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PROTIC ACID-INDUCED INTRAMOLECULAR REACTIONS OF 2-CYCLOPROPYLAZOBENZENES

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Treatment of 2-cyclopropylazobenzenes with concentrated sulfuric acid affords the intramolecular N- and C-alkylation products. Treatment of azobenzenes with trifluoroacetic acid results in quantitative rearrangement to arylindazoles via the intermediate formation of acid-stable indazolium ions. It is suggested that the formation of indazolium ions in trifluoroacetic acid results from the synchronous opening of the cyclopropane ring and stabilization of the developing carbocation by an internal nucleophile (the azo-group).

We have previously shown [1] that 2-cyclopropylazobenzene (I) on treatment with concentrated sulfuric acid is converted into the N-arylaminoindole (III) and the azoalcohols (IV) and (V), the percentages and ratios of which remain constant with time. We were able to show that the aminoindole (III) is formed, not from the arylcyclopropane (I), but from its isomerization product, ortho-propenylazobenzene (II). *(See scheme on top of next page.)

The key step in the conversion of compound (I) is the formation of the bemzyl carbocation (A). Azobenzenes containing in the orthoposition a substituent which is capable under the reaction conditions of giving rise to the corresponding carbocation are known to be able to undergo intramolecular reactions to give the indazoles [2, 3].*

*It should be pointed out that the synthesis of indazoles from the appropriate orthosubstituted azobenzenes is of limited usefulness as a result of the relative inaccessibility of the starting materials.

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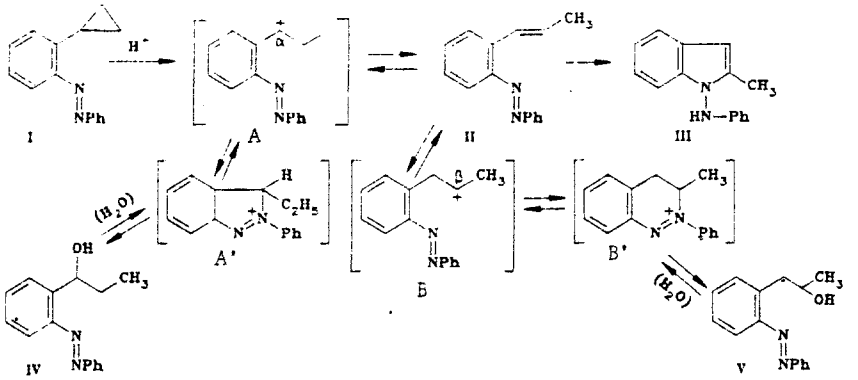
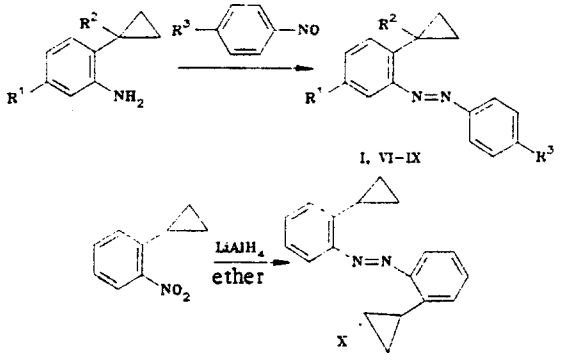


In order to establish the feasibility of synthesizing indazoles from 2-cyclopropylazobenzenes, and the effects of some factors on their formation, we have examined the reaction of 2-cyclopropylazobenzenes with protic acids of different strengths (sulfuric, trifluoroacetic, formic, and acetic) at a range of temperatures.

