The reaction was complete after 10 h. The catalyst was removed by filtration and washed with hot alcohol. The filtrate was evaporated to give 5.0 g (55%) of a product with mp 194-196° (from ethyl acetate). IR spectrum, cm^{-1}: 1635 (CO), 3290 (NH). Found: C 71.0; H 5.24; N 13.4%. $C_{19}H_{17}N_3O_2$. . Calculated: C 71.4; H 5.4; N 13.2%.

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CYCLIZATION OF N-(4-QUINAZOLYL)-α-AMINO CARBOXYLIC ACIDS

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2,3-Dihydroimidazo[1,2-c]quinazolin-2-one derivatives were obtained by cyclization of N-(4-quinazolyl)-α-amino carboxylic acids. A scheme including cleavage of the $C_{(2)}$—$N_{(3)}$ bond of the quinazoline ring and subsequent rearrangement is proposed for the mechanism of the cyclization. The structures of the synthesized compounds were established by means of chemical and physicochemical methods.

Continuing our research on the synthesis of imidazo[1,2-c]quinazoline derivatives [1], we attempted to cyclize N-(4-quinazolyl)-α-amino carboxylic acids [2], which were previously obtained from 4-chloroquinazoline and α-aminocarboxylic acids. Heating the N-(4-quinazolyl)-α-amino carboxylic acids in glacial acetic acid or in a mixture of glacial acetic acid and acetic anhydride gave 2,3-dihydroimidazo[1,2-c]quinazolinones (I-V), the hydrolytic cleavage of which in alkaline media did not regenerate the starting acids. Ammonia was evolved as a result of hydrolysis of imidazoquinazolinone I, and we obtained α-4-quinazolon-3-yl)propionic acid (X), which was identical to the acid synthesized from 4-quinazolone and α-bromopropionic acid by the method in [3]. We suppose that the imidazolone ring is opened in the first step of the hydrolysis to give α-(4-imino-3-quinazolyl)propionic acid, which is readily converted to quinazolinone X. The above-indicated cyclization of N-(4-quinazolyl)-α-amino carboxylic acids consequently does not lead to the formation of the expected 2,3-dihydroimidazo[1,2-c]quinazolin-3-ones but rather to the isomeric 2,3-dihydroimidazo[1,2-c]quinazolin-2-ones (I-V). (See scheme on following page.)

The formation of I-V can be explained by assuming destruction of the $C_{(2)}$—$N_{(3)}$ bond of the pyrimidine ring of the quinoline two-ring system, as in the cyclization of N-(2-pyrimidon-4-yl)amino carboxylic acids [4]. (See scheme on following page.)

It is known that 4-aminopyrimidines [5] and their benzo analogs — quinazolines [6] — exist in the form of systems in dynamic equilibrium. In our case N-(4-quinazolyl)-α-amino carboxylic acids A exist in tautomeric equilibrium with structure B. Since acetic anhydride and glacial acetic acid are not only dehydrating agents but also acylating agents, there is no doubt regarding the possibility of acetylation of the $N_{(1)}$ atom (structure C). Protonation of the latter at the $N_{(3)}$ atom probably leads to systems D and E. Specific interaction

