puted for these 290 reflections clearly showed the positions of 22 atoms. Structure factors based upon these positions were then calculated and the resulting phases used with the observed structure amplitudes to yield an electron density map which revealed the 10 remaining nonhydrogen atoms. At this point a conventional R of 26.6% was obtained. The structure was refined by full matrix least-squares techniques using a weighting scheme of the type suggested by Hughes.[10] All atoms were assigned anisotropic temperature factors. A final value for R of 12.9 was obtained using all 2067 observed reflections.

A copy of the final atomic parameters together with the observed and calculated structure factors can be obtained on request from R. T. P. Calculations were performed using the X-Ray 67 System of programs developed by Dr. James Stewart of the University of Maryland, together with the programs FAME and MAGIC by Fleisher, Dewar, and Stone, for the application of the symbolic addition procedure.

Registry No.—**2,** 532-55-8; **6,** 14205-42-6; **8a,** 30378-32-6; **8b,** 30428-03-6; **9,** 30428-04-7; **10,** 30428-05-8; **11,** 30428-06-9; **12,** 30428-07-0; **13,** 30428-08-1; **14,** 30428-09-2; **15,** 30428-10-5; **16,** 30428-11-6; **17,** 30428-12-7.

Acknowledgment.—The authors wish to express their appreciation to Mr. Louis Dorfman and the members of his staff for microanalytical and spectral determinations, and Professor Peter Yates, University of Toronto, for helpful discussions.

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[10] E. W. Hughes, *J. Amer. Chem. Soc.*, **63**, 1737 (1941).

Synthesis of a *dl* Polymer and an Active (+) Polymer Containing the 2,4,5,7-Tetranitrofluorenylideneaminooxysuccinic Moiety. Chromatographic Studies[1]

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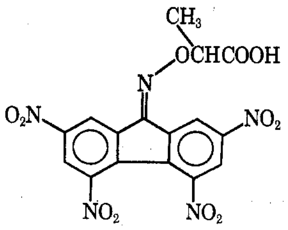
dl-Di-β-hydroxyethyl 2,4,5,7-tetranitrofluorenylideneaminooxysuccinate (6) was synthesized by the steps shown in Scheme I. Treatment with terephthalyl chloride yielded a solid polymer which was comparable to Woelm neutral alumina (grade I) in the separation of 6- and 10-methylbenz[*a*]anthracene by column chromatography. An optically active polymer was similarly prepared from (+)-6 but was ineffective in separating 2-butyl 1-naphthyl ether and hexahelicene into active forms in the range used, 3 g of polymer to 0.1 g of substrate.

α-(2,4,5,7-Tetranitro-9-fluorenylideneaminooxy)propionic acid (TAPA, **1**) was synthesized to provide a reagent suitable for resolution by complex formation of compounds lacking functional groups which would allow for resolution by standard method.[3] The resolution of hexahelicene (**2**) by TAPA was successful,[4] but very small amounts of optically pure **2** were obtained after considerable effort.

The object of the present study was to prepare a polymer containing the 2,4,5,7-tetranitrofluorene nucleus which might prove superior to alumina for the chromatographic separation of polynuclear hydrocarbons and to prepare an optically active polymer which might prove useful as a solid phase for column chromatographic resolution of **2** or other compounds[3] which have been resolved by the use of TAPA.[5]



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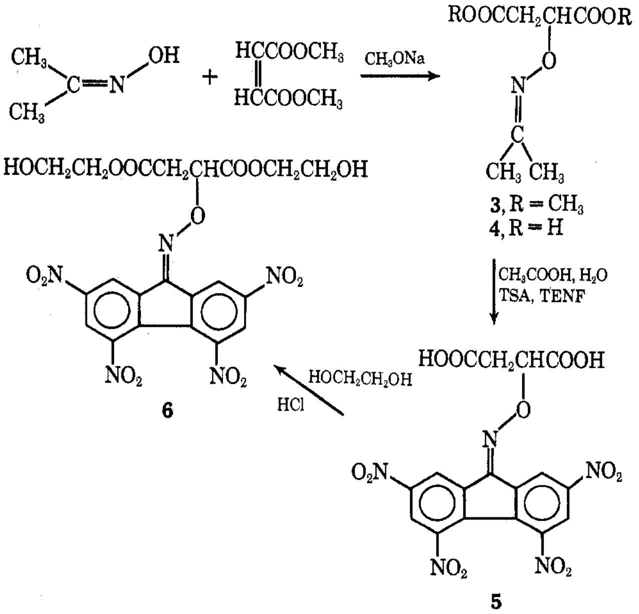
An optically inactive diol suitable for formation of a polyester polymer was prepared as shown in Scheme I.

