was 43%, assuming that four moles of aldehyde are required per mole of product.

Anal. Calcd. for $C_{19}H_{17}N$: C, 88.0; H, 6.55; N, 5.4. Found: C, 87.8; H, 6.5; N, 5.4.

The hydrochloride salt had a m.p. of 170–171°.

As summarized in Table II this method is applicable to other aromatic aldehydes to yield the corresponding 3,5-bis(arylmethyl)pyridines.

(b) *By condensation of N-benzoylpiperidine with benzaldehyde.* Rügheimer[3] reported a 72% yield of 3,5-dibenzylpyridine by heating a mixture of *N*-benzoylpiperidine and 2.15 mole equivalents of benzaldehyde in a sealed tube at 250° for 6 hr. However, it was necessary to employ conditions more stringent than those of Rügheimer when such a reaction was conducted in a stainless steel microbomb. Thus, in a reaction carried out at 250° for 6 hr., none of the dibenzylpyridine was isolated. Prolonging the heating time at 250° to 24 hr. gave 17.5% of the desired product. However, reaction at 300° for 12 hr. increased the yield of distilled (b.p. 177°/0.23 mm.) and recrystallized 3,5-dibenzylpyridine, m.p. 89–90° (lit.[3] m.p., 89°) to 54%. The hydrochloride salt melted at 168–171° (lit.[3] m.p., 164.5–166°) and did not depress the melting point of the product derived by reaction of benzaldehyde with piperidinium acetate.

(c) *From 3,5-pyridinedicarboxylic acid.* A mixture of 50 g. (0.3 mole) of 3,5-pyridinedicarboxylic acid and 150 ml. of thionyl chloride was refluxed for 16 hr. Excess thionyl chloride was removed by evaporation under reduced pressure; two 50-ml. portions of dry benzene were added and evaporated to remove the last of the thionyl chloride. The residual acid chloride was dissolved in 300 ml. of dry reagent benzene, and to this solution, cooled to 5 to 10°, was added 200 g. of anhydrous aluminum chloride with stirring. The reaction mixture was permitted to warm to room temperature and then refluxed with stirring for 6 hr. The dark brown mixture was poured cautiously onto ice and hydrochloric acid, and the solid 3,5-dibenzoylpyridine which formed was collected by filtration. Additional product was obtained by concentration of the benzene layer of the filtrate; yield, 78.7 g. (91%); m.p. 122.5–123.5°, unchanged after recrystallization from alcohol (lit.,[3] m.p., 123°).

A solution of 14.3 g. (0.05 mole) of 3,5-dibenzoylpyridine in 85 ml. of concd. hydrochloric acid and 100 ml. of water was added to 100 g. of amalgamated mossy zinc, and the mixture was boiled under reflux for 4 hr. in a manner similar to that described for reduction of 4-benzoylpyridine to 4-benzylpyridine.[7] A brown insoluble oil formed, and after separation by decantation, it was heated with 10% aqueous sodium hydroxide and benzene. The benzene layer was washed with water and the benzene removed by evaporation. The residual dark oil, which refused to crystallize, was distilled under reduced pressure, and the fraction boiling at 173–178° (0.2 mm.) was collected as a pale yellow oil which was crystallized and recrystallized from petroleum ether (b.p. 60–110°); yield, 5.8 g. (45%); m.p. 89–90° (reported,[3] m.p., 88.5–89°).

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(7) F. B. La Forge, *J. Am. Chem. Soc.*, **50**, 2484 (1928).

[CONTRIBUTION FROM THE DEPARTMENT OF CHEMISTRY, MASSACHUSETTS INSTITUTE OF TECHNOLOGY]

Stereoselective Synthesis of α-Substituted α,β-Unsaturated Esters

HERBERT O. HOUSE AND GARY H. RASMUSSON[1]

Received May 10, 1961

2-Methyl-*trans*-2,4-pentadienoic acid and its methyl ester have been prepared from acrolein and the ylid *3*. Reaction of the same ylid with acetaldehyde afforded an ester mixture composed of 96.5% methyl tiglate and 3.5% methyl angelate. Reaction of methyl pyruvate with the ylid *15* produced an ester mixture composed of 68% methyl tiglate and 32% methyl angelate.

