SYNTHESIS OF ACYL DERIVATIVES OF α-HYDRAZINO OXIMES AND 2-IMIDAZOLIDINONES BASED ON THEM

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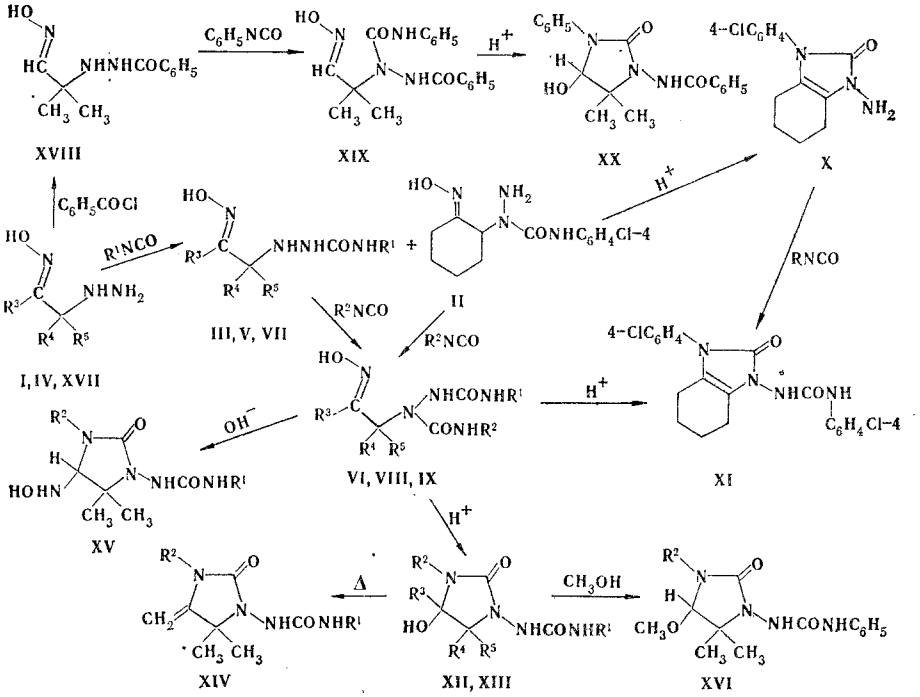
The reaction of N_1- or N_2-monoacyl derivatives of 2-hydrazinocyclohexanone, 3-hydrazino-3-methyl-2-butanone, and 2-hydrazino-2-methylpropanal oximes with isocyanates was used to synthesize, N,N-diacyl derivatives, which, under the influence of acids or bases, undergo cyclization to the corresponding substituted 2-imidazolinones or 4-methylene-, 4-hydroxylamino-, 4-hydroxy-, and 4-methoxy-2-imidazolidinones.

We have previously shown [1] that 2-hydrazinocyclohexanone oxime (I) reacts with an equimolar amount of an aryl isocyanate to give a mixture of two oximes, viz., 2-[N_1-(arylcarbamoyl)hydrazino]- and 2-[N_2-(arylcarbamoyl)-hydrazino]cyclohexanone (II, III), whereas 3-hydrazino-3-methyl-2-butanone (IV) gives only 3-[N_2-(arylcarbamoyl)hydrazino]-3-methyl-2-butanone (V).



I, III, VI $R^3+R^4=(CH_2)_4$, $R^5=H$; IV, V, VIII, XII $R^3=R^4=R^5=CH_3$; VII, IX, XIII, XVII $R^3=H$, $R^4=R^5=CH_3$; III $R^1=4\text{-}ClC_6H_4$; V, VII a $R^1=C_6H_5$, b $R^1=4\text{-}ClC_6H_4$, c $R^1=3\text{-}ClC_6H_4$, d $R^1=4\text{-}CH_3C_6H_4$, e $R^1=3,4\text{-}Cl_2C_6H_3$, f $R^1=2\text{-}Cl\text{-}4\text{-}CH_3C_6H_3$; VI $R^1=R^2=4\text{-}ClC_6H_4$; VIII, XII, XIV a $R^1=C_6H_5$, $R^2=4\text{-}ClC_6H_4$, b $R^1=R^2=4\text{-}ClC_6H_4$; IX, XIII, XV a $R^1=R^2=C_6H_5$, b $R^1=C_6H_5$, $R^2=3,4\text{-}Cl_2C_6H_3$, c $R^1=3\text{-}ClC_6H_4$, $R^2=CH_3$, d $R^1=4\text{-}ClC_6H_4$, $R^2=CH_3$, e $R^1=C_6H_5$, $R^2=4\text{-}Cl\text{—}C_6H_4$, f $R^1=R^2=4\text{-}ClC_6H_4$, g $R^1=4\text{-}CH_3C_6H_4$, $R^2=C_6H_5$, h $R^1=2\text{-}Cl\text{—}4\text{-}CH_3C_6H_3$, $R^2=CH_3$

