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Dedicated to Full Member of the Russian Academy of Sciences N.S. Zefirov on His 70th Anniversary

Reaction of 2-Alkoxypropenals with α-Hydroxyamino Oximes and 1,2-Bis(hydroxyamino)cyclohexane

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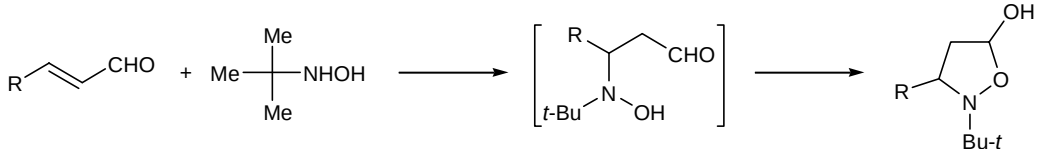
Abstract—Reactions of 2-alkoxypropenals with α-hydroxyamino oximes in neutral medium involve the aldehyde group of the former to afford both acyclic and cyclic azomethine oxides: *N*-(2-hydroxyiminoalkyl)-*N*-(2-alkoxy-2-propenylidene)amine oxides and 1-hydroxy-2,5-dihydroimidazole 3-oxides. The state of tautomeric equilibrium between the cyclic and acyclic products depends on the solvent nature and temperature. The reaction in acidic aqueous medium is accompanied by hydrolysis of the vinyl ether moiety in 2-alkoxypropenals with formation of 2-oxopropionaldehyde which reacts with α-hydroxyamino oxime at the hydroxyamino group to give substituted pyrazine 1,4-dioxides. The reaction of 2-alkoxypropenals with 1,2-bis(hydroxyamino)cyclohexane leads to formation of 2-(1-alkoxyvinyl)-1,3-dihydroxyperhydrobenzimidazoles. The structure of the products was proved by IR, UV, and ^{1}H and ^{13}C NMR spectroscopy and X-ray analysis.

[3+2]-Cycloaddition of nitrones to dipolarophiles underlies a widely used method for the synthesis of five-membered heterocycles [1]. In particular, cycloaddition of nitrones to unsaturated systems, including α,β-unsaturated carbonyl compounds, leads to formation of di- and tetrahydroisoxazole derivatives [2–4]. Some compounds of these series exhibit antiphlogistic, analgetic, and sedative activity [4]. Therefore, synthesis of open-chain and cyclic nitrones on the basis of 2-alkoxypropenals could extend the series of potential biologically active compounds.

The goal of the present work was to study the regioselectivity in reactions of α-hydroxyamino oximes and 1,2-bis(hydroxyamino)cyclohexane with 2-alkoxypropenals **I** whose molecules represent a combination of vinyl ether and α,β-unsaturated aldehyde fragments; we also planned to examine prospects in the synthesis of functionalized heterocyclic systems containing two nitrogen atoms on the basis of the above reaction.

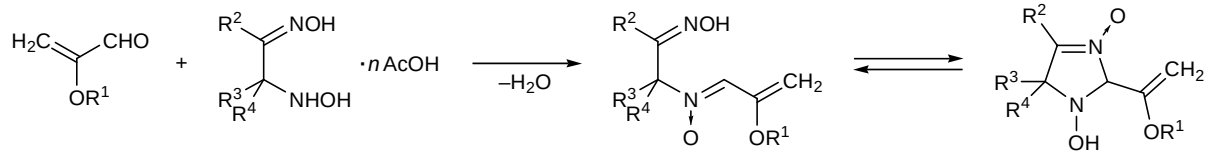
It is known that *N*-(*tert*-butyl)hydroxylamine reacts with propenal and 2-butenal to give the corresponding isoxazolidine derivatives via initial Michael 1,4-addition followed by intramolecular ring closure via attack by the hydroxyamino group on the carbonyl carbon atom (Scheme 1) [5]. In keeping with published data, reactions of aldehydes **I** with α-hydroxyamino oximes **II** or their salts **III** were expected to involve addition

Scheme 1.



R = H, Me.

Scheme 2.



Ia, **Ib** **IIa**, **IIb**, **IIIa**–**IIIc** **IVa**–**IVf** **Va**–**Vf**

I, R^1 = Et (**a**), Me (**b**); **II**, **III**, $R^2 = R^3 = R^4$ = Me (**a**); $R^2R^3 = (CH_2)_4$, R^4 = H (**b**); $R^2 = R^3$ = Me, R^4 = H (**c**); **IV**, **V**, $R^2 = R^3 = R^4$ = Me, R^1 = Et (**a**), Me (**b**); $R^2R^3 = (CH_2)_4$, R^4 = H, R^1 = Et (**c**), Me (**d**); $R^2 = R^3$ = Me, R^4 = H, R^1 = Et (**e**), Me (**f**); **II**, n = 0; **III**, n = 1.

of the hydroxyamino group of **II** both at the vinyl moiety in 2-alkoxypropenal, as is typical of vinyl ethers [6], and at the carbonyl group, as was reported for unsaturated aldehydes [5, 7, 8]. We have found that 2-alkoxypropenals react with α-hydroxyamino oximes in neutral medium at the aldehyde group of the former to afford equilibrium mixtures of tautomeric acyclic and cyclic azomethine oxides, *N*-(2-hydroxyiminoalkyl)-*N*-(2-alkoxy-2-propenylidene)amine oxides and 1-hydroxy-2,5-dihydroimidazole 3-oxides (Scheme 2).

According to the ^{1}H NMR and IR spectra and analytical data, products of the reactions of 2-alkoxypropenals **Ia** and **Ib** with oxime **IIa** or the corresponding acetate **IIIa** have the structure of cyclic azomethine oxides, 1-hydroxy-2,5-dihydro-1*H*-imidazole 3-oxides **Va** and **Vb**. The UV spectrum of **Vb** contained an absorption maximum at λ 232 nm, which indicates the presence of an alkylnitrone moiety (cf. [7]). The structure of compound **Vb** was proved by the X-ray diffraction data (Fig. 1).

