	R_1	R_2
a	H	H	Me
b	OMe	H	H
c	OMe	H	Br
d	Cl	H	OMe
e	Cl	Me	Cl

5	R	R_1	R_2
a	H	H	Me
b	H	H	OMe
c	H	H	OEt
d	OMe	H	H
e	OMe	H	Cl
f	OMe	H	Br
g	Cl	H	OMe
h	Br	H	OMe
i	Cl	Me	Cl

Under these conditions the products of the cyclization of the quaternary salts **4a-e** are the derivatives of 1,4-diaryl-5,6,7,8-tetrahydro-2a,4a-diazacyclopenta[*c,d*]azulene **5a-i** - pale yellow, stable, crystalline compounds. The ^{1}H-NMR spectra of **5a-i** showed a multiplet at 1.9 ppm corresponding to the 6- and 7-CH_2, and two triplets at 2.8 ppm and 3.6 ppm for 8- and 5-CH_2 respectively.

The signal of 2-H and 3-H can be identified among other aromatic protons in the region 6.7-7.6 ppm. The comparison of ^{1}H-NMR spectra for **5a-h** with base **5i** permits the identification of the signal of the imidazole proton 3-H at 7.0-7.22 ppm while the signal in the stronger field 6.75-6.91 corresponds to the pyrrole proton 5-H. The absorption of the carbonyl group in the IR spectrum of the bases **5a-i** is absent. The absorption in the range of 3000 cm^{-1} becomes more simple. This fact permits us to conclude that the peak at 3060 cm^{-1} for the quaternary salts is achieved due to the stretching frequencies of the methylene group at 9 position of the system of *N*-phenacylcyclimmonium salts **4**. The UV absorption spectrum of **5** contains two maximums at 260 nm (lg ε 4.0) and 310 nm (lg ε 3.9) while the starting quaternary salts **4** have only the short wave absorption at 260 nm (lg ε 4.1).

All the bases **5a-i** except compound **5i** condense with the *p*-dimethylaminobenzaldehyde in the presence of HCl (Erlich test on pyrrole ring) forming the dye-styryl with $\lambda_{max.}$ 511 nm.

Experimental

^{1}H-NMR were recorded at 100 MHz on a Bruker WR-100 with the tetramethylsilane as an internal

standard. The listed NMR data are given in the following order: chemical shift in ppm (δ), multiplicity (s, singlet; d, doublet; t, triplet; m, multiplet; dd, doublet of doublets; q, quartet; br, broad), number of protons (by integration), and interpretation. Spectra of compounds **2a** and **5a,b,c,h** were measured in $CDCl_3$; **2b,c,d, 4a-e, 5d-g,i** - in DMSO-d_6. The IR spectra were run as KBr pellets using a Pye Unicam SP3-300 spectrometer. The ultraviolet absorption spectra were determined with the Specord UV-VIS spectrometer in ethanol. The melting points were determined using the Boetius hot stage apparatus and are uncorrected. The elemental microanalysis was performed by the chemical department microanalysis laboratory.

The starting α-bromoacetophenones[7] and α-aminoacetophenones[8] were prepared as it was described earlier.

3-Phenyl-6,7,8,9-tetrahydro-5*H*-imidazo[1,2-*a*]azepine (2a).

The *O*-methylcaprolactime (7,0 g; 55 mmol) was mixed with the α-aminoacetophenone hydrochloride (3.55 g; 50 mmol). The viscous mass was left to stand at room temperature for 48 hrs. 100 ml of ether were added and the reaction mixture was stirred for 1 h. The crystalline residue was separated, washed with ether and then dissolved in 100 ml of 0.1 N HCl. The solution was refluxed for 2 hrs, cooled and adjusted to pH 10 with the aqueous ammonia. The precipitate was collected, then washed with water and dried in vacuo. After the recrystallization from hexane, 7.1 g (67%) of 2a were obtained, m.p. 102 °C (lit.[6] m.p.101 °C). ^{1}H-NMR: s 6.87 (1H,2-**H**); t 3.92 (2H,5-CH_2), m 1,82 (6H,6.7.8-$|CH_2|_3$); t 2.96 (2H,9-CH_2); m 7.24-7.49 (5H, aromatic). *Anal.* Calcd for $C_{14}H_{16}N_2$: C, 79.29; H, 7.60; N, 13.20%. Found: C, 78.55; H, 7.73; N, 12.98%.

3-(4'-Anisyl)-6,7,8,9-tetrahydro-5*H*-imidazo[1,2-*a*]azepine (2b).

