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Five-Membered 2,3-Dioxo Heterocycles: L.* Synthesis and Thermolysis of 3-Aroyl- and 3-Hetaroyl-5-phenyl-1,2,4,5-tetrahydropyrrolo[1,2-*a*]quinoxalin-1,2,4-triones

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Abstract—(*Z*)-3-Phenacylidene- and (*Z*)-3-hetaroylmethylidene-1-phenyl-1,2,3,4-tetrahydroquinoxalin-2-ones react with oxalyl chloride to give 3-acyl-5-phenyl-1,2,4,5-tetrahydropyrrolo[1,2-*a*]quinoxaline-1,2,4-triones. Thermolysis of the latter generates acyl(3-oxo-4-phenyl-3,4-dihydroquinoxalin-2-yl)ketenes which are stabilized via [4+2]-cyclodimerization followed by [1,3]-acylotropic shift to afford 4-acyl-3-acyloxy-2-(3-oxo-4-phenyl-3,4-dihydroquinoxalin-2-yl)-6-phenyl-5,6-dihydro-1*H*-pyrido[1,2-*a*]quinoxaline-1,5-diones.

Thermal decarbonylation of five-membered dioxo heterocycles [2–18] underlies a convenient method for generation of functionally substituted heterocumulenes, including acyl(imidoyl)ketenes. In continuation of our studies in the field of generation and stabilization of acyl(imidoyl)ketenes in which the imidoyl fragment is a part of a heterocyclic system, we examined methods of synthesis and thermal transformations of compounds belonging to a new class of five-membered dioxo heterocycles.

Reactions of heterocyclic enaminoketones with oxalyl chloride are widely used for the preparation of 4-acyl-2,3-dihydropyrrole-2,3-diones fused through the [*a*] side with nitrogen-containing heterocycles [2–7]. Such reactions with substituted 1-acylmethylidene-1,2,3,4-tetrahydroisoquinolines [3], 3-acylmethylidene-3,4-dihydro-2*H*-1,4-benzoxazin-2-ones [4], 2-acylmethylidene-3,4-dihydro-2*H*-1,3-benzoxazin-4-ones [5], 2-phenacylidene-1,2,3,5-tetrahydro-4,1-benzoxazepin-3-ones [6], and 2-phenacylidene-3-phenyl-1,2,3,4-tetrahydroquinazolin-4-ones [7] produce the corresponding 4-acyl-2,3-dihydropyrrole-2,3-diones fused through the [*a*] side to isoquinoline, 1,4- and 1,3-benzoxazine, 4,1-benzoxazepine, and quinazoline rings in almost quantitative yield. These reactions are generally smooth; however, if the initial enaminoketone possesses an additional center capable of being acylated with oxalyl chloride, by-products may be formed. For example, the reaction of (*Z*)-3-ethoxycarbonylmethylidene-1,2,3,4-tetrahydroquinoxalin-2-one with oxalyl chloride leads to formation of expected 3-ethoxycarbonyl-1,2,4,5-tetrahydropyrrolo[1,2-*a*]-quinoxaline-1,2,4-trione and 4-ethoxycarbonyl-3,5-dihydro-2*H*-pyrano[2,3-*b*]quinoxaline-2,3-dione as minor product [8].

As shown in recent publications [7, 9, 10], another pathway is possible in the reaction of heterocyclic enaminoketones with oxalyl chloride. (*Z*)-3-Aryl-2-phenacylidene-1,2-dihydroquinoxalines react with oxalyl chloride to afford compounds belonging to a new class of 4-heteryl-2,3-dihydrofuran-2,3-diones, 3-aryl-2-(2-aryl-4,5-dioxo-4,5-dihydro-3-furyl)quinoxalines [7], instead of the expected 3-aroyl-4-aryl-1,2-dihydropyrrolo[1,2-*a*]quinoxaline-1,2-diones [7]. Likewise, 2-(4,5-dioxo-2-phenyl-4,5-dihydro-3-furyl)-4*H*-3,1-benzoxazin-4-one was obtained from (*E*)-2-phenacylidene-2,4-dihydro-1*H*-3,1-benzoxazin-4-one and oxalyl chloride [9], while 2,3-bis(phenacylidene)-1,2,3,4-tetrahydroquinoxaline gave rise to 2,3-bis(2-phenyl-4,5-dioxo-4,5-dihydro-3-furyl)quinoxaline (bis-furandione) [7, 10]. The predominant pathway in the reaction with oxalyl chloride is likely to be determined by the structure of enaminoketone, and study of the effect of this factor is an important aspect of the present work.

In order to obtain an additional information which may be useful for prediction of the reaction pathway, we examined reactions of (*Z*)-3-phenacylidene- and

* For communication XLIX, see [1].

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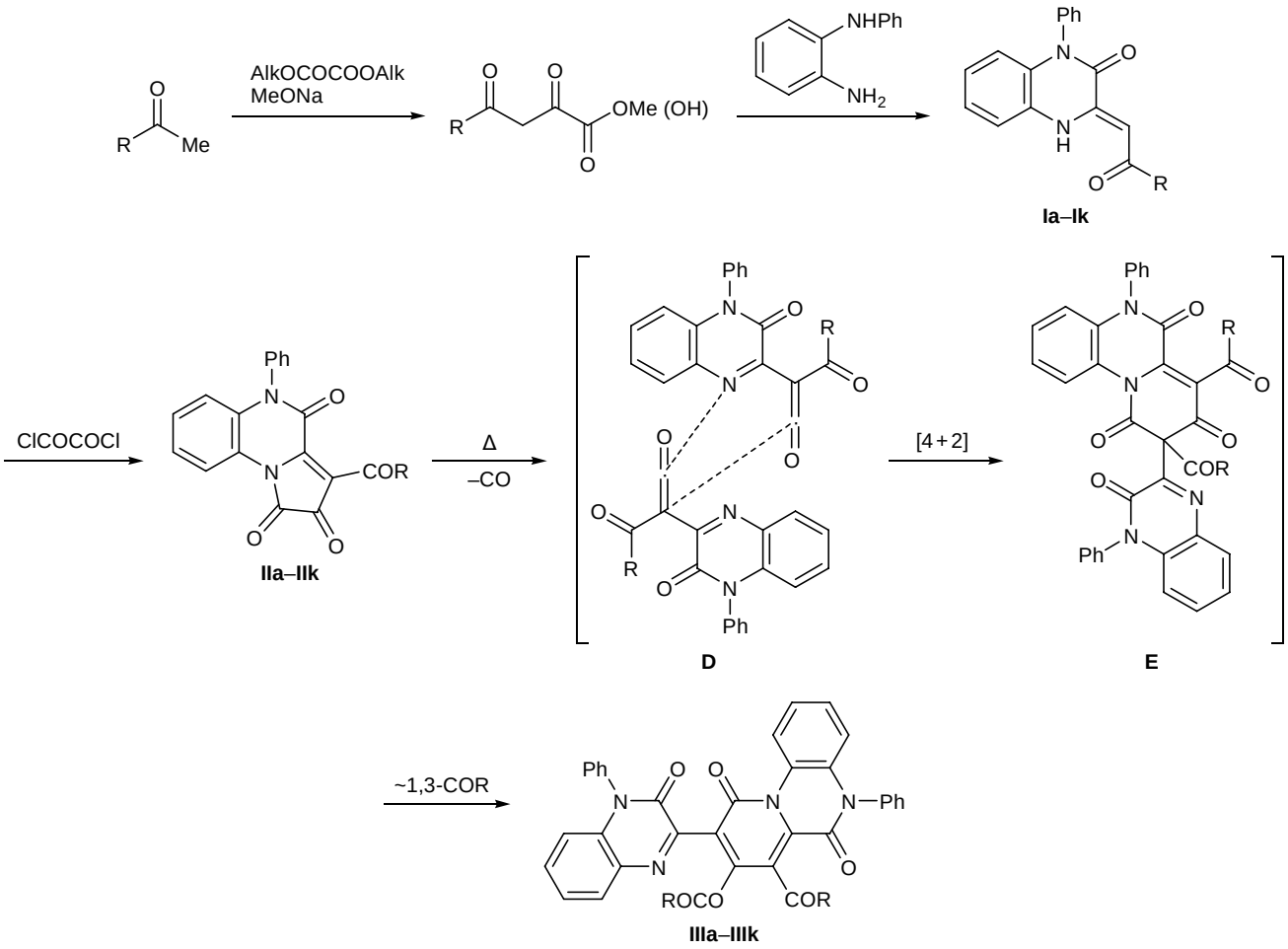
Scheme 1.



