Pyrolysis of *N*-[β-(*N''*-Phenylcarbamyl)ethyl]-*N*,*N'*-diphenylurea. Synthesis and Properties of the Decomposition Product, 2-Phenylimino-3-phenyloxazolidine and Its Analogs

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The pyrolysis of *N*-[β-(*N''*-phenylcarbamyl)ethyl]-*N*,*N'*diphenylurea proceeds by two simultaneous pathways. The major reaction is cyclization to 2-phenylimino-3-phenyloxazolidine with loss of aniline and carbon dioxide. The minor reaction is a dissociation into β-anilinoethyl *N*-phenylcarbamate and phenyl isocyanate. Further reactions of these intermediates lead to a complex mixture of other products. A simple and rapid method of synthesis of N,N'-disubstituted 2-iminooxazolidines consists in the acid cyclodehydration of *N*-(β-hydroxyethyl)-*N*,*N'*-diaryl- or -alkylarylureas. The 2-arylimino-3-aryloxazolidines are isomerized by heating with hydrobromic acid to 1,3-diaryl-2-imidazolidones. Other properties of the iminooxazolidines, including acid hydrolysis, reaction with aromatic amines, thermal isomerization, and polymerization, are reported.

Pyrolysis of *N*-[β-(*N''*-Phenylcarbamyl)ethyl]-*N*,*N'*-diphenylurea (1).—In line with our previous work on the thermal degradation of ethylene bis(*N*-phenylcarbamate),[1] it was expected that the pyrolysis of the urea carbamate **1** at 200–240° would give primarily carbon dioxide, aniline, and 1,3-diphenyl-2-imidazolidone by a one-step decomposition. However, a complex mixture of 11 products was found: carbon dioxide, aniline, 2-phenylimino-3-phenyloxazolidine (**2a**), 1,3-diphenyl-2-imidazolidone (**3a**), β-anilinoethyl *N*-phenylcarbamate (**4**), *N*,*N'*-diphenylpiperazine (**5**), *N*,*N'*-diphenylethylenediamine (**6**), *N*,*N'*-diphenylurea (**7**), 2-anilinoethanol (**8**), ethylene oxide (**9**), and 3-phenyl-2-oxazolidone (**10**) (Table I). All the products were completely identified by appropriate chemical reactions or by their melting points, ir spectra, nmr spectra, and comparison with authentic samples.

TABLE I
PYROLYSIS OF COMPOUND 1

Compd	mmol Run 1, 1 hr, 200°	Run 2, 2 hr, 200°	Run 3, 2.5 hr, 210°	Run 4, 1.5 hr, 230°	Run 5, 2 hr, 240°
Compound **1** submitted to pyrolysis	30.0	40.0	20.0	30.0	40.0
CO_2	22.3	27.7	15.4	25.0	33.7
Aniline	6.7	9.2	5.0	9.0	10.0
Compound **2a**	16.3	24.9	14.0	19.3	15.3
1,3-Diphenyl-2-imidazolidone (**3a**)	0.7	2.5	2.2	4.3	12.0
Carbamate **4**	4.1	2.8	0.3	0.0	0.0
N,*N'*-Diphenylpiperazine (**5**)	0.19	0.26	*a*	0.0	0.0
N,*N'*-Diphenylethylenediamine (**6**)	2.3	1.6	1.4	3.1	2.5
N,*N'*-Diphenylurea (**7**)	13.0	17.0	7.7	8.7	14.3
2-Anilinoethanol (**8**)	*a*	0.9	*a*	*a*	0.15
Ethylene oxide (**9**)[b]	*a*	*a*	*a*	*a*	*a*
3-Phenyl-2-oxazolidone (**10**)	0.6	1.7	1.0	1.0	2.5
Weight % recovered	89.5	92.1	95.4	91.2	92.8

[a] Isolated or detected but not determined quantitatively. [b] Detected by its reaction with KSCN, giving a characteristic pink color: W. Deckert, *Angew. Chem.*, **45**, 758 (1932).