Obtained as a white solid analogous to compound 2a. The recrystallization was from ether, (54%), m.p.125-6°C. ^{1}H-NMR: s 6.66 (1H,2-**H**); t 3.86 (2H,5-CH_2); m 1.72 (6H,6.7.8-$|CH_2|_3$); t 2.86 (2H,9-CH_2); dd 7.02-7.23 (5H,aromatic); s 3.79 (3H,O-CH_3). *Anal.* Calcd for $C_{15}H_{18}N_2O$: C, 74.35; H, 7.49; N, 11.56%. Found: C, 74.02; H, 7.33; N, 11.29%.

3-(4'-Chlorophenyl)-6,7,8,9-tetrahydro-5*H*-imidazo[1,2-*a*]azepine (2c).

Obtained analogous to compound 2a. The recrystallization was from ether, (70%), m.p.122-3 °C. ^{1}H-NMR: s 6.79 (1H,2-**H**); t 3.91 (2H,5-CH_2); m 1.74 (6H,6.7.8-$|CH_2|_3$); t 2.86 (2H,9-CH_2); dd 7.33-7.53 (5H,aromatic). *Anal.* Calcd for $C_{14}H_{15}ClN_2$: Cl, 14.21; N, 11.38%. Found: Cl, 14.12; N, 11.45%.

3-(4'-Bromophenyl)-6,7,8,9-tetrahydro-5*H*-imidazo[1,2-*a*]azepine (2d).

Obtained analogous to compound 2a. The recrystalization was from the mixture of the hexane-propanol-2 (1:1), (74%), m.p.118-9 °C. ^{1}H-NMR: s 6.78 (1H,2-**H**); t 3.92 (2H,5-CH_2), m 1.73 (6H,6.7.8-$|CH_2|_3$); t 2.86 (2H,9-CH_2); dd 7.35-7.53 (5H,aromatic). *Anal.* Calcd. for $C_{14}H_{15}BrN_2$: Br, 27.21; N, 9.66%. Found: Br, 26.84; N, 9.48%.

1-(4'-Methylphenacyl)-3-phenyl-6,7,8,9-tetrahydro-5*H*-imidazo[1,2-*a*]azepinium bromide (4a).

The solution of 4-methylphenacylbromide (0.4 g, 3.2 mmol) in acetone (30 ml) was added to the solution of the 3-phenyl-6,7,8,9-tetrahydro-5*H*-imidazo[1,2-*a*]azepine (**2a**) (0.71 g, 3.2 mmol) in acetone (30 ml). The obtained solution was refluxed for 1 h and then cooled. The precipitated product was filtered and washed with ether, affording 1.5 g of **4a**. The analytical sample was obtained by recrystallization from acetone, m.p. 247-8 °C. ^{1}H-NMR: s 7.71 (1H,2-**H**), t 4.22 (2H,5-CH_2); br s 1.87 (6H,6.7.8-$|CH_2|_3$); t 3.14 (2H,9-

CH_2); dd 7.49-7.98 (4H, aromatic); s 6.17 (2H,$COCH_2$); s 2.43 (3H,CH_3); m 7.51-7.63 (5H,aromatic). IR: ν 3080 cm^{-1},2930,2820 (C-H), 1680 (C=O), 1595 (C=C). *Anal.* Calcd. for $C_{23}H_{25}BrN_2O$: C, 64.94; H, 5.92; Br, 18.78; N, 6.59%.Found: C, 64.32; H, 5.65; Br, 18.83; N, 6.76%.

1-Phenacyl-3-(4"-anisyl)-6,7,8,9-tetrahydro-5*H*-imidazo[1,2-*a*]azepinium bromide (4b).

The base **2b** was converted into the salt **4b** by the same treatment as in the case for the preparation of compound **4a**. Yield 68%. The analytical sample was obtained by recrystallization from propanol-2, m.p. 237-8 °C. ^{1}H-NMR: s 7.68 (1H,2-H); t 4.18 (2H,5-CH_2); br s 1.85 (6H,6.7.8-[CH_2]3); t 3.14 (2H,9-CH_2); dd 7.16-7.40 (4H,aromatic); s 6.17 (2H,$COCH_2$); s 3.83 (3H,OCH_3); m 7.60-8.10 (5H,aromatic). IR: ν 3060 cm^{-1},2940,2820 (C-H), 1685 (C=O), 1590 (C=C). *Anal.* Calcd. for $C_{23}H_{25}BrN_2O_2$: C, 62.59; H, 5.71; Br, 18.10; N, 6.35%. Found: C, 62.12; H, 5.61; Br, 18.03; N, 6.19%.

