

ing to give colorless needles with mp 31-32° (after slow sublimation). The picrate was obtained as light-yellow prisms with mp 171-172° (from alcohol). Found: C 51.2; H 2.9; N 19.6%.  $C_{12}H_9N_3 \cdot C_6H_3N_3O_7$ . Calculated: C 51.0; H 2.9; N 19.8%.

2-Mercapto-1-phenyl-1H-imidazo[4,5-c]pyridine. This compound was obtained in 90% yield from 3-amino-4-phenylaminopyridine [3] by the method used to prepare 2-mercapto derivative IIb. The product was obtained as prisms with mp 310° (from aqueous alcohol). Found: C 62.9; H 4.1; S 14.0%.  $C_{12}H_9N_3S$ . Calculated: C 63.4; H 4.0; S 14.1%.

1-Phenyl-1H-imidazo[4,5]pyridine (IIIc). This compound was obtained in 88% yield by oxidation of the corresponding mercapto derivative as in the preparation of IIc. The product was obtained as colorless needles with mp 105-106° (from benzene-hexane). Found: C 73.5; H 4.4; N 21.4%.  $C_{12}H_9N_3$ . Calculated: C 73.8; H 4.6; N 21.5%.

General Method for Hydroxylation of Imidazopyridines (Table 1). A mixture of 10 mmole of imidazopyridine and 50 mmole of anhydrous KOH or NaOH was heated at 150-190°. At the end of hydrogen evolution, the fused mixture was cooled and dissolved in 2-3 ml of water. The aqueous solution was acidified to pH 5-6 with hydrochloric acid, and the resulting precipitate was removed by filtration, washed with a small amount of cold water, dried, and crystallized from a suitable solvent.

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#### SYNTHESIS AND PROPERTIES OF SUBSTITUTED 2-IMIDAZOLIDINONES

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4-Hydroxy- and 4-hydroxylamino-1-hydroxy-3-aryl-5,5-dimethyl-2-imidazolidinones, respectively, were obtained by treatment of N-arylcarbamoyl derivatives of N-(1-oximino-2-methyl-2-propyl)hydroxylamine with acids and alkalis. 4-Hydroxylaminoimidazolidinones react with p-nitrobenzaldehyde to give nitrones and are converted in acidic media to 4-hydroxy derivatives, which by the action of methanol in the presence of p-toluenesulfonic acid give 4-methoxy-2-imidazolidinones. Acylation and alkylation of 4-hydroxy- and 4-methoxy-2-imidazolidinones take place at the hydroxy group attached to N<sub>(1)</sub>.

In the present research we have studied the behavior of the previously obtained N-arylcarbamoyl derivatives of N-(1-oximino-2-methyl-2-propyl)hydroxylamine [1] (I) in acidic and alkaline media. It was established that dissolving of I in aqueous NaOH solution and subsequent neutralization lead to the isolation of white crystalline II, the results of the elementary analysis of which are in agreement with the data for starting I.

The absorption band of a carbonyl group at 1715 and 1720  $cm^{-1}$ , shifted to the high-frequency region by 50-60  $cm^{-1}$  as compared with the same band in the spectra of starting I,