SCHEME I



The base-catalyzed reaction of acetone oxime with dimethyl maleate produced dimethyl isopropylideneaminoxysuccinate (**3**) in 50% yield. Acid-catalyzed (toluenesulfonic acid, TSA) ketone exchange with 2,4,5,7-tetranitrofluorenone (TENF) in aqueous acetic acid proceeded in high yield to 2,4,5,7-tetranitrofluorenylideneaminooxysuccinic acid (**5**) which was esterified to the bis-2-hydroxyethyl ester **6**, the glycol

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[1] This work was supported in part by Grant No. CA 07394 from the U. S. Public Health Service and by the award of an Ohio State University Postdoctoral Research Fellowship to H. Junjappa, 1967–1968.

[2] Author to whom correspondence should be addressed.

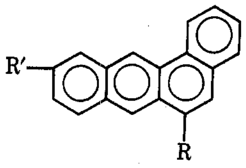
[3] M. S. Newman and W. B. Lutz, *J. Amer. Chem. Soc.*, **78**, 2469 (1956).

[4] M. S. Newman and D. Lednicer, *ibid.*, **78**, 4765 (1956).

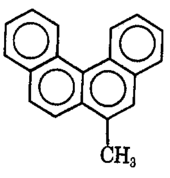
[5] Partial optical resolution of racemic 1-naphthyl 2-butyl ether and of 3,4,5,6-dibenzo-9,10-dihydrophenanthrene by chromatography on silicic acid impregnated with optically active TAPA has been reported by L. K. Klemm and D. Reed, *J. Chromatogr.*, **3**, 364 (1960).

desired for formation of polyester. By treatment of **6** with terephthaloyl chloride in dimethylformamide, a solid polymer (mp 85–90° dec) was obtained in high yield. No attempts were made to characterize the polymers obtained by conventional polymer techniques because the only point of interest in the present work was the chromatographic usefulness of the solids involved.

This polymer (of greenish yellow hue) was used as an adsorbent in a conventional chromatographic column and tested for its effectiveness in separating 6-methylbenz[*a*]anthracene (**7**) from 10-methylbenz[*a*]anthracene (**8**) and 6-methylbenzo[*c*]phenanthrene (**9**) from **8**, as compared to an equal weight of Woelm neutral alumina, grade I. The polymer proved to be slightly more effective than alumina in separating the very similar hydrocarbons **7** and **8**. The polymer was also effective in separating **9** from **8** but was slightly less effective than alumina for these hydrocarbons which differ from each other to a greater extent than do **7** and **8**. The success of these experiments prompted us to prepare an optically active polymer of similar structure.



7, R = CH_3; R′ = H
8, R = H; R′ = CH_3



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Attempts were also made to prepare polymers from **6** by reaction with 4,4′-diisocyanatodiphenylmethane, 2,2,4-trimethylhexamethylene diisocyanate, and 1,3-diisocyanato-1,5,5-trimethylcyclohexane. Although polymers were obtained in each case they were dark in color, of undesirable properties for chromatography, and were not further studied.

After many attempts to prepare **5** and **6** in optically active forms, the following procedure was developed. The acid **4** was obtained by careful hydrolysis of **3** with the calculated amount of sodium hydroxide. The acid **4** was resolved with dehydroabietylamine[6] and isolated as the (−)-dimethyl ester **3**. This was converted into (+)-**5** and the latter into (+)-**6** as for the racemic compounds. Treatment of (+)-**6** with terephthaloyl chloride yielded a (+) polymer which was very similar to the inactive polymer mentioned above.

