excellent agreement with a variety of data for hydrogen-transfer reactions and appears accurately to reproduce the chemical behavior of these transition states.

Comparison is also possible with end-atom effects in other nucleophilic displacements; examples were given in the paper on the reacting-bond rule.[7]

In addition, the model shows a decrease in BC strength with an increase in AB strength and *vice versa;* this is just the "alternating effect" of the reacting-bond rule.

Our earlier attempts at theoretical justification of the rule were limited to bond-dissociating or bond-associating reactions involving two atoms (two electrons and two nuclei) only.[7]

Figure 3 shows that electron withdrawal from the center atom of a nucleophilic-displacement transition state should result in the strengthening of the bonds to both entering and leaving groups, while conversely electron release to the center atom should weaken both bonds. Again the model is in exact agreement with the reacting-bond rule; examples of isotope effects and other data following the rule were given previously.[7]

Furthermore, the model predicts (Figure 3) that electron withdrawal at the center atom should produce an accumulation of negative charge on the center atom whereas electron release to the center atom should result in a loss of reacting-electron density and thus an increase in positive charge on B. If the Hammett reaction constant ρ is taken as a measure of the charge on B, then nucleophilic displacements on benzyl compounds fit the model very well. These reactions display curved Hammett plots generally possessing a minimum; thus relatively electron-releasing substituents produce a negative ρ (B relatively low in electron density) while relatively electron-withdrawing substituents produce a positive ρ (B relatively high in electron density).

We conclude that the model represents the substituent effects in hydrogen-transfer and other nucleophilic-displacement transition states in considerable detail.

Scope of the Model.—The reason for the failure of the model to account for normal, stable hydrogen-bond behavior in spite of its good success in reproducing substituent effects in transition states lies in an assumption made in constructing the figures: that the electron cloud is sufficiently polarizable and the nuclei sufficiently far apart that the electron shift due to changes in nuclear electronegativity greatly outweighs the increased or decreased nuclear repulsions arising from the same variation. Note that the nuclei were assumed to remain stationary in constructing all the figures.

This assumption is very poor for hydrogen bonds, where one pair of electrons is nearly localized in the lone-pair orbital of the base and the other is involved in a strong σ-bond. However, in transition states for nucleophilic displacement where the internuclear distances are abnormally long and where the electron cloud is especially polarizable, the assumption is apparently nearly enough true to produce excellent agreement with experiment.

The agreement of this model with the reacting-bond rule suggests that the success of the reacting bond rule may be due to (a) the high polarizability of the electron cloud in the transition states to which it was applied and (b) the relative constancy of the transition-state internuclear distances with the structural variations considered. If, in the future, cases arise which fail to follow the reacting-bond rule they should be examined for violation of one of these restrictions.

The Nitro Group as an *ortho* Participant in the Dissociation of Iodobenzene Dichloride

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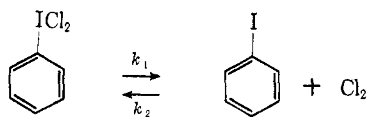
Evidence is presented that an o-NO_2 substituent participates as an internal nucleophile in promoting the dissociation of iodobenzene dichloride in acetic acid while the o-CN group does not. The capacities of the o-NO_2 and o-$COOCH_3$ groups to enhance the dichloride dissociation rate are comparable in magnitude.

The rate of dissociation of iodobenzene dichloride (eq. 1) in a polar solvent such as acetic acid is subject to marked rate enhancement when certain substituents which are nucleophilic in nature are located *ortho* to the reaction center.[1] Presumably these *ortho* substituents promote the reaction by releasing electrons to the iodine atom as it undergoes positive polarization in the activation process. Of all the *ortho* substituents investigated, those which contribute most to reactivity of the dichloride are the carbomethoxy and structurally closely related groups.[2,3] In fact o-carbomethoxyiodobenzene dichloride equilibrates with its components in acetic acid too rapidly to permit even a semiquantitative investigation of the reaction rate by the spectrophotometric method employed in earlier work on C_6H_5-ICl_2 and its derivatives. Evidence has also been presented that COOR groups participate as internal nucleo-



(1)

$K = k_1/k_2$

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[1] For a recent survey see L. J. Andrews, L. J. Spears, and R. M. Keefer, *J. Am. Chem. Soc.*, **86**, 687 (1964).

[2] L. J. Andrews and R. M. Keefer, *ibid.*, **81**, 4218 (1959).