In order to synthesize a sample of methyl 2-methyl-2,4-pentadienoate (*1*), needed for other synthetic work, we were led to consider various synthetic routes to α,β-unsaturated acids and their esters. The dienoic acid *2* has been prepared by the decarboxylative condensation of acrolein with malonic acid,[2] a reaction which is generally applicable to aldehydes forming the *trans* α,β-unsaturated acids.[3,4] Although substituted malonic acids have been successfully employed in comparable condensations with aromatic aldehydes,[5] the sensitivity[2] of the dienoic acid *2* to polymerization suggested that the direct formation of the desired ester *1* from acrolein and the Wittig reagent *3*[6–8] would be a more satisfactory route. In accord with this expectation, the ester *1* was produced in 60% yield. The liquid product, which exhibited a single peak on gas chromatography, yielded a single crystalline acid *4* on saponification; treatment of this acid *4* with diazomethane regenerated the ester *1*. Comparison of the ultraviolet spectra of the acid *4* [λ_{max} 251 mμ (ϵ 23,000)] and the ester *1* [λ_{max} 253 mμ (ϵ 24,300)] with the spectra of *2* [λ_{max} 244 mμ (ϵ 24,000)] and *5* [λ_{max} 247 mμ (ϵ 20,600)] indicated that product *4* possesses the indicated *trans* configuration.

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(1) National Science Foundation Predoctoral Fellow, 1958–61.

(2) I. E. Muskat, B. C. Becker, and J. S. Lowenstein, *J. Am. Chem. Soc.*, **52**, 326 (1930).

(3) J. R. Johnson, *Org. Reactions*, **1**, 210 (1942).

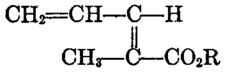
(4) Cf. H. E. Zimmerman and L. Ahramjian, *J. Am. Chem. Soc.*, **81**, 2086 (1959).

(5) W. J. Gensler and E. Berman, *J. Am. Chem. Soc.*, **80**, 4949 (1958).

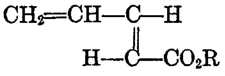
(6) O. Isler, H. Gutmann, M. Montavon, R. Rüegg, G. Ryser, and P. Zeller, *Helv. Chim. Acta*, **40**, 1242 (1957).

(7) U. Schöllkopf, *Angew. Chem.*, **71**, 260 (1959).

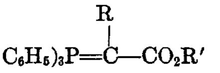
(8) S. Trippett in R. A. Raphael, E. C. Taylor, and H. Wynberg, *Advances in Organic Chemistry, Methods and Results*, Vol. **1**, Interscience Publishers, Inc., New York, N. Y., 1960, pp. 83–102.


$CH_2=CH-C(H)=C(CH_3)-CO_2R$

1, R = CH$_3$
4, R = H

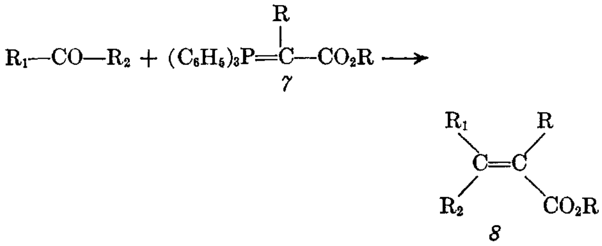

$CH_2=CH-C(H)=C(H)-CO_2R$

2, R = H
5, R = CH$_3$


$(C_6H_5)_3P=C(R)-CO_2R'$

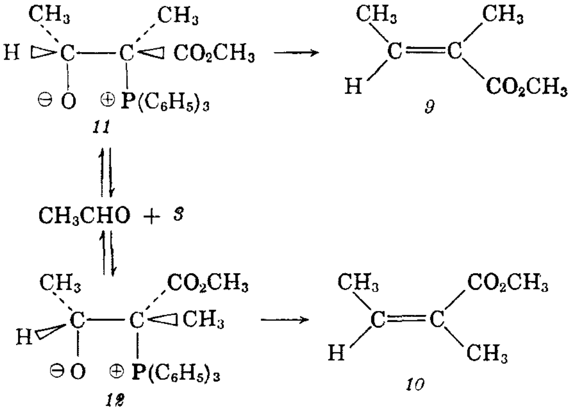
3, R = R' = CH$_3$
6, R = H, R' = C$_2$H$_5$