R = Ph (**a**), 4-MeC_6H_4 (**b**), 4-$MeOC_6H_4$ (**c**), 4-$EtOC_6H_4$ (**d**), 4-FC_6H_4 (**e**), 4-ClC_6H_4 (**f**), 4-BrC_6H_4 (**g**), 4-$O_2NC_6H_4$ (**h**), 2-furyl (**i**), 5-methyl-2-furyl (**j**), 5-chloro-2-thienyl (**k**).

(*Z*)-3-hetaroylmethylidene-1-phenyl-1,2,3,4-tetrahydroquinoxalin-2-ones **Ia**–**Ik** with oxalyl chloride. Quinoxalinones **Ia**–**Ik** were synthesized according to the known procedure [11] by reactions of aroyl- and hetaroylpyruvic acids or esters derived therefrom with *N*-phenyl-*o*-phenylenediamine (Scheme 1). Compounds **Ia**–**Ik** are bright yellow high-melting crystalline substances. They are readily soluble in dimethylformamide and dimethyl sulfoxide, poorly soluble in common organic solvents, and insoluble in water and saturated hydrocarbons.

The IR spectra of compounds **Ia**–**Ik** contain absorption bands belonging to stretching vibrations of the N–H group involved in intramolecular hydrogen bond (a broad band in the region 3040–3070 cm^{-1}, amide carbonyl group (C^2=O, 1670–1689 cm^{-1}), and ketone carbonyl groups (aroyl and hetaroyl moieties involved in intramolecular hydrogen bond; a broad band in the region 1606–1624 cm^{-1}). In the ^1H NMR spectra of solutions of **Ia**–**Ik** in DMSO-d_6 we observed signals from protons in the aromatic rings and substituents attached thereto, a singlet from the vinyl proton (C^3=CH) at δ 6.62–6.91 ppm, and a singlet from the N^4H proton; the latter appeared in a weak field (δ 13.37–14.05 ppm) due to formation of intramolecular hydrogen bond. Among the aromatic proton signals (δ 6.33–6.44 ppm), the doublet from 8-H in the quinoxaline ring was displaced upfield due to shielding by π-electrons in the benzene ring on N^1, and the doublet from two *ortho*-protons in the ArCO group was displaced downfield (δ 7.89–8.33 ppm; compounds **Ia**–**Ih**).

The spectral parameters of compounds **Ia**–**Ik** indicate that they exist in crystal and in solution as the corresponding *Z* isomers stabilized by a strong intramolecular hydrogen bond (like H-chelate) between the

N^4H proton and side-chain carbonyl group; such hydrogen bonding is typical of these compounds [4, 11].

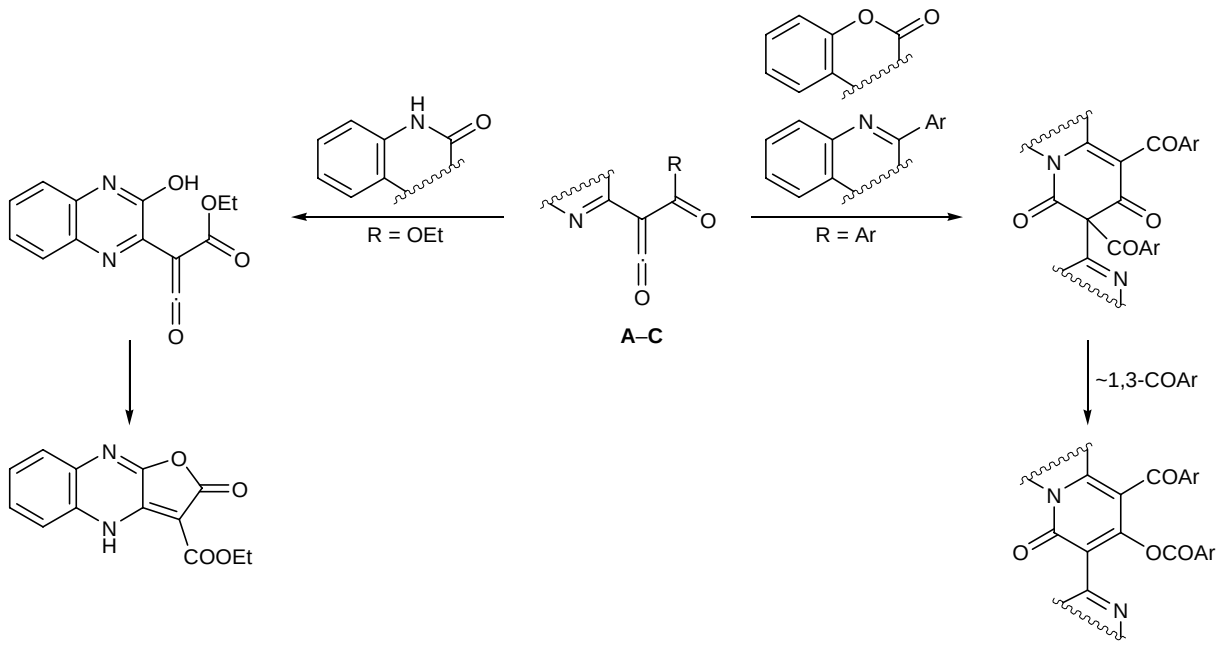
Quinoxalinones **Ia**–**Ik** were treated with oxalyl chloride on heating for 1 h in boiling anhydrous benzene. As a result, we obtained in almost quantitative yields representatives of a new class of 4-acyl-2,3-dihydropyrrole-2,3-diones fused through the [*a*] side with aza heterocycles, 3-aroyl- and 3-hetaroyl-5-phenyl-1,2,4,5-tetrahydropyrrolo[1,2-*a*]quinoxaline-1,2,4-triones **IIa**–**IIk** (Scheme 1). Compounds **IIa**–**IIk** were isolated as dark violet (almost black) crystalline substances with high melting points (with decomposition). The products are readily soluble in DMF and DMSO, poorly soluble in common aprotic organic solvents, and insoluble in alkanes. Compounds **IIa**–**IIk** reacted with water and alcohols and lose their color on storage due to hydrolysis by atmospheric moisture.

