SYNTHESIS AND PROPERTIES OF SOME DERIVATIVES OF 3,4-DIHYDRO-3-OXO-2-QUINOALINYLACETIC ACID*

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The paper describes opening of isoxazole ring with 1,2-diaminobenzene leading to 2-cyanomethylene-3-oxo-1,2,3,4-tetrahydroquinoxaline and preparation of its derivatives. Synthesis has been studied of substituted 3,4-dihydro-3-oxo-quinoxaline-oxides, and tautomers of 1-oxides of 2-ethoxycarbonylmethylquinoxalines have been obtained.

Our previous communications described preparation and properties of some 2-alkoxycarbonylmethylene- and 2-cyanomethylene-3-oxo-1,2,3,4-tetrahydroquinoxalines. The present paper gives further synthetic procedures and preparation of 1-oxides of the mentioned esters.

A new way of synthesis of the cyano derivatives was found which makes use of the opening of isoxazole ring by action of nucleophiles[1] involving splitting of the bond between the heteroatoms. Reaction of methyl isoxazole-5-carboxylate (*I*) with 1,2-diaminobenzene in boiling dimethyl sulphoxide gave 2-cyanomethylene-3-oxo-1,2,3,4-tetrahydroquinoxaline (*II*) identical with the product of reaction of ethyl 3-cyano-2-oxopropanoate with 1,2-diaminobenzene[2]. Our procedure gives a very pure product.

The nitrile *II* was alkylated with methyl iodide in acetone in the presence of potassium carbonate. Whereas the reaction with equimolar amount of methyl iodide gives 4-methyl derivative of the nitrile *II* (ref.[3]), excess of the reagent gave another product which was identified by elemental analysis and spectral data as 2-(1-cyanoethyl)-3,4-dihydro-4-methyl-3-oxoquinoxaline (*III*). The same product was obtained by analogous alkylation of 2-(1-cyanoethyl)-3,4-dihydro-3-oxoquinoxaline (*IV*) (ref.[4]). The ketimine tautomeric structure of compound *III* was proved by its ^{1}H NMR spectrum measured in chloroform; in dimethyl sulphoxide solution the both tautomers are present in the ratio 1 : 1. In contrast to the nitrile *II*, 2-ethoxycarbonylmethylene-3-oxo-1,2,3,4-tetrahydroquinoxaline (*V*) only gives the 4-methyl derivative[5], whereas the ester analogue of nitrile *III* is formed by alkylation of ester *V* in the

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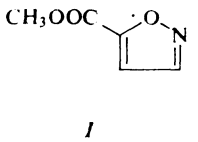
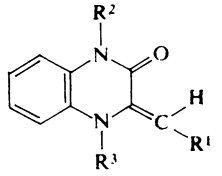
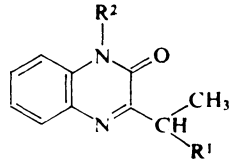
presence of silver oxide[4]. The alkylation at β-carbon atom of enamine group of the nitrile *II* indicates a higher activating effect of cyano group as compared with ester group. This fact also agrees with the result of our attempt at a long-term alkylation of the nitrile *III* from which we isolated 2-(1-cyano-1-methylethyl)-3,4-dihydro--4-methyl-3-oxoquinoxaline (*VI*).

From N,N'-dimethyl-1,2-diaminobenzene and ethyl 3-cyano-2-oxopropanoate we obtained 2-cyanoethylene-1,4-dimethyl-3-oxo-1,2,3,4-tetrahydroquinoxaline (*VII*) with the fixed enamine structure. Analogous reaction of the same diamine with diethyl 2-butine-1,4-dioate gave 2-ethoxycarbonylmethylene-1,4-dimethyl-3-oxo--1,2,3,4-tetrahydroquinoxaline (*VIII*) in a higher yield than that resulting from the analogous reaction of diethyl 2-oxo-1,4-butanedioate. On the contrary, the attempts at preparation of 2-ethoxycarbonylmethylene-1,4-dimethyl-3-oxopiperazine (*IX*) by reactions of N,N'-dimethyl-1,2-ethanediamine with both diethyl 2-butine-1,4--dioate and diethyl 2-oxo-1,4-butanedioate failed, although each of the two esters reacts with 1,2-ethanediamine to give the corresponding piperazine derivative[4]. In the former case the product was identified by elemental analysis, mass and IR spectra as tetraethyl 2,2'-[ethylene-bis(methylamino)]di-*trans*-2-butanedioate (*X*) The mass spectrum shows, besides the weak molecular ion, mainly the fragment *m/z* 214 formed by the *β* splitting of the tertiary amine and the ions formed by fragmentation of the ethoxycarbonyl groups. The IR spectrum shows, besides two *ν*(C=O) bands at 1 726 and 1 692 cm^{-1}, a very intensive band at 1 590 cm^{-1} belonging to the vibration *ν*(C=O) of the enaminoester.



