

## Fused Polycyclic Nitrogen-Containing Heterocycles: VII.\* Reaction Products of 3-( $\alpha$ -Chlorobenzyl)-1,2-dihydro- quinoxalin-2-one and Thioureas as Key Intermediate Compounds in the Synthesis of Thiazolo[3,4-*a*]quinoxalines

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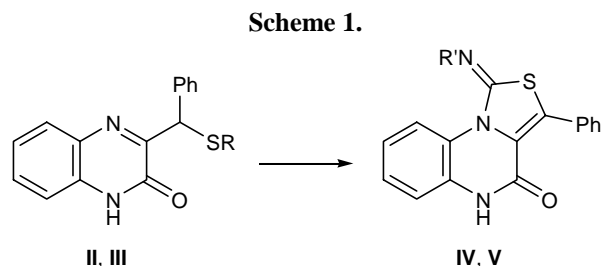
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**Abstract**—Products of the reactions of 3-( $\alpha$ -chlorobenzyl)-1,2-dihydroquinoxalin-2-one with thiourea, *N*-phenylthiourea, *N,N'*-diphenylthiourea, 2-sulfanylbenzimidazole, 2-sulfanylbenzothiazole, and potassium thiocyanate were brought into intramolecular cyclizations. Specific features of these reactions are discussed.

In the recent years, azolo[*a*]quinoxalines attract persistent interest. However, these compounds remain so far poorly studied despite the fact that a number of their derivatives exhibit various practically important properties, specifically pronounced biological activity [2]. Introduction into the fused ring system of heteroatoms other than nitrogen, e.g., sulfur, was expected to afford new azolo[*a*]quinoxaline analogs, thiazolo[*a*(or *b*)]quinoxalines, which could possess different chemical properties and physiological activity. Unlike thiazolo[*b*]quinoxalines (methods of synthesis and properties of which have been studied in sufficient detail), relatively limited published data are available on thiazolo[*a*]quinoxalines. Among the latter, mesoionic thiazolo[3,2-*a*]quinoxalines are known; they were obtained by intramolecular cyclization of 2-carboxymethylsulfanylquinoxalines [3, 4]. Thiazolo[3,4-*a*]quinoxalines were synthesized mainly via intramolecular condensation of complex and difficultly accessible thiazole and tetrahydrothiazole derivatives [5–7]. We recently described a new efficient procedure for the synthesis of thiazolo[3,4-*a*]quinoxalin-2-ones by reaction of 3-( $\alpha$ -chlorobenzyl)-1,2-dihydroquinoxalin-2-one (**I**) with potassium thiocyanate and *N,N'*-diphenylthiourea and subsequent cyclization of 3-( $\alpha$ -thiocyanato)- and 3-[ $\alpha$ -(*N,N'*-diphenylisothioureido)-benzyl]quinoxalin-2-ones **II** and **III**. The cyclization involves nucleophilic attack by the N<sup>4</sup> atom on the

electrophilic carbon atom of the thiocyanato or isothioureido group with formation of 1-imino (**IV**) and 1-phenylimino derivatives (**V**) [8] (Scheme 1).

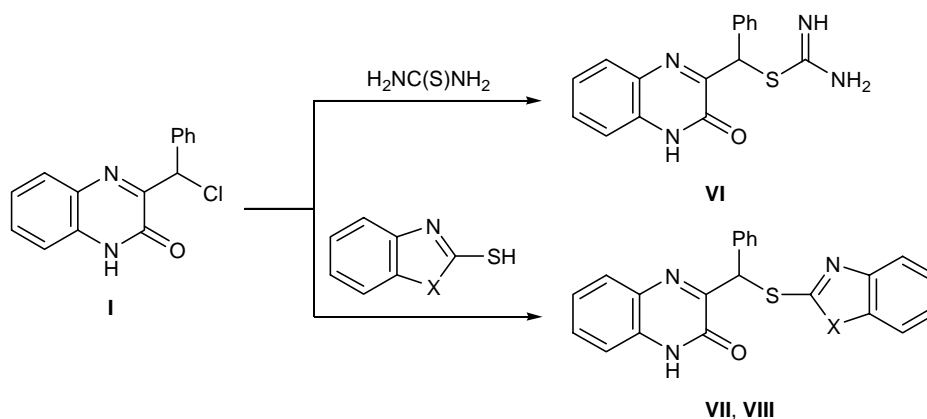


**II**, R = CN; **III**, R = C(=NPh)NHPh; **IV**, R' = H; **V**, R' = Ph.

In continuation of these studies, the present article reports on the behavior of 3-( $\alpha$ -chlorobenzyl)-1,2-dihydroquinoxalin-2-one (**I**) with various reagents which are synthetic equivalents of the  $\text{-N=C}^+\text{-S}^-$  species, in particular with thiourea, *N*-phenylthiourea, 2-sulfanylbenzimidazole, and 2-sulfanylbenzothiazole. The reactions of compound **I** with thiourea, 2-sulfanylbenzimidazole, and 2-sulfanylbenzothiazole in DMSO at room temperature afforded the corresponding 3-( $\alpha$ -RS-benzyl)-2-oxo-1,2-dihydroquinoxaline hydrochlorides which were treated with a solution of sodium carbonate to obtain free bases (Scheme 2). The structure of products **VI**–**VIII** is confirmed by the upfield shift of the benzylic proton signal in the <sup>1</sup>H NMR spectrum of **VI** ( $\delta$  6.0 ppm) and downfield shift of the same

\* For communication VI, see [1].