The data in Table I indicate that the initial degradation of **1** occurs by two simultaneous pathways. The major reaction is cyclization to **2a** with loss of aniline and carbon dioxide. Its extent was calculated to be about 68 and 78% at 200 and 240°, respectively, from the data in expt 2 and 5. The minor pathway is a dissociation to the anilinoethyl carbamate **4** and phenyl isocyanate. Compounds **5**–**10** arise from further decomposition of **4** as shown by separate pyrolysis experiments (Table II). The main reactions are shown in Scheme I.

TABLE II
PYROLYSIS OF COMPOUND 4

Compd	mmol Run 1, 2 hr, 200°	Run 2, 2 hr, 240°	Run 3,[a] 2 hr, 200°
Compound **4** submitted to pyrolysis	10.0	10.0	10.0
CO_2	4.5	7.3	2.0
Aniline	3.5	7.0	*a*
1,3-Diphenyl-2-imidazolidone (**3a**)	0.3	0.4	0.17
Compound **4** (unreacted)	0.7	0.1	0.0
N,*N'*-Diphenylpiperazine (**5**)	1.5	3.5	0.17
N,*N'*-Diphenylethylenediamine (**6**)	1.3	1.2	1.9
N,*N'*-Diphenylurea (**7**)	2.0	0.8	5.2
2-Anilinoethanol (**8**)	0.7	0.4	3.9
Ethylene oxide (**9**)	*b*	*b*	*c*
3-Phenyl-2-oxazolidone (**10**)	2.8	0.7	1.9
Weight % recovered	93.6	91.3	

[a] 21.5 mmol of aniline added. [b] Detected qualitatively by its reaction with KSCN: W. Deckert, *Angew. Chem.*, **45**, 758 (1932). [c] Amount of ethylene oxide increasing very clearly as proved by a very deep pink color developed in the KSCN solution.

The parallel increase of imidazolidone **3a** and decrease of **2a** at progressively higher temperature indicate that isomerization of **2a** is the major source of **3a** in the decomposition of **1**. This was confirmed by the quantitative conversion of **2a** into **3a** at 200–240°. The small quantity of **3a** found in the pyrolysis of **4** may be due to the reaction of *N*,*N'*-diphenylethylenediamine (**6**) with *N*,*N'*-diphenylurea (**7**).[1]

N,*N'*-Diphenylpiperazine (**5**) probably arises from the intermolecular condensation of 2 mol of **4**. This reaction is suppressed to a large extent by dilution of **4** with aniline as shown by expt 3 (Table II). This

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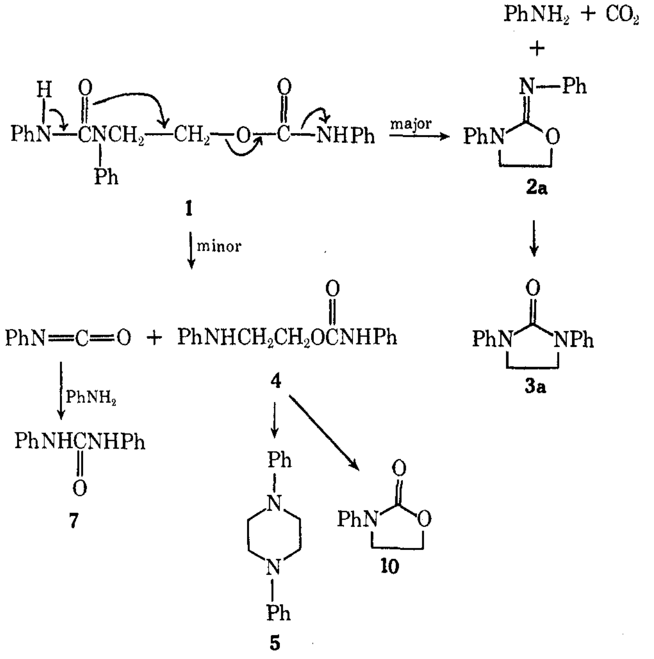
[1] H. C. Beachell and C. P. Ngoc Son, *J. Polym. Sci., Part A*, **2**, 4773 (1964).

SCHEME I



explains the very small amount of **5** found in the decomposition of **1**.