Pyrroloquinoxalines **IIa**–**IIk** showed in the IR spectra absorption bands at 1760–1775 cm^{-1} due to stretching vibrations of the lactam carbonyl group (C^1=O), 1721–1740 cm^{-1} (ketone carbonyl group C^2=O), 1682–1700 cm^{-1} (amide carbonyl C^4=O), and 1638–1670 cm^{-1} (side-chain ketone carbonyl group). The frequency ranges corresponding to stretching vibrations of the carbonyl groups in the pyrroledione fragment, as well as the higher frequency of stretching vibrations of the lactam carbonyl group as compared to the endocyclic ketone carbonyl group, are consistent with published data for monocyclic 2,3-dihydropyrrole-2,3-diones [12] and isatins [13].

The ^{1}H NMR spectra of **IIa–IIk** in DMSO-d_6 contained signals from aromatic protons and substituents in the aromatic rings. The doublet from the 6-H proton was located in a strong field, δ 6.36–6.45 ppm, due to shielding by π-electrons of the benzene ring on N^5, while the doublet from two *ortho*-protons in the ArCO group (δ 7.82–8.07 ppm, compounds **IIa**–**IIh**) and the doublet from 9-H (deshielded by the C^1=O group; δ 8.01–8.56 ppm) were observed in a weak field.

It is known that stabilization of acyl(imidoyl)-ketenes generated by thermal decarbonylation of substituted 2,3-dihydropyrrole-2,3-diones and 2,3-dihydrofuran-2,3-diones can follow different paths, depending on the substituent in the dioxo heterocycle. Two stabilization pathways were reported for acyl(imidoyl)-ketenes in which the imidoyl fragment constitutes a part of a heterocyclic system (Scheme 2). Ethoxycarbonyl(3-oxo-3,4-dihydroquinoxalin-2-yl)ketene **A** stabilizes via transformation of the quinoxalinone fragment from the amide to hydroxyimino form, followed by intramolecular acylation of the hydroxyimino group by the ketene moiety [14]. Aroyl(2-oxo-3,4-dihydro-2*H*-1,4-benzoxazin-2-yl)ketenes **B** and aroyl(3-arylquinoxalin-2-yl)ketenes **C** undergo [4+2]-cyclodimerization where one ketene molecule acts as diene (imidoylketene fragment), and the other, as dienophile

Scheme 2.



A, R = OEt; **B**, **C**, R = Ar.

(ketene C=C bond) [15, 16]; next follows thermal [1,3]-migration of the aroyl group in the primary cycloadduct.

Intramolecular cyclization like that described in [14] is impossible (for structural reasons) for aroyl- and hetaroyl(3-oxo-4-phenyl-3,4-dihydroquinoxalin-2-yl)ketenes **D** which are expected to be formed as a result of thermal decarbonylation of 3-aroyl- and 3-hetaroyl-5-phenyl-1,2,4,5-tetrahydropyrrolo[1,2-*a*]-quinoxaline-1,2,4-triones **IIa**–**IIk** (Scheme 1). However, there are alternatives for participation in intermolecular reactions through both acylketene and imidoylketene fragments.

When pyrroloquinoxalinetriones **IIa–IIk** were heated for 30–60 min at 172–173°C in an inert aprotic solvent (decane or decane–Dowtherm A, 5:1), we obtained in good yields 4-acyl-3-acyloxy-2-(3-oxo-4-phenyl-3,4-dihydroquinoxalin-2-yl)-6-phenyl-5,6-dihydro-1*H*-pyrido[1,2-*a*]quinoxaline-1,5-diones **IIIa**–**IIIk** [17] (Scheme 1). According to the TLC data, the reaction mixtures contained traces of initial quinoxalinones **Ia**–**Ik**; presumably, they were formed by hydrolysis of pyrroloquinoxalinetriones **IIa**–**IIk** with traces of water present in the solvent. Compounds **IIIa**–**IIIk** are yellow high-melting crystalline substances which melt with decomposition and are readily soluble in DMF and DMSO, poorly soluble in common organic solvents, and insoluble in water and saturated hydrocarbons.

The IR spectra of **IIIa**–**IIIk** contained absorption bands due to stretching vibrations of the ester carbonyl group (1759–1770 cm^{-1}) and amide and aroyl (hetaroyl) carbonyl groups (one or two broad bands in the region 1656–1680 cm^{-1}). Compounds **IIIa**–**IIIk** showed in the ^1H NMR spectra signals from aromatic protons and protons in the substituents attached to aromatic rings. The doublet signals from 7-H and 5'-H appeared in a strong field (δ 6.52–6.55 and 6.65–6.87 ppm, respectively) due to shielding by *p*-electrons in the benzene rings on the nitrogen atoms. The 10-H proton is deshielded due to interaction with the C^1=O carbonyl group, and its signal is displaced downfield (δ 9.05–9.20 ppm). The spectral parameters of compounds **IIIa–IIIk** are similar to structurally related dimers derived from ketenes **B** and **C**; the structure of the latter was proved by X-ray analysis [15, 16].

Presumably, ketenes **D** undergo [4+2]-cyclodimerization through the imidoylketene fragment in a way similar to that reported in [15, 16], provided that no other partner is present. Primary [4+2]-cycloadducts **E** are stabilized via [1,3]-C–O migration of the acyl group, which is typical of ketenes like **B** and **C** [15, 16]. The described reaction may be regarded as a convenient and regioselective method for building up difficultly accessible functionalized pyrido[1,2-*a*]-quinoxaline system.