These observations as well as the previous reports[7,8] that unsaturated esters from the ylid *6* possessed the configuration in which the carboalkoxy group was *trans* to the larger group at the *beta*-carbon atom, suggested that the reaction of ylids such as *7* with aldehydes and ketones might generally be expected to produce unsaturated esters *8* in which R$_1$ is larger than R$_2$. In other words the stereochemical outcome of this reaction would parallel the Perkin and related reactions.[3,4]

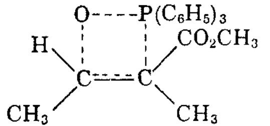


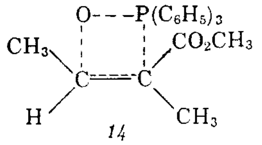
To explore this idea the reaction of the ylid *3* with acetaldehyde was studied since the expected products methyl tiglate *9* and methyl angelate *10* have thoroughly established configurations.[9]

The product of this reaction was methyl tiglate (*9*) containing no more than 3.5% of methyl angelate (*10*). Furthermore, we were able to demonstrate that methyl angelate *10* was not isomerized by the conditions employed for the reaction and product isolation.

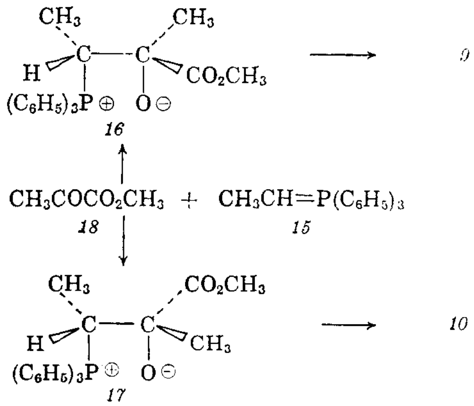


These results are to be contrasted with those obtained with alkylidene and arylidene ylids[8,10] where significant amounts of both stereoisomers of the olefinic product are formed. For the reaction considered here, the difference in steric interactions[10] resulting from the formation of intermediates *11* and *12* appears much too small to account for the degree of stereoselectivity observed. However, the observed stereoselectivity would be in accord with the *reversible* formation of intermediates *11* and *12* if one assumes that the activation energy required for conversion of the intermediate *11* to the tiglic ester *9* is less than the energy required for the conversion *12* to *10*.[11] This assumption may be justified if the transition state *13* for the conversion *11* to *9* is considered. In this transition state *13* overlap between the π orbitals of the carbonyl group and the incipient carbon-carbon double bond would be expected to lower the energy of the transition state. However, this overlap is possible only when the carbonyl function lies in the plane of the developing carbon-carbon double bond. In the corresponding transition state *14* for the conversion *12* to *10*, the β-methyl group offers interference to this coplanarity requirement leading to the expectation that the activation energy required for the change *12* to *14* would be greater than for the change *11* to *13*.


13


14

These considerations suggest that substantial stereoselectivity may be anticipated in Wittig reactions forming conjugated systems if the starting ylid (*e.g.*, *3*) is sufficiently stable to permit reversible formation of the intermediate betaines (*e.g.*, *11* and *12*).[12] On the other hand use of a highly reactive ylid (*e.g.*, *15*) should oppose reversible formation of the intermediate betaines (*e.g.*, *16* and *17*) and the stereochemical outcome of



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(9) R. E. Buckles, G. V. Mock, and L. Locatell, Jr., *Chem. Revs.*, **55**, 659 (1955).

(10) J. P. Dusza, *J. Org. Chem.*, **25**, 93 (1960).

(11) For discussion of a comparable situation in the Darzen's Condensation see H. E. Zimmerman and L. Ahramjian, *J. Am. Chem. Soc.*, **82**, 5459 (1960).