[3] R. M. Keefer and L. J. Andrews, *ibid.*, **81**, 5329 (1959).

philes in the hydrolysis of *ortho*-substituted benzhydryl halides.[4,5]

The nitro group is so constituted electronically that it seems a likely candidate to function effectively as an internal nucleophile.[6] Experimental attention has been given to the possibility that an o-NO_2 substituent may participate in the hydrolysis of benzyl[7] and benzhydryl[5] halides, but no definitive results have been obtained. Because of close similarities in the spectra of the free iodobenzene and its dichloride it has not proved feasible to investigate the ring substituent effect on the dissociation rate of o-nitroiodobenzene dichloride by spectrophotometric procedures. A volumetric method has now been applied in establishing the fact that the o-NO_2 group does make an important contribution as an internal nucleophile to the course of this reaction. This method has been used successfully in making a semiquantitative determination of the k_1-values for the reactions of the o-carbomethoxy- as well as for the o- and p-nitroiodobenzene dichlorides, and it has also been adapted to the study of the dissociation rates of the o- and p-cyanoiodobenzene dichlorides.

Experimental

Dry Acetic Acid.—A mixture of 2 l. of acetic acid and 200 ml. of benzene was fractionally distilled on a column packed with glass helices. Fractions with freezing points below 16.6° were discarded. The remaining distillate was retained for use as solvent in the kinetic experiments.

Chlorine and Hydrogen Chloride.—Commercial samples of these two gases were dried before use by passing them through concentrated sulfuric acid. Solutions of chlorine in acetic acid were analyzed for the halogen by iodometric methods which have been described previously.[3] Solutions of hydrogen chloride in acetic acid were analyzed for the hydrogen halide by diluting samples with water and titrating the resultant mixtures with standard silver nitrate solutions by the customary procedures.

Durene.—Eastman Organic Chemicals durene was recrystallized several times from ethanol before use in rate work, m.p. 79°.

The Iodobenzenes.—The iodonitrobenzenes, the methyl iodobenzoates, and p-iodobenzonitrile were obtained from Eastman Organic Chemicals. The o- and p-iodonitrobenzenes were recrystallized from aqueous ethanol before use and had m.p. 53° and 172–173°, respectively.

A sample of o-iodobenzonitrile was prepared from o-iodobenzamide[3] by a procedure similar to that used in converting nicotinamide to nicotinonitrile.[8] A mixture of 2.0 g. of the iodobenzamide and 1.0 g. of phosphorus pentoxide was heated to the melting point in a distillation apparatus. The pressure in the system was then reduced to 15 mm. and o-iodobenzonitrile was distilled at 145–147°. The crude product, which solidified on cooling, was recrystallized several times from aqueous ethanol. The nitrile, m.p. 53.5–54.5° (lit.[9] m.p. 54–55°), was obtained in 60% yield.

Anal. Calcd. for C_7H_4IN: C, 36.70; H, 1.76; N, 6.12. Found: C, 36.77; H, 1.65; N, 5.80.

The Dichlorides.—The solid dichlorides of the substituted iodobenzenes generally were prepared immediately before use in rate work. The methods of preparation of the derivatives of p-iodonitrobenzene and of the methyl o- and p-iodobenzoates have been described previously.[2,10] The other dichlorides were obtained by passing chlorine gas through solutions of 0.3–0.5-g. samples of the iodo compounds in small volumes (2–7 ml.) of dry acetic acid. Precipitation of the yellow crystalline solids generally occurred within 0.5 hr. The crystals were filtered, washed with carbon tetrachloride, and air dried briefly before analysis. The equivalent weights of the freshly isolated solids were determined iodometrically.[10] The equivalent weights and melting points of dichlorides not described in previous reports by the authors are listed in Table I.

TABLE I

PROPERTIES OF THE DICHLORIDES ($XC_6H_4ICl_2$)

X	Equiv. wt. Calcd.	Equiv. wt. Found	M.p., °C.
o-NO_2	160	165	80–82
o-CN	150	146	73–75
p-CN	150	150	88–90

Kinetic Studies.—The experimental procedures employed in rate runs to determine k_1-values for the several dichlorides under investigation were designed to take advantage of the fact that free chlorine reacts extremely rapidly with durene (at moderate concentration) in acetic acid; the undissociated aryliodoso dichlorides, on the other hand, do not undergo direct reaction with durene in acetic acid.[1] In the presence of durene the iodoso dichlorides undergo complete decomposition to the free iodobenzene rather than to an equilibrium mixture of the dichlorides and their components.

