REACTIONS FORMING AND RECYCLIZING HETEROCYCLES

II.* REACTION OF AZOLIDINEDIONES WITH HYDRAZINE

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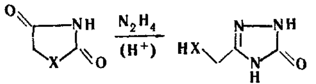
It has been shown that 1,3-thiazolidine-2,4-dione and 1,3-oxazolidine-2,4-dione react with hydrazine with a rearrangement of the ring to form a triazolinone ring. In each case, the reaction begins with the formation of an ionic adduct of hydrazine with the azolidinedione. On heating, the adduct undergoes decyclization (water catalyzes the process). The further course of the reaction is governed by the temperature and pH of the medium, which determine whether dehydration of the intermediate product with the formation of a triazolinone ring (in acid media) or rearrangement into a 1-acylsemicarbazide (at elevated temperatures) predominates. The influence of substituents in the thiazolidinone ring on the course of the reaction has been studied.

An account of the competing cyclization of diacylhydrazines has been given previously [1]. The present paper reports investigations of a reaction discovered by us – the rearrangement of thia- and oxazolidinedione rings (I) into the triazolinone ring (II) [2] (recyclization reaction) under the action of hydrazine:



Ia, b IIa, b

I, II a X = S, b X = O

The structure of compounds II follows from their elementary compositions and chemical properties and has been shown by the independent synthesis of IIa from isorhodanine and hydrazine [2], and it is also confirmed by the physicochemical properties of the latter – in particular its IR spectra, in which the sulfhydryl group is represented by a narrow medium-intensity absorption band at 2575 cm^{-1}. The same group is revealed by the reaction with sodium nitroprusside. Compounds II do not give reaction for a primary hydrazine group with Fehling's reagent and do not form condensation products with aldehydes in an ethanolic medium, which shows the absence of free amino groups, and they dissolve in alkalies, being reprecipitated by acids.

The recyclization of Ib takes place more readily than that of Ia and can be performed even without mineral acids.

When the reaction of the azolidinediones with hydrazine is carried out below 0°C, it is possible to isolate compounds to which, from their chemical and physicochemical properties and elementary compositions, it is possible to ascribe the structure of ionic adducts of the corresponding azolidinedione with hydrazine (III, see scheme). Thus, in the action of benzaldehyde on III in neutral medium at room temperature the hydrazine is eliminated almost quantitatively in the form of benzylidene azine. Compounds III give a positive reaction for a hydrazine group with Fehling's reagent (accompanied by the evolution of nitrogen) and they possess the high electrical conductivity characteristic for organic salts and a high solubility in water and insolubility in organic solvents. On acidification, the corresponding azolidinedione is recovered.

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*For Communication I, see [1].

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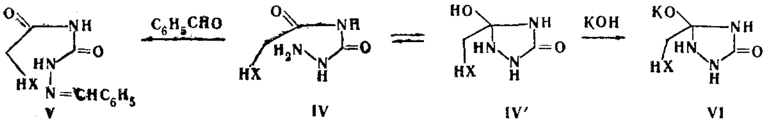
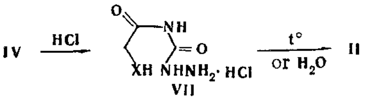
When the temperature is raised to that of the room in the presence of water, compounds III are converted into the isomeric compounds IV. Since in anhydrous solvents this transformation takes place very slowly even on prolonged heating, it may be assumed that water catalyzes it. The electrical conductivity of compounds IV, in contrast to III, is very low. Their properties are obviously connected with the ring-chain tautomerism IV ⇌ IV'. The existence of the form IV, possessing a primary hydrazine group, is confirmed by the quantitative formation of N-benzylidene derivatives V, even at room temperature.

However, Fehling's reagent (i.e., in an alkaline medium) does not detect the hydrazine group. This may show that the hydrazine group is present in the bound state in an alkaline medium, i.e., the cyclic form IV' is stabilized by the formation of a salt VI, and the equilibrium is shifted completely in its direction. The potassium salt of this form (VIa) has actually been isolated from an ethanolic solution of KOH; on careful neutralization with acetic acid the initial compound reprecipitated.



In absolute ethanol containing hydrogen chloride, compounds IV form the stable insoluble chlorides VII, i.e., the acyclic form is stabilized by salt formation at the hydrazine group. On being heated in anhydrous solvents, the latter are slowly converted into II, obviously because of solvolysis.