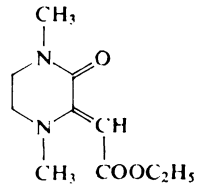
I



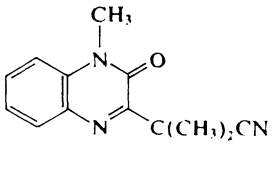


II, $R^1 = CN$, $R^2 = R^3 = H$
III, $R^1 = CN$, $R^2 = CH_3$
IV, $R^1 = CN$, $R^2 = H$
V, $R^1 = CO_2C_2H_5$, $R^2 = R^3 = H$
VII, $R^1 = CN$, $R^2 = R^3 = CH_3$
VIII, $R^1 = CO_2C_2H_5$, $R^2 = R^3 = CH_3$
XI, $R^1 = CO_2C_2H_5$, $R^2 = CH_3$, $R^3 = H$
XIII, $R^1 = CO_2CH_3$, $R^2 = CH_3$, $R^3 = H$
XIV, $R^1 = COCH_3$, $R^2 = CH_3$, $R^3 = H$
XV, $R^1 = CO_2C_2H_5$, $R^2 = H$

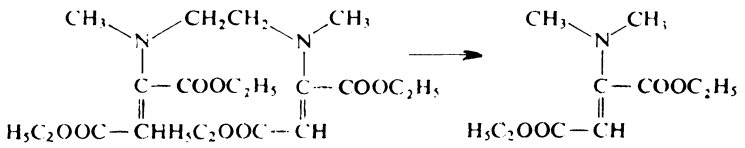
For studies of tautomerism and potential biologic activity we tried to prepare 1-oxide derivative of ester V and its 4-methyl derivative *XI*. Out of general procedures we chose, first of all, the direct oxidation of 1-nitrogen atom. The ester *V* reacted with peracetic acid to give the C-oxidation product, 2,3-dioxo-1,2,3,4-tetrahydroquinoxaline (*XII*). The oxidative degradation of 2-substituent was also observed with 2-



IX



VI



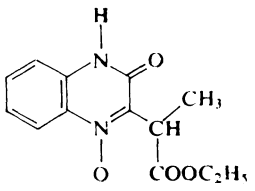
X *m/z* 428 (6 %) *m/z* 214 (100 %)