Two different types of rate runs were made. One set of runs was based on the use of freshly precipitated samples of the crystalline dichlorides as starting materials. Solutions of the dichlorides (0.02–0.05 M) in acetic acid were prepared and brought to the temperatures at which the runs were to be conducted. The total halogen content (free halogen and dichloride) of these solutions was established by iodometric methods by adding 2-ml. aliquots of the solutions to large volumes of dilute aqueous potassium iodide solutions and titrating for liberated iodine with standard (0.01 N) sodium thiosulfate solution to the starch end point. To start runs, samples of these solutions were mixed with equal volumes of solutions of durene (0.2–0.5 M) in acetic acid which previously had been adjusted to the appropriate reaction temperature. At suitable time intervals, known volumes of the reaction mixtures were removed for iodometric analysis (eq. 2) for undissociated dichloride.

$$\mathrm{ArICl_2} + 3\mathrm{I^-} \longrightarrow \mathrm{ArI} + \mathrm{I_3^-} + 2\mathrm{Cl^-} \qquad (2)$$

In other runs, measured volumes of dilute acetic acid solutions of free chlorine and free iodobenzene of known concentration were mixed at the temperature of the rate measurement. These solutions were allowed to stand sufficiently long (usually overnight) to provide for equilibration of the reactants with the dichloride. Runs were initiated by mixing known volumes of these equilibrated solutions with known volumes of durene in acetic acid. Rate samples were removed from time to time for analysis for undissociated dichloride (again by the iodometric method). Rate constants k_1 (eq. 3) were calculated from the slopes of plots of values of log $[\mathrm{ArICl_2}]_{\mathrm{undissocd}}$ *vs.* time ($k_1 = 2.3 \times$ slope). The

$$-\mathrm{d}[\mathrm{ArICl_2}]/\mathrm{d}t = k_1[\mathrm{ArICl_2}] \qquad (3)$$

runs generally were followed to completion. In very fast runs in which a number of rate samples were taken in a relatively short period the reaction time was recorded when half the sample had been transferred to the potassium iodide solution. In some cases rate constants calculated towards the ends of the runs were observed to drift upward with time. This was ascribed to catalysis by hydrogen chloride accumulating in the medium from the reaction of chlorine and durene. That such catalysis occurred was readily demonstrated by conducting runs in which substantial amounts of hydrogen chloride were initially present in the reaction mixtures. The rate constants for runs at 15° in which the initial concentrations of p-nitroiodobenzene dichloride and durene were both about 0.01 M were 0.12×10^{-4} sec.$^{-1}$ (initially) in the absence of added hydrogen chloride and 0.46×10^{-4} sec.$^{-1}$ when the initial hydrogen chloride concentration was

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[4] A. Singh, L. J. Andrews, and R. M. Keefer, *J. Am. Chem. Soc.*, **84**, 1179 (1962).

[5] E. A. Jeffery, R. K. Bansal, L. J. Andrews, and R. M. Keefer, *J. Org. Chem.*, **26**, 3365 (1964).

[6] It has been suggested that the nitro group acts in this manner in promoting ionic scission of the sulfur-chlorine bond of 2,4-dinitrobenzenesulfenyl chloride (see N. Kharasch, "Organic Sulfur Compounds," Vol. 1, Pergamon Press Ltd., Oxford, England, 1961, p. 379). This proposal is supported by recent findings of W. C. Hamilton and S. J. La Placa [*J. Am. Chem. Soc.*, **86**, 2289 (1964)].

[7] J. B. Hyne and R. Wills, *ibid.*, **85**, 3650 (1963).

[8] P. C. Teague and W. A. Short, "Organic Syntheses," Coll. Vol. IV, John Wiley and Sons, Inc., New York, N. Y., 1963, p. 706.

[9] I. Heilbron, "Dictionary of Organic Compounds," Vol. III, Oxford University Press, New York, N. Y., 1953, p. 22.

[10] R. M. Keefer and L. J. Andrews, *J. Am. Chem. Soc.*, **80**, 277 (1958).

0.13 *M*. Generally the k_1-values reported below are based on those phases of the runs in which the plots of log $[ArICl_2]$ *vs.* time remained linear.