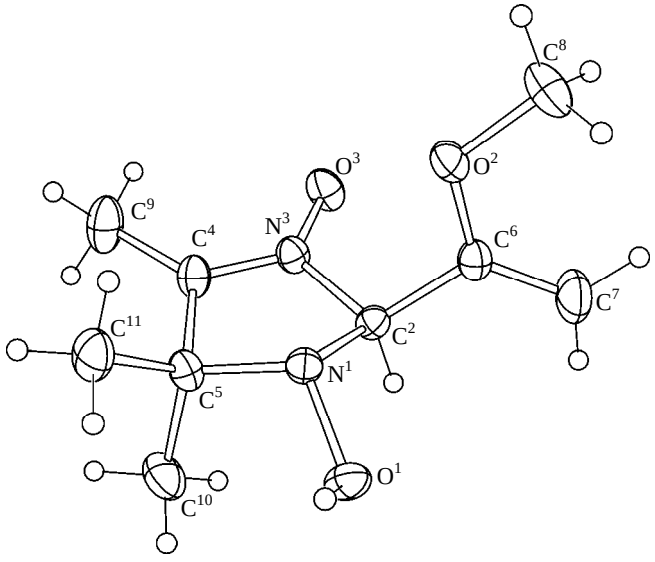


Fig. 1. Structure of the molecule of 1-hydroxy-2-(1-methoxyethenyl)-4,5,5-trimethyl-2,5-dihydro-1*H*-imidazole 3-oxide (**Vb**) according to the X-ray diffraction data.

1-Hydroxy-2,5-dihydro-1*H*-imidazole 3-oxides **Va** and **Vb** were found to exist in tautomeric equilibrium with their open-chain isomers **IVa** and **IVb**. In $CDCl_3$, the ratio of tautomers **Vb** and **IVb** is 8.3:1 or 6.3:1 (11–14% of **IVb**). After 12 h at room temperature, the fraction of acyclic tautomer **IVb** increases to 18%. Taking into account that the equilibration time is fairly long, in some cases ^{1}H NMR spectroscopy detected only cyclic tautomers **Va** and **Vb** (immediately after dissolution in $CDCl_3$). The condensation products obtained from α-hydroxyamino oxime **IIa** (or its acetate **IIIa**) exist in DMSO and CD_3CN exclusively as cyclic structures **V**, while no open-chain tautomers **IVa** and **IVb** were detected by ^{1}H NMR spectroscopy, regardless of the time elapsed before recording the spectrum.

The state of the ring–chain equilibrium also depends on the temperature. According to published data [9], raising the temperature shifts the equilibrium toward open-chain structure. In fact, heating of a solution (in $CDCl_3$) containing tautomers **Vb** and **IVb** in an NMR ampule for 1 h at 58°C resulted in change of the tautomer ratio from 6.3:1 to 2.2:1. After cooling to 27°C and keeping for 1 h at that temperature, the ratio increased to 3.2:1, and after 25 h, to 4.4:1; i.e., the equilibrium was slowly displaced toward the cyclic tautomer.

α-Hydroxyamino oxime **IIb** (or acetate **IIIb**) in which the hydroxyamino group is attached to a secondary carbon atom is more reactive than compound **IIa** (**IIIa**) with the hydroxyamino group at a tertiary carbon atom. The reaction of 2-methoxypropenal with hydroxylamine **IIb** at 80°C is complete in 15 min (according to the ^{1}H NMR data), whereas compound **IIa** does not react under the same conditions; even after heating for 2 h at 78°C, up to 50% of the initial reactants remained unchanged.

Acyclic tautomers **IVc** and **IVd** formed by reaction with secondary hydroxyamino oxime **IIIb** are more stable than tautomers **IVa** and **IVb** derived from

tertiary hydroxylamine **IIa**. Compounds **IVc** and **IVd** can be isolated in the crystalline state (yield 28–37%) by precipitation from diethyl ether or ethyl acetate. The fraction of isomers **IVc** and **IVd** in the reaction mixture reaches ~40–50% (or sometimes ~70–80%; $CDCl_3$, ^1H NMR data). They characteristically show in the ^1H NMR spectra two doublets at δ 4.7–4.8 and 6.40 ppm (=CH_2) and a doublet in the region δ 6.9–7.0 ppm (N=CH). The structure of compound **IVc** was also confirmed by the two-dimensional NMR technique (CH–CORR) (Fig. 2), as well as by ^{13}C (JMOD) spectroscopy. The UV spectrum of a crystalline sample of **IVc** (in KBr) displayed an absorption maximum at λ 264 nm, presumably due to the presence of a conjugated alkylnitrone moiety.

Several factors determine the possibility for isolation of individual isomers **IVc**/**Vc** and **Vd**/**IVd** which occur in a dynamic equilibrium with each other. These factors include the nature of the solvent taken for precipitation or recrystallization, temperature of the solution, time of crystallization (precipitation), and, probably, concentration. For example, the crystalline substance isolated by precipitation with diethyl ether from the reaction mixture obtained from aldehyde **Ia** and hydroxylamine **IIIb** in THF was identified as open-chain tautomer **IVc** by the ^1H NMR spectrum in DMSO-d_6, while in $CDCl_3$ it was a mixture of tautomers **IVc** and **Vc** at a ratio of 1:0.4 to 1:2. In the UV spectrum of the crystalline product in KBr we observed two maxima at λ 243 and 256 nm, which are typical of conjugated nitrone moiety in **IVc**. As with isomers **IVb** and **Vb**, the state of tautomeric equilibrium between **IVc** and **Vc** in $CDCl_3$ depends on the temperature. The initial **IVc**:**Vc** ratio equal to 0.55:1 increases to 1.3:1 on heating for 15 min at 58°C in an NMR ampule.

An analogous ^1H NMR pattern was observed for tautomer couple **IVd** and **Vd**. The crystalline condensation product obtained from aldehyde **Ib** and α-hydroxyamino oxime acetate **IIIb** (isolated by precipitation from ethyl acetate) in DMSO-d_6 was isomer **IVd** (according to the ^1H NMR data), while in $CDCl_3$ a mixture of tautomers **IVd** and **Vd** at a ratio of 1:0.4 was identified. The UV spectrum of the product (KBr) contained two maxima at λ 285 and 238 nm (cf. [7]).

Secondary α-hydroxyamino oxime acetate **IIIc** reacted with aldehydes **Ia** and **Ib** quickly and quantitatively. The product isolated by crystallization from acetone was identified as cyclic isomer **Ve** (in DMSO-d_6 and D_2O). In $CDCl_3$ we observed an equilibrium mixture of tautomers **IVe** and **Ve** at a ratio of about 1:1. The product obtained in an analogous experiment by crystallization from ethyl acetate was also cyclic tautomer **Ve** in DMSO-d_6, while in $CDCl_3$ the ratio **IVe**:**Ve** was 1:3. The crystals showed an absorption maximum at λ 236 nm in the UV spectrum (KBr) in support of its cyclic structure **Ve** (cf. [7]).

