Role of aromatic substituent in the photochemical reactions of naphthyl isoxazolines fused with saturated heterocyclic rings

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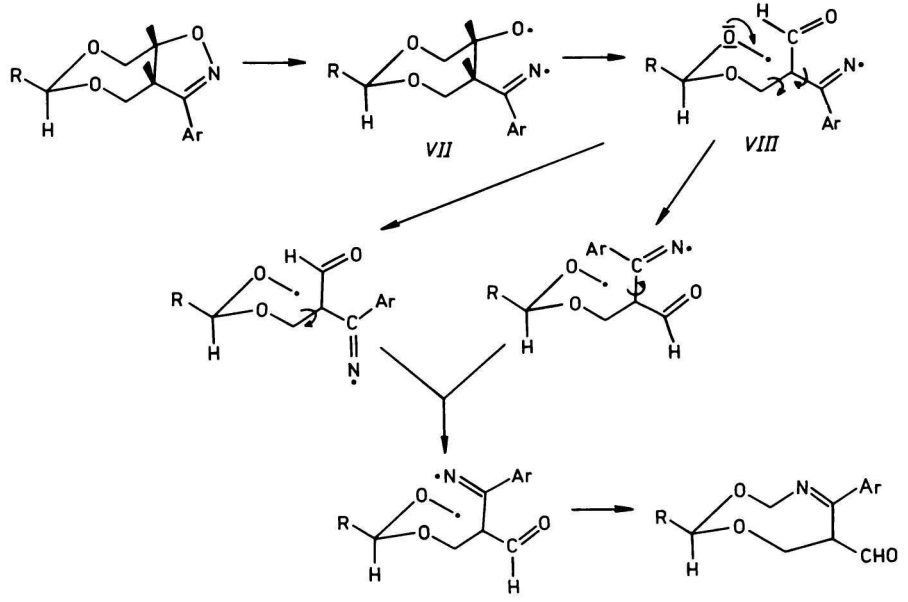
Character of aromatic substituent in the position 3 of 2-isoxazolines fused with the saturated heterocyclic rings in 4,5-position exerts strong influence on the outcome of the photochemical reaction. In contrast to phenyl-substituted isoxazolines those having α-naphthyl or β-naphthyl substituents in position 3 are either photostable or suffer gradual photodestruction, depending on the UV radiation used.

Характер ароматического заместителя в положении 3 2-изоксазолинов с конденсированными насыщенными гетероцыклами в положении 4,5 значительно влияет на результат фотохимической реакции. Обмен фенила α- и β-нафтилом способствует в большинстве случаев фотоустойчивости или постепенной деструкции, в зависимости от характера источника УФ-излучения.

In the majority of cases by the irradiation of isoxazolines the biradical resulting from the homolytic N—O bond cleavage was established as the primary product. Its further fate depends on the substitution pattern of the starting isoxazolines [1—10]. We have found that if the attached heterocyclic ring contains oxygen in β-position to the oxygen of isoxazoline the ensuing stabilization of the secondary biradical *VIII* (Scheme 1) can change the course of reaction to produce cyclic enamino aldehydes selectively and in high yields [11—16]. Hence derivatives of 2,3-dihydro-6*H*-1,3-oxazine (*I*→*II*), (*III*→*IV*) [11, 12, 15, 16] and 2,4,5,8-tetrahydro-1,3-dioxa-5-azocine (*V*→*VI*) were obtained. The selectivity of the rearrangement was rather unexpectedly influenced by the substituent on the aromatic moiety in the position 3 of isoxazoline. In the presented paper we set out to explore the role of aromatic substituent using naphthyl-substituted 2-isoxazolines with fused rings as models.

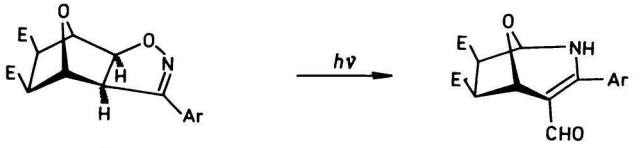
Model compounds synthesized by 1,3-dipolar cycloaddition comprised 9-(1-naphthyl)- (*Ib*) and 9-(2-naphthyl)-3,4-dimethoxycarbonyl-7,10-dioxa-8-azatricyclo-