EXPERIMENTAL

The IR spectra were recorded on a UR-20 spectrometer from samples dispersed in mineral oil. The ^1H and ^{13}C NMR spectra were measured at the *Ural–YaMR* Center (Russian Foundation for Basic Research project no. 00-03-40139) on a Bruker AM-400 instrument (400 MHz for ^1H) from solutions in DMSO-d_6 using tetramethylsilane as internal reference. The purity of the products was checked by thin-layer chromatography on Silufol plates using ethyl acetate, ethyl acetate–benzene (1:5), and benzene as eluents; spots were visualized by treatment with iodine vapor or under UV light.

(*Z*)-3-Phenacylidene-1-phenyl-1,2,3,4-tetrahydroquinoxalin-2-one (Ia). A solution of 2.00 g (9.7 mmol) of methyl 2,4-dioxo-4-phenyl-2-butenoate and 1.79 g (9.7 mmol) of *N*-phenyl-*o*-phenylenediamine in 50 ml of 2-propanol was heated for 60 min under reflux. The mixture was cooled, and the precipitate was filtered off and recrystallized from ethyl acetate. Yield 2.41 g (73%), mp 217–218°C; published data [4]: mp 212–213°C. IR spectrum, ν, cm^{-1}: 1610 br (COPh), 1670 (C^2=O), 3060 br (NH). ^1H NMR spectrum, δ, ppm: 6.40 d (1H, 8-H, J = 8.0 Hz), 6.89 s (1H, C^3=CH), 7.01–7.70 m (11H, H_{arom}), 7.99 d (2H, *o*-H in PhCO, J = 8.7 Hz), 13.91 s (NH). Found, %: C 77.68; H 4.70; N 8.22. $C_{22}H_{16}N_2O_2$. Calculated, %: C 77.63; H 4.74; N 8.23.

Compounds **Ib**–**Ik** were synthesized in a similar way.

(*Z*)-3-(4-Methylphenacylidene)-1-phenyl-1,2,3,4-tetrahydroquinoxalin-2-one (Ib). Yield 2.83 g (80%), mp 206–207°C (from ethyl acetate); published data [18]: mp 205–207°C. IR spectrum, ν, cm^{-1}: 1613 br (COAr), 1689 (C^2=O), 3040 br (NH). ^1H NMR spectrum, δ, ppm: 2.49 s (3H, Me), 6.39 d (1H, 8-H, J = 8.1 Hz), 6.86 s (1H, C^3=CH), 6.99–7.70 m (10H, H_{arom}), 7.89 d (2H, *o*-H in ArCO, J = 8.8 Hz), 13.89 s (NH). Found, %: C 77.99; H 5.14; N 7.88. $C_{23}H_{18}N_2O_2$. Calculated, %: C 77.95; H 5.12; N 7.90.

(*Z*)-3-(4-Methoxyphenacylidene)-1-phenyl-1,2,3,4-tetrahydroquinoxalin-2-one (Ic). Yield 2.37 g

(64%), mp 218–219°C (from ethyl acetate). IR spectrum, ν, cm^{-1}: 1620 br (COAr), 1688 (C^2=O), 3040 br (NH). ^{1}H NMR spectrum, δ, ppm: 2.49 s (3H, Me), 6.39 d (1H, 8-H, *J* = 8.1 Hz), 6.86 s (1H, C^3=CH), 6.99–7.70 m (10H, H_{arom}), 7.89 d (2H, *o*-H in ArCO, *J* = 8.8 Hz), 13.89 s (NH). Found, %: C 74.54; H 5.07; N 7.59. $C_{23}H_{18}N_2O_3$. Calculated, %: C 74.58; H 5.00; N 7.56.

(*Z*)-3-(4-Ethoxyphenacylidene)-1-phenyl-1,2,3,4-tetrahydroquinoxalin-2-one (Id). Yield 2.92 g (76%), mp 204–205°C (from ethyl acetate). IR spectrum, ν, cm^{-1}: 1606 br (COAr), 1680 (C^2=O), 3050 br (NH). ^{1}H NMR spectrum, δ, ppm: 1.36 t (3H, Me), 4.12 q (2H, CH_2O), 6.33 d (1H, 8-H, *J* = 8.1 Hz), 6.83 s (1H, C^3=CH), 7.00–7.67 m (10H, H_{arom}), 7.96 d (2H, *o*-H in ArCO, *J* = 8.8 Hz), 13.77 s (NH). Found, %: C 74.96; H 5.27; N 7.28. $C_{24}H_{20}N_2O_3$. Calculated, %: C 74.98; H 5.24; N 7.29.

(*Z*)-3-(4-Fluorophenacylidene)-1-phenyl-1,2,3,4-tetrahydroquinoxalin-2-one (Ie). Yield 2.54 g (71%), mp 204–205°C (from ethyl acetate). IR spectrum, ν, cm^{-1}: 1608 br (COAr), 1683 (C^2=O), 3070 br (NH). ^{1}H NMR spectrum, δ, ppm: 6.40 d (1H, 8-H, *J* = 7.9 Hz), 6.84 s (1H, C^3=CH), 7.01–7.68 m (10H, H_{arom}), 8.07 d (2H, *o*-H in ArCO, *J* = 8.9 Hz), 13.82 s (NH). Found, %: C 73.77; H 4.20; N 7.83. $C_{22}H_{15}FN_2O_2$. Calculated, %: C 73.73; H 4.22; N 7.82.

(*Z*)-3-(4-Chlorophenacylidene)-1-phenyl-1,2,3,4-tetrahydroquinoxalin-2-one (If). Yield 1.87 g (50%), mp 212–213°C (from ethyl acetate); published data [4]: mp 211–213°C. IR spectrum, ν, cm^{-1}: 1620 br (COAr), 1686 (C^2=O), 3050 br (NH). ^{1}H NMR spectrum, δ, ppm: 6.35 d (1H, 8-H, *J* = 8.3 Hz), 6.85 s (1H, C^3=CH), 7.06–7.68 m (10H, H_{arom}), 8.01 d (2H, *o*-H in ArCO, *J* = 8.7 Hz), 13.84 s (NH). Found, %: C 70.50; H 4.06; Cl 9.44; N 7.45. $C_{22}H_{15}ClN_2O_2$. Calculated, %: C 70.50; H 4.03; Cl 9.46; N 7.47.