Unfortunately, all attempts to effect resolution of 1-naphthyl 2-butyl ether or hexahelicene by chromatography failed to yield any fraction which had even small activity. In one experiment, the optically active polymer proved about as effective for separation of **7** and **8** as the inactive polymer. Hence, the failure to effect resolution cannot be due to greatly different dispersed forms of the two polymers.[7] Undoubtedly, the asymmetric center in the repeating unit of the polymer is not in a suitable position relative to the fluorene nucleus to induce preferential complex formation as is the case when TAPA (**1**) is used in solution,[7] and, therefore, even though the polymer is active, no resolution occurs on chromatography. It should be recognized that our attempts to effect resolution were carried out with small amounts of polymer and very low ratio of adsorbing agent to racemic compound. Possibly, if very high ratios were used, a small amount of resolution, comparable to that obtained by Prelog in the resolution of Troeger's base on lactose,[8] might be obtainable. The failure of an optically active polymer, which was used to resolve polymers by chromatography, to resolve a monomer should be noted.[9]

Experimental Section[10]

Dimethyl Isopropylideneaminooxysuccinate (3).—To the stirred solution under nitrogen prepared by dissolving 21.9 g (0.3 mol) of acetone oxime and 1.0 g of sodium methoxide in 70 ml of pure dry dioxane at 15–20° was added in one portion 43.2 g (0.3 mol) of dimethyl maleate. Within 2 min the temperature rose to 70°. The solution was left at ambient temperature overnight and then poured into 300 ml of ice water and acidified to litmus with hydrochloric acid. After the usual work-up, two distillations afforded 32.5 g (50%) of **3**, bp 118° (4 mm).

Anal. Calcd for $C_9H_{15}O_5N$: C, 49.7; H, 6.9. Found: C, 49.8; H, 7.2.

Isopropylideneaminooxysuccinic Acid (4).—In the best of several runs 21.7 g of **3** was added at one time to 200 ml of stirred 5% sodium hydroxide at 60° with cooling to keep the temperature below 70°. Within 5 min the mixture was homogenous. After 20 min the mixture was cooled, acidified to congo red, and saturated with ammonium sulfate (about 60 g). The product was taken into ether–benzene and worked up as usual to yield 16.0 g (85%) of **4**, mp 123–124°. The analytical sample, mp 123–124°, was obtained by crystallization from ether.

Anal. Calcd for $C_7H_{11}NO_5$: C, 44.4; H, 5.8; N, 7.4. Found: C, 44.7; H, 6.1; N, 7.7.

***dl*-2,4,5,7-Tetranitrofluorenylideneaminooxysuccinic Acid (TASA) (5).**—A solution of 36 g of TENF,[11] 21.7 g of **3**, and 3 g of toluenesulfonic acid in 400 ml of acetic acid in a three-necked flask connected to a packed fractionating column was refluxed for for 3 hr, the acetone formed being slowly removed. Then, at 1-hr intervals 10-, 5-, 5-, 3-, and 2-ml portions of water were added. After 3 hr more at reflux the mixture was poured on ice. After standing overnight, the yellow precipitate was collected, washed with cold water, and dried. Recrystallization from benzene–methanol yielded 44.0 g (89%) of **5**, mp 170–172°.

Anal. Calcd for $C_{17}H_9N_5O_{13}$. C, 48.5; H, 2.6; N, 12.3. Found: C, 48.2; H, 3.0; N, 12.5.

Di-β-hydroxyethyl 2,4,5,7-Tetranitrofluorenylideneaminooxysuccinate (6).—A solution of 49.1 g of **5** in 250 ml of ethylene glycol was added to 500 ml of ethylene glycol which had been saturated with dry hydrogen chloride at 0°. The mixture was left overnight at room temperature and was then diluted with water and extracted thoroughly with methylene chloride. After the usual work-up (a water wash) there was obtained 50.0 g (86%) of **6** as light yellow needles, unsharp melting point at 80°.

Anal. Calcd for $C_{21}H_{17}N_5O_{15}$: C, 43.6; H, 2.9; N, 12.1. Found: C, 43.4; H, 2.8; N, 12.1.