Scheme 1

-[4,3,0,$1^{2,5}$]dec-8-ene (*Ic*), 4-(1-naphthyl)-2,7-dioxa-3-azabicyclo[3,3,0]oct-3-ene (*IIIb*), and 4-R-10-(1-naphthyl)-3,5,8-trioxa-9-azabicyclo[5,3,0]dec-9-ene (*Vb*, *Vd*—*Vg*) (Scheme 2). Nitrile oxides used in the 1,3-dipolar cycloadditions were prepared directly by treating the corresponding oximes with hypochlorite and triethylamine [17]. Structures of thus synthesized model compounds were established by ^{1}H and ^{13}C NMR spectroscopy, corroborated by the spectra of the corresponding phenyl derivatives. By the cycloadditions of benzonitrile oxide [13, 14, 18] and antracenonitrile oxide [19] to the 2-substituted 1,3-dioxep-5-ene we have found *endo-exo* stereoselectivity towards the substituent in the position 2, producing both stereoisomers. α-Naphthonitrile oxide, on the other hand, produced exclusively *endo* derivatives *Vd*—*Vg*, having the substituent and isoxazoline on the opposite sides of the dioxepine skeleton. The structural assignment was based on ^{13}C NMR data analyses, using C-2 and C-6 signals as indicators. Compared to nonsubstituted derivatives of the type *V exo* isomers of the analogous phenyl and 9-antryl derivatives display C-2, C-6 signals at about the same δ value, shifted approximately 5 ppm upfield. Naphthyl-substituted derivatives possess those signals at $\delta = 71.53$ ppm and 66.33 ppm, indicating an *endo* arrangement. The analysis corresponds to that made in detail for phenyl analogues [15].

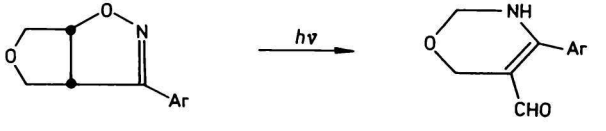
To our knowledge there is only one paper so far dealing with the photochemistry



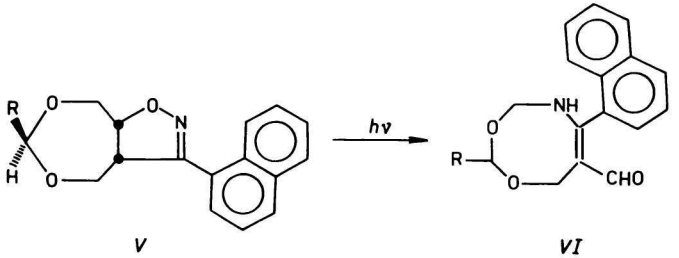
I

E = $COOCH_3$

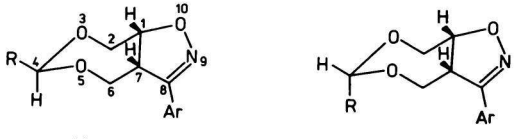
II



III *IV*



V *VI*



V endo *V exo*

I—VI	Ar	*V, VI*	R
a	Phenyl	*d*	Methyl
b	α-Naphthyl	*e*	i-Propyl
c	β-Naphthyl	*f*	i-Butyl
		g	Phenyl

Scheme 2

of α-naphthyl-substituted 2-isoxazoline [10]. In contrast to other 3-aryl-substituted 2-isoxazolines 3-(1-naphthyl)-2-isoxazoline was found to be photostable under various irradiation conditions. Having found a linear relationship between the singlet energy and the quantum yield of the photoreaction the authors proposed

the lack of reactivity to be due to the too low singlet energy of the naphthyl-substituted derivative. Besides that intensive fluorescence was observed at room temperature as well as phosphorescence at liquid nitrogen temperature. However structural modifications can enhance photoreactivity, as can be seen from the example of strained 4-(1-naphthyl)-2-oxa-3-azabicyclo[3,1,0]hex-3-ene which was found to undergo photofragmentation at room temperature and photorearrangement at -50 °C to -70 °C in methanol [20]. Since configuration of the fused heterocyclic ring can have influence on the absorption spectra [18] we have irradiated the model compounds utilizing two different sources of UV light, namely a low-pressure Hg lamp with the principal emission line at $\lambda = 254$ nm and medium-pressure Hg lamp in quartz vessel.

As seen from Table 1, when irradiated at $\lambda = 254$ nm 1-naphthyl-substituted derivatives *IIIb, Ib, Vb, Vd* were photostable, whereas *Ve* and *Vf* decomposed to intractable material gradually. Possible products in the case of *Ve* indicated by LC analysis were too unstable to be identified.