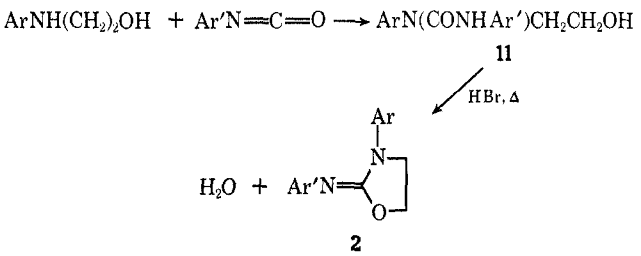
The diamine **6**, the urea **7**, 2-anilinoethanol (**8**), and ethylene oxide (**9**) are likely formed by the attack of aniline on **4** at the methylene adjacent to the oxygen[2,3] or at the carbonyl of the urethane function. This is supported by expt 3 (Table II), showing a net increase of these products in the pyrolysis of **4** in the presence of aniline. However, comparison of the data in Tables I and II shows that in the pyrolysis of **1**, the major portion of **7** must be formed from the reaction of aniline with phenyl isocyanate liberated during the partial dissociation of **1**. Compounds **6** and **7** may also be obtained from the reaction of aniline with **2a**; in fact, **2a** treated with an excess of aniline gave **6** and **7** in good yields.

Finally 3-phenyl-2-oxazolidone (**10**) would arise by the intramolecular cyclization of **4**.

Synthesis and Properties of *N*,*N'*-Disubstituted 2-Iminooxazolidines 2. A. Synthesis.—Although compound **2a** is one of the main decomposition products, it is not readily isolated from the reaction mixture. Various other time-consuming methods of synthesis of 2-arylimino-3-aryloxazolidines have been described such as the reaction of bis(β-anilinoethyl)-phenylphosphonite or ethyl bis(β-anilinoethyl)phosphite with 2 equiv of phenyl isocyanate,[4,5] and the cyclization of β-chloroethyl-*N*,*N'*-diarylureas in boiling water.[6]

We found a very rapid and simple method of synthesis of compounds **2** which consists of the ring closure in acid medium of *N*-(β-hydroxyethyl)-*N*,*N'*-diarylureas **11**. The latter were quantitatively obtained by slow addition at room temperature of a dilute solution of aryl isocyanate into a dilute solution of 1 equiv of *N*-arylaminoethanol in dry benzene or dichloromethane as solvent (Scheme II).

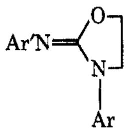
SCHEME II



The cyclization must be carried out quickly to avoid further isomerization of **2** into 1,3-diaryl-2-imidazolidones **3**. In general, compounds **11** were heated in 48% HBr until dissolution, then reflux was continued for about 10 min. In most of the cases, the iminooxazolidines **2** were easily obtained with fairly good yields (65–75%) (Table III).

TABLE III[a]

2,



(prepared from **11**)

Compd	Ar	Ar'	Mp, °C	Yield, %
2a	C_6H_5	C_6H_5	115–116[b]	75
2b	C_6H_5	*p*-$CH_3C_6H_4$	95–96	70
2c	C_6H_5	*m*-$CH_3C_6H_4$	114–115	78
2d	C_6H_5	*p*-ClC_6H_4	143–144	61
2e	C_6H_5	*p*-$CH_3OC_6H_4$	91–92	72
2f	C_6H_5	*o*-$CH_3C_6H_4$	90–91	65
2g	C_6H_5	*p*-$NO_2C_6H_4$	164–166	27
2h	C_6H_5	*o*-$C_2H_5OC_6H_4$	105–106	40
2i	*p*-ClC_6H_4	*p*-ClC_6H_4	135–136	42

[a] Satisfactory analytical values (±0.40 for C, H, N) were reported for compounds listed in this table except for **2a** and **2c** (C found: 0.43 and 0.48 high) and **2b** (N found: 0.48 low): Ed.
[b] Lit.[6] 115–116°.

Compounds **2** show characteristic infrared absorption around 1670 (C=N) and 1410 cm^{-1} ($-N{=}\overset{|}{C}ON<$).[5] Their nmr spectra present two heptets centered around 4.2 ppm (A_2B_2 system).