Scheme 2.

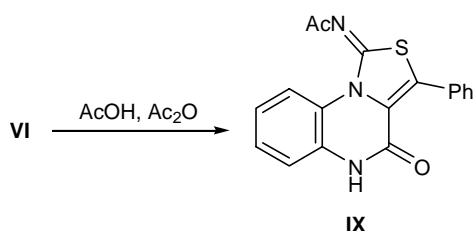


**VII**, X = NH; **VIII**, R = S.

proton signal in the spectra of **VII** and **VIII** ( $\delta$  6.64 and 6.87 ppm, respectively) relative to the corresponding signal of quinoxaline **I** ( $\delta$  6.53 ppm), as well as by the data of elemental analysis (see Experimental).

Compounds **VI**–**VIII** turned out to be stable under the conditions ensuring intramolecular cyclization of thiocyanate **II** and diphenylisothioureide **III** to thiazoloquinoxalines, i.e., on heating in 10% hydrochloric acid or in acetic acid [8]. We succeeded in obtaining 1-acetyliminothiazolo[3,4-*a*]quinoxaline **IX** in a poor yield only by prolonged (~25 h) heating of quinoxaline **VI** in boiling acetic acid (Scheme 3). Replacement of acetic acid by acetic anhydride allowed us not only to strongly shorten the reaction time (to one minute) but also to raise the yield of compound **IX** to almost quantitative.

Scheme 3.



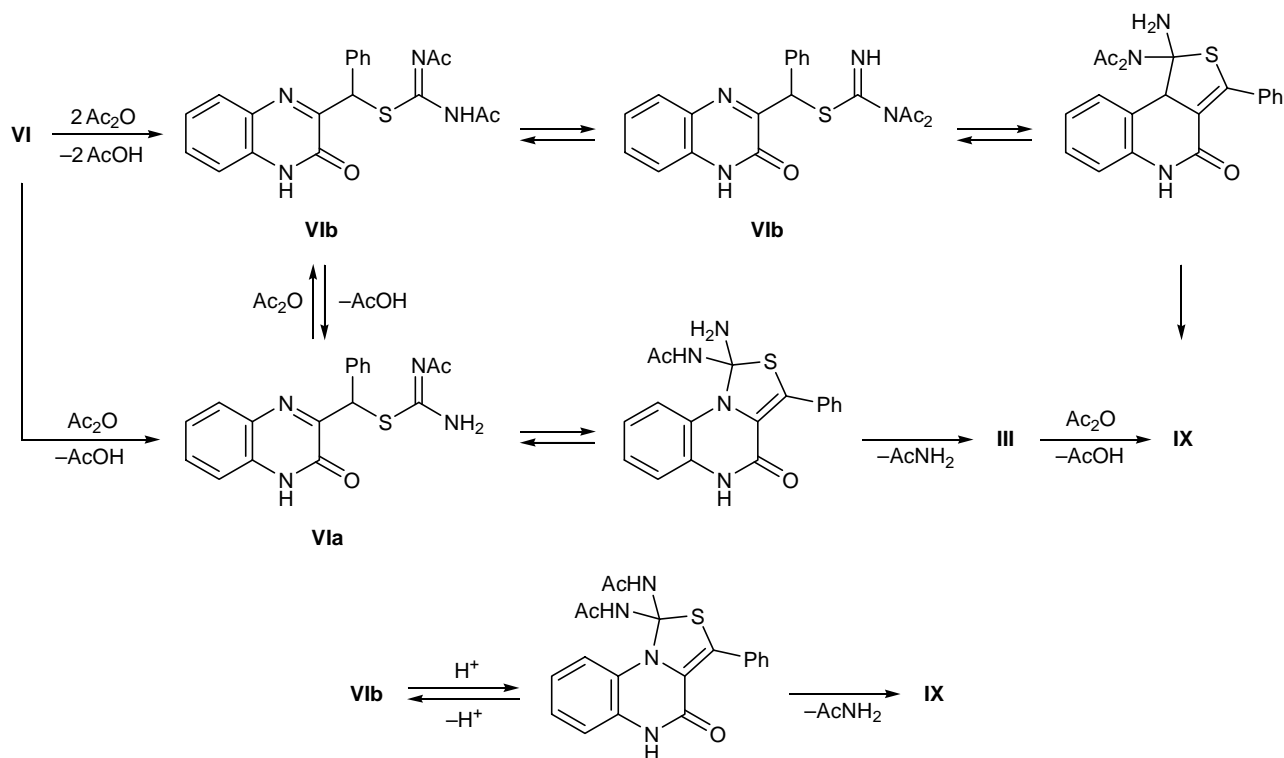
Presumably, the cyclization of **VI** to **IX** is preceded by acetylation with formation of intermediate mono- (**VIa**) and/or diacetylisothioureide (**VIb**) having readily departing  $\text{AcNH}$  groups (Scheme 4). Unlike compounds **VI**, no cyclization of **VII** and **VIII** occurred in boiling acetic anhydride. Here, compound **VII** was acetylated to give the corresponding *N*-acetyl derivative **XII** (Scheme 5). The lack of intramolecular cyclization is likely to result from the nature of the