1-(4'-Bromphenacyl)-3-(4"-anisyl)-6,7,8,9-tetrahydro-5*H*-imidazo[1,2-*a*]azepinium bromide (4c).

The compound **4c** was prepared in the same order as compound **4a**. Yield 80%. The analytical sample was obtained by recrystallization from propanol-2, m.p. 241-2 °C. ^{1}H-NMR: s 7.64 (1H,2-H); t 4.21 (2H,5-CH_2); br s 1.86 (6H,6.7.8-[CH_2]3); t 3.16 (2H,9-CH_2); dd 7.15-7.43 (4H,aromatic); s 6.19 (2H,$COCH_2$); s 3.84 (3H,OCH_3); m 7.84-8.02 (4H,aromatic). IR: ν 3060 cm^{-1},2940,2830 (C-H), 1690 (C=O), 1580 (C=C). *Anal.* Calcd. for $C_{23}H_{24}Br_2N_2O_2$: C, 53.10; H, 4.65; Br, 30.72; N, 5.38%. Found: C, 52.91; H, 4.48; Br, 30.28; N, 5.24%.

1-(4'-Methoxyphenacyl)-3-(4"-chlorophenyl)-6,7,8,9-tetrahydro-5*H*-imidazo[1,2-*a*]azepinium bromide (4d).

The treatment of **2d** in the same order as described for the preparation of the **4a** gave **4d** (79%) as a colourless powder. The analytical sample was obtained by recrystallization from propanol-2, m.p. 232-3 °C. ^{1}H-NMR: s 7.72 (1H,2-H); t 4.19 (2H,5-CH_2); br s 1.83 (6H,6.7.8-[CH_2]3); t 3.14 (2H,9-CH_2); dd 7.18-8.03 (4H,aromatic); s 6.13 (2H,$COCH_2$); s 3.88 (3H,OCH_3); m 7.54-7.66 (4H,aromatic). IR: ν 3050 cm^{-1},2940,2830 (C-H), 1690 (C=O), 1595 (C=C). Anal. Calcd. for $C_{23}H_{24}BrClN_2O_2$: C, 58.06; H, 5.08; N, 5.89%. Found: C, 57.65; H, 4.94; N, 5.77%.

1-[1-(4'-Chlorophenyl)-propan-1-on-2-yl]-3-(4"-chlorophenyl)-6,7,8,9-tetrahydro-5*H*-imidazo[1,2-*a*]azepinium bromide (4e).

The base **2c** was converted into salt **4e** by the same treatment as for the preparation of compound **4a**. Yield 76%. The analytical sample was obtained by recrystallization from propanol-2, m.p. 208-9 °C. ^{1}H-NMR: s 8.05 (1H,2-H); t 4.19 (2H,5-CH_2); br s 1.87 (6H,6.7.8-[CH_2]3); t 3.20 (2H,9-CH_2); dd 7.55-7.68 (4H,aromatic); d 1.82 (3H,$CHCH_3$); q 6.73 (1H,$CHCH_3$); dd 7.50-8.15 (4H,aromatic). IR: ν 3050 cm^{-1},2940,2830 (C-H), 1690 (C=O), 1595 (C=C). *Anal.* Calcd. for $C_{23}H_{23}BrCl_2N_2O$: C, 55.89; H, 4.69; N, 5.67%. Found: C, 55.63; H, 4.61; N, 5.58%.

1-(4'-Tolyl)-4-phenyl-5,6,7,8-tetrahydro-2a,4a-diazacyclopenta[*c,d*]azulene (5a).

The solid salt **4a** (1.06 g, 2,5 mmol) was added as a single portion to the solution of 5% NaOH (150 ml). The obtained suspension was boiled for 1 h and then cooled. The formed precipitate was collected, washed with water and dried *in vacuo*. Yield 0.71 g (87%). The analytical sample was obtained by recrystallization

from benzene/heptane (1:1), m.p.123-4 °C. ^{1}H-NMR: s 6.75 (1H,2-H), s 6.96 (1H,3-H); s 3.74 (2H,5-CH_2); br s 2.00 (4H,6+7|CH_2|2), s 2.90 (2H,8-CH_2); br m 7.14-7.42 (9H,aromatic); s 2.37 (3H,C-CH_3). IR: ν 2810 cm^{-1},2915 (C-H), 1600,1520 (C=C). *Anal.* Calcd. for $C_{23}H_{22}N_2$: C, 84.63; H, 6.79; N, 8.58%. Found: C, 84.21; H, 6.68; N, 8.43%.