The k_1-values for the dichlorides of *o*-nitroiodobenzene and methyl *o*-iodobenzoate which are reported are based on runs in which the dichloride initially present was generated by mixing solutions of the free iodo compound and a substantial excess of chlorine. In dilute solution in acetic acid these dichlorides dissociate so extensively and so rapidly that freshly prepared solutions of the dichlorides themselves cannot be used successfully in the determination of k_1-values.

Estimation of *K* for *o*-Nitroiodobenzene Dichloride.—Known volumes of solutions of known concentrations of *o*-nitroiodobenzene and of chlorine in acetic acid were mixed and allowed to equilibrate with respect to dichloride formation. A sample of known volume of the equilibrated solution was mixed with a known volume of a dilute solution of durene in acetic acid, and the concentration of dichloride in the resultant solution *vs.* time was established by a series of iodometric analyses. The dichloride content of the solution at initial time (when durene was first added) was then determined by extrapolation of a plot of values of log $[ArICl_2]_{\text{undissocd}}$ *vs.* time. This provided the information essential for the calculation of values of $[Cl_2]$, $[ArI]$, and $[ArICl_2]$ in the durene-free equilibrated solution.

The Recovery of *o*-Nitroiodobenzene from a Solution of Its Dichloride.—The following experiment was conducted to demonstrate that *o*-nitroiodobenzene could be recovered from its dichloride solutions without alteration in structure. A 0.24-g. sample of the dichloride was dissolved in acetic acid, and the resultant solution was allowed to stand for 15 min. to allow time for equilibration with the free iodo compound and chlorine. A 4-ml. sample of a 1.2 *M* solution of potassium iodide was added followed by 2.2 ml. of 0.5 *N* sodium thiosulfate solution. The yellow oil which separated during the course of these additions soon solidified to provide an 80% yield of crystalline *o*-nitroiodobenzene, m.p. 51–52°.

Results

A summary of the runs conducted in connection with this investigation is presented in Table II. The k_1-values listed in this table are reported to two significant figures. The values for the *o*-nitro- and *o*-carbomethoxyiodobenzene dichlorides are, however, probably reliable only to one significant figure. The half-lives for these compounds (under the conditions of the runs which were made) were less than 1 min., and the times recorded in sampling the reaction mixtures were subject to considerable error.

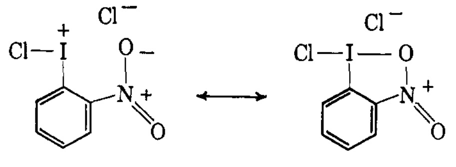
TABLE II

RATE CONSTANTS FOR DISSOCIATION OF $XC_6H_4ICl_2$ IN ACETIC ACID[a]

$[ArI]_i$, *M*	$[Cl_2]_i$, *M*	$[ArICl_2]_i$, *M*	[Durene]$_i$, *M*	Temp., °C.	$10^3 \times k_1$, sec.$^{-1}$
X = *p*-NO_2					
0.0123	0.0421		0.067	15.0	0.022[b]
0.0133	0.0215		0.066	15.0	0.018[b]
		0.0120	0.204	15.0	0.011
		0.0113	0.098	15.0	0.012
			Av.	(15.0)	0.016 ± 0.004
		0.0069	0.102	24.9	0.041
X = *o*-NO_2[c]					
0.0247	0.0612		0.211	15.0	13
0.0131	0.122		0.236	15.0	14
0.0131	0.0428		0.228	15.0	17
0.0123	0.0784		0.220	15.0	14
0.0121	0.0392		0.109	15.0	18
			Av.	(15.0)	15 ± 2
0.0129	0.0465		0.228	25.0	38
X = *p*-CN					
		0.0121	0.094	15.0	0.015
		0.0108	0.092	25.0	0.046
		0.0104	0.194	25.0	0.041
			Av.	(25.0)	0.044 ± 0.003
X = *o*-CN					
		0.0125	0.091	15.0	0.021
		0.0167	0.187	25.0	0.064
		0.0118	0.102	25.0	0.067
			Av.	(25.0)	0.065 ± 0.002
X = *p*-$COOCH_3$					
0.0114	0.0612		0.208	25.0	0.077
X = *o*-$COOCH_3$					
0.0109	0.0580		0.227	15.0	28
0.0108	0.0580		0.102	15.0	23
			Av.	(15)	26 ± 3
X = H					
		0.0140	0.099	25.0	0.091