The cyclopropylazobenzenes (I) and (VI-X) required for this investigation were obtained as follows:



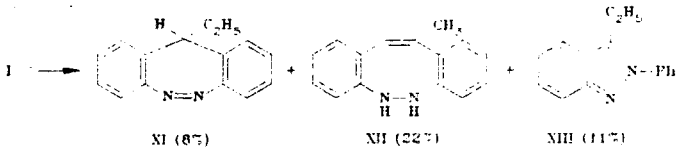
I, VI–IX

X

I $R^1=R^2=R^3=H$; VI $R^1=R^3=H$, $R^2=CH_3$; VII $R^1=Br$, $R^2=R^3=H$; VIII $R^1=i\text{-}C_4H_9$, $R^2=R^3=H$; IX $R^1=R^2=H$, $R^3=Cl$

The conversion of (I) into the N-arylaminoindole (III) on treatment with concentrated sulfuric acid occurred at relatively low temperatures (−20 to −25°C). On the assumption that reducing the temperature would promote the regioselectivity of the reaction and increase the yield of N-phenylaminoindole, and that increasing it would modify the course of the reaction, we examined the reaction of (I) with concentrated sulfuric acid at temperatures below and above those employed in [1].

It was found that at lower temperatures (−30 to −40°C), substantial amounts of (I) were recovered unchanged, and the relative yield of N-phenylaminoindole even decreased. Increasing the reaction temperature to −10°C resulted in the mixture of products becoming more complex, and in addition to the products (III-V) previously reported [1], there were also obtained 11-ethyl-11H-dibenzo[c, f][1,2]diazepine (XI), 5,6-dihydro-11-methyldibenzo[c,g][1,2] diazocine (XII) and 3-ethyl-2-phenylindazole (XIII).

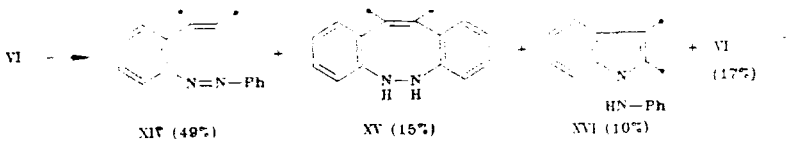


XI (8%) XII (22%) XIII (11%)

+ IV (9%) + III (30%) + V (5%)

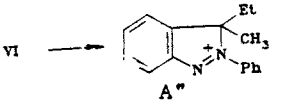
Further increases in temperature to 0°C or even 20°C had no effect on the qualitative composition of the mixtures obtained, but the yields of products were reduced considerably as a result of the formation of water-soluble materials.

In contrast to the substrate (I), at −10°C (VI) gave a mixture of three products (XIV-XVI), and substantial amounts of the starting material (VI) were recovered unchanged. No alkylarylindazole was obtained in this case.



XII (49%) XV (15%) XVI (10%)

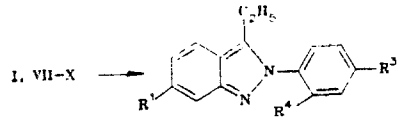
It is important to note that the azoalcohols (IV) and (V) and the alkenylazobenzene (XIV) are not direct products of the reaction of cyclopropanes (I) and (VI) with concentrated sulfuric acid, being formed only following hydrolysis of the reaction mixtures. It was found that treatment under the conditions adopted of the pure compounds (IV), (V), and (XIV) with concentrated sulfuric acid followed by water gave in each instance mixtures of products formed by the reaction of the cyclopropylazobenzenes (I) and (VI) with the same reagent. In other words, during the reaction of (I) and (VI) with concentrated sulfuric acid, the intermediate carbocations at C_α (A) and C_β (B) gave intermediates which are reasonable for the formation of the azoalcohols (IV) and (V), and the alkenylazobenzene (XV), and are stable under the reaction conditions until the reaction mixture is treated with water. In the case of the azobenzene (I), these intermediates are clearly the cyclic ions A' and B', and in the case of (VI), the cyclic ions A". It is known [4] that in trifluoroacetic acid (VI) gives a quantitative yield of the salt with the organic cation A".



A"

Following treatment of the reaction mixture with water, the azobenzene (VI) gives the alkenylazobenzene (XIV) rather than the corresponding azoalcohol (XVII), as is the case in the reaction of the azobenzene (I). In all likelihood, nucleophilic attack of water on the potential carbenium center at ions C' is sterically hindered. In this case, water functions as a base, removing a proton from the β-position of the alkyl group, with the consequent formation of the alkenyl moiety.