1-(Phenyl)-4-(4'-anisyl)-5,6,7,8-tetrahydro-2a,4a-diazacyclopenta[*c,d*]azulene (5d).

The salt **4b** was converted into the base **5d** by the same treatment as in the case for the preparation of the compound **5a**. Yield 92%. The analytical sample was obtained by recrystallization from benzene, m.p. 170-2 °C. ^{1}H-NMR: s 6.89 (1H,2-H); s 7.22 (1H,3-H); s 3.68 (2H,5-CH_2); br s 1.93 (4H,6+7|CH_2|2); s 2.79 (2H,8-CH_2); br m 7.02-7.50 (9H,aromatic); s 3.81 (3H,O-CH_3). IR: ν 2940 cm^{-1},2830 (C-H), 1610,1500 (C=C). *Anal.* Calcd. for $C_{23}H_{22}N_2O$: C, 80.67; H, 6.48; N, 8.18%. Found: C, 81.02; H, 6.36; N, 8.03%.

1-(4'-Bromophenyl)-4-(4'-anisyl)-5,6,7,8-tetrahydro-2a,4a-diazacyclopenta[*c,d*]azulene (5f).

The salt **4c** was converted into the base **5f** by the same treatment as for the preparation of the compound **5a**. Yield 87%. The analytical sample was obtained by recrystallization from pyridine, m.p. 193-4 °C. ^{1}H-NMR: s 6.90 (1H,2-H); s 7.18 (1H,3-H); s 3.64 (2H,5-CH_2); br s 1.95 (4H,6+7|CH_2|2); s 2.80 (2H,8-CH_2); br m 7.00-7.60 (9H,aromatic); s 3.83 (3H,O-CH_3). IR: ν 2930 cm^{-1}, 2840 (C-H), 1600,1500 (C=C). *Anal.* Calcd. for $C_{23}H_{21}BrN_2O$: C, 65.57; H, 5.02; Br, 18.96; N, 6.75%. Found: C, 65.33; H, 4.96; Br, 18.45; N, 6.64%.

1-(4'-Anisyl)-4-(4'-chlorophenyl)l-5,6,7,8-tetrahydro-2a,4a-diazacyclopenta[*c,d*]azulene (5g).

The salt **4d** was converted into base **5g** by the same treatment as for the preparation of the compound **5a**. Yield 84%. The analytical sample was obtained by recrystallization from pyridine, m.p. 193-4 °C. ^{1}H-NMR: s 6.82 (1H,2-H); s 7.35 (1H,3-H); s 3.69 (2H,5-CH_2); br s 1.88 (4H,6+7|CH_2|2); s 2.74 (2H,8-CH_2); br m 6.91-7.55 (9H,aromatic); s 3.76 (3H,O-CH_3). IR: ν 2930 cm^{-1}, 2830 (C-H), 1610,1520 (C=C). *Anal.* Calcd. for $C_{23}H_{21}ClN_2O$: C, 73.30; H, 5.62; Cl, 9.41; N, 7.43%. Found: C, 73.00; H, 5.53; Cl, 9.26; N, 7.29%.

1,4-Di-(4'-chlorophenyl)-2-methyl-5,6,7,8-tetrahydro-2a,4a-diazacyclopenta[*c,d*]azulene (5i).

The salt **4e** was converted into base **5i** by the same treatment as for the preparation of the compound **5a**. Yield 62%. The analytical sample was obtained by recrystallization from benzene, m.p. 199-200 °C. ^{1}H-NMR: s 1.93 (3H,2-CH_3); s 7.08 (1H,3-H); s 4.00 (2H,5-CH_2); br s 1.73 (4H,6+7|CH_2|2); s 2.34 (2H,8-CH_2); br m 7.33-7.57 (9H,aromatic). IR: ν 2940 cm^{-1},2830 (C-H), 1600,1510 (C=C). *Anal.* Calcd. for $C_{23}H_{20}Cl_2N_2$: C, 69.88; H, 5.10; Cl, 17.94; N, 7.09%. Found: C, 69.48; H, 5.12; Cl, 17.48; N, 6.97%.

1-(4'-Anisyl)-4-phenyl-5,6,7,8-tetrahydro-2a,4a-diazacyclopenta[*c,d*]azulene (5b).