B. Properties of 2. 1. Acid Hydrolysis and Isomerization.—In boiling 1.2 *N* aqueous HCl, all 2-arylimino-3-phenyloxazolidines are hydrolyzed to afford 3-phenyl-2-oxazolidone (**10**, yield 30–50%).

In boiling 48% HBr, hydrolysis does not occur; instead isomerization affords 1,3-diaryl-2-imidazolidones **3**, characterized by their ir spectra (C=O around 1690 cm^{-1}) and nmr spectra showing one singlet around 3.95 ppm (4 methylenic H). For example with **2a**, **2c**, **2g**, and **2i**, the corresponding imidazolidones were isolated with yields of 71, 75, 20, and 84%, respectively. The isomerization failed, however, with **2** having an ortho substituent on the ring of the aryl imino group.

2. Nucleophilic Reaction of Aromatic Amines.—Primary aromatic amines react with **2** to give N,N'-disubstituted ureas and diamines.

For instance, a fourfold excess of aniline heated at reflux with **2a** yielded *N*,*N'*-diphenylethylenediamine

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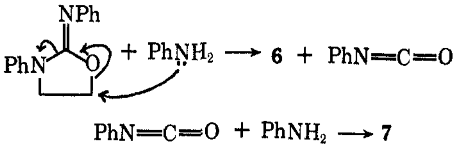
[2] E. Dyer and G. C. Wright, *J. Amer. Chem. Soc.*, **81**, 2138 (1959).
[3] C. P. Ngoc Son and T. T. Kim Lan, *Ann. Fac. Sci. Saigon*, 53 (1963–1964); Abstracts, 152nd National Meeting of the American Chemical Society, New York, N. Y., Sept 1966, No. V96.
[4] O. Mitsunobu, T. Ohashi, M. Kikuchi, and T. Mukaiyama, *Bull. Chem. Soc. Jap.*, **39**, 214 (1966).
[5] O. Mitsunobu, T. Ohashi, and T. Mukaiyama, *ibid.*, **39**, 708 (1966).
[6] H. Nohira, Y. Nishikawa, and T. Mukaiyama, *ibid.*, **37**, 797 (1964).

(6, 75%) and N,N'-diphenylurea (7, 80%). By analogy with the mode of attack of aromatic amines on 10[1,3] leading to its fragmentation into carbon dioxide and the corresponding diamine, the ring opening of **2a** in the presence of aniline likely occurs at the methylene adjacent to the oxygen with subsequent formation of **6** and phenyl isocyanate. The latter reacts immediately with aniline in excess to yield **7** (Scheme III).

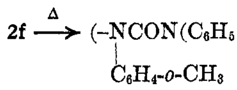
SCHEME III



$$PhN{=}C{=}O + PhNH_2 \longrightarrow \mathbf{7}$$

Evidence for the proposed mechanism was the detection of phenyl isocyanate when **2a** was treated with aniline at 190–210° in a 2-ml flask having a curved side arm. After a few minutes of heating, the isocyanate partly liberated from the reaction medium reacted with aniline to yield **7** on the upper part of the flask and in the side arm.

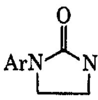
3. Thermal Isomerization of 2.—At high temperature (220–240°), under a nitrogen atmosphere, compounds **2** isomerize quantitatively to **3** (Table IV) except **2f** and **2h**, which have an ortho substituent on the ring of the arylimino group. In the latter case,



$$\mathbf{2f} \xrightarrow{\Delta} (\text{-}\underset{\underset{C_6H_4\text{-}o\text{-}CH_3}{|}}{N}CON(C_6H_5)CH_2CH_2\text{-})_n$$

polymerization occurs to give poly(ethylene-N,N'-diarylureas) characterized by elemental analyses and infrared spectra which display urea absorption around 1660 cm^{-1}. It appears that steric crowding of the ortho substituents plays an important role in the determination of the reaction pathways. Further investigations will help us to understand the mechanism of these reactions.