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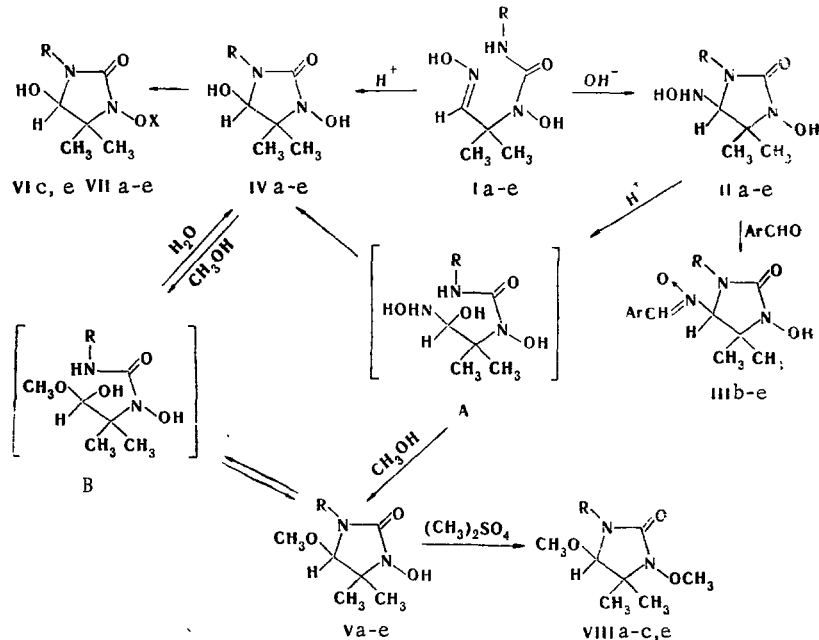
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is observed in the IR spectra of II; this is characteristic for conversion of the linear fragment of the hydroxyurea molecule to a cyclic fragment [2]. However, the practically complete coincidence of the UV spectra of II and the corresponding N-aryl-N'-hydroxyureas may serve as a confirmation of the invariability of the N-aryl-N'-hydroxyurea skeleton. For example, IIe and N-3,4-dichlorophenyl-N'-hydroxyurea have absorption maxima at 256 ( $\epsilon$  18,860) and 248 nm ( $\epsilon$  19,900), respectively. Compounds II react with 2,3,5-triphenyltetrazole to give bright-red complexes characteristic for the hydroxylamino group. On the basis of these data, the 4-hydroxylamino-1-hydroxy-3-aryl-5,5-dimethyl-2-imidazolidinone structure was assigned to II.

In fact, in addition to two singlets of a gem-dimethyl grouping at 1.1-1.29 ppm, signals of protons of an aryl substituent at 7.0-7.7 ppm, and a singlet of the protons of the OH group (8.5-9.0 ppm) in the 1 position of the heteroring, two doublets of the protons of the >CHNH fragment in the 4 position at 4.66-4.70 and 5.92-6.42 ppm ( $J$  = 3.5-4.5 Hz) and, finally the singlet of the proton of the OH group of a hydroxylamino group at 7.3-7.5 ppm are observed in the PMR spectra of solutions of II in hexadeuterodimethyl sulfoxide. It should be noted that the signal of the 4-H proton appears as a singlet in the PMR spectra of solutions of II in deuteroacetonitrile. However, the absence of signals of the protons of the NOH and NHOH groups is evidently associated with their volume [3].

A chemical confirmation of the structure of II was provided by their condensation [4] with p-nitrobenzaldehyde, which leads to N-(1-hydroxy-3-aryl-5,5-dimethyl-2-imidazolidonene-4-yl)-p-nitrophenylnitrones (III). The presence in the IR spectra of III of intense absorption bands at 1110 and 1605  $\text{cm}^{-1}$  and of a band at 349 nm ( $\epsilon$  10,110) in the UV spectra constitutes evidence for the presence of a p-nitrophenylnitron grouping in the molecule [4, 5].

We were unable to find information in the literature regarding cyclizations that would include intramolecular nucleophilic attack of the carbon atom of the oxime group leading to heterocyclic hydroxylamines. However, cyclization of this sort involving a ketone function leading to five- and six-membered hydroxy heterocycles is known [6-9]; ring-chain tautomerism, which, as shown in [10], depends on the temperature, polarity, and acid-base properties of the medium, was observed in the several cases in this cyclization.



I-VIII a  $R = \text{C}_6\text{H}_5$ ; b  $R = p\text{-CH}_3\text{C}_6\text{H}_4$ ; c  $R = p\text{-ClC}_6\text{H}_4$ ; d  $R = m\text{-ClC}_6\text{H}_4$ ; e  $R = 3,4\text{-Cl}_2\text{C}_6\text{H}_2$ .  
VIc, e  $X = \text{COC}_6\text{H}_5$ ; VIIa-e  $X = \text{CH}_3$

White crystalline IV with distinct melting points, which were insoluble in water and only slightly soluble in nonpolar organic solvents, were isolated when hydrochloric acid solutions of II were neutralized. According to the results of elementary analysis, IV correspond to products of replacement of the hydroxylamino group in II by a hydroxy group. The disappearance of the characteristic reaction for the NHOH group is a confirmation of this.