(*Z*)-3-(4-Bromophenacylidene)-1-phenyl-1,2,3,4-tetrahydroquinoxalin-2-one (Ig). Yield 1.84 g (44%), mp 242–243°C (from ethyl acetate). IR spectrum, ν, cm^{-1}: 1620 br (COAr), 1684 (C^2=O), 3040 br (NH). ^{1}H NMR spectrum, δ, ppm: 6.36 d (1H, 8-H, *J* = 7.8 Hz), 6.85 s (1H, C^3=CH), 7.06–7.74 m (10H, H_{arom}), 7.93 d (2H, *o*-H in ArCO, *J* = 8.6 Hz), 13.85 s (NH). Found, %: C 63.02; H 3.61; Br 19.06; N 6.68. $C_{22}H_{15}BrN_2O_2$. Calculated, %: C 63.04; H 3.62; Br 19.08; N 6.67.

(*Z*)-3-(4-Nitrophenacylidene)-1-phenyl-1,2,3,4-tetrahydroquinoxalin-2-one (Ih). Yield 2.35 g (61%), mp 226–227°C (from ethyl acetate). IR spectrum, ν, cm^{-1}: 1612 br (COAr), 1687 (C^2=O), 3050 br (NH). ^{1}H NMR spectrum, δ, ppm: 6.44 d (1H, 8-H, *J* = 7.9 Hz), 6.91 s (1H, C^3=CH), 7.05–7.70 m (10H, H_{arom}), 8.33 d (2H, *o*-H in ArCO, *J* = 8.6 Hz), 8.34 d (2H, *m*-H in ArCO, *J* = 9.1 Hz), 14.05 s (NH). Found, %: C 68.59; H 3.90; N 10.94. $C_{22}H_{15}N_3O_4$. Calculated, %: C 68.57; H 3.92; N 10.90.

(*Z*)-3-(2-Furoylmethylidene)-1-phenyl-1,2,3,4-tetrahydroquinoxalin-2-one (Ii). Yield 2.75 g (86%), mp 242–243°C (from ethyl acetate). IR spectrum, ν, cm^{-1}: 1625 br (COHt), 1677 (C^2=O), 3040 br (NH). ^{1}H NMR spectrum, δ, ppm: 6.34 d (1H, 8-H, *J* = 8.3 Hz), 6.69 s (1H, C^3=CH), 6.71–7.64 m (11H, H_{arom}), 13.45 s (NH). Found, %: C 72.69; H 4.28; N 8.45. $C_{20}H_{14}N_2O_3$. Calculated, %: C 72.72; H 4.27; N 8.48.

(*Z*)-3-(5-Methyl-2-furoylmethylidene)-1-phenyl-1,2,3,4-tetrahydroquinoxalin-2-one (Ij). Yield 2.44 g (71%), mp 212–214°C (from ethyl acetate). IR spectrum, ν, cm^{-1}: 1624 br (COHt), 1685 (C^2=O), 3040 br (NH). ^{1}H NMR spectrum, δ, ppm: 2.40 s (3H, Me), 6.34 d (1H, 8-H, *J* = 8.2 Hz), 6.62 s (1H, C^3=CH), 6.36–7.64 m (10H, H_{arom}), 13.42 s (NH). Found, %: C 73.22; H 4.66; N 8.14. $C_{21}H_{16}N_2O_3$. Calculated, %: C 73.24; H 4.68; N 8.13.

(*Z*)-3-(5-Chloro-2-thenoylmethylidene)-1-phenyl-1,2,3,4-tetrahydroquinoxalin-2-one (Ik). Yield 1.66 g (72%), mp 250–251°C (from ethyl acetate). IR spectrum, ν, cm^{-1}: 1618 br (COHt), 1675 (C^2=O), 3050 br (NH). ^{1}H NMR spectrum, δ, ppm: 6.35 d (1H, 8-H, *J* = 8.3 Hz), 6.69 s (1H, C^3=CH), 7.04–7.82 m (10H, H_{arom}), 13.37 s (NH). Found, %: C 63.08; H 3.43; Cl 9.32; N 7.38; S 8.44. $C_{20}H_{13}ClN_2O_2S$. Calculated, %: C 63.07; H 3.44; Cl 9.31; N 7.35; S 8.42.

3-Benzoyl-5-phenyl-1,2,4,5-tetrahydropyrrolo-[1,2-*a*]quinoxaline-1,2,4-trione (IIa). A solution of 1.72 g (0.005 mol) of compound **Ia** and 0.45 ml (0.00525 mol) of oxalyl chloride in 30 ml of anhydrous benzene was heated for 60 min under reflux (until the originally yellow solution turned dark violet). The mixture was cooled, and the precipitate was filtered off. Yield 1.16 g (59%), mp 224–227°C (from benzene, decomp.); published data [18]: mp 210–211°C. IR spectrum, ν, cm^{-1}: 1660 (C^3=O), 1682 (C^4=O), 1721 (C^2=O), 1760 (C^1=O). ^{1}H NMR spectrum, δ, ppm: 6.36 d (1H, 6-H, *J* = 8.2 Hz), 7.05–7.82 m (12H, H_{arom}, 7-H, 8-H), 8.01 d (1H, 9-H, *J* = 7.2 Hz). Found, %: C 73.08; H 3.57; N 7.14. $C_{24}H_{14}N_2O_4$. Calculated, %: C 73.09; H 3.58; N 7.10.

Compounds **IIb**–**IIk** were synthesized in a similar way.

3-(4-Methylbenzoyl)-5-phenyl-1,2,4,5-tetrahydropyrrolo[1,2-*a*]quinoxaline-1,2,4-trione (IIb). Yield 1.83 g (90%), mp 207–209°C (from benzene, decomp.); published data [18]: mp 208–210°C (from dichloroethane). IR spectrum, ν, cm^{-1}: 1661 (C^3=O), 1698 (C^4=O), 1731 (C^2=O), 1770 (C^1=O). ^{1}H NMR spectrum, δ, ppm: 2.40 s (3H, Me), 6.41 d (1H, 6-H, *J* = 8.1 Hz), 7.05–7.75 m (9H, H_{arom}, *m*-H in ArCO, 7-H, 8-H), 7.85 d (2H, *o*-H in ArCO, *J* = 7.9 Hz), 8.55 d (1H, 9-H, *J* = 7.6 Hz). Found, %: C 73.58; H 3.98; N 6.86. $C_{25}H_{16}N_2O_4$. Calculated, %: C 73.52; H 3.95; N 6.86.