In an aqueous medium, these salts (just like IV on acidification in the presence of water) are converted very rapidly into II. In the absence of acids the heating of IVa also forms a certain amount of IIa – the acid is a catalyst of the process.

In an aqueous medium at room temperature, compounds IV gradually hydrolyze to glycolic acids and semicarbazide.

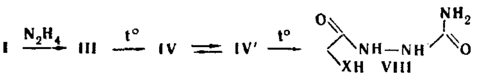
$$\text{IV} \xrightarrow{\text{HOH}} \text{HX—CH}_2\text{—COOH} + \text{H}_2\text{N—CO—NHNH}_2$$

The IR spectra of IVa clearly show the absorption band of a sulfhydryl group at 2495 cm^{-1}.

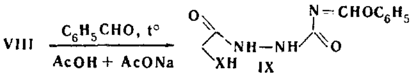
The course of the recyclization reaction of the azolidinediones under the action of hydrazine hydrate may therefore be represented by the following scheme:

$$\text{I} \xrightarrow{N_2H_4} \underset{\text{III}}{\text{I}\cdot N_2H_4} \xrightarrow{t^\circ} \text{IV} \rightleftharpoons \text{IV}' \xrightarrow[-H_2O]{(H^+)} \text{II}$$

When the temperature is raised the reaction of the azolidinediones with hydrazine takes place predominantly with the formation of 1-acylsemicarbazides (VIII), into which III and IV are likewise converted on heating. These transformations represent a new rearrangement (a semicarbazide rearrangement) which is alternative to recyclization and takes place on heating both in organic solvents and in water.



From their elementary compositions, compounds VIII are isomers of IV, but differ in properties. Like compound IV, they do not react with Fehling's solution, but their benzylidene derivatives are formed under more severe conditions, which may show the absence of a primary hydrazine group.



Compounds VIII do not undergo further cyclization under the conditions of the conversion of IV into II. On being boiled in acetic acid, compounds VIII are converted into dicarbamoylhydrazine, while compounds IV form triazolinones; X is obtained from semicarbazide under the same conditions.

$$\mathrm{VIII}\ \xrightarrow[\mathrm{AcOH}]{t^\circ}\ \underset{\mathrm{X}}{\mathrm{H_2N{-}CO{-}NHNH{-}CO{-}NH_2}}\ \xleftarrow[\mathrm{AcOH}]{t^\circ}\ \mathrm{2\,H_2N{-}CO{-}NHNH_2}$$

The structure of VIII has also been shown by independent synthesis from the fragments.

$$\mathrm{HX{-}CH_2COOH + H_2NNH{-}CO{-}NH_2 \longrightarrow VIII}$$

The IR spectra of VIIIa have the band of a sulfhydryl group at 2580 $\mathrm{cm^{-1}}$.

It must be mentioned that the synthesis of 4-mercaptoacetylsemicarbazide reported by Wella is not correct. The synthetic product with mp 132°C mentioned by this author is actually its isomer VIIIa: the assumed product should have melted at 120°C (see Experimental).

Alkyl substituents in position 5 of 1,3-thiazolidine-2,4-dione direct the process in an ethanolic medium to the formation of rearrangement products and in water to decomposition into substituted thioglycolic acids and semicarbazide. The decomposition of the ring to thioglycolic acid and the corresponding semicarbazide derivative both in ethanolic (slow) and in aqueous (very fast) media is favored by substituents in position 3, in analogy with previous work [4].

It follows from what has been said above that the reaction of the azolidinediones with hydrazine can take place in the direction either of recyclization or of rearrangement: the direction of the reaction is determined by the temperature of the process and the pH of the medium mainly at the stage of the transformation of IV' by dehydration into triazolinones (in an acid medium) or by rearrangement into 1-acylsemicarbazides (at elevated temperature). From this, the features of the course of the reaction with substituted azolidinediones also become understandable. Since substituents in position 5 stabilize the ring, to perform decyclization (the stage of the formation of IV) a higher temperature is necessary or else the starting material is recovered. But under these conditions rearrangement also takes place, and the intermediate compounds can no longer be isolated. The course of the reaction when substituents are present in position 3 can be explained analogously, but in this case it appears that the hydrolytic decomposition of IV takes place more rapidly.