Well-known wavelength-dependent photochemistry of oxazoles and isoxazoles and the presence of two absorption maxima in the UV spectra of our derivatives prompted us to investigate their behaviour when irradiated with medium-pressure Hg lamp, the light of which would be absorbed exclusively by the long wavelength maximum at 295—300 nm. Under such conditions *IIIb* behaved like its phenyl-substituted analogue, producing besides polymeric material isomerization product, enamino aldehyde (*IV*) [11, 12]. ^{1}H NMR spectrum of *IVb* features aldehydic proton signal at $\delta = 8.66$ ppm doublet of the CH_2 group ($J_{2,3} = 1.5$ Hz) and NH proton at $\delta = 4.28$ ppm. Derivatives *Ib, Ic, Vb, Vf* suffered quick decomposition, *Ve* decomposed within 1 h. *Vd*, when irradiated with the medium-pressure Hg lamp furnished besides polymers 33 % of isomerization product 4-methyl-6-(1-naphthyl)-7-formyl-2,4,5,8-tetrahydro -1,3-dioxa-5-azocine (*VId*). The identification was facile due to comparison with its 6-phenyl-substituted analogue (see Experimental) (Scheme 1).

Comparison of naphthyl- and phenyl-substituted 2-isoxazolines with fused oxygen-containing saturated rings demonstrates clearly lower photoreactivity of naphthyl derivatives, although there is no clear cut division as with the 3-aryl-2-isoxazolines [10].

The presence of two separated maxima in the absorption spectra of derivatives *I, III,* and *V* implies existence of two different excited states, both with $\Pi^* \leftarrow \Pi$ character. At $\lambda = 254$ nm there is a relative minimum with $\log(\varepsilon/(\mathrm{dm^3\ mol^{-1}\ cm^{-1}}))$ values ≈ 2.4 making the excitation of 220 nm aryl group maximum very inefficient. Moreover there is a fairly efficient fluorescence ($\Phi = 0.34$ in ethanol, $\lambda_{exc} = 280$ nm), competing with the quantum efficiency of the $\Pi^* \leftarrow \Pi$ excited state. Thus within irradiation time 10—12 h derivatives *IIIb, Ib, Ic, Vb,* and *Vd* were photostable (LC analysis, Table 1), derivatives possessing

Table 1

Course of irradiation of naphthyl-substituted fused isoxazolines in methanol

Compound	Irradiation conditions λ/nm	Fraction of the starting compound/%							Irradiation outcome
		1/2 h	1 h	1 1/2 h	2 h	3 h	4 h	5 h	
IIIb	254	—	100.0	—	97.6	96.7	96.7	96.3	Photostable
	polychr.	8.8	18.6	25.8	38.3	50.3	62.1	72.6	*IIIb*
Ib	254	—	99.7	—	99.0	97.2	93.0	90.0	Photostable
	polychr.	91.0	85.6	79.1	73.3	63.0	52.7	44.0	Decomposition
Ic	254	—	100.0	—	100.0	100.0	100.0	99.8	Photostable
	polychr.	—	77.0	—	61.3	55.5	50.5	—	Decomposition
Vb	254	—	99.7	—	99.3	97.0	95.5	93.8	Photostable
	polychr.	89.4	81.3	65.0	59.6	—	—	—	Decomposition
Vd	254	—	91.0	—	90.7	90.3	90.1	90.0	Photostable
	polychr.	93.1	80.6	71.0	60.4	37.4	28.5	—	*VId*
Ve	254	—	82.7	—	79.7	79.2	79.3	76.5	Nonisolable product
	polychr.	—	0	—	—	—	—	—	Total decomposition
Vf	254	—	82.6	—	82.4	74.0	71.0	68.3	Decomposition
	polychr.	97.9	97.1	96.7	92.6	75.0	70.3	60.0	Decomposition
Vg	254	—	96.2	—	—	77.6	70.1	57.8	Decomposition
	polychr.	83.7	53.4	39.4	31.1	28.5	—	—	Rapidly decomposing product