Concentrated sulfuric acid is used in the isomerization of arylcyclopropanes bearing strong electron acceptor groups in the orthoposition to the small ring, such as the nitro- [5] or azoxy-groups [1]. If, however, such groups are absent from the phenylcyclopropane, then opening of the three-membered ring to give the corresponding carbonium intermediate may also occur with weaker acids. Our studies have shown that 2-cyclopropylazobenzene (I) does not react with either acetic or formic acids even at their boiling points, but treatment with trifluoroacetic acid at temperatures as low as 20°C results in partial (~30%) conversion to the indazole (XIII). When the temperature is increased to 60-70°C, this reaction is complete after 2 h. It is interesting that although treatment of the azoalcohol (IV) with concentrated sulfuric acid gives a complex mixture of (III-V) and (XI-XIII), similar treatment with trifluoroacetic acid affords exclusively the indazole (XIII). The nature of the substituents present in the aromatic rings of the azobenzenes (I) in the 5, 2', or 4' positions has no effect on the course of the reaction, and little effect on the yields of arylindazoles (Table 1).



XIII, XVIII–XXI

The high regioselectivity of the reactions of the 2-cyclopropylazobenzenes (I) and (VII-X) in the presence of trifluoroacetic acid is probably due to the following factors: 1) the existence of an equilibrium between the nitrogen-protonated and unprotonated forms of the azobenzene; 2) the ability of the azo-group to display nucleophilic assistance to the opening of the protonated cyclopropane ring; and 3) the ability of cyclic ions such as A not to undergo modification on treatment with base (water). The fact that an important (or decisive) role in the opening of the cyclopropane ring is played by nucleophilic assistance by the azo-group is shown by the rearrangement of 2,2'-dicyclopropylazobenzene (X) on treatment with trifluoroacetic acid. It was found that, under these conditions only one of the cyclopropane rings reacted.

TABLE 1. 3-Ethyl-2-phenylindazoles (XIII) and (XVIII-XXI)

Starting material	Products	R¹	R²	R³	mp, °C	Found, %			Empirical formula	Calculated, %			Yield, %
						C	H	N		C	H	N	
I	XIII	H	H	H	40—41	80.8	6.5	12.4	$C_{15}H_{14}N_2$	81.0	6.4	12.6	91
VII	XVIII	Br	H	H	42—43	59.8	4.4	9.5	$C_{15}H_{13}BrN_2$	59.8	4.4	9.3	93
VIII	XIX	*t*-C_4H_9	H	H	160—162	82.1	8.2	10.2	$C_{19}H_{22}N_2$	82.0	7.9	10.1	88
IX	XX	H	Cl	H	99—100	70.2	5.2	11.0	$C_{15}H_{13}ClN_2$	70.2	5.1	10.9	74
X	XXI	H	H	Cyclopropyl	Oil	82.5	6.8	10.7	$C_{18}H_{16}N_2$	82.4	6.9	10.7	85

TABLE 2. Spectral Properties of Indazoles (XIII) and (XVIII-XXI)

Compound	PMR spectrum, ppm (CCl_4)			UV spectrum, λ_{max}, nm (ε)
	CH_3 (τ)	CH_2 (κ)	arom. protons	
XIII	1,16	2,93	6,7—7,75	292 (29 000), 314 (20 500)
XVIII	1,15	2,92	6,75—7,70	292 (31 000), 308 (24 000), 502 (168)
XIX*	1,21	2,96	6,90—7,65	294 (22 500), 495 (805)
XX	1,23	3,02	6,8—7,8	296 (27 300), 478 (105)
XXI†	1,12	2,82	6,6—7,7	336 (25 000), 450 (1200)

*1.43 ppm (9H, s, t-C_4H_9).
†0.5-0.9 (4H, m); 2.6-3.0 ppm (1H, m, small ring protons).

Bearing in mind the regioselectivity of the formation of indazoles in the reactions of 2-cyclopropylazobenzenes with trifluoroacetic acid, and the important part played by nucleophilic assistance by the azo-group in the rearrangement, it may be concluded that under these conditions no open ions of type A are formed, i.e., the opening of the small ring and the formation of the cyclic ion A' occur synchronously, the nitrogen atom of the azo-group attacking intramolecularly the protonated small ring, followed by the synchronous breaking of the $C_{(1)}$-$C_{(2)}$ bond of the cyclopropane ring and formation of the N—C bond.