Inactive Polymer.—To a stirred solution of 11.58 g of **6** and 4.1 g of triethylamine in 75 ml of pure dimethylformamide (DMF) at room temperature was added rapidly a solution of 4.06

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(6) B. Sjöberg and S. Sjöberg, *Ark. Kemi*, **22**, 447 (1964).

(7) We are indebted to Dr. Lloyd R. Snyder, Union Oil Co., Brea, Calif., for this suggestion.

(8) V. Prelog and P. Wieland, *Helv. Chim. Acta*, **27**, 1127 (1944), used 2.7 kg of lactose for 6 g of base. See also G. M. Henderson and H. G. Rule, *J. Chem. Soc.*, 1568 (1939), for a review of early literature; C. W. Roberts and D. H. Haigh, *J. Org. Chem.*, **27**, 3375 (1962), and J. A. Loft and W. Riemann, III, *ibid.*, **31**, 561 (1966), for references to more recent work in this area.

(9) P. Pino, F. Ciardelli, G. P. Lorenzi, and G. Natta, *J. Amer. Chem. Soc.*, **84**, 1487 (1962).

(10) All melting points and boiling points are uncorrected. The term "worked up as usual" means that an ether or ether–benzene extract of the reaction mixture was washed with dilute acid and/or base, with saturated salt solution, and then filtered through a cone of anhydrous magnesium sulfate. The solvents were removed by distillation or rotary evaporation and the residue was processed as indicated. Microanalyses were performed by Galbraith Laboratories, Knoxville, Tenn. All new compounds had ir and nmr spectra consistent with the proposed formulas.

(11) M. S. Newman and W. B. Lutz, *J. Amer. Chem. Soc.*, **78**, 2469 (1956).

TABLE I

SEPARATION OF **7** AND **8** OVER 3 g OF POLYMER

Fraction	Wt,[a] mg	Vol[b]	% **7**[c]
1	0.0	4	
2	0.8	2	100[d]
3	6.0[e]	2	100
4	14.2	2	93
5	12.2	2	85
6	8.5	2	80
7	8.8	2	75
8	6.2	2	50
9	9.7	5	24
10	9.4	6.5	0
11–19	18.7[f]	81	0[g]
	Total 94.5		

[a] Weight in milligrams weighed to 4 decimals on an analytical balance. [b] Volume is approximate. [c] The difference between 100% and value shown consists of **8**. [d] This sample added to sample 3. [e] A total of 6.8 mg (13.6% of starting **7**) isolated pure. [f] A total of 28.1 mg (56.2% of starting **8**) isolated pure. [g] In a similar experiment run on an earlier day with the same polymer, there were isolated 4.8 mg (9.6%) of pure **7** and 29.3 mg (58.6%) of pure **8**, with a total of 87.7 mg recovered.

g of pure terephthaloyl chloride in 25 ml of DMF. After standing for 2 hr, the precipitated salt was filtered and washed with DMF. The filtrate and washings were then poured on crushed ice. After standing for 3 hr, the greenish yellow solid was collected, washed well with water, and air-dried. This polymer was then washed with methanol and dried (below 80°) to yield 13.5 g of polymer which decomposed when heated at 85–90°. After further washing with hot benzene and methanol repeatedly 7.0 g of polymer was obtained, suitable for use as the solid phase in chromatography.

Anal. Calcd for polymer unit as $C_{29}H_{19}N_5O_{17}$: C, 49.1; H, 2.7; N, 9.9. Found: C, 50.2; H, 2.9; N, 7.7.