isothioureide and isodithiocarbamate moieties both which are parts of aromatic systems.

All known methods for the preparation of thiazolo[3,4-*a*]quinoxalines are based on either thiazole derivatives [5, 9, 10] or, as we showed in [7, 8], quinoxaline derivatives. Retrosynthetic analysis of the structure of thiazolo[3,4-*a*]quinoxalines suggests that such systems could be built up not only through synthon **A** [whose synthetic equivalents are thiocyanato (**II**) and isothioureido derivatives **III** and **VI**] but also from synthons **B1** and **B2** using compound **I** and the corresponding thiourea derivatives (Scheme 6) as equivalents. In fact, by heating thiourea, *N*-phenylthiourea, or *N,N'*-diphenylthiourea with quinoxaline **I** in dioxane with subsequent addition of acetic anhydride we obtained thiazoloquinoxalines **V** and **IX** (Scheme 7). This reaction may be regarded as a one-pot procedure for the synthesis of such compounds. It should be noted that, like unsubstituted thiourea, *N*-phenylthiourea reacts with quinoxaline **VI** to give acetylimino rather than phenylimino derivative, i.e., elimination of aniline rather than acetamide occurs.

However, the yield of thiazolo[3,4-*a*]quinoxaline **IX** is considerably lower than in the two-step synthesis through isothioureide **VI**, presumably due to side processes. Heating of quinoxaline **I** with thiourea over a period of 4 h gave in a good yield a crystalline substance. The high-resolution mass spectrum of the product contained the molecular ion peak with  $m/z$  236.094 ( $I_{\text{rel}}$  100%) which corresponded to the elemental composition  $\text{C}_{15}\text{H}_{12}\text{N}_2\text{O}$  ( $M_{\text{calc}}$  236.095). In addition, in the mass spectra of the same sample, obtained at lower temperature of the direct inlet probe, we observed peaks belonging to molecular sulfur ( $\text{S}_8$ ). We failed to obtain an analytically pure sample even

Scheme 4.

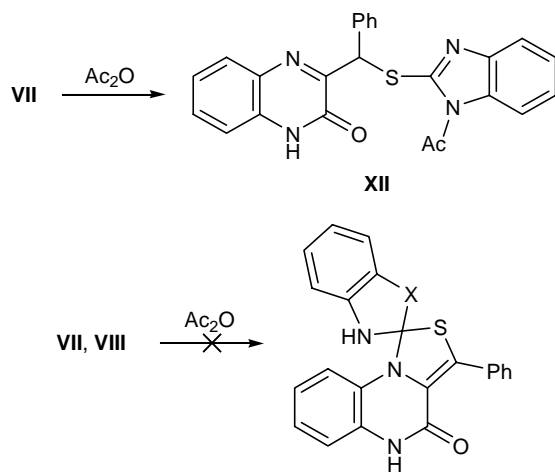


by repeated crystallizations from dioxane, although the <sup>1</sup>H NMR spectra were identical, regardless of the number of recrystallizations. The spectra contained singlets at δ 4.39 (2H) and 12.45 ppm (1H) from the CH<sub>2</sub> and NH protons, respectively, and a multiplet at δ 7.19–7.77 ppm from 9 aromatic protons. These data confirm the formation of 3-benzyl-1,2-dihydroquinoxalin-2-one (**XIII**) as the major product (Scheme 8).