[a] In the particular batch of solvent which was used in these rate runs, reactions were generally a bit slower than they were in the samples of acetic acid used in earlier work. The differences are assumed to reflect differences in trace-water content of different batches of "purified" acetic acid. Values of k_1 (25°) which have been reported previously[1,10] [see also R. M. Keefer and L. J. Andrews, *J. Am. Chem. Soc.*, **80**, 5350 (1958)] for the dichlorides of iodobenzene and its *p*-NO_2 and *p*-$COOCH_3$ derivatives are 0.17×10^{-3}, 0.089×10^{-3}, and 0.15×10^{-3}, respectively. [b] These values are somewhat in error, on the high side, because of catalysis by hydrogen chloride generated at the start by reaction of free chlorine and durene. [c] *K*-values evaluated as outlined in the Experimental section were 0.12, 0.18, 0.13, and 0.16 (av. 0.14) l./mole at 15.0° (based on the first five runs on *o*-nitroiodobenzene dichloride) and 0.18 l./mole at 25.0°.

In cases in which the substituents are not functionally capable of participation, the dissociation rates of *ortho*-substituted iodobenzene dichlorides are not much different from those of their *para* isomers.[11] At 15° the *ortho* isomer of nitriodobenzene dichloride is of the order of 750–1000 times as reactive as its *para* isomer. This reactivity ratio is considerably larger than most of those (18 to 170) which have been cited[1,3] as evidence of *ortho* substituent (*e.g.*, $-CH_2OH$, $-CH_2OCH_3$, –CO–R) participation. Through an oversight not discovered until this investigation was terminated, the reactivities of the *o*- and *p*-carbomethoxyiodobenzene dichlorides were not measured at the same temperature. As noted above, however, the k_1-value reported (Table II) for the *ortho* isomer at 15° is probably reliable to no more than one significant figure. A comparison of the *o*/*p* reactivity ratios of the carbomethoxy dichlorides, roughly as adequate as that based on rate studies of the two compounds at the same temperature, can therefore be made by assuming that k_1 at 15° for the *para* isomer is one-third that found (Table II) for reaction at 25°. This is approximately the ratio of k_1-values at 15 and 25° for the *o*-cyano- and *p*-cyano- and the *p*-nitroiodobenzene dichlorides. On this basis it is calculated that at 15° the *o*-carbomethoxy compound is about 10^3 times as reactive as its *para* isomer. The *o*-nitro and *o*-carbomethoxy groups are, therefore, about equally effective, and dramatically so, as participating nucleophiles in promoting iodobenzene dichloride dissociation. The suggested contribution of the *o*-NO_2 group in providing for transition state stabilization is depicted below.



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[11] R. M. Keefer and L. J. Andrews, *J. Am. Chem. Soc.*, **81**, 2374 (1959).

The relative ineffectiveness of the nitro group as a participant in benzhydryl bromide hydrolysis[5] is tentatively ascribed to a geometric situation in the transition state which is unfavorable for electron release by the *ortho* substituent to the vacant p-orbital on the exocyclic carbon atom.[12] Because of the much larger size of the iodine orbital in question this geometric problem apparently does not arise in the *o*-nitroiodobenzene dichloride dissociation reaction.

It is particularly interesting to observe that the *o*- and *p*-cyanoiodobenzene dichlorides are comparable in reactivity. The *o*-CN group, like the *o*-NO_2 and *o*-$COOCH_3$ groups, is potentially nucleophilic in character. The fact that it is nonparticipating can only be explained on geometric grounds. The two atoms of the substituent and the ring carbon to which it is attached are linear. Neither the two π-orbitals of the triple bond nor the p-orbital of the nitrogen atom are properly oriented to release electrons to iodine in the activation process for dichloride dissociation.

Thermodynamic Constants.—Activation energies and entropies for dissociation of the *o*- and *p*-cyano- and *p*-nitroiodobenzene dichlorides (calculated from the results presented in Table II) are listed in Table III. The E_a and ΔS^* values are similar to those reported previously for other dichlorides.

TABLE III

THERMODYNAMIC CONSTANTS FOR DISSOCIATION OF $XC_6H_4ICl_2$ IN ACETIC ACID

X	E_a, kcal./mole	$-\Delta S^*$, e.u.
p-NO_2	21 ± 1	10 ± 3
p-CN	18 ± 2	20 ± 6
o-CN	19 ± 1	16 ± 3

Acknowledgment.—The authors are indebted to the National Science Foundation for a grant in support of this research.