Dimethyl (−)-Isopropylideneaminooxysuccinate (3).—A suspension of 262 g of purified dehydroabietylamine acetate[6,12] in 1 l. of water was treated with a solution of 65 g of sodium hydroxide in 100 ml of water in portions. The cooled reaction mixture was extracted with ether. After drying over potassium hydroxide pellets, the ether solution was added to a solution of 75.6 g of *dl*-**4** in 1 l. of ether–benzene (1:1). After standing at room temperature for 4 hr, the colorless solid was collected by filtration. Successive crops were obtained from the filtrate until a total of 130.8 g had been obtained. After four recrystallizations from methanol–chloroform (1:1), 76 g of the (−)(−) salt of **4**, mp 165–168°, was obtained. Attempts to obtain the (+)(−) salt from the mother liquors were unsuccessful. A solution of 75.9 g of the (−)(−) salt in 200 ml of dry methanol was added to 800 ml of methanol saturated at 0° with dry HCl. After standing overnight at room temperature 400 ml of methanol was distilled under reduced pressure. The mixture was diluted with water and extracted with ether. After the usual work-up distillation afforded 12.3 g (56%) of (−)-**3**, bp 145° (11 mm), $[\alpha]^{24}\mathrm{D}$ 23.01° (*c* 3, CCl_4).

(+)-**5**.—In a manner similar to that described for *dl*-**5**, a solution of 10.85 g of (−)-**3** and 18.0 g of TENF in 300 ml of acetic acid containing 1.0 g of toluenesulfonic acid yielded 23.2 g (94%) of (+)-**5**, mp near 180°, $[\alpha]^{25}\mathrm{D}$ 34.97° (*c* 4.86, methanol).

(+)-**6**.—Esterification of 19.64 g of (+)-**5** in 100 ml of ethylene glycol as in the case of *dl*-**5** yielded 20.1 g (87%) of (+)-**6**, melting point unsharp near 80°, $[\alpha]^{25}\mathrm{D}$ 29.06° (*c* 12.1, THF).

(+) **Polymer.**—A solution of 6.09 g of pure terephthaloyl chloride in 50 ml of pure DMF was added all at once to a solution of 17.37 g of (+)-**6** in 150 ml of DMF and 6.06 g of triethylamine in a 500-ml erlenmeyer flask with shaking. The active polymer was isolated as described for the inactive polymer. There was obtained 15.5 g of (+) polymer, $[\alpha]^{25}\mathrm{D}$ 31.18° (*c* 17.2, THF).

TABLE II

SEPARATION OF **7** AND **8** OVER 3 g OF ALUMINA

Fraction	Wt,[a] mg	Vol[b]	% **7**[c]
1–4	0.0	8	
5–7	7.6[d]	7	100
8	4.5	4	85
9	4.0	5	80
10–11	16.2	13	73
12–15	27.4	45	53
16–19	23.0	60	24
20	3.6	15	0
21	10.0[e]	15	0
	Total 96.3		

[a] Weight in milligrams. [b] Volume is approximate; larger volumes are those of combined fractions. [c] The difference between 100% and the number shown consists of **8**. [d] A total of 7.6 mg (15.2% of starting **7**) isolated pure in three fractions. [e] A total of 13.6 mg (27.2% of starting **8**) isolated pure. [f] In a similar experiment 8.6 mg (17.2%) of **7** and 12.0 mg (24%) of pure **8** were isolated, with a total recovery of 94.3 mg of starting **7** and **8**.

TABLE III

SUMMARY OF CHROMATOGRAPHIC SEPARATIONS

A. Separation of **7** and **8**[a]

Adsorbent	Amt of adsorbent, g	%[b] of pure **7**	% of pure **8**
Polymer	1.0	7	48
		9	54
Alumina	1.0	*c*	21
		c	22
Polymer	3.0	14	56
		10	59
Alumina	3.0	15	27
		17	24

B. Separation of **9** and **8**[a]

Adsorbent	Amt of adsorbent, g	% of pure **9**	% of pure **8**
Polymer	1.0	49	40
Alumina	1.0	71	65

[a] 50 mg of each used. [b] Percentages rounded off to nearest whole number. [c] No pure **7** obtained as first fractions were about 85% **7**.

Chromatographic Experiments.—Many experiments were run on known mixtures of 6-methylbenz[*a*]anthracene (**7**), 10-methylbenz[*a*]anthracene (**8**), and 6-methylbenzo[*c*]phenanthrene (**9**) over the polyester polymer prepared from inactive **6** and terephthaloyl chloride and over Woelm neutral alumina, grade I. In a typical experiment 50-mg each of the components to be separated was mixed with a small amount of adsorbent and placed on the chromatographic column which was covered by about 1 cm of cyclohexane. The internal diameter of the columns was about 1 cm. The height of absorbent was about 3–4 cm/g of polymer and 1–1.5 cm/g of alumina. Pure distilled cyclohexane was used as solvent and eluent. The eluate fractions, at a flow rate of about 1 ml/1–2 min, were collected in small weighed beakers which were weighed after evaporation of solvent in a draft.