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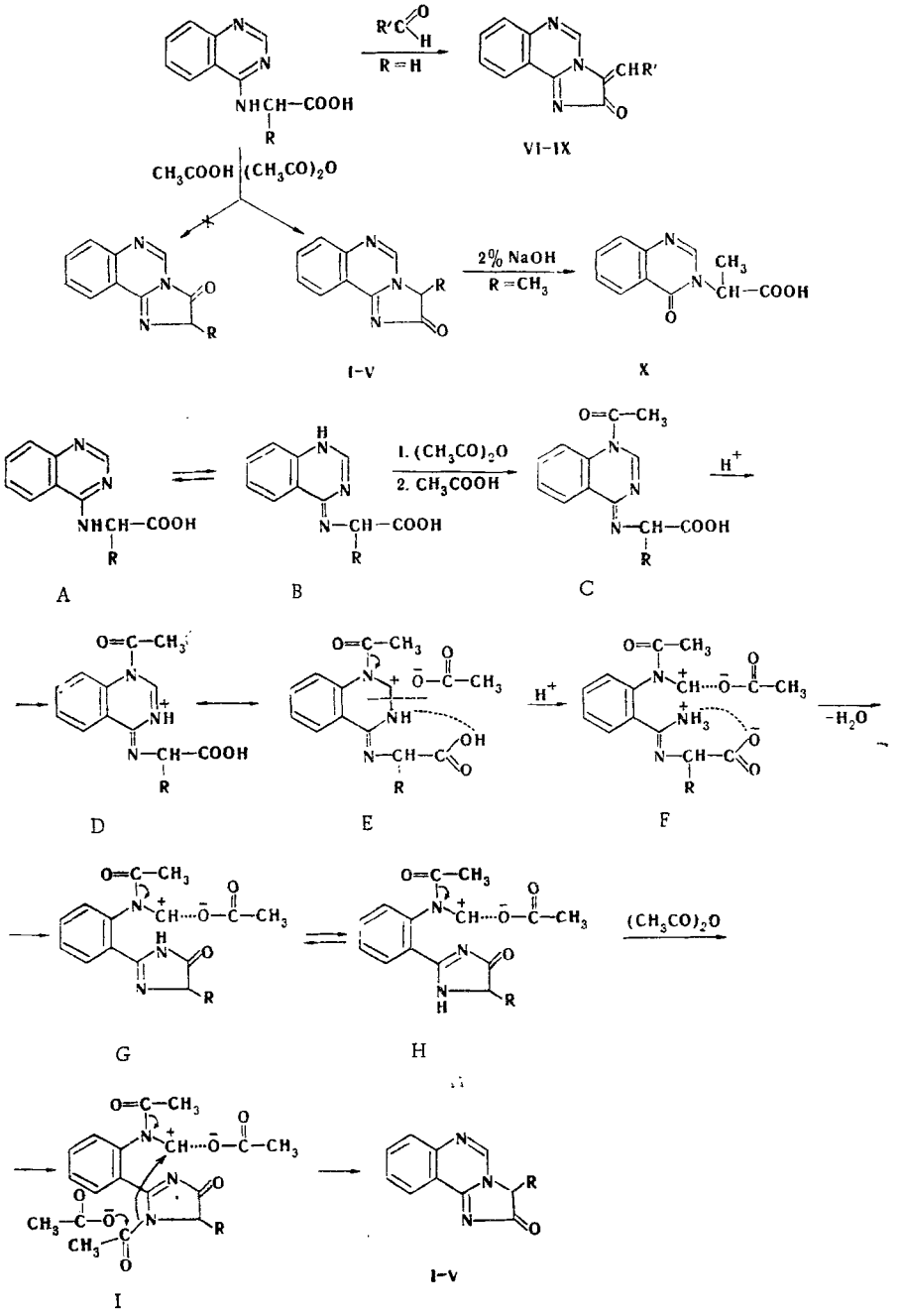

of the acetate anion with the $C_{(2)}$ atom and the probable formation of intermediate F may promote cleavage of the $C_{(2)}$-$N_{(3)}$ bond. Dehydration of intermediate F gives rise to imidazolone ring G in which migration of the active hydrogen from $N_{(1)}$ to $N_{(3)}$ and acetylation of H to give I are possible. In addition, attack on the acetate anion by the carbonyl carbon atom of the acetyl group gives rise to redistribution of the electron density over the entire system, ensuring the possibility of static approach of the $N_{(1)}$ atom of the imidazolone ring to the electron-deficient carbon atom. This conversion is accompanied by deacetylation and the formation of a three-ring system (I-V). The IR spectra of I-V contain distinct absorption bands of a CO group at 1650-1660 cm^{-1}, and absorption of the OH group is completely absent; this constitutes evidence for the existence of imidazolinoquinazolinones I-V in the oxo form.