TABLE IV[a]

3,  (prepared from **2**)

Compd	Ar	Ar′	Mp, °C	Yield, %
3a	C_6H_5	C_6H_5	210–212[b]	80
3b	C_6H_5	p-$CH_3C_6H_4$	188–189[c]	70
3c	C_6H_5	m-$CH_3C_6H_4$	136–137	73
3d	C_6H_5	p-ClC_6H_4	202–203[d]	82
3e	C_6H_5	p-$CH_3OC_6H_4$	211–212[e]	92
3f	C_6H_5	p-$NO_2C_6H_4$	208–209	80
3i	p-ClC_6H_4	p-ClC_6H_4	206–207	76

[a] Satisfactory analytical values (±0.40 for C, H, N) were reported for compounds listed in this table: Ed. [b] Lit.[1] 210–212°. [c] Lit.[6] 191–193°. [d] Lit.[6] 200–202°. [e] Lit.[6] 212–213°.

Experimental Section

Synthesis. A. Products of Degradation.—Aniline, N,N'-diphenylethylenediamine, and 2-anilinoethanol were commercially available from Eastman.

1,3-Diphenyl-2-imidazolidone (**3a**), mp and mmp 210–212°, ir (KBr) 1690 cm^{-1} (C=O), nmr ($CDCl_3$) 3.95 ppm (s, 4 methylenic H), N,N'-diphenylurea (**7**), mmp 242–243°, N,N'-diphenylpiperazine (**5**), mp 164–165° (lit.[7] 163–164°), nmr ($CDCl_3$) 3.5 ppm (s, 8 methylenic H), and 3-phenyl-2-oxazolidone (**10**), mp 119–120° (lit.[1] 120–121°), ir (KBr) 1750 cm^{-1} (C=O), were synthesized from known procedures.[1,7]

B. N-[β-(N''-Phenylcarbamyl)ethyl]-N,N'-diphenylurea (1).—Phenyl isocyanate (71.4 g, 0.6 mol) was added dropwise at reflux and under a nitrogen atmosphere to a solution of 2-anilinoethanol (41.1 g, 0.3 mol) in 150 ml of dry benzene. The solution was heated for 7 hr, then solvent was partially removed. Recrystallization from benzene–hexane afforded colorless needles: mp 137°; ir (KBr) 3300 (NH), 1706 (-NHCOO-), 1634 cm^{-1} (-NHCON<); yield 90%.

Anal. Calcd for $C_{22}H_{21}N_3O_3$: C, 70.40; H, 5.60; N, 11.20. Found: C, 70.35; H, 5.63; N, 11.22.

C. β-Anilinoethyl N-Phenylcarbamate (4).—β-Bromoethyl N-phenylcarbamate, mp 70° (24.4 g, 0.1 mol) (obtained by the reaction of 2-bromoethanol with an equimolar quantity of phenyl isocyanate in benzene solution), was treated with aniline (11.2 g, 0.12 mol) at reflux in dry benzene in the presence of pyridine for 5 hr. Pyridinium bromide was then removed and the solution was extracted with 1.2 N HCl. Neutralization of the aqueous layer with solid KOH at 0° yielded the carbamate **4** which was recrystallized from chloroform–hexane, mp 81–82°, yield 51%.

Anal. Calcd for $C_{15}H_{16}N_2O_2$: C, 70.31; H, 6.25; N, 10.92. Found: C, 70.18; H, 6.25; N, 10.76.

D. 2-Arylimino-3-aryloxazolidines 2.—All the compounds **2** were already recorded in Table III. A typical synthesis is described below.

E. 2-Phenylimino-3-phenyloxazolidine (2a).—(β-Hydroxyethyl)-N,N'-diphenylurea, mp 83–84° (lit.[1] 82–83°), was first synthesized by a very slow dropwise addition of 1 equiv of phenyl isocyanate to 1 equiv of 2-anilinoethanol in very dilute benzene solution at room temperature and under a nitrogen atmosphere. A mixture of 5 g of the urea and 15 ml of 48% HBr was heated until complete dissolution occurred. Heating was continued for about 10 min, then the solution was diluted with distilled water and filtered. Neutralization of the filtrate with solid KOH at 0° provided **2a**. Recrystallization from chloroform–hexane afforded colorless crystals: mp 115–116° (lit.[5] 116°); ir (KBr) 1670 cm^{-1} (C=N); nmr ($CDCl_3$) two symmetrical multiplets entered at 4.1 ppm; yield 75%.