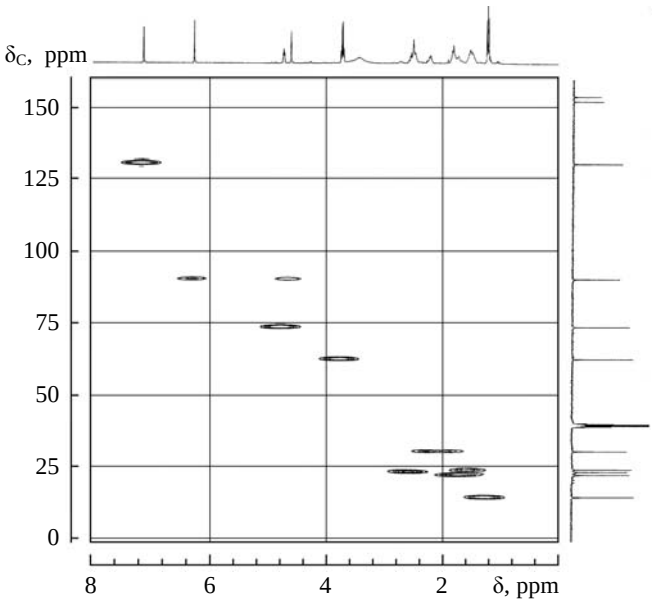


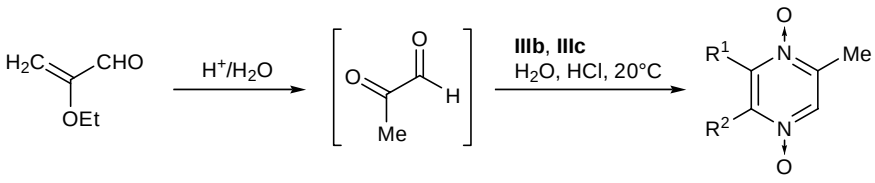
Fig. 2. Two-dimensional CH–CORR NMR spectrum of compound **IVc** in DMSO-d_6.

When the equilibrium mixture of **IVe** and **Ve** (~1:1; $CDCl_3$, 2 h at room temperature) was heated for 1 h at 58°C in an NMR ampule, the ratio **IVe**:**Ve** changed to 2.6:1. After cooling to 27°C and keeping for 1 h at that temperature, the ratio decreased to 1.8:1, and after 3 days, to 1.3:1. Thus, rise in temperature leads to reversible displacement of the tautomeric equilibrium toward the open-chain structure.

The equilibrium between isomers **IVf** and **Vf** in DMSO-d_6 is displaced toward the latter, while in going to $CDCl_3$ the tautomer ratio becomes 1:1. In the ^1H NMR spectra of condensation products obtained from aldehydes **Ia** and **Ib** and hydroxylamine **IIIc** we identified two cyclic structures which were *cis* and *trans* isomers with respect to substituents in positions *2* and *5* (**Ve**/**Ve'** and **Vf**/**Vf'**).

When the reactions of 2-alkoxypropenal **Ia** with secondary α-hydroxyamino oximes **IIIb** and **IIIc** were carried out in water in the presence of hydrochloric acid, the first stage was likely to be hydrolysis of aldehyde **Ia** to 2-oxopropionaldehyde. The subsequent reaction of hydroxylamine acetates **IIIb** and **IIIc** involved both carbonyl groups of the substrate to

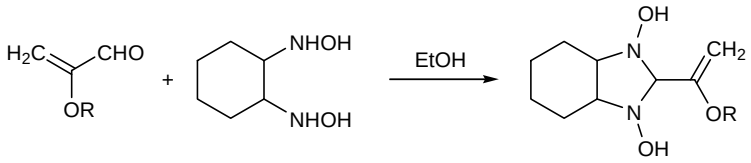
Scheme 3.



Ia **VIa**, **VIb**

IIIb, **VIb**, $R^1R^2 = (CH_2)_4$; **IIIc**, **VIa**, $R^1 = R^2 = Me$.

Scheme 4.



Ia **VIII** **IXa**, **IXb**

R = Et (**a**), Me (**b**).

produce substituted pyrazine 1,4-dioxides **VIa** and **VIb** (Scheme 3).

1,2-Bis(hydroxyamino)cyclohexane (**VIII**) reacted with 2-alkoxypropenals **Ia** and **Ib** in ethanol at 20°C. The reaction regioselectively occurred at the carbonyl group of the latter, and the products were 1,3-dihydroxyimidazolidines **IXa** and **IXb**, respectively (Scheme 4). The structure of 2-(1-ethoxyethenyl)-1,3-dihydroxyperhydrobenzimidazole (**IXa**) was proved by X-ray analysis (Fig. 3).

We can conclude that 2-alkoxypropenals react with α-hydroxyamino oximes in neutral medium at the aldehyde group to give isomeric acyclic and cyclic azomethine oxides, *N*-(2-hydroxyiminoalkyl)-*N*-(2-alkoxy-2-propenylidene)amine oxides and 1-hydroxy-2,5-dihydroimidazole 3-oxides, which occur in tautomeric equilibrium. The best conditions for the condensation of α-hydroxyamino oximes with 2-alkoxypropenals are equimolar reactant ratio, tetrahydrofuran as solvent, temperature 78°C, and reaction time 3 h. The reaction in aqueous medium in the presence of hydrochloric acid is accompanied by hydrolysis of the vinyl ether moiety in 2-alkoxypropenal with formation of 2-oxopropionaldehyde whose subsequent condensation at both carbonyl groups with α-hydroxyamino oxime leads to pyrazine 1,4-dioxides. This reaction demonstrates the possibility of using alkoxyacroleins as synthetic equivalents of 2-oxopropionaldehyde. Reactions of 2-alkoxypropenals with 1,2-bis(hydroxyimino)cyclohexane in alcohol at room temperature (24 h) afforded 2-(1-alkoxyethenyl)-1,3-dihydroxyperhydrobenzimidazoles.