Ylidene derivatives of 2,3-dihydroimidazo[1,2-c]quinazolin-2-one (VI-IX) were obtained by reaction of N-(4-quinazolyl)aminoacetic acid or its esters with aromatic and heterocyclic aldehydes in glacial acetic acid in the presence of anhydrous sodium acetate. The IR spectra of VI-IX contain characteristic bands of absorption of the CO group at 1670-1745

TABLE 1. 2,3-Dihydroimidazo[1,2-c]quinazolin-2-ones and Their Ylidene Derivatives

Compound	R or R'	mp, °C (dec.)	Empirical formula	Found, %			Calc., %			Yield, %
				C	H	N	C	H	N	
I	CH_3	238—240	$C_{11}H_9N_3O$	66,8	4,6	21,4	66,3	4,6	21,1	76
II	C_2H_5	144—146	$C_{12}H_{11}N_3O \cdot 0{,}5H_2O$	64,8	5,3	18,9	64,8	5,4	18,9	56
III	iso-C_4H_9	232—234	$C_{14}H_{15}N_3O$	69,4	6,6	17,6	69,7	6,3	17,4	66
IV	$CH_2CH_2SCH_3$	113—115	$C_{13}H_{13}N_3OS$*	54,7	5,6	14,6	54,5	5,6	14,7	34
V	3-Methylindolyl	148—150	$C_{19}H_{14}N_4O$	66,3	4,5	16,3	65,9	4,1	16,2	69
VI	p-HOC_6H_4	289—291	$C_{17}H_{11}N_3O_2$	70,7	4,0	14,9	70,6	3,8	14,5	53
VII	o-$CH_3OC_6H_4$	223—225	$C_{18}H_{13}N_3O_2$	71,5	4.2	13.9	71,3	4,3	13,9	36
VIII	p-$(C_2H_5)_2NC_6H_4$	221—223	$C_{21}H_{20}N_4O$	73.5	6,2	15,8	73,2	5,9	16,2	68
IX	5-Nitro-2-furyl	Above 350	$C_{15}H_8N_4O_4$	58,7	3,0	—	58,4	2.6	—	41

*Found: S 10.9%. Calculated: S 11.2%.

cm^{-1}; the spectrum of VI also contains a broad band of an associated OH group at 3100-3390 cm^{-1}.

EXPERIMENTAL

The IR spectra of mineral oil suspensions of the compounds were recorded with a UR-20 spectrometer.

2,3-Dihydroimidazo[1,2-c]quinazoline-2-ones (I-V, Table 1). A) A 0.01-mole sample of N-(4-quinazoyl)-α-amino carboxylic acid was heated in 15-20 ml of a mixture of glacial acetic acid and acetic anhydride (9:1) for 1 h (in the presence of activated charcoal in the last 5 min) after which the charcoal was removed by filtration, and the filtrate was evaporated to dryness on a water bath. Water (40-50 ml) was added to the residue, and the solid material was removed by filtration, washed with water, and dried.

B) A solution of 2.17 g (0.01 mole) of N-(4-quinazolyl)-α-aminopropionic acid in 25-30 ml of glacial acetic acid was refluxed for 1 h, after which it was worked up as in experiment A to give 1.4 g (70%) of I.

Compounds I-V were obtained as colorless crystalline substances that were only slightly soluble in organic solvents and insoluble in water. They were purified for analysis by crystallization from DMF (I and III), DMF-water (1:2) (II and IV), or DMF-water (1:3) (V).

α-(4-Quinazolon-3-yl)propionic Acid (X). A) A solution of 0.8 g (4 mmole) of 3-methyl-2,3-dihydroimidazo[1,2-c]quinazolin-2-one (I) in 15 ml of 2% sodium hydroxide solution was heated on a boiling-water bath for 1.5 h, after which it was cooled and acidified to pH 2-3 with 10% hydrochloric acid. The resulting precipitate was removed by filtration and washed with water to give 0.7 g (80%) of colorless crystals of acid X with mp 221-223° (dec., from water). Found: C 60.5; H 4.8; N 12.8%. $C_{11}H_{10}N_2O_3$. Calculated: C 60.5; H 4.6; N 12.8%.

