form, producing a light yellow coloration, while they are insoluble in water and petroleum ether.

Derivatives of Anils. *p*- Nitrophenyl hydrazones, 2,4-dinitrophenyl hydrazones, semicarbazones, and the oximes of the above anils were prepared by the usual methods. These derivatives were crystallized from absolute alcohol except the *p*- nitrophenyl hydrazone of *o*- toluidine which could be crystallized from methanol. The yield of the derivatives was almost quantitative. The characteristics of the above derivatives are given in Table II.

The oximes give color reactions with alcoholic solution of heavy metal ions, especially copper, cobalt, nickel, and iron (ic). The colors are all green of varying shades ranging from yellow green (nickel), to bright green (copper) and dark green (cobalt). With ferric ions color response is given by only two of the above oximes—*viz.*, phenacylidene β-naphthylamine and phenacylidene *o*- toluidine oximes which give reddish orange and violet colors.

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Chemistry of Imidoyl and Amide Chlorides

On the Preparation and Properties of *N*-Substituted 2-Furimidoyl Chlorides. *N,N′*-Disubstituted 2-Furamidines

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N-Monosubstituted-2-furamides C_4H_9O—CONHR, where R = aryl or *n*-C_3H_7—, *n*-C_4H_9—, or $C_6H_5CH_2$—, on reaction with PCl_5 gave new imidoyl chlorides, C_4H_3—C(Cl)=NR; with R = CH_3—, C_2H_5—, and *iso*-C_3H_7—, however, amide chlorides, C_4H_3O—CCl_2NHR, subsequently convertible to imidoyl chlorides, were obtained; with R = *tert*-C_4H_9—, the alkyl group was eliminated to form 2-furonitrile and *tert*-butyl chloride. The new imidoyl chlorides were converted with primary amines to a number of new *N,N′*-disubstituted-2-furamidines and their salts.

ALTHOUGH a large number of imidoyl chlorides have been prepared (*3, 15*), the present paper reports the first isolation and characterization of such derivatives obtained from *N*-substituted amides of 2-furoic acid.

Depending upon the nature of the *N*-substituent in the 2-furamide, the product of the reaction with phosphorus pentachloride may be either the imidoyl or the amide chloride (*9*), or fragmentation products.

The eleven *N*-arylimidoyl chlorides (I–XI) were obtained in good yield, either as viscous yellow oils or as crystalline solids. The reaction between *N*-alkyl-2-furamides and PCl_5, however, does not follow the same pattern. Thus, *N*-(*n*-propyl)- and *N*-(*n*-butyl)-2-furamides reacted with evolution of hydrogen chloride yielding the imidoyl chlorides (XIV, XVI), but *N*-methyl-, *N*-ethyl-, and *N*-isopropyl-2-furamides reacted without evolution of hydrogen chloride to yield the amide chlorides. The amide chlorides, similar to the ones reported previously (*3, 9*), were extremely sensitive to moisture and not isolable in a pure state. Heating of the amide chlorides in dry benzene resulted in the imidoyl chlorides (XII, XIII, XV). Dry pyridine can be used in place of benzene with the same result.

N-alkyl-2-furimidoyl chlorides are thermally less stable and more sensitive than the *N*-aryl analogs. The ease of thermal decomposition on varying the nature of the alkyl group increases in the order prim. < sec. < tert.

Consistent with the results of Degnan and Pope (*8*) is the authors' observation that the reaction of *N*-*tert*-butyl-2-furamide with PCl_5 gives 2-furonitrile and *tert*-butyl chloride, presumably by immediate decomposition of the unstable intermediate imidoyl chloride. This can be rationlized in terms of a considerable fragmentation effect of the *tert*-butyl group. The instability of *N*-phenylsulfonyl-2-furimidoyl chloride, which spontaneously decomposes into furonitrile and benzenesulfonyl chloride, may also be noted. This had not been anticipated since the corresponding *N*-phenylsulfonylimidoyl chlorides of other aliphatic and aromatic counterparts are very stable (*4*).