-methoxycarbonylmethylene-4-methyl-3-oxo-1,2,3,4-tetrahydroquinoxaline (*XIII*) (ref.[6]) and 2-acetylmethylene-4-methyl-3-oxo-1,2,3,4-tetrahydroquinoxaline (*XIV*) (ref.[3]). The dioxo derivative *XII* was also obtained by oxidation of ethyl 2-(3,4-dihydro-3-oxo-2-quinoxalinyl)propanoate (*XV*). With respect to the oxidation of alkyl 2- and 4-pyridyl acetates giving 2- and 4-carboxypyridine 1-oxides, respectively[7], we presume that the reactions of quinoxalines *V*, *XIV*, and *XV* go through the 2-carboxy--3,4-dihydro-3-oxo derivatives which give[8] the C-oxidation product *XII*. In further experiments we chose the more selective N-oxidation with 3-chloroperbenzoic acid in aprotic solvent. A small amount of 2-(1-ethoxycarbonylethyl)-3,4-dihydro-3-oxoquinoxaline 1-oxide (*XVI*) was isolated from the reaction mixture after oxidation of ester *XV* with ketimine tautomeric structure. The N-oxyketimine structure of the solid compound is indicated by the band ν(C=O) at 1 748 cm^{-1} and the signals 4·63 and 1·55 ppm in ^{1}H NMR spectrum of the compound in chloroform. A different spectral behaviour was shown by the oxidation product from the isomeric ester *XI* which exists in the enamine form. The proton signals of =CH groups at 5·36 ppm and NOH 24°C) at 3·62 ppm indicate the N-hydroxyenamine form, and we suppose 2-ethoxycarbonylmethylene-1-hydroxy-4-methyl-3-oxo-1,2,3,4-tetrahydroquinoxaline (*XVII*) to be the oxidation product. So far, the heterocyclic N-hydroxide tautomers have been found[9,10] only with compounds lacking conjugated carbonyl group. The N-hydroxyenamine form is also assigned the bands ν(OH) 3 380 cm^{-1} and ν(C=O) 1 760 and 1 660 cm^{-1} in the IR spectrum of ester *XVII*. The wave number of the band at 1 760 cm^{-1} has higher value (by about 100 cm^{-1}) than ν(C=O) of ester group of the enamino esters *V* and *XIII*, which is obviously due to the absence of intramolecular hydrogen bond and electronegativity of N-hydroxy group. These conclusions also agree with the found wave numbers of the bands ν(C=O) 1 760, 1 775 (shoulder) and 1 651 cm^{-1} for carbonyl group of ethoxycarbonyl and N-acetate group and amidic

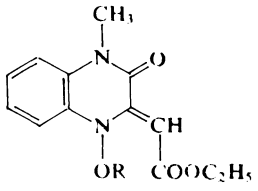
carbonyl group at 3-position of the 1-acetoxy-2-ethoxycarbonyl-4-methyl-3-oxo-1,2,3,4-tetrahydroquinoxalines (*XVIII*) prepared from ester *XVII* by action of acetic anhydride. Our oxidation experiments with the esters *XI* and *XV* also show that application of 3-chloroperbenzoic acid does not eliminate fully the C-oxidation of heterocyclic ring, however, the proportion of dioxo derivative *XII* in the oxidation products is distinctly lower.

We also tried to prepare the methyl ester analogue of the derivative *XVII* by condensation of 1,3-dimethyl-2-oxo-1,2-dihydroquinoxaline 1-oxide (*XIX*) with dimethyl carbonate. However, the reaction did not proceed even with application of sodium hydride as the base, although at the same conditions 1,3-dimethyl-2-oxo--1,2-dihydroquinoxaline gave smoothly the ester *XIII*. The reactivity lowering of 2-methyl group in 1-oxides was described also for the reaction with formaldehyde[11].

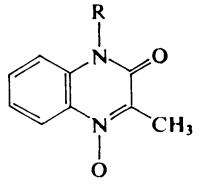
The starting N-oxide *XIX* is accessible by a multistep synthesis[12], and we tried to prepare it by a new way. Quinoxaline-1,4-dioxide is known[13] to undergo photochemical isomerization to give 3,4-dihydro-3-oxoquinoxaline 1-oxide which, on action of hydrogen chloride in aqueous methanol, can be isomerized further[14] to the dioxo derivative *XII* in the same way as the starting quinoxaline 1,4-dioxide. We tried to apply the two isomerization reactions to the preparation of the N-oxide *XIX*, starting from the 2-methylquinoxaline 1,4-dioxide which can be prepared easily from benzofurazane 1-oxide. Photochemical reaction in aqueous medium gave 2-methyl-3-oxo--3,4-dihydroquinoxaline 1-oxide (*XX*) whose methylation to the dimethyl derivative *XIX* is known[12]. On the contrary, the isomerization of 2-methylquinoxaline-1,4-dioxide with methanolic hydrogen chloride did not take place. The N-oxide *XIX* was also prepared by oxidation of 1,3-dimethyl-2-oxo-1,2-dihydroquinoxaline with 3--chloroperbenzoic acid.