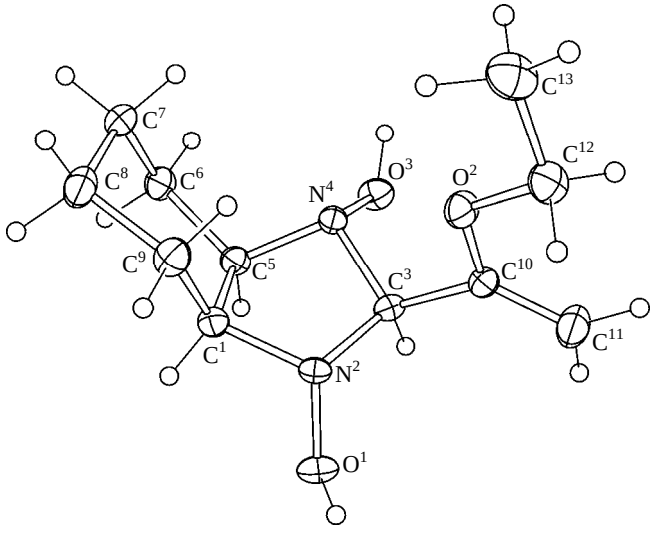


Fig. 3. Structure of the molecule of 2-(1-ethoxyethenyl)-1,3-dihydroxyperhydrobenzimidazole (**IXa**) according to the X-ray diffraction data.

EXPERIMENTAL

The ^1H and ^{13}C NMR spectra were recorded on a Bruker DPX-400 spectrometer at 400 MHz for ^1H and 100.6 MHz for ^{13}C using $CDCl_3$ or DMSO-d_6 as solvent and HMDS as internal reference. The IR spectra were measured on a Specord 75IR spectrometer. The UV spectra of crystalline samples (in KBr) were obtained on a Specord UV-Vis spectrophotometer. The X-ray diffraction data for compounds **Vb** and **IXa** were acquired on a Bruker P4 diffractometer (MoK_α irradiation, graphite monochromator, 2θ/θ scanning).

Crystallographic data for compound Vb. Crystal habit 0.90×0.20×0.10 mm. Triclinic crystal system with the following unit cell parameters: a = 11.139(2), b = 12.557(2), c = 16.572(3) Å; α = 90.037(15), β = 104.082(12), γ = 105.502(14)°; V = 2161.2(7) Å^3; space group $P\bar{1}$; Z = 8; $C_9H_{16}N_2O_3$, M 200.24; d_{calc} = 1.231 g/cm^3; μ = 0.093 mm^{-1}. Intensities of 5647 independent reflections were measured in the range 2θ < 45°. Correction for absorption was introduced empirically by Ψ curves (transmission 0.880–0.976). The structure was finally refined with respect to all F^2 to wR_2 = 0.1609, S = 1.036; 522 parameters were refined (R = 0.0545 for 4017 reflections with $F > 4\sigma$).

Crystallographic data for compound IXa. Crystal habit 0.30×0.22×0.18 mm. Monoclinic crystals with the following unit cell parameters: a = 10.473(3), b = 9.981(3), c = 12.144(4) Å; β = 108.16(2)°; V = 1206.3(6) Å^3; space group $P2_1/n$; Z = 4; $C_{11}H_{20}N_2O_3$, M 228.29; d_{calc} = 1.257 g/cm^3; μ = 0.091 mm^{-1}. Intensities of 2118 independent reflections were measured in the range 2θ < 50°. Correction for absorption was introduced empirically by Ψ curves (transmission 0.535–0.892). The structure was finally refined with respect to all F^2 to wR_2 = 0.1728, S = 1.046; number of refined parameters 226 (R = 0.0663 for 1195 reflections with $F > 4\sigma$).

The structures were solved by the direct method using SHELXS-97 program; the positions of hydrogen atoms in molecule **Vb** were determined from the difference synthesis. The structures were refined by the least-squares procedure in full-matrix anisotropic (isotropic for hydrogen atoms) approximation using SHELXL-97 program. The parameters of hydrogen atoms in molecule **Vb** were set from the geometry considerations. The coordinates and equivalent thermal parameters of non-hydrogen atoms in molecules **Vb** and **IXa** are available from the authors.

The independent part of a unit cell of 1-hydroxy-2-(1-methoxyethenyl)-4,5,5-trimethyl-2,5-dihydro-1*H*-imidazole 3-oxide (**Vb**) in crystal contains four crystallographically independent molecules; the structure of one of these is shown in Fig. 1. The bond lengths and bond angles in these molecules coincide with the error of determination (3σ). The dihydroimidazole ring in molecule **Vb** adopts an *envelope* conformation with the N^1 atom deviating from the double bond plane by 0.444(4)–0.484(4) Å. The double bond in the ethoxyethenyl group is eclipsed by the hydrogen atom on C^2, and the torsion angles $HC^2C^6C^7$ range from –7 to 10°. The most structurally related compounds deposited to the Cambridge Crystallographic Data Center [10] (except for structures with a π system at position *4*) are 4-dibromomethyl-1-hydroxy-2,2,5,5-tetramethyl-2,5-dihydro-1*H*-imidazole 3-oxide (**VI**) [11] and 8-hydroxy-1,4,5,7-tetramethyl-6,8-diazabicyclo[3.2.1]oct-6-ene 6-oxide (**VII**) [12]. The bond lengths in molecules **Vb**, **VI,** and **VII** are fairly similar. A slight shortening of the $N^3{=}C^4$ bond and slight lengthening of the $N^3{\rightarrow}O^3$ bond in **Vb** may be noted [1.291 and 1.308, 1.316 and 1.271, and 1.323 and 1.293 Å in structures **Vb**, **VI**, and **VII**, respectively; for compound **Vb**, average values for 4 independent molecules are given). Molecules **Vb** in crystal are linked along the *b* axis via hydrogen bonds to give infinite chains; the hydrogen bond parameters are as follows: O^1–H···O^{3B} [O–H 1.00(4), H···O 1.67(4), O···O 2.666(3) Å, ∠OHO 177(4)°], O^{1A}–H···O^{3C} [0.99(4), 1.68(4), 2.661(4), 172(4)], O^{1B}–H···O^3 [0.99(5), 1.70(5), 2.675(3), 170(4)], O^{1C}–H···O^{3A} [1.03(5), 1.65(5), 2.661(3), 165(4)].

The dihydroimidazole ring in molecule **IXa** has a conformation intermediate between *twist* and *envelope*. The hydroxy group O^1H occupies pseudoequatorial position, and the the O^3H group is pseudoaxial. Only one structure containing a 1,3-dihydroxyimidazolidine fragment was found in the Cambridge Crystal Structure Database [10], namely 1,3-dihydroxy-2-(4-*p*-nitrophenyl)-4,4,5,5-tetramethylimidazolidine [13]. Both the latter and compound **IXa** are characterized by almost similar conformations of the imidazolidine ring and orientations of the 2-substituents. The bond lengths in the 1,3-dihydroxyimidazolidine fragment are also similar. Molecules **IXa** in crystal are linked to centrosymmetric pairs by the O^1–H···N^2 hydrogen bonds [O–H 0.97(4), H···N 1.88(5), O···N 2.810(4) Å, ∠OHN 160(4)°]. These pairs are linked in turn about the symmetry centers to give infinite chains along the *a* axis through weaker hydrogen bonds O^3–H···N^4 [O–H 0.87(4), H···N 2.17(5), O···N 2.994(4) Å, ∠OHN 157(4)°].