Existence of any syn-anti equilibria in the prepared imidoyl chlorides, otherwise found in sterically similar compounds (*11, 13*), was not apparent. A recent attempt to detect the presence of syn-anti isomers of imidoyl chlorides indicated only the presence of anti isomers (*10*). This appears to be consistent with the stereochemistry of imidoyl chlorides prepared from oximes (*7*).

In addition to direct identification, all the imidoyl chlorides were converted to *N,N′*-disubstituted-2-furamidines (*8, 12*). Contrary to Degnan and Pope's observation (*8*), *N*-aryl-2-furamides can be converted to *N*-aryl-*N′*-alkyl-2-furamidines via the imidoyl chlorides in the presence of an alkyl amine. In this case, however, a considerable excess of the alkyl amine has to be used.

The experimental results obtained are presented in Tables I and II. All melting points are uncorrected. Crystalline

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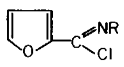
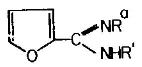
Table I. *N*-Substituted-2-Furimidoyl Chlorides



Compound	R	Reaction Conditions		Yield, %	M.P., ° C.	B.P., ° C./Mm.	Formula		Composition, %				Misc.	
		Temp., ° C.	Time, min.						C	H	N	Cl[a]		
I	C_6H_5—	100	30	93	. . .	161-3/15	$C_{11}H_8ClNO$	Calcd.	64.24	3.92	6.81	17.24	n^{D}_{25}	1.6312
								Found	64.03	3.66	7.03	17.37	d^{25}_{25}	1.2360
II	2′-CH_3·C_6H_4—	100	30	92	. . .	162-4/16	$C_{12}H_{10}ClNO$	Calcd.	65.62	4.59	6.38			
								Found	65.22	4.49	6.58			
III	3′-CH_3·C_6H_4—	100	35	77	. . .	168-70/12	$C_{12}H_{10}ClNO$	Calcd.	65.62	4.59	6.38			
								Found	65.35	4.75	6.61	. . .		
IV	4′-CH_3·C_6H_4—	100	40	78	36-7	176-8/17	$C_{12}H_{10}ClNO$	Calcd.	65.62	4.59	6.38			
								Found	65.74	4.49	6.40	. . .		
V	3′-Cl·C_6H_4—	80	20	86	. . .	126-30/0.5	$C_{11}H_7Cl_2NO$	Calcd.	55.02	2.94		14.77	n^{D}_{25}	1.6354
								Found	55.03	3.04	. . .	14.73	d^{D}_{25}	1.3343
VI	4′-Cl·C_6H_4—	100	25	96	83-4	145-8/1	$C_{11}H_7Cl_2NO$	Calcd.	55.02	2.94		14.77		
								Found	55.00	3.09	. . .	14.68		
VII	3′-NO_2·C_6H_4—	80-90	40	85	97-8	175-80/0.1	$C_{11}H_7ClN_2O_3$	Calcd.	52.71	2.81		14.15		
								Found	52.86	2.88	. . .	14.19		
VIII	4′-NO_2·C_6H_4—	110-140	60	83	120-1	185-8/0.1	$C_{11}H_7ClN_2O_3$	Calcd.	52.71	2.81		14.15		
								Found	52.89	2.92	. . .	14.05		
IX	4′-CH_3O·C_6H_4—	70-85	60	85	44-5	155-60/0.1	$C_{12}H_{11}ClNO_2$	Calcd.	61.15	4.28		15.04		
								Found	61.23	4.27	. . .	14.74		
X	1-$C_{10}H_7$— (α-naphthyl-)	100	50	41	62-3	182-5/2	$C_{15}H_{10}ClNO$	Calcd.	70.46	3.94	5.48			
								Found	70.45	3.88	5.77			
XI	2-$C_{10}H_7$—[b] (β-naphthyl-)	70-90	180	74	60-2	. . .	$C_{15}H_{10}ClNO$	Calcd.	70.46	3.94				
								Found	71.13	3.98	. . .			
XII	CH_3—[c]	60	15	71	. . .	85-7/20	C_6H_6ClNO	Calcd.	50.19	4.21	9.76	24.70		
								Found	50.44	4.14	9.49	24.86		
XIII	C_2H_5—[c]	20	30	74	. . .	82-5/14	C_7H_8ClNO	Calcd.	53.34	5.12		22.50		
								Found	53.20	5.16	. . .	22.70		
XIV	*n*-C_3H_7—	75	60	95	. . .	96-7/13	$C_8H_{10}ClNO$	Calcd.	55.98	5.87		20.66	n^{25}_{D}	1.5206
								Found	55.67	5.78	. . .	20.90	d^{25}_{25}	1.1170
XV	*iso*-C_3H_7—[d,c]	130	60	93	. . .	92-3/13	$C_8H_{10}ClNO$	Calcd.	55.98	5.87		20.66	d^{25}_{D}	1.5188
								Found	55.67	5.82	. . .	20.86		
XVI	*n*-C_4H_9—	100	60	79	. . .	112-13/14	$C_9H_{12}ClNO$	Calcd.	58.22	6.52		19.09	n^{25}_{D}	1.5152
								Found	58.42	6.64	. . .	19.20	d^{25}_{25}	1.0901
XVII	$C_6H_5CH_2$—[e]	100	15	. . .	. . .	Decomp.	$C_{12}H_{10}ClNO$		. . .	. . .	. . .	. . .		
									. . .	. . .	. . .	. . .		
XVIII	*N*-Isopropylbenzimidoyl chloride[f]	100	20	94	. . .	105-7/13	$C_{10}H_{12}ClN$	Calcd.	66.10	6.66				
								Found	66.30	6.67	. . .	. . .	n^{25}_{D}	1.5368