(12) For a discussion and an experimental demonstration that the betaine from triphenylphosphinemethylene and benzaldehyde is not formed reversibly see G. Wittig. H. Weigmann, and M. Schlosser, *Chem. Ber.*, **94**, 676 (1961),

such reactions should be determined by the relative ease of formation of the betaines. To test this prediction the reaction of methyl pyruvate (*18*) with the ylid *15* was studied. In contrast to the high degree of stereoselectivity obtained in the reaction of acetaldehyde with the ylid *3* the ester mixture obtained from the pyruvic ester *18* and *15* was composed of 68% methyl tiglate (*9*) and 32% methyl angelate (*10*).

EXPERIMENTAL[13]

2,4-Pentadienoic acid (*2*). The reaction of 118 g. (0.53 mole) of potassium metabisulfite, 29 g. (0.52 mole) of acrolein and 52 g. (0.50 mole) of malonic acid in 200 ml. of water as previously described[2] afforded the crude dienoic acid which was recrystallized from an ether–petroleum ether (b.p. 30–60°) mixture. The pure acid separated as 9.78 g. of pale yellow needles, m.p. 71.5–72.5° (lit.[2] m.p. 72°), which exhibit infrared absorption[14] at 3000 cm.$^{-1}$ (broad, assoc. O—H), at 1690 cm.$^{-1}$ (conj. carboxyl C=O), at 1635 cm.$^{-1}$ (conj. C=C), at 930 cm.$^{-1}$ with a shoulder at 950 cm.$^{-1}$ (*trans* CH=CH) and at 875 cm.$^{-1}$ (conj. CH$_2$=CH—). The acid has an ultraviolet maximum[15] at 244 mμ (ϵ 24,000). Concentration of the mother liquors afforded an additional 4.09 g. of crude acid, m.p. 66.5–69.5° (total yield 13.87 g. or 27.4%).

An 8.4-g. (0.10 mole) sample of the acid *2* in ether was allowed to react with a slight excess of diazomethane. After the resulting solution had been washed successively with aqueous sodium bicarbonate and water, dried over magnesium sulfate and concentrated, distillation of the residue separated 5.85 g. (61%) of the *methyl ester 5*, b.p. 36–38° (10 mm.) [lit.[16] b.p. 77–80° (25 mm.)], n_{D}^{23} 1.4833, with infrared absorption[17] at 1720 cm.$^{-1}$ (conj. ester C=O), at 1645 and 1605 cm.$^{-1}$ (conj. C=C), at 925 cm.$^{-1}$ (*trans* CH=CH) and at 870 cm.$^{-1}$ (conj. CH$_2$=CH–) and an ultraviolet maximum[15] at 247 mμ (ϵ 20,600). A gas chromatogram[18] of the ester exhibits a single peak.

Methyl 2-methyl-2,4-pentadienoate (*1*). The reaction of 209 g. (0.80 mole) of triphenylphosphine with 126 g. (0.75 mole) of methyl α-bromopropionate in 960 ml. of benzene followed by reaction of the crude phosphonium salt with aqueous potassium hydroxide as previously described[6] yielded the crude phosphorane *3*, m.p. 146–152°, yield 111.3 g. (44%). Recrystallization of a 91-g. portion of this material from an ethyl acetate–petroleum ether mixture separated 83 g. of the pure *phosphorane 3*, m.p. 152–154.5° (lit.[6] m.p. 152–153°), which exhibits broad, intense infrared absorption[14] at 1600 cm.$^{-1}$ with an ultraviolet maximum [15] at 226 mμ (ϵ 26,000) and a series of low intensity peaks (ϵ 4000 to 4900) in the region 260–275 mμ.