In a continuation of our research in this direction we have studied the reactivities of monocarbamoyl derivatives of α-hydrazino oximes and the ability of their acylation products to undergo cyclization under the influence of acids and bases. The reaction of 2-[N_1-(4-

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chlorophenylcarbamoyl)]- and 2-[N_2-(4-chlorophenylcarbamoyl)hydrazino]cyclohexanone oxime with 4-chlorophenyl isocyanate leads to VI, the presence in the IR spectrum of which of two absorption bands of carbonyl groups at 1680 and 1690 cm^{-1} and the presence in the PMR spectrum of which of a singlet of the proton of an HON=C group at 10.6 ppm make it possible to assign the 2-[N_1,N_2-bis(4-chlorophenylcarbamoyl)hydrazino]cyclohexanone oxime structure to this compound.

The carbamoylation of V and 2-[N_2-(arylcarbamoyl)hydrazino]-2-methylpropanal oxime (VII) with aryl (alkyl) isocyanates (1 mole) also leads to 3-[N_1,N_2-bis(arylcarbamoyl)hydrazino]-3-methyl-2-butanone (VIII) and 2-{N_1,N_2-bis[aryl(alkyl)carbamoyl]hydrazino}-2-methylpropanol (IX) oximes, respectively. Compounds VI, VIII, and IX were also synthesized by the action of 2 moles of the isocyanate on the corresponding α-hydrazino oximes.

Monocarbamoyl derivatives III, V, and VII are resistant to the action of alkalis and acids under ordinary conditions. However, III and VII upon treatment with acids under severe conditions (at the boiling points of the reaction mixtures) undergo destruction, in contrast to V, which are known [2] to undergo cyclization to perhydro-1,2,4-triazin-2-one derivatives. At the same time, treatment of II with hydrochloric acid in alcohol leads to X, to which the 4,5-tetramethylene-1-amino-3-(4-chlorophenyl-2-imidazolone structure was assigned on the basis of data from the IR and PMR spectra. N_1,N_2-Dicarbamoyl derivative VI, which under the influence of sulfuric acid in alcohol gives 4,5-tetramethylene-3-(4-chlorophenyl)-1-(N-4-chlorophenyl-N'-ureido)-2-imidazolinone (XI), behaves similarly in an acidic medium. Compound X reacts with 4-chlorophenyl isocyanate to give XI.

Compounds XII and XIII, respectively, were obtained when VIII and IX were treated with sulfuric acid in alcohol. Absorption bands of carbonyl groups at 1660 and 1710 cm^{-1} are observed in the IR spectrum of, for example, XIIa, and three singlets of three methyl groups at 1.16, 1.21, and 1.24 ppm, two singlets of two NH protons at 8.24 and 8.55 ppm, a singlet of an OH group at 6.47 ppm, and a multiplet of aromatic ring protons at 8.30-8.73 ppm are present in the PMR spectrum. The 1-(N-aryl-N'-ureido)-3-aryl(alkyl)-5,5-dimethyl-4-hydroxy-2-imidazolidinone and 1-(N-aryl-N'-ureido)-3-aryl-4,5,5-trimethyl-4-hydroxy-2-imidazolidinone structures, respectively, were assigned to XIII and XII on the basis of these data.

Heat treatment of solutions of XII in dimethyl sulfoxide (DMSO) or chlorobenzene leads to their dehydration and the formation of 4-methylene-5,5-dimethyl-3-(4-chlorophenyl)-1-(N-aryl-N'-ureido)-2-imidazolidinones (XIV). Thus when a solution of XIIa is heated directly in the sensor of the spectrometer, the appearance of a singlet of the protons of geminal methyl groups at 1.42 ppm and the AB system (4.09 and 4.27 ppm, J = 2 Hz) of the protons of a methylene group is observed in the PMR spectrum as the intensities of the signals of the starting compound become weaker.