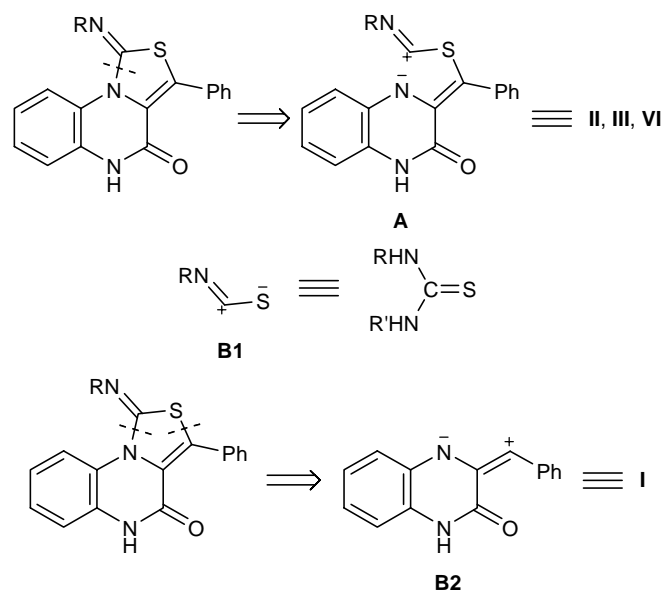
The formation of compound **XIII** may be rationalized in terms of a reaction sequence including

initial intramolecular nucleophilic condensation with participation of one exocyclic and one endocyclic imino group in hydrochloride **VI** in a way similar to the condensation of the imino group and γ-carbonyl group in compounds having an N–C–S–C–C fragment [11, 12]. By analogy with six-membered sulfur-containing systems [13, 14], acid-catalyzed isomerization

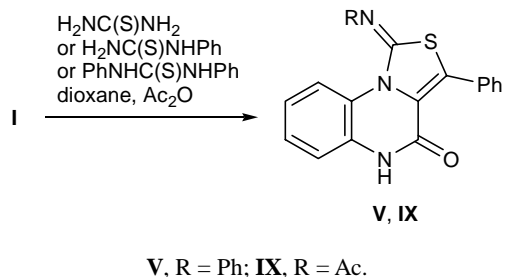
Scheme 5.



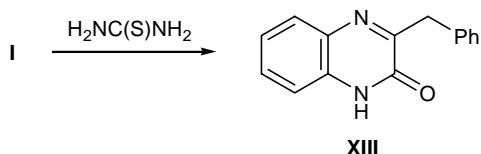
Scheme 6.



Scheme 7.



Scheme 8.

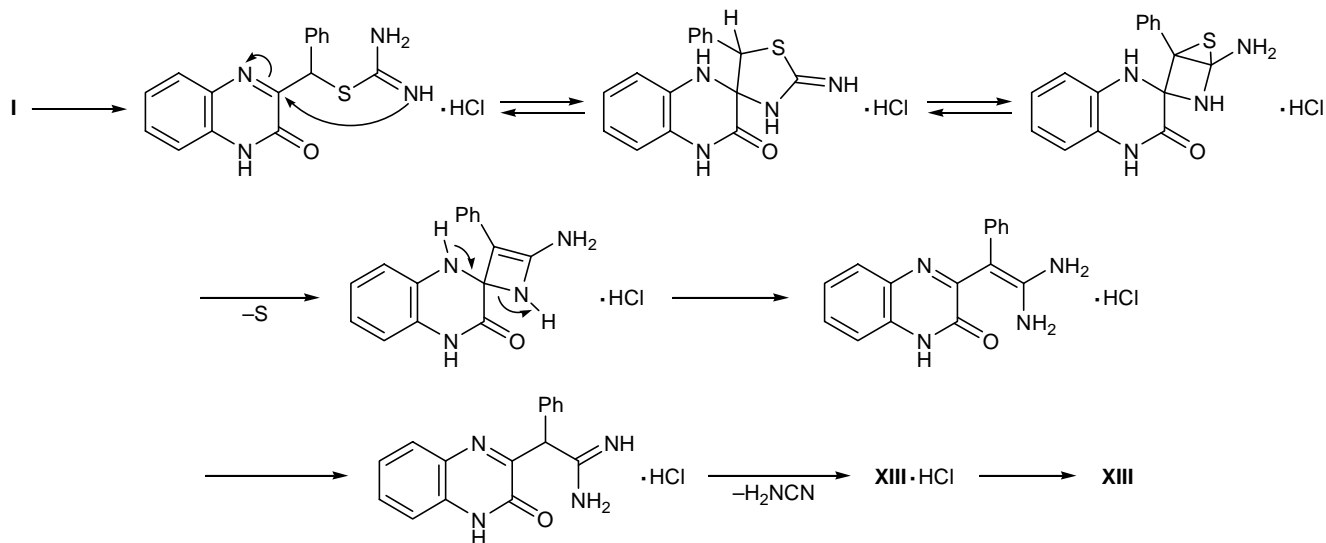


of the spiro-fused thiazolidine system leads to bicyclic structure which loses sulfur atom to give less stable azetine system. The latter is stabilized via elimination of cyanamide as shown in Scheme 9.