To a solution of 64.0 g. (0.184 mole) of the ylid *3* in 110 ml. of methylene chloride was added, dropwise and with stirring, 12.6 g. (0.225 mole) of acrolein. After the resulting mixture had been refluxed for 3.5 hr., the solution was concentrated by the distillation of approximately half of the methylene chloride from the mixture through a 30-cm. Vigreux column. The residual solution was diluted with 250 ml. of petroleum ether and filtered to remove the triphenylphosphine oxide which separated. After the precipitated triphenylphosphine oxide had been washed with an additional 100 ml. of petroleum ether, the combined organic solutions were concentrated. Distillation of the residue afforded 13.78 g. (60%) of the ester, b.p. 50–50.5° (7–8 mm.), n_{D}^{26} 1.4882. The gas chromatogram[19] exhibited a major peak corresponding to 96% of the volatile material. A pure sample was collected from the chromatograph for analysis. The material has infrared absorption[17] at 1715 cm.$^{-1}$ (conj. ester C=O) at 920 cm.$^{-1}$ (CH$_2$=CH—) with an ultraviolet maximum[15] at 253 mμ (ϵ 24,300).

Anal. Calcd. for $C_7H_{10}O_2$: C, 66.64; H, 7.99. Found: C, 66.59; H, 8.04.

A solution of 1.382 g. (0.011 mole) of the methyl ester *1* in 15 ml. of a 6% solution of sodium hydroxide in methanol was refluxed for 2 hr. under a nitrogen atmosphere and then concentrated under reduced pressure and diluted with 35 ml. of water. The resulting solution was acidified at 0° and then extracted with chloroform. After the extract had been washed with water, dried over magnesium sulfate and concentrated, the crude solid residue (1.12 g.) was recrystallized from an ether–petroleum ether mixture. The acid *4* separated as white needles, m.p. 60.4–63°, yield 0.82 g. (67%). Sublimation of this material under reduced pressure afforded the pure *acid 4*, m.p. 63.8–65°, which exhibits infrared absorption[17] at 3000 cm.$^{-1}$ (broad, assoc. O—H), at 1685 cm.$^{-1}$ (conj. carboxyl C=O) and at 925 cm.$^{-1}$ (CH$_2$=CH—) and an ultraviolet maximum[15] at 251 mμ (ϵ 23,000).

Anal. Calcd. for $C_6H_8O_2$: C, 64.27; H, 7.19. Found: C, 63.99; H, 7.19.

An ethereal solution of 1.03 g. (0.0090 mole) of the acid *4* was treated with a slight excess of diazomethane and the crude product was isolated in the usual manner. Distillation afforded 0.761 g. (67%) of the methyl ester *1*, b.p. 48–48.5° (7 mm.), n_{D}^{25} 1.4901, which had infrared and ultraviolet absorption identical with the previously described sample.

Methyl tiglate (*9*). An authentic sample of tiglic acid[20] [infrared bands[17] at 2950 cm.$^{-1}$ (broad, assoc. O—H), at 1680 cm.$^{-1}$ (conj. carboxyl C=O) and at 1640 cm.$^{-1}$ (conj. C=C), ultraviolet maximum[15] at 212 mμ (ϵ 11,400)] was converted to the methyl ester *9* by reaction with diazomethane in the usual manner. After distillation through a short path still, the ester *9*, n_{D}^{26} 1.4344 (lit.[9] n_{D}^{20} 1.4370), exhibits infrared absorption[18] at 1720 cm.$^{-1}$ (conj. ester C=O) and at 1635 cm.$^{-1}$ (conj. C=C) with an ultraviolet maximum[15] at 215 mμ (ϵ 11,500). The gas chromatogram[21] of the ester exhibits a single peak.

Methyl angelate (*10*). A sample of 2-bromo-*trans*-2-butene, b.p. 39–41.5° (133–137 mm.), $n_{\mathrm{D}}^{25.5}$ 1.4552, containing[21] 98% of the desired isomer, was prepared as previously described.[22] A solution of 40.6 g. (0.30 mole) of the bromobutene in 100 ml. of ether was added, dropwise and with stirring under a nitrogen atmosphere, to a suspension of 4.2 g. (0.61 mole) of lithium in 150 ml. of ether cooled in an ice bath. The resulting solution of the organolithium compound was poured onto a slurry of excess Dry Ice in 200 ml. of ether. The resultant mixture was allowed to warm to room temperature and then diluted with aqueous hydrochloric acid. After the ether layer had been separated and extracted with sodium bicarbonate, the bicarbonate

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(13) All melting points are corrected and all boiling points are uncorrected. The infrared spectra were determined with either a Baird, Model B, or a Perkin-Elmer, Model 21, infrared recording spectrophotometer fitted with a sodium chloride prism. The ultraviolet spectra were determined with a Cary recording spectrophotometer, Model 11MS. The microanalyses were performed by Dr. S. M. Nagy and his associates and by the Scandinavian Microanalytical Laboratory.