[a] Chlorine analyses were determined by potentiometric titrations of the water–acetic acid hydrolyzates of the imidoyl chlorides; under the conditions given, only imidoyl chloride chlorine was hydrolyzed and determined. [b] The compound was crystallized from ether (110–120° C.). [c] The compound was prepared by treatment (dehydrohalogenation) of the intermediate amide chloride with either benzene or pyridine. [d] The compound was prepared from hitherto unreported *N*-isopropyl-2-furamide, obtained by the Schotten-Baumann procedure from 2-furoyl chloride and isopropyl amine in a 93% yield. The amide crystallized from aqueous ethanol as colorless needles, m.p. 125–126° C. Anal. Calcd. $C_8H_{11}NO_2$: C, 62.72; H, 7.24. Found: C, 62.72; H, 7.24. [e] Owing to the extensive decomposition during the vacuum distillation; the imidoyl chloride was identified as amidine (see Table II, compounds XVIIa and XVIIb). [f] Thionyl chloride instead of phosphorus pentachloride was used.

Table II. *N,N'*-Disubstituted-2-Furamidines



Compound	R	R	R'/R^b	Reaction Conditions: Time, hours^c	Reaction Conditions: Product formed as:^d	Yield, %	M.P., ° C.	Formula		Composition, %: C	H	N	Misc.
Ia	C_6H_5—	C_6H_5—			Hychl.	72^e	91-2	$C_{17}H_{14}N_2O$	Calcd.	77.84	5.38	10.68	Crystl.: pet. ether
									Found	77.72	5.10	10.87	
Ia-Hydrochloride			2	72	Hychl.	85	235-6	$C_{17}H_{15}ClN_2O$	Calcd.	68.33	5.06	9.38	Crystl.: ethanol-ether
									Found	68.22	5.00	9.65	
Ib	C_6H_5—	$C_6H_5\cdot CH_2$—	2.9	1	F.b.	96	89-90	$C_{18}H_{16}N_2O$		...	...	...	Crystl.: pet. ether; no. m.p. depression with XVIIa
Ic	C_6H_5—	CH_3—	9.2	12	F.b.	82	99-100	$C_{12}H_{12}N_2O$	Calcd.	71.98	6.04	13.93	Crystl.: aq. methanol, pet. ether
									Found	72.17	5.88	14.14	
Id	C_6H_5—	C_2H_5—	4.8	12	F.b.	91	80-81	$C_{13}H_{14}N_2O$	Calcd.	72.87	6.59	13.07	Crystl.: aq. ethanol
									Found	73.12	6.57	13.22	
Ie	C_6H_5—	*n*-C_3H_7—	4.2	12	F.b.	100	64-5	$C_{14}H_{16}N_2O$		...	...	...	Crystl.: aq. ethanol; no m.p. depression with XIVb
If	C_6H_5—	*iso*-C_3H_7—	4.2	12	F.b.	100	87-8	$C_{14}H_{16}N_2O$		...	...	...	Crystl.: aq. ethanol; no m.p. depression with XVa
Ig	C_6H_5—	*n*-C_4H_9—	3.9	12	F.b.	76	70-1	$C_{15}H_{18}N_2O$		...	...	...	Crystl.: aq. ethanol; no m.p. depression with XVIa
Ih	C_6H_5—	*tert*-C_4H_9—	4.5	240	F.b.	94	75-6	$C_{15}H_{18}N_2O$	Calcd.	74.35	7.49	11.56	Crystl.: aq. ethanol, pet. ether
									Found	74.39	7.48	11.31	