The formation of the indazole system can only take place via cyclic ions of type A', possessing a sufficiently acidic hydrogen atom in the "benzyl" position. This explains the absence of indazoles among the products of the reaction of 2-(1-methylcyclopropyl)azobenzene (VI) with concentrated sulfuric acid. The cyclic ion A" formed in this instance is stabilized, either by attack of an external nucleophile on the developing carbenium center, or by removal of a proton from the β-position of the alkyl groups.

EXPERIMENTAL

IR specta were obtained on a UR-20 spectrometer (in Vaseline oil or in liquid films), UV spectra on a Cary-15 instrument, and PMR spectra on a Tesla BS-467 (60 MHz, in CCl_4, $CDCl_3$, or CF_3COOH), internal standard HMDS. The homogeneity of the compounds and the composition of the reaction mixtures were established by TLC on Silufol UV-254 plates in the system ether—pentane (1:2); LC was carried out on a Tsvet-105 apparatus. The reaction mixtures were separated by column chromatography on alumina grade (II) and on silica gel, eluents ether or a mixture of ether and pentane (1:2 or 1:1). The starting materials were synthesized by literature methods: 2-aminophenylcyclopropane as in [6], 1-methyl-1-(2-aminophenyl)cyclopropane as in [7], and 2-(1-hydroxypropyl)azobenzene, 2-cyclopropylazobenzene, and 2,2'-dicyclopropylazobenzene as in [1]. The yields and physicochemical constants of the products were in accordance with the literature values.

2-Amino-4-bromophenylcyclopropane was obtained by reducing 4-bromo-2-nitrophenylcyclopropane with iron in hydrochloric acid as described in [6], yield 63%, bp 152-154°C (11 mm), n_D^{20} 1.6131. IR spectrum: 3385, 3480 cm^{-1} (NH_2). PMR spectrum (CCl_4): 0.3-1.0 (4H, m, CH_2); 1.3-1.7 (1H, m, CH); 3.95 (2H, s, NH_2); 6.65-6.90 ppm (3H, m, Ar). Found, %: C 50.3, H 4.9, N 6.4. $C_9H_{10}BrN$. Calculated, %: C 51.0, H 4.8, N 6.6.

2-Amino-4-tert-butylphenylcyclopropane was obtained as described in [6], yield 85%, bp 141-143°C (8 mm), n_D^{20} 1.5496. IR spectrum: 3390, 3475 cm^{-1} (NH_2). PMR spectrum (CCl_4):

0.45-0.84 (4H, m, CH_2); 1.25 (9H, s, $C(CH_3)_3$]; 1.2-1.5 (1H, m, CH); 3.7 (2H, s, NH_2); 6.45-6.90 ppm (3H, m, Ar). Found, %: C 82.1, H 9.7, N 7.2. $C_{13}H_{19}N$. Calculated, %: C 82.5, H 10.1, N 7.4.

5-Bromo-2-cyclopropylazobenzene (VII). A mixture of 3 g (0.01 mole) of 2-amino-4-bromophenylcyclopropane and 1.07 g (0.01 mole) of nitrosobenzene in 30 ml of glacial acetic acid was stirred at 20°C for 6 h, poured into 200 ml of water, and extracted with chloroform. The extract was washed with water, dried over $MgSO_4$, the solvent removed, and the residue chromatographed on a silica column, eluent ether—pentane (1:2). Yield 37%: Dark-colored oil, IR spectrum: 1490, 1620 cm^{-1} (N=N); UV spectrum, λ_{max} (ε): 322 (49,000), 433 nm (2250). PMR spectrum (CCl_4): 0.75-1.38 (4H, m, CH_2); 2.88-3.10 (1H, m, CH); 6.72-8.31 ppm (8H, m, Ar). Found, %: C 60.0, H 4.4, N 9.5. $C_{15}H_{13}BrN_2$. Calculated, %: C 59.8, H 4.4, N 9.3.