3-(4-Methoxybenzoyl)-5-phenyl-1,2,4,5-tetrahydropyrrolo[1,2-*a*]quinoxaline-1,2,4-trione (IIc). Yield 1.86 g (88%), mp 199–200°C (from benzene, decomp.). IR spectrum, ν, cm^{-1}: 1652 (C^3=O), 1698 (C^4=O), 1732 (C^2=O), 1774 (C^1=O). ^{1}H NMR spectrum, δ, ppm: 3.83 s (3H, Me), 6.37 d (1H, 6-H, *J* = 8.0 Hz), 6.98–7.89 m (9H, H_{arom}, *m*-H in ArCO, 7-H, 8-H), 8.03 d (2H, *o*-H in ArCO, *J* = 7.8 Hz), 8.53 d (1H, 9-H, *J* = 7.8 Hz). Found, %: C 70.77; H 3.83; N 6.57. $C_{25}H_{16}N_2O_5$. Calculated, %: C 70.75; H 3.80; N 6.60.

3-(4-Ethoxybenzoyl)-5-phenyl-1,2,4,5-tetrahydropyrrolo[1,2-*a*]quinoxaline-1,2,4-trione (IId). Yield 1.92 g (88%), mp 203–205°C (from benzene, decomp.). IR spectrum, ν, cm^{-1}: 1653 (C^3=O), 1696 (C^4=O), 1728 (C^2=O), 1769 (C^1=O). ^{1}H NMR spectrum, δ, ppm: 1.29 t (3H, Me), 4.23 q (2H, CH_2O), 6.45 d (1H, 6-H, *J* = 8.1 Hz), 7.04–7.60 m (9H, H_{arom}, *m*-H in ArCO, 7-H, 8-H), 7.82 d (2H, *o*-H in ArCO, *J* = 7.9 Hz), 8.50 d (1H, 9-H, *J* = 7.7 Hz). Found, %: C 71.20; H 4.15; N 6.36. $C_{26}H_{18}N_2O_5$. Calculated, %: C 71.23; H 4.14; N 6.39.

3-(4-Fluorobenzoyl)-5-phenyl-1,2,4,5-tetrahydropyrrolo[1,2-*a*]quinoxaline-1,2,4-trione (IIe). Yield 2.02 g (98%), mp 210–211°C (from benzene, decomp.). IR spectrum, ν, cm^{-1}: 1660 (C^3=O), 1697 (C^4=O), 1735 (C^2=O), 1770 (C^1=O). ^{1}H NMR spectrum, δ, ppm: 6.40 d (1H, 6-H, *J* = 8.4 Hz), 7.05–7.92 m (11H, H_{arom}, *m*-H in ArCO, 7-H, 8-H, *o*-H in ArCO), 8.56 d (1H, 9-H, *J* = 8.3 Hz). Found, %: C 69.91; H 3.16; N 6.82. $C_{24}H_{13}FN_2O_4$. Calculated, %: C 69.90; H 3.18; N 6.79.

3-(4-Chlorobenzoyl)-5-phenyl-1,2,4,5-tetrahydropyrrolo[1,2-*a*]quinoxaline-1,2,4-trione (IIf). Yield 2.03 g (95%), mp 211–212°C (from benzene, decomp.). IR spectrum, ν, cm^{-1}: 1665 (C^3=O), 1689 (C^4=O), 1740 (C^2=O), 1771 (C^1=O). ^{1}H NMR spectrum, δ, ppm: 6.41 d (1H, 6-H, *J* = 8.2 Hz), 7.05–7.82 m (9H, H_{arom}, *m*-H in ArCO, 7-H, 8-H), 8.07 d (2H, *o*-H in ArCO, *J* = 8.7 Hz), 8.56 d (1H, 9-H, *J* = 8.1 Hz). Found, %: C 67.22; H 3.09; Cl 8.25; N 6.56. $C_{24}H_{13}ClN_2O_4$. Calculated, %: C 67.22; H 3.06; Cl 8.27; N 6.53.

3-(4-Bromobenzoyl)-5-phenyl-1,2,4,5-tetrahydropyrrolo[1,2-*a*]quinoxaline-1,2,4-trione (IIg). Yield 2.10 g (89%), mp 216–218°C (from benzene, decomp.). IR spectrum, ν, cm^{-1}: 1670 (C^3=O), 1697 (C^4=O), 1744 (C^2=O), 1777 (C^1=O). ^{1}H NMR spectrum, δ, ppm: 6.39 d (1H, 6-H, *J* = 8.3 Hz), 7.11–7.88 m (9H, H_{arom}, *m*-H in ArCO, 7-H, 8-H), 8.01 d (2H, *o*-H in ArCO, *J* = 8.9 Hz), 8.54 d (1H, 9-H, *J* = 8.3 Hz). Found, %: C 60.90; H 2.74; Br 16.86; N 5.96. $C_{24}H_{13}BrN_2O_4$. Calculated, %: C 60.91; H 2.77; Br 16.88; N 5.92.

3-(4-Nitrobenzoyl)-5-phenyl-1,2,4,5-tetrahydropyrrolo[1,2-*a*]quinoxaline-1,2,4-trione (IIh). Yield 2.08 g (95%), mp 201–203°C (from benzene, decomp.). IR spectrum, ν, cm^{-1}: 1666 (C^3=O), 1690 (C^4=O), 1739 (C^2=O), 1775 (C^1=O). ^{1}H NMR spectrum, δ, ppm: 6.42 d (1H, 6-H, *J* = 8.2 Hz), 7.10–8.38 m (11H, H_{arom}, *o*-H and *m*-H in ArCO, 7-H, 8-H), 8.52 d (1H, 9-H, *J* = 8.0 Hz). Found, %: C 65.63; H 2.96; N 9.53. $C_{24}H_{13}N_3O_6$. Calculated, %: C 65.61; H 2.98; N 9.56.

3-(2-Furoyl)-5-phenyl-1,2,4,5-tetrahydropyrrolo-[1,2-*a*]quinoxaline-1,2,4-trione (IIi). Yield 1.84 g (96%), mp 194–196°C (from benzene, decomp.). IR spectrum, ν, cm^{-1}: 1629 (C^3=O), 1680 (C^4=O), 1721 (C^2=O), 1763 (C^1=O). ^{1}H NMR spectrum, δ, ppm: 6.40 d (1H, 6-H, *J* = 8.3 Hz), 6.68–8.03 m (10H, H_{arom}), 8.53 d (1H, 9-H, *J* = 7.1 Hz). Found, %: C 68.77; H 3.14; N 7.25. $C_{22}H_{12}N_2O_5$. Calculated, %: C 68.75; H 3.15; N 7.29.