IIa	2'-$CH_3C_6H_4$—	C_6H_5—			Hychl.	71^e	78-9	$C_{18}H_{16}N_2O$	Calcd.	78.23	5.84	10.13	Crystl.: aq. methanol
									Found	78.46	5.56	10.37	
IIa-Hydrochloride			2.2	72	Hychl.	83	191-2	$C_{18}H_{17}ClN_2O$	Calcd.	69.11	5.48	8.96	Crystl.: ethanol-ether
									Found	69.18	5.60	9.19	
IIIa	3'-$CH_3C_6H_4$—	C_6H_5—			Hychl.	77^e	71-2	$C_{18}H_{16}N_2O$	Calcd.	78.23	5.84	10.13	Crystl.: aq. methanol
									Found	78.48	5.73	10.39	
IIIa-Hydrochloride			2.0	72	Hychl.	87	215-16	$C_{18}H_{17}ClN_2O$	Calcd.	69.11	5.48	8.96	Crystl.: ethanol-ether
									Found	69.32	5.41	9.20	
IVa	4'-$CH_3\cdot C_6H_4$—	C_6H_5—			Hychl.	64^e	91-2	$C_{18}H_{16}N_2O$	Calcd.	78.23	5.84	10,13	Crystl.: pet. ether
									Found	78.56	5.97	10.38	
IVa-Hydrochloride			2.1	72	Hychl.	75	216-17	$C_{18}H_{17}ClN_2O$	Calcd.	69.11	5.48	8.96	Crystl.: ethanol-ether
									Found	69.16	5.20	8.95	
IVb	4'-$CH_3\cdot C_6H_4$—	$C_6H_5CH_2$—	4.0	12	Hychl. + F.b.	94	80-81	$C_{19}H_{18}N_2O$	Calcd.	78.59	6.25	9.65	Crystl.: pet. ether
									Found	78.79	5.99	9.83	
Va	3'-Cl$\cdot C_6H_4$—	C_2H_5—	4.2	12	F.b.	96	104-5	$C_{13}H_{13}ClN_2O$	Calcd.	62.77	5.27	11.27	Crystl.: aq. ethanol
									Found	62.98	5.25	11.46	
VIa	4'-Cl$\cdot C_6H_4$—	C_2H_5—	4.2	12	F.b.	96	105-6	$C_{13}H_{13}ClN_2O$	Calcd.	62.77	5.27	11.27	Crystl.: aq. ethanol
									Found	63.00	5.32	11.29	
VIIa	3'-$NO_2\cdot C_6H_4$—	C_6H_5—	2.0	240	F.b.	58	148-9	$C_{17}H_{13}N_3O_3$	Calcd.	66.44	4.26	13.68	Crystl.: aq. ethanol
									Found	66.24	4.13	13.97	
VIIIa	4'-$NO_2\cdot C_6H_4$—	C_2H_5—	4.0	12	F.b.	97	107-8	$C_{13}H_{13}N_3O_3$	Calcd.	60.22	5.05	16.21	Crystl.: aq. ethanol
									Found	60.46	5.01	16.44	
VIIIb	4'-$NO_2\cdot C_6H_4$—	C_6H_5—	2.0	5 at 80° C.	F.b.	51	154-5	$C_{17}H_{13}N_3O_3$	Calcd.	66.44	4.26		Crystl.: aq. ethanol
									Found	66.70	4.34	...	
IXa	4'-$CH_3O\cdot C_6H_4$—	C_2H_5—	3.8	12	F.b.	100	86-7	$C_{14}H_{16}N_2O_2$	Calcd.	68.83	6.60	11.47	Crystl.: aq. ethanol
									Found	68.94	6.44	11.17	
IXb	4'-$CH_3O\cdot C_6H_4$—	C_6H_5—	2.0	Very fast	F.b.	77	91-2	$C_{18}H_{16}N_2O_2$	Calcd.	73.95	5.52		Crystl.: aq. ethanol
									Found	73.75	5.57	...	
Xa	1-$C_{10}H_7$— (α-naphthyl-)	C_6H_5—	2.2	80	F.b.	85	115-16	$C_{21}H_{16}N_2O$	Calcd.	80.75	5.17	8.98	Crystl.: aq. ethanol
									Found	80.47	4.90	9.07	
XIa	2-$C_{10}H_7$— (β-naphthyl-)	C_6H_5—	2.9	96	F.b.	85	85-6	$C_{21}H_{16}N_2O$	Calcd.	80.75	5.17	8.98	Crystl.: pet. ether
									Found	81.10	4.93	9.09	