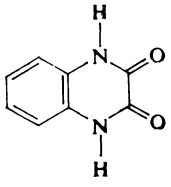
XVI



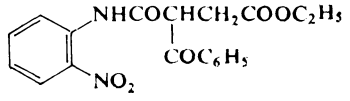
XVII, R = H
XVIII, R = $COCH_3$



XIX, R = CH_3
XX, R = H



XII



XXI

We also tried to prepare the N-oxide derivative *XVII* by cyclization. Benzoylaceto -2-nitroanilide was alkylated with ethyl iodoacetate to give ethyl 3-benzoyl-N-(2--nitrophenyl)succinamate (*XXI*) which, however, did not undergo cyclization by action of sodium ethoxide in spite of the fact that the reaction is considered general for various acylaceto-2-nitroanilides[15].

EXPERIMENTAL

The melting points are corrected. The IR spectra were measured in Nujol with a Spectromom 2000 apparatus. The ^{1}H NMR spectra were measured in deuteriochloroform and ($^2H_6$)-dimethyl sulphoxide with a Tesla BS 487 B apparatus at 80 MHz with hexamethyldisiloxane as internal standard. The mass spectra were measured with an MX 1303 apparatus (U.S.S.R.) at the ionization energy of 50 eV. 3-Chloroperbenzoic acid was analyzed iodometrically.

2-Cyanomethylene-3-oxo-1,2,3,4-tetrahydroquinoxaline (*II*)

Solution of 0·13 g (0·001 mol) methyl 5-isoxazolcarboxylate[16] and 0·11 g (0·001 mol) 1,2-diaminobenzene in 1 ml dimethyl sulphoxide was boiled 5 min (the reaction does not take place in the lower-boiling dimethylformamide). The product separated on addition of 2 ml methanol and cooling was collected and washed with methanol. Yield 0·17 g (70%) yellow crystals, m.p. 296–300°C (decomp.); the authentic substance[2] has identical melting point and IR spectrum.

2-(1-Cyanoethyl)-3,4-dihydro-4-methyl-3-oxo-quinoxaline (*III*)

a) 5 g Cyano derivative *II* (0·03 mol), 7 g freshly annealed potassium carbonate, 100 ml 2-butanone, and 10 ml (0·07 mol) methyl iodide were mixed and boiled 4 h. The separated salts were collected by suction, washed with acetone, and the filtrate was evaporated. The evaporation residue was crystallized three times from ethanol (charcoal) to give 2 g product (38%), m.p. 147–149°C. For $C_{12}H_{11}N_3O$ (213·2) calculated: 67·59% C, 5·20% H, 19·71% N; found: 67·33% C, 5·43% H, 19·65% N. ^{1}H NMR spectrum in deuteriochloroform: 1·71 ppm $CHCH_3$ (d), 4·56 ppm $CHCH_3$ (q), 3·74 ppm N—CH_3 (s); in ($^2H_6$)-dimethyl sulphoxide: 1·60 ppm $CHCH_3$ (d), 4·64 ppm $CHCH_3$ (q), 1·88 ppm = CCH_3 (s), 3·33 and 3·63 ppm N—CH_3 (s).

b) The same product was obtained in the yield of 59% by analogous alkylation of compound *IV*.

2-(1-Cyano-1-methylethyl)-3,4-dihydro-4-methyl-3-oxoquinoxaline (*VI*)

0·3 g (0·0016 mol) 2-cyanomethyl-4-methyl-3-oxo-1,2,3,4-tetrahydroquinoxaline, 30 ml acetone, 5 ml methyl iodide, and 1 g freshly annealed powdered potassium carbonate was boiled with exclusion of atmospheric moisture 36 h. The separated salts were hot filtered and washed with acetone. The combined filtrates were evaporated under reduced pressure, and the residue was boiled with 70 ml water. The solid separated on cooling was collected by suction and washed with water. For $C_{13}H_{13}N_3O$ (227·2) calculated: 68·70% C, 5·77% H, 18·49% N; found: 68·89% C, 5·71% H, 18·86% N. After crystallization from 60% ethanol 0·25 g (73%) needles, m.p. 142 to 144°C. ^{1}H NMR spectrum: C—CH_3 1·87 ppm, N—CH_3 3·74 ppm.