Compounds VI and VIII, just like monocarbamoyl derivatives II, III, V, and VII, are resistant to heating in 5% solutions of alkali or sodium ethoxide. At the same time, dicarbamoyl derivatives (IX) of an α-hydrazino aldoxime upon heating in 5% alkali solution with subsequent neutralization gave XV, which are isomers of the starting compounds. A 20-30 cm^{-1} shift of the absorption band of the carbonyl group to the high-frequency region is observed in the IR spectra of XV. Two singlets of geminal methyl groups at 1.20 and 1.42 ppm, two doublets of CH–NH protons at 4.68 and 6.50 ppm, two singlets of two NH groups at 8.36 and 8.40 ppm, a signal of the proton of the HON group (7.74 ppm), and a multiplet of aromatic ring protons at 6.78-7.80 ppm are present in the PMR spectrum of, for example, XVa. The compounds give a positive test for the presence of an unsubstituted hydroxylamino group in the structure with 2,4,5-triphenyltetrazolium chloride [3]. The 1-(N-aryl-N'-ureido)-3-aryl-3,5-dimethyl-4-hydroxylamino-2-imidazolidinone structure was assigned to XV on the basis of these data.

When XV are treated with hydrochloric acid, they readily lose hydroxylamine and form 4-hydroxy-2-imidazolidinones (XIII). When XIII are refluxed in methanol, they are converted to the corresponding 4-methoxy-substituted 2-imidazolidines (XVI).

The acylation of α-hydrazino oximes with the participation of benzoyl chloride leads to different results. The reaction of 2-hydrazino-2-methylpropanaladoxime (XVII) with benzoyl chloride in the presence of triethylamine gave 2-[N_2-(benzoyl)hydrazino]-2-methylpropanaldoxime (XVIII), the subsequent carbamoylation of which led to 2-[N_2-(benzoyl)-N_1-(phenylcarbamoyl)-hydrazino]-2-methylpropanaldoxime (XIX). Under the influence of acid, XIX undergoes cyclization to 1-benzamido-3-phenyl-5,5-dimethyl-4-hydroxy-2-imidazolidinone (XX). However, the

TABLE 1. Chemical Shifts of the Protons in the PMR Spectra of the Compounds Obtained

Compound	δ, ppm					
	germ-$(CH_3)_2$	Ar*	>CH	CH_3	NH, NH, NH	OH
VIIa	1,18	6,69—7,35	7,23		4,9; 7,3; 8,4	10,46
VIIb	1,16	6,74—7,71	7,23		4,87; 7,27; 8,51	10,38
VIIc	1,12	6,64—7,66	7,25		4,90; 7,31; 8,58	10,42
VIIe	1,22	7,12—7,80	7,23		4,90; 7,30; 8,58	10,35
VIIIa	1,35	6,75—7,65		1,83	7,65; 8,01; 8,70	10,00
VIIIb	1,47; 1,61	6,80—7,61		2,02	7,50; 7,95; 8,77	9,67
IXa	1,46	6,52—7,50	7,65		8,21; 8,25; 8,55	9,87
IXb	1,54	6,75—7,98	7,65		8,21; 8,55; 8,73	10,00
IXe	1,42	6,70—7,63	7,65		8,36; 8,70; 8,77	10,20
IXg	1,39	6,72—7,68	7,65		8,16; 8,75; 8,85	10,01
XVIII	1,21	7,05—7,61	7,35		5,36; 7,33	9,82
XIX	1,52	6,56—7,51	7,61		8,21; 7,90	10,02
XXI	1,35	6,60—7,95	7,85		5,21; 7,43; 8,25	—

*Multiplet that includes the signals of the protons of the aromatic rings.