(Continued on page 214)

Compound	R	R′	R′/R[b]	Reaction Conditions		Yield, %	M.P., ° C.	Formula	Composition, %				Misc.
				Time, hours[c]	Product formed as:[d]					C	H	N	
XIb	2-$C_{10}H_7$— (β-naphthyl-)	$C_6H_5CH_2$—	2.0	12	F.b.	96	121-2	$C_{22}H_{18}N_2O$	Calcd. Found	80.95 80.80	5.56 5.53	5.58 8.61	Crystl.: aq. ethanol
XIIa	CH_3—	C_6H_5—	1.4	Very fast	Hychl.	88	99-100	$C_{12}H_{12}N_2O$	Calcd. Found	. . . . . .	. . . . . .	16.31 16.40	Crystl.: pet. ether; no m.p. depression with Ic
XIIa-Picrate			. . .	. . .	. . .	74	106 8	$C_{18}H_{15}N_5O_8$	Calcd. Found			16.31 16.40	Crystl.: aq. ethanol
XIIIa	C_2H_5—	C_6H_5—	2.0	Very fast	Hychl.	85	80-1	$C_{13}H_{14}N_2O$		. . .	. . .	. . .	Crystl.: aq. ethanol; no m.p. depression with Id
XIVa	*n*-C_3H_7—	*iso*-C_3H_7—	2.2	12	F.b.	99	B.P.,	$C_{11}H_{18}N_2O$	Calcd. Found	68.01 67.96	9.34 9.27		
XIVb[f]	*n*-C_3H_7—	C_6H_5—	2.1	Very fast	Hychl.	72	64-5	$C_{14}H_{16}N_2O$		. . .	. . .	. . .	Crystl.: aq. ethanol
XVa	*iso*-C_3H_7—	C_6H_5—	1.9	Very fast	Hychl.	70	87-8	$C_{14}H_{16}N_2O$	Calcd. Found	73.65 73.60	7.07 7.17	12.27 12.55	Crystl.: aq. ethanol
XVIa[f]	*n*-C_4H_9—	C_6H_5—	1.2	Very fast	Hychl.	84	70-1	$C_{16}H_{18}N_2O$		. . .	. . .	. . .	Crystl.: aq. ethanol
XVIIa	$C_6H_5CH_2$—	C_6H_5—	. . .	Very fast	Hychl.	. . .	89-90	$C_{18}H_{16}N_2O$	Calcd. Found	78.24 78.51	5.84 5.68	10.14 10.18	Crystl.: aq. ethanol
XVIIb	$C_6H_5CH_2$—	4′-$CH_3\cdot C_6H_4$—	~1	96	Hychl.	. . .	80-1	$C_{19}H_{18}N_2O$		. . .	. . .	. . .	Crystl.: pet. ether; no m.p. depression with IVb
	N-Isopropyl-*N*′-phenylbenzamidine			Very fast	Hychl.	90	96 7	$C_{16}H_{18}N_2$	Calcd. Found	80.63 80.89	7.61 7.84	11.76 12.04	Crystl.: aq. ethanol