3-(5-Methyl-2-furoyl)-5-phenyl-1,2,4,5-tetrahydropyrrolo[1,2-*a*]quinoxaline-1,2,4-trione (IIj). Yield 1.95 g (98%), mp 200–202°C (from benzene, decomp.). IR spectrum, ν, cm^{-1}: 1638 (C^3=O), 1700 (C^4=O), 1735 (C^2=O), 1772 (C^1=O). ^{1}H NMR spectrum, δ, ppm: 2.36 s (3H, Me), 6.34 d (1H, 6-H, *J* = 8.1 Hz), 6.38–7.87 m (9H, H_{arom}), 8.53 d (1H, 9-H, *J* = 8.4 Hz). Found, %: C 69.36; H 3.55; N 7.01. $C_{23}H_{14}N_2O_5$. Calculated, %: C 69.35; H 3.54; N 7.03.

3-(5-Chloro-2-thenoyl)-5-phenyl-1,2,4,5-tetrahydropyrrolo[1,2-*a*]quinoxaline-1,2,4-trione (IIk). Yield 2.13 g (98%), mp 208–209°C (from benzene, decomp.). IR spectrum, ν, cm^{-1}: 1632 (C^3=O), 1684 (C^4=O), 1723 (C^2=O), 1770 (C^1=O). ^{1}H NMR spec-

trum, δ, ppm: 6.40 d (1H, 6-H, *J* = 8.2 Hz), 7.12–8.03 m (9H, H_{arom}), 8.53 d (1H, 9-H, *J* = 8.3 Hz). Found, %: C 60.76; H 2.56; Cl 8.17; N 6.40; S 7.39. $C_{22}H_{11}ClN_2O_4S$. Calculated, %: C 60.77; H 2.55; Cl 8.15; N 6.44; S 7.37.

4-Benzoyl-3-benzoyloxy-2-(3-oxo-4-phenyl-3,4-dihydroquinoxalin-2-yl)-6-phenyl-5,6-dihydro-1*H*-pyrido[1,2-*a*]quinoxaline-1,5-dione (IIIa). A solution of 1.00 g (2.5 mmol) of compound **IIa** in 20 ml of decane was heated for 60 min at 172–173°C (until the originally violet solution turned yellow). The mixture was cooled, and the precipitate was filtered off and recrystallized from acetonitrile–dichloroethane (1:1). Yield 0.73 g (78%), mp 329–330°C (decomp.). IR spectrum, ν, cm^{-1}: 1670 br, 1680 (CON, C^4–CO); 1749 (COO). ^{1}H NMR spectrum, δ, ppm: 6.54 d (1H, 7-H, *J* = 7.8 Hz), 6.60 d (1H, 5'-H, *J* = 8.3 Hz), 7.24–7.98 m (25H, H_{arom}), 9.21 d (1H, 10-H, *J* = 7.6 Hz). ^{13}C NMR spectrum, δ_C, ppm: 115.13–136.90 (C_{arom}); 151.72, 154.90, 160.40, 161.69 (C^1, C^3–O–**C**=O, C^5, $C^{3'}$); 189.10 (C^4–**C**=O). Found, %: C 75.43; H 3.87; N 7.65. $C_{46}H_{28}N_4O_6$. Calculated, %: C 75.40; H 3.85; N 7.66.

Compounds **IIIb**–**IIIk** were synthesized in a similar way.

4-(4-Methylbenzoyl)-3-(4-methylbenzoyloxy)-2-(3-oxo-4-phenyl-3,4-dihydroquinoxalin-2-yl)-6-phenyl-5,6-dihydro-1*H*-pyrido[1,2-*a*]quinoxaline-1,5-dione (IIIb). Yield 1.29 g (69%), mp 244–245°C (from acetonitrile, decomp.). IR spectrum, ν, cm^{-1}: 1670 br (CON, C^4–CO), 1770 (COO). ^{1}H NMR spectrum, δ, ppm: 2.30 s (3H, Me), 2.39 s (3H, Me), 6.52 d (1H, 7-H, *J* = 8.3 Hz), 6.58 d (1H, 5'-H, *J* = 8.6 Hz), 7.16–7.85 m (23H, H_{arom}), 9.17 d (1H, 10-H, *J* = 7.9 Hz). Found, %: C 75.76; H 4.26; N 7.33. $C_{48}H_{32}N_4O_6$. Calculated, %: C 75.78; H 4.24; N 7.36.

4-(4-Methoxybenzoyl)-3-(4-methoxybenzoyloxy)-2-(3-oxo-4-phenyl-3,4-dihydroquinoxalin-2-yl)-6-phenyl-5,6-dihydro-1*H*-pyrido[1,2-*a*]quinoxaline-1,5-dione (IIIc). Yield 0.65 g (70%), mp 319–321°C (from acetonitrile, decomp.). IR spectrum, ν, cm^{-1}: 1668 br, 1680 (CON, C^4–CO); 1750 (COO). ^{1}H NMR spectrum, δ, ppm: 3.77 s (3H, MeO), 3.90 s (3H, MeO), 6.52 d (1H, 7-H, *J* = 7.7 Hz), 6.58 d (1H, 5'-H, *J* = 7.7 Hz), 7.18–7.83 m (23H, H_{arom}), 9.16 (1H, 10-H, *J* = 8.2 Hz). Found, %: C 72.75; H 4.09; N 7.06. $C_{48}H_{32}N_4O_8$. Calculated, %: C 72.72; H 4.07; N 7.07.

4-(4-Ethoxybenzoyl)-3-(4-ethoxybenzoyloxy)-2-(3-oxo-4-phenyl-3,4-dihydroquinoxalin-2-yl)-6-phenyl-5,6-dihydro-1*H*-pyrido[1,2-*a*]quinoxaline-1,5-dione (IIId). Yield 0.66 g (73%), mp 278–279°C (from DMSO, decomp.). IR spectrum, ν, cm^{-1}: 1680 br (CON, C^4–CO), 1765 (COO). ^{1}H NMR spectrum, δ, ppm: 1.32 t (3H, Me, *J* = 7.0 Hz), 1.35 t (3H, Me, *J* = 7.0 Hz), 4.01 q (2H, CH_2O, *J* = 7.0 Hz), 4.01 q (2H, CH_2O, *J* = 7.0 Hz), 6.54 d (1H, 7-H, *J* = 8.3 Hz), 6.59 d (1H, 5'-H, *J* = 8.3 Hz), 7.18–7.86 m (23H, H_{arom}), 9.18 (1H, 10-H, *J* = 8.1 Hz). Found, %: C 73.15; H 4.44; N 6.80. $C_{50}H_{36}N_4O_8$. Calculated, %: C 73.16; H 4.42; N 6.83.