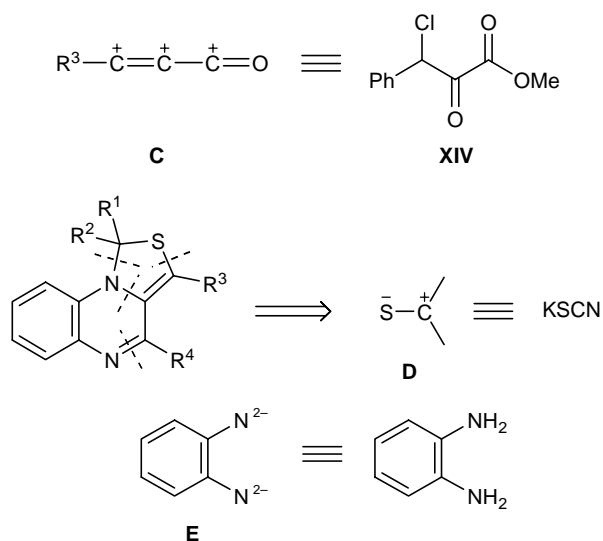
Retrosynthetic analysis shows that not only thiazole [5, 9, 10] or quinoxaline derivatives [7, 8] may be precursors of thiazolo[3,4-*a*]quinoxalines. These compounds can be built up from structures containing no such heterocyclic systems, e.g., from synthons **C**, **D**, and **E** whose synthetic equivalents are methyl chloro(phenyl)pyruvate (**XIV**), potassium thiocyanate (KSCN), and *o*-phenylenediamine, respectively (Scheme 10). The reaction of chloropyruvate **XIV** with potassium thiocyanate gave a tarry material, treatment of which with *o*-phenylenediamine afforded 1-(2-thia-

zolylimino)thiazolo[3,4-*a*]quinoxaline **XV** (Scheme 11). The electron impact mass spectrum of **XV** contained the molecular ion peak with  $m/z$  528 ( $I_{\text{rel}}$  30%). The precise  $m/z$  value (528.091) corresponds to the formula  $\text{C}_{27}\text{H}_{20}\text{N}_4\text{O}_4\text{S}_2$  ( $M_{\text{calc}}$  528.093). The presence of a thiazoloquinoxaline fragment gives rise to the following characteristic fragment ions,  $m/z$  ( $I_{\text{rel}}$ , %): 293 (42), 266 (100), 236 (68), 235 (39), 234 (40), 206 (26), 205 (31). The fragment ions with  $m/z$  237 (11) and 121 (67) are formed by decomposition of the thiazole ring [6, 7]. In the near-molecular region of the spectrum, ion peaks with  $m/z$  510 (25) and 452 (55) were present. The first of these results from elimination of water molecule from the molecular ion, indicating the presence of a hydroxy group, and the second corresponds to expulsion of  $\text{C}_2\text{H}_2\text{O}_2$  from the ion with  $m/z$  510. A number of abundant ions in the mass spectrum of **XV** appear due to localization of the positive charge on both thiazolo[3,4-*a*]quinoxaline fragment and that containing the thiazole ring (originally located at position 1 of the tricyclic system). Thus the thiazolo[3,4-*a*]quinoxaline fragment gives rise to the following fragment ions,  $m/z$  ( $I_{\text{rel}}$ , %): 293 (42), 266 (100), 236 (68), 235 (39), 234 (40), 206 (26), 205 (31); the ion peak with  $m/z$  237 (11) belongs to the thiazole ring [15, 16]. An important information can be deduced from the presence of an ion peak with  $m/z$  320 (33). The elemental composition of this ion is  $\text{C}_{17}\text{H}_{10}\text{N}_3\text{O}_2\text{S}$ ; it includes the thiazolo[3,4-*a*]quinoxaline ring system, imino nitrogen atom, and  $\text{C}^2\text{-OH}$  fragment of the thiazole ring. Therefore, the product has structure **XV** rather than isomeric structure **XVI** or **XVII**.

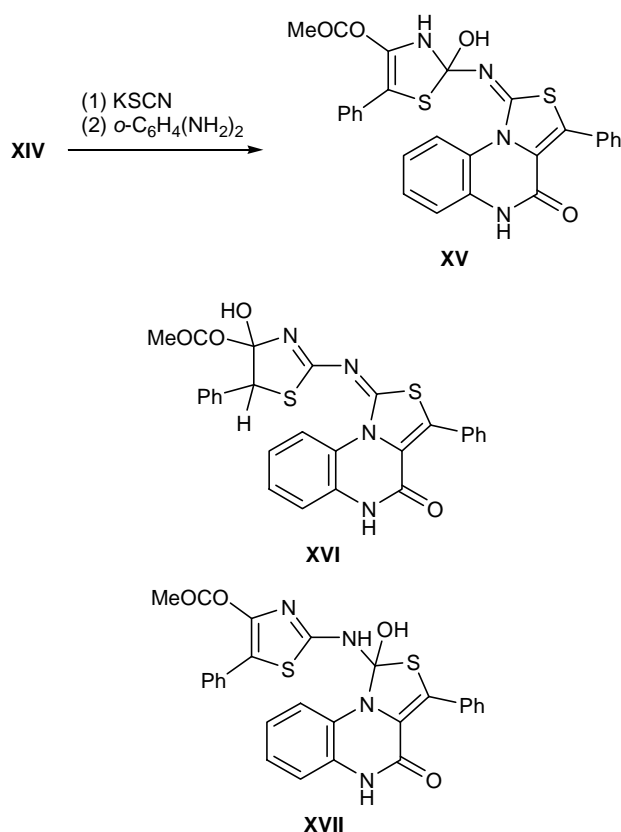
Scheme 9.