2-(1-Ethoxyethenyl)-1-hydroxy-4,5,5-trimethyl-2,5-dihydro-1*H*-imidazole 3-oxide (Va). 2-Ethoxypropenal (**Ia**), 0.194 g (1.94 mmol), was added to a solution of 0.37 g (1.94 mmol) of hydroxylamine acetate **IIIa** in 4 ml of THF. The mixture was stirred for 1 h at room temperature, and the solvent was carefully distilled off under reduced pressure. The oily residue was dissolved in chloroform. On storage, a solid precipitated from the solution. The precipitate was compound **Va**. Yield 41%, mp 99–100°C. IR spectrum, ν, cm^{-1}: 3400, 3230, 2900, 1630, 1545, 1410, 1220, 1140, 1030, 950, 835, 680. ^{1}H NMR spectrum ($CDCl_3$), δ,

ppm: 1.29 t (3H, C**H**$_3$CH$_2$, J = 7.0 Hz), 1.32 s (3H, 5-CH$_3$), 1.35 s (3H, 5-CH$_3$), 2.02 d (3H, 4-CH$_3$, J = 1.8 Hz), 3.82 m (2H, OCH$_2$), 4.37 d (1H, CH$_2$=, J = 2.5 Hz), 4.47 d (1H, CH$_2$=, J = 2.5 Hz), 5.04 q (1H, 2-H, J = 1.8 Hz), 6.93 br.s (1H, NOH). ^{1}H NMR spectrum of minor tautomer **IVa** ($CDCl_3$), δ, ppm: 1.23 s (3H, CH$_3$), 1.63 s (3H, CH$_3$), 1.84 s (3H, CH$_3$), 3.5 m (2H, OCH$_2$), 4.66 s (1H, =CH$_2$), 6.4 (1H, =CH$_2$), 6.29 s (1H, N=CH); triplet signal of the ethyl CH$_3$ group could not be identified. ^{1}H NMR spectrum of **Va** in DMSO-d_6, δ, ppm: 1.18 s (3H, 5-CH$_3$), 1.21 t (3H, C**H**$_3$CH$_2$, J = 7.0 Hz), 1.22 s (3H, 5-CH$_3$), 1.89 d (3H, 4-CH$_3$, J = 1.6 Hz), 3.72 q (2H, CH$_2$O, ^3J = 7.0 Hz), 4.33 d (1H, CH$_2$=, J = 2.0 Hz), 4.35 d (1H, CH$_2$=, J = 2.0 Hz), 4.85 q (1H, 2-H, J = 1.6 Hz), 8.43 br.s (1H, NOH). Found, %: C 56.16; H 8.57; N 13.64. $C_{10}H_{18}N_2O_3$. Calculated, %: C 56.05; H 8.47; N 13.08.

1-Hydroxy-2-(1-methoxyethenyl)-4,5,5-trimethyl-2,5-dihydro-1*H*-imidazole 3-oxide (Vb). *a*. 2-Methoxypropenal (**Ib**), 0.166 g (1.94 mmol), was added to a solution of 0.37 g (1.94 mmol) of hydroxylamine acetate **IIIa** in 4 ml of THF, and the mixture was heated for 3 h 15 min under reflux. The mixture was cooled and dried over $MgSO_4$, and the solvent was carefully distilled off under reduced pressure. The oily residue was dissolved in diethyl ether, and the solid separated on storage was compound **Vb**. Yield 52%, mp 150°C. IR spectrum, ν, cm^{-1}: 3050, 2960, 2920, 2830, 1645, 1622, 1450, 1320, 1285, 1170, 1140, 1070, 1010, 915, 830. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 1.27 s and 1.33 s (3H each, 5-CH$_3$), 2.01 d (3H, 4-CH$_3$, J = 1.8 Hz), 3.63 s (3H, CH$_3$O), 4.42 d (1H, CH$_2$=, J = 2.7 Hz), 4.50 d (1H, CH$_2$=, J = 2.7 Hz), 5.01 q (1H, 2-H, J = 1.8 Hz), 6.95 br.s (1H, NOH). After storage, the ^{1}H NMR spectrum of a solution of **Vb** in $CDCl_3$ contained signals of open-chain isomer **IVb**, δ, ppm: 1.27 s (3H, CH$_3$), 1.62 s (3H, CH$_3$), 1.83 s (3H, CH$_3$), 3.57 s (3H, OCH$_3$), 4.67 s (1H, =CH$_2$), 6.39 s (1H, =CH$_2$), 6.92 s (1H, N=CH). ^{1}H NMR spectrum of **Vb** in DMSO-d_6, δ, ppm: 1.14 s and 1.20 s (3H each, 5-CH$_3$), 1.86 d (3H, 4-CH$_3$, J = 1.7 Hz), 3.50 s (3H, CH$_3$O), 4.36 d (1H, CH$_2$=, J = 2.0 Hz), 4.85 q (1H, 2-H, J = 1.7 Hz), 8.38 s (1H, 1-OH). ^{13}C NMR spectrum (JMOD, DMSO-d_6), δ_C, ppm: 9.55 (CH$_3$), 18.64 (CH$_3$), 24.89 (CH$_3$), 55.64 (OCH$_3$), 69.66 (C^5), 90.92 (=CH$_2$), 92.46 (C^2), 147.91 (C^4), 155.15 (O–C=CH$_2$). ^{1}H NMR spectrum (CD_3CN), δ, ppm: 1.20 s and 1.25 s (3H each, 5-CH$_3$), 1.90 d (3H, 4-CH$_3$, J = 1.8 Hz), 4.38 d and 4.41 d (2H, CH$_2$=, J = 2.7 Hz), 4.88 q (1H, 2-H, J = 1.8 Hz), 6.8 br.s (1H, 1-OH). UV spectrum: λ_{max} 232 nm. Found, %: C 54.40; H 8.68; N 14.10. $C_9H_{16}N_2O_3$. Calculated, %: C 54.00; H 8.00; N 14.00.

b. Likewise, compound **Vb** was synthesized from oxime **IIa** by heating for 4.5 h in boiling THF.