[a] Owing to the tautomerism of *N,N*′-disubstituted amidines, R and R′ cannot be distinguished. However, in this table, R indicates the substituent originally present in the imidoyl chloride and R′ the substituent originally present in the primary amine. [b] R′/R indicates the molar ratio of primary amine and imidoyl chloride in the reaction mixture. [c] Unless otherwise indicated, the reactions were performed at room temperature. [d] This column indicates form (Hychl.: amidine hydrochloride, F.b.: amidine free base) in which the product is originally obtained from the reaction mixture. [e] An over-all yield. [f] See reference (*8*).

analytical samples were derived by 2 to 3 recrystallizations from a suitable solvent followed by drying at 0.5 to 15 mm. for 2 to 10 hours at room temperature over P_2O_5.

The *N*-monosubstituted-2-furamides were prepared from 2-furoyl chloride and the appropriate primary amine, according to the Schotten-Baumann method, or by use of pyridine as solvent (*1, 2, 5, 6, 14*).

N-Substituted-2-furimidoyl chlorides (I–XVII). The imidoyl chlorides (Table I) were prepared by the following general procedure: In a Claisen flask equipped with a reflux condenser and a drying tube, an intimate mixture of the *N*-monosubstituted amide and PCl_5, in approximately 1- to -1 ratio, was heated from 15 minutes to 3 hours over 60° to 140° C. After completion of the reaction, the phosphorus oxychloride was removed under reduced pressure at 40° to 50° C., after which the remaining imidoyl chloride was distilled under vacuum. An analytical sample was obtained by redistillation.

If the reaction product was a stable amide chloride (XII, XIII, XV), the $POCl_3$ was removed under vacuum and the residue boiled under reflux of benzene for 1 to 2.5 hours, followed by removal of solvent; the resulting imidoyl chloride was purified as before.

N,N′-Disubstituted-2-furamidines. 2-Furamidines (Table II) were prepared in the following general way: In an Erlenmeyer flask equiped with a drying tube, the imidoyl chloride was dissolved in 2.5–10-fold amount of dry benzene, and a sufficient amount of primary amine was added to give a 20 to 400% molar excess. The mixture was allowed to stand at room temperature. The crystalline amidine hydrochloride was filtered, washed with benzene, and recrystallized from a mixture of ethanol-ether. The free amidine was liberated from an aqueous solution by addition of concentrated ammonia. The crude solid was filtered, washed with water, and recrystallized from either ethanol-water, methanol-water, or 60° to 80° C. petrol ether. If the crystalline reaction product consists of the amine hydrochloride, the free amidine may be obtained by filtration followed by evaporation of the benzene, and the amidine purified as above.

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