4-Methoxyphenacylbromide (2.29 g, 10 mmol) was added to the mixture of **2a** (2.12 g, 10 mmol) in 150 ml of 2-propanol. The reaction mixture was refluxed for 1 h (last for 15 min. - with charcoal). After hot filtration the solvent was removed *in vacuo* and 5% NaOH (150 ml) added. The mixture was boiled for 1 h, cooled, while a precipitate was formed, and was collected by filtration, washed with water, and dried *in vacuo*. The crude product was purified by crystallization from benzene-hexane (1:1) to give **5b**. The desired product (2.4 g,70%) was obtained as a pale-yellow solid. The analytical sample was obtained by recrystallization from

benzene/hexane (1:1), m.p. 162-4 °C. ^{1}H-NMR: s 6.75 (1H,2-H); s 6.98 (1H,3-H); s 3.72 (2H,5-CH_2); br s 2.07 (4H,6+7[CH_2]$_2$); s 2.89 (2H,8-CH_2); br m 6.92-7.43 (9H,aromatic); s 3.83 (3H,O-CH_3). IR: ν 2930 cm^{-1},2815 (C-H), 1600,1510 (C=C). *Anal.* Calcd. for $C_{23}H_{22}N_2O$: C, 80.67; H, 6.48; N, 8.18%. Found: C, 80.22; H, 6.35; N, 8.00%.

1-(4'-Phenethyl)-4-phenyl-5,6,7,8-tetrahydro-2a,4a-diazacyclopenta[*c,d*]azulene (5c).

The mixture of the base 2a and 4'-ethoxyphenacyl bromide was converted into base 5c by the same treatment as for the preparation of the compound 5b. Yield 67%. The analytical sample was obtained by recrystallization from benzene, m.p. 133-5 °C. ^{1}H-NMR: s 6.70 (1H,2-H); s 6.97 (1H,3-H); s 3.75 (2H,5-CH_2); br s 2.02 (4H,6+7[CH_2]$_2$); s 2.89 (2H,8-CH_2); br m 6.89-7.43 (9H,aromatic); t 1.44 (3H,CH_2-CH_3); k 4.07 (2H,CH_2-CH_3). IR: ν 2960 cm^{-1},2810 (C-H), 1615,1520 (C=C). *Anal.* Calcd. for $C_{24}H_{24}N_2O$: C, 80.87; H, 6.79; N, 7.86%. Found: C, 80.34; H, 6.66; N, 7.79%.

1-(4'-Chlorophenyl)-4-(4'-anisyl)-5,6,7,8-tetrahydro-2a,4a-diazacyclopenta[*c,d*]azulene (5e).

The mixture of base 2b and 4'-chlorophenacyl bromide was converted into base 5e by the same treatment as for the preparation of compound 5b. Yield 73%. An analytical sample was obtained by recrystallization from DMF, m.p. 165-6 °C. ^{1}H-NMR: s 6.91 (1H,2-H); s 7.22 (1H,3-H); s 3.68 (2H,5-CH_2); br s 1.95 (4H,6+7[CH_2]$_2$); s 2.78 (2H,8-CH_2); br m 7.02-7.41 (9H,aromatic); s 3.81 (3H,O-CH_3). IR: ν 2910 cm^{-1},2830 (C-H), 1600, 1500 (C=C). *Anal.* Calcd. for $C_{23}H_{21}ClN_2O$: C, 73.30; H, 5.62; Cl, 9.41; N, 7.43%. Found: C, 73.09; H, 5.56; Cl, 9.19; N, 7.38%.

1-(4'-Anisyl)-4-(4"-bromophenyl)-5,6,7,8-tetrahydro-2a,4a-diazacyclopenta[*c,d*]azulene (5h).

The mixture of base 2d and 4'-methoxyphenacyl bromide was converted into base 5h by the same treatment as in the case for the preparation of compound 5b. Yield 71%. An analytical sample was obtained by recrystallization from benzene, m.p. 186-8 °C. ^{1}H-NMR: s 6.74 (1H,2-H); s 7.00 (1H,3-H); s 3.69 (2H,5-CH_2); br s 1.94 (4H,6+7[CH_2]$_2$); s 2.88 (2H,8-CH_2); br m 6.92-7.57 (9H,aromatic); s 3.83 (3H,O-CH_3). IR: ν 2940 cm^{-1},2810 (C-H), 1605,1500 (C=C). *Anal.* Calcd. for $C_{23}H_{21}BrN_2O$: C, 65.57; H, 5.02; Br, 18.96; N, 6.65%. Found: C, 65.11; H, 4.94; Br, 18.46; N, 6.51%.

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