EXPERIMENTAL

The infrared spectra of the compounds were recorded on a UR-10 double-beam instrument (Zeiss) in tablets with potassium bromide. The specific electrical conductivities* were determined at a current frequency of 1 kHz.

Adduct of Hydrazine with 1,3-Thiazolidine-2,4-dione (IIIa). A suspension of 11.7 g (0.1 mole) of 1,3-thiazolidine-2,4-dione (Ia) in 30 ml of methanol was cooled to −10°C and, with stirring, a cooled solution of 5 ml (0.1 mole) of hydrazine hydrate in 10 ml of methanol was added. After 20 min, the white precipitate that had formed was filtered off. Yield quantitative. mp about 60°C. The reaction with Fehling's reagent for a hydrazine group was positive (accompanied by the evolution of nitrogen); with benzaldehyde benzylidene azine with mp 92°C was formed. On careful neutralization with acetic acid, the initial Ia was recovered. Compound IIIa is readily soluble in water and less readily in ethanol and is insoluble in other organic solvents. The specific electrical conductivity at 25°C of a 0.1 M solution of IIIa in methanol $\varkappa = 2.28 \cdot 10^{-3}$ $\mathrm{ohm^{-1}/cm^{-1}}$, i.e., of the order of the electrical conductivity of organic salts and considerably exceeding the conductivity (under the same conditions) of the components – thiazolidinedione $9.3 \cdot 10^{-5}$ $\mathrm{ohm^{-1}/cm^{-1}}$ and hydrazine hydrate $2.13 \cdot 10^{-5}$ $\mathrm{ohm^{-1}/cm^{-1}}$. Found %: N 27.94; S 21.20. $\mathrm{C_3H_7N_3O_2S}$. Calculated %: N 28.13; S 21.47.

4-Mercaptoacetylsemicarbazide (IVa). a. With stirring, 2.5 ml (0.05 mole) of hydrazine hydrate was added in small portions to a suspension of 5.85 g (0.05 mole) of 1,3-thiazolidine-2,4-dione in 25 ml of methanol. After an hour, the resulting homogeneous solution deposited a precipitate. After standing for 3 h, it was filtered off and was washed with methanol and ether. Yield 4.5 g (61%), mp 120°C (decomp.; the

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*The authors express their gratitude to O. I. Kachurin for the help given in the electrical conductivity measurements.

capillary was introduced close to the melting point). White lamellar crystals, soluble in dimethylformamide and dimethyl sulfoxide, sparingly soluble in water, insoluble in the usual organic solvents. The reaction with Fehling's reagent for a primary hydrazine group is negative. Found %: C 24.30; H 4.95; N 28.33; S 21.61. $C_3H_7N_3O_2S$. Calculated %: C 24.16; H 4.69; N 28.18; S 21.47.

In water, IVa decomposes to thioglycolic acid (which can be detected by smell after only a few minutes) and semicarbazide, identified in the form of benzaldehyde semicarbazone, mp 222°C.

b. At room temperature in ethanol (in the presence of water), IIIa is converted almost quantitatively into IVa, mp 120°C, giving no depression of the melting point with the preceding sample.

Benzaldehyde 4-Mercaptoacetylsemicarbazone (Va). To a suspension of 0.75 g (0.005 mole) of IVa in 10 ml of methanol was added 0.53 ml (0.005 mole) of benzaldehyde. The yellow crystalline precipitate obtained as a result of the reaction was filtered off and washed with ethanol and ether. Yield 1.11 g (94%). Colorless crystals insoluble in water and soluble on heating in alcohols and dioxane, mp 151°C (from ethanol). Found %: N 17.55; S 13.80. $C_{10}H_{11}N_3O_2S$. Calculated %: N 17.72; S 13.50.

Potassium Salt of 5-Hydroxy-5-mercaptomethyl-1,2,4-triazolidin-3-one (VIa). One gram (0.0067 mole) of IVa was added to a solution of 0.4 g (0.007 mole) of caustic potash in 5 ml of methanol. First the substance dissolved, and then a voluminous white precipitate deposited, which was filtered off and washed with methanol. Yield 1.1 g (87%). mp 175-180°C. It did not give the reaction for a hydrazine group with Fehling's reagent. It deliquesced in the air. On treatment with an equimolar amount of acetic acid in methanol, the starting material reprecipitated. Found %: N 22.08; S 16.81. $C_3H_6KN_3O_2S$. Calculated %: N 22.34; S 17.02.