TABLE 2. Chemical Shifts (ppm) of the Protons in the PMR Spectra of the Compounds Obtained

Compound	gem-$(CH_3)_2$	CH_3	>CH	OH	Ar[a]	NH	NH
XIIa	1,16; 1,27	1,21		6,47	8,30—8,73	8,24	8,55
XIIb	1,12; 1,23	1,21		6,48	7,07—7,55	8,40	8,62
XIIIa	1,12; 1,20		5,04	—	6,78—7,76	8,26	8,38
XIIIb	1,21; 1,27		5,06	—	6,76—7,83	8,06	8,32
XIIIc	1,08		4,55	—	6,95—7,84	8,44	8,66
XIIId[b]	1,08; 1,16		4,38	—	6,73—7,95	8,13	8,36
XIIIe	1,27; 1,46		5,02	—	6,75—7,95	8,17	8,51
XIIIg[c]	1,05; 1,16		5,06	—	6,82—7,72	8,25	8,32
XVa	1,21; 1,42		4,68[d]	7,74	6,78—7,80	8,36	8,41
XVb	1,21; 1,42		4,72[d]	—	6,75—7,98	8,17	8,28
XVd	1,12; 1,27		3,92 d	7,53	6,97—7,58	8,21	8,32
XVIe	1,27		5,13	—	6,78—7,76	8,06	8,47
XX	1,23		5,10	—	6,93—7,98	—	—

[a]Multiplet that includes the signals of the protons of the aromatic rings. [b]The N—CH_3 signal is at 2.81 ppm. [c]The CH_3Ar signal is at 2.13 ppm (3H). [d]The doublet of the 4-NH proton: at 6.5 ppm (J = 3 Hz) for XVa, at 6.63 ppm (J = 3Hz) for XVb, and 6.39 ppm (J = 3 Hz) for XVd. [e]The OCH_3 signal is at 3.37 ppm.

acylation of monophenylcarbamoyl derivative VII with benzoyl chloride leads to XXI, the IR spectrum of which contains absorption bands of an ester group at 1735 and 1245 cm^{-1}, whereas the PMR spectrum does not contain the signal of the proton of an oxime group. These data constitute evidence for O-acylation and make it possible to assign the O-benzoyl-2-[N_2-(phenylcarbamoyl)hydrazino]-2-methylpropanaldoxime structure, which does not undergo cyclization under the influence of acid, to XXI.

EXPERIMENTAL

The IR spectra of KBr pellets and solutions of the compounds in CCl_4 and dioxane were recorded with a Perkin—Elmer 457 spectrometer. The PMR spectra of 10% solutions of the compounds in $(CD_3)_2SO$ were recorded with Tesla BS-487C and Brucker-90E spectrometers with tetramethylsilane as the internal standard. The starting α-hydrazino oximes were obtained by the method in [4]. The course of the reaction was monitored on Silufol UV-254 plates in a tetrahydrofuran—hexane system (1:1). The results of elementary analysis and the yields and characteristics of the compounds are presented in Tables 1-3.