An intense band of carbonyl absorption at  $1700\text{--}1710\text{ cm}^{-1}$  is present in the IR spectra of IV, and their UV spectra are practically completely identical to the UV spectra of I and the corresponding N-aryl-N'-hydroxyureas. The PMR spectra of solutions of these compounds in  $(\text{CD}_3)_2\text{SO}$  contain two singlets of gem-dimethyl groups at 1.08–1.22 ppm, two doublets at 5.10–5.30 and 6.30–6.60 ppm ( $J = 6\text{--}8\text{ Hz}$ ) from the  $>\text{CHOH}$  group in the 4 position of the ring, a multiplet of protons of an aryl substituent attached to  $\text{N}_{(3)}$  at 7.15–7.65 ppm, and, finally, a singlet of the proton of the OH group in the 1 position of the ring at 9.0 ppm. The doublet at 6.30–6.60 ppm and the singlet related to the proton of the NOH group vanish when  $\text{D}_2\text{O}$  is added to the solution or when the spectrum of a solution in  $\text{CD}_3\text{CN}$  is recorded, and the doublet at 5.10–5.30 ppm becomes a singlet.

We were unable to observe the open form of II and IV by means of the IR and PMR spectra. However, it is known [10, 11] that some hydroxy heterocycles in methanol solution exchange a hydroxy group for a methoxy group, and this process is evidently accompanied by intermediate ring opening.

Compounds IV in methanol solution are converted to 4-methoxy derivatives V in the presence of catalytic amounts of p-toluenesulfonic acid, whereas II forms a mixture of IV and V in the presence of hydrochloric acid. These facts make it possible to assume that an open linear form (A or B) is also formed in our case during the transformation. It should be noted that the conversion of IV to V is reversible, and they are also readily converted entirely to IV when they are allowed to stand in acidic media; IV can also be obtained by direct treatment of I with hydrochloric acid. It is extremely likely that intermediate A, which leads to IV, is also formed during the transformation. However, one cannot exclude the possibility that I is initially deoximated to give its carbonyl analog, which subsequently is cyclized to IV.

The relative reactivities of the two hydroxy groups of IV were ascertained in the case of acylation and alkylation. Thus, white crystalline VI, which, according to the results of elementary analysis, correspond to the monobenzoyl derivatives of IV, are obtained in the reaction with benzoyl chloride under the conditions of the Schotten-Baumann reaction. The IR spectra of VI contain intense bands of carbonyl absorption at 1730–1745 and 1765–1770  $\text{cm}^{-1}$ , and bands of free and associated hydroxyl groups appear at 3580 and 3325–3330  $\text{cm}^{-1}$ , respectively (the band at 3325–3330  $\text{cm}^{-1}$  does not vanish on dilution). The PMR spectra of these compounds contain two doublets of protons of a  $>\text{CHOH}$  group at 5.37–5.48 and 7.0–7.1 ppm ( $J = 7.0\text{--}7.5\text{ Hz}$ ); the singlet of the proton of the OH group attached to  $\text{N}_{(1)}$  is absent. This makes it possible to assume that benzylation occurs at the hydroxy group attached to  $\text{N}_{(1)}$  and that VI have the 1-benzoyloxy-4-hydroxy-3-aryl-5,5-dimethyl-2-imidazolidinone structure. This is also confirmed by the absence, in the case of VI, of the positive color test with ferric chloride that is characteristic for hydroxamic acid fragments.

Methylation of IV and V with dimethyl sulfate in the presence of equimolar amounts of sodium hydroxide leads to 1-hydroxy-(VII) and 1,4-dimethoxy(VIII) derivatives. Intense bands of carbonyl absorption at 1700–1725 and 1720–1740  $\text{cm}^{-1}$ , are observed in the IR spectra of KBr pellets of VII and VIII, whereas the IR spectra of  $\text{CCl}_4$  solutions of VII contain the band of a free 4-OH group at 3585–3595  $\text{cm}^{-1}$ .

It is interesting to note that the IR spectra of  $\text{CCl}_4$  solutions of V contain, in addition to a band at 3560–3570  $\text{cm}^{-1}$  corresponding to a free OH group in the 1 position, a broad band of an associated OH group at 3230–3245  $\text{cm}^{-1}$ . The invariance of the bands of the stretching vibrations of the free and associated OH groups in the IR spectra of  $\text{CCl}_4$  solutions (0.01–0.001%) makes it possible to assume the presence of a hydroxy group tied up in an intramolecular hydrogen bond (IHB) for V, in addition to the presence of a free hydroxy group.