Hydrochloride of 4-Mercaptoacetylsemicarbazide (VIIa). 0.75 g (0.005 mole) of IVa was treated with 5 ml of concentrated methanolic hydrogen chloride. The resulting product was filtered off and washed with methanol and ether. Yield 0.73 g (79%), colorless crystals, mp 215°C. Found %: C 19.63; H 4.59; Cl 18.92; S 17.05. $C_3H_8ClN_3O_2S$. Calculated %: C 19.41; H 4.31; Cl 19.14; S 17.25.

The treatment of VIIa with an equimolecular amount of hydrazine hydrate in ethanol led to the precipitation of the starting material, yield 95%, mp 120°C.

5-Mercaptomethyl-1,2,4-triazolin-3-one (IIa). a. With stirring, 2.5 ml (0.05 mole) of hydrazine hydrate was added to a suspension of 5.8 g (0.05 mole) of 1,3-thiazolidin-2,4-dione in 20 ml of methanol. A homogeneous solution was first formed, but after an hour this crystallized. After 3 h, 40 ml of a 1N solution of hydrochloric acid (to give pH ~4) was rapidly added with stirring. A homogeneous solution was again formed from which a white crystalline precipitate deposited, and this was filtered off and was washed with water, ethanol, and ether. Yield 3.9 g (60%). mp 218°C (from water), giving no depression of the melting point with a sample obtained by the method described previously [2]. It dissolves in dimethylformamide and dimethyl sulfoxide and, with more difficulty, in water, and it is insoluble in alcohols, dioxane, and benzene. Fehling's reaction for a hydrazine group is negative; it does not react with benzaldehyde in ethanol at room temperature, and with sodium nitroprusside in the presence of sodium carbonate, unlike thiazolidinedione, it gives a crimson coloration. Found %: C 27.40; H 4.04; N 32.12; S 24.68. $C_3H_5N_3OS$. Calculated %: C 27.48; H 3.81; N 32.06; S 24.42.

b. 7.45 g (0.05 mole) of IVa was added to 50 ml of 1N aqueous hydrochloric acid and the mixture was stirred until dissolution was complete. The white crystalline precipitate that deposited after 20 min was filtered off and was washed with water, ethanol, and ether. Yield 6.1 g (93%). mp 218°C (from water).

c. The action of water on VIIa first formed a solution, which then deposited IIa, mp 218°C.

d. A suspension of 0.93 g (0.005 mole) of VIIa in 10 ml of dioxane was boiled for an hour, after which the insoluble residue (0.85 g) was filtered off and suspended in 5 ml of methanol, and the suspension was neutralized with 0.25 ml of hydrazine hydrate. This gave 0.25 g (33%) of a white lamellar precipitate of 1-mercaptoacetylsemicarbazide, mp 132°C showing no depression of the melting point with an authentic sample. Neutralization of the mother liquor precipitated 0.3 g (46%) of IIa, mp 218°C, giving no depression of the melting point with either of the above-obtained samples of IIa.

1-Mercaptoacetylsemicarbazide (VIIIa). a. A suspension of 5.85 g (0.05 mole) of 1,3-thiazolidine-2,4-dione in 10 ml of methanol was treated with 2.5 ml (0.05 mole) of hydrazine hydrate, and the resulting solution was boiled for 30 min. The precipitate that deposited on cooling was filtered off and was washed with methanol and ether. Yield 4.55 g. The product obtained was dissolved in 20 ml of ethanol and the undissolved residue was filtered off; it consisted of IIa [0.75 g (12%), mp 218°C]. The filtrate deposited a voluminous precipitate of VIIIa. Yield 3.7 g (50%). White crystals readily soluble in water, more sparingly in ethanol, mp 132°C (from ethanol). Fehling's reaction for a hydrazine group was negative. Found %: C 24.27; H 4.81; N 28.50; S 21.26. $C_3H_7N_3O_2S$. Calculated %: C 24.16; H 4.69; N 28.28; S 21.47.