TABLE 3. Characteristics of VII-IX, XII-XVI, and XVIII-XXI

Compound	mp, °C	IR spectrum (KBr), CO, cm^{-1}	Found, %				Empirical formula	Calc., %				Yield, %
			C	H	Cl	N		C	H	Cl	N	
VIIa	134—135	1665	56,0	6,9	—	23,9	$C_{11}H_{16}N_4O_2$	55,8	6,8	—	23,7	58
VIIb	153—154	1665	49,1	5,7	12,6	20,7	$C_{11}H_{15}ClN_4O_2$	48,8	5,6	13,1	20,7	60
VIIc	129—130	1665	49,4	5,6	13,3	20,6	$C_{11}H_{15}ClN_4O_2$	48,8	5,6	13,1	20,7	80
VIId	155—156	1670	57,2	7,4	—	22,0	$C_{12}H_{18}N_4O_2$	57,6	7,2	—	22,3	90
VIIe	155—157	1690	44,0	4,5	23,2	18,1	$C_{11}H_{14}Cl_2N_4O_2$	43,3	4,6	23,2	18,4	66
VIIf	148—149	1665	50,7	6,6	12,7	19,6	$C_{12}H_{17}ClN_4O_2$	50,6	5,9	12,4	19,6	71
VIIIa	142—143	1700[a]	56,5	5,5	7,8	17,2	$C_{19}H_{22}ClN_5O_3$	56,5	5,5	8,8	17,4	84
VIIIb	204—205	1700[a]	52,0	4,7	16,6	16,1	$C_{19}H_{21}Cl_2N_5O_3$	52,1	4,8	16,2	15,9	92
IXa	175—176	1660, 1695	60,5	6,0	—	19,4	$C_{18}H_{21}N_5O_3$	60,8	5,9	—	19,7	76
IXb	168—169	1645, 1690	49,7	4,5	16,2	16,5	$C_{18}H_{19}Cl_2N_5O_3$	50,9	4,5	16,7	16,5	68
IXc	145—146	1650, 1690	47,6	5,4	11,0	21,3	$C_{13}H_{18}ClN_5O_3$	47,6	5,5	10,8	21,4	94
IXd	174—175	1660, 1685	47,6	5,8	11,2	21,4	$C_{13}H_{18}ClN_5O_3$	47,6	5,5	10,8	21,4	80
IXe	160—161	1650, 1690	55,4	5,1	8,9	17,9	$C_{18}H_{20}ClN_5O_3$	55,4	5,2	9,1	17,9	93
IXf	123—124	1680[a]	50,9	4,8	16,6	16,5	$C_{18}H_{19}Cl_2N_5O_3$	50,9	4,5	16,7	16,5	94
IXg	168—169	1645, 1685	61,5	6,5	—	18,4	$C_{19}H_{23}N_5O_3$	61,7	6,3	—	18,9	82
IXh	144—145	1650, 1695	50,6	6,1	11,1	18,0	$C_{14}H_{20}ClN_5O_3$	50,6	6,0	10,7	18,1	84
XIIa	182—183	1660, 1710	58,4	5,2	9,7	14,5	$C_{19}H_{21}ClN_4O_3$	58,6	5,4	9,1	14,6	83
XIIb	286—287	1660, 1715	53,7	4,4	17,1	13,0	$C_{19}H_{20}Cl_2N_4O_3$	53,9	4,7	16,7	13,1	76
XIIIa	244—245	1660, 1715	63,6	6,1	—	16,6	$C_{18}H_{20}N_4O_3$	63,5	5,9	—	16,4	93
XIIIb	168—169	1670, 1725	52,8	4,4	11,3	13,3	$C_{18}H_{18}Cl_2N_4O_3$	52,8	4,4	11,7	13,7	92
XIIIc	117—118	1700[b]	50,1	5,4	10,9	17,9	$C_{13}H_{17}ClN_4O_3$	49,9	5,4	11,3	17,9	66
XIIId	159—160	1700[b]	50,0	5,4	11,0	18,2	$C_{13}H_{17}ClN_4O_3$	49,9	5,4	11,3	17,9	62
XIIIe	211—212	1660, 1715	57,6	5,1	9,9	14,9	$C_{18}H_{19}ClN_4O_3$	57,6	5,1	9,5	14,9	80
XIIIf	220—221	1685, 1715	52,7	4,4	11,4	13,6	$C_{18}H_{18}Cl_2N_4O_3$	52,8	4,4	11,7	13,7	84
XIIIg	232—233	1665, 1715	64,3	6,2	—	15,8	$C_{19}H_{22}N_4O_3$	64,0	6,2	—	16,0	70
XIIIh	91—92	1700[b]	50,8	5,5	11,1	18,1	$C_{14}H_{19}ClN_4O_3$	50,8	5,7	10,7	18,2	87
XIVa[c]	218—219	1665, 1710	61,1	4,7	10,1	12,1	$C_{19}H_{19}ClN_4O_2$	61,5	5,1	9,5	12,8	94
XIVb[c]	313—314	1670, 1710	51,8	4,2	16,7	15,5	$C_{19}H_{18}Cl_2N_4O_2$	52,2	4,3	17,1	15,5	89
XVa	225—226	1695, 1715	60,8	5,9	—	19,5	$C_{18}H_{21}N_5O_3$	60,8	5,9	—	19,7	86
XVb	203—204	1670, 1720	50,8	4,4	16,4	16,9	$C_{18}H_{19}Cl_2N_5O_3$	50,9	4,5	16,7	16,5	79
XVc	192—193	1685, 1710	47,4	5,4	11,0	21,1	$C_{13}H_{18}ClN_5O_3$	47,6	5,5	10,8	21,4	68
XVd	194—195	1685, 1710	47,6	5,5	11,1	21,4	$C_{13}H_{18}ClN_5O_3$	47,6	5,5	10,8	21,4	63
XVI	168—169	1700, 1735[d]	64,1	5,7	—	16,1	$C_{19}H_{22}N_4O_3$	64,0	6,2	—	16,0	94
XVIII	130—131	1665	59,7	6,7	—	18,9	$C_{11}H_{15}N_3O_2$	59,7	6,8	—	18,9	82
XIX	155—156	1660, 1690	63,4	6,3	—	15,9	$C_{18}H_{20}N_4O_3$	63,5	5,9	—	16,4	90
XX	110—111	1680, 1720	66,3	5,8	—	12,9	$C_{18}H_{19}N_3O_3$	66,4	5,9	—	12,9	92
XXI	128—129	1700, 1735	63,3	5,9	—	16,5	$C_{18}H_{19}N_3O_3$	63,5	5,9	—	16,5	81