(14) Determined in chloroform solution.

(15) Determined as a solution in 95% ethanol.

(16) E. P. Kohler and F. R. Butler, *J. Am. Chem. Soc.*, **48**, 1036 (1926).

(17) Determined in carbon tetrachloride solution.

(18) A column packed with 1,2,3-tris(2-cyanoethoxy)propane suspended on ground firebrick was employed.

(19) A column packed with Dow Silicone Fluid No. 550 suspended on ground firebrick was employed.

(20) Aldrich Chemical Co., Inc., Milwaukee, Wis.

(21) A column packed with 20 M Carbowax suspended on ground firebrick was employed.

(22) H. O. House and R. S. Ro, *J. Am. Chem. Soc.*, **80**, 2428 (1958) and references therein.

solution was acidified and extracted with ether. The ethereal solution was dried over magnesium sulfate and concentrated to leave 12.3 g. of crude angelic acid, m.p. 38–43°, which was recrystallized to separate 10.4 g. (35%) of angelic acid as white prisms, m.p. 40–43° (lit.[9] m.p. 45.0–45.5°), with infrared absorption[17] at 2950 cm.$^{-1}$ (broad, assoc. O—H), at 1685 cm.$^{-1}$ (conj. carboxyl C=O) and at 1640 cm.$^{-1}$ (conj. C=C) and an ultraviolet maximum[15] at 213 mμ (ε 7,400). A 5.0-g. (0.05 mole) sample of this acid, after esterification with diazomethane in the usual manner, yielded 4.48 g. (79%) of ester *10*, b.p. 45° (26–28 mm.), $n^{25.5}_D$ 1.4292 (lit.[9] b.p. 127.6–128°, n^{25}_D 1.4321) which contained[21] more than 98% of methyl angelate.[23] The ester *10* has infrared bands[17] at 1715 cm.$^{-1}$ (conj. ester C=O) and at 1645 cm.$^{-1}$ (conj. C=C) with an ultraviolet maximum at 216 mμ (ε 9,200).

Reaction of the phosphorane 3 with acetaldehyde. To a solution of 30.0 g. (0.086 mole) of the ylid *3* in 100 ml. of methylene chloride was added, dropwise under a nitrogen atmosphere, 7.8 g. (0.18 mole) of acetaldehyde. The resulting solution was allowed to stand overnight and then concentrated, diluted with 200 ml. of petroleum ether and filtered to remove the precipitated triphenylphosphine oxide. Distillation of the filtrate separated 8.84 g. (90%) of a product mixture, b.p. 68–69° (62–65 mm.), n^{25}_D 1.4274, containing[21] 93.5% methyl tiglate and 3.2% methyl angelate corresponding to an ester mixture containing 96.5% methyl tiglate and 3.5% methyl angelate. Fractional distillation of this mixture separated a sample of pure methyl tiglate, $n^{25.5}_D$ 1.4337, which had infrared absorption identical with that of the authentic sample.

To establish that methyl angelate would be stable under the conditions of this preparation, a solution of 3.94 g. (0.0345 mole) of methyl angelate, 9.6 g. (0.0345 mole) of triphenylphosphine oxide and 3 mg. of the ylid *3* in 25 ml. of methylene chloride was refluxed for 4 hr. and then worked up as previously described. The recovered methyl angelate, 2.15 g. (55%), b.p. 43.5–44.5° (26 mm.), n^{25}_D 1.4281, contained[18] less than 1% of methyl tiglate.