The shift of the band of the carbonyl absorption for IV–VII by 20–40  $\text{cm}^{-1}$  to the high-frequency region on passing from KBr pellets to solutions in dioxane and the practically complete coincidence of this band in the latter case with the corresponding band in the spectra of VIII, in which it is known that the carbonyl group does not participate in the formation of hydrogen bonds, constitute evidence that the carbonyl groups of IV–VII evidently participate in the formation of intermolecular hydrogen bonds but do not participate in the formation of intramolecular hydrogen bonds.

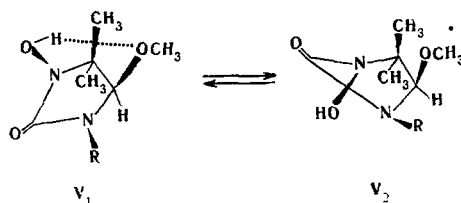
However, the presence of a free and intramolecularly bonded OH group in V can be explained by the existence of an equilibrium between two possible forms  $\text{V}_1$  and  $\text{V}_2$ , in which the OH and  $\text{OCH}_3$  groups are found either on the same side of the plane of the ring (intra-

TABLE 1. Results of Elementary Analysis, Melting Points, Yields, and Spectral Characteristics of 2-Imidazolidinones

Compound	mp, °C	Empirical formula	Found, %				Calc., %				IR spectrum, $\nu_{\text{CO}}$ , cm <sup>-1</sup>		Yield, %
			C	H	Cl	N	C	H	Cl	N	KBr	Dioxane	
IIa	145—147 <sup>a</sup>	C <sub>11</sub> H <sub>15</sub> N <sub>3</sub> O <sub>3</sub>	55.5	6.3	—	17.9	55.7	6.4	—	17.6	1720	—	67
IIb	102—104 <sup>a</sup>	C <sub>12</sub> H <sub>17</sub> N <sub>3</sub> O <sub>3</sub>	—	—	—	16.5	—	—	—	16.7	1715	—	73
IIc	148—150 <sup>b</sup>	C <sub>11</sub> H <sub>14</sub> ClN <sub>3</sub> O <sub>3</sub>	48.7	5.2	13.1	15.4	48.6	5.2	13.1	15.5	1720	—	69
IId	166—167 <sup>a</sup>	C <sub>11</sub> H <sub>14</sub> ClN <sub>3</sub> O <sub>3</sub>	48.6	5.1	13.2	16.0	48.6	5.2	13.1	15.5	1720	—	67
IIe	110—112 <sup>b</sup>	C <sub>11</sub> H <sub>13</sub> Cl <sub>2</sub> N <sub>3</sub> O <sub>3</sub>	43.4	5.1	—	13.9	43.2	5.1	—	13.7	1720	—	70
IIIf	211—213 <sup>c</sup>	C <sub>19</sub> H <sub>20</sub> N <sub>4</sub> O <sub>5</sub>	58.3	5.1	—	14.6	59.4	5.2	—	14.6	1735	—	81
IIIf	180—182 <sup>c</sup>	C <sub>18</sub> H <sub>17</sub> ClN <sub>4</sub> O <sub>5</sub>	53.5	4.5	8.7	13.8	53.4	4.2	8.8	13.8	1730	—	81
IIId	226—228 <sup>c</sup>	C <sub>18</sub> H <sub>17</sub> ClN <sub>4</sub> O <sub>5</sub>	52.6	4.2	8.7	13.5	53.4	4.2	8.8	13.8	1738	—	95
IIIf	210—212 <sup>c</sup>	C <sub>18</sub> H <sub>16</sub> Cl <sub>2</sub> N <sub>4</sub> O <sub>5</sub>	48.4	3.9	16.0	12.1	49.2	3.7	16.1	12.8	1740	—	91
IVa	178—180 <sup>b</sup>	C <sub>11</sub> H <sub>14</sub> N <sub>2</sub> O <sub>3</sub>	59.7	6.2	—	12.5	59.4	6.3	—	12.6	1705	1746	68
IVb	191—193 <sup>c</sup>	C <sub>12</sub> H <sub>16</sub> N <sub>2</sub> O <sub>3</sub>	61.6	6.7	—	11.7	61.0	6.8	—	11.9	1700	1745	88