b. A mixture of 2.98 g (0.02 mole) of IIIa in 10 ml of absolute ethanol was boiled in a flask with a reflux condenser fitted with a calcium chloride tube for 20 min. A homogeneous solution was formed. After the ethanol had been distilled off in vacuum, the residue was treated with ether. A white crystalline precipitate deposited, which was filtered off and washed with methanol and ether. Yield 2.1 g. On recrystallization from ethanol, 0.1 g of undissolved IIa was separated off, after which 1.9 g (61%) of VIIIa, mp 131°C was obtained.

c. A mixture of 1.49 g (0.01 mole) of IVa and 10 ml of water was boiled for 10 min and was then evaporated on the water bath. The residue crystallized on treatment with methanol. The crystals were washed with ethanol, yield 1.05 g. On crystallization from ethanol, traces of insoluble IIa were separated off, after which 1.0 g (67%) of VIIIa, mp 131°C (from ethanol) was obtained.

The methanolic filtrate obtained first was treated with 0.5 ml of benzaldehyde. A yellow precipitate of benzylidine azine deposited; yield 0.3 g (15%), mp 92°C. The mother liquor was treated with water, whereupon a white precipitate of benzaldehyde semicarbazone deposited; yield 0.25 g (15%), mp 220°C (from methanol).

d. A suspension of 1.49 g (0.01 mole) of IVa in 10 ml of absolute tert-butanol in the presence of 0.2 ml (0.02 mole) of piperidine was boiled with stirring in a flask with a reflux condenser fitted with a calcium chloride tube for 40 min. After cooling, a white pulverulent precipitate of VIIIa was filtered off. Yield 1.2 g (80%), mp 130-132°C.

e. A mixture of 3.75 g (0.05 mole) of semicarbazide and 3.5 ml (0.05 mole) of thioglycolic acid in 10 ml of ethanol was boiled for 2 h. The white precipitate that deposited after cooling was washed with ethanol and ether. Yield 5.6 g (75%), mp 132°C (from ethanol), mixtures with the preceding samples giving no depression of the melting point.

N-Benzylidene-1-mercaptoacetylsemicarbazide (IX). A solution of 1.49 g (0.01 mole) of VIIIa in 15 ml of glacial acetic acid was treated with 0.5 g of sodium acetate and 1.06 ml (0.01 mole) of benzaldehyde. The mixture was boiled for 30 min and cooled, and the precipitate that deposited was filtered off and washed with water, ethanol, and ether. Yield 1.5 g (63%), mp 249°C. Found %: N 17.73; S 13.40. $C_{10}H_{11}N_3O_2S$. Calculated %: N 17.72; S 13.50.

1,2-Dicarbamoylhydrazine (X). A mixture of 0.005 mole of VIIIa or b and glacial acetic acid was boiled for 2 h (VIIIa) or 6 h (VIIIb). The white precipitate that deposited was filtered off; yield 70%; mp 248°C (from water) (247°C [5]).

The same substance was obtained on boiling the semicarbazide in acetic acid.

1-α-Mercaptobutyroylsemicarbazide. A mixture of 0.7 g (0.005 mole) of 5-ethyl-1,3-thiazolidine-2,4-dione and 0.25 ml (0.005 mole) of hydrazine hydrate in 2 ml of methanol was boiled for 1 h, after which the methanol was evaporated off on the water bath. The resulting oily product was treated with dichloroethane and the precipitate formed was filtered off; yield 0.5 g (28%). White acicular crystals, mp 158-160°C (from ethanol). Fehling's reaction for a hydrazine group was negative, and the substance did not react with benzaldehyde in ethanol. Found %: C 34.01; H 6.44; N 23.77; S 17.81. $C_5H_{11}N_3O_2S$. Calculated %: C 33.89; H 6.21; N 23.73; S 18.08.

At room temperature the reaction of 5-ethyl-1,3-thiazolidine-2,4-dione with hydrazine hydrate did not take place either in ethanol or water, and the starting materials were recovered (the hydrazine in the form of benzylidene azine).