2-Cyanomethylene-1,4-dimethyl-3-oxo-1,2,3,4-tetrahydroquinoxaline (*VII*)

Solution of 1·4 g (0·01 mol) N,N'-dimethyl-1,2-diaminobenzene[17] and 1·9 g (0·011 mol) potassium salt of ethyl 3-cyano-2-oxopropanoate[18] in 10 ml diluted (1 : 1) acetic acid was boiled 5 min.

The product precipitated on 1 h standing was filtered, washed with water, and recrystallized from ethanol (charcoal). Yield 0·5 g (22%) yellowish needles, m.p. 216–218°C. For $C_{12}H_{11}N_3O$ (213·2) calculated: 67·59% C, 5·20% H, 19·71% N; found: 67·81% C, 5·55% H, 19·93% N.

2-Ethoxycarbonylmethylene-1,4-dimethyl-3-oxo-1,2,3,4-tetrahydroquinoxaline (*VIII*)

Solution of 1 g (0·074 mol) N,N'-dimethyl-1,2-diaminobenzene[17] and 1·7 g (0·01 mol) ethyl 2-butine-1,4-dioate in 5 ml methanol was left to stand overnight. The precipitated solid was collected by suction and dried to give 1·4 g (75%) yellow crystals, m.p. 122–124°C (ref.[4] gives m.p. 122 to 124°C).

Tetraethyl 2,2'-[ethylene-bis(methylamino)]di-*trans*-2-butenedioate (*X*)

Mixture of 1·8 g (0·02 mol) N,N'-dimethyl-1,2-ethanediamine and 2·9 g (0·02 mol) diethyl 2-butine-1,4-dioate in 15 ml methanol was boiled 10 min. After cooling the product was separated by filtration and recrystallized from ethanol to give 3·3 g (87%) crystals, m.p. 114–116°C. For $C_{20}H_{32}N_2O_8$ (428·5) calculated: 56·06% C, 7·53% H, 6·54% N; found: 56·29% C, 7·19% H, 6·81% N.

2,3-Dioxo-1,2,3,4-tetrahydroquinoxaline (*XII*)

a) 2·31 g (0·01 mol) ester *V* and 50 ml 40% peracetic acid was heated at 50°C 8 h. The mixture was cooled, and the separated solid was collected by suction, washed with acetic acid and water, and dried in air to give 1·5 g (93%) product, m.p. < 360°C, showing identical IR spectrum with that of the authentic substance[19].

b) The same product was obtained also by analoguos oxidation of ethyl 2-(3,4-dihydro-3-oxo-2-quinoxalinyl)propanoate (*XV*); yield 89%.

2-Methoxycarbonylmethylene-4-methyl-3-oxo-1,2,3,4-tetrahydroquinoxaline (*XIII*)

1·1 g (0·0063 mol) 1,3-dimethyl-2-oxo-1,2-dihydroquinoxaline and 0·5 g (0·02 mol) sodium hydride in 5 ml dimethyl carbonate and 40 ml toluene was boiled 5 h. The mixture was treated with 1 ml water, acidified with acetic acid, evaporated, and the residue was recrystallized from ethyl acetate (charcoal) to give 0·7 g (61%) yellow needles, m.p. 160–162°C (ref.[4] gives m.p. 162–164°C).

2-(1-Ethoxycarbonylethyl)-3,4-dihydro-3-oxoquinoxaline 1-oxide (*XVI*)

Solution of 2 g *m*-chloroperbenzoic acid (90%, i.e. 10% excess) in 50 ml chloroform was added to solution of 2·45 g (0·01 mol) ester *XV* in 5 ml chloroform during 5 min. The mixture was left to stand overnight, boiled 5 min, concentrated, and separated by column chromatography (alumina activity II; cyclohexane–chloroform 1 : 1). The starting compound is eluted first, being followed by the reaction product. Evaporation of the solvents gave 0·4 g (15%) raw product, m.p. 135–147°C, which was recrystallized from cyclohexane–tetrachloromethane mixture; m.p. of the pure product 147–148°C. ^{1}H NMR spectrum: 4·63 ppm **CH**–CH_3 (q), 4·18 ppm $\mathbf{CH_2}$–CH_3 (q), 1·55 ppm CH–$\mathbf{CH_3}$ (d), 1·16 ppm CH_2–$\mathbf{CH_3}$ (t). For $C_{13}H_{14}N_2O_4$ (262·3) calculated: 59·53% C, 5·38% H, 10·68% N; found: 59·42% C, 5·88% H, 10·91% N.