Anal. Calcd for $C_{15}H_{14}N_2O$: C, 75.63; H, 5.88; N, 11.76. Found: C, 75.20; H, 5.93; N, 11.85.

Reactions. A. Acid Hydrolysis of 2.—A typical reaction is described below.

A 1-g sample of **2a** was heated with 10 ml of 1.2 N HCl at reflux for 3 hr and then cooled in the refrigerator. 3-Phenyl-2-oxazolidone (**10**) precipitated and was recrystallized from chloroform–hexane, mp 119–120°, yield 44%.

B. Acid Isomerization of 2.—The isomerization was performed with 48% HBr. With **2a**, 1,3-diphenyl-2-imidazolidone (**3a**) precipitated during the reaction. Recrystallization from chloroform–hexane afforded yellowish crystals, mp and mmp 210–212°, yield 71%.

C. Reaction of Aniline with 2a.—A 1-g sample of **2a** was treated with 3 ml of aniline for 1 hr at 200° and under a nitrogen atmosphere. The residue was extracted with cold ether. The insoluble fraction was impure N,N'-diphenylurea (80%) which was then recrystallized from chloroform–hexane, mp and mmp 242°. The filtrate was treated with 2 N HCl. Neutralization of the aqueous layer at 0° with solid KOH gave the diamine **6**, mp 57–58°, yield 75%. Recrystallization from ether–hexane yielded pale yellow crystals, mp and mmp 62°.

D. Thermal Isomerization of 2.—General procedure: A 1-g sample of **2** was heated at 220–240° for 2 hr under a nitrogen atmosphere. The residue was washed with cold ether to eliminate the unreacted iminooxazolidine and recrystallized from chloroform–hexane to yield **3**. All compounds **3** were recorded in Table IV.

From 2-o-tolylimino-3-phenyloxazolidine (**2f**), poly(ethylene-N-o-tolyl-N'-phenylurea) was obtained in 30% yield.

Anal. Calcd for $(C_{16}H_{16}N_2O)_n$: C, 76.03; H, 6.50; N, 11.01. Found: C, 76.18; H, 6.34; N, 11.11.

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[7] H. W. Heine, B. L. Kapur, and C. S. Mitch, *J. Amer. Chem. Soc.*, **76**, 1173 (1954).

E. Pyrolysis of Compounds 1 and 4.—The pyrolysis of **1** and **4** was carried out in a 100-ml three-necked flask equipped with thermometer, nitrogen inlet, and condenser. The latter was connected to a $CaCl_2$ U-tube, a three-way stopcock carrying at each end an ascarite U-tube. The weighing of the latter at regular intervals of time permitted us to measure carbon dioxide liberated during the pyrolysis. The residue after the pyrolysis was partially dissolved in cold ether. The insoluble portion was composed of the compounds **2a**, **3a**, and **7**. The filtrate contained aniline, and the products, **2a**, **4**, **5**, **6**, **8**, **10**, small amounts of **3a** and **7** slightly soluble in cold ether. The degradation products were separated quantitatively by column chromatography on alumina of these two portions with hexane, benzene, chloroform, ethanol, and their mixtures as eluents.

Registry No.—**1**, 748-84-5; **2b**, 32974-53-1; **2c**, 32974-54-2; **2d**, 32974-55-3; **2e**, 32974-56-4; **2f**, 13468-08-1; **2g**, 32974-58-6; **2h**, 32974-59-7; **2i**, 32974-60-0; **3c**, 32974-61-1; **3f**, 32974-62-2; **3i**, 5198-55-0; **4**, 33020-71-2; **6**, 150-61-8; **7**, 102-07-8; β-bromoethyl *N*-phenylcarbamate, 32353-12-1; poly(ethylene-*N*-*o*-tolyl-*N*′-phenylurea), 33029-39-9.

The Transannular Neophyl Rearrangement[1,2]

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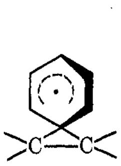
The radical rearrangement in solution of a phenyl group across a cyclohexane ring *via* bicyclo[2.2.1]heptyl and [2.2.2]octyl species has been achieved. This transannular rearrangement did not occur in analogs *via* smaller sized bicyclic species. In these cases the parent structures were retained or ring opening occurred.