***N*-(2-Ethoxy-2-propenylidene)-*N*-(2-hydroxyiminocyclohexyl)amine oxide (IVc).** 2-Ethoxypropenal (**Ia**), 0.29 g (2.9 mmol), was added to a solution of 0.59 g (2.9 mmol) of hydroxylamine acetate **IIIb** in 3 ml of THF. The mixture was heated for 15 min under reflux and cooled, and the solvent was carefully distilled off under reduced pressure. According to the ^{1}H NMR data, the mixture contained no initial compounds. After addition of diethyl ether, a solid precipitated and was filtered off. The ^{1}H NMR spectrum of the product dissolved in DMSO-d_6 corresponded to compound **IVc**. Yield 26%, mp 146°C. UV spectrum (ethanol): λ_{max} 264 nm (log ε 4.86). IR spectrum, ν, cm^{-1}: 3400, 3250, 2960, 2930, 2850, 1590, 1570, 1450, 1430, 1260, 1170, 1070, 970, 950, 820. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 1.28 t (3H, C**H**$_3$CH$_2$, J = 7.0 Hz), 1.58 m (2H, CH$_2$), 1.63 m (1H, CH$_2$), 1.92 m (2H, CH$_2$), 2.58 m (2H, CH$_2$), 2.76 m (1H, CH$_2$), 3.78 q (2H, OCH$_2$, J = 7.0 Hz), 4.50 t (1H, NCH, J = 5.4 Hz), 4.68 d.d (1H, CH$_2$=, J = 1.6, 1.8 Hz), 6.40 d (1H, CH$_2$=, J = 1.8 Hz), 6.92 d (1H, N=CH, J = 1.6 Hz). The solution in $CDCl_3$ contained a mixture of isomers **IVc** and **Vc** at a ratio of 2:1. ^{1}H NMR spectrum of **Vc** ($CDCl_3$), δ, ppm: 1.39 t (3H, C**H**$_3$CH$_2$, J = 7.0 Hz), 1.90 m (3H, CH$_2$), 2.07 m (2H, CH$_2$), 2.56 m (2H, CH$_2$), 2.80 m (1H, CH$_2$), 3.75 q (2H, OCH$_2$, J = 7.0 Hz), 3.98 m (1H, CH$_2$C**H**N), 4.38 d (1H, CH$_2$=, J = 2.5 Hz), 4.48 d (1H, CH$_2$=, J = 2.5 Hz), 5.17 s (1H, 2-H). ^{1}H NMR spectrum of **IVc** in DMSO-d_6, δ, ppm: 1.22 t (3H, C**H**$_3$CH$_2$, J = 7.0 Hz), 1.53 m (3H, CH$_2$), 1.65 m (1H, CH$_2$), 1.80 m (1H, 6-H), 2.2 m (1H, 6-H), 2.50 m (2H, CH$_2$), 3.71 q (2H, OCH$_2$, J = 7.0 Hz), 4.60 s (1H, =CH$_2$), 4.72 t (1H, CHN, J = 5.5 Hz), 6.26 s (1H, =CH$_2$), 7.14 s (1H, N=CH), 10.98 (1H, =NOH). ^{13}C NMR spectrum (JMOD, DMSO-d_6), δ_C, ppm: 14.32 (CH$_3$), 21.90 (CH$_2$), 22.93 (CH$_2$), 23.76 (CH$_2$), 30.14 (CH$_2$), 62.14 (OCH$_2$), 73.28 (NCHC), 90.22 (CH$_2$=), 129.73 (N=CH), 151.88 (O–C= or C=N), 153.57 (C=N or O–C=). Found, %: C 59.48; H 8.02; N 12.71. $C_{11}H_{18}N_2O_3$. Calculated, %: C 58.39; H 8.02; N 12.38.

***N*-(2-Hydroxyiminocyclohexyl)-*N*-(2-methoxy-2-propenylidene)amine oxide (IVd).** 2-Methoxypropenal (**Ib**), 0.25 g (2.91 mmol), was added to a solution of 0.419 g (2.91 mmol) of oxime **IIb** in 3 ml of THF. After 4 h, the solvent was carefully distilled off under reduced pressure. Addition of ethyl acetate to the oily

residue resulted in separation of a solid product which was filtered off. It was dissolved in DMSO-d_6, and the ^{1}H NMR spectrum of the solution corresponded to compound **IVd**. Yield 28%, mp 130°C. ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 1.50 m (3H, CH_2), 1.80 m (2H, CH_2), 2.19 m (1H, CH_2), 2.55 m (2H, CH_2), 3.49 s (3H, OCH_3), 4.62 s (1H, $=CH_2$), 4.74 t (1H, CHN, J = 5.5 Hz), 6.26 s (1H, $=CH_2$), 7.17 s (1H, N=CH), 11.00 (1H, =NOH). A solution of the product in $CDCl_3$ contained a mixture of isomers **IVd** and **Vd** at a ratio of 1:1. ^{1}H NMR spectrum of **IVd** ($CDCl_3$), δ, ppm: 1.58 m (2H, CH_2), 1.74 m (1H, CH_2), 1.90 m (2H, CH_2), 2.56 m (2H, CH_2), 2.76 m (1H, CH_2), 3.56 s (3H, OCH_3), 4.49 t (1H, NCH, J = 5.0 Hz), 4.69 s (1H, CH_2=), 6.39 s (1H, $=CH_2$), 6.92 s (1H, N=CH). ^{1}H NMR spectrum of **Vd** ($CDCl_3$), δ, ppm: 1.37 m (2H, CH_2), 1.55 m (1H, CH_2), 2.12 m (2H, CH_2), 2.54 m (2H, CH_2), 2.78 m (1H, CH_2), 3.59 s (3H, OCH_3), 4.16 m (1H, $CH_2C\mathbf{H}N$), 4.37 d (1H, $=CH_2$, J = 3.0 Hz), 4.43 d (1H, $=CH_2$, J = 3.0 Hz), 5.24 s (1H, 2-H). Found, %: C 57.21; H 7.41; N 13.11. $C_{10}H_{16}N_2O_3$. Calculated, %: C 56.59; H 7.60; N 13.20.

The reaction of 2-methoxypropenal (**Ib**) with oxime **IIb** was complete (100% conversion) on heating for 15 min at 80°C. According to the ^{1}H NMR data, the product in DMSO-d_6 was a mixture of compounds **IVd** and **Vd** at a ratio of 1:0.4, and in $CDCl_3$, at a ratio of 1:1. The UV spectrum of the crystalline product isolated by precipitation from ethyl acetate contained absorption maxima at λ 238 and 285 nm.