5-tert-Butyl-2-cyclopropylazobenzene (VIII) was obtained as for (VII), yield 52%, oil. IR spectrum: 1480, 1535 cm^{-1} (N=N). UV spectrum, λ_{max} (ε): 322 (24,000), 444 nm (1350). PMR spectrum (CCl_4): 0.7-1.05 (4H, m, CH_2); 1.18 [9H, s, $C(CH_3)_3$]; 2.7-3.05 (1H, m, CH); 6.6-8.1 ppm (8H, m, Ar). Found, %: C 82.1, H 8.0, N 10.1. $C_{19}H_{22}N_2$. Calculated, %: C 82.0, H 8.0, N 10.1.

2-Cyclopropyl-4'-chlorobenzene (IX) was obtained as for (VII), from 2-aminophenylcyclopropane and 4-chloronitrosobenzene, yield 24%, mp 90-91°C (from alcohol). IR spectrum: 1495-1580 cm^{-1} (N=N). PMR spectrum (CCl_4): 0.7-0.95 (4H, m, CH_2); 2.8-3.10 (1H, m, CH); 6.7-8.25 ppm (3H, m, Ar). Found, %: C 70.2, H 5.2, N 11.1. $C_{15}H_{13}ClN_2$. Calculated, %: C 70.2, H 5.1, N 10.9.

Reaction of Azobenzenes (I) and (VI) with Concentrated Sulfuric Acid. To 30 ml of concentrated sulfuric acid, cooled to −10°C, was added in small portions with vigorous stirring 0.01 mole of the appropriate azobenzene, the temperature being kept at its original level. The mixture was stirred at −10°C for 1 h 30 min, poured into 100 ml of crushed ice, and basified with cooling with sodium hydroxide until just alkaline. It was then extracted with ether, the ether extracts combined, washed with water, and dried over $MgSO_4$. The solvent was removed, and the residue chromatographed on silica, eluent ether—pentane (1:3).

From 4.5 g of 2-cyclopropylazobenzene (I) there were obtained: 1.3 g (30%) of 2-methyl-1-anilinoindole (III), mp 123-124°C (from alcohol); 0.4 g (9%) of 2-(1-hydroxypropyl)azobenzene (IV), mp 74-76°C, and 0.3 g (7%) of 2-(2-hydroxypropyl)azobenzene (IV), an oil. These products were identified by comparison with the data given in [1]. In addition, there were also isolated from the reaction mixture 0.5 g (11%) of 2-phenyl-3-ethylindazole (XIII) (for physicochemical properties, see Table 2) and 1 g (22%) of 5,7-dihydro-11-methyldibenzo[c, g][1, 2]-diazocine (XII) [oil; IR spectrum: 3400 cm^{-1} (NH); UV spectrum, λ_{max} (ε): 290 (23,000); 325 (17,000); 441 nm (700; PMR spectrum ($CDCl_3$): 2.05 (3H, s, CH_3); 3.58 (2H, s, NH_2); 6.9-7.8 ppm (9H, m, C=CH); 0.36 g (8%) of 11-ethyl-11H-dibenzo[c, f][1, 2]-diazepine [oil. IR spectrum: 1460, 1535 cm^{-1} (N=N). PMR spectrum (CCl_4): 1.5 (3H, t, CH_3); 3.55 (2H, q, CH_2); 6.6-7.75 ppm (9H, m, Ar)].