Scheme 10.



Scheme 11.



The formation of thiazolo[3,4-*a*]quinoxaline system is also confirmed by the presence of a doublet signal from 9-H ( $J = 8.5$  Hz), which is typical of such systems [8, 17, 18]: this proton resonates separately from the other 13 aromatic protons, in a weaker field. It should be noted that, regardless of the order of

mixing of the reactants (KSCN + *o*-phenylenediamine + XIV or *o*-phenylenediamine + XIV + KSCN), thiazolo[3,4-*a*]quinoxaline system is formed. In both cases, from the reaction mixture we isolated a small amount of 1-acetylimino derivative IX rather than thiazolylimino derivative XV. However, this approach to thiazoloquinoxalines requires further study which will be the subject of our subsequent publications.

## EXPERIMENTAL

The melting points were determined on a Boetius device. The IR spectra were recorded on a UR-20 spectrometer from samples dispersed in mineral oil. The  $^1H$  NMR spectra were measured on a Bruker-250 spectrometer (250.13 MHz). The electron impact mass spectra (70 eV) were obtained on an MKh-1310 instrument at a resolution  $R$  of 1500; electron collector current 30–60  $\mu A$ ; vaporizer temperature 80–200°C; SVP-5 direct inlet probe.

**3-Phenyl-1-phenylimino-4,5-dihydrothiazolo[3,4-*a*]quinoxalin-4-one (V).** *a.* A solution of 0.30 g of quinoxaline I and 0.27 g of *N,N'*-diphenylthiourea in 5 ml of dioxane was heated for 0.5 h under reflux, 2 ml of acetic anhydride and 2 ml of acetic acid were added, and the mixture was refluxed for 0.5 h, cooled, and poured into water. The solution was separated from the tarry material, 10 ml of 2-propanol was added, and the mixture was heated to the boiling point. After cooling, the precipitate was filtered off and washed with an aqueous solution of sodium carbonate, water, and 2-propanol. Yield 41 mg (10%).

*b.* A solution of 1.00 g of quinoxaline I and 0.92 g of *N,N'*-diphenylthiourea in 20 ml of dioxane was heated for 25 h under reflux. The mixture was cooled and poured into water. The tarry material was separated from the solution, 10 ml of 2-propanol was added, and the mixture was heated to the boiling point. After cooling, the precipitate was filtered off and washed with a solution of sodium carbonate, water, and 2-propanol. Yield 50 mg (12%). The physical constants of samples of V prepared by the two methods were identical to those reported in [7].

**3-( $\alpha$ -Isothioureidobenzyl)-1,2-dihydroquinoxalin-2-one (VI).** A solution of 2.00 g of compound I and 0.62 g of thiourea in 20 ml of DMSO was stirred for 6 h, and the mixture was left overnight. It was then poured into water and made alkaline by adding a 5% aqueous solution of sodium carbonate. The product was filtered off and washed with water. Yield

2.11 g (92%), mp 207–208°C. IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 1620 (C=N); 1680 (C=O); 2300–3220 (NH, carbamoyl); 3380, 3440 (NH,  $\text{NH}_2$ ).  $^1\text{H}$  NMR spectrum (DMSO- $d_6$ ),  $\delta$ , ppm: 5.86 s (1H, NH), 6.00 s (1H, PhCH), 6.57–7.53 m (9H,  $\text{C}_6\text{H}_5$ , 6-H, 7-H, 8-H), 11.46 br.s (1H, NH, carbamoyl). Found, %: C 61.65; H 4.83; N 17.84; S 10.22.  $\text{C}_{16}\text{H}_{14}\text{N}_4\text{OS}$ . Calculated, %: C 61.81; H 4.54; N 18.02; S 10.29.