***N*-(2-Hydroxyimino-1-methylpropyl)-*N*-(2-ethoxy-2-propenylidene)amine oxide (IVe).** 2-Ethoxypropenal (**Ia**), 1.94 g (1.94 mmol), was added to a solution of 0.346 g (1.94 mmol) of hydroxylamine acetate **IIIc** in 4 ml of THF, and the mixture was heated for 25 min at 80°C. When the reaction was complete, the solvent was carefully distilled off under reduced pressure. The product isolated by crystallization from acetone and dissolved in $CDCl_3$ showed in the ^{1}H NMR spectrum signals from isomers **IVe** and **Ve** at a ratio of 1.2:1. In DMSO-d_6, only tautomer **Ve** was present. Conversion 100%. mp 139°C. ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 1.25 t (3H, $C\mathbf{H}_3CH_2$, J = 7.0 Hz), 1.60 d (3H, $C\mathbf{H}_3CH$, J = 6.9 Hz), 1.95 s (3H, $C\mathbf{H}_3C$=NOH), 3.82 m (2H, OCH_2, J = 7.0 Hz), 4.58 q (1H, $C\mathbf{H}CH_3$, J = 6.9 Hz), 4.68 s (1H, CH_2=), 6.37 s (1H, CH_2=), 6.96 s (1H, N=CH). The ^{1}H NMR spectrum in $CDCl_3$ also contained signals from cyclic tautomer **Ve**.

2-(1-Ethoxyethenyl)-1-hydroxy-4,5-dimethyl-2,5-dihydro-1*H*-imidazole 3-oxide (Ve). ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 1.25 t (3H, $C\mathbf{H}_3CH_2$, J = 7.0 Hz), 1.34 d (3H, 5-CH_3, J = 7.0 Hz), 2.03 s (3H, CH_3C=N), 3.79 m (2H, OCH_2), 4.11 q (1H, 5-H, J = 7.0 Hz), 4.35 d (1H, CH_2=, J = 2.5 Hz), 4.44 d (1H, CH_2=, J = 2.5 Hz), 5.23 (1H, 2-H), 6.79 s (1H, NOH). The ^{1}H NMR spectrum of the second cyclic isomer (**Ve'**) in $CDCl_3$ differed from the spectrum of **Ve** by the position of the $=CH_2$ signals (δ 4.32 i 4.38 ppm) and the signal from 2-H (δ 5.24 ppm). The isomer ratio **Ve**:**Ve'** varied from 1:1 to 1:2, depending on the conditions. ^{1}H NMR spectrum in DMSO-d_6, δ, ppm: 1.18 t (3H, $C\mathbf{H}_3CH_2$, J = 7.0 Hz), 1.23 d (3H, 5-CH_3, J = 7.0 Hz), 1.88 s (3H, 4-CH_3), 3.70 q (2H, OCH_2, J = 7.0 Hz), 3.98 q (1H, 5-H, J = 7.0 Hz), 4.30 d (1H, CH_2=, J = 1.8 Hz), 4.34 d (1H, CH_2=, J = 1.8 Hz), 4.88 s (1H, 2-H), 11.37 s (1H, =NOH). ^{13}C NMR spectrum (DMSO-d_6), δ_C, ppm: 9.35 (CH_3), 9.64 (CH_3), 14.07 (CH_3), 16.90 (CH_3), 62,99 (OCH_2), 70.35 (C^5), 89.38 ($=CH_2$), 96.49 (C^2), 141.36 (C^4), 155.05 (=**C**OEt). ^{1}H NMR spectrum (D_2O), δ, ppm: 1.09 t (3H, $C\mathbf{H}_3CH_2$, J = 7.0 Hz), 1.23 d (3H, 5-CH_3, J = 7.1 Hz), 1.92 s (3H, 4-CH_3), 3.67 m (2H, OCH_2), 4.11 q (1H, 5-H, J = 7.1 Hz), 4.36 d (1H, CH_2=, J = 2.5 Hz), 4.37 d (1H, CH_2=, J = 2.5 Hz), 5.09 s (1H, 2-H). Found, %: C 53.81; H 8.66. $C_9H_{16}N_2O_3$. Calculated, %: C 53.98; H 8.06.

***N*-(2-Hydroxyimino-1-methylpropyl)-*N*-(2-methoxy-2-propenylidene)amine oxide (IVf).** 2-Methoxypropenal (**Ib**), 0.22 g (2.56 mmol), was added to a solution of 0.45 g (2.53 mmol) of hydroxylamine acetate **IIIc** in 4 ml of THF, and the mixture was stirred for 1 h at room temperature. When the reaction was complete, the solvent was carefully distilled off under reduced pressure. According to the ^{1}H NMR data, the product dissolved in $CDCl_3$ was a mixture of tautomers **IVf** and **Vf** at a ratio of 1.1:1, and tautomer **Vf** was a mixture with its isomer **Vf'**. Conversion 100%, mp 150°C (from ethyl acetate). ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 1.61 d (3H, $C\mathbf{H}_3CH$, J = 6.8 Hz), 1.95 s (3H, CH_3), 3.57 s (1H, OCH_3), 4.60 q (1H, $CH_3C\mathbf{H}N$, J = 6.8 Hz), 4.71 d.d (1H, $=CH_2$, ^4J = 2.1, ^2J = 1.9 Hz), 6.35 d (1H, $=CH_2$, ^2J = 1.9 Hz), 6.98 d (1H, CH=N, ^4J = 2.1 Hz). Found, %: C 51.85 H 7.80; N 14.97. $C_8H_{14}N_2O_3$. Calculated, %: C 51.60; H 7.50; N 15.00.

1-Hydroxy-2-(1-methoxyethenyl)-4,5-dimethyl-2,5-dihydro-1*H*-imidazole 3-oxide (Vf). ^{1}H NMR spectrum ($CDCl_3$), δ, ppm: 1.36 d (3H, 5-CH_3, ^3J = 7.1 Hz), 2.08 s (3H, 4-CH_3), 3.57 s (3H, OCH_3), 4.13 q (1H, 5-H, ^3J = 7.1 Hz), 4.41 d (1H, CH_2=, J = 2.9 Hz), 4.50 d (1H, CH_2=, J = 2.5 Hz), 5.23 s (1H, 2-H). ^{1}H NMR spectrum of isomer **Vf'** ($CDCl_3$), δ, ppm: 1.39 d (3H, 5-CH_3, ^3J = 7.2 Hz), 2.08 s (3H, 4-CH_3),

3.57 s (3H, OCH_3), 4.13 q (1H, 5-H, ^3J = 7.2 Hz), 4.36 d (1H, CH_2=, J = 2.9 Hz), 4.43 d (1H, CH_2=, J = 2.5 Hz), 5.33 s (1H, 2-H). The ratio of isomers **Vf** and **Vf'** was 1:1. Found, %: C 51.00; H 7.67; N 14.71. $C_8H_{14}N_2O_3$. Calculated, %: C 51.60; H 7.50; N 15.00.