[a]Broad band. [b]In dioxane: 1710, 1735 cm^{-1}. [c]The C=C stretching vibrations are at 1635 cm^{-1}. [d]In dioxane.

2-[N_1,N_2-Bis(4-chlorophenylcarbamoyl)hydrazino]cyclohexanone Oxime (VI). A) A solution of 3.06 g (2 mmole) of 4-chlorophenyl isocyanate in 20 ml of dioxane was added gradually to a solution of 1.43 g (1 mmole) of I in 50 ml of dioxane, and the mixture was allowed to stand for 10 min. The solvent was evaporated, and the resulting oil was treated with diethyl ether to give 4.15 g (90%) of VI with mp 220-221°C. IR spectrum (KBr): 1680, 1690 cm^{-1} (C=O). PMR spectrum: complex bands at 0.78-2.28 [8H, $(CH_2)_4$] and 4.80 (1H, CH), 7.12-7.68 (8H, m C_6H_4), 7.42 (1H, s, NH), 9.03 (1H, s, NH), and 10.6 ppm (1H, s, OH). Found, %: C 53.3; H 4.62; Cl 15.9; N 13.0. $C_{20}H_{21}Cl_2N_5O_3$. Calculated, %: C 53.3; H 4.71; Cl 15.7; N 13.3.

B) Compound VI was also obtained by the action of an equimolar amount of 4-chlorophenyl isocyanate on II or III under the conditions in method A. The product was obtained in 83 and 84% yields, respectively.

2-[N_2-(Arylcarbamoyl)hydrazino]-2-methylpropanaldoxime (VII). A 1-mmole sample of the aryl isocyanate was added in the course of an hour to a solution of 1 mmole of XVII in 50 ml of dioxane. At the end of the reaction (according to chromatographic monitoring) the solvent was evaporated *in vacuo*, and the residue was treated with benzene or ether to give VII.

3-[N_1,N_2-Bis(arylcarbamoyl)hydrazino]-3-methyl-2-butanone Oximes (VIII) and 2-{N_1,N_2-Bis[aryl(alkyl)carbamoyl]hydrazino}-2-methylpropanaldoximes (IX). These compounds were obtained from IV and V and VII and XVII, respectively, by methods A and B.

4,5-Tetramethylene-1-amino-3-(4-chlorophenyl)-2-imidazolone (X). Three to five drops of concentrated H_2SO_4 were added to a solution of 2.96 g (1 mmole) of II in 25 ml of alcohol, and the reaction mixture was heated to the boiling point, neutralized with 2 N NaOH, and treated with 50 ml of water. The resulting precipitate was removed by filtration to give 2.3 g (86%) of X with mp 165-166°C. IR spectrum (KBr): 1670 (C=O); 3200, 3300 cm^{-1} (NH_2). PMR spectrum:

complex bands centered at 1.68 and 2.21 [8H, $(CH_2)_4$], 4.87 (2H, s, NH_2), and 7.20-7.65 ppm (4H, m, C_6H_4). Found, %: C 59.1; H 5.3; Cl 14.0; N 15.8. $C_{13}H_{14}ClN_3O$. Calculated, %: C 59.2; H 5.3; Cl 13.5; N 15.9.