Analyses of the various fractions of eluate were made by tracing the nmr spectra (taken in micro tubes for samples containing 3 mg or less) on heavy trace paper. The peaks were cut out and weighed in order to determine the relative amounts when mixtures were at hand. The nmr spectra were taken on a Varian A-60 with 250 sweep width, appropriate offset, and fast scanning (sweep time 100). The optimum rf energy was fed just in the region of resonance with suitable adjustment of the filter band width and phase to reduce the signal-to-noise ratio. Thus, it was possible to use fairly dilute solutions. The first fractions to be eluted were more difficult to estimate because of the dilution factor. Often several fractions were combined for analysis. The exact chemical shifts of the methyl groups (τ: **7**, *ca.* 7.4; **8**, *ca.* 7.6; **9**, 7.3)[13] were assigned both separately and in mixtures by using tetramethylsilane as internal standard. After assigning τ values for the methyl groups, it was unnecessary to include TMS in each tube for measurements of the composition of eluates.

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(12) L. F. Fieser and M. Fieser, "Reagents for Organic Synthesis," Wiley, New York, N. Y., 1967, p 183. We thank the Hercules Powder Co., Wilmington, Del., for a generous gift of Amine D.

(13) The exact τ value varied as in mixtures slight shifts were observed relative to the values in pure solvent (CCl_4 and $CDCl_3$).

Typical experiments are summarized in Tables I and II. The results of a series of experiments are listed in Table III.

Registry No.—(±)-**3**, 30256-03-2; (−)-**3**, 30318-67-3; (±)-**4**, 30256-04-3; (±)-**5**, 30247-99-5; (+)-**5**, 30248-00-1; (±)-**6**, 30275-69-5; (+)-**6**, 30248-01-2; (±) inactive polymer, 30228-77-4; (+) inactive polymer, 30228-78-5.

Synthesis of 2- and 3-Keto-5-*endo*-(2-imidazolyl)bicyclo[2.2.2]octane

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Received August 31, 1970

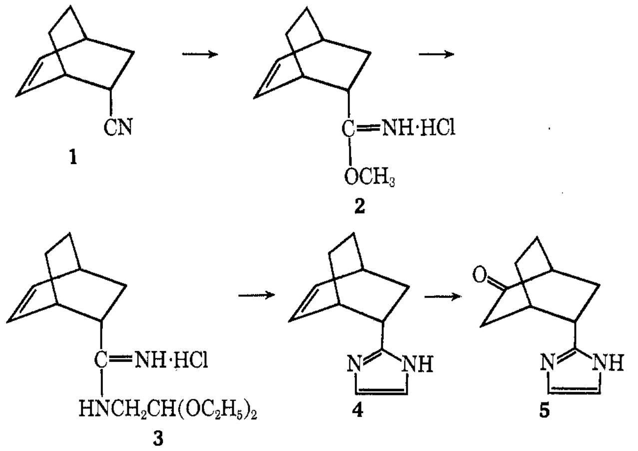
On the basis of a concept of attributes that make compounds attractive candidates for biological screening, 2-keto- and 3-keto-5-*endo*-(2-imidazolyl)bicyclo[2.2.2]octane were synthesized on a relatively large scale for conversion to a series of corresponding 2- or 3-substituted analogs.

In order to raise the probability of finding biological activity among candidates for pharmacometric screening, a family of compounds with structural features that should *a priori* enhance their effectiveness was synthesized. Three features included in this particular series were (1) a rigid bicyclic framework for favorable entropy of binding to a receptor, (2) an imidazole nucleus, attractive for its bifunctional nature and participation at active sites of enzymes, and (3) a second functional group chosen from those found among naturally occurring compounds. Although the biological activity of the group was disappointing, certain aspects of this synthetic organic chemistry are worthy of note.