a bulky substituent in the position 4 (*Ve—Vg*) decomposed without forming products. Although absorption spectra of the latter compounds do show a weak shoulder at $\lambda = 250$ nm, as do their 10-phenyl-substituted analogues [13, 14, 18], of as yet unknown origin, their bulkiness probably affects more the "vibronic assistance" needed to enhance the photoreactivity [10]. The longer wavelength maximum of 10-naphthyl-substituted fused 2-isoxazolines, belonging to the aryl—C=N—O chromophore, is red-shifted by about 20—30 nm compared to 10-phenyl derivatives. However, while the latter undergo upon irradiation clean isomerization to cyclic enamino aldehydes [11—16] naphthyl-substituted derivatives do so in only two cases (*IIIb, Vd*). Other examined derivatives gradually decompose without forming products. Thus ring-fused 3-naphthyl-substituted 2-isoxazolines proved to be more photoreactive than the parent 3-(1-naphthyl)-2-isoxazoline, albeit their singlet energy was still lower than the 376 kJ limit found experimentally for 3-aryl-2-isoxazolines [10]. Their inability to produce isomerization products may be due to steric hindrance encountered in the ring-forming step (Scheme 2) aggravated by bulky R.

Experimental

^{1}H NMR spectra were measured on the Tesla 80 MHz BS 487 C model, ^{13}C NMR spectra on the Jeol XF 100 in deuteriochloroform with tetramethylsilane as internal standard.

Electronic spectra were taken with a Perkin—Elmer model 323 in methanol, ε values are in $dm^3\ mol^{-1}\ cm^{-1}$. Irradiation was performed in forced circulation reactors [21], total solvent volume 270 cm^3 with a low-pressure Hg lamp Toshiba G1 15 (15 W). All irradiation experiments were done under nitrogen atmosphere, the irradiated solution was checked with HPLC (Spectra Physics, inversion column, eluted with methanol—water (volume ratio = 7 : 3)). Products were isolated on a silica gel column, eluent ethyl acetate—cyclohexane (volume ratio = 7 : 3).

Preparation of polycyclic isoxazolines by 1,3-dipolar cycloaddition of naphthonitrile oxides

Equimolar amounts of naphthaldehyde oxime and the dipolarophile were dissolved in small volume of dichloromethane, triethylamine added (mole ratio = 1 :10) and an excess of hypochlorite solution (mole ratio = 5 : 1) was dropwise added to the stirred reaction mixture. During the hypochlorite addition the temperature is maintained at 25—30 °C, the reaction mixture is then left to stand overnight. Cycloaddition product is then extracted by dichloromethane, combined extracts dried by magnesium sulfate and concentrated *in vacuo*. Oily residue solidifies after treatment with ether.

3,4-Dimethoxycarbonyl-9-(1-naphthyl)-7,10-dioxa-8-azatricyclo[4,3,0,$1^{2,5}$]dec-8-ene (*Ib*), yield = 40 %, m.p. = 186—189 °C. For $C_{21}H_{19}NO_6$ ($M_r = 381.37$) w_i(calc.): 66.13%

C, 5.02 % H, 3.67 % N; w_i(found): 66.24 % C, 4.96 % H, 3.81 % N. Ultraviolet spectrum, λ_{max}/nm (log {ε}): 224 (3.22), 303 (3.21). ^{1}H NMR spectrum, δ/ppm: 7.37—8.90 (m, 7H, aromatic protons), 5.06 (d, 1H, $J_{1,6}$ = 6.5 Hz, H-6), 5.01 (s, 1H, H-5), 4.77 (s, 1H, H-2), 4.52 (d, 1H, H-1), 3.5 and 3.6 (s, s, 6H, 2 × $COOCH_3$), 3.25 and 3.32 (s, s, 2H, H-3 and H-4). ^{13}C NMR spectrum, δ/ppm: 171.22 (s, C=O), 155.83 (s, C=N), 131.16, 129.27, 128.56, 127.91, 127.45, 127.13, 125.89 (aromatic carbons), 85.57 (d, C-2 and C-5), 80.83 (d, C-6), 61.42 (d, C-1), 52.13 and 52.00 (q, CH_3), 50.38 and 47.07 (s, s, C-3 and C-4).

3,4-Dimethoxycarbonyl-9-(2-naphthyl)-7,10-dioxa-8-azatricyclo[4.3.0.$1^{2,5}$]dec-8-ene (*Ic*), yield=42 %, m.p.=196—199 °C. For $C_{21}H_{19}NO_6$ w_i(found): 65.98 % C, 5.31 % H, 3.44 % N. Ultraviolet spectra, λ_{max}/nm (log {ε}): 266 (3.25), 288 (3.16), 300 (3.15). ^{1}H NMR spectrum, δ/ppm: 7.22—8.37 (m, 7H, aromatic protons), 5.16 (s, 1H, H-5), 5.08 (s, 1H, H-2), 4.96 (d, 1H, $J_{1,6}$=8.0 Hz, H-6). 4.05 (d, 1H, H-1), 3.67 and 3.70 (s, s, 6H, 2 × $COOCH_3$), 3.16 (d, d, 2H, H-3 and H-4). ^{13}C NMR spectrum, δ/ppm: 171.33 (s, C=O), 155.48 (s, C=N), 87.12 (d, C-5), 85.37 (d, C-2), 80.97 (d, C-6), 58.28 (d, C-1), 52.17 and 50.68 (q, CH_3), 47.07 (d, C-3 and C-4).