From 4.1 g of 2-(1-methylcyclopropyl)azobenzene (VI) there were obtained: 2.0 g (49%) of 2-(1-methylpropenyl)azobenzene (XIV) [oil; IR spectrum: 1450, 1490, 1520 cm^{-1} (N=N); UV spectrum, λ_{max} (ε): 319 (52,500), 446 nm (1130); PMR spectrum (CCl_4): 1.15-2.0 (6H, m, CH_3); 5.41 (1H, m, C=CH); 7.2-7.95 ppm (9H, m, Ar). Found, %: C 81.4, H 6.2, N 12.0. $C_{16}H_{16}N_2$. Calculated, %: C 81.3, H 6.8, N 11.9]; 0.6 g (15%) of 5,6-dihydro-11,12-dimethyldibenzo[c, g][1, 2]diazocine (XV) [oil, IR spectrum: 3380 cm^{-1} (N—H). PMR spectrum (CCl_4): 1.41 (6H, s, CH_3); 4.74 (2H, s, NH_2); 6.4-7.2 ppm (8H, m, Ar). Found, %: C 81.5, H 6.2, N 11.9. $C_{16}H_{16}N_2$. Calculated, %: C 81.3, H 6.8, N 11.9]; 0.4 g (10%) of 1-anilino-2,3-dimethylindole (XVI) [mp 111-113°C. IR spectrum: 3335 cm^{-1} (NH). PMR spectrum ($CDCl_3$): 2.08 (3H, s, CH_3); 2.2 (3H, s, CH_3); 5.7 (1H, s, NH); 6.2-7.7 ppm (9H, m, Ar). Found, %: C 80.8, H 7.0, N 11.9. $C_{16}H_{16}N_2$. Calculated, %: C 81.3, H 6.8, N 11.9.

Cyclization of 2-Cyclopropylazobenzenes (I) and (VII-X) to 2-Phenyl-3-ethylindazoles (XIII) and (XVIII-XXI). The azobenzene (0.01 mole) was dissolved in 30 ml of trifluoroacetic acid, heated to 60-70°C, and kept at this temperature for 2-3 h. The mixture was cooled, poured into 150 ml of water, neutralized with sodium hydroxide solution, and extracted with chloroform (3 × 50 ml). The chloroform extracts were washed with water, dried over $MgSO_4$, the solvent removed, and the residue chromatographed on a column of alumina, eluent ether. The yields and physicochemical properties of (XIII) and (XVIII-XXI) are given in Tables 1 and 2.

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SYNTHESES OF 2,2'-DIIMIDAZOLE

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UDC 547.223'281'785.1.07

New methods of preparation of 2,2'-diimidazole have been developed, by the reaction of ammonia with 1,1-dibromoacetaldehyde (20% yield) or with glyoxal sulfate (40%), and by the cyclization of 1,2-dihydroxyethylenediamine dihydrochloride in the presence of sodium acetate (yield 60%).

2,2'-Diimidazole (I) was synthesized long ago, but there are no satisfactory methods for its preparation. Its chemistry has received little attention, although its derivatives include compounds with useful biological properties [1, 2]. One method of preparation is from 2,2,2-trichlorolactic acid and aqueous ammonia [3], and another is based on the reaction of glyoxal with ammonia [4] or ammonium salts [5]. Common disadvantages of these methods are the low yields obtained (no greater than 25%) and the formation of highly contaminated (I), which can be purified only by sublimation. A method of synthesis developed recently [6] involving the aromatization of 2,2'-di-(2-imidazoline), although it gives high yields of (I), is complicated even for preparative use.

Despite the apparent diversity of the first three methods [3-5], they are in our view united in the ability of the starting materials to produce during the reaction intermediates which are structurally analogous and have similar properties, namely alkylamines containing a halogen or hydroxy group in the α-position. We therefore addressed ourselves to reactions in which such compounds are possible intermediates. Such reactions include the condensation of ammonia with 1,1-dibromoacetaldehyde and glyoxal sulfate. It was in fact found that 1,1-dibromoacetaldehyde reacts with aqueous ammonia on prolonged heating at a temperature not exceeding 60°C to give (I). However, this reaction is attended the same deficiencies as are inherent in the reaction of glyoxal with ammonia, namely similar yields of crude diimidazole, and the difficulty of its purification. When the reaction was carried out at temperatures above 60°C, a mixture of resinous materials was obtained which contained no (I). The best yields (up to 40%) were obtained by reacting glyoxal sulfate with aqueous ammonia at 60-80°C.

(See scheme on following page.)

In these syntheses of (I), intermediates of similar chemical structure and properties are undoubtedly present, one of which could be 1,2-dihydroxyethylenediamine (II). The latter compound is quite stable as its hydrochloride [7], but has not been obtained in the free state. It appears that in neutral or basic media, as with the simplest geminal aminoalcohols [8], (II) decomposes to glyoxal and ammonia, which may give (I), as described in [4]. In

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