2-Ethoxycarbonylmethylene-1-hydroxy-4-methyl-3-oxo-1,2,3,4-tetrahydroquinoxaline (*XVII*)

Solution of 1·5 g (0·0063 mol) ester *XI* in 30 ml chloroform was mixed with solution of 1·15 g *m*-chloroperbenzoic acid (90%, *i e* 5% excess) in 15 ml chloroform at 0°C. After standing at

0°C 1 h, the mixture was separated by chromatography on alumina (activity IV) with benzene–ethyl acetate 1 : 2 mixture as eluent. After a small amount of yellow forerun the product was isolated in the yield 0·35 g (15%), m.p. 78–82°C. After recrystalization from cyclohexane–ethyl acetate m.p. 82–84°C. For $C_{13}H_{14}N_2O_4$ (262·3) calculated: 59·53% C, 5·38% H, 10·68% N; found: 59·78% C, 5·04% H, 10·77% N.

1-Acetoxy-2-ethoxycarbonylmethylene-4-methyl-3-oxo-1,2,3,4-tetrahydroquinoxaline (*XVIII*)

Mixture of 0·15 g (0·0057 mol) hydroxy derivative *XVII* and 2 ml acetic anhydride was left to stand at room temperature 14 days. The excess acetic anhydride was distilled off in vacuum from water bath, and the evaporation residue was separated by chromatography (silica gel; chloroform–ethyl acetate 1 : 1). Evaporation of the eluate corresponding to the higher spot gave the raw product which was recrystallized from diluted ethanol. Yield 0·095 g (54%) plates, m.p. 91–94°C. ^{1}H NMR spectrum: 6·55 ppm (s) =CH—, 4·30 ppm (q) CH_2—CH_3, 3·70 and 2·21 ppm (s) N—CH_3 and $COCH_3$, 1·25 ppm (t) CH_2CH_3. For $C_{15}H_{16}N_2O_5$ (304·3) calculated: 9·21% N; found: 8·73% N.

1,3-Dimethyl-2-oxo-1,2-dihydroquinoxaline 1-oxide (*XIX*)

Solution of 1·7 g (0·01 mol) 1,3-dimethyl-2-oxo-1,2-dihydroquinoxaline in 50 ml chloroform was treated with 2 g *m*-chloroperbenzoic acid (90%, *i.e.* 10% excess) at 0°C. The mixture was left to stand at room temperature 3 h, and the reaction was completed by boiling 1/2 h. Acidic products were extracted with 2 × 50 ml 3% NaOH, and the organic layer was evaporated. The residue was recrystalized twice from ethanol to give 0·5 g (28%) crystals, m.p. 206–207°C (ref.[12] gives m.p. 207°C).

1,2-Dihydro-3-methyl-2-oxoquinoxaline 4-oxide (*XX*)

Solution of 1·76 g (0·01 mol) 2-methylquinoxaline 1,4-dioxide[20] in 20 ml water was irradiated with a Tungsram 125 W mercury discharge lamp located above the surface in a closed box for 48 h. The mixture was filtered with charcoal, and the filtrate was submitted to column chromatography (silica gel; benzene–ethyl acetate 1 : 1). After a small amount of yellow forerun, the main fraction was taken which gave 1·5 g (85%) product with m.p. 246–248°C (ref.[13] gives m.p. 250°C). Further elution with methanol gave 0·2 g of the starting material.

Ethyl-3-benzoyl-N-(2-nitrophenyl)succinamate (*XXI*)

Mixture of 22·9 g (0·1155 mol) benzoylaceto-2-nitroanilide[21], 27 g ethyl iodoacetate (0·1205 mol), 17·5 g freshly annealed potassium carbonate, and 150 ml acetone was boiled 3 h. After filtration the filtrate was diluted with water and the precipitated product was collected by suction. Yield 10·5 g (33%) crystals, m.p. 110–113°C. For $C_{17}H_{19}N_3O_2$ (297·3) calculated: 61·61% C, 4·90% H, 7·56% N; found: 61·66% C, 5·18% H, 7·97% N.

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