On the basis of the criteria outlined above, 5-*endo*-(2-imidazolyl)bicyclo[2.2.2]oct-2-ene (**4**) was selected as an appropriate starting compound. The most logical way to synthesize **4** is by a Diels–Alder condensation between cyclohexadiene-1,3 and a 2-vinylimidazole,[1] but this method was soon abandoned in favor of a stepwise synthesis starting from 5-*endo*-cyanobicyclo[2.2.2]-oct-2-ene.

The target compound **4** was synthesized starting with the conversion of 5-*endo*-cyanobicyclo[2.2.2]oct-2-ene[2] (**1**) to 5-*endo*-carbiminomethoxybicyclo[2.2.2]-oct-2-ene hydrochloride (**2**) on treatment with methanol and hydrogen chloride in ether solution. Reaction of the imino ether **2** with β-aminoacetaldehyde diethyl acetal yielded *N*-(β,β-diethoxyethyl)-5-*endo*-carbamidinobicyclo[2.2.2]oct-2-ene hydrochloride (**3**), which at 120° in glacial acetic acid–acetic anhydride gave 5-*endo*-(2-imidazolyl)bicyclo[2.2.2]oct-2-ene (**4**).

A critical step in the scheme required the introduction at an appropriate position on the bicyclic framework of a carbonyl group which could then be converted into a variety of functional groups. The unsaturation at the 2,3 position of the bicyclic skeleton was a logical point to attack, but more difficulty was encountered than anticipated in applying typical reactions for this transformation. The imidazole moiety appeared to be largely responsible for the difficulty by either preventing reaction at the 2 or 3 position or by being susceptible to attack by the reagent. For example, in a variety of hydroboration studies, stable aminoborane derivatives were isolated and the unsaturated moiety was intact. In other instances involving mild oxidizing agents, the imidazole ring was attacked. Facile conversion of the unsaturated compound **4** to 2-keto-5-*endo*-(2-imidazolyl)bicyclo[2.2.2]octane (**5**) was achieved by palladium chloride oxidation in aqueous medium.[3] The reaction is usually accomplished by using catalytic quantities of palladium chloride in the presence of large amounts of cupric chloride and an air stream. The propensity of the keto imidazole derivative **5** to chelate with copper, however, made it prudent to use palladium chloride in at least stoichiometric amount and eliminate the cupric salt.



The location of the carbonyl group in **5** could not be established on theoretical grounds and was not immediately obvious from nmr spectra data in $CDCl_3$. In the absence of model compounds, chemical shifts could not be exploited with assurance, since the positions of protons adjacent to the ketone would be similar in both the 2 and 3 isomers. Furthermore, any difference in multiplicity between the bridgehead protons could not be used as an approach to structure assignment because of the coincidence of the C_1 and C_4 proton peaks in several solvents. These difficulties were overcome by taking advantage of the long-range coupling

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(1) The relatively poor dienophilic nature of vinylimidazoles and their propensity to polymerize at relatively low temperature did not augur well for a successful Diels–Alder condensation. In addition, considerable difficulty was justifiably anticipated in the prepartion of certain 2-vinylimidazoles on a reasonably large scale. Nevertheless, 1-benzyl-2-vinylimidazole was synthesized from 1-benzyl-2-lithioimidazole by reaction with acetaldehyde and dehydration over fused $KHSO_3$ at 251–230° (25–45 mm). The distilled product (*ca.* 60% pure) did not undergo Diels–Alder condensation with cyclohexadiene-1,3 at temperatures up to 170° or at pressures up to 138,000 psi. In a parallel attempt, methyl β-(4-imidazolyl)acrylate was prepared from L-histidine and heated with cyclohexadiene-1,3 at temperatures up to 210° without success.

(2) K. Alder, H. Heimbach, and R. Reubke, *Chem. Ber.*, **91**, 1516 (1958).

(3) For a review see J. Smidt, W. Hafner, R. Jira, R. Sieber, J. Sedlmeier, and A. Sabel, *Angew. Chem., Int. Ed. Engl.*, **1**, 80 (1962).