4,5-Tetramethylene-3-(4-chlorophenyl)-1-(N-4-chlorophenyl-N'-ureido)-2-imidazolinone (XI). A) This compound, with mp 168-169°C, was obtained in 63% yield from VI by the method used to synthesize X. IR spectrum (KBr): 1670, 1700 cm^{-1} (C=O). PMR spectrum: complex bands centered at 1.68 and 2.21 [8H, $(CH_2)_4$], 7.08-7.57 (8H, m, C_6H_4), 8.55 (1H, s, NH), and 9.22 ppm (1H, 5, NH). Found, %: C 57.5; H 4.6; Cl 17.2; N 13.4. $C_{20}H_{18}Cl_2N_4O_2$. Calculated, %: 57.4; H 4.3; Cl 16.9; N 13.4.

B) Compound XI was also obtained by the action of 4-chlorophenyl isocyanate on X by the method used to synthesize VII.

1-(N-Aryl-N'-ureido)-3-aryl-4,5,5-trimethyl-4-hydroxy-2-imidazolininones (XII) and 1-(N-Aryl-N'-ureido)-3-aryl(alkyl)-5,5-dimethyl-4-hydroxy-2-imidazolidinones (XIII). These compounds were obtained, respectively, from VIII and IX and SV by the method used to synthesize X.

4-Methylene-5,5-dimethyl-1-(N-aryl-N'-ureido)-2-imidazolidinones (XIV). A 1-mmole sample of XII was refluxed in chlorobenzene until it dissolved, after which the solution was cooled, and the precipitated XIV was removed by filtration. PMR spectrum: XIVa: 1.42 (6H, s, CH_3); 4.09, 4.24 (2H, AB, J = 2 Hz, >C=CH_2); 6.90-7.65 (9H, m, Ar); 8.43 (1H, s, NH); 8.85 ppm (1H, s, NH); XIVb: 1.40 (6H, s, CH_3); 4.09, 4.24 (2H, AB, J = 2 Hz, >C=CH_2); 7.01-7.65 (8H, m, Ar); 8.41 (1H, s, NH); 8.85 ppm (1H, s, NH).

1-(N-Aryl-N'-ureido)-3-aryl(alkyl)-5,5-dimethyl-4-hydroxylamino-2-imidazolidinone (XV). A 1-mmole sample of IX was heated in 20 ml of 1 N NaOH to 100°C, after which the mixture was cooled and neutralized with 1 N HCl, and the precipitate was removed by filtration.

1-(N-Aryl-N'-ureido)-3-aryl-5,5-dimethyl-4-methoxy-2-imidazolidinone (XVI). A 1-mmole sample of XII was refluxed in 50 ml of methanol in the presence of a catalytic amount of p-toluenesulfonic acid for 2 h. At the end of the reaction (according to chromatographic monitoring) the alcohol was evaporated, and the residue was crystallized in a mixture of absolute ether and hexane.

2-[N_2-(Benzoyl)hydrazino]-2-methylpropanaldoxime (XVIII). A 1.54-g (1.1 mmole) sample of benzoyl chloride was added to a solution of 1.17 g (1 mmole) of XVII and 1.01 g (1 mmole) of triethylamine in dioxane, and the mixture was allowed to stand for 1 h. The dioxane was evaporated *in vacuo*, and the residue was washed with several portions of hot water, air dried, and recrystallized from benzene.

2-[N_2-(Benzoyl)-N_1-(phenylcarbamoyl)hydrazino]-2-methylpropanaldoxime (XIX). A 1.19-g (1 mmole) sample of phenyl isocyanate was added to a solution of 2.21 g (1 mmole) of XVIII in THF. At the end of the reaction (according to chromatographic monitoring) the solvent was evaporated *in vacuo*, the resulting oil was treated with ether, and the precipitate was removed by filtration and recrystallized from benzene.

1-Benzamido-3-phenyl-5,5-dimethyl-4-hydroxy-2-imidazolidinone (XX). This compound was obtained by the method presented for X.

O-Benzoyl-2-[N_2-(phenylcarbamoyl)hydrazino]-2-methylpropanaldoxime (XXI). A 1.54-g (1 mmole) sample of benzoyl chloride was added with stirring to a solution of 2.36 g (1 mmole) of VII in 20 ml of pyridine, and the mixture was allowed to stand for 2 h. It was then diluted with water, and the precipitate was removed by filtration, washed with hot water, air dried, and recrystallized from benzene.

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