Reaction of the phosphorane 15 with methyl pyruvate (18). Reaction of 39 g. (0.15 mole) of triphenylphosphine with 22 g. (0.20 mole) of ethyl bromide in 50 ml. of benzene at 135° in an autoclave for 20 hr. followed by recrystallization of the precipitated phosphonium salt from water and subsequent drying afforded 49.5 g. (89%) of ethyltriphenylphosphonium bromide as white prisms, m.p. 209–210.5° (lit.[24] m.p. 203–204°).

An ethereal solution (90 ml.) containing 0.105 mole of phenyllithium was added, with stirring in a nitrogen atmosphere, to a suspension of 40.0 g. (0.108 mole) of the phosphonium salt in 200 ml. of ether. After the addition was complete, the resulting red solution was stirred for 1 hr. at which time the mixture gave a negative Gilman color test for phenyllithium. The ethereal solution of the ylid was added, dropwise and with stirring in a nitrogen atmosphere, to a solution of 9.0 g. (0.088 mole) of methyl pyruvate in 10 ml. of ether until the red color of the mixture persisted in the reaction mixture. At this point the reaction mixture was heated to reflux, the remainder of the ylid solution was added, and the resulting mixture was refluxed overnight with stirring. The bulk of the ether was distilled from the pasty reaction mixture and 400 ml. of tetrahydrofuran was added. The resulting mixture was refluxed for 36 hr. under a nitrogen atmosphere and again concentrated to about 75 ml. by distillation of the tetrahydrofuran through a 30-cm. Vigreux column. The reaction mixture was diluted with ether and the ethereal solution was separated from an insoluble semisolid and distilled. The ester mixture obtained, b.p. 34–36° (25 mm.), $n^{24.5}_D$ 1.4396–1.4405, amounted to 2.10 g. (21%) and contained[19] 68% methyl tiglate and 32% methyl angelate. Samples collected from the chromatograph[19] were identified by comparison of their spectra with the spectra of authentic samples.

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[23] The substantial contamination with methyl tiglate noted in the previous preparation [A. S. Dreiding and R. J. Pratt, *J. Am. Chem. Soc.*, **76**, 1902 (1954)] of angelic acid by this procedure was almost certainly due to the use of impure bromo olefin as the starting material (see ref. 22).

[24] G. Wittig and D. Wittenberg, *Ann.*, **606**, 1 (1957).

[CONTRIBUTION FROM THE DEPARTMENT OF CHEMISTRY, UNIVERSITY OF COLORADO]

Diels-Alder Reactions of 9-Substituted Anthracenes. V. Adducts of Ethylene[1]

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MARY F. WILCOX, WESLEY G. CLARK, AND TRENT TIEDEMAN

Received December 22, 1960

The condensation of ethylene with 9- and 9,10-disubstituted anthracenes at 170–200° under pressure is a convenient way of preparing bridgehead compounds. A variety of anthracenes are readily available and undergo this reaction.

Some time ago we announced a convenient synthesis of bridgehead compounds by the condensation of a 9-substituted anthracene with ethylene.[2] This method has been used since then in two different studies by others.[3,4]

The ready availability of a variety of 9-substituted anthracenes, together with satisfactory yields, makes this method a general one for obtaining 9-substituted 9,10-dihydro-9,10-ethanoanthracenes.

Ethylene, the simplest possible dienophile, was first used in a Diels-Alder reaction by Joshel and Butz in 1941.[5] They condensed it with 1,3-butadiene, 2,3-dimethyl-1,3-butadiene and cyclopentadiene at temperatures of almost 200°.

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[1] Supported in part by the National Science Foundation.

[2] Presented in part at the 123rd meeting of the American Chemical Society, Los Angeles, Calif., March 1953.

[3] M. Wilhelm and D. Y. Curtin, *Helv. Chim. Acta*, **40**, 2129 (1957).

[4] T. W. Campbell, E. V. McCoy, J. C. Kauer, and V. S. Foldi, *J. Org. Chem.*, **26**, 1422 (1961).

[5] L. M. Joshel and L. W. Butz, *J. Am. Chem. Soc.*, **63**, 3350 (1941).