**3-[ $\alpha$ -(2-Benzimidazolylsulfanyl)benzyl]-1,2-dihydroquinoxalin-2-one (VII).** A solution of 0.50 g of quinoxaline **I** and 0.31 g of 2-sulfanylbenzimidazole in 10 ml of DMSO was stirred for 6 h, and the mixture was left to stand for 2 days. The mixture was poured into water, and the precipitate was filtered off and washed with a 5% aqueous solution of sodium carbonate, water, and 2-propanol. Yield 0.60 g (85%), mp >160°C (decomp., from dioxane). IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 1615 (C=N), 1665 (C=O), 2200–3570 (NH).  $^1\text{H}$  NMR spectrum (DMSO- $d_6$ ),  $\delta$ , ppm: 6.64 s (1H, PhCH), 7.05–7.47 m (12H,  $\text{C}_6\text{H}_5$ , 6-H, 7-H, 8-H, 4'-H, 5'-H, 6'-H, 7'-H), 7.78 d (1H, 5-H,  $J = 7.83$  Hz), 12.46 br.s (1H, NH, carbamoyl). Found, %: C 68.71; H 4.25; N 14.82; S 8.50.  $\text{C}_{22}\text{H}_{16}\text{N}_4\text{OS}$ . Calculated, %: C 68.73; H 4.20; N 14.57; S 8.34.

**3-[ $\alpha$ -(2-Benzothiazolylsulfanyl)benzyl]-1,2-dihydroquinoxalin-2-one (VIII).** A solution of 0.80 g of quinoxaline **I** and 0.55 g of 2-sulfanylbenzothiazole in 10 ml of DMSO was stirred for 6 h, and the mixture was left to stand for 7 days. The precipitate was filtered off and washed with 2-propanol, a 5% aqueous solution of sodium carbonate, and water. Yield 1.06 g (90%), mp >170°C (decomp., from DMSO). IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 1610 (C=N), 1670 (C=O), 2300–3220 (NH).  $^1\text{H}$  NMR spectrum (DMSO- $d_6$ ),  $\delta$ , ppm: 6.87 s (1H, PhCH), 7.25–7.66 m (10H,  $\text{H}_{\text{arom}}$ ), 7.88 d (1H,  $\text{H}_{\text{arom}}$ ,  $J = 7.50$  Hz), 7.98 d.d (1H,  $\text{H}_{\text{arom}}$ ,  $J = 7.50$ , 7.50 Hz), 8.07 d (1H,  $\text{H}_{\text{arom}}$ ,  $J = 7.50$  Hz), 12.73 br.s (1H, NH). Found, %: C 65.96; H 3.75; N 10.26; S 15.90.  $\text{C}_{22}\text{H}_{15}\text{N}_3\text{OS}_2$ . Calculated, %: C 65.81; H 3.77; N 10.47; S 15.97.

**1-Acetylimino-4-oxo-3-phenyl-4,5-dihydrothiazolo[3,4-*a*]quinoxaline (IX).** *a.* A solution of 1.00 g of isothioureide **VI** in 20 ml of acetic acid was heated for 25 h under reflux. The mixture was cooled, and the precipitate was filtered off and washed with 2-propanol. Yield 0.12 g (11%), mp 341–343°C.

*b.* A solution of 1.00 g of isothioureide **VI** in 10 ml of acetic anhydride was heated for 1 min under reflux (after 20 s, crystals began to separate from the solution). The mixture was cooled, and the precipitate

was filtered off and washed with 2-propanol. Yield 1.08 g (95%).

*c.* A solution of 0.30 g of quinoxaline **V** and 0.09 g of thiourea in 5 ml of dioxane was heated for 0.5 h under reflux, 2 ml of acetic anhydride and 2 ml of acetic acid were added, and the mixture was refluxed for 0.5 h. It was then cooled, and the precipitate was filtered off, washed with a solution of sodium carbonate, water, and 2-propanol. Yield 0.11 g (35%).

*d.* A solution of 0.3 g of quinoxaline **V** and 0.16 g of *N*-phenylthiourea in 5 ml of dioxane was heated for 0.5 h under reflux, 2 ml of acetic anhydride and 2 ml of acetic acid were added, and the mixture was refluxed for 0.5 h. It was then cooled, and the precipitate was filtered off, washed with a solution of sodium carbonate, water, and 2-propanol. Yield 32 mg (10%).

*e.* A solution of 0.9 g of compound **XV** in 5 ml of acetic acid was added to a mixture of 0.41 g of potassium thiocyanate, 0.45 g of *o*-phenylenediamine and 10 ml of acetic acid. The mixture was heated for 2 h under reflux and left overnight. The precipitate was filtered off and washed with 2-propanol. Yield 0.11 g (8%).

*f.* A solution of 0.7 g of chloropyruvate **XIV** in 5 ml of acetic acid was added to a mixture of 0.35 g of *o*-phenylenediamine and 10 ml of acetic acid, and, after 15 min, 0.32 g of potassium thiocyanate was added. The mixture was heated for 2 h under reflux and left overnight. The precipitate was filtered off and washed with 2-propanol. Yield 20 mg (2%). The physical constants of compound **IX** obtained by the above procedures (*a–f*) were identical to those reported in [8].