4-Phenylsemicarbazide. a. A mixture of 1.93 g (0.01 mole) of N-phenyl-1,3-thiazolidine-2,4-dione and 10 ml of 25% hydrazine hydrate (0.05 mole) was brought to the boil, forming a homogeneous solution from which, after cooling, a white crystalline precipitate of 4-phenylsemicarbazide deposited; yield 1.1 g (73%), mp 122°C, giving no depression of the melting point with an authentic sample.

b. A mixture of 1.93 g (0.01 mole) of N-phenyl-1,3-thiazolidine-2,4-dione, 0.6 g (0.012 mole) of hydrazine hydrate, and 10 ml of n-propanol was boiled for 5 h. The precipitate that deposited was filtered off and washed with hot methanol and ether. Yield 0.9 g (66%). White crystalline substance, mp 280°C, consisting of 1,2-bis(n-phenylcarbamoyl)hydrazine. The mother liquor deposited 0.4 g (20%) of unchanged N-phenyl-1,3-thiazolidine-2,4-dione, mp 144°C. From the filtrate, benzaldehyde precipitated 0.3 g (12%) of white acicular crystals of benzaldehyde 4-phenylsemicarbazone, mp 170-172°C.

4-Hydroxyacetylsemicarbazide (IVb). At −10°C, 0.25 ml (0.005 mole) of hydrazine hydrate was added to a suspension of 0.5 g (0.005 mole) of 1,3-oxazolidine-2,4-dione in 1 ml of methanol. An oily liquid deposited which apparently consisted of an adduct of hydrazine with 1,3-oxazolidine-2,4-dione: Fehling's reaction for a hydrazine group was positive; on acidification, the initial Ib with mp 89-90°C deposited, and benzaldehyde yielded benzylidene azine.

The mixture was dissolved in 2 ml of methanol and the solution was kept at room temperature for 4 h. The white lamellar precipitate that deposited was washed with methanol and ether. Yield 0.3 g (47%), mp 121°C (decomp., from methanol). Found %: C 27.33; H 5.48; N 31.70. $C_3H_7N_3O_3$. Calculated %: C 27.07; H 5.26; N 31.58.

Compound IVb does not give the Fehling reaction for a primary hydrazine group, but with benzaldehyde in ethanol at room temperature it forms benzaldehyde 4-hydroxyacetylsemicarbazone, in almost quantitative yield, mp 188°C. Found %: C 54.40; H 5.21; N 18.84. $C_{10}H_{11}N_3O_3$. Calculated %: C 54.29; H 4.98; N 19.00.

5-Hydroxyethyl-1,2,4-triazolin-4-one (IIb). a. A solution of 0.5 g (0.005 mole) of 1,3-oxazolidine-2,4-dione in 1 ml of methanol was treated with 0.25 ml (0.05 mole) of hydrazine hydrate. The resulting solution was kept at room temperature for 4 h, acidified with 3 ml of 10% hydrochloric acid, and evaporated on the water bath until crystallization began. After cooling, the precipitate was filtered off and washed with methanol and ether. Yield 0.4 g (70%); colorless crystals, readily soluble in water, less readily in ethanol, and insoluble in ether, chloroform, and carbon tetrachloride. mp 187°C (from ethanol). Compound IIb does not give Fehling's reaction for a primary hydrazine group, and in an ethanolic medium it does not form a benzylidene derivative. Found %: C 31.17; H 4.62; N 36.52. $C_2H_5N_3O_2$. Calculated %: C 31.30; H 4.35; N 36.32.

b. A mixture of 0.5 g (0.005 mole) of 1,3-oxazolidine-2,4-dione and 0.25 ml (0.005 mole) of hydrazine hydrate in 3 ml of methanol was boiled for 30 min. The white precipitate that deposited after cooling was filtered off (the filtrate was kept for the following experiment); yield of IIb 0.2 g (35%), mp 185-187°C (from ethanol), giving no depression of the melting point with the preceding sample.

1-Hydroxyacetylsemicarbazide (VIIIb). a. After evaporation of the solvent at room temperature, the filtrate obtained in the preceding experiment deposited 0.25 g (38%) of a white crystalline substance, mp 149°C (from ethanol). Fehling's reaction for a primary hydrazine group was negative, and no benzylidene derivative was formed in ethanol. Found %: C 27.28; H 5.44; N 31.34. $C_3H_7N_3O_3$. Calculated %: C 27.07; H 5.26; N 31.58.

b. A mixture of 7.5 g (0.1 mole) of semicarbazide and 8 g (0.1 mole) of glycolic acid in 20 ml of methanol was boiled for an hour. The solvent was evaporated off on the water bath until the residue crystallized. Yield 11 g (63%). Colorless crystals, mp 149°C (from ethanol), giving no depression of the melting point with the preceding sample.

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