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[12] For evidence of *o*-NO_2 group participation in certain reactions of benzhydrol, see, however, W. B. Dickinson, *J. Am. Chem. Soc.*, **86**, 3580 (1964).

Mechanism of the Gabriel–Colman Rearrangement

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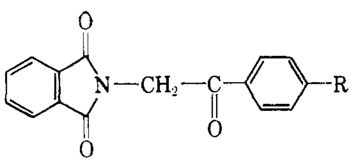
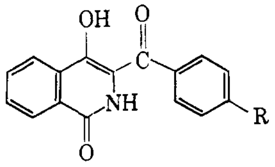
The kinetics of the methoxide-promoted rearrangement of six *p*-substituted N-phenacylphthalimides to the corresponding 3-benzoyl-4-hydroxyisocarbostyrils have been investigated. A mechanism involving opening of the phthalimide ring followed by rearrangement of the resulting imide anion to the carbanion and ring closure is suggested by the identical rates of reaction and products from rearrangement of N-phenacylphthalimide and of methyl N-phenacylphthalamate and by the decreased rate observed when *t*-butoxide was used with the former. The reaction exhibits a linear Hammett plot with $\rho = 1.98$. Cleavage of the 3-benzoyl-4-hydroxyisocarbostyrils by methoxide is a minor side reaction.

Alkoxides effect rearrangement of N-phenacylphthalimides (I) to 3-benzoyl-4-hydroxyisocarbostyrils (II).[1] This, the Gabriel–Colman rearrangement, affords a convenient though little used[2] method for synthesis of isoquinolines. This ring expansion appears to be general when the group attached to nitrogen has an enolizable hydrogen.[3]

Interest in this reaction arose from its similarity to the facile methoxide-promoted rearrangement of N-tosylamidophthalimide to phthalhydrazide[4] and because its mechanism had not been investigated in detail before. Accordingly, the kinetics of the Gabriel–Colman rearrangement of six *p*-substituted N-phenacylphthalimides (Ia–f) to the corresponding 3-benzoyl-4-hydroxyisocarbostyrils (IIa–f) in anhydrous methanol containing sodium methoxide were measured. The rates of formation of IIa–f were determined spectrophotometrically. After removal from ampoules, reaction aliquots were quenched in 10% concentrated hydrochloric acid in methanol and the concentrations of II were estimated at their absorbance maxima in the ultraviolet. A recent report[5] states that comparisons of the spectra of 1,4-naphthoquinone and 1,4-dihydroxynaphthalene with those of Gabriel–Colman rearrangement products indicates that these products exist in the 2,3-dihydro-1,4-dione form in acidic solution and as the enolates of the 4-hydroxy form in basic solution. Consequently in the quenched solutions, which were acidic, the dione form was the only form present during estimation. The reaction products were stable in acidic solution and obeyed Beer's Law with no apparent sensitivity to the acid concentration over a wide range. The kinetic measurements indicate that the reaction is first order in both I and methoxide over about a fourfold ratio of concentrations.[6] The kinetic data are presented in Table I.



I



II

a, R = OCH_3
b, R = CH_3
c, R = H
d, R = C_6H_5
e, R = Br
f, R = NO_2

Two mechanisms of reaction have been suggested previously. In the first,[7] reaction of methoxide with I produces an equilibrium concentration of carbanion

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[1] S. Gabriel and J. Colman, *Ber.*, **33**, 980 (1900).

[2] A. Ulrich, *ibid.*, **37**, 1689 (1904); H. Kusel, *ibid.*, **37**, 1971 (1904).

[3] Saccharine derivatives are converted into 1,2-benzothiazines [K. Abe, S. Yamamoto, and K. Matsui, *Yakugaku Zasshi*, **86**, 1058 (1956)] or 1,3-benzothiazines [H. Zinnes, R. A. Comes, and J. Shavel, Jr., *J. Org. Chem.*, **29**, 2068 (1964)], and during the current work it was found that N-phenacylbenzoyleneureas yield benzodiazepindiones.

[4] Unpublished results.

[5] L. R. Caswell and R. D. Campbell, *J. Org. Chem.*, **26**, 4175 (1961).

[6] See Experimental section.

[7] W. J. Gensler, "Heterocyclic Compounds," Vol. 4, R. C. Elderfield, Ed., John Wiley and Sons, Inc., New York, N. Y., 1952, p. 378.