Temperature effect on the tautomeric equilibrium. *a*. A mixture of tautomers **Vb** and **IVb** at a ratio of 6.3:1 (in $CDCl_3$) was heated in an NMR ampule for 1 h at 58°C; as a result, the ratio decreased to 2.2:1. The solution was cooled to 27°C and was left to stand for 1 h at that temperature. The ratio of tautomers **Vb** and **IVb** increased to 3.2:1; after 25 h, it was 4.4:1.

b. A mixture of tautomers **IVc** and **Vc** at a ratio of 0.55:1 (in $CDCl_3$) was heated in an NMR ampule for 15 min at 58°C. The isomer ratio changed to 1.3:1. After cooling to 26°C, the ratio remained unchanged. Presumably, the time was insufficient for equilibration at that temperature.

c. An equimolar mixture of tautomers **IVe** and **Ve** (2 h after dissolution in $CDCl_3$) was heated in an NMR ampule for 1 h at 58°C. The ratio **IVe**:**Ve** changed to 2.6:1. The ampule was cooled to 27°C and was left to stand for 1 h at that temperature; the ratio changed to 1.8:1, and after 3 days, to 1.3:1.

2,3,5-Trimethylpyrazine 1,4-dioxide (VIa). 2-Ethoxypropenal (**Ia**), 0.291 g (2.91 mmol), was added to a solution of 0.51 g (2.91 mmol) of hydroxylamine acetate **IIIc** in 16 ml of distilled water containing 0.5 ml of 33% hydrochloric acid. The mixture was stirred for 24 h at room temperature and extracted with chloroform, the extract was dried over $MgSO_4$, and the solvent was distilled off under reduced pressure. Yield 10%, mp 160°C; published data [14]: mp 136–137°C. ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 2.43 (3H, CH_3), 2.52 (3H, CH_3), 2.55 (3H, CH_3), 8.17 (1H, 6-H). ^{13}C NMR spectrum (DMSO-d_6), δ_C, ppm: 13.83, 14.34, 15.06, 132.72, 143.21, 143.57, 145.06.

2-Methyl-5,6,7,8-tetrahydroquinoxaline 1,4-dioxide (VIb). 2-Ethoxypropenal (**Ia**), 0.291 g (2.91 mmol), was added to a solution of 0.59 g (2.91 mmol) of hydroxylamine acetate **IIIb** in 16 ml of distilled water containing 0.5 ml of 33% hydrochloric acid. The mixture was stirred for 30 h at room temperature and extracted with chloroform, the extract was dried over $MgSO_4$, and the solvent was distilled off under reduced pressure. Yield 10%, mp 187°C; published data [15]: mp 180–181°C. IR spectrum, ν, cm^{-1}: 3400, 2950, 1420, 1350, 1200, 1090, 970, 860, 720. ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 1.72 m (4H, β-CH_2), 2.25 s (3H, CH_3), 2.68 m (4H, α-CH_2), 8.40 s (1H, 3-H).

2-(1-Ethoxyethenyl)-1,3-dihydroxyperhydrobenzimidazole (IXa). 2-Ethoxypropenal (**Ia**), 0.3 g (3.0 mmol), was added to a solution of 0.3 g (2.05 mmol) of bis-hydroxylamine **VIII** in 4 ml of ethanol, and the mixture was stirred for 24 h at room temperature. Compound **IXa** crystallized on careful evaporation of the reaction mixture. Yield 52%, mp 192–194°C. IR spectrum, ν, cm^{-1}: 3400, 3250, 2920, 1590, 1570, 1450, 1255, 1160, 1070, 970, 940, 820. ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 1.22 t (3H, C**H**$_3$CH$_2$, ^3J = 6.77 Hz), 1.20 m and 1.65 m (4H, 4-CH_2, 7-CH_2), 1.47 m (4H, 5-CH_2, 6-CH_2), 3.12 m (2H, 3a-H, 7a-H), 3.68 q (2H, OCH_2, ^3J = 6.77 Hz), 3.93 and 4.01 (2H, CH_2=), 4.36 s (1H, 2-H), 8.31 s (2H, OH). ^{13}C NMR spectrum (JMOD, DMSO-d_6), δ_C, ppm: 14.23 (**C**H$_3$CH$_2$), 21.81 (C^5, C^6), 25.85 (C^4, C^7), 62.3 (C^{3a}, C^{7a}), 64.72 (OCH_2), 83.53 (C^2), 95.02 (CH_2=), 160.71 (OC=). Found, %: C 57.92; H 8.82; N 12.17. $C_{11}H_{20}N_2O_3$. Calculated, %: C 57.87; H 8.83; N 12.27.

1,3-Dihydroxy-2-(1-methoxyethenyl)perhydrobenzimidazole (IXb) was synthesized in a similar way from 2-methoxypropenal (**Ib**). Yield 40%, mp 201°C. IR spectrum, ν, cm^{-1}: 3380, 3260, 2920, 2840, 1630, 1445, 1315, 1195, 1110, 1070, 885, 825. ^{1}H NMR spectrum (DMSO-d_6), δ, ppm: 1.20 m and 1.61 m (4H, 4-CH_2, 7-CH_2), 1.48 m (4H, 5-CH_2, 6-CH_2), 3.12 br.s (2H, 3a-H, 7a-H), 3.48 s (3H, CH_3O), 3.95 and 4.04 (2H, CH_2=), 4.40 s (1H, 2-H), 8.33 s (2H, OH). ^{13}C NMR spectrum (JMOD, DMSO-d_6), δ_C, ppm: 22.07 (C^5, C^6), 26.26 (C^4, C^7), 54.95 (OCH_3), 65.04 (C^{3a}, C^{7a}), 83.33 (C^2), 95.05 (CH_2=), 162.04 (O–C=). Found, %: C 56.00; H 8.46; N 12.78. $C_{10}H_{18}N_2O_3$. Calculated, %: C 56.06; H 8.47; N 13.07.

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