The vicinal migration of a phenyl group *via* **1** (the neophyl rearrangement) is well known.[3] Less common, but still recorded,[4] are analogous further rearrangements *via* **2**. It was the intent of the present study to seek phenyl shifts *via* **3**, a process we term the "transannular neophyl rearrangement."[5] Quite clearly, such rearrangements will be sensitive to the bicyclic ring size of the intermediate; so a series of these were investigated.



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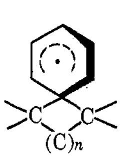

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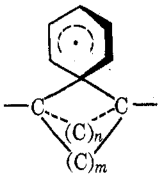
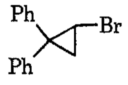


3

Any of several extant free radical processes that exhibited rearrangement *via* **1** or **2** could be chosen for application to **3**. However, in recent years the generation of carbon radicals from the reaction of halides with organotin hydrides has become widespread.[6] This method recommended itself for the present purpose for several reasons: the temperature of the reaction can be varied readily, as can the concentration of the tin hydride. Generally speaking, higher temperatures and lower concentrations of the hydride favor radical rearrangement processes.[7] Also, as one of its best features, the process produces the radical of interest directly and not *via* intervening species.

As precursors to the radicals under study, the series of bromides **4–8** was prepared and characterized. The rationale for using a *gem*-diphenyl function was twofold. First, one of the phenyls is always situated appropriately for migration. Second, the other phenyl group serves as a stabilizer for the radical center formed subsequent to rearrangement. Coincidentally, this second phenyl group serves also as a rearrangement marker through its influence on the nmr spectrum of the product. Secondary bromides sometimes present a problem in their synthesis because carbonium ion rearrangements can occasionally plague the customary routes to them.[8] For this reason, a noncarbonium ion process (eq 1) was chosen to prepare most of those needed for this study.



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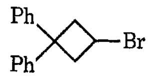

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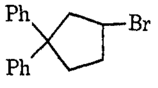

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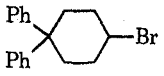
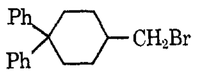


7



8

$$\underset{11,\ 17,\ 19}{\mathrm{ROTs}} + \mathrm{LiBr} \xrightarrow[\text{or diglyme}]{\text{acetone}} \underset{5\text{–}7}{\mathrm{RBr}} + \mathrm{LiOTs} \tag{1}$$

The requisite tosylates were made as shown (eq 2–4). The sequences shown are straightforward and will not be discussed. Details may be found in the Experimental Section. Diagnosis by spectral and chromatographic methods indicated that reaction 1 proceeded without rearrangement in every instance. Bromides **4** and **8** were, however, prepared alternatively (eq 5, 6). In all cases the structures were supported by combustion analytical and spectral data.

One may notice that the transannular neophyl re-

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[1] Taken from (a) the Dissertation of R. A. D., 1970; and (b) the M.S. Thesis of K. C. W., 1970.

[2] Presented at the Third Great Lakes Meeting of the American Chemical Society, Northern Illinois University, De Kalb, Ill., June 1969, Abstracts of Papers, paper 58.

[3] *Cf.* R. Kh. Freidlina in "Advances in Free-Radical Chemistry," Vol. 1, G. H. Williams, Ed., Academic Press, New York, N. Y., 1965, pp 249–260.

[4] S. Winstein, R. Heck, and S. Lapporte, *Experientia*, **12**, 138 (1956).

[5] Although perhaps properly applied only to the rearrangement of the β-phenylisobutyl ("neophyl") radical itself, the term "neophyl rearrangement" is used here for the radical migration of an aromatic group from a carbon atom origin to any other carbon atom terminus.

[6] H. G. Kuivila, *Accounts Chem. Res.*, **1**, 229 (1968).

[7] L. Kaplan, *J. Amer. Chem. Soc.*, **88**, 4531 (1966).

[8] *Cf.* J. Cason and J. S. Correia, *J. Org. Chem.*, **26**, 3645 (1961).