4-(1-Naphthyl)-2,7-dioxa-3-azabicyclo[3.3.0]oct-3-ene (*IIIb*), yield=46 %, m.p.= =110—112 °C. For $C_{15}H_{13}NO_2$ (M_r = 239.26) w_i(calc.): 75.30 % C, 5.48 % H, 5.86 % N; w_i(found): 75.21 % C, 5.66 % H, 5.94 % N. Ultraviolet spectra, λ_{max}/nm (log {ε}): 229 (3.33), 304 (2.91). ^{1}H NMR spectrum, δ/ppm: 8.77—9.05 and 7.40—8.10 (m, 7H, aromatics), 5.40 (dd, 1H, $J_{1,5}$ = 9.0 Hz, $J_{1,8}$=3.0 Hz, H-1), 4.72 (m, 1H, H_A-8), 4.25 (d, 1H, H-5), 3.65—4.07 (m, 3H, H_B-8, H_A-6, H_B-6). ^{13}C NMR spectrum, δ/ppm: 158.07 (s. C=N), 85.82 (d, C-1), 77.12 (t, C-8), 72.44 (t, C-6), 57.30 (d, C-5).

10-(1-Naphthyl)-3,5,8-trioxa-9-azabicyclo[5,3,0]dec-9-ene *Vb*), yield = 60 %, m.p.= = 145—146 °C. For $C_{16}H_{15}NO_3$ (M_r=269.29) w_i(calc.): 71.36 % C, 5.61 % H, 5.20 % N; w_i(found): 71.12 % C, 5.87 % H, 5.31 % N. Ultraviolet spectra, λ_{max}/nm (log {ε}): 293 (2.93). ^{1}H NMR spectrum, δ/ppm: 8.40—8.72 and 7.42—8.10 (m, 7H, aromatic protons), 4.75—5.17 (m, 2H, H_A-4, H-7), 3.70—4.62 (m, 6H, H_B-4, H-1, H_2-2 and H_2-6). ^{13}C NMR spectrum, δ/ppm: 157.49 (s, C=N), 99.60 (t, C-4), 83.23 (d, C-7), 72.18 (t, C-6), 68.28 (t, C-2), 55.74 (d, C-1).

endo-4-Methyl-10-(1-naphthyl)-3,5,8-trioxa-9-azabicyclo[5,3,0]dec-9-ene (*Vd*), yield= = 31 %, m.p. = 165—166 °C. For $C_{17}H_{17}NO_3$ (M_r=283.31) w_i(calc.): 72.06 % C, 6.05 % H, 4.94 % N; w_i(found): 71.93 % C, 6.16 % H, 5.07 % N. Ultraviolet spectrum, λ_{max}/nm (log {ε}): 226 (3.34), 293 (2.83). ^{1}H NMR spectrum, δ/ppm: 8.37—8.62 and 7.35—7.82 (m, 7H), 4.72—5.02 (m, 1H, H-7), 4.50 (q, 1H, H-4), 3.75—4.40 (m, 5H, H_2-2, H_2-6, H-1), 1.10 (d, 3H, CH_3). ^{13}C NMR spectrum, δ/ppm: 157.42 (s, C=N), 105.77 (d, C-4), 83.10 (d, C-7), 71.53 (t, C-6), 66.33 (t, C-2), 55.55 (d, C-1), 21.76 (q, CH_3).