**3-[ $\alpha$ -(1-Acetoxy-2-benzimidazolyl)benzyl]-1,2-dihydroquinoxalin-2-one (XII).** A solution of 0.2 g of quinoxaline **VII** in a mixture of 2 ml of acetic anhydride and 2 ml of acetic acid was heated for 1 min under reflux and left overnight. The precipitate was filtered off and washed with 2-propanol. Yield 0.21 g (95%), mp 252–254°C (from DMSO). IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 1620 (C=N), 1670 (C=O, lactam), 1715 (C=O, acetyl), 2380–3220 (NH).  $^1\text{H}$  NMR spectrum (DMSO- $d_6$ ),  $\delta$ , ppm: 2.81 s (3H,  $\text{CH}_3$ ), 6.97 s (1H, PhCH), 7.25–7.39 m (7H,  $\text{H}_{\text{arom}}$ ), 7.48–7.59 m (2H,  $\text{H}_{\text{arom}}$ ), 7.67 d (2H,  $\text{H}_{\text{arom}}$ ,  $J = 6.90$  Hz), 7.72–7.78 m (1H,  $\text{H}_{\text{arom}}$ ), 7.84 d (1H,  $\text{H}_{\text{arom}}$ ,  $J = 7.34$  Hz), 12.47 br.s (1H, NH). Found, %: C 67.46; H 4.08; N 13.36; S 7.67.  $\text{C}_{24}\text{H}_{18}\text{N}_3\text{O}_2\text{S}$ . Calculated, %: C 67.59; H 4.25; N 13.14; S 7.52.

**3-Benzyl-1,2-dihydroquinoxalin-2-one (XIII).** A solution of 1.55 g of compound **I** and 0.48 g of

thiourea in 15 ml of dioxane was heated for 4 h under reflux. The solution was separated from the tarry material, 7 ml of dioxane was added to the latter, the mixture was heated to the boiling point, and the remaining tarry material was separated. The dioxane solution was cooled and left overnight. The precipitate was filtered off and washed with a solution of sodium carbonate, water, and 2-propanol. Yield 1.05 g (78%), mp 190–192°C (from dioxane). IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 1610 (C=N), 1660 (C=O), 2520–3220 (NH).  $^1\text{H}$  NMR spectrum (DMSO- $d_6$ ),  $\delta$ , ppm: 4.39 s (2H, PhCH<sub>2</sub>), 7.19–7.39 m (7H, C<sub>6</sub>H<sub>5</sub>, 6-H or 7-H, 8-H), 7.48 d.d (1H, 7-H or 6-H,  $J = 7.50, 7.50$  Hz), 7.77 d (1H, 5-H,  $J = 8.00$  Hz), 12.45 br.s (1H, NH). Satisfactory analytical data were obtained after repeated recrystallizations.

**Methyl 2-hydroxy-2-(4-oxo-3-phenyl-4,5-dihydro-1H-thiazolo[3,4-a]quinoxalin-1-ylideneamino)-5-phenyl-2,3-dihydrothiazole-4-carboxylate (XV).** A solution of 1.20 g of KSCN in 5 ml of acetonitrile was added to a mixture of 2.50 g of chloropyruvate **XIV** and 3 ml of acetonitrile. The mixture was stirred for 6 h and left overnight. The precipitate of inorganic salts was filtered off, a solution of 1.27 g of *o*-phenylenediamine in 10 ml of acetic was added to the filtrate, and the mixture was stirred for 1 h under reflux. It was then cooled, and the precipitate was filtered off and washed with 2-propanol. Yield 0.40 g (8%), mp 339–341°C (DMSO). IR spectrum,  $\nu$ ,  $\text{cm}^{-1}$ : 1672 (C=O, amide), 1722 (C=O, ester), 2650–3220 (NH, OH).  $^1\text{H}$  NMR spectrum (DMSO- $d_6$ ),  $\delta$ , ppm: 3.37 s (3H, CH<sub>3</sub>), 7.28–7.65 m (15H, 2C<sub>6</sub>H<sub>5</sub>, 6-H, 7-H, 8-H, NH, OH), 9.74 d (1H, 9-H,  $J = 8.5$  Hz), 11.52 br.s (1H, NH, lactam). Mass spectrum,  $m/z$  ( $I_{\text{rel}}$ , %): 529 (10), 528 (30), 510 (25), 543 (24), 452 (55), 350 (11), 320 (33), 293 (42), 266 (100), 237 (11), 236 (68), 235 (39), 234 (40), 206 (26), 205 (31), 121 (67), 105 (18), 89 (12), 77 (19). Found, %: C 65.96; H 3.75; N 10.26; S 15.90. C<sub>25</sub>H<sub>17</sub>N<sub>4</sub>O<sub>4</sub>S<sub>2</sub>. Calculated, %: C 65.81; H 3.77; N 10.47; S 15.97.

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