IVc	198—200 <sup>c</sup>	C <sub>11</sub> H <sub>13</sub> ClN <sub>2</sub> O <sub>3</sub>	51.5	5.0	13.9	10.8	51.5	5.1	13.8	10.9	1710	—	88
IVd	183—185 <sup>c</sup>	C <sub>11</sub> H <sub>13</sub> ClN <sub>2</sub> O <sub>3</sub>	51.5	5.1	13.8	10.8	51.5	5.1	13.8	10.9	1705	1750	94
IVe	228—230 <sup>c</sup>	C <sub>11</sub> H <sub>12</sub> Cl <sub>2</sub> N <sub>2</sub> O <sub>3</sub>	43.4	4.2	24.9	9.8	43.6	4.2	24.4	9.6	1710	1747	68
Va	118—119 <sup>a</sup>	C <sub>12</sub> H <sub>16</sub> N <sub>2</sub> O <sub>3</sub>	60.7	6.8	—	11.9	61.0	6.8	—	11.9	1723	1743	89
Vb	157 <sup>a</sup>	C <sub>13</sub> H <sub>18</sub> N <sub>2</sub> O <sub>3</sub>	62.3	7.2	—	11.2	62.4	7.2	—	11.2	1698	1745	64
Vc	132—134 <sup>a</sup>	C <sub>12</sub> H <sub>15</sub> ClN <sub>2</sub> O <sub>3</sub>	53.2	5.6	12.6	10.3	53.2	5.6	13.1	10.4	1698	1743	87
Vd	88—90 <sup>a</sup>	C <sub>12</sub> H <sub>15</sub> ClN <sub>2</sub> O <sub>3</sub>	53.5	5.6	12.3	10.0	53.3	5.6	13.1	10.4	1705	1745	62
Ve	119—121 <sup>a</sup>	C <sub>12</sub> H <sub>14</sub> Cl <sub>2</sub> N <sub>2</sub> O <sub>3</sub>	—	—	—	9.1	—	—	—	9.2	1730	1755	73
VIf	134—135 <sup>a</sup>	C <sub>18</sub> H <sub>17</sub> ClN <sub>2</sub> O <sub>4</sub>	60.2	4.7	9.5	8.0	59.9	4.8	9.8	7.8	1730	1760	78
VIf	163—164 <sup>a</sup>	C <sub>18</sub> H <sub>16</sub> Cl <sub>2</sub> N <sub>2</sub> O <sub>4</sub>	54.7	4.1	18.3	6.0	54.7	4.1	17.9	7.1	1748	1760	74
VIIa	135—136 <sup>a</sup>	C <sub>12</sub> H <sub>16</sub> N <sub>2</sub> O <sub>3</sub>	61.1	6.4	—	11.9	61.0	6.8	—	11.9	1724	1742	63
VIIb	131—132 <sup>b</sup>	C <sub>13</sub> H <sub>18</sub> N <sub>2</sub> O <sub>3</sub>	62.5	7.0	—	11.4	62.4	7.2	—	11.2	1710	1750	60
VIIc	136—137 <sup>a</sup>	C <sub>12</sub> H <sub>15</sub> ClN <sub>2</sub> O <sub>3</sub>	53.1	5.4	12.8	10.4	53.2	5.6	13.1	10.4	1725	1742	58
VId	176—179 <sup>a</sup>	C <sub>12</sub> H <sub>15</sub> ClN <sub>2</sub> O <sub>3</sub>	52.9	5.2	13.0	10.2	53.2	5.6	13.1	10.4	1718	—	85
VIIe	145—147 <sup>a</sup>	C <sub>12</sub> H <sub>14</sub> Cl <sub>2</sub> N <sub>2</sub> O <sub>3</sub>	47.3	4.5	22.5	9.1	47.2	4.6	23.2	9.2	1726	1753	73
VIIIa	51—53 <sup>d</sup>	C <sub>13</sub> H <sub>18</sub> N <sub>2</sub> O <sub>3</sub>	—	—	—	11.1	—	—	—	11.2	1740	1745	84
VIIIb	79—79.5 <sup>d</sup>	C <sub>14</sub> H <sub>20</sub> N <sub>2</sub> O <sub>3</sub>	63.2	7.8	—	10.2	63.6	7.6	—	10.6	1720	1745	70
VIIIc	126 <sup>d</sup>	C <sub>13</sub> H <sub>17</sub> ClN <sub>2</sub> O <sub>3</sub>	54.8	5.9	12.4	10.0	54.8	5.3	12.5	9.8	1731	1743	81
VIIIe	74.5—75 <sup>d</sup>	C <sub>13</sub> H <sub>16</sub> Cl <sub>2</sub> N <sub>2</sub> O <sub>3</sub>	48.5	5.3	22.6	9.1	48.9	5.1	22.2	8.8	1728	1749	58