endo-4-i-Propyl-10-(1-naphthyl)-3,5,8-trioxa-9-azabicyclo[5,3,0]dec-9-ene (*Ve*), yield = 16 %, m.p. = 161.5 °C. For $C_{19}H_{21}NO_3$ (M_r=311.37) w_i(calc.): 73.29 % C, 6.80 % H, 4.50 % N; w_i(found): 73.41 % C, 7.04 % H, 4.52 % N. Ultraviolet spectrum, λ_{max}/nm (log {ε}): 228 (3.34), 292 (2.91). ^{1}H NMR spectrum, δ/ppm: 8.47—8.75 and 7.35—7.80 (m, 7H), 4.95 (m, 1H, H-7), 3.70—4.50 (m, 6H, H_2-2, H_2-6, H-1, H-4), 2.67 (m, 1H, CH), 0.8 (d, 6H, 2 × CH_3). ^{13}C NMR spectrum, δ/ppm: 157.42 (s, C=N), 112.46 (d, C-4), 83.16 (d, C-7), 71.86 (t, C-6), 66.92 (t, C-2), 55.55 (d, C-1), 17.72 and 17.6 (q, CH_3).

endo-4-i-Butyl-10-(1-naphthyl)-3,5,8-trioxa-9-azabicyclo[5,3,0]dec-9-ene (*Vf*), yield = = 20 %, m.p.=103—106 °C. For $C_{20}H_{23}NO_3$ (M_r=325.39) w_i(calc.): 73.83 % C,

7.12 % H, 4.30 % N, w_i(found): 74.04 % C, 7.17 % H, 4.60 % N. Ultraviolet spectrum, λ_{max}/nm (log {ε}): 227 (3.32), 292 (2.81). ^{1}H NMR spectrum, δ/ppm: 8.45—8.70 and 7.40—8.07 (m, 7H), 4.96 (m, 1H, H-7), 3.82—4.60 (m, 6H, H_2-2, H_2-6, H-4, H-1), 2.85 (m, 1H, CH), 1.42 (m, 2H, CH_2), 0.82 (d, 6H, $2\times CH_3$). ^{13}C NMR spectrum, δ/ppm: 107.58 (d, C-4), 82.85 (d, C-7), 71.29 (t, C-6), 66.36 (t, C-2), 55.32 (d, C-1), 44.28 (d, CH), 24.60 (t, CH_2), 22.85 and 22.66 (q, $2\times CH_3$).

endo-Phenyl-10-(1-naphthyl)-3,5,8-trioxa-9-azabicyclo[5,3,0]dec-9-ene (*Vg*), yield = 34 %, m.p. = 183—185 °C. For $C_{22}H_{19}NO_3$ (M_r = 345.38) w_i(calc.): 76.50 % C, 5.55 % H, 4.06 % N; w_i(found): 73.38 % C, 5.39 % H, 4.18 % N. Ultraviolet spectrum, λ_{max}/nm (log {ε}): 294 (3.21). ^{1}H NMR spectrum, δ/ppm: 7.42—8.72 (m, 7H), 7.27 (s, 5H), 5.45 (s, 1H, H-4), 4.07—5.22 (m, 5H, H_2-2, H_2-6, H-7), 2.87 (m, 1H, H-1). ^{13}C NMR spectrum, δ/ppm: 106.35 (d, C-4), 81.99 (d, C-7), 69.19 (t, C-6), 64.77 (t, C-2), 54.77 (d, C-1).

4-(1-Naphthyl)-5-formyl-2,3-dihydro(6*H*)-1,3-oxazine (*IVb*) was obtained by the irradiation of *IIIb* in methanol for 8 h, yield = 8 %, colourless oil after chromatography. For $C_{15}H_{13}NO_2$ (M_r = 239.26) w_i(calc.): 75.30 % C, 5.48 % H, 5.85 % N; w_i(found): 75.53 % C, 5.71 % H, 5.77 % N. ^{1}H NMR spectrum, δ/ppm: 8.66 (s, 1H, CHO), 7.35—8.17 (m, 7H), 4.92 (d, 2H, J = 1.5 Hz, H_2-2), 4.60 (s, 2H, H_2-6), 4.28 (d, 1H, NH).

4-Methyl-6-(1-naphthyl)-7-formyl-2,4,5,8-tetrahydro-1,3-dioxa-5-azocine (*VId*) was obtained by the irradiation of *Vd* for 4 1/2 h in 33 % yield, colourless oil after chromatography. For $C_{17}H_{17}NO_3$ (M_r = 283.31) w_i(calc.): 72.06 % C, 6.05 % H, 4.94 % N; w_i(found): 72.14 % C, 6.01 % H, 5.21 % N. ^{1}H NMR spectrum, δ/ppm: 8.82 (s, 1H, CHO), 6.87—8.00 (m, 7H), 3.92—5.65 (m, H-2, H_2-4, H_2-8, NH), 1.46 (d, 3H, J = 4.0 Hz).

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