4-(4-Fluorobenzoyl)-3-(4-fluorobenzoyloxy)-2-(3-oxo-4-phenyl-3,4-dihydroquinoxalin-2-yl)-6-phenyl-5,6-dihydro-1*H*-pyrido[1,2-*a*]quinoxaline-1,5-dione (IIIe). Yield 0.67 g (72%), mp 293–294°C (from acetonitrile, decomp.). IR spectrum, ν, cm^{-1}: 1670 br (CON, C^4–CO), 1750 (COO). ^{1}H NMR spectrum, δ, ppm: 6.55 d (1H, 7-H, *J* = 7.2 Hz), 6.63 d (1H, 5'-H, *J* = 8.2 Hz), 7.05–7.99 m (23H, H_{arom}), 9.20 (1H, 10-H, *J* = 8.0 Hz). Found, %: C 71.85; H 3.45; N 7.27. $C_{46}H_{26}F_2N_4O_6$. Calculated, %: C 71.87; H 3.41; N 7.29.

4-(4-Chlorobenzoyl)-3-(4-chlorobenzoyloxy)-2-(3-oxo-4-phenyl-3,4-dihydroquinoxalin-2-yl)-6-phenyl-5,6-dihydro-1*H*-pyrido[1,2-*a*]quinoxaline-1,5-dione (IIIf). Yield 0.65 g (69%), mp 338–340°C (from acetonitrile, decomp.). IR spectrum, ν, cm^{-1}: 1670 br (CON, C^4–CO), 1760 (COO). ^{1}H NMR spectrum, δ, ppm: 6.53 d (1H, 7-H, *J* = 7.8 Hz), 6.55 d (1H, 5'-H, *J* = 8.0 Hz), 7.25–7.98 m (23H, H_{arom}), 9.18 d (1H, 10-H, *J* = 7.9 Hz). Found, %: C 68.93; H 2.28; Cl 8.88; N 6.95. $C_{46}H_{26}Cl_2N_4O_6$. Calculated, %: C 68.92; H 2.27; Cl 8.85; N 6.99.

4-(4-Bromobenzoyl)-3-(4-bromobenzoyloxy)-2-(3-oxo-4-phenyl-3,4-dihydroquinoxalin-2-yl)-6-phenyl-5,6-dihydro-1*H*-pyrido[1,2-*a*]quinoxaline-1,5-dione (IIIg). Yield 0.68 g (72%), mp 310–311°C (from acetonitrile, decomp.). IR spectrum, ν, cm^{-1}: 1676 br (CON, C^4–CO), 1759 (COO). ^{1}H NMR spectrum, δ, ppm: 6.53 d (1H, 7-H, *J* = 7.6 Hz), 6.59 d (1H, 5'-H, *J* = 8.0 Hz), 7.26–7.90 m (23H, H_{arom}), 9.18 d (1H, 10-H, *J* = 7.9 Hz). Found, %: C 62.09; H 2.97; Br 17.90; N 6.28. $C_{46}H_{26}BrN_4O_6$. Calculated, %: C 62.04; H 2.94; Br 17.95; N 6.29.

4-(4-Nitrobenzoyl)-3-(4-nitrobenzoyl)-2-(3-oxo-4-phenyl-3,4-dihydroquinoxalin-2-yl)-6-phenyl-5,6-dihydro-1*H*-pyrido[1,2-*a*]quinoxaline-1,5-dione (IIIh). Yield 0.64 g (68%), mp 314–315°C (from benzene, decomp.). IR spectrum, ν, cm^{-1}: 1680 br (CON, C^4–CO), 1760 (COO). ^{1}H NMR spectrum, δ, ppm: 6.57 d (1H, 7-H, *J* = 6.1 Hz), 6.63 d (1H, 5'-H, *J* = 8.2 Hz), 7.17–8.30 m (23H, H_{arom}), 9.23 d (1H, 10-H,

J = 8.2 Hz). Found, %: C 67.18; H 3.15; N 10.22. $C_{46}H_{26}N_6O_{10}$. Calculated, %: C 67.15; H 3.19; N 10.21.

4-(2-Furoyl)-3-(2-furoyloxy)-2-(3-oxo-4-phenyl-3,4-dihydroquinoxalin-2-yl)-6-phenyl-5,6-dihydro-1*H*-pyrido[1,2-*a*]quinoxaline-1,5-dione (IIIi). Yield 0.62 g (67%), mp 320–321°C (from toluene, decomp.). IR spectrum, ν, cm^{-1}: 1665 br (CON, C^4–CO), 1755 (COO). ^{1}H NMR spectrum, δ, ppm: 6.45 d (1H, 7-H, *J* = 7.5 Hz), 6.65 d (1H, 5'-H, *J* = 7.5 Hz), 7.17–7.88 m (21H, H_{arom}), 9.15 d (1H, 10-H, *J* = 8.3 Hz). Found, %: C 70.77; H 3.37; N 7.87. $C_{42}H_{24}N_4O_8$. Calculated, %: C 70.79; H 3.39; N 7.86.

4-(5-Methyl-2-furoyl)-3-(5-methyl-2-furoyloxy)-2-(3-oxo-4-phenyl-3,4-dihydroquinoxalin-2-yl)-6-phenyl-5,6-dihydro-1*H*-pyrido[1,2-*a*]quinoxaline-1,5-dione (IIIj). Yield 0.67 g (72%), mp 325–327°C (from toluene, decomp.). IR spectrum, ν, cm^{-1}: 1656 br, 1682 (CON, C^4–CO); 1759 (COO). ^{1}H NMR spectrum, δ, ppm: 6.35 d (1H, 7-H, *J* = 8.3 Hz), 6.55 d (1H, 5'-H, *J* = 8.2 Hz), 6.87–7.85 m (19H, H_{arom}), 9.05 d (1H, 10-H, *J* = 8.3 Hz). Found, %: C 71.36; H 3.84; N 7.58. $C_{44}H_{28}N_4O_8$. Calculated, %: C 71.35; H 3.81; N 7.56.

4-(5-Chloro-2-thenoyl)-3-(5-chloro-2-thenoyloxy)-2-(3-oxo-4-phenyl-3,4-dihydroquinoxalin-2-yl)-6-phenyl-5,6-dihydro-1*H*-pyrido[1,2-*a*]quinoxaline-1,5-dione (IIIk). Yield 0.65 g (70%), mp 323–324°C (from toluene, decomp.). IR spectrum, ν, cm^{-1}: 1660 br, 1680 (CON, C^4–CO); 1765 (COO). ^{1}H NMR spectrum, δ, ppm: 6.46 d (1H, 7-H, *J* = 7.3 Hz), 6.66 d (1H, 5'-H, *J* = 7.9 Hz), 7.19–7.94 m (19H, H_{arom}), 9.17 d (1H, 10-H, *J* = 8.2 Hz). Found, %: C 62.06; H 2.75; Cl 8.70; N 6.84; S 7.94. $C_{42}H_{22}Cl_2N_4O_6S_2$. Calculated, %: C 62.00; H 2.73; Cl 8.71; N 6.89; S 7.88.

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