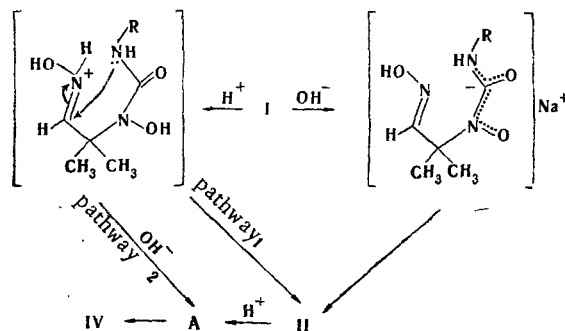
- a) From benzene.  
b) From ether.  
c) From acetone.  
d) From petroleum ether.

molecularly bonded OH group) or on opposite sides (free OH group).



The formation of an intramolecular hydrogen bond in monobenzoyl derivatives VI is possible with respect to both the ester oxygen atom and the carbonyl oxygen atom of the benzoyl group. The band of the intramolecular hydrogen bond in these compounds is also probably associated with the existence of two forms similar to forms V<sub>1</sub> and V<sub>2</sub>, the conversion of which occurs as a result of ring inversion with simultaneous pyramidal inversion with simultaneous pyramidal inversion of both nitrogen atoms of the urea fragment (see [12]).

The cyclization of I in acids and alkali is evidently associated with the fact that in alkalis salt formation at the NOH group occurs initially, and, in connection with this, the electron density over the entire hydroxylurea fragment of the molecule increases, and this facilitates nucleophilic attack of the nitrogen atom at the carbon atom of the oxime group. In the case of acids, however, prior protonation of the oxime group leads to an increase in the fraction of positive charge on the carbon atom. This promotes cyclization to give II, which being unstable in acids, is converted to IV (path 1). On the other hand, the possibility of the formation of intermediate linear product A, which can cyclize with the loss of a hydroxylamine molecule to give IV (path 2), is not excluded.



## EXPERIMENTAL

The IR spectra of KBr pellets and carbon tetrachloride and dioxane solutions of the compounds were recorded with UR-10 and Perkin-Elmer-457 spectrometers. The UV spectra of alcohol solutions were recorded with a Specord UV-vis spectrophotometer. The PMR spectra of 10% solutions of the compounds in  $(\text{CD}_3)_2\text{SO}$  and  $\text{CD}_3\text{CN}$  were recorded with Tesla BS-487C and Bruker HX-90E spectrometers with hexamethyldisiloxane as the internal standard. The results of elementary analysis, the melting points, the yields, and the spectral characteristics of II-VIII are presented in Table 1. The synthesis of Ia-e was previously described in [1].

4-Hydroxylamino-1-hydroxy-3-aryl-5,5-dimethyl-2-imidazolidinones (IIa-e). A solution of 2 mmole of Ia-e in 20 ml of 1 N NaOH was heated to the boiling point, after which it was cooled, acidified to pH 7 with 10% hydrochloric acid solution, and extracted with four 15-ml portions of ether. The ether extract was dried with magnesium sulfate, and the solvent was evaporated to give crystalline IIa-d. According to the results of elementary analysis and the PMR spectrum, IIe was isolated in the form of the  $\text{IIe} \cdot (\text{C}_2\text{H}_5)_2\text{O}$  complex with mp 72-74°. Found: Cl 18.7; N 11.2%.  $\text{C}_{15}\text{H}_{23}\text{Cl}_2\text{N}_3\text{O}_4$ . Calculated: Cl 18.6; N 11.0%. IR spectrum,  $\nu$  (in KBr): 1740 (C=O) and 1095 (C-O-C)  $\text{cm}^{-1}$ . PMR spectrum ( $\text{CD}_3\text{CN}$ ): 1.23 (t,  $J = 7.0$  Hz) and 3.43 (q,  $J = 7.0$  Hz)  $[(\text{CH}_3\text{CH}_2)_2]$ ; 1.23 and 1.33  $[>\text{C}(\text{CH}_3)_2]$ ; 4.67 ( $>\text{CH}$ ); 7.47-7.80 (aromatic ring protons). Compound IIe was obtained in the free state by the method described above, except that chloroform was used as the extractant.