B) A 2.92-g (0.02 mole) sample of 4-quinazolone and 3.06 g (0.02 mole) of α-bromopropionic acid were added to a solution of 1.6 g (0.04 mole) of sodium hydroxide in 20 ml of water, and the mixture was refluxed for 2 h. It was then cooled and acidified to pH 2-3 with a 10% solution of hydrochloric acid, and the resulting precipitate was removed by filtration and washed with water to give 3 g (66%) of product. No melting-point depression was observed for a mixture of this product with the compound obtained in experiment A.

Ylidene Derivatives of 2,3-Dihydroimidazo[1,2-c]quinazolin-2-one (VI-IX, Table 1). An equimolar amount of p-hydroxy-, o-methoxy-, or p-dimethylaminobenzaldehyde or 5-nitrofurfural and 0.03 mole of anhydrous sodium acetate were added to a solution of 0.01 mole of N-(4-quinazolyl)aminoacetic acid or its ester in glacial acetic acid, and the mixture was refluxed for 3 h. It was then cooled, and the resulting precipitate was removed by filtration, washed with water, and dried. The orange (VI), yellow (VII), red (VIII), or dark-brown (IX) crystalline products were insoluble in water and only slightly soluble in most organic solvents. They were purified for analysis by crystallization from dioxane (VI and VII) and butanol (VIII) or by reprecipitation from DMF by the addition of ether (IX).

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IR SPECTRA AND STRUCTURE OF 6-METHYLIMIDAZO[1,2-a]PYRIMIDINE-2, 5-DIONE AND ITS 3-YLIDENE DERIVATIVES

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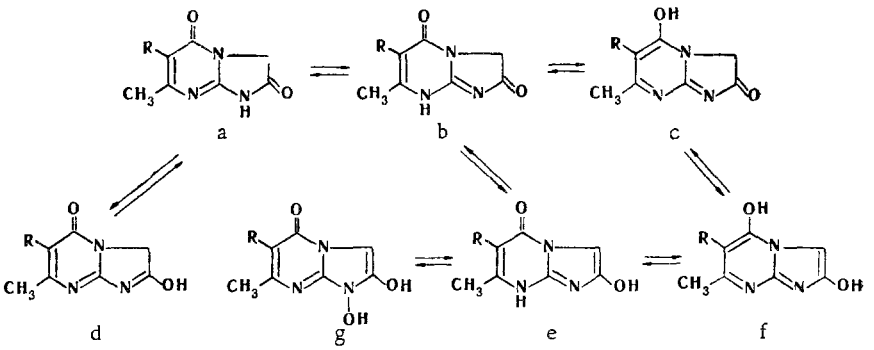
The structures of 6-R-7-methylimidazo[1,2-a]pyrimidine-2,5-diones and their 3-alkylidene and 3-arylidene derivatives were studied by IR spectroscopy. It was established that these compounds exist in the solid phase in the form of a mixture of tautomers. The most probable tautomers were identified. It is shown that the $\nu_{C=O}$ band of the imidazole fragment correlates with the σ parameters of the substituents in the arylidene grouping. The geometry of this grouping is discussed.

Recently, one of us described the synthesis of imidazo[1,2-a]pyrimidine-2,5-diones [1] and 3-ylidene derivatives based on them [2]. In the present communication we set out to discuss the results of a study of the structure of these compounds by means of IR spectroscopy. The spectra of KBr pellets of the compounds (2 mg of the compound in 100 mg of KBr) were measured with a UR-20 spectrometer at 700-3700 cm^{-1}. The results are presented in Table 1.

A primary feature of this heterocyclic system is the possibility of its existence in the form of several tautomeric forms:



The literature does not contain information regarding the ratio of these forms. It is asserted in [3] on the basis of data on the acidities, behavior with respect to solvents, and hydrolysis that the labile hydrogen atom is attached to the nitrogen atom in the imid-

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