N-(1-Hydroxy-3-aryl-5,5-dimethyl-2-imidazolidinon-4-yl)-p-nitrophenylnitrones (IIIb-e). A solution of 2 mmole of IIb-e and 2 mmole of p-nitrobenzaldehyde in 15 ml of alcohol was refluxed for 30 min, after which it was allowed to stand at 25° for 24 h. The resulting yellow precipitate was removed by filtration and washed with ether.

1,4-Dihydroxy-3-aryl-5,5-dimethyl-2-imidazolidinones (IVa-e). A) A 3-mmole sample of Ia-e was heated to the boiling point in 40 ml of 10% hydrochloric acid, after which the mixture was cooled, and the precipitated IVa-e were removed by filtration and recrystallized from acetone.

B) A 2-mmole sample of IIa-e was dissolved by heating in 10 ml of 10% HCl solution, after which the solution was heated to the boiling point. It was then cooled, and the precipitated IVa-e were removed by filtration.

C) Four to five drops of concentrated  $\text{H}_2\text{SO}_4$  were added to a solution of 2 mmole of Va-e in 30 ml of  $\text{CCl}_4$ , the mixture was stirred at 25° for 2-3 h, and the precipitated IVa-e were removed by filtration.

1-Hydroxy-4-methoxy-3-aryl-5,5-dimethyl-2-imidazolidinones (Va-e). A) A 5-10-mg sample of p-toluenesulfonic acid was added to a solution of 4 mmole of IVa-e in 30 ml of absolute methanol, and the mixture was allowed to stand at 25° for 24 h. It was then made alkaline to pH 8.5-9.0 with 10% ammonium hydroxide, the solvents were removed by vacuum distillation to dryness, and the residue was treated two to three times with absolute ether. The ether solution was vacuum evaporated to give Va-e.

B) Dry hydrogen chloride was bubbled into a solution of 4 mmole of IIa-e in 10 ml of absolute methanol for 5 min, after which the mixture was heated to the boiling point, cooled, made alkaline to pH 8.0-8.5 with 10% ammonium hydroxide, and worked up as in method A to give Va-e. A mixture of IV and V [thin-layer chromatography (TLC) data] was obtained when hydrochloric acid was used in place of dry hydrogen chloride.

1-Benzoxo-4-hydroxy-3-aryl-5,5-dimethyl-2-imidazolidinones (VIc,e). A 2.2-mmole sample of triethylamine and 2.2 mmole of benzoyl chloride were added to a suspension of 2 mmole of IVc or IVe in 80 ml of methylene chloride, and the mixture was stirred at 23° for 3 h. It was then poured into water, and 150 ml of ether was added to the aqueous mixture. The organic layer was separated and dried with magnesium sulfate, and the solvent was evaporated to give VIc or VIe.

1-Methoxy-4-hydroxy-3-aryl-5,5-dimethyl-2-imidazolidinones (VIIa-e). A solution of 2.2 mmole of dimethyl sulfate in 10 ml of methanol was added dropwise in the course of 10 min to a solution of 2 mmole of IVa-e and 2.2 mmole of sodium hydroxide in 20 ml of methanol, after which the mixture was stirred at 25° for 2 h. It was then treated again with 2.2 mmole of sodium hydroxide and (dropwise) a solution of 2.2 mmole of dimethyl sulfate in 10 ml of methanol. The mixture was stirred at 25° for 1 h, after which the solvent was vacuum evaporated, and the residue was treated with 40 ml of dry chloroform. The insoluble material was removed by filtration, and the chloroform was evaporated from the filtrate to give VIIa-e.

1,4-Dimethoxy-3-aryl-5,5-dimethyl-2-imidazolidinones (VIIIa-c, e). A solution of 4.4 mmole of dimethyl sulfate in 10 ml of methanol was added dropwise to a solution of 4 mmole of Va-c,e and 4.4 mmole of sodium hydroxide in 30 ml of methanol, and the mixture was stirred at 25° for 2 h. It was then worked up as in the preceding example to give VIIIa-c,e.

Testing of the Complexing Ability of II-VII. A greenish-blue coloration was observed when an alcohol solution of ferric chloride was added to acetone solutions of II-VII, whereas a bright-red coloration was observed when 2,3,5-triphenyltetrazole, obtained by neutralization of a 1% alcohol solution of its hydrochloride with 2% NaOH solution, was added to acetone solutions of II.

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